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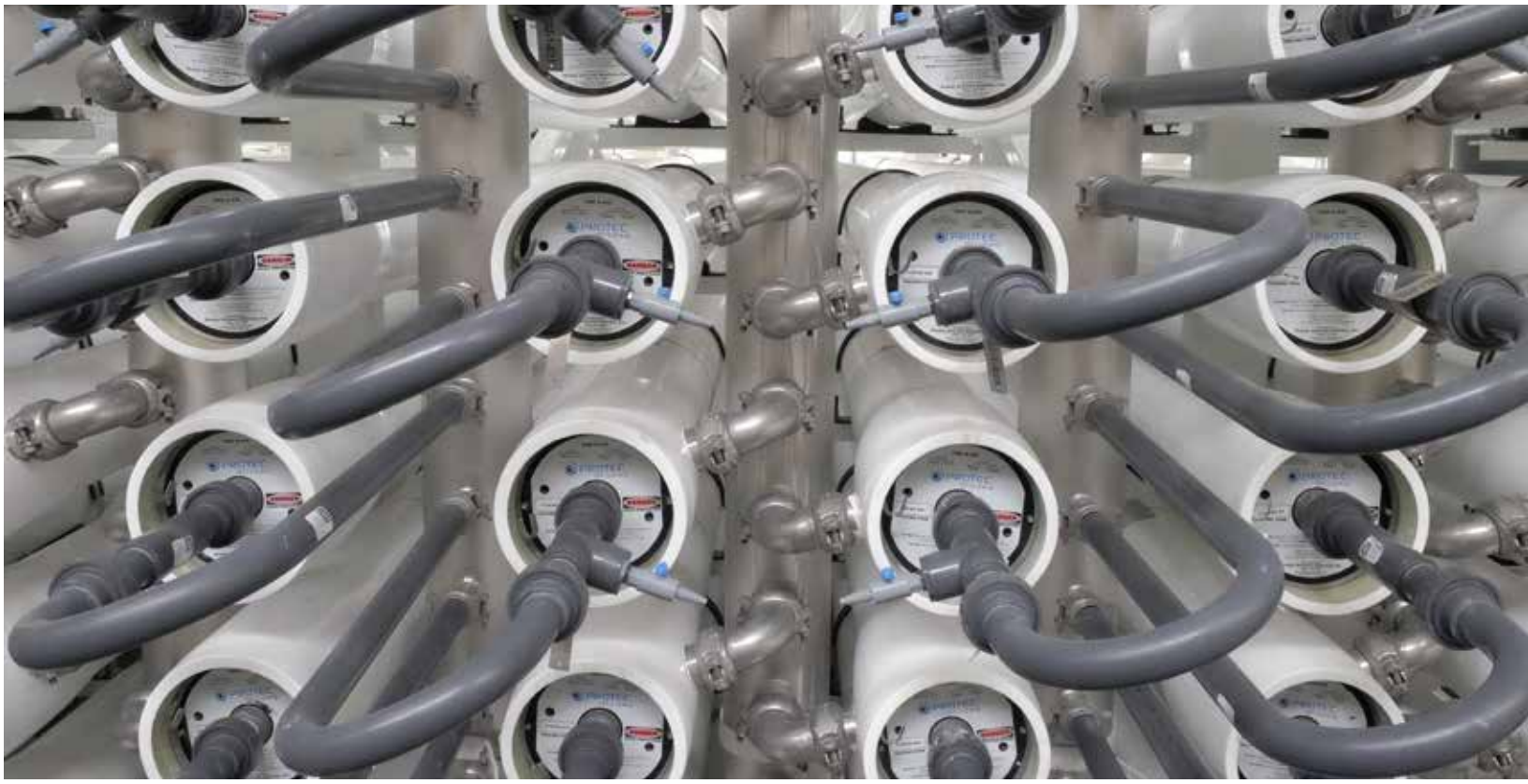

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New Techniques, Tools, and Validation Protocols for Achieving Log Removal Credit Across High-Pressure Membranes

CALIFORNIA Water Boards
STATE WATER RESOURCES CONTROL BOARD
REGIONAL WATER QUALITY CONTROL BOARDS

New Techniques, Tools, and Validation Protocols for Achieving Log Removal Credit Across High-Pressure Membranes

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Abstract and Benefits

Abstract:
Integral reverse osmosis (RO) and nanofiltration (NF) membranes are capable of robust rejection of dissolved ions in water and are therefore sufficient to act as a significant barrier to pathogens such as viruses, *Cryptosporidium*, and *Giardia.* Pathogens of concern are much larger than the molecular weight cutoff values for RO and NF membranes, thus it is expected that RO and NF consistently achieve significant log-removal credits for virus and protozoa. However, meeting the United States Environmental Protection Agency (USEPA) membrane filtration guidance manual (MFGM) integrity monitoring requirements and demonstrating more than 2-log removal has proven a challenge with NF and RO membranes.

The goal of this research was to identify native markers or surrogates that can be implemented at scale now and have the resolution and sensitivity sufficient to demonstrate a log reduction value (LRV) greater than 2-log by NF and RO. In addition, the recommended surrogates were evaluated against established USEPA MFGM criteria of resolution (i.e., how small a defect can be verified), sensitivity (i.e., what magnitude of LRV is able to be demonstrated), and frequency (i.e., can the technique verify integrity and allow corrective action at least daily).

A literature review was conducted to identify practical integrity monitoring approaches, followed by pilot- and/or full-scale testing of various promising approaches. This included a novel technique to calculate LRV from conductivity profiling results that differentiates between defect and nominal diffusive flow of conductivity, increasing sensitivity. The direct removal of native markers including sulfate, strontium, sucralose, phosphate, and magnesium, as well as a spiked fluorescent marker, uranine, were also investigated. Integrity monitoring approaches were evaluated at three full-scale sites and supplemented with pilot scale investigations to quantify maximum sensitivity.

Results suggest that native markers and surrogates would be limited to demonstrating a maximum LRV between 3.0 to 4.0-log. The limitation to a maximum of 4.0-log was due to diffusion across intact parts of the membrane. In practice, the maximum sensitivity of native markers was 3.0-log and was limited by their abundance in feedwater and the detection limit available in RO permeate, characteristics consistent with other studies in the literature. Nevertheless, a potential regulatory credit of 3.0-log represents a meaningful improvement over the more typical 2-log removal. The maximum sensitivity of conductivity profiling to calculate an LRV was conservatively limited to 5.0 log. In practice, intact RO arrays studied demonstrated typical conductivity profiling LRVs of 3.0 to 5.0-log. Thus, diffusion-adjusted conductivity profiling appears promising for further study and demonstration.

Benefits:

- Practical and available surrogates were evaluated against USEPA MFGM criteria.
- A novel diffusion-adjusted conductivity profiling approach to calculate LRV was investigated at multiple full-scale facilities and was able to demonstrate LRVs between 3.0 and 5.0-log.
- Practical limitations prevent demonstration of LRVs greater than 3.0 log with native surrogates.
- Of the native surrogates investigated, sulfate appears to provide consistently high LRVs.

- Approaches for regulatory approval based on the surrogates investigated in this report were proposed.

Keywords: Reverse Osmosis, Integrity Monitoring, LRVs, Water Reuse

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Acronyms and Abbreviations

◦F	degrees Fahrenheit
°C	degrees Celsius
ANSI	American National Standards Institute
AOP	advanced oxidation process
ASDWA	Association of State Drinking Water Administrators
ASTM	ASTM International
ATP	adenosine triphosphate
AWPF	advanced water purification facility
BGRS	Beenyup Groundwater Replenishment Scheme
C12H19Cl3O8	sucralose
cATP	cellular Adenosine Triphosphate
CDOM	chromophoric dissolved organic matter
CF	concentrate flow
CIIM	Continuous indirect integrity monitoring
CIP	clean-in-place
Cl2	chlorine
CO2	carbon dioxide
Co3+	Cobalt(III)
Da	dalton
DI	deionized
DIT	direct integrity test
DLS	dynamic light scattering
DOC	dissolved organic carbon
DOM	dissolved organic matter
DPR	direct potable reuse
EC	electrical conductivity
EEM	excitation-emission matrix
FCM	flow cytometry
FF	feed flow
ft2	square feet
g/L	grams per liter
g/mole	grams per mole
gfd	gallons per square foot per day
gpm	gallons per minute
GWRS	Groundwater Replenishment System
H+	hydronium
Hg	mercury

HPLC/IC	high-performance liquid chromatography ion chromatography
IC	ion chromatography
IPR	indirect potable reuse
L	liter(s)
LRV	log reduction value
LT2ESWTR	Long Term 2 Enhanced Surface Water Treatment Rule
MDL	method detection limit
MF	microfiltration
MFGM	Membrane Filtration Guidance Manual
mg/L	milligrams per liter
mgd	million gallons per day
mg-h/L	milligrams per liter times hours
mL	milliliter
mm Hg	millimeters
MRL	method reporting limit
mS/cm	millisiemens per centimeter
MS2	surrogate virus MS2 bacteriophage
MW	molecular weight
N/A	not applicable, analysis not performed
NaCl	sodium chloride
NCWRP	North City Water Reclamation Plant
NDP	normalized differential pressure
NF	nanofiltration
ng/L	nanograms per liter
nm	nanometer
NPF	normalized permeate flow
NSF	National Science Foundation
NTU	Nephelometric Turbidity Unit
OC San	Orange County Sanitation District
OCWD	Orange County Water District
PBS	phosphate buffered saline
PDT	pressure decay tests
PES	polyethersulfone
PF	permeate flow
PFU	plaque forming unit
pg/mL	picograms per milliliter
PM-MIMo	pulsed-marker membrane integrity monitoring
ppb	parts per billion
ppm	parts per million

psi	pounds per square inch
PV	pressure vessel
QCRV	quality control removal value
qPCR	quantitative polymerase chain reaction
R&D	research and development
RF	recycle flow
RL	reporting limit
RO	Reverse osmosis
ROF	reverse osmosis feed
ROP	reverse osmosis permeate
R-WT	Rhodamine WT
SCADA	supervisory control and data acquisition
SNWA	Southern Nevada Water Authority
SO4	Sulfate
SPI	Separation Processes, Inc, Carlsbad, CA
TDS	total dissolved solids
TOC	total organic carbon
TRASAR	3D TRASAR
UF	ultrafiltration
USEPA	United States Environmental Protection Agency
UV	ultraviolet
UV/H2O2	ultraviolet disinfection and hydrogen peroxide addition
VCF	volumetric concentration factor
WCWA	Water Corporation of Western Australia
WRWRF	Wochholz Regional Water Recycling Facility
WWTP	wastewater treatment plant
YVWD	Yucaipa Valley Water District
µg/L	micrograms per Liter
µm	micrometer
µS/cm	microsiemens per centimeter
σ	standard deviation

Executive Summary

Traditionally used as a key component in desalination processes, intact reverse osmosis (RO) and nanofiltration (NF) membranes are capable of robust rejection of dissolved ions in water and are therefore sufficient to act as a significant barrier to pathogens such as viruses, *Cryptosporidium*, and *Giardia.* Currently, California and other U.S. states (i.e., Colorado, Florida, Texas, New Mexico, and Arizona) are developing direct potable reuse regulations and/or guidelines with similar pathogen LRV requirements. Because of the acute public health implications associated with pathogens, drinking water regulations for low-pressure membrane systems (i.e., microfiltration or ultrafiltration) require performing both continuous indirect integrity monitoring (CIIM) using turbidity and a once-daily direct integrity test (DIT) using pressure decay tests [PDT]. Based on the USEPA's Membrane Filtration Guidance Manual (MFGM) (USEPA 2005), which was developed for use in conjunction with implementation of the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) for *Cryptosporidium* removal, a DIT must meet criteria of resolution (i.e., how small of a defect can be verified), sensitivity (i.e., what magnitude of LRV is able to be demonstrated), and frequency (i.e., can the technique verify integrity and allow corrective action at least daily).

Development of DITs according to MFGM criteria for RO and NF systems (high-pressure membranes) has been challenging due to the differences in how these systems operate compared to low-pressure membranes. As the fundamental basis for regulation of membrane systems, requiring state regulators to interpret the MFGM language to grant approval of LRVs for RO/NF results in significant differences in regulatory approval and may not be the proper use of the MFGM. Evolving potable reuse regulations require the water industry to adapt existing tools and develop new strategies to achieve the very high LRVs (particularly for direct potable reuse) required to protect public health. In the absence of a federal revision to the MFGM or a supplemental document, states will continue to develop their own analyses and conclusions related to pathogen removal by NF and RO.

Monitoring and verification of membrane integrity is necessary for high-pressure membrane systems as they can be compromised or breached through a variety of mechanisms such as damaged O-rings, glue line leaks, and particulate damage. These breaches may be sufficient to allow pathogens to pass through the membrane system and into the permeate (Jacangelo et al., 2019; Jacangelo and Gray, 2015). Nominal RO membrane systems, when challenged with the surrogate virus MS2 bacteriophage (MS2), typically demonstrate LRVs of 5.0 to >6.0 log. Removal of O-rings during challenge testing with MS2 was reported to result in a reduction of LRV from approximately 4.0 to 2.0-log, and a corresponding change in conductivity LRV from 1.5 to approximately 1.0-log was observed (Carollo Engineers, Inc. 2016). Challenge testing cannot feasibly be performed at a frequency sufficient to protect public health and is impractical for larger systems. For RO, LRVs of 1.5 to 2.0-log units for viruses, *Giardia*, and *Cryptosporidium* are typically credited based on the LRV of conductivity and/or total organic carbon (TOC) from the bulk RO feed to the RO permeate (Vickers et al., 2019). There continues to be a gap between the actual pathogen removal of high-pressure membrane systems and what is able to be demonstrated with accepted integrity monitoring approaches. Integrity

testing approaches with sensitivity to demonstrate higher LRVs, that can be feasibly implemented to verify performance daily are needed to bridge this gap.

ES.1 Project Goals

It is worth noting that several reviews on high-pressure membrane integrity have been published (Jacangelo et al. 2019; Kitis et al. 2003a; Niewersch et al. 2020; Yoon 2019). In particular, a recently conducted literature review provided a comprehensive list of high-pressure membrane integrity approaches (Jacangelo et al. 2019). Compared to prior efforts to develop integrity monitoring and crediting frameworks for RO and NF, the goals of this report differ and focus on:

- Shortlisting and evaluation of surrogates with respect to criteria established in the MFGM in an effort to facilitate use of the proposed surrogates.
- Evaluation of a shortlist of surrogates that may not require significant research or innovation and are available for commercial application at full scale now and
- Demonstration and evaluation of a novel approach that utilizes the data from conductivity profiling of RO arrays to calculate a diffusion adjusted LRV (herein also referred to as "profile LRV" or "LRV_{defect}") consistent with the defect flow dilution approach established in the MFGM.

The direct removal of native markers including sulfate, strontium, sucralose, phosphate, and magnesium, as well as a spiked fluorescent marker, uranine, were shortlisted from literature review and investigated during this project. The integrity monitoring approaches were evaluated at three full-scale sites, Orange County Water District (OCWD), Water Corporation of Western Australia (WCWA), and Yucaipa Valley Water District (YVWD) and supplemented with systematic pilot-scale investigations conducted by Southern Nevada Water Authority (SNWA) to quantify the maximum sensitivity of high-pressure membranes under intact and oxidized conditions.

ES.2 Automated Conductivity Profiling as a Basis for RO LRV Verification

Conductivity profiling involves testing the permeate specific to an RO pressure vessel, which is the blend of permeate from all membrane elements within that vessel and doing this for all of the pressure vessels within the particular RO unit or system. Conductivity probing involves sampling the permeate conductivity at set lengths along a pressure vessel and can isolate damage to individual membrane elements. Procedures for both profiling and probing are well established as related to RO troubleshooting and maintenance and are described in detail elsewhere (AWWA 2007). A more recent innovation is an approach to use conductivity profiling as a means to calculate the LRV of an RO array and was described in previous peer reviewed literature (Vickers 2018). The basis for this approach is described briefly here. Slower diffusion of dissolved ions relative to water molecules across intact RO membranes results in their apparent rejection from feed to permeate. The conductivity profiling methodology utilizes the dilution of defect flow models established and used for pressure decay testing in the MFGM. To establish the defect flow contribution, a statistical approach was developed whereby the

median permeate conductivity of a stage was assumed to represent the nominal rejection due to diffusion. In a normal system, a small proportion of vessels may feature defects and would produce a conductivity higher than the median. If a significant number of defects are present, the average conductivity would then be pushed to higher values than the median. The difference between the average and median conductivity of a stage can then be used to represent the difference between nominal and defect flow. A maximum sensitivity of 5.0-log reduction is assigned corresponding with the lower range of MS2 challenge LRVs reported for intact RO systems. The larger the difference between the average and median conductivities, the more the LRV reduces from its maximum of 5.0-log. As well as establishing this calculation methodology for diffusion adjusted LRV determination, a number of potential alarm setpoints based either on number of standard deviations or absolute distance from the median have been proposed as a means to interpret the conductivity profiling dataset and were evaluated in this study.

Conductivity profiling was evaluated at YVWD where an automated system is installed and has been operating since May 2019. A number of intact and damaged conditions were evaluated both with direct conductivity, sulfate, and uranine removal as well as MS2 challenge testing. Based on the five MS2 bacteriophage integrity tests that were performed on the YVWD RO system, it was demonstrated that the RO unit attained at least 5.1-log of MS2 bacteriophage reduction. When comparing sulfate to bulk conductivity, the RO unit attained 2.8-log removal of sulfate compared to 1.3-log removal of conductivity. This difference in sulfate LRVs to conductivity was due to the fact that reduction of monovalent ions in RO is lower than that of divalent ions. Therefore, conductivity is easily measured but it creates a disadvantage because it uses the smallest entities possible and uses them as proxy for enteric viruses which are much larger. Even in the case of sulfate or uranine, in which the RO system attained 2.8-log and 3.5-log removals, respectively, these values do not approach the actual expected reduction of viruses nor the at least 5.1-log removal observed for MS2 in the challenge testing. Automated conductivity profiling was typically able to demonstrate LRVs higher than 3.0-log at YVWD and did not require spiking or offsite analysis of constituents. Fundamentally, the gap in LRV between the above surrogates and actual virus is caused by comparing reduction of viruses with constituents that are of a different nature.

The application of conductivity profiling to calculate an LRV necessitates arrays with at least six vessels in the final stage. As a consequence, conductivity profiling for the purposes of LRV calculation may only be suitable at sites with capacities on the order of 0.5 - 1.0 million gallons per day (mgd) or higher. Further work is needed to validate the technique for evaluation of NF integrity. Notwithstanding the limitations of minimum system size, the automated conductivity profiling at YVWD would meet all MFGM criteria for a DIT and demonstrate LRVs, depending on array integrity, between 3.0 to 5.0-log.

ES.3 Conductivity and Uranine Profiling at Full Scale

At OCWD, conductivity profiling was measured manually (i.e., no installation of an automated conductivity profiling system as done for YVWD) and profiling at a vessel level was also performed with uranine. The statistical analysis method applied to weekly RO permeate conductivity profiles (over six months) from the full-scale 5-mgd RO unit demonstrated 3.6-log removal on average. This was a significant increase to potential RO pathogen credit based on direct LRV for electrical conductivity (EC) (bulk permeate compared to feed) of 1.9 to 2.0-log for the same dates, as well as the typical LRV for RO credited at OCWD based on TOC of approximately 2.2-log based on daily average TOC LRV. All observed LRVs calculated from EC profile results were above 3.2-log and ranged between 3.2 and 4.7-log. Here we note for reference that a ~0.5-log difference in removal (when it's above 3-log) is a *very* small difference in terms of percent removal, i.e., when transformed back to percent removal, a range of 3.2 to 4 4.7-log corresponds to a range of 99.937% (3.2-log) to 99.997% (4.5-log) removal which is just a 0.06% difference. Thus, an observation of 4.5-log compared to 3.2-log is actually not likely to be significantly different and is probably within the 5% error of the analytical accuracy of the EC instrument. Thus, for log crediting (regulatory) purposes, some consideration should be paid to uncertainty analysis to appropriately round measured log removal to a conservatively credited value.

At OCWD, uranine spike tests in the same full-scale OCWD RO unit resulted in LRVs, calculated from profile data, up to 3.5-log. Given the similarity in profile LRV for uranine, which necessitated spiking, to conductivity, there was little demonstrated advantage to its use at scale. Sulfate and strontium concentrations were measured at the RO system level (i.e., bulk, or combined RO permeate) and stage level. The observed removals for strontium and sulfate were similar for the two test dates, 2.9 and 2.92 LRVs for sulfate and 3.13 and 3.15 for strontium. Future work could perform complete RO system profiling of sulfate and strontium (similar to the EC and uranine profiling that was conducted) in order to determine the impact of diffusion versus defects on calculated LRV to understand whether LRV may be significantly increased. This would require measuring sulfate or strontium in all vessels for a given RO unit (e.g., 150 vessels in the case of a single 5-mgd RO unit at OCWD advanced water purification facility [AWPF]), i.e., sulfate/strontium profiling, which may be cost-prohibitive.

ES.4 Long Term Surrogate Monitoring

At WCWA, an extensive sampling campaign for two full-scale RO skids was conducted over a 6-month period. From the surrogates analyzed, strontium and magnesium were not present at sufficient concentration in the RO feed and did not have low enough reporting limits to demonstrate improved LRVs. Sulfate and phosphate were able to demonstrate LRVs of 2.6 to >3.1 and 3.0 to >3.2-log, respectively. Phosphate was detected in almost half the permeate samples, whereas sulfate was only detected in approximately 30% of the samples.

The LRV calculated from conductivity profiling at WCWA suggested that the RO membranes which had been in operation for 5-6 years had maintained very good integrity, as most of the results were capped at 5-log. The conductivity profile calculation approach for diffusion-adjusted LRV was capped at 5-log due to the very small standard deviations and difference between average and median permeate conductivities of the arrays. However, there were events where the diffusion adjusted LRV (i.e., conductivity profiling LRV) reduced to 3.4-log. Surrogate result reproducibility was high when comparing the WCWA results to OCWD and YVWD where the same markers were measured. In addition, the sensitivity demonstrated for sulfate was very similar to results from OCWD and YVWD. Strontium results did not compare well to other sites due to its low abundance in the RO feed water at WCWA. Uranine was evaluated at pilot scale and was able to demonstrate steady state LRVs between 3.6 to 3.8-log, close to the 3.5-log reported at OCWD and YVWD.

ES.5 Pilot Testing to Determine Maximum Marker Sensitivity

At all full-scale sites, the sensitivity of native markers had the potential to be limited either by high method reporting limits in RO permeate or limited abundance in feedwater. In an effort to address this shortfall, systematic pilot testing with spiking of all markers was conducted at SNWA.

At SNWA, a number of different membrane products were tested under various hydraulic conditions to represent different locations in a full-scale RO array. In addition, the membranes were subjected to intentional oxidation using sodium hypochlorite to compare their rejection before and after damage of the selective polyamide layer. The maximum observed LRV for the Hydranautics ESPA2-LD-4040 and Toray TMG10D new membrane tests for sulfate, sucralose, uranine, and conductivity were very comparable (with sulfate and uranine consistently the highest of the four parameters), 3.1–3.5-log for the lead tests, and 2.7–2.9-log for the tail tests. While having lower LRVs, the CSM NE4040-40 and Filmtec BW30XFRLE 4040 exhibited similar trends across all parameters and sulfate and uranine LRVs were consistently higher than other tested surrogates. The oxidized membranes demonstrated similar MS2 rejection pre and post oxidation, demonstrating that the underlying support layer can act as an effective virus removal barrier even if the polyamide layer is impaired.

It is important to note that the BW30XFRLE 4040 membrane may not be the same membrane used for full-scale applications as the 4-inch element was custom made and issues with membrane operation stabilization were experienced. However, consistent data was observed for the randomized duplicate tests throughout the project.

Another key finding of the SNWA testing was the inability to demonstrate LRVs of 4 or greater for the tested molecular markers due to diffusion limitations. Each molecular marker was dosed and tested so that a 4-log removal could be demonstrated by the analytical instruments. LRVs calculated by EC (nominal diffusion), sulfate, conductivity, sucralose, and uranine were unable to reach LRVs of 4-log removal due to limitations in the background concentration and/or method detection limit. It can also be concluded that other similar compounds would be expected to be unable to reach LRVs of 4.0-log. MS2 challenge testing confirmed that most intact and oxidized membrane products achieved virus LRVs of more than 5.0-log.

ES.6 Proposed Approach to RO Crediting

In light of the above summarized results from the different sites, system scales, and surrogates evaluated, a potential approach suitable for regulatory framework where different RO LRV credit can be sought based upon choice of surrogate was proposed as follows:

- Tier 1 - an LRV of 1.0 - 2.0 is granted based on the direct LRV of conductivity (and/or total organic carbon) from bulk feed to bulk permeate:
  - This approach is conservative with respect to the actual level of virus removal anticipated across an RO system.
- Tier 2 - an LRV of 2.0 - 3.0 is granted based on daily measurement and analysis of results of surrogates such as sulfate, strontium, or phosphate across an RO array:
  - In order to be consistent with the MFGM, limits for conductivity reduction or absolute permeate conductivity would need to be established as a continuous indirect integrity monitoring (CIIM)to verify performance between surrogate sampling events and enable a response to gross membrane integrity failures.
  - Daily measurement and analysis of results will either require grab sampling and an in-house lab or investment in multiplexing online surrogate meters that have sufficiently low detection limits to demonstrate the proposed LRV.
- Tier 3 - an LRV of 3.0 - 5.0 is granted based on the LRV calculated from conductivity profiling:
  - Limits for conductivity reduction or absolute permeate conductivity would need to be established as a CIIM to verify performance between conductivity profiles.
  - Use of an automated profiler is recommended and must be capable of testing all operating skids at least once per day.
  - Periodic (i.e., monthly) grab samples of a selected surrogate are recommended to ensure that LRV always remains above 3.0.
  - Further research may be needed on interpretation of conductivity profile results and conversion to an LRV for full scale nanofiltration arrays. At this time, and with the available information, Tier 3 is proposed to only apply to RO membranes.

The recommended requirements for a Tiered approach to RO crediting are summarized in Table ES-1 below.

Table ES-1. Summary of Options for Crediting RO.

Parameter	Tier 1	Tier 2	Tier 3
Validation Type	1:1 pathogen relationship between direct conductivity LRV (Combined Feed to Combined Permeate) and pathogen LRV.	LRV of surrogates (e.g SO_4^{2-}, Sr) measured in RO Feed and Combined Permeate of each array adopted for pathogen LRV.	LRV calculated from conductivity profiling each vessel in each array adopted for pathogen LRV.
Anticipated LRV Range	1.0 – 2.0	2.0 – 3.0	3.0 – 5.0
Requirements	Continuous online measurements of Conductivity. Optional online monitoring of TOC LRV for the purpose of enhancing credit in low conductivity waters.	Enhancement of in-house laboratories and/or addition of complex online instrumentation for RO arrays. Must verify surrogate LRV daily for each array. Indirect continuous conductivity monitoring to detect and respond to gross failures.	Daily conductivity profiling every day for each array. Indirect continuous conductivity monitoring to detect and respond to gross failures. Periodic checks with Tier 2 surrogates recommended as a secondary verification. May not be suitable for NF or for very small systems (<<1 mgd)

Tiers 1 and 2 are already accepted in California, with direct conductivity and TOC monitoring for LRV compliance, and strontium is now approved at other sites. Tier 3, use of conductivity profiling, is yet to gain acceptance but should be considered in light of the results of this report. In some other states outside California, the MFGM is strictly adhered to and, as a result, RO systems are typically not credited for virus LRV. The most realistic approach to achieve widespread acceptance of a framework for RO crediting would be to revise and amend the USEPA MFGM. Amendments could include details on new approaches to verify virus removal of RO and clarification on requirements that have potentially been misinterpreted as a basis to avoid granting RO credit. However, to trigger and enact such an effort may not occur for some time.

ES.7 General Project Conclusions

Other overall conclusions from the project are summarized as follows:

- Findings for the measurement of overall removal performance (feed to permeate) indicated that the use of sulfate as a surrogate indicator (nominally 2.8-log) appears to allow for higher LRVs compared to EC (nominally 1.3-1.5-log) due to EC encompassing ions that have lower rejection rates than sulfate. As evidenced from data available from operating facilities, sulfate removal is higher than TOC removal. Thus, higher LRVs are obtainable if sulfate is used as a surrogate indicator. Based on the study results, strontium is expected to provide similar direct LRV as sulfate or even slightly higher (e.g., per OCWD results), assuming it is present at a sufficient concentration in the RO feed.

- Fluorescent compounds, such as uranine, provide a higher level of demonstrated removal (nominally 3.5-log). From a practical perspective, the use of fluorescent compounds poses operational challenges.
- The hypothesis that the membrane itself is not a source of virus passage appears to be consistent with the data. As described in the later in the report introduction, the composite structure (polyamide membrane on top of an ultrafiltration membrane) used for spiral wound NF/RO elements provides a multiple barrier approach for removal. Results of testing suggest the integrity issues arise from compromised sealing within the element or external O-rings. These types of issues would pass a significant amount of conductivity that could be proactively identified in operation with appropriate membrane element integrity testing methods. The salt rejection of the membrane element used should not affect virus reduction. It can be inferred from the testing of the NF elements that integrity for virus removal is not a function of the salt rejection characteristics of the membrane. Furthermore, membrane cleaning, which temporarily changes conductivity removal performance does not appear to substantially change virus removal.
- It is noted that the membranes tested at the YVWD were approximately seven years old. The membranes tested at WCWA were five to six years old. Loss of virus and surrogate rejection through membrane aging and cleaning does not appear to be substantial within typical RO membrane lifetimes at well operated facilities.
- Conductivity profiling alongside MS2 challenge testing demonstrated a correlation between high permeate conductivity vessels (outliers) and virus passage demonstrating that conductivity profiling is a useful diagnostic tool to determine potential integrity issues. It is logical to further investigate those vessels that are associated with the highest permeate conductivity as potential integrity issues. Sulfate, uranine, or other surrogate indicators of larger molecular weight can also be used for profiling; however, their implementation is more involved and costly and thus it may be preferred to reserve this effort for non-routine monitoring purposes. Data seems to be consistent with the literature that the defects (i.e., integrity compromises) have to be significant in size to drastically lower the LRV.

As the necessity to consider and implement potable reuse projects becomes more prevalent as water resources availability continues to diminish, there is a clear need for pragmatic RO and NF integrity monitoring techniques with a sensitivity that correlates and is equivalent to the results from MS2 challenge testing. This project has identified and proposes a tiered approach for surrogates that can achieve 3 to 5-log LRV, which would be an improvement over the current (and inconsistent) implementation of pathogen credits for RO and NF.

ES.8 Related WRF Research

- Guidance for Implementing Action Spectra Correction with Medium Pressure UV Disinfection (4376)
- Methods for Integrity Testing and Online Monitoring of NF and RO membranes (4757)
- New Techniques for Real-Time Monitoring of Membrane Integrity for Virus Removal (1663/1664)
- Demonstrating Virus Log Removal Credit for Wastewater Treatment and Reverse Osmosis for Potable Reuse at OCWD (5041)

CHAPTER 1

Introduction

High-pressure membranes, including RO and NF, are widely used in water treatment (e.g., groundwater, surface water, seawater) and water reuse (indirect and direct potable reuse) applications. Traditionally used as a key component in desalination processes, intact RO and NF membranes are capable of robust rejection of dissolved ions in water and are therefore sufficient to act as a significant barrier to pathogens such as viruses, *Cryptosporidium*, and *Giardia* (i.e., protozoa). The control of waterborne pathogens is of particular importance to the protection of human health, and thus is the focus of several regulations that impact treatment systems employing RO/NF, including:

- California Code of Regulations (Title 22) Groundwater Replenishment and Surface Water Augmentation Regulations - Requires at a minimum, 12-log virus, 10-log *Giardia,* and 10-log *Cryptosporidium* removal by reclaimed water systems used for indirect potable reuse (IPR). Each treatment process can be given up to 6-log removal credits (SWRCB 2018).
- USEPA LT2ESWTR - Requires 4-log removal of enteric viruses, 3-log *Giardia* removal, and up to 5.5-log *Cryptosporidium* removal by drinking water treatment systems depending on source water quality (USEPA 2007).
- USEPA Groundwater Rule - Requires 4-log removal of enteric viruses for groundwater sources determined susceptible to fecal contamination (USEPA 2006).

Currently, California and other U.S. states, (i.e., Colorado, Florida, Texas, New Mexico, and Arizona) are developing direct potable reuse regulations and/or guidelines with similarly pathogen LRV requirements. Because of the acute public health implications associated with pathogens, drinking water regulations for microfiltration (MF) and ultrafiltration (UF) membrane systems require performing both CIIM using turbidity and a once-daily DIT using PDT. Based on the USEPA's Membrane Filtration Guidance Manual (MFGM) (USEPA 2005), which was developed for use in conjunction with implementation of the Long Term 2 Enhanced Surface Water Treatment Rule (LT2ESWTR) for Cryptosporidium removal, a DIT must meet criteria of resolution (i.e., how small of a defect can be verified), sensitivity (i.e., what magnitude of LRV is able to be demonstrated), and frequency (i.e., can the technique verify integrity and allow corrective action at least daily).

Based on the size of pathogens and the molecular weight cutoff values for RO and NF membranes (Figure 1-1), both RO and NF should consistently achieve significant log-removal credits for virus and protozoa. However, meeting the MFGM integrity monitoring requirements and demonstrating more than 2-log removal has proven a challenge. In order to satisfy the MFGM, chemical markers that are "discretely quantifiable" or of a known molecular weight are preferred. Typical markers used for water quality characterization are orders of magnitude smaller than the targeted pathogen and will diffuse across the RO and NF membranes under normal operating conditions, thereby reducing sensitivity. One difficulty has been identifying native markers or surrogates that have the resolution and sensitivity necessary to demonstrate

an LRV value greater than 2-log in a high-pressure membrane system's feed and permeate due to a variety of reasons (e.g., analytical measurement limitations, cost of implementation, lower than ideal rejection, non-detects in the permeate, etc.). In addition, high-pressure membrane systems can be compromised or breached through a variety of mechanisms such as damaged O-rings, glue line leaks, and particulate damage. These breaches may be sufficient to allow pathogens to pass through the membrane system and into the permeate (Jacangelo et al. 2019; Jacangelo and Gray 2015).

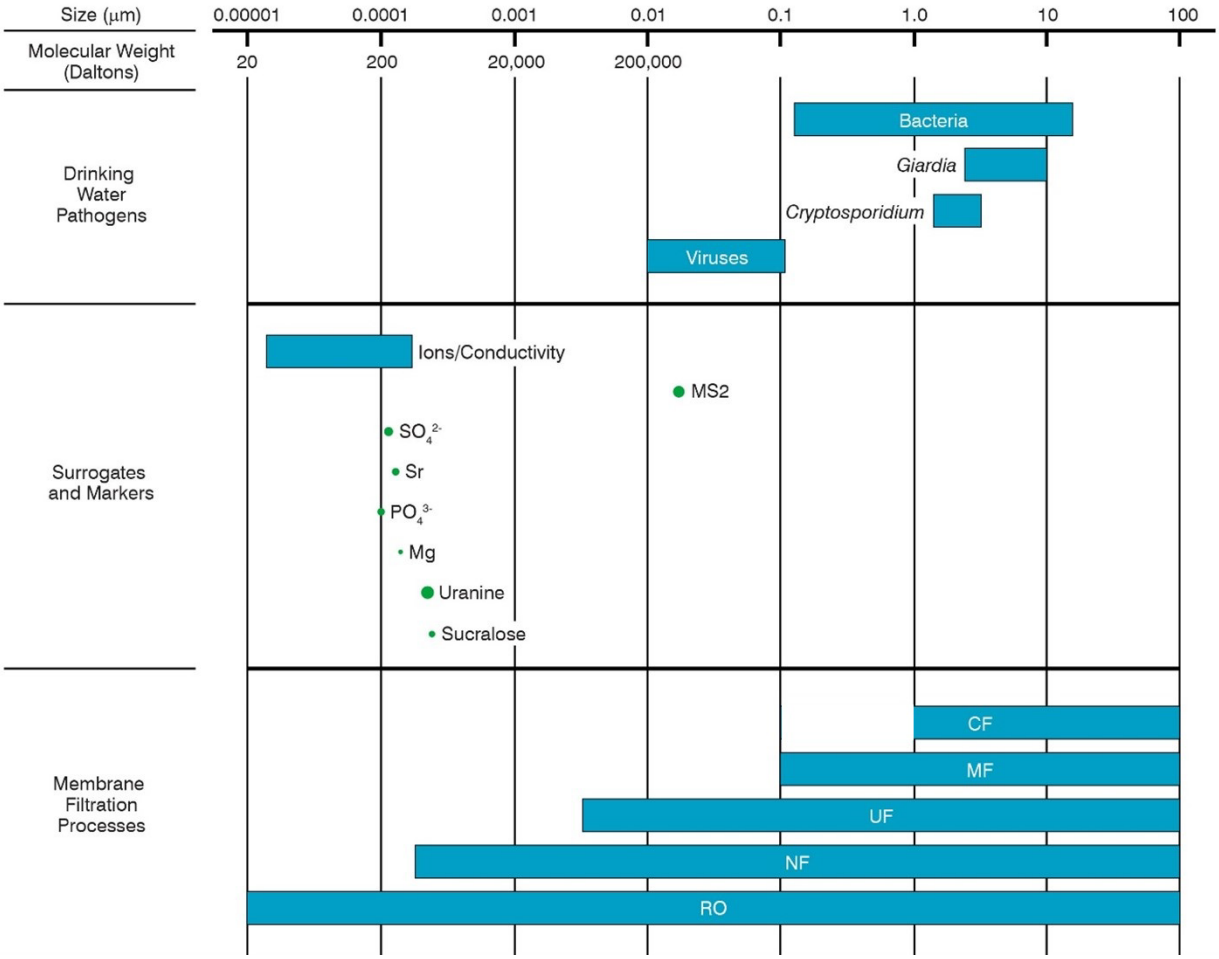

Figure 1-1. Modified Filtration Application Guide for Pathogen Removal Indicating Size of Surrogates Evaluated in This Study.

For RO, LRVs of 1.5-2.0-log for viruses, *Giardia*, and *Cryptosporidium* are typically credited based on the LRV of conductivity and/or TOC from the bulk RO feed to the RO permeate (Vickers et al. 2019). However, during spiked challenge testing using MS2 bacteriophage, an MFGM approved virus surrogate, removals of greater than 6-log at the bench scale (Hornstra et al. 2019; Kitis et al. 2003a; Pype et al. 2016a; Steinle-Darling et al. 2016) and greater than 5-log (this study and previously by Vickers et al. 2019), have been observed.

Findings from past research on NF and RO membranes indicate that identifying indigenous molecular markers that can meet the USEPA MFGM's requirements for resolution and sensitivity in a DIT method to demonstrate removal of 3-logs or higher is challenging. Note, native parameters such as TOC/DOC, UV_{254}, and conductivity, which are approved for potable reuse projects in California, do not meet the USEPA MFGM's requirements for a DIT's resolution since the size of these parameters are not uniform and therefore not comparable to the size of viruses or pathogens. In addition, integrity methods that may not reside within guidance but are historically associated with the operation of RO systems such as conductivity profiling were investigated.

1.1 RO and NF Membrane Characteristics

As shown in Figure 1-2, RO membranes are made with a thin-film polyamide layer, an underlying polysulfone ultrafiltration layer, and a polyester non-woven backing material which provide support for the membrane (AWWA 2007). RO membranes do not have pores in the conventional sense, as solutes pass through interstitial spaces between molecular structures based upon molecular weight and charge, which are controlled by principles of diffusion. Pathogen reduction by size exclusion is theoretically infinite; however, differences in manufacturing (i.e., glue lines, O-rings) or plumbing defects may limit the overall reduction performance of the RO membrane element.

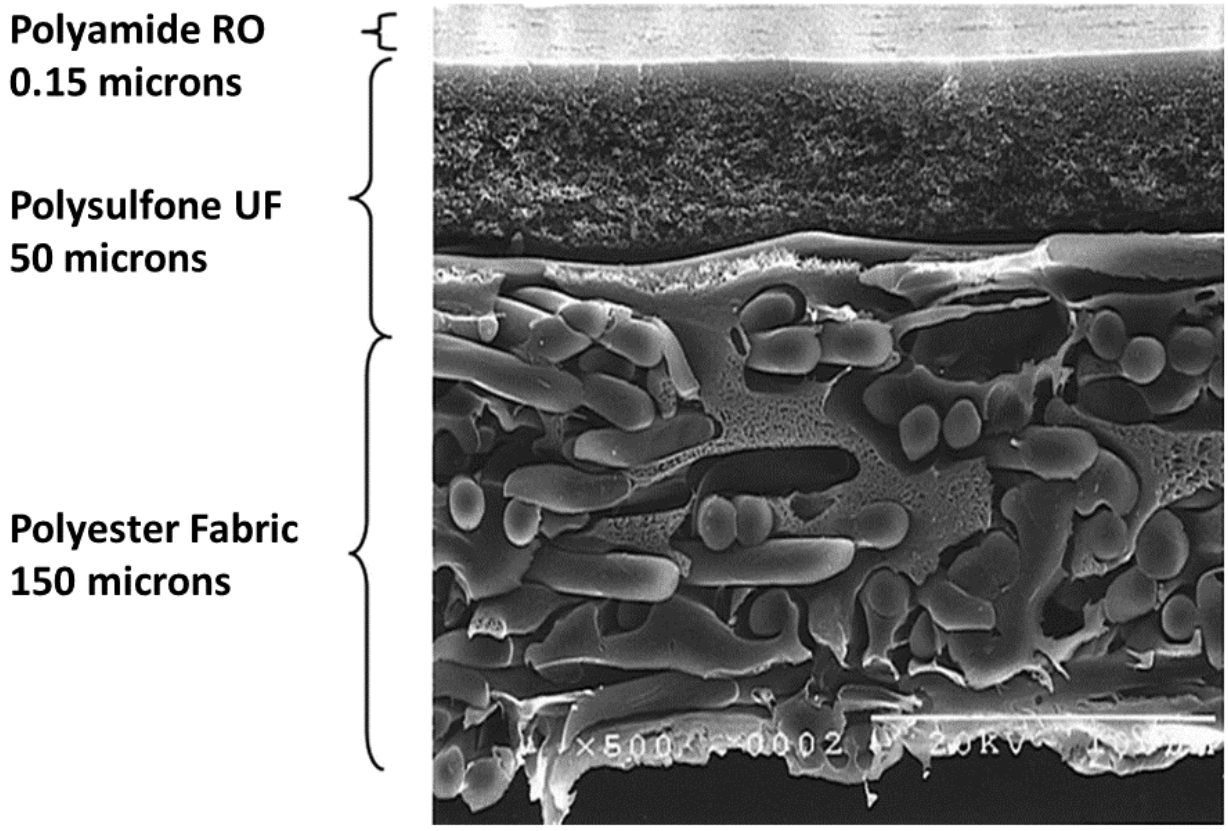

Figure 1-2. Reverse Osmosis Membrane Cross Section.
(Photo Courtesy of Hydranautics).

Studies have shown that commercial RO membranes can achieve greater than 6-log removal of MS2 bacteriophage, a commonly accepted surrogate for enteric virus (DeCarolis et al. 2006; Adham et al. 1997; Jacangelo and Gray 2015; Jacangelo et al. 2019). Lower LRVs of 3.0 to 4.0-log, have also been observed during MS2 challenge testing for demonstration scale RO systems that were believed to have nominal integrity (Carollo Engineers, Inc. 2016). Virus challenge tests with an oxidized RO membrane suggest that reduction of viruses is possible and defects in the form of O-ring failure are most likely the mechanism of significant integrity loss (Steinle-Darling et al. 2016). Removal of O-rings during challenge testing with MS2 reported a reduction of LRV from 4.0-log to 2.0-log and a corresponding change in conductivity LRV from 1.5-log to approximately 1.0-log (Carollo Engineers, Inc. 2016). Virus challenges with an oxidized RO membrane also suggested that oxidation of the RO membrane does not appear to compromise the virus rejection ((Steinle-Darling et al. 2016). An oxidized membrane might retain its virus rejection because the polyamide structure "loosened" enough for more salts to diffuse, but not enough for viruses to pass through.

A polyamide membrane could become oxidized as a result of exposure to free chlorine or a high pH solution (during a clean-in-place [CIP]). The reduction characteristics of RO membranes is usually reflected as a higher electrical conductivity in the permeate water immediately after a CIP. It has also been observed that the change in reduction performance after a CIP is temporary, and that membrane rejection stabilizes once the membrane reaches equilibrium after operating under typical feedwater conditions for several days. Similar to the oxidized condition, RO membranes after a CIP will produce permeate of higher electrical conductivity because salt passage is facilitated by the RO membrane being temporarily altered (Ying et al. 2014). Even though a higher salt passage exists in a Post-CIP membrane due to "loosening" of the membrane, virus passage is still limited because viruses are orders of magnitude larger than salts. This behavior is also similar to experiencing higher salts/TOC concentration in the permeate caused by higher feed temperatures - where an increase in virus passage is not expected to occur.Therefore, in order to establish a safe LRV, it is necessary to demonstrate the virus reduction performance after a CIP, oxidation, and with a damaged interconnector to confirm that the virus reduction is not changed as a result of these conditions.

MS2 bacteriophage is a well-established challenge testing organism that functions as a surrogate for enteric viruses because of their size similarity (~25-28 nanometers [nm]) (USEPA 2005). Therefore, testing with MS2 can verify the resolution and sensitivity required to attain the appropriate LRV for enteric viruses. Since viruses are smaller than other pathogens, the suitable LRVs established for virus could also be applied for other pathogenic organisms (e.g., protozoa) regulated in drinking water (either from traditional or potable reuse sources). This project utilized MS2 challenge testing in conjuction other surrogate indicators with different molecular weights to demonstrate performance in a variety of operating conditions.

Although electrical conductivity may be useful as an indicator, this parameter includes all ions irrespective of their size. Monovalent or divalent ions are orders of magnitude smaller than an enteric virus, thus making the assumption of virus LRV based on electrical conductivity is conservative although as a bulk parameter, this has not been considered appropriate (or

allowable) in some states based on the interpretation of current regulatory guidance (i.e., the MFGM). Strontium, sulfate, and other native indicators can demonstrate RO reduction of divalent ions. These indicators, coupled with other parameter testing provides a benchmark for what is expected with low molecular weight compounds to cover a range of conditions that will support the establishment of practical guindance for virus reduction credit.

1.2 Quantifying Performance

The apparent rejection of a membrane system is calculated using Equation 1-1:

$$R = 1 - \frac{C_p}{C_f}\ x\ 100\% \qquad \text{(Equation 1-1)}$$

Where:

R = Rejection (%).

C_p = Concentration in the RO combined permeate, (plaque forming unit [PFU]/milliliter [mL], milligrams per liter [mg/L], or parts per billion [ppb]).

C_f = Concentration in the RO feed, (PFU/mL, mg/L, or ppb).

Performance of pathogen removal by membrane filtering systems can be calculated by the logarithmic ratio of the concentration in the feed to the concentration in the *combined* permeate (i.e., the comingled RO permeate from all pressure vessels from both stages), as shown in Equation (1-2), which is used to calculate the LRV of a unit.

$$LRV = \log C_f - \log C_p \qquad \text{(Equation 1-2)}$$

Where:

LRV = Log Reduction Value (dimensionless).

Equation 1-2 could refer to: (1) MS2 bacteriophage (PFU/mL), (2) specific compounds such as sulfate (mg/L) or uranine (ppb), or (3) a bulk water quality parameter such as conductivity (microsiemens per centimeter [µS/cm]) or TOC (mg/L).

Although LRVs are by definition assigned to an RO unit rather than a stage or vessel, this study also quantified the MS2 reduction of specific pressure vessels. For those instances, C_f continues to be the feed to the first stage, but the C_p term is the permeate produced by a particular pressure vessel. The traditional measurement of removal for compliance reporting is from the bulk feed to combined permeate. For the purposes of this study, additional data was collected to characterize the removal performance at intermediate locations (i.e., first stage permeate, interstage feed, second stage permeate), as well as from selected pressure vessels to provide a better understanding and characterization of the removal performance across the system.

1.3 MFGM and Challenges with Applying the MFGM to RO and NF

Although the MFGM provides the fundamental guidance by which state regulators have considered assigning LRV credits for high-pressure membrane systems, the MFGM also poses several challenges for RO and NF applications. The MFGM was written nearly 20 years ago and has not undergone any revisions or amendments since it was first published. The MFGM was written with the primary intent of guiding the verification of the performance of MF/UF membranes to assure *Cryptosporidium* removal of greater than 3.0-log. The MFGM was not designed as a tool to regulate virus removal by UF (which is scientifically proven but not credited in most locations) or virus and protozoa removal by NF or RO (which, as previously mentioned, is also proven). Although the MFGM can be applied to RO and NF, a good question may be "should the MFGM be applied to RO and NF?" There are a number of practical issues with respect to rejection mechanisms, system configurations, and operations that pose challenges to directly applying the MFGM as guidance.

Relevant sections of the MFGM are presented in Table 1-1.

Table 1-1. MFGM Requirements for LRV Certification and DITs.

Requirement	Relevance	MFGM Section
Product-specific Certification	Establishes stable quality control removal value (QCRV) for a membrane product which is then demonstrated by DITs.	1.3
Challenge Testing	• Limits monitoring for surrogates that are mass based and discretely quantifiable to the size of protozoa or enteric virus for which LRVs are credited. • Prohibits use of 'bulk parameters' such as conductivity and TOC.	1.3 3.2 3.4 3.6 3.7 3.13
Pressure-Based DITs	• Air and vacuum pressure tests are not practicable for RO/NF systems. • 'Dilution model' presented as part of Pressure Based Tests permits users to remove the influence of air diffusion from DIT calculations, this approach can be adapted to conductivity in a non-pressure-based testing approach.	4.2.1 4.3.1
Marker-Based DITs	• *Resolution* and *sensitivity* require the use of 'discretely quantifiable' markers, such as particles or molecules, that can be demonstrated to have a size less than the size of protozoa or enteric virus for which LRVs are credited. • Does not discuss making provisions for an allowance to account for the influence of diffusion when evaluating DIT results.	4.2.2 4.3.2

As the fundamental basis for regulation of membrane systems, requiring state regulators to interpret the MFGM language to grant approval of LRVs for RO/NF results in significant differences in regulatory approval and may not be the proper use of the MFGM. Evolving potable reuse regulations require the water industry to adapt existing tools and develop new strategies to achieve the very high LRVs (particularly for direct potable reuse [DPR]) required to protect public health. In the absence of a federal revision to the MFGM or a supplemental document, states will continue to develop their own analysis and conclusions related to pathogen removal by NF and RO.

In practice, the direct applicability of the MFGM for use in an IPR/DPR treatment system involves consideration of fundamental differences between removal credits and the manner in which they can be obtained. For example, in drinking water disinfection, chlorine or chloramine can be used to obtain 4-log virus removal. In the California IPR/DPR scenario, the virus removal required is significantly higher and necessitates multiple processes and the maximum individual log removal that can be credited to a single treatment technique is limited to 6-log.

1.3.1 Product-Specific Certification

MF and UF membranes are challenge tested to establish a QCRV that is tied to a pressure-based test (i.e., air pressure decay) result and a marker-based challenge test result. This testing is performed as part of the product-specific certification process required by the MFGM. Its purpose is to isolate the impact of any manufacturing defects so that, when applied, the pressure-based DIT used is evaluating only the impacts of breaches that may pass protozoa.

In drinking water applications, product-specific challenge testing is required for membrane products to benchmark log-removal (MFGM Section 1.3 and National Science Foundation [NSF] 419). While MF and UF manufacturers have embraced and followed through with this testing, NF and RO manufacturers have not. Lack of product specific testing has not been uniformly enforced by State regulatory agencies. Without appropriate product-specific benchmarking, the effect of membrane age on DIT method resolution and sensitivity is uncertain when using molecular tracers or other chemical markers (i.e., LRV credit cannot be confirmed). Even for DIT methods that are intended only for defect detection (e.g., 'bubble test' for MF/UF or a 'dilution model' based statistical analysis of conductivity profiling data for RO/NF), a benchmark for product-specific performance should be required to establish the range of acceptable operating conditions (i.e., testing pressure or conductivity, respectively).

1.3.2 The 'Dilution Model' Approach

This approach is only referenced in the Pressure Based Test sections of the MFGM (Section 4.2.1 and 4.3.1 [USEPA 2005]). The dilution model allows MF and UF membrane users to isolate the loss of air pressure due to diffusion from those of a 'discretely quantifiable' defect (e.g., air flow through an orifice with a size ≥ 3-micron). If we wish to isolate the diffusion of another parameter, like conductivity, in an alternative DIT method (or marker-based DIT method), it will require an understanding and application of diffusion science. As demonstrated by the previous and current work, the LRV of electrical conductivity can be increased from between 1.5 to 2-log removal to between 3.0 to 4.5-log by removing the influence of diffusion to isolate the conductivity associated with possible membrane defects in a challenge test or DIT. Isolating the difference between "defects" versus "diffusion" or nominal rejection of these constituents is permitted by the MFGM.

1.3.3 Bulk Water Quality Parameters

The MFGM requires CIIM for which turbidity is used as the indicator for membrane processes that are used for obtaining *Cryptosporidium* removal of 3.0-log or higher. In drinking water, the limits for turbidity were established at less than 0.15 Nephelometric Turbidity Units (NTU) for two consecutive readings 15 minutes apart.

Bulk parameters such as conductivity (and TOC or turbidity), are not recognized by the MFGM as being acceptable for use in challenge tests because they are not based upon mass per unit volume that is quantifiable to a 'discrete size' of particle smaller than the protozoan or virus for which LRV is to be credited (refer to *MFGM* Sections 3.2 and 3.9). Alternative methods (e.g., electrical conductivity or TOC) that would be more applicable for monitoring of NF or RO permeate on a continuous basis "must be approved by the State" (page 5-2 of the MFGM). However, some State regulators interpret the MFGM such that conductivity is also not acceptable for use in a DIT, which is contrary to the MFGM's stated intent.

1.4 Project Goals

The goal of this project is to identify methods and procedures for long-term validation testing to demonstrate enteric viruses (as well as *Giardia* and *Cryptosporidium*) LRV credit by RO and NF membrane systems. Long term testing of DIT methods conducted at both pilot-scale and full-scale focused on answering key questions to support alternative regulatory consideration. At the onset of this project, we recognized the regulatory obstacles that these issues pose, and the study was structured to address these challenges in a manner that supports a pathway for future regulatory acceptance. Our approach considered how to achieve lower cost and simple to use testing methods that will achieve greater LRV credit than would otherwise be possible.

The regulatory obstacles pertaining to the need for/value of product-specific certification, using bulk water quality parameters (i.e., conductivity), and the influence of diffusion are the fundamental goal of this project work. The results of the research on RO and NF DIT methods demonstrate that it is possible to develop a marker-based testing method that address the practical and regulatory hurdles associated with increased LRV credit.

1.4.1 Test Facilities

A fundamental component of this project focused on testing of various selected methods at 4 participating utilities:

- Orange County Water District (OCWD),
- Yucaipa Valley Water District (YVWD),
- Water Corporation of Western Australia (Water Corporation), and
- Southern Nevada Water Authority (SNWA).

1.5 Report Contents

This report is organized into the chapters below:

- Chapter 2 - Literature Review
  - Contains a focused literature review of the integrity monitoring approaches investigated during this project and notes other recent RO integrity monitoring projects.
- Chapter 3 - Testing and Calculation Methods
  - Briefly summarizes common standard test methods, method reporting limits, and the test schedule from the field investigations from this project.
- Chapter 4 – Conductivity Profiling to Credit RO
  - Describes the RO vessel profiling approach and how it could be applied as a DIT per the MFGM.
- Chapter 5 – Orange County Water District
  - Describes the full-scale field investigations at OCWD including profiling with both conductivity and uranine, as well as sulfate, strontium, and TOC monitoring.
- Chapter 6 - Yucaipa Valley Water District
  - Describes the full-scale field investigations at YVWD including profiling with both conductivity and uranine, as well as sulfate and MS2 challenge testing. The MS2 challenge testing and response of conductivity profiling was conducted under nominal and impaired conditions.
- Chapter 7 - Water Corporation of Western Australia
  - Contains the results of a six-month monitoring campaign where inorganic markers including sulfate and strontium, among others, were monitored for two arrays at the Beenyup GWR Scheme. Automated conductivity profiling results and conventional monitoring were also summarized for the same period.
- Chapter 8 - Southern Nevada Water Authority
  - Systematic experiments were conducted on a single element pilot rig to investigate the maximum sensitivity of sulfate, strontium, and sucralose on new and intentionally damaged membranes.
- Chapter 9 - Approaches for Crediting RO and NF
  - This chapter combines the results from all field investigations to evaluate surrogates against MFGM and general monitoring criteria. In addition, perspectives from regulator workshops are summarized. Practical recommendations as to the use of the surrogates evaluated in this project are made.
- Chapter 10 - Conclusions and Recommendations
  - Contains the project level conclusions and recommendations from this work.

CHAPTER 2

Literature Review

This literature review summarizes the previous studies and research pertaining to integrity testing regulatory framework and development of approaches for ensuring membrane system integrity, and ultimately claiming LRV for high-pressure membrane systems greater than 2-log and ideally equal to or greater than 4-log. It is worth noting that several reviews on high pressure membrane integrity have recently been published (Jacangelo et al. 2019; Kitis et al. 2003a; Niewersch et al. 2020; Yoon 2019). In particular, a recently conducted literature review provided a comprehensive list of high-pressure membrane integrity approaches (Jacangelo et al. 2019).

2.1 Integrity Testing Regulatory Framework

LT2ESWTR was introduced in 2006 to provide additional requirements for drinking water systems to control *Cryptosporidium* and other pathogens in surface water sources (USEPA 2007). As part of the LT2ESWTR, the USEPA released the MFGM to provide guidance for regulators and municipalities to understand how membrane technologies fit into the LT2ESWTR framework, as well as establishing guidance regarding their use and implementation. The MFGM also adopted a means of communicating the removal efficiencies of membranes through the LRV.

The LRV that a membrane receives is the lower of two demonstrated values: (1) the removal efficiency demonstrated during challenge testing or (2) the maximum LRV that can be verified by a DIT used during the course of normal operation. Challenge testing was established to be product specific and is typically performed a single time by the membrane manufacturer in accordance with NSF/American National Standards Institute (ANSI) 419. Membrane manufacturers typically sponsor challenge testing of their products, which is then performed by a qualified third party such as NSF. DITs, on the other hand, must be applied to all physical elements of the entire membrane units, including all non-membrane components that could result in the contamination of the filtrate (i.e., membrane permeate) if compromised, on a frequency of once per day. Additionally, a membrane unit must have a method for CIIM which involves the monitoring of some aspect of the permeate water quality. The chosen water quality aspect serves as a surrogate to membrane integrity, as changes in permeate quality can indicate a compromise in membrane integrity (USEPA 2005). It is worth noting that the MFGM does not explicitly define acceptable DIT and CIIM approaches for high-pressure membranes (RO and NF), and, as a result, there is some ambiguity regarding whether a specific method constitutes a DIT versus a CIIM approach. The following sections define DITs and CIIMs in accordance with the MFGM. Additionally, many of the requirements set forth by the MFGM focus on MF and UF membranes as opposed to RO and NF membranes. Furthermore, consideration to the use of chloramines as a means to control biofouling is almost always employed at advanced treatment facilities and may impact some types of tests under consideration. The following discussions consider the challenges in applying these methods to high-pressure membrane systems.

2.1.1 Direct Integrity Testing

DIT is defined by the LT2ESWTR as a physical test applied to a membrane unit in order to identify and isolate integrity breaches. DIT is required to be completed on every membrane actively used at a frequency of at least once per day, except if State approval has been given for less frequent testing based on demonstrated process reliability, the use of multiple barriers against *Cryptosporidium*, or reliable process safeguards. The chosen DIT must have the resolution to detect an integrity breach relating to the removal credit given. This corresponds to less than 3 micrometers (µm) for *Cryptosporidium* and *Giardia*, and less than 0.01 µm for viruses. The test must also have the sensitivity to verify an LRV greater than or equal to the removal credit awarded to the entire membrane filtration process.

2.1.1.1 Pressure Based Testing

One of the most common DIT methods is PDT. The MFGM allows pressure-based direct integrity testing based upon a calculation of air flow through an orifice, the size of which can be equated to the pathogen size for which removal is sought. In the case of PDT, a test pressure that corresponds to a resolution of 3 µm is targeted. Air flow is equated to air pressure decay in these tests, and allowances for air diffusion through the intact membrane are also considered and incorporated into the test method. This allows isolation of "diffusion" related air pressure decay from the "defect" related pressure decay. These tests are common for their relatively low cost and acceptance by State and Federal regulators. However, the tests must be done offline and do not have the resolution required to detect virus-sized defects without causing damage to the membrane element. According to Table E.4 of the MFGM, the required test pressure to target virus sized pathogens, the smallest of which are 0.01 µm, would range from 768 to 4,344 pounds per square inch (psi) (USEPA 2005). The required test pressure to allow resolution of virus sized defects exceeds the pressure that a typical commercially available MF/UF membrane system can withstand without damage.

While PDT is a standard practice for assurance of protozoa removal by MF/UF, it has logistical challenges that make it difficult to implement on full-scale RO and NF. Specifically, the system must be drained in order for the results to be reproducible and the orifice (pressure or vacuum decay) calculation to work. Vacuum decay testing on the permeate size of RO membranes is possible, but there are concerns about the practicality of a vacuum decay test on large arrays and the potential for RO membrane damage due to routine draining and air entrapment in the permeate channels upon refill. Since it is challenging to completely drain horizontally oriented RO and NF systems, this form of testing is not practical to permit a system with NF/RO membranes to receive *Cryptosporidium* and *Giardia* removal credit if intended to be performed on a routine basis (Ostarcevic et al. 2018).

2.1.1.2 Marker-Based Tests

The second type of DIT presented in the MFGM is a marker-based test. In this test, a surrogate is periodically applied to the feed water in order to verify the membrane system integrity. In order to qualify for virus LRVs, the surrogate must be in the size range of 0.03 to 0.1 µm (i.e., resolution). An approved surrogate for viruses in the MFGM is the male-specific coliphage (MS2), with a size of 0.025 µm (USEPA 2005). In addition to being a similar size, MS2

bacteriophage also has a similar shape and composition to viruses, making it an attractive substitute (Ostarcevic et al. 2018).

Rejection of MS2 bacteriophage by RO has been widely studied, and LRVs of greater than five are typically achieved across intact membranes, with some reports of greater than seven (Kitis et al. 2003a; Ostarcevic et al. 2018; Trussell et al. 2017). Despite its wide use to characterize virus removal, there are practical limitations with respect to frequent use of MS2 bacteriophage at large facilities including (Gitis et al. 2002; Trussell et al. 2017):

- Production of a sufficient amount of MS2 to challenge test a full-scale facility is time-consuming, expensive, and logistically challenging.
- The assay turn-around time is at least 24 hours meaning that verification of daily performance is not possible.
- False positives have been reported as treated water sample contamination becomes more likely when handling concentrated stock solution.
- Although unfounded, as MS2 is not pathogenic and replicates by infecting *E. coli* and not humans, there are concerns about subsequent release of MS2 during challenge testing if diversion of water is not possible during the test.
- RO permeate alone may degrade MS2 over time resulting in erroneous results.

For these reasons MS2 testing is more often used for challenge testing purposes to compare other surrogates against, as opposed to daily integrity testing. However, the LT2ESWTR does not require the use of a particular DIT for rule compliance, but rather that any test used must meet the specified performance criteria for resolution, sensitivity, and frequency (USEPA 2005). Therefore, a significant amount of research has been undertaken to identify surrogate markers (e.g., molecular markers) that meet the requirements of the MFGM and result in appropriate LRV credits. These alternate surrogates are discussed in more detail herein.

2.1.1.3 Dilution or Imperfection LRV Calculations

Recently, several approaches have been developed to improve LRVs calculated from DIT and CIIM monitoring data. This concept is analogous to pressure decay test data evaluation where the rate of diffusion of air through an intact membrane is subtracted from measured air diffusion rates during DIT measurements (i.e., dilution approach, MFGM Section 4.3.1.3) (USEPA 2005). Following a similar approach, statistical analysis of conductivity profiling data was performed to calculate the permeate conductivity due to diffusion and the permeate conductivity due to a defect for LRV calculations (Vickers 2018). More information on this approach is provided in Chapter 4, but studies have demonstrated LRVs for RO greater than three using this approach (Vickers 2018; Vickers et al. 2019). A component of this project will be to explore whether this diffusion adjusted approach could be applied to molecular markers with greater sensitivity (e.g., fluorescent dyes) to achieve LRVs greater than two and, ideally, LRVs equal to four or higher.

Other researchers have applied the solution-diffusion-imperfection model as an approach for high-pressure membrane system DIT (Frenkel et al. 2014; Niewersch et al. 2020; Surawanvijit et al. 2015). Because the removal mechanism of dissolved constituents is governed by diffusion for RO and NF membranes, the rate of diffusion can be calculated based upon the

mineral composition in the feed water adjacent to the membrane surface and temperature. Accounting for the water flux through the membrane, the concentration of easy-to-measure constituents in RO or NF permeate water can be calculated. If the measured concentration matches or is less than the calculated value, integrity can be demonstrated in a manner equivalent to that used for a pressure-based DIT. With this methodology, the model is calibrated to intact membrane specimens and applied to permeate water quality obtained semi-continuously along with membrane system operating parameters. From this approach, solute specific model parameters (e.g., solute permeability and solute leakage factor) can be calculated and compared to values obtained from intact membrane specimens with deviations indicating an integrity issue. While studies have demonstrated the effectiveness of this approach, actual LRVs that could be obtained from this method have not yet been firmly established and regulatory approval has not yet been requested (Frenkel and Cohen 2018; Niewersch et al. 2020). To meet the individual membrane DIT requirements of the MFGM, a diffusion adjusted approach to LRV crediting would likely require profiling at a vessel level in a high-pressure membrane system.

Although it has particular relevance to RO and NF systems, profiling at a vessel level is a concept that requires additional explanation since it is not described in sufficient detail in the MFGM. The MFGM describes PDT or marker-based testing approaches as being applied at a single point or unit level. RO and NF systems are typically provided with a sample location at each vessel which can be used as part of a multiple point integrity test to enhance the information associated with any applied testing methodology.

2.1.2 Continuous Indirect Integrity Monitoring

In the MFGM, CIIM is defined as “monitoring some aspect of filtrate water quality as a surrogate measure of membrane integrity” (USEPA 2005). This definition focuses on low-pressure membrane monitoring, as indicated by the use of the term filtrate, as opposed to permeate for a high-pressure system. Although the methodology of CIIM does not have the same level of sensitivity and resolution constraints that a DIT method does, a marked decline in filtrate quality can serve as an indicator for membrane integrity problems. While low-pressure membrane filtrate quality is very consistent and is largely independent of fluctuations in feed water quality, the same is not true for high-pressure membranes. While fluctuations in a turbidity would be indicative of an integrity breach in a UF/MF membrane system, the same response to a defect cannot be guaranteed for a RO/NF membrane. For example, it is known that under warmer water conditions, RO permeate conductivity will be higher (for the same feedwater quality) due to accelerated mass transport of salts. CIIM must be separately conducted on each membrane unit at a frequency of at least once every 15 minutes. While LT2ESWTR and the MFGM don’t specify any criteria for resolution and sensitivity relating to CIIM, they do require the establishment of performance-based control limits. If readings fall outside these control limits for a period of longer than 15 minutes (two consecutive readings), a DIT must be done to assess a potential integrity problem. It should be noted that while CIIM is currently the standard, the MFGM has acknowledged that the development of a sufficiently sensitive and reliable continuous DIT is feasible, and a proven test of this caliber could preclude CIIM at a federal level, state requirements notwithstanding (USEPA 2005). Conductivity

monitoring of the permeate would qualify as CIIM for an RO system but is discussed in a subsequent section.

2.1.2.1 Turbidity

The MFGM states that unless there is a State-approved alternative parameter in use, filtrate turbidity monitoring must be implemented. Turbidity monitoring has well-defined control limits, with consecutive readings above 0.15 NTU indicating a loss in integrity and requiring direct integrity testing in MF/UF membranes. Despite these well-defined parameters, it has a low sensitivity to breaches that would allow the passage of virus particles (USEPA 2005). Even laser turbidimetry, capable of measuring much lower turbidity values than traditional turbidimeters, is limited to particles over approximately 1000 nm, ten times larger than even the largest virus particle (Ostarcevic et al. 2018). Additionally, while laser turbidity has been shown to be able to detect a single broken fiber on a UF membrane module with several thousand fibers (pore size ~0.1 microns), it is unclear how well this would apply to higher pressure membranes like RO or NF (Banerjee et al. 1999). In practice, the use of turbidity, even at the detection level allowed by laser turbidimetry, would be unlikely to work for high-pressure membranes, as feed waters typically undergo pre-treatment though MF/UF that substantially reduces the number of particles that can be detected (Adham et al. 1998).

2.2 Molecular Markers for DIT

2.2.1 Fluorescent Dyes as Markers for Membrane Integrity

Fluorescent dyes are common tracers in water studies, having been used to study flow, mixing, and other properties of natural and man-made water systems. Fluorescent molecules work by absorbing light of a certain frequency, then emitting light at a different frequency. In order to be used as a tracer, a fluorescent molecule or dye must be able to emit at a frequency that will be unique from other fluorescing compounds potentially in the water source. The molecule must be stable under a wide variety of conditions, and not be altered by exposure to oxidizers, acids, bases, sunlight, microorganisms, or changes in pH, conductivity, hardness, or temperature. Additionally, the molecule must remain in the water, and not be removed by corrosion byproducts or precipitate out of solution (Reggiani et al. n.d.).

While the USEPA MFGM requires that a DIT must have the resolution to determine if particles the size of virus are removed, if an increased concentration of a molecular tracer in the membrane permeate is uniformly distributed across a membrane train as a result of oxidation and aging damage, the uniformity of this concentration can be attributed to increased diffusion as opposed to a defect. If a challenge test demonstrates sufficient LRV, the challenge test LRV credit can still be used for compliance. The acceptable concentration of a molecular tracer in membrane permeate under this oxidized or aged condition should be included as a boundary limit within the challenge testing protocol, meaning that a maximum molecular tracer concentration in the membrane permeate should be attributed to an acceptable challenge test LRV for MS2.

All of the fluorescent dyes considered for use as a molecular marker DIT surrogate are significantly smaller than virus particles, meaning they provide conservative estimates of membrane integrity. Dyes with larger molecular weight would theoretically yield higher

calculated rejection values (i.e., LRVs), and be able to provide more accurate estimates of virus rejection, giving a more accurate representation of membrane integrity. Due to the sensitivity of fluorophore analysis and availability of online fluorescent analyzers, it may be possible to apply either: (1) a dilution model approach similar to that described in the MFGM for pressure-based testing (i.e., using air) and via conductivity profiling (Vickers 2018) or (2) the imperfection model approach (Frenkel and Cohen 2018) using an organic fluorophore. Such approaches would require instrumentation to conduct fluorescence profiling on a high-pressure membrane system (e.g., RO system), but could potentially yield higher LRVs by accounting for diffusion and separating this from defect caused fluorescence.

However, it should be recognized that the primary consideration with the use of fluorescent markers is the underlying fact that they are dyes. As such they are added to the feed to the RO/NF system and can accumulate to a visually unacceptable levels in the concentrate that is discharged from the RO/NF system. Thus, there may be inherent hesitancy upon the user towards the use of fluorescent markers that significantly alter the visual appearance of the water that is passed through the system.

2.2.1.1 Uranine

A number of studies have been performed evaluating the use of fluorophores for monitoring membrane integrity (Frenkel et al. 2014; Frenkel and Cohen 2018; Jacangelo et al. 2019; Lozier et al. 2013; Yoon 2019). Of four commercially available fluorescent dye candidates (eosin, fluorescein, rhodamine-WT, and uranine) tested in prior literature (Table 2-1) (Frenkel et al. 2014), uranine was selected for further testing as part of this work due to its high sensitivity.

Membrane filtration evaluations using uranine as a marker with the Hydranautics ESPA2 membrane demonstrated LRVs between 3.8 and 4.4-log at a nominal feed concentration of 40 mg/L, depending on permeate flux. Subsequent evaluation of intentional integrity breaches (i.e., pin holes and membrane degradation due to oxidation) demonstrated that uranine fluorescent monitoring was sensitive enough to detect relatively small breaches (Frenkel et al. 2014). These results were comparable to independent investigations, where uranine demonstrated an average LRV of 4.0-log with a continuous feed concentration of 1 mg/L (Jacangelo and Gray 2015). The very similar uranine LRVs across RO for the different feed concentrations suggest that lower feed concentrations may be possible while maintaining sufficiently high LRV.

Table 2-1. Various Fluorescent Markers Proposed for Inclusion in Membrane Integrity Testing.
Source: Frenkel et al. 2014

Fluorescent Marker	Excitation/Emission Wavelength (nm)	Molecular Weight (g/mole)	Aqueous Solubility (g/L)
Rhodamine-WT	554/580	480.55	n.r.
Rhodamine-B	554/576	479.02	50
Rhodamine-6G	526/552	497.02	20
Sulforhodamine-B	554/576	580.65	10
Amidorhodamine-G	530/551	552.59	Very Soluble
Fluorescein	490/520	332.31	0.3
Uranine	491/512	376.28	40
Eosin-B	516/538	691.88	40
Pyranine	455/512	524.39	178
Tinopal CBS-X	346/435	526.57	25
Erythrosine	525/547	879.87	20
Sodium Naphtionate	320/430	245.23	240
Lanaperl Fast Yellow	469/508	549.55	Very soluble
Lissamine-FF	432/508	404.38	40
Bengal Rose	518/535	1017.67	100
Fluorescent Brightener	349/430	960.96	Very soluble
Abbreviations: g/L - grams per liter; g/mole - grams per mole.			

In addition to identifying viable fluorescent markers for integrity monitoring, a testing approach termed pulsed-marker membrane integrity monitoring (PM-MIMo) using uranine as the fluorophore was reported (Frenkel and Cohen 2018). The PM-MIMo approach monitors the permeate fluorescence of a pulsed mass of uranine into the membrane feed water (although it could be any fluorescent marker) to determine the fraction of mass breaching the membrane in a specified time period. This mass fraction is continuously compared with experimentally determined mass fraction values of an 'intact' membrane to determine if integrity has been breached. In addition, the researchers evaluated the use of the Spiegler-Kedem (or phenomenological) model to distinguish between diffusive and convective transport (Frenkel et al. 2014; Frenkel and Cohen 2018).

2.2.1.2 3D TRASAR®

Nalco Water originally developed 3D TRASAR (TRASAR) as a monitoring additive for cooling towers and other water circulation systems, but it has also shown promise as a molecular tracer to monitor the integrity of high-pressure membranes. TRASAR contains an organic fluorophore that can be tracked through fluorescence measurements in near real-time (Steinle--Darling et al. 2016). Although originally scoped for investigation in this project, TRASAR was discontinued. However, the concept could be implemented with other fluorophores and is reviewed in this section.

The addition of TRASAR results in fluorescence at a level that is distinct and easily correlated to a concentration of the molecular tracer in the membrane feed and permeate water while being easily distinguished from natural fluorescent particles that may be present (Zeiher et al. 2003). The molecule can be detected at a concentration as low as 10 micrograms per liter (µg/L) with an online sensor (Steinle-Darling et al. 2016). The fluorophore in TRASAR is considered chemically inert and is blended with other select Nalco products for use in RO systems (e.g., antiscalant, biocide) (Reggiani et al. n.d.). When used for high-pressure membrane monitoring, TRASAR is typically combined with an antiscalant, though studies have shown improved LRV's without the presence of the antiscalant (Ostarcevic et al. 2018, 2013; Trussell et al. 2017). TRASAR is considered a conservative measure of membrane integrity, as the fluorophore has a molecular weight of 614 g/mol, significantly smaller than virus particles (20,000 to 200,000 g/mol or Daltons). The molecule is also negatively charged, which can help mimic the charge repulsion experienced by similarly charged virus particles, but also increases rejection across the system (Steinle-Darling et al. 2016). TRASAR fluorescence also has the advantage of not requiring reagents, and measurements are relatively simple and rapid to obtain, typically with immediate results (Reggiani et al. n.d.).

During operation, the TRASAR dye is added as a continuous dose to the feed water and the fluorescence output is monitored with an online sensor, which is correlated to the mass concentration of the molecular tracer in the water through standard calibration methods. A raw water production facility in Big Spring, Texas was able to collect readings as often as once per minute as part of their investigation into TRASAR technology; fluorescence data collection on such a rapid scale led to an immediate identification of an introduced cut O-ring. This facility measured LRVs of 4.3 across intact membranes and saw lowered removals in the second stage due to fouling from long-term use prior to experimentation (Steinle-Darling et al. 2016). Similar LRVs were seen during other studies, with typical values between 4.0 and 4.5-log when TRASAR was added with the antiscalant (Jacangelo et al. 2019; Trussell et al. 2017), but with values as low as 2-log also reported (Ostarcevic et al. 2018). When uncoupled from antiscalant addition, TRASAR was able to provide LRVs of greater than 6.0-log, providing some evidence that exclusion of antiscalant may provide better information on membrane integrity (Ostarcevic et al. 2018). Pulsing TRASAR into the system also allowed for higher concentrations to be used without incurring additional costs or risk of membrane fouling. In one study, the researchers found that LRVs increased with pulsed TRASAR addition from 4.6 to 4.9-log (Jacangelo et al. 2019).

While TRASAR does provide a conservative estimate of membrane integrity due to the size of the fluorophore relative to a pathogen, evidence has shown that the effect of aging and oxidation disproportionately affects TRASAR removals as compared to MS2. The study conducted at Big Spring showed that intentionally oxidized membranes had an LRV reduction to 3.6-log for TRASAR (compared to 4.3-log before oxidation occurred), while MS2 removal reportedly remained steady (Steinle-Darling et al. 2016). Oxidation damage and aging effects therefore appear to be conservatively captured by TRASAR before integrity breaches negatively impact the actual LRVs of pathogens. It is important to note that TRASAR was initially proposed as the fluorescent marker to be tested, however the product was discontinued prior to initial testing.

2.2.1.3 Rhodamine WT

Rhodamine WT (R-WT) is another fluorescent molecular tracer for use in integrity monitoring. R-WT is a non-reactive tracer chemical that has been approved by the USEPA for use in drinking water systems. It was specifically formulated for water tracing applications, both natural and man-made (Smart and Laidlaw 1977; Sutton et al. 2001). R-WT has a molecular weight of 487 g/mol, smaller than TRASAR, and is negatively charged at pH typical of RO and NF membrane feed water (i.e., pH 6 to 7). R-WT is expected to be rejected in a membrane system via charge repulsion and size exclusion and can be detected in the nanograms per liter (ng/L) range (Kitis et al. 2003b, 2003a). These aspects combined with its stability under a wide range of environmental conditions make R-WT a promising molecular tracer to be used for membrane integrity monitoring and, as such, it has been widely studied (Magal et al. 2008; Stanbro and Pyrch 1979). A summary of these studies and their removal values are shown in Table 2-2 below. Feed concentrations have been included for reference, as evidence shows that higher feed concentrations result in high LRVs. However, there is evidence that higher concentrations of dye in the feed can lead to adsorption and increase the risk of membrane fouling (Pype et al. 2016b).

Table 2-2. Summary of Rhodamine WT Studies and Associated RO LRV.

Feed Concentration (mg/L)	Dosing Mode	LRV	Reference
0.1 - 1.0	Continuous	3.5 - 5.3	(Kitis et al. 2003b)
1.0 - 2.0	Continuous	2.7 - 3.9	(Kitis et al. 2003a)
0.1 - 1.0	Continuous	2.0 - 5.0	(Lozier et al. 2013)
0.1	Continuous	2.6	(Lozier et al. 2011)
1	Continuous	3.6 - 4.8	(Lozier 2016)
1.0	Continuous	4.2	(Jacangelo et al. 2019)
5.0	Pulsed	4.8	
5.0 - 10.0	Pulsed	>4.0	(Ostarcevic et al. 2013)

2.2.2 Conductivity Monitoring (Combined Permeate)

A common alternative to turbidity measurements (discussed in Section Turbidity 2.1.2.1) used in high-pressure membrane applications for both DITs and CIIM is EC monitoring. Conductivity is considered a surrogate for total dissolved solids (TDS), and monitoring is relatively easy to

implement because conductivity instruments are relatively inexpensive, stable, reliable, and require limited maintenance. A survey of five utilities utilizing RO membranes as part of their treatment process was also conducted as part of this study. Of these facilities, all five had conductivity monitoring in place as a RO permeate monitoring technique, although conductivity measurements were not being used to monitor membrane integrity and instead focused on plant performance (Jacangelo and Gray 2015). Conductivity monitoring is also an accepted method of integrity monitoring because of the relative confidence regulators have in TDS rejection due to dissolved solids and ions being significantly smaller than known virus or protozoa. Sodium chloride, for example, has a Van der Waals radius of 0.282 nm, two orders of magnitude smaller than MS2 (Trussell et al. 2017). Conductivity monitoring is further supported as a measure of membrane integrity because integral RO membranes are highly efficient at removing salts. Feed in reuse waters tend to have high ambient levels of salts, meaning that additional seeding is not required to achieve up to 2-log LRV. Conductivity measurements are a common method of integrity monitoring despite relatively low LRV credit currently awarded, but there has been little research in the past focused on the process. It is not uncommon to see conductivity results reported in past research, but they are often reported as comparisons for surrogates or alternative methods. A summary of conductivity LRV's reported as part of other research projects can be found in Table 2-3.

Table 2-3. Observed LRVs Using Conductivity in Various Studies.

Observed Conductivity LRV	Facility Information	Primary Integrity Test Evaluated	Reference
1.7	Big Spring, Texas, 2-Stage, Pilot Scale	TRASAR	(Steinle-Darling et al. 2016)
2.4	Kamerik, Netherlands Parallel 8-inch RO Elements, Recirculated Water, Temperature Controlled	Natural Viruses	(Hornstra et al. 2019)
1.7 - 1.8	Gippsland Water Factory	R-WT	(Zornes et al. 2010)
1.5	Orange County Water District, California, 5 mgd Skid, 3-Stage	Naturally Occurring Surrogates	(Safarik 2020)
~1.5	Big Spring, Texas 2.4 mgd, 2-Stage	TRASAR	(Steinle-Darling et al. 2016)

Conductivity measurements can be collected either continuously online at the stage level or through periodic profiling (i.e., testing of each pressure vessel in a membrane train), with both methods having benefits and drawbacks. These approaches are described in the subsequent sections.

2.2.2.1 Continuous Online Conductivity Analysis on Combined Permeate Streams (Stage Testing)

Continuous online conductivity measurements provide a real-time evaluation of a system's gross integrity, as online conductivity has been shown to be sensitive enough to detect major breaches in a system (i.e., damaged O-rings) (Adham et al. 1998). The sensitivity of this method may vary depending on the size of a train and the number of pressure vessels contributing permeate water to the online instrument. Figure 2-1 shows an example of a typical three-stage RO unit. Conductivity analyzers are typically installed at locations where waters from multiple vessels have already combined.

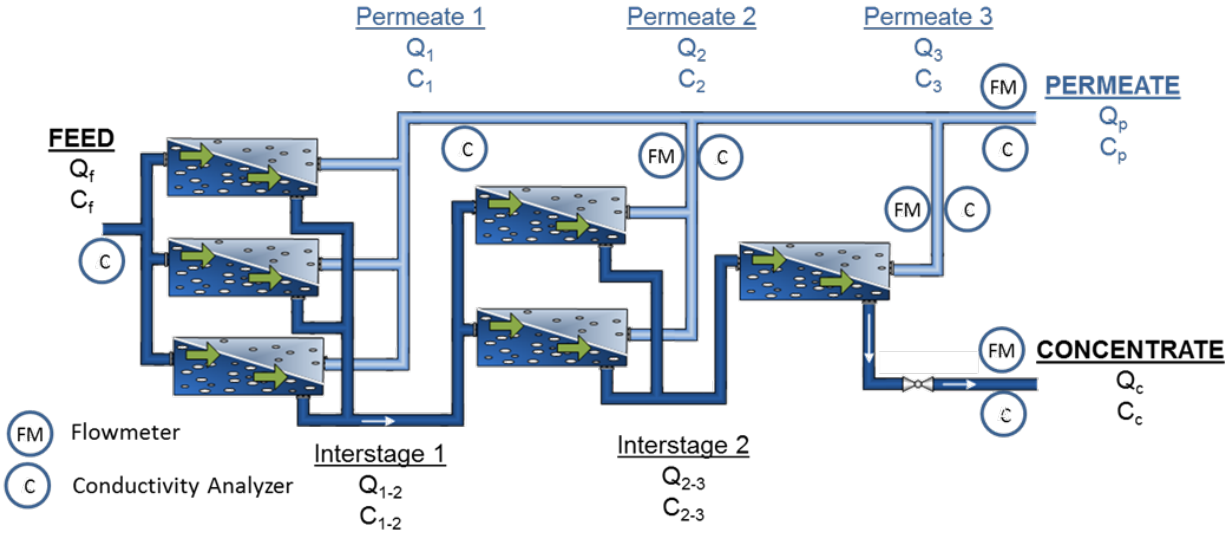

Figure 2-1. Typical Three-Stage Reverse Osmosis Unit Instrumentation.
Source: Vickers 2018

Permeate conductivity measurement can be affected by changes in temperature, feed TDS composition and/or concentration, pressure, flux, fouling, and even damage to the probe components (Ostarcevic et al. 2018). This variability can be addressed by monitoring water flow and conductivity for each stage, allowing performance to be calculated and/or normalized to a set of standard conditions. However, because permeate conductivity readings are typically taken on combined permeate, small defects in individual vessels can be masked by dilution. As such, conductivity monitoring by this method provides low sensitivity for integrity monitoring purposes, with facilities typically achieving LRVs between 1- and 2-log (90 to 99% removal). The State of California has granted 1.5-log virus removal for seawater desalination plants using RO in Carlsbad and Santa Barbara and 2-log virus removal for a seawater desalination plant in Avalon. In these cases, despite the MFGM's requirement that a DIT have the resolution necessary to attribute the size of a defect and its restriction on using non-mass-based bulk parameters as surrogates for challenge tests, conductivity is used as a surrogate for DIT. For Carlsbad and Santa Barbara, 1.5-log removal is granted based on the requirement that the permeate from not more than eight pressure vessels can be combined for a composite DIT. For Avalon, 2-log virus credit is granted but every vessel must be tested individually.

2.2.2.2 Conductivity Profiling of Vessels

Conductivity profiling involves testing the permeate specific to a pressure vessel, which is the blend of permeate from all membrane elements within a vessel. Conductivity profiling is not commonly performed online but can be automated through the use of valves to multiplex a single conductivity probe. Profiling is often used to find the location of a vessel with integrity failure, and then vessel probing is conducted to identify the location of a damaged element (Adham et al. 1998; AWWA 2007).

Conductivity profiles (e.g., as a DIT) can be generated in response to a change in combined permeate conductivity (e.g., as CIIM) as a way to identify an outlier element or generated as part of regular monitoring on a weekly, monthly, or quarterly basis. Currently, operators typically identify conductivity outliers through non-statistical means. Work is being undertaken to apply formal statistical limits to these profiles as a way to standardize the process and is discussed in detail in sections 4.2 and 4.3.

Automation of conductivity profiling can reduce the time required for profile generation. The YVWD, located in Southern California, has installed a novel automated conductivity probing system capable of collecting conductivity data across an RO system as frequently as every two hours. The automated system is a solenoid manifold installed on a two-stage (52:20) 1,650 gpm RO train at the Wochholz Regional Water Recycling Facility (WRWRF). The system is programmed to index pressure vessels one at a time to measure permeate conductivity across the entire system using only a single conductivity probe (Vickers and Dummer 2020). The combination of automated profile generation and the implementation of standardized statistical evaluation, as discussed in section 4.4, has the potential as a significantly improved DIT resulting in higher LRVs without additional work by operators at a low cost.

2.2.2.3 Vessel Probing

Conductivity probing involves sampling the permeate conductivity at set lengths along a pressure vessel and can isolate damage to individual membrane elements. Unlike online conductivity monitoring, conductivity probing does not provide real time (continuous) monitoring but is more sensitive to the extent and location of membrane element specific breaches in a system.

2.2.3 Total Organic Carbon Monitoring

TOC monitoring is another common method of CIIM monitoring used by the reuse industry. However, TOC monitoring is not an accepted approach for DIT in the drinking water industry because TOC that occurs naturally does not have a uniform size that can be compared to the size of virus (Cai et al. 2020). Therefore, the use of TOC as a monitoring method does not meet the MFGM's resolution criteria. It should be noted that DIT methods that are described in the MFGM apply to systems that require 3-log or greater removal of *Cryptosporidium*, and that the practical limit for TOC removal LRV credit is a maximum of two.

In applications where TOC is an accepted approach for integrity verification, TOC monitoring involves the oxidation of carbon containing molecules in the water stream and the detection of carbon dioxide generated in the process. Naturally occurring TOC in reuse applications has been found to have a greater resolution than conductivity monitoring and is able to achieve an LRV of 1.6-log without spiking (Adham et al. 1998). Since the TOC in the feed and the permeate streams are at concentrations on different orders of magnitude, it is common for the implementation of two TOC analyzers - one able to detect higher TOC concentrations present in the feed stream and a second to monitor the lower TOC concentrations in the permeate stream. However, the cost of two instruments may limit some facilities from implementing TOC monitoring (Ostarcevic et al. 2018). Like conductivity, TOC is not considered directly representative of virus transport through a membrane since a portion of organic matter is able to pass through the membrane by diffusion - a transport mechanism not available to viruses (Trussell et al. 2017).

2.2.4 Sulfate Monitoring

Sulfate (SO_4^{2-}) has been identified as a potential naturally occurring surrogate for high pressure membrane integrity monitoring. Sulfate monitoring is an attractive surrogate because it is commonly present at high enough ambient concentrations in membrane feed waters, in part due to sulfuric acid dosing. Sulfuric acid is a common method of lowering the pH of feed waters to reduce the risk of forming calcium carbonate mineral scale (Ning and Netwig 2002). Constant dosing of sulfuric acid has the added benefit of ensuring a more consistent sulfate feed concentration in addition to increasing observable LRV.

Online sulfate monitoring does exist but currently must be done by ion chromatography (IC) which is the only instrument with a low enough detection limit to measure sulfate in the RO permeate. Online IC is expensive and usually cost-prohibitive (Jacangelo et al. 2013). Collection of grab samples for ex-situ testing can be done for same-day analysis, but this method would not be able to provide real-time information on membrane integrity. Despite this, it has the potential to fulfill the requirement for a daily DIT (Trussell et al. 2017). A number of online and automated sulfate monitoring systems have been developed including the TitriLyzer and EZ Series analyzer (Hach USA), online IC (Metrohm, Switzerland), and Mettler Toledo (Columbus, OH). Among these only the Metrohm online IC has a low enough detection limit for RO permeate. The TitriLyzer can monitor up to eight streams sequentially and the EZ analyzer can monitor three streams sequentially although upgrades to automated sample valves can increase the number of streams that can be monitored with each system (Applitek 2016, 2013).

Sulfate ions are significantly smaller than the smallest virus particles, therefore monitoring would be a conservative estimate of membrane integrity. But sulfate also has the potential to pass through the membrane via diffusion as well as through defects. It may be possible to differentiate between sulfate diffusion and the passage of sulfate through defects using the method proposed for conductivity profiling (Vickers 2018). Online sulfate monitoring combined with conductivity profiling has the potential to increase removal credits, however, no work has yet been performed to combine the methods.

Current work with sulfate monitoring that have not differentiated between diffusion and defect related sulfate concentrations has observed removals typically around 3-log (See Section 9.1.1). Sulfate monitoring of the Western Corridor Recycled Water Scheme in Queensland, Australia observed 3-log reductions of sulfate in daily integrity testing. Removal may have been higher, but sulfate concentrations in the feed were not high enough to achieve more than 3-log (Jacangelo and Gray 2015). A test performed at San Diego's North City Water Reclamation Plant (NCWRP) observed sulfate rejections ranging from 2.4- to 3.1-log. While these results are in line with typical values, the data was collected over a 24-hour period and may not be representative of the LRVs the plant would achieve with monitoring over a longer period of time.

The Heemskerk water treatment plant in the Netherlands was able to establish 3-log removal credits over a six-month monitoring period in 1998. This was lower than the tested MS2 removal of 4.8-log, demonstrating that the measurements were conservative but still above the 2-log LRV measured by conductivity removal (Kruithof et al. 2001). Limitations for this compound include background concentration in the feed as well as the method detection limit for the permeate.

2.2.5 Strontium Monitoring

Strontium has also shown promise as a surrogate for integrity monitoring in high-pressure membrane systems because strontium is generally naturally found in wastewater at sufficiently high enough concentrations that additional spiking is not required. Several investigations involving strontium removal have been performed without accounting for diffusion related strontium transport, which conservatively represent the LRV this molecular marker can achieve. For example, investigations into strontium removal at the Orange County Water District Groundwater Replenishment System found that strontium removal was typically between 3.0 and 3.3-log, with minimal variation seen during the 24-hour sampling period (Safarik 2020). Additional studies on strontium removal have found similar results, with ranges from 2.1 to 3.6-log (Bernados 2018; Liang et al. 2011; Subramani et al. 2010; Trussell et al. 2017). Limitations for this ion include background concentration in the feed as well as the method detection limit for the permeate.

Strontium is also found at elevated concentrations in radioactive wastewater, with work being performed to assess removal by membranes across a wide range of conditions. NF experimentation performed on simulated low-level radioactive waste material found strontium rejections were near 100% at high pH, and rejections from 80-85% at more neutral pH levels (Chen et al. 2018). Similar investigations found that strontium rejection reached a minimum of 97.2% rejection (approximately 1.98-log) at a pH of 5.0, where the membrane in use for the experiment reached its isoelectric point. Rejection values of 99.0% (2-log) and higher were observed below a pH of 5 and above a pH of 9.0. Experiments with spiked surface waters obtained 97.5% rejection (1.99-log), as well (Ding et al. 2015). While the latter experiments are of relatively extreme conditions, they do support the evidence that strontium is a viable surrogate for membrane integrity monitoring. Online strontium analyzers are currently available but are costly and would not be anticipated to sample and measure at a rate sufficient to be used for CIIM.

2.2.6 Sucralose Monitoring

The artificial sweetener sucralose ($C_{12}H_{19}Cl_3O_8$) is an organic compound that is widely used as a food additive, but it is not well adsorbed or metabolized by the human body, leading to the majority of it being passed into wastewater (Roberts et al. 2000) and subsequently into the environment due to incomplete removal during wastewater treatment (Soh et al. 2011; Torres et al. 2011).

Artificial sweeteners such as sucralose are currently being used as indicator compounds for the presence of wastewater in drinking water sources (Oppenheimer et al. 2011). Sucralose has been found in surface waters in 27 European countries, as well as being shown to be widespread in the United States (Mawhinney et al. 2011). Because of its ubiquity in wastewater effluent, sucralose has promise as an integrity monitoring surrogate.

Like many of the other molecular markers discussed in this section, sucralose's low molecular weight (397.6 g/mol - (NLM 2020)) means it is able to diffuse through high pressure membranes, a method of transmission not available to much larger virus particles. Therefore, without isolating the effects of diffusion from the sucralose that may result from defects, the work that has been performed to date represents a conservative basis for assessing membrane integrity using sucralose as a molecular marker. Sucralose removals at a target feed concentration of 5 mg/L with permeate monitoring able to quantify LRV of 2.9-log across both unused and aged membranes, an increase of 1.0-log from conductivity monitoring done as part of the same experiment (Trussell et al. 2017). The Raw Water Production Facility in Big Springs, Texas saw log removals of 2.4 from facility influent to product water, with virtually no change from where the water entered the plant to entering the RO feed (Steinle-Darling et al. 2016). Sucralose monitoring does have the drawback of requiring liquid chromatography tandem mass spectrometry (LC-MS/MS) analyses. The coordination, cost, and turnaround time required for analysis with a contractor or in-house labs limits the feasibility of daily integrity testing. The cost of regular LC-MS/MS analysis may also deter smaller facilities from adopting regular sucralose monitoring.

2.2.7 Fluorescence Excitation-Emission Matrix Spectroscopy

Excitation-emission matrix (EEM) results from fluorescence spectroscopy. An EEM is a three-dimensional contour plot of thousands of data points generated by measuring fluorescence intensity across a range of excitation and emission wavelengths (Rosario-Ortiz and Korak 2017). EEM results have been used to characterize dissolved organic matter (DOM) in water systems. Use for EEM spectroscopy has been reported for water quality monitoring (Stedmon et al. 2003; Yan et al. 2000), effluent flow tracking (Baker 2001), and in monitoring recycled water schemes (Hambly et al. 2010) and has been investigated for its capacity as a high-pressure membrane integrity monitoring technique (Pype et al. 2013).

DOM, which is being measured as part of EEMs, is a heterogeneous mixture of aromatic and aliphatic hydrocarbon structures containing different functional groups (Pype et al. 2013). However, only a small fraction of DOM molecules have the ability to absorb wavelengths greater than 200 nm (chromophoric DOM [CDOM]), and only a fraction of CDOM will fluoresce. In fact, less than 3% of absorbed photons are emitted as fluorescence (Del Vecchio and Blough

2004). As such, EEM analysis should be done with great caution and oversimplifying data from such a small subset of DOM particles risks bias in the analysis and masking integrity issues (Rosario-Ortiz and Korak 2017). Research on EEM as a monitoring technique has found a calculated LRV of 3-log (99.9%) when combined with size exclusion chromatography (Yoon 2019).

A recent review concluded that use of natural fluorescence is unlikely to yield LRVs greater than conductivity measurements (Yoon 2019). Natural DOM also fails to meet the MFGM's resolution requirement for a DIT method as the size of DOM is not uniform and therefore not comparable to the size of a virus. In addition, there is currently no standardized method of interpreting EEM and additional work is needed to verify its applicability to high-pressure membrane monitoring.

2.2.8 Nanoparticles

Nanoparticles are becoming increasingly accepted as potential surrogates, because they can be produced at sizes similar to the smallest virus particle and remove some of the risk involved in challenge tests using live MS2 bacteriophage. A wide range of potential nanoparticles exist, including fluorescent micro – and nanoparticles (Acker et al. 2001; Gitis et al. 2006; Kitis et al. 2003b, 2003a; Mi et al. 2004; Ostarcevic et al. 2013; Pontius et al. 2009), gold nanoparticles (Gitis et al. 2006), and magnetic nanoparticles (Deluhery and Rajagopalan 2008; Guo et al. 2010). While these particles offer the potential for real-time integrity monitoring, further investigation needs to be done on the process. For example, the effects of solubility, surface chemistry, agglomeration, and other factors influencing rejection is still largely unknown as most challenge tests to date have not considered their effects.

It has been reported that some nanoparticles aggregate and foul the surface of membranes, providing overestimated calculations of LRVs and system integrity (Lohwacharin and Takizawa 2009). If the fluorescent markers are improperly attached to the nanoparticles, there is a risk of the fluorescence leaching from the nanoparticle's surface into the surrounding waters, again providing incorrect estimates of membrane integrity (Jacangelo et al. 2019). Production of the large amounts of fluorescent nanoparticles needed for full scale monitoring would be expensive, particularly for smaller facilities, despite the relative ease of using fluorescence monitors (Ostarcevic et al. 2018).

There is additional work needed to determine the fate and toxicity of the synthetic or engineered nanomaterials in the environment. Such information in relation to nanoparticles is largely unknown with the exception of common inorganic nanoparticles such as titanium dioxide (O'Malley 2015). The use of magnetic nanoparticles could potentially address many of these concerns, as they could potentially be recovered for reuse, but testing is limited and has largely been performed on UF membranes (Guo et al. 2010). The cost of implementing the required instruments for full-scale implementation of magnetic nanoparticle detection and recovery would be of considerable cost to facilities.

Despite this, nanoparticles have shown the ability to provide the required sensitivity for high pressure membrane integrity monitoring. Silver nanoparticle feed concentrations of >8 mg/L were able to obtain maximum LRVs of 2.6-log, though the detection mode was limited to off-

line analysis (Antony et al. 2012). A more recent test was able to calculate an LRV of >5-log at a pulsed feed concentration of approximately 2 mg/L, but offline analysis and integration of results was required (Jacangelo and Gray 2015).

2.2.9 Adenosine Triphosphate

Adenosine Triphosphate (ATP) is a nucleotide that transfers energy within cells, is present in all living cells, and degrades rapidly when cells die (Abushaban et al. 2017). ATP measurements are useful as surrogate measure of all active and unculturable microbial cells, thus providing better estimates of biomass than heterotrophic plate counts (Siebel et al. 2008). This makes ATP measurement a useful surrogate for membrane integrity monitoring. Total ATP in a water sample is the sum of cellular ATP (cATP) that is still bound within living cells combined with extra-cellular ATP (free ATP) from dead or lysed cells. Free ATP, with a molecular weight of 507 g/mole, should theoretically be well removed by RO membranes with a molecular cutoff of 150 dalton (Da) or lower (Ozaki and Li 2002) and is abundant in feed waters (Safarik 2020).

Currently ATP is used as a biomass indicator in RO systems to quantify biomass on membrane surface, diagnose biofouling, measure biomass in feedwater, and as a biomass parameter in bacterial growth potential measurements. Up until recently, ATP levels could not be measured online, and ATP concentration measurements relied on a semi-rapid (15 minutes) bioluminescence method in batch mode, with the level of light output detected correlating directly to a sample's ATP concentration (Steinle-Darling et al. 2016). The method does not work for high ionic strength waters, such as seawater, due to inhibition of the ATP reaction and interference with background luminescence (Abushaban et al. 2017). Waters at the Big Spring Raw Water Production Facility were found to have significant reductions in ATP levels across the RO system, with permeate samples falling below 0.1 picograms per milliliter (pg/mL) of cATP from approximately 25 pg/mL in the feed (Steinle-Darling et al. 2016). Similarly, work done as part of WRRF Reuse-12-07/WRF4757 calculated 2.5-log reductions in ATP levels across an intact system (Jacangelo et al. 2019). ATP measurements also showed sensitivity to common integrity damage tests, with significant LRV reductions from introduced damage. More recently, several companies have offered online ATP monitoring devices such as the HACH EZ-ATP Online Microbial Analyzer (Loveland, Colorado). In practice, such a device could be used to monitor ATP in near-real time as a membrane integrity monitoring approach. Orange County Water District has reportedly been evaluating online ATP monitoring for integrity monitoring of RO systems at their Groundwater Replenishment System.

2.2.10 Additional Methods

A comprehensive review of membrane integrity verification approaches, that were in development prior to 2019, was summarized as part of WRRF Reuse-12-07/WRF4757, See (Jacangelo et al. 2019). The lengthy list of 'monitoring techniques' is not included herein; however, it is worth noting that significant effort has been put towards developing more effective integrity monitoring approaches across a number of research projects (Frenkel et al. 2014; Jacangelo et al. 2019; Trussell et al. 2017). The goal of this research project was to identify DIT options for RO that are commercially viable and available now. Consequently, many of the methods proposed in prior projects were not included in this review due to high cost of implementation, impracticality for large-scale membrane systems, and/or the requirement of additional research and development prior to deployment.

Flow cytometry (FCM) among other pathogen quantification methods has been investigated as a tool for indirect integrity monitoring as it can measure total virus particles. FCM measurements at a water reuse facility employing MF-RO indicated that approximately 3-log removal could be quantified on the combined MF-RO system using FCM measurements. In addition, the researchers evaluated dynamic light scattering (DLS) as a method for submicron particle counting. Both FCM and DLS methods were reported as relatively expensive relative to other CIIM methods (i.e., conductivity and TOC) and required additional development for deployment (Huang et al. 2015).

Removals of naturally occurring viruses in fresh surface water across RO membranes were investigated as a method of DIT. Surface water analyses identified two species that were at high concentrations in the six European surface waters being tested. Once identified, quantitative polymerase chain reaction (qPCR) assays were developed to allow testing for the selected natural viruses. LRVs of more than 6.0-log for both natural virus species across intact membranes were observed. Both natural viruses also showed significant reductions in LRVs when damage was intentionally introduced into the system (Hornstra et al. 2019). The identification of natural viruses in an RO system would allow for similar monitoring sensitivity as achieved by MS2 but with the advantage of avoiding the cost and practical challenges of spiking a non-indigenous organism. Use of qPCR remains an interesting approach for quantification of viral surrogates. However, the following uncertainties remain with respect to use of indigenous viruses:

- The indigenous virus must be at sufficient concentration to allow detection in the feed and ideally in the permeate, or an automated permeate sample concentration step needs to be developed to improve sensitivity.
- Ideally, a ubiquitous indigenous virus would be selected such that performance could be compared between sites.
- The size and other characteristics of indigenous viruses should be quantified such that their use as a surrogate can be associated with a particular defect resolution.
- The removal of indigenous viruses should be demonstrated to either correlate with and/or be a more conservative indicator of virus removal than MS2.
- The cost, reliability and sensitivity of semi-automated qPCR instruments needs to be defined.

Commercially available, online, and semi-continuous water quality monitoring devices have become more prevalent over the past several years. Several commercial systems have been developed for detection of anomalous water quality events in drinking water distribution systems and wastewater collection systems including S::CAN™, LiquID™, and Kando, or software coupled to water quality measurements such as CANARY. Based mainly on spectrophotometric analysis, these systems are typically used to detect changes in water quality and alerting operators to potential water and wastewater system upsets. A review of real-time water quality monitoring to aid in decision making at wastewater treatment facilities concluded that combining water quality monitoring data with machine learning may aid in the rapid detection of high-pressure membrane integrity issues and potentially, higher LRVs (Newhart et al. 2019).

2.3 Summary of Findings

Facilities employing NF or RO typically rely on conductivity monitoring to obtain a base-level LRV credit of 1.0 to 2.0-log. A significant amount of fundamental and applied research has been undertaken to develop DIT and CIIM approaches to improve integrity monitoring and obtain greater LRVs in accordance with the MFGM. Because the LT2ESWTR does not require the use of a particular DIT for rule compliance but stipulates that any test used meet must the specified performance criteria for resolution, sensitivity, and frequency, researchers and practitioners have focused on alternative surrogate markers (e.g., molecular markers) that meet the requirements of the MFGM and result in appropriate LRV credits.

The MFGM also does not consider the concept of multiple location DIT such as the use of individual and automated conductivity profiling (Vickers 2018). The work done at YVWD has shown that multiple location DIT has the potential to more accurately describe removals than traditional methods of measuring combined permeates, thus addressing a traditionally overlooked method of integrity characterization. The MFGM largely focuses on verification of *Cryptosporidium* removal by MF/UF processes. There are a number of areas where the guidance in the MFGM could be improved to better account for the practical differences involved in monitoring RO for virus and protozoa removal when compared to MF/UF.

The objective of this literature review was to screen pertinent sources of information to identify promising membrane integrity monitoring approaches to be evaluated through experimentation during the course of this project. Various indigenous and exogenous molecular markers have been evaluated for use in DIT and CIIM of RO and NF membranes. Several markers were identified to yield quantifiable LRVs of 3-log or higher including certain fluorescent dyes, sulfate, strontium, and wastewater derived organic contaminants (sucralose). Considering the practicality (e.g., cost and measurement ease) and regulatory (e.g., resolution and sensitivity) requirements associated with marker adoption, it appears feasible to achieve close to 3-log LRVs through either dosing and/or monitoring of an appropriate fluorescent dye (e.g., uranine) or sulfate.

Although molecular markers have been identified that can be used in DITs or CIIM to quantify LRVs greater than those obtained through conductivity monitoring, findings of this literature review suggest that routine monitoring of these markers is unlikely to yield LRVs significantly

greater than 3-log. This is largely due to the fact that diffusion of these markers through the membrane constrains measurement of LRVs to around 3-log, even with no loss of integrity. Furthermore, the sensitivity (i.e., the LRV able to be demonstrated) by a majority of these markers is intrinsically linked to the commercially available detection limits for RO permeate and their RO feedwater abundance.

Approaches analogous to the dilution model correction applied to pressure-based DITs outlined in the MFGM have been developed. The conductivity profiling method uses statistical analysis to differentiate between diffusive and advective solute mass transfer and calculates LRVs based on permeate conductivity resulting from an integrity breach. The combination of automated conductivity profile generation and this monitoring approach for the system at YVWD improved the DIT sensitivity and LRV increased from 1.6 to 3.2-log based on conductivity (Vickers 2018). A similar concept also identified a membrane imperfection model to assess integrity through permeate monitoring (Frenkel et al. 2014). Combination of one or more of these approaches with measurement (i.e., profiling) of a more sensitive molecular marker (e.g., fluorophore or sulfate) that by itself yields quantifiable LRVs greater than 3-log, could improve the DIT sensitivity and possible RO LRV credit significantly.

2.4 Conclusions

Despite the ability of RO, and to a lesser extent NF, to act as a robust barrier to dissolved solids (e.g., sodium and chloride), high-pressure membrane systems are routinely granted conservative pathogen LRVs by regulators (1 to 2 LRV credit) when using the integrity monitoring guidelines outlined in the MFGM. A significant amount of work has been performed to demonstrate higher LRV credits including application of sophisticated and sensitive analytical techniques, statistical data analysis, and the use of membrane transport models. The main challenges associated with successful adaption of these approaches are 1) finding a molecular marker that meets the resolution requirements of a DIT as specified in the MFGM, meaning its size can be compared directly to that of virus; 2) finding a molecular marker and/or analytical approach that meets the sensitivity requirements of the MFGM, ideally demonstrating at least 4-log removal; 3) applying a marker monitoring approach to a system which could potentially include many pressure vessels on a daily or semi-continuous basis; and 4) obtaining regulatory approval.

The major findings of this literature review that provide framework for the full scale and pilot scale testing to be conducted as part of this project are:

1. The combination of a dilution or imperfection model approach similar to Vickers et al. (2018) or Frenkel et al. (2014) with multiple location measurement (i.e., profile generation from individual vessels) of a more sensitive molecular marker (e.g., fluorophore or sulfate) can be accomplished within the context of the MFGM's criteria and when accepted by regulators can enhance LRV sensitivity by 1 to 2-logs (or more) for many easy-to-use marker-based DIT methods.
2. Marker based DIT methods such as fluorophore, sulfate, or strontium are the most developed, meet the MFGM's requirements for resolution (i.e. they can demonstrate breaches smaller than a virus), and when used in combination with the dilution model approach by Vickers et al. (2018) or the imperfection model approach by Frenkel et al. (2014) (i.e., Spieler-Kedem or phenomenological model), have analytical methods that can provide sensitivity sufficient to demonstrate at approximately 4-log (or more) LRV. Therefore, these methods are recommended for further testing as part of this project.
3. To address the question of how aging can affect the sensitivity of DIT methods, challenge testing with molecular tracers and MS2 should be conducted under a variety of conditions that represent aging impacts. As per the MFGM, because the LRV that a membrane system will receive is the lower of two demonstrated values: (1) the removal efficiency demonstrated during challenge testing or (2) the maximum LRV that can be verified by a DIT used during the course of normal operation, the challenge testing to be conducted for this project may likely also need to account for increased diffusion that result from aging to validate DIT results in the context of MS2 challenge test data.

CHAPTER 3

Testing and Calculation Methods

Testing was carried out at four different facilities as a critical component of this research report. During integrity testing field work, methods were replicated across different facilities to the greatest extent possible, but given the geographic differences, absolute alignment on test methods was not possible. In addition, conductivity profiling data, collected as part of regular process checks, were also provided from three additional full scale RO facilities. This chapter summarizes the common methods used to analyze surrogates.

3.1 Overview

The following integrity testing approaches were investigated during field work as part of this study:

- Continuous Indirect Integrity Monitoring:
  - Reporting of online conductivity from the following possible process locations:
    - RO feed
    - Combined RO permeate
    - Permeate from each stage
  - Total Organic Carbon monitoring measured in the RO feed and combined RO permeate.
- Marker Based Integrity Surrogates Measured on RO feed and combined permeate:
  - Sulfate
  - Strontium
  - Phosphate
  - Magnesium
  - Uranine
    - Also measured on the permeate from individual vessels at OCWD and YVWD.
  - Sucralose
  - MS2
- Direct Integrity Testing Methods:
  - Vacuum decay testing
  - Conductivity profiling

For the four project testing locations, the nominal analytical method as well as the frequency of analysis and method reporting limit (MRL) are summarized in Table 3-1.

Table 3-1. Summary of Test Schedule, Methods and MRLs from Field Studies.

Parameter	Equivalent Analysis Method	Site Specific Number of Tests (n) and MRL[1]			
		SNWA[6]	YVWD	OCWD	WCWA
		Jan - Apr 2021	Feb - Dec 2020	Feb - Sept 2021	Oct 2020 - Apr 2021
Conductivity	SM 2510B	Online	Online	Online	Online
Conductivity Profiling	(Vickers 2018)	N/A	Online[2]	1/week[3]	n = 27[2]
Sulfate	EPA 300.0	n = 48 [0.25 mg/L]	n = 1 [0.24 mg/L]	n = 2 [0.5 mg/L]	n = 52 [0.11 mg/L]
Uranine[8]	Fluorometer Calibrated with Uranine	n = 48 [0.1 µg/L]	n = 1 [0.01 µg/L]	n = 2 [0.02 µg/L]	n = 10 [0.01 µg/L]
Strontium	EPA 200.8	N/A	N/A	n = 2 [0.3 µg/L]	n = 52 [0.002 mg/L]
MS2[7]	(Adams 1959)	n = 99 [0.1 pfu/mL]	n = 7 [1 pfu/mL]	N/A	N/A
Sucralose	EPA 1694M[4]	n = 33 [5 ng/L]	N/A	N/A	N/A
Magnesium	EPA 200.8	N/A	N/A	N/A	n = 52 [0.1 mg/L]
Phosphate	EPA 300.0	N/A	N/A	N/A	n = 52 [0.005 mg/L]
TOC	SM 5310C	N/A	N/A	Online [0.03 µg/L][5]	N/A

Notes:

MRL for conductivity (and conductivity profiling) is not specified as all conductivity readings were detected. However, maximum LRV due to conductivity profiling was capped at five in all cases.

Profile results automatically generated. WCWA test frequency every two weeks, data for two of four trains analyzed.

Profile results manually generated via grab samples for a specific test skid.

Solid-phase extraction coupled with high-performance LC/MS/MS in tandem.

Practical reporting limit for online TOC monitor reported.

Total number of Tests spread across four different membrane products under both new and intentionally damaged conditions.

MS2 stock and analysis provided by GAP environmental laboratories, Ontario, Canada. Samples were shipped on ice and analyzed within hold times.

Uranine profiles of each studied vessel were conducted at OCWD and YVWD.

Abbreviations: N/A - not applicable, analysis not performed. Online means analysis and reporting of results from a dedicated apparatus connected to the system. Frequency of reporting depends on the analytical parameter.

CHAPTER 4

Conductivity Profiling to Credit RO

As noted in Chapter 2, a number of investigations have previously taken place to validate new approaches for crediting LRV for RO (Frenkel et al. 2014; Jacangelo et al. 2019; Trussell et al. 2017). A unique aspect of this research report contains a detailed evaluation across multiple facilities of a method that utilizes vessel specific profiling to verify the integrity of an RO array and system performance as a whole. The proposed approach described in this chapter was adapted from previous peer reviewed literature (Vickers 2018). In general terms, testing of this type is based on the sampling of individual pressure vessels and is characterized as a multiple point test method versus the single point test method that is described in the MFGM.

It is noted that conductivity profiling is different from conductivity monitoring which involves the sampling of the combined permeate. As noted in Section 2.2.2.2, conductivity profiling involves testing the permeate specific to a pressure vessel, which is the blend of permeate from all membrane elements within a vessel.

Automatic conductivity profiling can be used to characterize the removal performance of an RO system. The majority of results within this chapter refer to a device that was designed, constructed, and installed at the YVWD WRWRF. The automated sampling apparatus performs sampling of each vessel in this system every two hours, which resulted in twelve samples collected per day. As an operational practice, YVWD has performed routine conductivity profiling of the RO train on a monthly basis since installation in January 2013. In January 2017, there was one observed incident where performance of a single vessel became questionable. One RO element and its interconnectors were removed and replaced. Other than routine conductivity profiles, there were no other modifications of the RO system prior to the installation of the automatic conductivity profiling unit.

The goal of this chapter is to describe the method of how conductivity profiling results can be statistically interpreted to verify array integrity and to infer a LRV for the RO system. In addition, this chapter provides information on the performance of an automated conductivity profiling system that would allow the use of conductivity profiling at a frequency sufficient to qualify as a DIT per the MFGM (i.e., at least once per day). Also, the sensitivity enhancement to LRV calculated from conductivity profiling, compared to conventional direct feed to permeate conductivity monitoring, was quantified based on data from other contributing utilities. Finally, a sensitivity analysis is provided for the conductivity LRV approach based on results from the automated system at YVWD.

Conductivity probing involves sampling the permeate conductivity at set lengths along a pressure vessel and can isolate damage to individual membrane elements. Procedures for both profiling and probing are well established RO troubleshooting and maintenance approaches and are described in detail elsewhere (AWWA 2007).

4.1 Conductivity Profiling in Practice

Conductivity profiling is intended to be straight forward with standard sampling, analyzing, and recording system components:

- The permeate and concentrate flow from each stage requires:
  - Installed and calibrated online flow monitoring to fully define these flows or calculate unspecified flows via material balance.
- The conductivity of individual pressure vessels requires:
  - Valves and a dedicated sampling panel to allow sampling of permeate from individual vessels. See Figure 4-1 below showing the end of the vessels for the YVWD system and the profiling sample taps mounted on a single panel to allow sampling from each RO vessel.

Figure 4-1. RO Pressure Vessels and Sample Panel (bottom left) at YVWD to Allow Manual Conductivity Profiling.

After sampling and recording conductivity from each vessel a profile of the entire system is generated. Conventionally, these are shown (and recorded) in a manner that matches the physical system arrangement. An example profile for a 3-stage system is included in Figure 4-2, below, showing incrementally increasing permeate conductivity in each stage.

Stage 1

16	16	19	17	17	16	17	16	16	17	17	Average
16	17	18	17	17	15	16	16	17	17	19	17.5
19	17	17	17	18	17	18	15	18	18	18	
20	17	19	17	18	17	16	16	19	18	19	
19	17	18	18	18	17	16	18	18	18	18	
17	17	19	17	18	18	15	19	17	19	17	
17	19	17	18	19	21	16	18	17	19	19	

Stage 2

55	56	54	56	54	52	54	Average
56	55	54	55	53	54	54	52.4
55	56	44	62	44	58	56	
55	40	48	56	45	56	56	
55	42	43	52	45	57	57	
56	42	47	45	52	56	57	
56	44	48	48	62	56	56	

Stage 3

191	177	144	142	Average
163	179	171	149	166
186	186	166	149	
186	192	150	148	
180	172	149	151	
172	172	145	163	

Figure 4-2. Example Conductivity Profile Results from a 3-Stage RO Array.
Each box represents the measured conductivity in uS/cm from each pressure vessel.

The discussion to follow considers the statistical approach that can be applied to the measured conductivity across all vessels within a stage (i.e., the results in each box) and how they can be interpreted to verify the array integrity.

4.2 Defining a Statistical Framework to Determine Array Integrity

Off specification performance in RO systems is associated with vessels that have higher conductivity than normal, otherwise known as outliers. Conceptually, the performance of an integral stage (i.e., a stage of an RO system where all membrane integrity is nominal) should fit a statistical distribution and/or limit, thus a straightforward statistical analysis can be applied to identify the outlier vessels. Currently, there are no specific rules associated with this practice. However, after an examination of all 21 RO units from the Orange County Water District, the following approaches were developed and appear appropriate to correlate operational practice to statistical methods.

One approach is to apply the Central Limit Theorem and the “3-Sigma rule” (i.e., in any normal distribution of data, 99.73% of the samples should fall between three standard deviations [σ] of the mean) regarding the sample in order to test the validity of the population. The Central Limit Theorem is valid for an RO unit, as there are small differences in membrane flow and salt rejection properties between pressure vessels.

Another approach is to state that the highest conductivity should be no more than 50% greater than the median conductivity of the stage, referred to subsequently as 1.5*Median. This type of test is commonly applied to situations such as a new membrane installation which may have multiple questionable vessels. Use of a percent above the median as an approach to set alarms may be more applicable for use in smaller systems where there are an insufficient number of pressure vessels available for determination of conductivity statistics that benefit from a higher number of observations.

Skew is the measure of the distribution above and below the average, with positive values representing higher than anticipated values, outliers of a distribution, or vessels that may not be integral. A value of 0 represents an ideal or symmetric distribution around the average and a value of greater than +1 represents conditions where a defect may be present. Provided the assumption that most RO membranes in an array are nominal is valid; in an RO system, outliers associated with defects are anticipated to cause a skew in the positive direction (i.e., most modules are defect free and are producing low conductivity and a small number are defective and produce a higher conductivity).

Figure 4-3 illustrates the characteristics of normal and outlier data groups. In a data set that exhibits a normal distribution the following statements can be made:

There is data that exceeds +3σ standard deviations (outliers).

The average of the data becomes greater than the median.

There is a positive skew in the data.

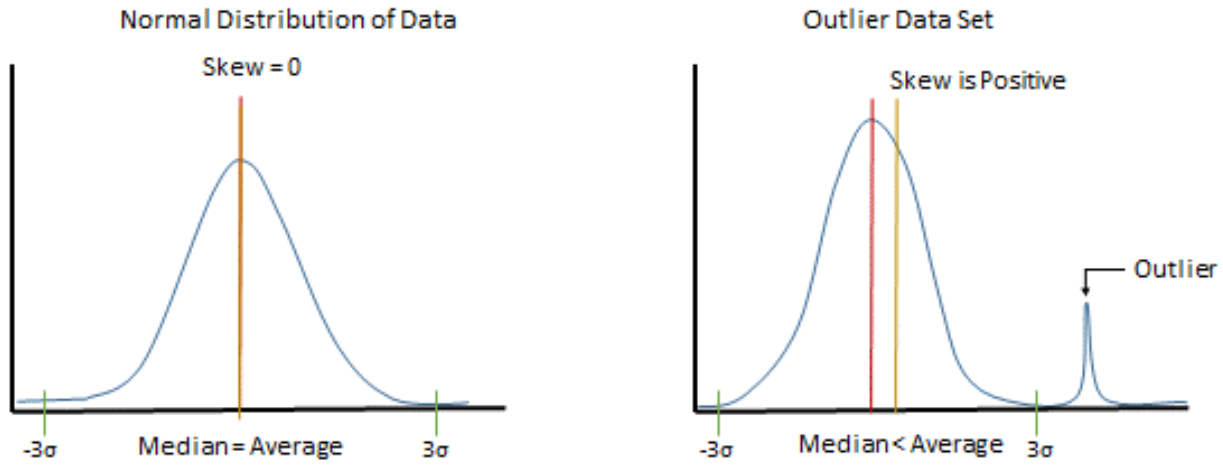

Figure 4-3. Characteristics of Normal and Outlier Data Sets.

4.3 Theoretical LRV Calculated from Profile Data

While LRV is typically calculated by equation 1-2 (see Section 1.2), permeate concentration can be more accurately calculated by using the flow-weighted average of the individual stage permeate concentrations instead of the overall permeate conductivity. For example, in a three-stage system the following would apply:

$$C_p = \frac{C_1 Q_1}{Q_p} + \frac{C_2 Q_2}{Q_p} + \frac{C_3 Q_3}{Q_p} \quad \text{(Equation 4-1)}$$

Where:

C_p = Concentration of the permeate (mg/L, µS/cm)

C_1 = Concentration of Stage 1 (mg/L, µS/cm)

C_2 = Concentration of Stage 2 (mg/L, µS/cm)

C_3 = Concentration of Stage 3 (mg/L, µS/cm)

Q_p = Membrane unit design capacity permeate flow (gpm)

Q_1 = Stage 1 Flow (gpm)

Q_2 = Stage 2 Flow (gpm)

Q_3 = Stage 3 Flow (gpm)

The underlying approach is to isolate the conductivity that is associated with diffusion that is not of pathogenic concern from conductivity that is associated with a defect that would be associated with an increased potential for pathogen pass-through. Calculating the LRV that would be associated with a system integrity defect, the mass flow rate for the permeate (Q_pC_p) can be rewritten in the following manner (equation 4-2).

$$Q_p C_p = Q_{diff}\, C_{diff} + Q_{defect} C_{defect} \quad \text{(Equation 4-2)}$$

Where:

Q_p= Membrane unit design capacity permeate flow (gpm)

Q_{diff} = Diffusive flow of constituents in the water matrix through the membrane (gpm)

Q_{defect} = Flow from an integrity defect associated with the smallest integrity test response that can be reliably measured (gpm)

C_p= Concentration of the permeate (mg/L, µS/cm)

C_{diff}= Concentration of Diffusion (mg/L, µS/cm)

C_{defect} = Concentration of the Defect (mg/L, µS/cm)

The above equation (Equation 4-2) separates the overall mass flow rate into its component (diffusion and size exclusion) related terms. Building on this approach to calculate the LRV, the dilution model approach that is described in the MFGM as equation 4-2 can be used. Shown below, where the Q_{breach} from the MFGM is the same as Q_{defect}.

$$LRV_{DIT} = log\left(\frac{Q_p}{VCF \bullet Q_{defect}}\right) \quad \text{(Equation 4-3)}$$

Where:

LRV_{DIT} = Direct integrity test sensitivity in terms of LRV (dimensionless)

Q_p = Membrane unit permeate flow (gpm)

Q_{defect} = Flow from the defect associated with the smallest integrity test response that can be reliably measured, referred to as the critical breach size (gpm)

VCF = Volumetric concentration factor (dimensionless)

In the dilution model, the flow through the defect can be calculated from the feed concentration that would pass untreated into the permeate. This condition would be associated with individual vessels determined to be outliers.

The equation above can be rewritten and solved for any given stage using the relationship that permeate flow is equal to the sum of diffusive flow and defect flow and would apply to the vessel outlier data points. The equation below is written for the vessels that were determined to be outliers and would be applied to each stage, as the diffusion for each stage is different.

$$\sum_{i=1}^{n} Q_{defect} = \frac{Q_{pn}C_{pn} - Q_{pn}C_{diffn}}{C_{defect} - C_{diffn}} \quad \text{(Equation 4-4)}$$

Where:

Q_{defect} = Flow from an integrity defect associated with the smallest integrity test response that can be reliably measured at stage n (gpm)

Q_{pn} = Membrane unit permeate flow at stage n (gpm)

C_{pn} = Concentration of the Permeate at stage n (mg/L, µS/cm)

C_{diffn} = Concentration of Diffusion at stage n (mg/L, µS/cm)

C_{defect} = Concentration of the Feed at stage n (mg/L, µS/cm)

The defect flow, Q_{defect}, can be calculated using the outlier vessel(s) and the feedwater conductivity for a given stage. If there are no stage outliers, the stage would be provided with the maximum LRV credit. For an array that contains outliers, the method requires an approach about the value that is associated with stage diffusion. The use of the median stage value is

suggested as a conservative approximation for the diffusive component C_{diff}, although that assumption can be challenged with the rationale that a higher standard deviation, up to +2σ, could be used based upon the premise that RO membranes that would fit within the normal distribution would also be integral. A value of +3σ is not believed to be appropriate, as all the data does not fit within the bounds of a normal distribution, thereby violating the underlying Central Limit Theorem. The implication is that the use of a higher (or lower) diffusion value would yield higher (or lower) LRVs. However, the magnitude of C_{diffn} is relatively small in comparison to the feed conductivity C_{defect}.

In the dilution model approach, the overall relationship that is associated with the VCF requires a discussion to understand in context with the operation of an RO unit, as the multiplier (Q_{defect} x C_{defect}) changes as feed water is concentrated through the system. The VCF, as discussed in Section 2.5 of the MFGM (USEPA 2005), was a term used to describe the increase in suspended solids at the membrane surface. In the case of RO, the increase in dissolved solids is a measured value that increases as well, and as such, the VCF is equal to 1. In some RO systems, interstage conductivity (i.e., the feed to a subsequent stage is the concentrate from the prior stage) is not monitored. In this instance, a pseudo VCF to account for TDS increase across the RO stages should be adopted based on flow and conductivity balance. Therefore, for systems intending on pursuing automated conductivity profiling as a means to verify LRV, online monitoring of interstage and concentrate conductivities is recommended. Although this value can be obtained from a mass balance equation using the feed conductivity and flows.

A simpler but less sensitive approximation is also possible. As previously stated, and using Figure 4-3 as a reference, one of the properties of normal versus outlier data is that the average of the group is higher than the median of the group. This characteristic of the conductivity profile data can be used as the basis of an LRV calculation. The approach is logical, as the conductivity that would be associated with an outlier defect such as $Q_{defect}C_{defect}$ (low volume, high concentration) would be diluted into the composite stage permeate, creating a measurable increase in the average conductivity for a stage, and the median would remain the same.

$$C_{defect} = \frac{(C_{p1} - C_{diff1})Q_1}{Q_p} + \frac{(C_{p2} - C_{diff2})Q_2}{Q_p} + \frac{(C_{p3} - C_{diff3})Q_3}{Q_p} \quad \text{(Equation 4-5)}$$

Where:

C_{defect} = Concentration of the Defect (mg/L, µS/cm)

C_{p1} = Concentration of the Permeate at stage 1 (mg/L, µS/cm)

C_{p2} = Concentration of the Permeate at stage 2 (mg/L, µS/cm)

C_{p3} = Concentration of the Permeate at stage 3 (mg/L, µS/cm)

C_{diff1} = Concentration of Diffusion at stage 1 (mg/L, µS/cm)

C_{diff2} = Concentration of Diffusion at stage 2 (mg/L, µS/cm)

C_{diff3} = Concentration of Diffusion at stage 3 (mg/L, µS/cm)

Q_1 = Stage 1 Permeate Flow (gpm).

Q_2 = Stage 2 Permeate Flow (gpm).

Q_3 = Stage 3 Permeate Flow (gpm).

Q_p = Membrane unit Total Permeate flow (gpm).

Where the average stage values are used for C_{pn} and the median stage value is used for C_{diffn}, and where *n* is the stage of the RO unit. The LRV for the defect (LRV_{defect}) can be written in the following manner. Unlike the individual vessel approach, the use of the median is an underlying requirement, as the combined permeate from all vessels is used as a basis for the calculation. This approach yields a more conservative approximation of the LRV.

$$LRV_{defect} = \log C_f - logC_{defect} \quad \text{(Equation 4-6)}$$

Where:

LRV_{defect} = Log Reduction Value due to the defect (dimensionless)

C_f = Concentration of the Feed (mg/L, µS/cm)

C_{defect} = Concentration of the Defect (mg/L, µS/cm)

For this calculation, the median conductivity of the stage is being used as the basis for $C_{diffusion}$. The median is normally less than the average and is less likely to yield infinite LRVs or mathematical errors, which would be assigned the maximum LRV as determined by the regulatory authority. This statement also applies if the average conductivity of the combined permeate is used as the basis for $C_{diffusion,}$ as the resulting LRV would be infinity. In the example below, train values are flow-weighted by stage.

Table 4-1. Example Calculation of LRV_{defect} Using Median Stage TDS.

Parameter	Symbol	Units	Stage 1	Stage 2	Stage 3	Train[1]
Feed TDS	C_f	mg/L				970
Log_{10} Feed TDS	$log_{10}C_f$	-				2.99
Average Permeate EC		uS/cm	24.6	63.4	275	59.6
Average Permeate TDS		mg/L	14.7	38.1	165	35.8
Median TDS		mg/L	14.4	36.3	163.8	34.9
Adjusted Permeate TDS[2] (average - median)	C_{defect}	mg/L	0.33	1.76	1.10	
Log_{10} Adjusted Permeate TDS	$Log_{10}C_{defect}$	-	-0.48	0.25	0.04	-0.2
Log Removal Value (defect)	LRV_{defect}	-				3.19[3]

Notes:

1. Train permeate parameters are flow weighted.
2. This is the effective residual TDS that is assumed to be due to a defect if the median is adequately represented by diffusion.
3. Use of the flow weighted $Log_{10}C_{defect}$ in place of Log_{10} average permeate - which includes both nominal diffusive conductivity as well as defect flow- increases calculated LRV from 1.4 to 3.2 in this example.

In the table above, there are no condition where the median is greater than the average; such a condition would result in the maximum LRV assignment for the stage. Based on the conductivity profiling data analysis in this study (six full scale systems) this method of LRV calculation has a practical upper limit for sensitivity of 5.0-log. A higher LRV can be obtained using the individual vessel calculation as a result of removing the median-diffusion related conductivity to calculate the defect-related conductivity.

4.4 Automation of Conductivity Profiling

Although valuable as a means of calculating higher LRVs, manual sampling for conductivity profiling is labor intensive and not practical for daily testing, particularly for large systems. Design and implementation of an automated conductivity profiling unit has proven to be successful as demonstrated by the system installed at the YVWD. A schematic of the unit is shown in Figure 4-4 and an image of the physical unit is shown mounted to the RO sample panel in Figure 4-5.

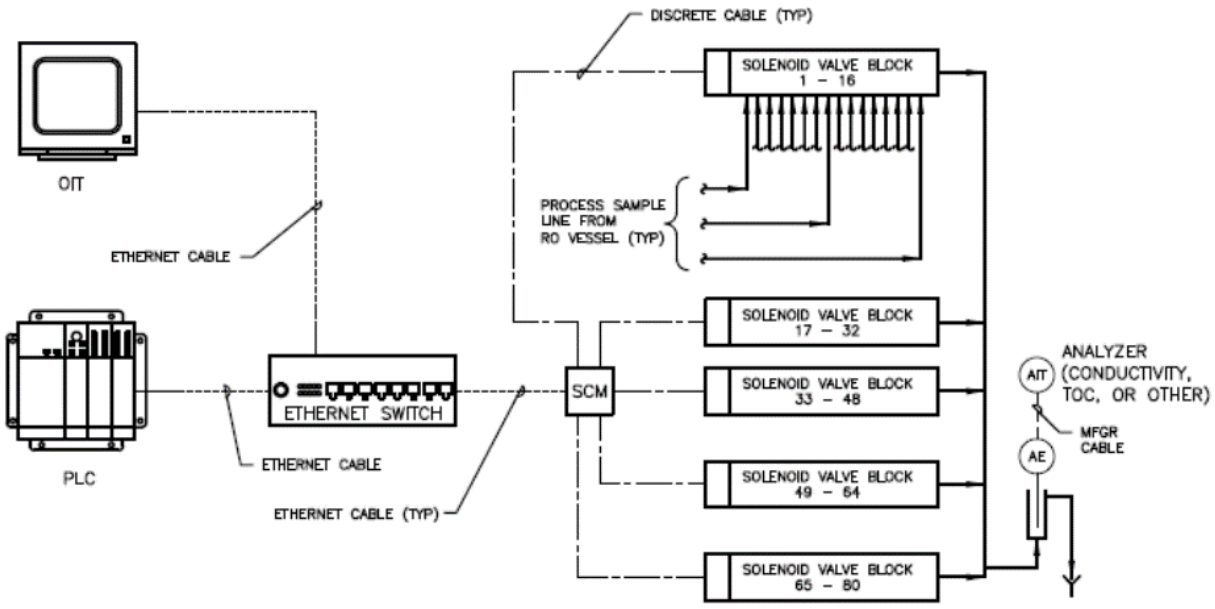

Figure 4-4. Schematic of Automated Conductivity Profiling Apparatus.

Figure 4-5. Automated Conductivity Profiling Apparatus at YVWD.

Each pressure vessel is connected to a solenoid valve and a total of 80 solenoids are connected to the automated conductivity profiling apparatus which includes spare solenoids available for a future expansion. While the system requires individual sample lines and solenoid valves, only a single conductivity probe is required for the system. It is important for the conductivity probe to be flushed before and after the analysis of each conductivity vessel sample. In order to establish an adequate amount of time for water to pass through the tubing, a "time-of-flight study" can be performed using a salt solution to set the sampling times by stage (as shown in Figure 4-6). At the YVWD, the first stage sampling period was set to 60 seconds and second stage sampling periods were set to 90 seconds due to the distance of the tubing from the sample panel.

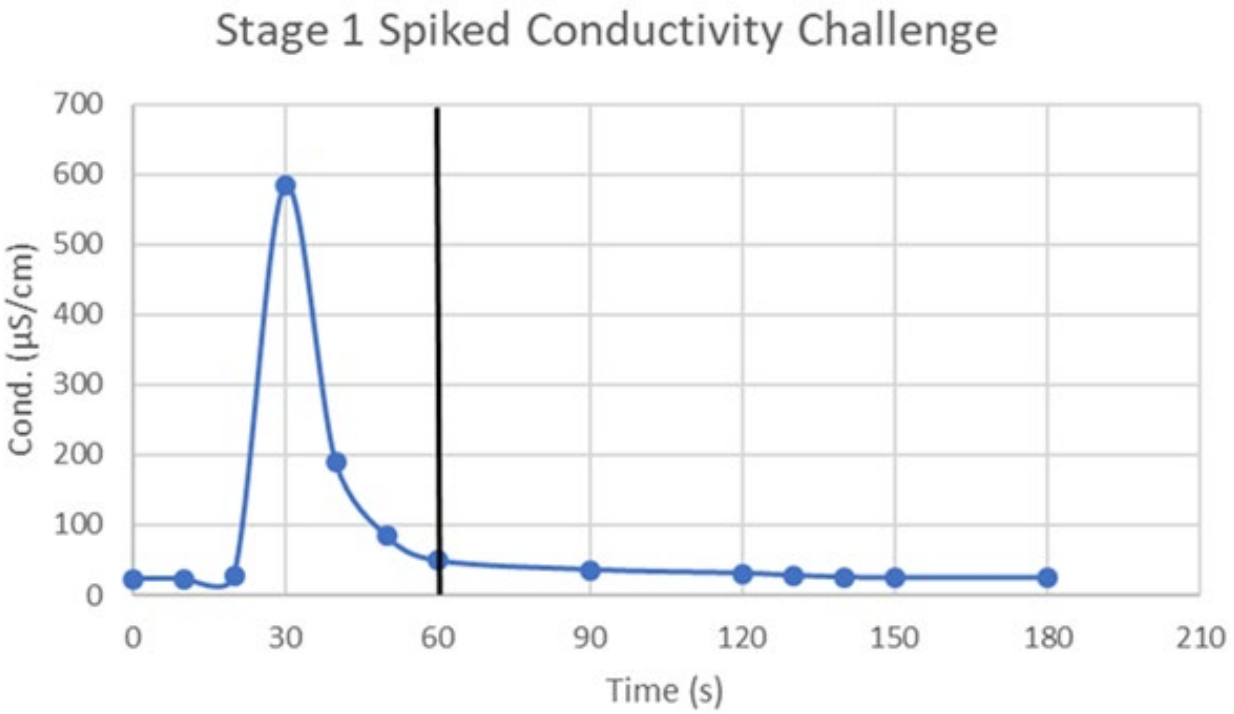

Figure 4-6. Stage 1 "Time-of-Flight" Study to Determine Solenoid Sample Duration.

4.4.1 Automated Profile Results

Another benefit of this system is that automated reports for each RO conductivity profile, including the conductivity of all pressure vessels, the test number, date, starting and ending time as well as supporting data for analysis can be automatically generated. Based on the collected raw data, the program calculates the average, standard deviation, median, and skew as well as the preestablished alarm limits (i.e., Average + 3σ, +1 Skew, and 1.5*Median). Each report also provides bulk stage flows and other salient information which can simply be extracted from the supervisory control and data acquisition (SCADA) system (Figure 4-7). The report also processes the data to provide the LRV calculated from the conductivity profile according to the approach detailed in Section 4.3. as a performance indicator. Conductivity Profile Reports maintain operators informed of the existence of outliers by underlining which pressure vessels may have integrity issues and potentially affect virus removal.

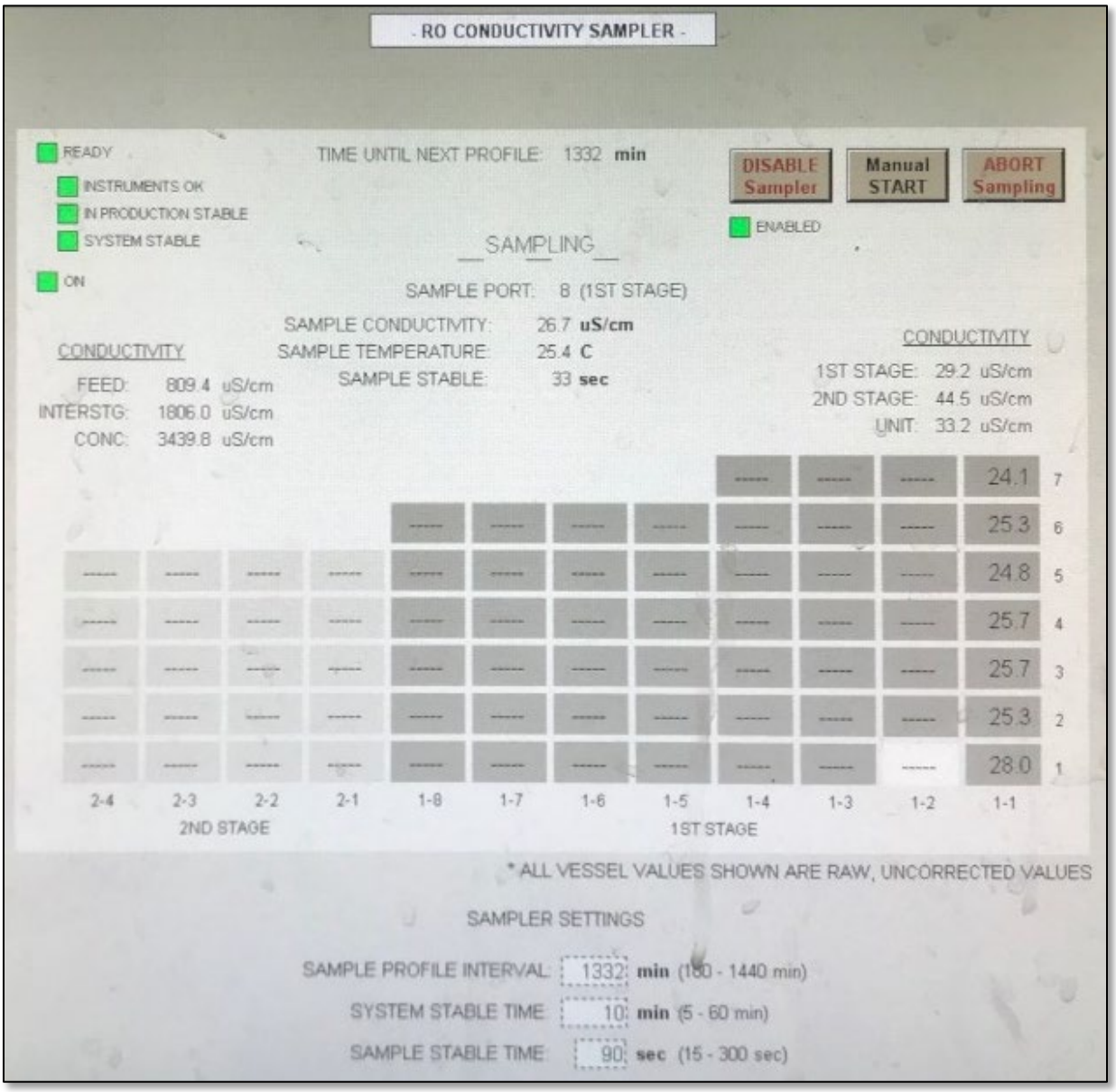

Figure 4-7. Example of RO Automated Conductivity Profile SCADA Screen.

Figure 4-8 shows an example conductivity profile report which does not have any warnings or alarms. Warnings would be shaded in yellow while alarms would be shaded in red. Alarms occur after two or more warnings are detected within a single vessel. There are three warning and alarm limits that are calculated for each report and are a result of the conductivity values obtained for that particular profile. In other words, the value for each alarm itself will be different for each report. During the study, alarms and warnings occurred at low frequencies, and overall, the RO conductivities were stable during this period.

Yucaipa Valley Water District

RO CONDUCTIVITY PROFILE REPORT

START TIME: 3 / 30 / 2020 05:17:15

END TIME: 3 / 30 / 2020 07:18:47

Average Feed Conductivity: 752.8 µS/cm

TEST NO: 2137

				24.8 52	23.3 46	23.3 40	22.6 34	23.4 27	23.2 20	24.7 13	24.9 6	
42.7 72	34.3 67	40.3 62	36.4 57	23.9 51	23.4 45	23.0 39	21.9 33	23.8 26	24.5 19	25.0 12	23.6 5	
37.3 71	43.0 66	37.2 61	39.8 56	24.3 50	24.5 44	25.7 38	24.8 32	23.7 25	25.2 18	25.2 11	25.3 4	
40.3 70	39.2 65	35.6 60	39.1 55	24.3 49	23.9 43	23.7 37	23.9 31	23.9 24	24.4 17	26.1 10	25.5 3	
37.4 69	39.5 64	38.9 59	40.3 54	22.6 48	24.7 42	25.4 36	24.6 30	27.0 23	24.4 16	23.5 9	24.9 2	
35.7 68	35.3 63	35.5 58	38.3 53	22.9 47	24.2 41	25.7 35	24.3 29	23.3 22	25.0 15	27.4 8	27.8 1	
SECOND STAGE				FIRST STAGE								

STAGE 2 CURRENT RESULTS

AVERAGE	38.3 µS/cm
STANDARD DEV.	2.4
MEDIAN	38.6 µS/cm
SKEW	0.22

STAGE 1 CURRENT RESULTS

AVERAGE	24.3 µS/cm
STANDARD DEV.	1.2
MEDIAN	24.3 µS/cm
SKEW	0.67

STAGE 2 CURRENT ALARM LIMITS

AVG + (3 x STDEV)	45.4
MEDIAN + 50%	57.9

STAGE 1 CURRENT ALARM LIMITS

AVG + (3 x STDEV)	28.0
MEDIAN + 50%	36.4

STAGE 2 VESSEL SUMMARY

VESSEL WARNING COUNT:	0
VESSEL ALARM COUNT:	0
AVG FEED CONDUCTIVITY:	755.9 µS/cm
AVG FEED BRINE COND:	2,276.4 µS/cm

STAGE 1 VESSEL SUMMARY

VESSEL WARNING COUNT:	0
VESSEL ALARM COUNT:	0
AVG FEED CONDUCTIVITY:	751.5 µS/cm
AVG FEED BRINE COND:	1,090.5 µS/cm

STAGE 2 FLOW

PERMEATE:	296 gpm
CONCENTRATE:	284 gpm

STAGE 1 FLOW

PERMEATE:	854 gpm
CONCENTRATE:	588 gpm

STAGE 2 LRV VALUES

LRV:	5.00
BETA:	100,000
PERCENT REMOVAL:	99.9990%

SYSTEM LRV:	4.71
SYSTEM PERCENT REMOVAL:	99.9981%

STAGE 1 LRV VALUES

LRV:	4.33
BETA:	21,553
PERCENT REMOVAL:	99.9954%

CONDUCTIVITY & TEMPERATURE (AVERAGE)

FEED:	752.8 µS/cm
	68.0 °F
INTERSTAGE:	1,669.6 µS/cm
CONCENTRATE:	3,197.9 µS/cm
STAGE 1:	24.9 µS/cm
STAGE 2:	39.8 µS/cm

Figure 4-8. Example Automated Conductivity Profile Report from YVWD.

4.4.2 Automated Profiler Workflow

For the YVWD, unit test results are forwarded electronically after the conductivity profile is complete. The conductivity profile report is emailed, and data is extracted using an Excel Macro program. The system LRV is calculated using a conservative flow-weighted feed-to-permeate calculation for regulatory purposes (as described in Section 4.3). Graphical analysis is reviewed with observations of conductivity range divergence from averages as well as outliers which will be shown through warnings and alarms. Gathered data can be used to graphically illustrate trends and the overall conditions of the membranes (i.e., fouling and/or scaling issues). Given that the system is automated, further programing could be conducted to assign new tags to log calculated LRVs and system warnings into the historian. At the Water Corporation of Western Australia, a similar automated approach is applied to profile the vessels with the output directed to SCADA. However, the WCWA system is scheduled such that conductivity profiling is only used for monitoring of array integrity every two weeks and quantitative calculation of the outputs is not presently automated.

4.4.3 Automated Profiler Alarms and Warnings

Conductivity profiling provides information as to the extent of potential damage, as well as its location. Although the principle focus of this evaluation is to document the sensitivity of the LRV able to be demonstrated by profiling, the outputs also serve as guide to key statistics that are recommended to be quantified on each profile as a fault detection diagnostic tool.

Table 4-2 contains a summary of the statistics of the permeate conductivity per vessel from the WRWRF during the months of July 2019 through March 2021. These values are averages of all 4,768 automatic conductivity profiles during the testing period, including the following:

- The average of averages of the permeate conductivity per vessel,
- the average standard deviation of the permeate conductivity per vessel,
- the average of the median values of the permeate conductivity distribution,
- the average skew, and
- the average values of the resulting limits to which each pressure vessel was checked against.

The YVWD data summarized in Table 4-2 is positively skewed, exhibited as an average skew value of +0.7 for Stage 1. The +1 Skew alarm was triggered on only 14% of the tests for Stage 1 and was triggered on 4.4% of the tests for Stage 2. The Average + 3σ alarm was triggered 35.8% of all the tests for Stage 1.

Part of the intent of using three different alarm types was to identify what types of alarms are more useful for operational practice and better understand the nature of systems in actual operation. This information can be subsequently used to establish appropriate alarm limits based on simplified criteria which notifies operators of a potential integrity breach and minimize the occurrence of “false positives”.

For example, if an array has a natural positive skew, there would be a higher incidence of alarms that may occur. Similarly, the Central Limit Theorem test is based on the underlying assumption that the data fits the binomial probability distribution, which, by its definition, has a skew of zero. The Yucaipa array exhibited a positive skew, with increased "false positive" notifications, particularly with stage 1 membranes.

Table 4-2. Summary of Warnings and Alarms from 4,768 Automated Conductivity Profiles Conducted at YVMD between July 2019 - March 2021

Statistic/Parameter	Units	Stage 1	Stage 2
Average	µS/cm	30.5	48.7
Standard Deviation	µS/cm	1.3	2.8
Median	µS/cm	30.4	48.4
Skew	-	0.7	0.6
Limit: Average + 3σ	µS/cm	34.6	57.2
Limit: 1.5*Median	µS/cm	45.7	72.6
Percentage Warning Average + 3σ	%	35.8	0
Percentage Warning 1.5*Median	%	0.19	0
Percentage Warning +1 Skew	%	14	4.40
Percentage Alarm Two warnings	%	14	0
Percentage Alarm Three warnings	%	0	0

4.4.4 Sensitivity Analysis of Individual Vessel Performance

The third test that involves checking each vessel against the value of 1.5*Median remained inactivated throughout all tests for both stages, as shown in Table 4-2. It may be plausible that the limit of 1.5*Median surpasses any reasonable values and that such approach is too lenient for this group of membranes. In order to determine a more accurate representation of an alarm condition, a sensitivity analysis was carried out to explore how different benchmarks based on the median would correlate to the data. Therefore, in addition to comparing each vessel to the 1.5*Median we calculated the following:

- The median plus 20% of the median (1.2*Median),
- the median plus 30% of the median (1.3*Median), and
- the median plus 40% of the median (1.4*Median).

Table 4-3. Evaluation of Vessel Permeate Conductivity at Different Potential Benchmarks for Array Integrity Based on Distance from the Median.

Date	Median	Maximum Conductivity	Skew	1.2*Median	1.3*Median	1.4*Median	1.5*Median
1/6/2020	20.8	25.6	1.7	25.0	27.1	29.1	31.2
12/30/2019	21.6	26.2	1.5	25.9	28.1	30.3	32.4
12/23/2019	22.6	26.9	1.4	27.1	29.3	31.6	33.9
12/16/2019	19.4	23.4	1.4	23.2	25.2	27.1	29.0
11/28/2019	17.3	20.6	1.1	20.7	22.5	24.2	25.9

Table 4-3 shows the results of the sensitivity analysis when comparing several dates in which vessel 33 had the maximum permeate conductivity which triggered the Average +3σ alarm, and the +1 skew alarm. It was observed that for two of the listed tests (1/6/20 and 12/30/19), besides exceeding the other two alarms, the maximum conductivity exceeds the 1.2*Median, and the other three tests would also nearly trigger the warning of a vessel performing higher than the 1.2*Median. In the case of the 1.3*Median, no vessel exceeds such value or the 1.4*Median.

Considering that warnings should accurately diagnose an issue without causing unnecessary alarms, it might be appropriate to establish the 1.3*Median as a conservative or more realistic approach rather than the initial test of 1.5*Median. These alarm conditions would be established as site-specific baseline conditions and would be the basis for demonstrating the justification for granting increased LRV credit for the system as part of a regulatory approval process (see Chapter 9).

4.4.5 Confirming Outliers are Integrity Issues

Data evaluation of outlier vessels at YVWD from November 2019 through March 2020 are depicted in Figure 4-9, which illustrates the ratio of the vessel permeate conductivity to the stage Average + 3σ alarm. A value below 100% implies the Average +3σ was not triggered. Vessel 33 (1-5-5) began to increase above 100% on November 26th, 2019, hence triggering the Average + 3σalarm. For the following three months, Vessel 33 continuously had alarms due to the skew being greater than +1 and for being above the Average + 3σ. Vessel 1 and Vessel 8 rarely triggered the Average +3σ alarm but were nominally higher than the other vessels (as shown in Figure 4-9).

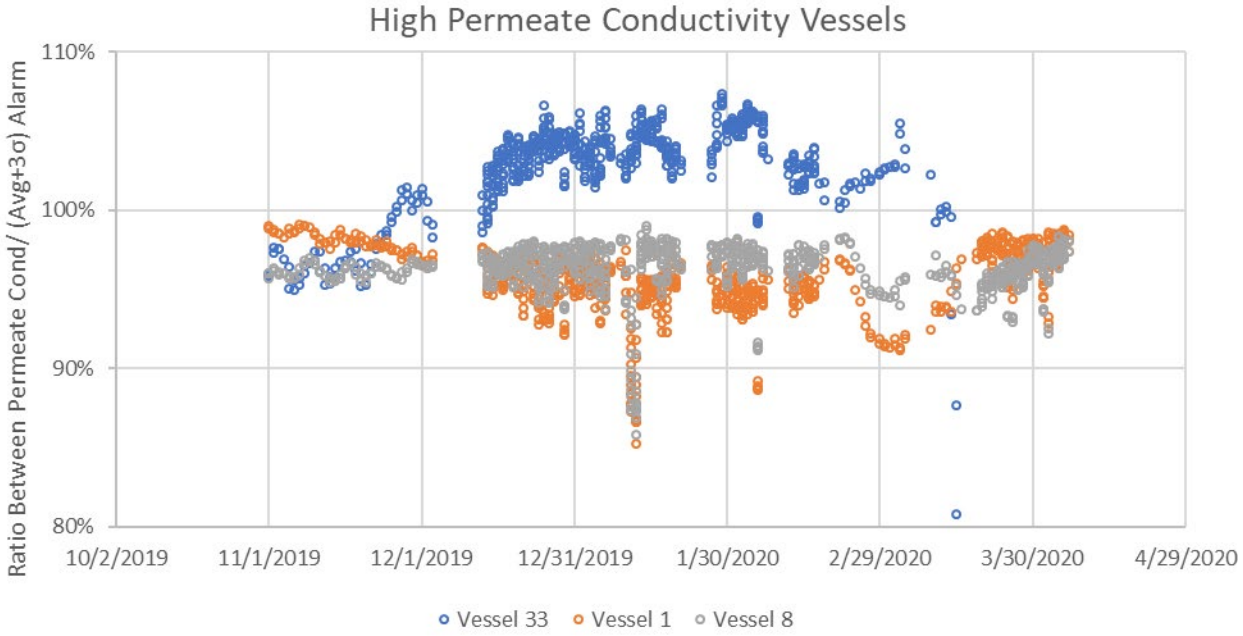

Figure 4-9. Ratio of the Vessel Permeate Conductivity to the Average + 3σ Alarm.

Values greater than 100% mean alarms were triggered. The automated conductivity profiling system provided the ability to identify potential integrity failure in Vessel 33 as indicated by the

upward trending of results with time. After noting the increasing trend in conductivity profile results, Vessel 33 was probed on February 18, 2020, and it was determined that integrity issues existed near the tail element of that particular vessel. A 30-foot long, 1/4-inch diameter polyethylene tube was inserted into the permeate carrier of Vessel Number 1-5-5 to take conductivity measurements every 20 inches. In order to avoid erroneous readings from the handheld device, permeate conductivity readings were first recorded at the lowest concentrations. For this reason, the tube was inserted all the way through until it reached the lead-element end adapter. At that point, the probing tube was extracted 20 inches at a time and another conductivity measurement was recorded. The conductivity profile of Vessel Number 1-5-5 is illustrated in Figure 4-10, with the lead element having a permeate conductivity of approximately 22 µS/cm and increasing by the tail end to 32 µS/cm.

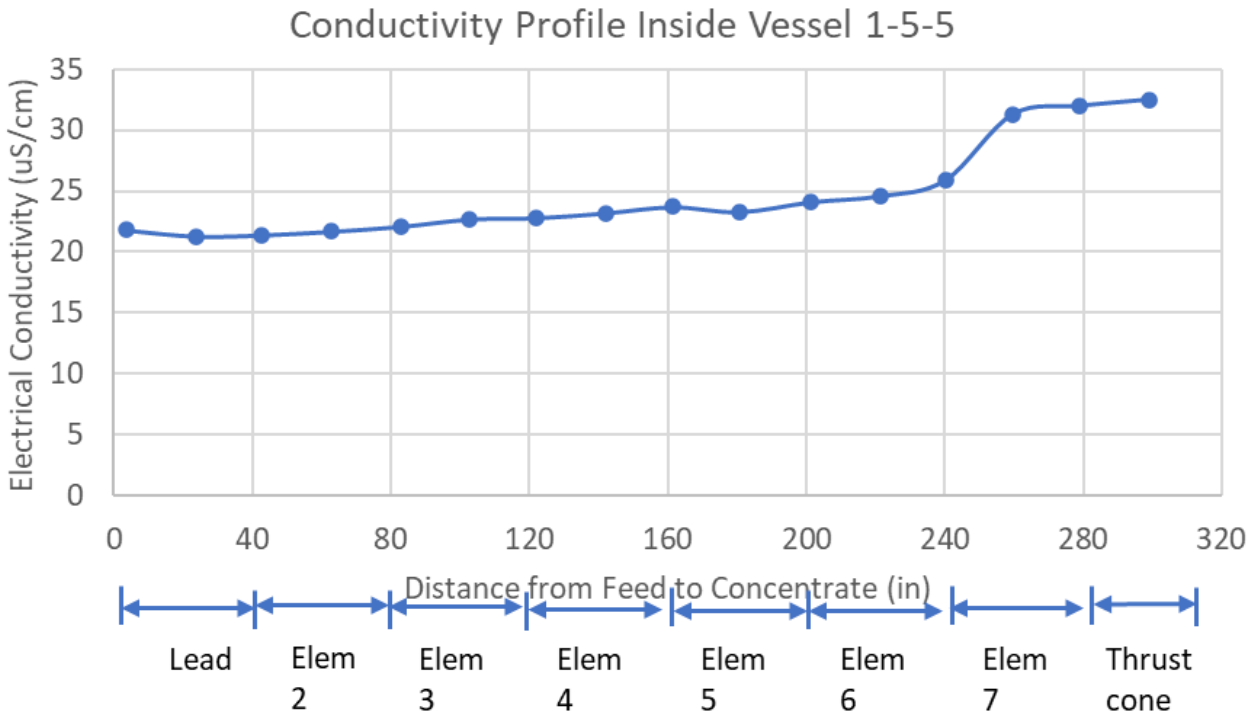

Figure 4-10. Permeate Conductivity Measurements Inside Pressure Vessel Number 1-5-5 During Probing.

On March 17, 2020, Wochholz operators removed the tail element of Vessel 33, inspected the element, the interconnectors, and end adapter. Pictures of the tail element, shown below (Figure 4-11), demonstrate that the element was cracked around its entire circumference. The cracked element was replaced and a new element, which had been stored at Wochholz, was installed as the lead element for vessel 33. After replacement, the performance of Vessel 33 improved dramatically and began to produce water with much lower permeate conductivity.

The cracked-tail element was sent to Avista Technologies for further testing and analysis. Wet tests were performed (the feed pressure set at 225 psi with a recovery of 15%) which showed slightly lower salt rejection than manufacturer specifications. Figure 4-12 illustrates the two vacuum tests which shows the rate of pressure decay over time. A vacuum decay test is used to detect leaks or to confirm the mechanical integrity of an element. The element failed the vacuum test and was unable to hold a steady vacuum, with values beginning at 21 millimeters (mm) of mercury (Hg), decreasing to 3 mm Hg, and at 4 mm Hg after only 120 seconds.

The investigation into the performance of Vessel 33 served as evidence that the Average +3σ alarm had accurately warned of integrity issues regarding this vessel for the last three months.

Figure 4-11. Cracked Tail element of Vessel 33.
The failure was identified by conductivity profiling (see Figure 4-9) and the specific element located by probing.

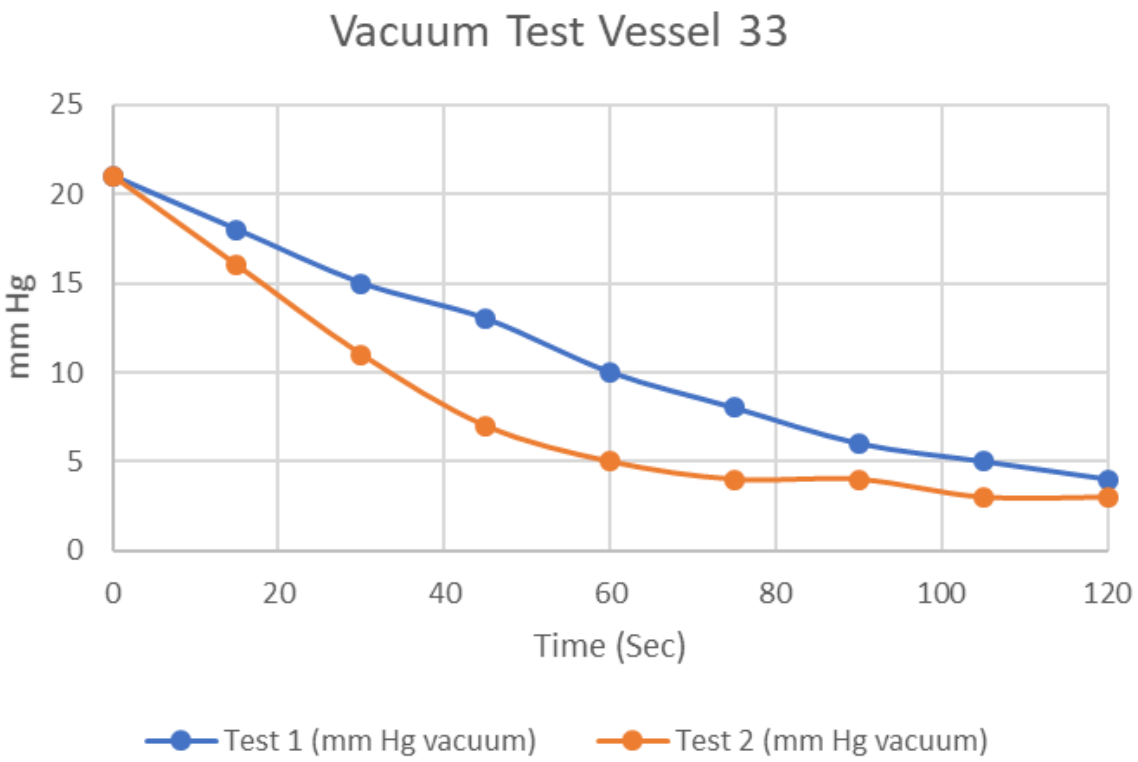

Figure 4-12. Results of Vacuum Decay Test of Vessel 33.
The rapid decay in vacuum indicates integrity failure.

4.4.6 Long Term Monitoring of Conductivity Profiles

Observations and analysis of the YVWD RO continued for more than 1.5 years after initial installation. Additional monitoring involved the performance of the RO unit as a whole. Figure 4-13 shows the average conductivity for the RO feed as well as the permeate conductivity for both stages. Seasonal changes are experienced as a higher feed conductivity and lower permeate conductivities. Even though the feed conductivity was higher, the lower feed temperature in colder months slowed diffusion rates across the RO membrane, resulting in less salt transport and a lower conductivity in the permeate. This is an important aspect for regulators to recognize the changes in permeate conductivity during summer months, which although display higher conductivity (due to faster diffusion rates, do not necessarily reflect a higher passage of pathogens.

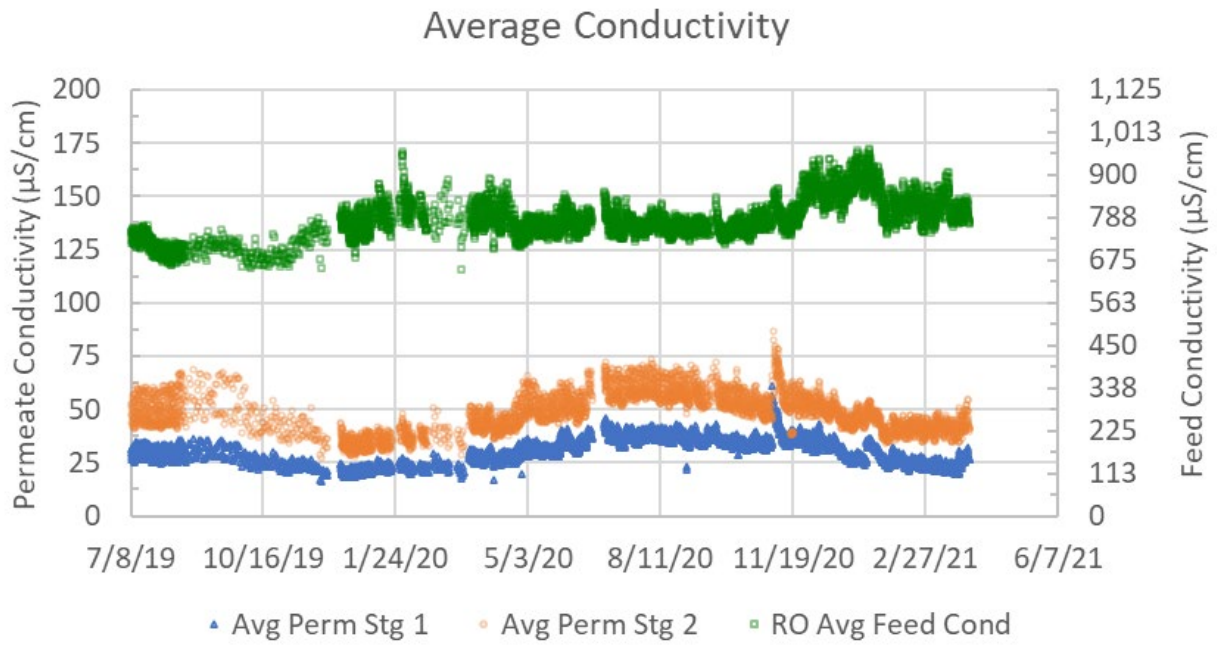

Figure 4-13. Long Term RO Feed and Permeate Conductivity for the YVWD RO.
Note the seasonal variation in feed and also permeate conductivity due to temperature impacts.

Figure 4-14 depicts the permeate conductivity of Stage 1 and its two alarms Average +3σ and 1.5*Median. A close overlap between the permeate conductivity for Stage 1 and the Average + 3σ alarm is shown which confirms how realistic this alarm is with respect to the group, whereas the 1.5*Median alarm is consistently much higher. Note that the higher values seen on the graph in September 2020 and November 2020 relate to the virus challenge testing that was being performed at the facility on those dates.

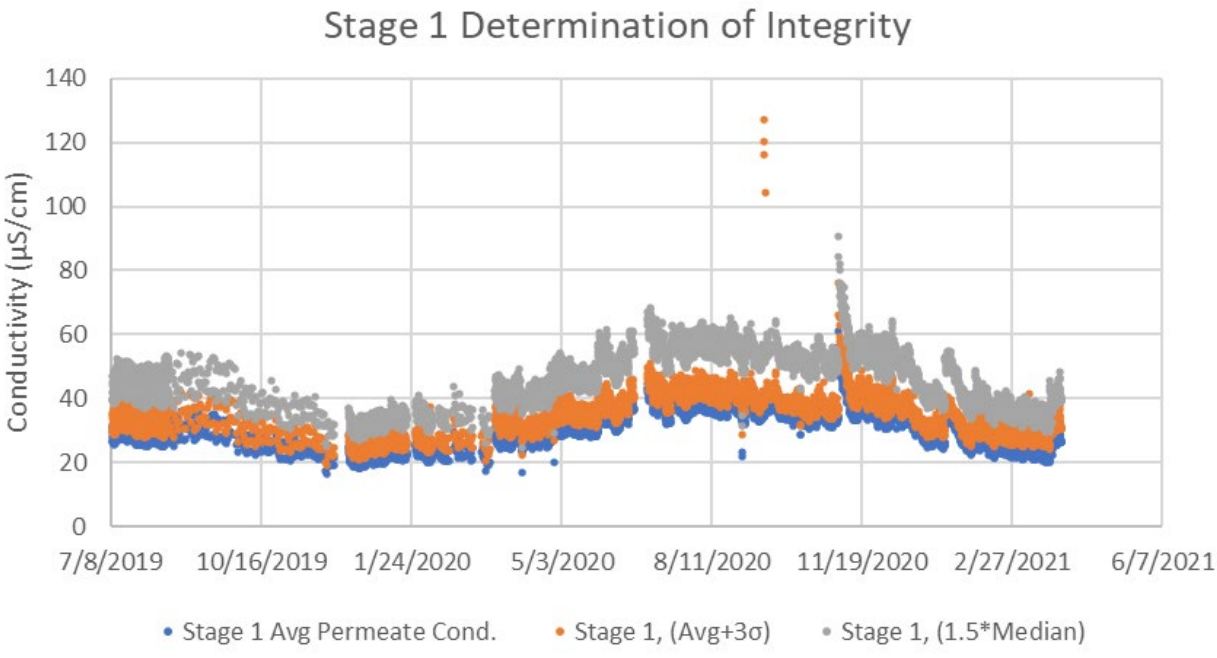

Figure 4-14. YVWD Stage 1 Average Permeate Conductivity, Average +3σ, and 1.5*Median.

Figure 4-15 shows the permeate conductivity for Stage 2 and its respective alarms. A similar trend to Stage 1 was observed, in which the 1.5*Median exceeds both the permeate conductivity and the Average + 3σ alarm, rendering the 1.5*Median alarm less practical.

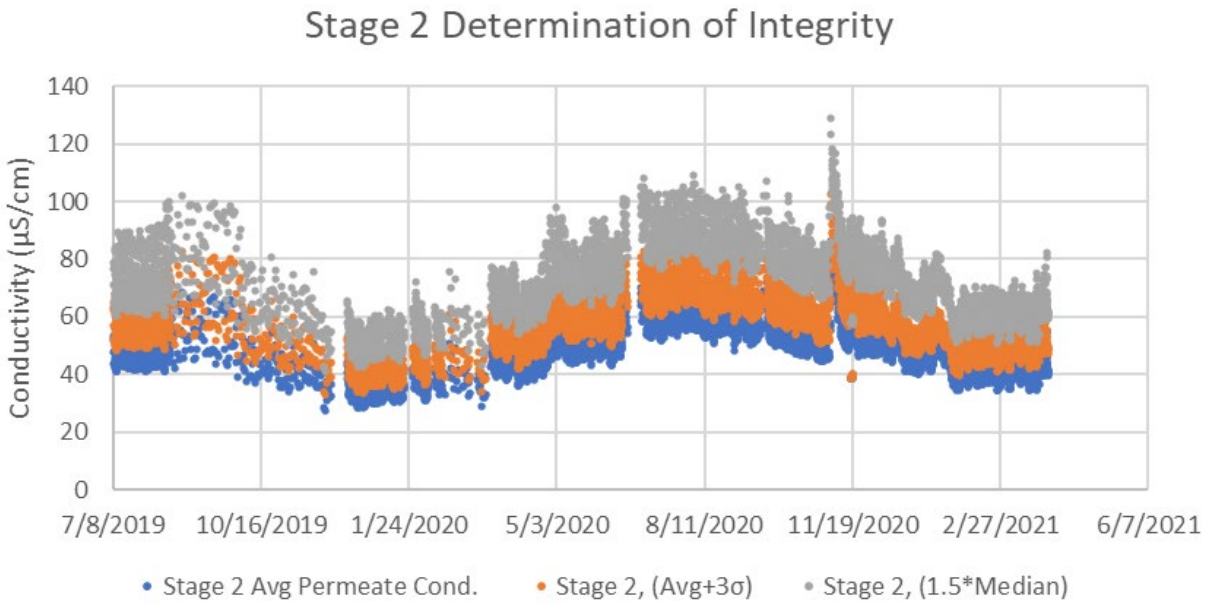

Figure 4-15. YVWD Stage 2 Average Permeate Conductivity, Average +3σ, and 1.5*Median.

The typical direct LRV of conductivity (RO Feed to Combined Permeate) at YVWD was close to 1.4-log. Figure 4-16 shows the LRV calculated based on conductivity profiling for each stage consistently demonstrating at least 2.8-log.

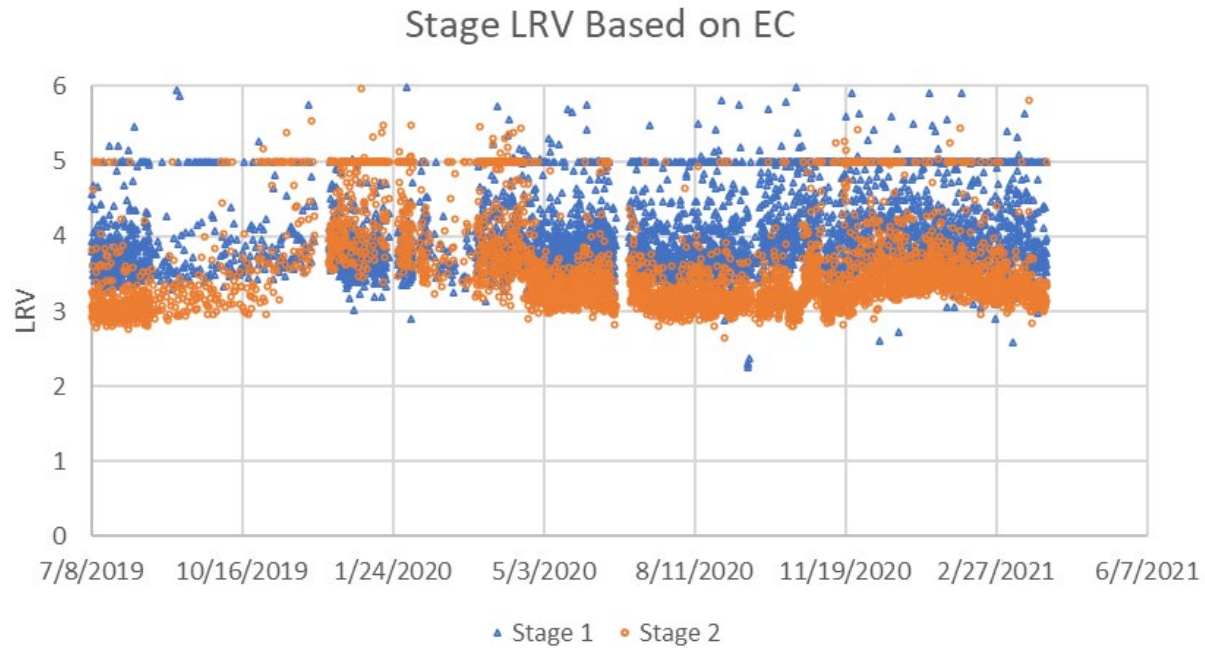

Figure 4-16. YVWD Stage 1 and Stage 2 LRV Calculated from Conductivity Profiling Results Demonstrating more than 2.8-Log For each Stage.
Note that combined RO feed to permeate LRV was close to 1.4.

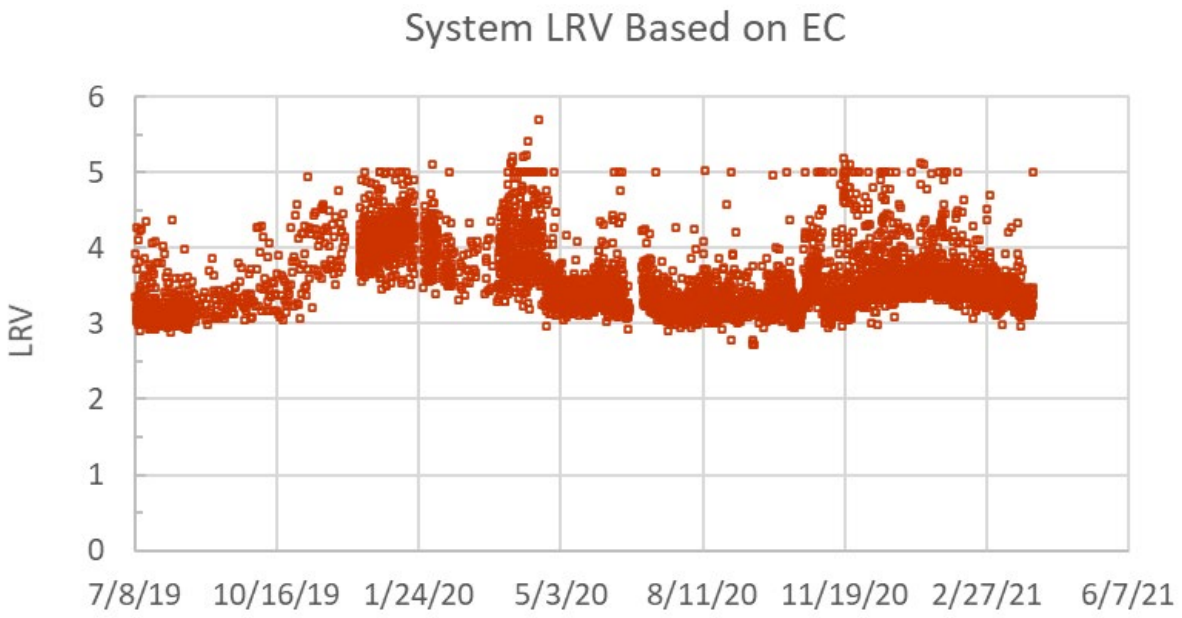

Figure 4-17. Overall System LRV Calculated from Conductivity Profiling at YVWD Demonstrating more than 3-log. Seasonal variation in profiling LRV is observed. Note that combined RO feed to permeate LRV was close to 1.4 indicating a sensitivity improvement by conductivity profiling of more than 1.6-log.

Figure 4-17 illustrates the LRVs for the system based on EC, which shows the system obtains approximately 3-log consistently, with certain performances capped at the maximum of 5-log. Note summer LRVs are lower than winter LRVs due to the lower diffusion rates across the membrane with colder feed temperatures. Future method development to introduce temperature normalization into the calculation may further improve sensitivity of the conductivity profiling technique.

4.5 Sensitivity of Conductivity Profiling at Other Facilities

Currently, operating facilities perform conductivity profiles as part of routine operations. Criteria for acceptable operation is normally established in the individual operating plans, and as such there will be differences in profile results that initiate further investigation for a potential integrity issue. This point aside, there is value in reviewing the data that is routinely collected to identify if the methodology described could work within a regulatory framework as a means to increase the LRV credited to a system.

In addition to the conductivity profiling results from YVWD, results were also available from other facilities including:

- Orange County Water District (OCWD):
  - A random subset of these results is discussed in this chapter and a more detailed discussion is in Chapter 5.
- Water Corporation of Western Australia (WCWA):
  - These results are shown in detail in Chapter 7 and a subset from RO train 1 is reproduced here to highlight relative sensitivity improvement compared to direct measurement of conductivity.

    - WCWA results are from an automated profiling arrangement which is conducted every two weeks for the purposes of qualitative assessment of array integrity.
- Site 1 - Northern California:
    - Overall System Recovery 80%.
    - 2 - Stage 52:28 Arrays.
    - Results from multiple RO arrays shown.
- Site 2 - Southern California:
    - Overall System Recovery 80 - 85%.
    - 2 - Stage 68:30 Arrays.
    - Results from multiple RO arrays shown.
- Site 3 - Southern California:
    - Primary RO Recovery 85%.
    - 2 - Stage 72:30 Primary RO Arrays.
    - Results from multiple primary RO arrays shown.
    - 3rd Stage Secondary RO.
    - Note - 3rd Stage RO could not be flow weighted with Primary RO results due to profiles conducted on separate days. Results are reported for the Primary RO only.
- Site 4 - Northern California:
    - Overall System Recovery 80%.
    - 2 - Stage 26:13 Arrays.
    - Results from multiple RO arrays shown.

A combination of conductivity profiling LRVs for the whole array from the sites listed above, relative to the direct LRV calculated from RO feed to combined RO permeate is shown below in Figure 4-18. Most sites displayed conductivity profiling LRVs over 3-log. When compared to direct LRV calculation, the conductivity profile LRV appeared to increase sensitivity by 1 to 2-log. Site 2 contained conductivity profiling LRVs between 2.5 - 3.0-log. For each of these LRVs, that were less than or equal to 3.0-log for Site 2, there were 2 or more vessels with at least one stage, typically the second, where the vessel conductivity was greater than 1.3 times the median suggesting potential integrity failure.

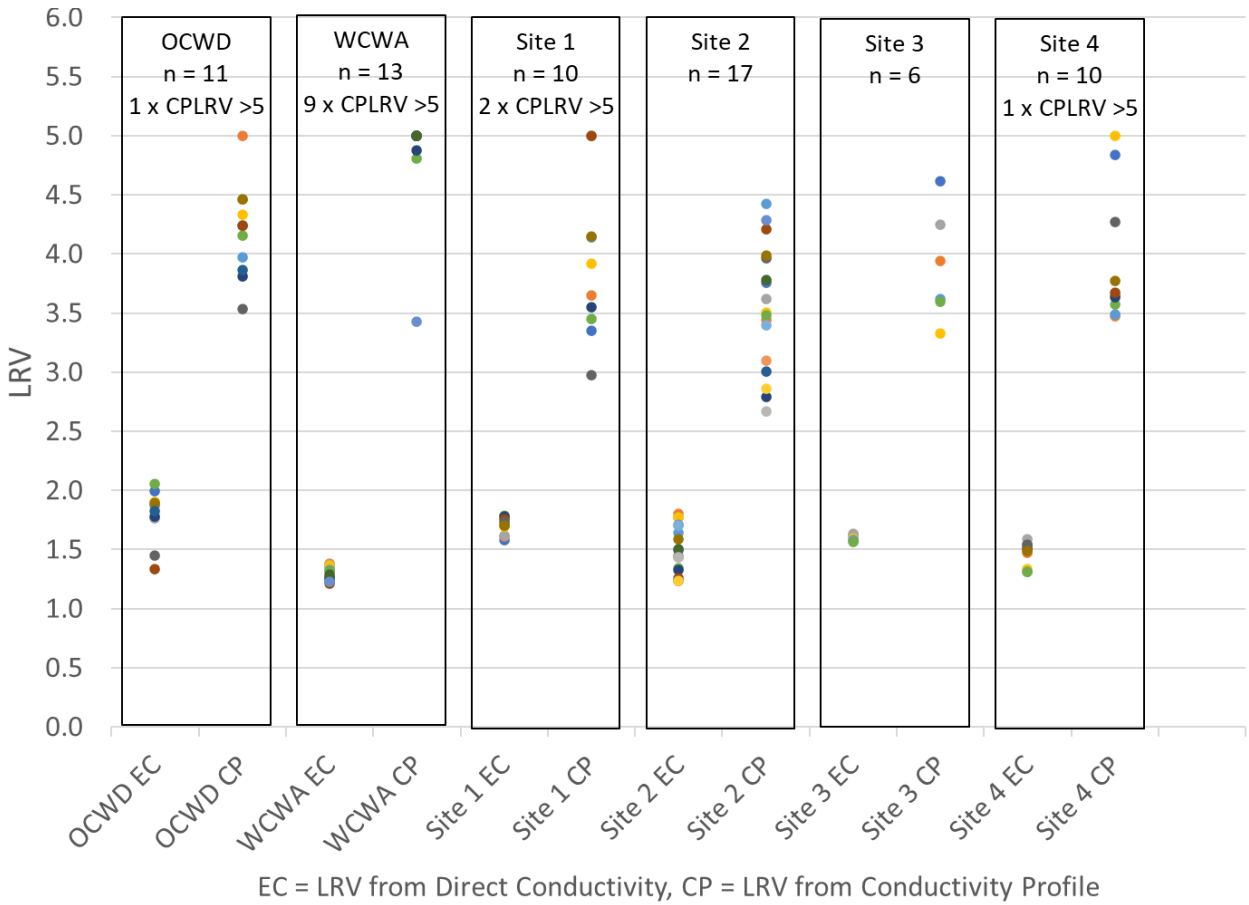

Figure 4-18. Comparison of Conductivity Profile LRVs (CP) vs Direct Conductivity LRVs (EC) from Multiple Sites. Typically, CP LRVs were greater than 3-log, representing a 1 - 2-log increase in LRV sensitivity compared to the direct LRV calculation method. Dots of the same color at the same site are from the same data set.

4.6 Considerations for Applying Conductivity Profiling for Increased RO Credit

This section contains the research teams recommendations for adoption of an RO LRV Credit based upon conductivity profiling.

4.6.1 MFGM Criteria for DIT

As stated previously, the MFGM established frequency, sensitivity, and resolution criteria for a monitoring technique to qualify as a DIT. This section outlines the research team's justification for conductivity profiling to meet the MFGM criteria.

4.6.1.1 Frequency

To meet the daily DIT frequency criteria, each array at a plant intending to claim credit for the RO would need to be profiled at least once per day with statistical summaries and calculations performed. This could be performed manually, however, development of an automated system would likely be the most appropriate for meeting frequency goals. The automated system at YVWD is able to perform a complete array profile in approximately two hours which means that 12 similarly sized arrays (each 2 mgd capacity) could be tested within one day with a single profiling system. For larger facilities, multiple automated profiling systems may be required.

In between DITs with conductivity profiling, a CIIM would be required. It is proposed that continuous online monitoring of the RO feed and combined permeate from an individual array is sufficient for the purposes of CIIM. Control criteria to divert and trigger additional DIT could be set for the combined permeate conductivity according to the alarm limits proposed for array integrity, e.g., 3 times the standard deviation or 1.3 times the median.

4.6.1.2 Resolution

Resolution refers to the smallest defect able to be identified with a DIT. In the MFGM, it is stated that "the effective size of the marker can be established through any accepted methodology such as size distribution analysis of particulate markers or estimation techniques based on the molecular weight and geometry of molecular markers" (MFGM pages 5 to 6 of Section 4).

Conductivity profiling receives a conductivity signal. Conductivity originates from charged ionic species in the feed water. While there are some larger charged organic molecules such as humic acids, ionic salts are responsible for a majority of the conductivity in municipal wastewater. The hydrodynamic radii of common salts (e.g., sodium, chloride, calcium, carbonate, sulfate, etc. have been reported to vary between 0.268 angstroms for hydronium (H^+) to 2.7 angstroms for Cobalt(III) (Co^{3+}) (Kadhim and Gamaj 2020). 1 angstrom is 10 nm, so common ions will be between 0.027 to 0.27 nm. MS2 is 25 nm (Antony et al. 2012), approximately 100 times larger than the larger dissolved ions. *Cryptosporidium* oocysts are 3 µm (3000 nm), approximately 10,000 times larger than dissolved ions. Given that the smaller dissolved ions would be expected to pass through defects smaller than target viruses or protozoa, the research team suggests that the resolution of conductivity profiling should be considered appropriately conservative to represent both virus and protozoa removal. It is also important to note that some states (e.g., Colorado) already allow protozoa credit for RO.

4.6.1.3 Sensitivity

Sensitivity is the magnitude, or how much LRV is able to be demonstrated. Improved sensitivity of the conductivity measurement for compliance purposes can be obtained through monitoring of individual vessels and subtracting the naturally occurring diffusion. The sensitivity of conductivity profiling will, in some respect, be influenced by the precision of conductivity meters. However, this impact is anticipated to be accounted for by the inherent maximum cap of 5-log for virus assumed in the conductivity profiling LRV calculation. Based on the performance of the RO systems considered in this chapter, a realistic sensitivity for conductivity profiling is between 3-5 log. Although the conductivity profiling approach proposed will not be

possible to verify more than 5-log, it does appear to be able to demonstrate a higher LRV than a number of other surrogates tested. In addition, the conductivity profiling approach, requires simple and reliable instrumentation (i.e., conductivity and flow meters) making it comparatively cheaper and more practical than other surrogates. With a possible range of 3-5-log able to be demonstrated, conductivity profiling is more sensitive than direct monitoring of conductivity removal, which can typically only demonstrate 1-2-log.

4.6.1.4 Adoption of an LRV Credit

Consistent with the MFGM, we suggest that the LRV credit for an RO system is the lower of challenge test results or the DIT. If conducted, challenge test results for an intact RO system are presumed to achieve more than 5-log virus and protozoa removal. To that end, the LRV credit would be the conductivity profiling LRV used as a DIT. It should be noted that all the RO membranes systems used for the above discussions were operated with membranes that were not previously tested or certified for their ability to remove virus. The data from MS2 challenge tests support the premise that an RO system that exhibits an integral conductivity profile will be able to exhibit a high level of virus and protozoa removal.

The profile is specific to an array. For a system with multiple arrays, it is proposed that the overall RO system credit (based on conductivity profile) should be the instantaneous flow weighted average of the most recent conductivity profile LRV from each array in parallel.

4.6.2 RO System Design Limitations

There may be system design limitations that impact the feasibility or practicality of implementing automated conductivity profiling, including:

- New high recovery cyclic RO technologies - In systems such as pulsed flow or closed-circuit reverse osmosis the recovery is transient. This means that the volumetric concentration of feed changes over a cycle which will impact permeate conductivity. To obtain a representative conductivity sample from a vessel, the permeate conductivity would have to be sampled at a set recovery for each vessel to be able to assume a normal distribution. As a result, there would be more complexity with appropriately sampling and triggering conductivity profiling.
- Separated primary and secondary RO systems - Site 3 from Section 4.5 is an example of where a primary and secondary RO is installed, with the secondary RO acting as a 3rd stage for 1 or more blended 2nd stage concentrates to boost overall system recovery. The design rationale to separate the third stage is to achieve a baseline product flow and allow more frequent cleaning of the third stage. In this manner, the production capacity of the 1st and 2nd stage are decoupled from the third stage (which produces a lower volume). All permeate from the primary and secondary systems is still blended and, as such, a flow weighted LRV calculation would have to take into account the contribution of both primary and secondary RO. This would likely necessitate more than one conductivity monitoring point and careful mapping/pairing of primary trains that contribute to the secondary train in order to appropriately group vessels actually contributing to LRV.
- Minimum Vessel Requirement - The LRV and alarm validation approach proposed via conductivity profiling requires representative median and average values. This necessitates

at a minimum 3 vessels per stage. For a 2-stage system, the minimum array size would be 6:2. However, to obtain meaningful statistics on the final stage, more than six vessels is recommended. The minimum size requirement will be a barrier to the use of conductivity profiling for smaller demonstration systems. However, typical full-scale systems treating 1 mgd or more would be anticipated to have array sizes of approximately 18:12:6 for a 3-stage system or 20:10 for a 2-stage system. To that end, a three stage RO system may need to be close to 1 mgd to have enough vessels to calculate a median and average for the third stage with n = 6. A 2-stage system for 1 mgd would have approximately 10 vessels in the third stage, meaning that conductivity profiling to verify LRV might be possible on smaller volume 2-stage skids. As noted in Section 4.4.3, smaller systems could make use of general integrity rules, e.g., 1.3 times the median, but it may not be appropriate to calculate an LRV.

4.6.3 Technique Limitations

4.6.3.1 Median Greater Than or Equal to Mean

The proposed methodology for calculating an LRV from a conductivity profile would cap reduction at a maximum value of 5-log if the median was greater than or equal to the mean. If the median was greater than the mean for an RO system, this could be interpreted as a system in which a majority of elements are producing lower quality water than a minority producing higher quality water. If a system was producing primarily poor-quality water, use of a secondary surrogate (such as a marker or MS2 testing) may assist in determination of whether array removal is adequate. Alternatively, the performance of the array could be checked against RO anticipated salt passage based on manufacturer specific nominal rejection data. If the projected result suggested much higher than actual removal this could serve as a relatively inexpensive trigger for further trouble shooting.

The median greater than or equal to the mean did occur for some of the profiles evaluated. However, in instances where it did occur, it was largely due to the fact that arrays were very integral (low standard deviation) and therefore the difference between the mean and median (although negative) was very small and potentially within error associated with the precision of conductivity meters.

4.6.3.2 Seasonal Variation of Results with Temperature

As shown from the long term and frequent monitoring conducted at YVWD (Section 4.4.6), the conductivity profile results vary with temperature. There are ASTM standard approaches to normalize RO data, so using manufacturer specific constants could mediate seasonal variability (ASTM D4516-19A 2019).

Normalization is suggested to be an appropriate and optional addition prior to calculation of an LRV and is anticipated to further reduce outliers due to temperature fluctuation. Further method development would be required to appropriately incorporate the ASTM D4516 salt passage normalization approach into the LRV calculation approach.

4.6.3.3 Application to Nanofiltration

NF membranes are not designed to remove divalent salts and allow some passage of monovalent salts. The rejection mechanisms for nanofiltration membranes include size

exclusion, charge repulsion, and diffusion. Given that size exclusion will play a role for NF compared to RO, it may not be appropriate to calculate an LRV for NF units using the conductivity profiling approach. Principally due to the assumption that nominal flow through integral portions of the membrane solely due to diffusion may not be valid. Charge repulsion of NF membranes has been shown to be sensitive to pH (Askari et al. 2018). To that end, conductivity profiling activities for NF membranes may benefit from being performed under reference/constant pH conditions as well. The work in this report demonstrated conductivity profiling as a means to calculate an LRV for RO systems only. Consequently, further method development is recommended to investigate the applicability of this approach to NF integrity verification.

4.7 Conductivity Profiling Conclusions

An automatic conductivity profiler was installed in May 2019 at YVWD to monitor the integrity of its full-scale RO system. Utilizing a normal distribution approach, a statistical analysis framework was developed to categorize vessel performance as integral or as an outlier based on the group performance of each stage. During the last 21 months, over 4,000 conductivity profile reports were automatically generated and analyzed by Separation Processes, Inc., Carlsbad, CA (SPI) to demonstrate the ability of this diagnostic tool to verify integrity of the RO system. Similarly, during this test period, the validity of the statistical framework had been evaluated to determine the robustness of the different alarms that were put into place. In other words, if a vessel was reported as an outlier, does that vessel actually constitute an integrity breach? Or are the alarms too lenient that no alarm is triggered yet a compromise does exist? If an alarm is too conservative, too many alarms (i.e., false positives) will be unnecessarily generated. On the other hand, if alarms are too lenient, integrity issues are undermined. A sensitivity analysis was performed to answer these questions by testing different alarms and it was determined that the 1.3*Median alarm would be a more accurate integrity predictor than the currently used 1.5*Median which was rarely activated.

The Average +3σ alarm has proven to be an accurate alarm because it diagnosed vessel 33 as compromised since November 2019. This integrity issue was confirmed with MS2 testing, vessel probing, and pressure tests demonstrating the effectiveness of the automatic conductivity profiler and the statistical framework to accurately pinpoint outlier vessels (Discussed further in Chapter 6).

The automatic conductivity profiler has shown to be a useful diagnostic tool for RO integrity monitoring with the ability to identify outlier vessels and signal operators to correct integrity issues.

If automated, conductivity profiling can be used at a frequency sufficient to qualify as a DIT per the MFGM (i.e., once per day). The sensitivity of the LRV calculated from conductivity profiling is conservatively capped at a maximum of 5-log, corresponding with low end of values typically observed during MS2 challenge tests of intact RO systems. In practice, considering the calculated LRV results reported in this chapter, LRVs from conductivity profiling could demonstrate LRVs between 3 and 5-log. This is an improvement over LRVs of 1 to 2-log demonstrated from monitoring bulk feed to permeate conductivity. The resolution of conductivity is based on the size range of ions responsible for conductivity in the source water. Typically, ions are 100 times smaller than MS2 and 10,000 times smaller than *Cryptosporidium*. Although the particular ions are not discretely quantifiable (i.e., conductivity is a blend of charged constituents), there are 2 and 5 orders of magnitude margins of safety to verify virus and *Cryptosporidium* removal sized defects respectively. The research team recommends that, due to this added level of conservatism with respect to target resolution, an allowance should be made to waive discreetly quantifiable requirements for the use of conductivity. Consequently, it is proposed, subject to the best practices described in Section 4.6, that use of an automated conductivity profiling unit should qualify as a DIT per the MFGM.

Conductivity profiling as a monitoring technique to calculate LRV may be limited to systems where arrays produce 0.5 - 1.0 mgd (using 8-inch elements) to ensure that there are a sufficient number of comparable vessels to calculate meaningful statistics. Systems with primary and secondary RO would need further RO flow monitoring to adequately account for the flow contribution between stages and weight ultimate contribution to the whole plant LRV. Cyclic high recovery RO processes will require further investigation as rejection performance of these systems is dynamic within a cycle. It may be overwhelmingly complex to design conductivity profiling apparatus for these systems to ensure that samples are taken at representative and reproducible points within a production cycle. In addition, further work evaluating conductivity profiling and calculation of NF arrays would help to understand whether the inherent assumptions regarding diffusion being responsible for median conductivity is appropriate for NF.

Use of conductivity profiling to verify RO removal credit would likely benefit from periodic verification against other surrogates or comparison to vendor projection standards to eliminate the possibility of false negatives (when the median is greater than the mean). Temperature correction approaches based on standardized normalization (ASTM D4516) could be further investigated and integrated within the calculation framework applied herein. Temperature correction would be anticipated to marginally increase LRV, by mitigating the impact of low rejection outliers due to higher RO feed temperatures.

CHAPTER 5

Orange County Water District

5.1 Site Description and Testing Objectives

Full scale testing was conducted at the OCWD Groundwater Replenishment System (GWRS) Advanced Water Purification Facility (AWPF) located in Fountain Valley, CA. The AWPF produces high quality recycled water as part of the GWRS, a potable reuse project jointly operated by OCWD and Orange County Sanitation District (OC San). GWRS is currently the world's largest water reclamation facility for potable reuse and a recognized industry standard. As a groundwater augmentation project, the finished water is recharged into the local groundwater aquifer (a drinking water source), as opposed to being directly used for drinking (i.e., delivery straight to tap).

The GWRS AWPF facility utilizes a multiple-barrier approach to recycle 100 mgd of secondary-treated wastewater that would otherwise be discharged to the ocean. The AWPF treatment train is comprised of MF, UF, RO, and ultraviolet (UV) disinfection with hydrogen peroxide addition (UV/H2O2), referred to as the advanced oxidation process (AOP), followed by decarbonation and lime stabilization. The AWPF RO process principally uses two types of membranes, Hydranautics ESPA2-LD (Nitto Denko) and Dupont FilmTec BW30XFRLE-400i (DuPont). There are a total of 21 RO treatment units at the facility, each with 5 mgd rated capacity, running in parallel to produce the total 100 mgd of RO permeate. Each 5-mgd unit operates over a range of recovery from 80 to 85%. The RO is a three-stage process, in which the water recovery is approximately 50% for the first two stages and 30% in the third stage. Each RO unit contains a total of 150 pressure vessels (PV): 78 PVs in parallel in the 1st stage, 48 PVs in the 2nd stage, and 24 PVs in the 3rd stage. Each PV holds seven RO membranes (membrane elements) for a total of 1050 RO membranes per 5 mgd unit. Figure 5-1 shows a simplified diagram of a 5 mgd RO unit. The RO unit sampled for this study was equipped with Dupont FilmTec BW30XFRLE-400i membranes.

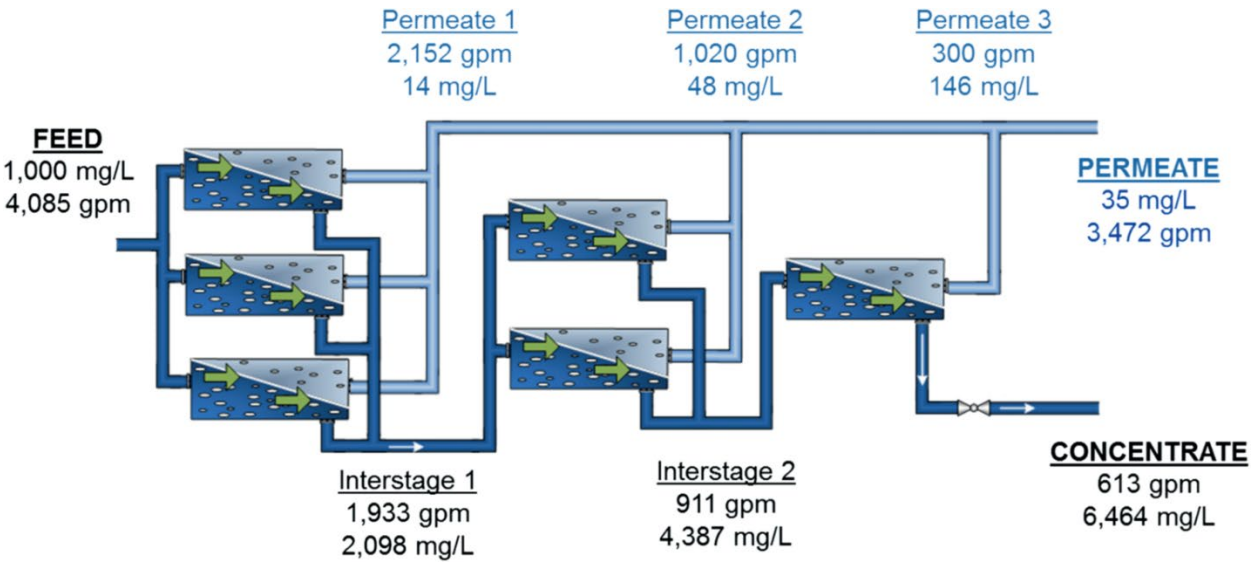

Figure 5-1. Typical Three-Stage Reverse Osmosis Unit and Instrumentation at the OCWD GWRS AWPF.

5.2 Orange County Water District Test Methods

Testing for this study at OCWD consisted of full-scale RO unit profiling of conductivity and uranine, where conductivity was present as a native surrogate and uranine was spiked. The profiling of conductivity or uranine enabled determination of diffusion adjusted LRV. Additionally, LRV was determined directly using TOC, sulfate, and strontium from the RO unit bulk feed and permeate.

5.2.1 Conductivity Profiling

EC measurements were performed manually by OCWD staff on a weekly basis for a six-month period. The manual sampling on all 150 PVs in the RO unit provided conductivity profiling data. The sampling locations included PVs 1-150 RO permeate; stage 1, 2, 3 RO concentrate; the combined permeate; and the first stage RO feed. EC measurements were collected using a handheld EC meter (Myron L, Ultrameter II 6PII, Carlsbad, CA).

The profiling data sets were entered into a standardized Excel spreadsheet provided by SPI to calculate various parameters based on the EC readings to support assessment of RO integrity. The excel template calculates alarm warnings and a conductivity profiling LRV consistent with the method described in Section 4.3. A total of 28 separate conductivity profiling events for the same RO unit were collected over the six-month study period (weekly, except for 2.5 weeks of GWRS shutdown period in August 2021).

5.2.2 TOC Analysis

For determination of LRV based on TOC, two portable TOC analyzers (Suez Sievers M9, Boulder, CO) were installed to measure the feed and combined permeate streams of the same 5-mgd RO unit that was evaluated for conductivity profiling and all other surrogates. TOC was monitored as a control for comparison with the various other LRV surrogates and methods evaluated in the study since TOC is conventionally used for LRV crediting purposes in RO at potable reuse facilities. The analysis frequency for the portable TOC instruments was every 2 minutes with an accuracy of ± 2% or ± 0.5 ppb. The analyzer oxidizes organic matter to gaseous carbon dioxide (CO_2) and measures the TOC in mg/L.

5.2.3 Uranine Spiking

Uranine (Bright Dyes Fluorescent FLT Yellow/Green powder, Kingscote Chemicals, Miamisburg, OH) was injected into the full-scale RO feed water of one 5 mgd RO unit (same unit used for other above surrogates) using a prepared stock solution with a concentration of 154.8 g/L. The uranine powder is a specially formulated Xanthene dye (generic name for a class of fluorescent dyes including uranine), certified by ANSI/NSF 60 for use in drinking water. It has a molecular weight of approximately 376 g/mol. The maximum use level in potable water is 1 µg/L (or 1 ppb). The ANSI/NSF 60 certification is based on brief and infrequent use of the product.

To prepare the stock solution, the dye powder was gradually mixed with deionized (DI) water in a five-gallon plastic bucket lined with aluminum foil to prevent degradation by light exposure and plastic bags due to the intense staining ability of the solution. The stock solution was dosed at 200 mL per minute using a metering pump (Grundfos DDA 12-10, Grundfos holding AG, Denmark) into a sampling port on the feed side of the high-pressure pump of the full-scale 5 mgd RO unit.

Uranine sample collection began 10 minutes after dosing commenced to ensure a stable feed concentration. RO permeate samples for uranine analysis were collected from all pressure vessels of the RO unit (1-150). Also, the concentrate from Stages 1, 2, and 3, as well as the combined feed and permeate (bulk ROP) were sampled to fully specify a uranine array profile. All samples were collected in 40-mL amber glass vials and were stored in the dark until analysis. To avoid a pH effect on the measurement of the fluorescence signal, the pH of all samples was adjusted to approximately 8.1 by adding 1.0 M Tris Base buffer (Millipore-Sigma, St. Louis, MO) to the sampling vials prior to the sampling. Uranine was dosed for approximately 40 minutes for the 4/28/2021 event and 25 minutes for the 6/3/2021 event.

Uranine samples were analyzed within 48 hours of sample collection using fluorescence spectroscopy at the OCWD Research and Development (R&D) Department laboratory (Horiba Aqualog, Irvine, CA). The feed, interstage feed, and concentrate samples were diluted 100, 133, and 400 times before analysis. An excitation wavelength of 490 nm and absorbance wavelength of 512 nm were used to quantify uranine concentration. The peak absorbance occurs at 512 nm. The emission measurement of the samples was converted into uranine concentration using a calibration curve generated using standard uranine solutions of known concentration.

5.2.4 Sulfate and Strontium

Sulfate and strontium samples were collected during the 4/29/2021 and 6/3/2021 sampling events. The water samples were collected prior to the uranine dosing experiments. Sulfate and strontium samples were analyzed by an accredited commercial laboratory (Eurofins Eaton Analytical, Monrovia, CA), whereas conductivity and uranine were analyzed in-house.

Not all PVs were sampled for sulfate and strontium due to the cost associated with external lab analysis and instead a representative subset of PVs was selected. Samples were collected directly from RO unit concentrates from stage 1, 2 and 3; reverse osmosis feed (ROF); and combined reverse osmosis permeate (ROP) sampling points. Additional RO permeate samples were collected from selected PVs (eight PVs in stage 1: numbers 1, 7, 13, 37, 43, 61, and 67; five PVs from stage 2: numbers 78, 79, 85, 97, 103, and 126; and three PVs from Stage 3: numbers 133, 134, and 135) using 250mL amber glass bottles. These stage samples were equal volume composited to create stage 1, 2, and 3 permeate composite sample since there is no stage combined permeate sampling port on the RO unit. The composite permeate samples were analyzed for sulfate and strontium.

5.3 Orange County Water District Results

5.3.1 Conductivity Profiling

Each RO conductivity profile report included the conductivity of all pressure vessels, the test number, date, and starting and ending time as well as supporting data for analysis. Based on the collected raw data, the program calculates the average, standard deviation, median, and skew as well as the preestablished alarm limits (i.e., Average + 3σ, +1 Skew, and 1.5*Median) as discussed in Chapter 4. Each report also summarized bulk stage flows and other properties which were extracted from SCADA for the tested RO unit.

Table 5-1 shows the activated alarms and log removal value calculation results from a representative conductivity profile from February 4, 2021 (Event 1 of 28). In this EC profiling result summary, there is one PV from stage 1 (PV 33) and another from stage 2 (PV 98) where permeate EC values were observed to be higher than three standard deviations from the mean. One PV from stage 1 (PV 33), two PVs from stage 2 (PV 98, 121), and two PVs from stage 3 (PV 127, 130) had permeate EC values that were 50% greater than the stage's median conductivity. The skew from all stages was larger than 1.

Table 5-1. Summary of the Activated Alarms and LRV Calculation Results for February 4, 2021, Conductivity Profiling Event.

Parameter	Units	Stage 1	Stage 2	Stage 3	Array
Activated Alarms					
Number of PVs with Avg + 3σ Warning [1]	Vessels	1	1	0	
Number of PVs with 1.5*Median Warning [2]	Vessels	1	2	2	
Skew [3]	-	1.9	4.3	1.6	
Log Removal Value Calculation					
Median Permeate Conductivity	µS/cm	12	21	43	17.9
Median Permeate TDS	mg/L	6.6	11.6	23.7	10.8
Adjusted Permeate TDS	mg/L	0.2	1.2	1.1	0.55
Adjusted Perm Log TDS	mg/L	-0.8	0.1	0	
Diffusion-Adjusted LRV	-	3.7	3.2	3.6	
Percent Removal	%	99.982	99.942	99.972	99.970
Full unit Diffusion-Adjusted LRV	-				3.5

Notes:

1. This statistical criterion counts the number of samples (PVs) where permeate conductivity falls outside of the three standard deviations of the mean.
2. This statistical criterion counts the number of samples (PVs) where permeate conductivity is more than 50% greater than the stage's median conductivity.
3. The final criterion is to establish whether skew, or the measure of distribution above and below the average, is an ideal or symmetric distribution around the average at zero; a value greater than +1 indicates that a defect may be present.

Table 5-2 contains a summary of the statistical analysis of the permeate conductivity per vessel from the RO unit from February 2021 to October 2021. These values are averages of all 28 manual conductivity profiles conducted weekly during the testing period. The statistical analysis interprets the data in terms of warnings and alarms based on degree of any deviation from median permeate conductivity. It should be noted that OCWD uses different operational criteria for investigation and resolution of outliers with respect to the RO system performance, thus the methodology below developed by SPI for interpreting conductivity profiles may be a more stringent set of criteria for operational intervention. While the focus of the present study was to determine diffusion-adjusted LRV from the conductivity profiles (e.g., as shown in Table 5-1 for one of the events), the (separate) statistical evaluation (review of warnings/alarms) was also completed and summarized here.

Table 5-2. Summary of Warnings and Alarms During February 2021 - October 2021.
28 Conductivity Profiling Events

Statistical Parameters[1]	Units	Stage 1	Stage 2	Stage 3
Average Permeate Conductivity	µS/cm	13.46	28.77	67.69
Standard Deviation Permeate Conductivity	µS/cm	1.48	3.59	6.89
Median Permeate Conductivity	µS/cm	12.93	27.82	65.52
Skew	-	1.66	2.67	2.23
Limit: Average + 3σ	µS/cm	17.89	39.54	88.37
Limit: 1.5*Median	µS/cm	19.39	41.73	98.28
% of Sampling Events with Average + 3σ Warning	%	96.4%	92.9%	78.6%
% of Events with 1.5*Median Warning	%	28.6%	42.9%	28.6%
% of Events with >1 Skew Warning	%	85.7%	92.9%	96.4%
% of Events with Two Warnings 3σ and 1.5*Median	%	27.6%	39.8%	22.4%
% of Events with Three Warnings	%	23.6%	37.0%	21.6%

Notes:
1. The average, standard deviation, median, and skew values here refer to average values from all 28 Conductivity profiling events of the array studied.

It is worth noting that the data are positively skewed, exhibited as an average skew value of +1.66 for stage 1, +2.67 for stage 2, and +2.23 for stage 3 (as shown in Figure 5-2). The +1 Skew alarm was triggered in 85.7% of the EC profiling events for stage 1, 92.9% of the tests for stage 2, and 96.4% for stage 3. This means, in the case of stage 1, that of the 28 weekly profiling events, 24 events (or 85.7% of the time) showed a skew above 1. The EC distribution for each stage was non-uniform and the second stage had the highest average skew. A skew greater than one indicates asymmetric distribution around the average and that a defect may be present.

The Average + 3σ alarm was triggered in 96.4% of all the tests for Stage 1, 92.9% of all the tests for Stage 2, and 78.6% of all the tests for Stage 3. This means, in the case of Stage 1, that of the 28 weekly profiling events, 27 events (or 96.4% of the time) showed at least one PV featured permeate conductivity more than three standard deviations away from the average. The 1.5*Median alarm was triggered in 28.6% of all the tests for Stage 1, 42.9% of all the tests for Stage 2, and 28.6% of all the tests for Stage 3.

As shown in the Figures below, one or two vessels within each RO stage were responsible for triggering the Average + 3σ and +1.5*Median alarms in each EC profiling event, and these same PVs were observed to be consistent with each event (i.e., conductivity profiling appeared to reproducibly indicate that particular vessels may contain off-specification elements). For example, vessels 33 and 34 in stage 1, vessels 98 and 121 in stage 2, and vessels 127 and 130 in stage 3 nearly continuously had alarms for skews and Average + 3σ. For the 1.5*Median warning, the alarms were much less frequent after 5/6/21, but vessels 34, 98, and 121 still triggered these alarms. As noted above, it is not surprising that a high permeate EC value stays high for the next conductivity profiling event given no intervention between events, and thus the same PVs appear as outliers multiple times. However, the conductivity profiles completed

for this study indicate a fair amount of variability in permeate conductivity between the PVs within this 5-mgd RO unit compared to others in the plant. OCWD worked with Dupont (the membrane manufacturer) to examine this variability. The possibility of a membrane manufacturing defect among some of the elements in this unit is under further investigation with the manufacturer. Overall, statistical evaluation of conductivity profiling was valuable to identify and quantitatively assess the variability; importantly, it should be noted that the RO unit permeate quality was acceptable despite the noted variability in the elements' (PVs) performances.

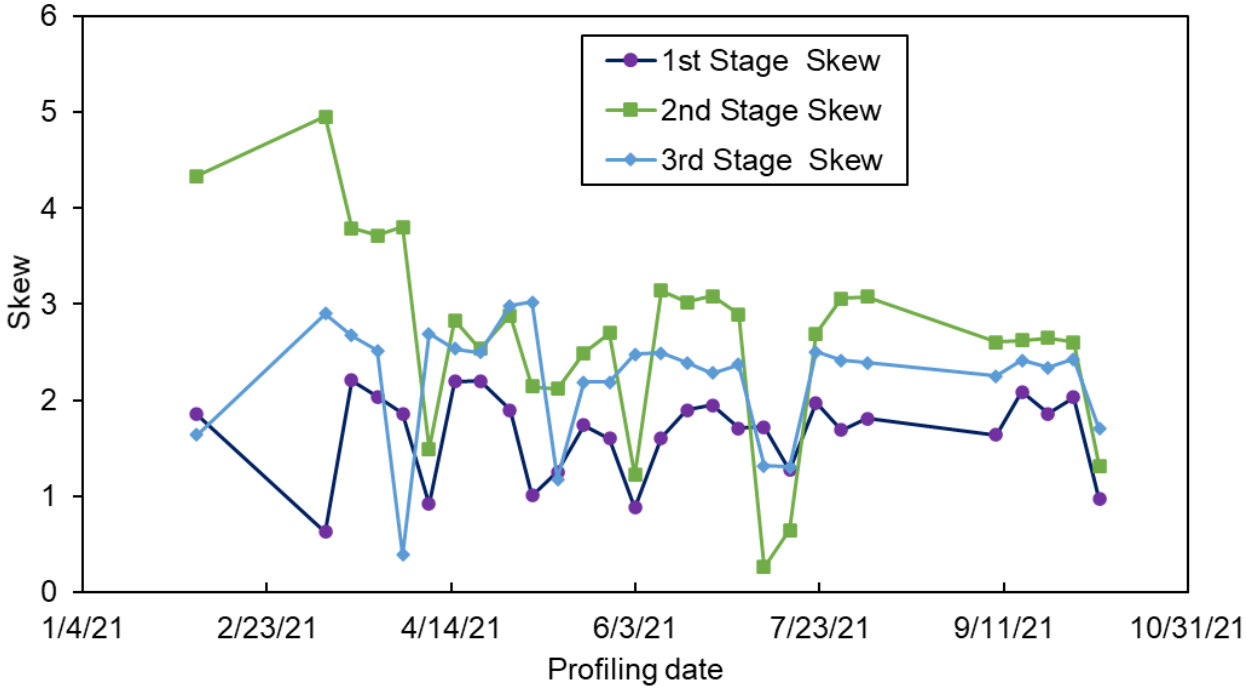

Figure 5-2. The Stage Average Skew Calculated from the RO Permeate Conductivity Per Pressure Vessel for each Manual Conductivity Profiling Event (Approximately Monthly) at OCWD.

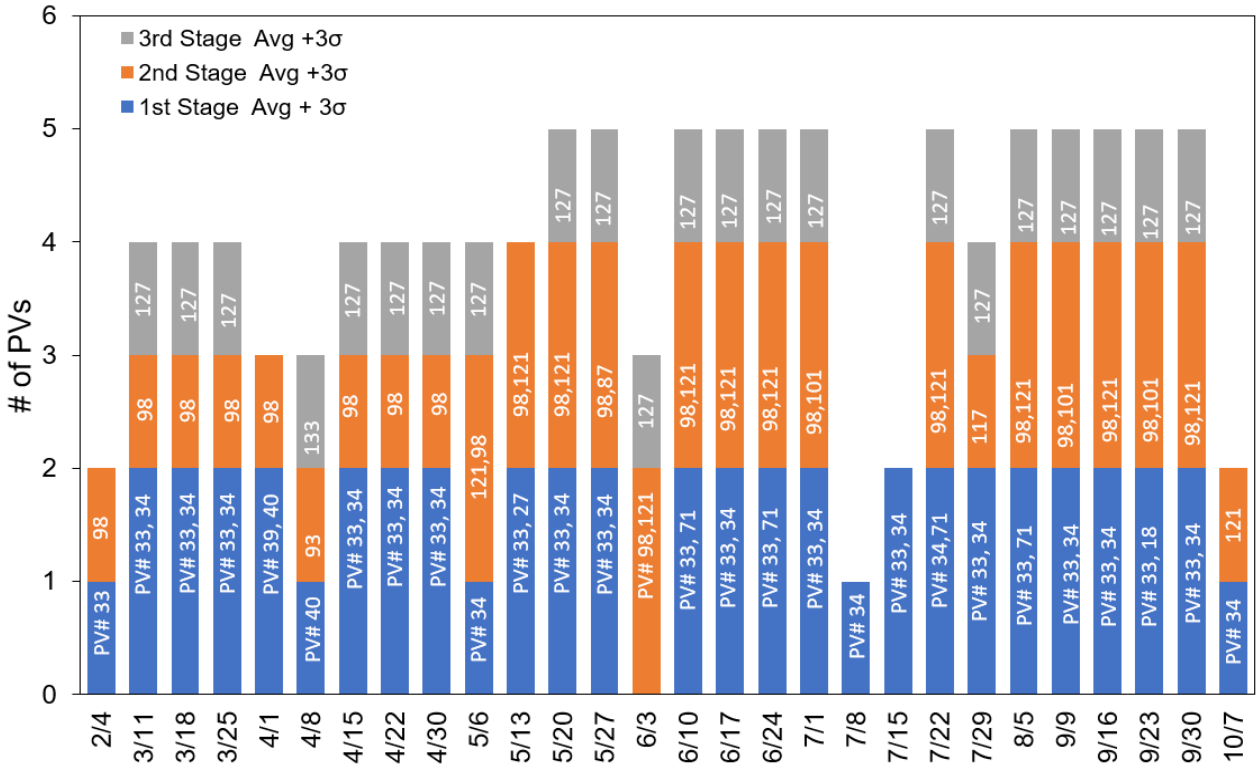

Figure 5-3. Number of RO Pressure Vessels Observed with Permeate EC Exceeding the Limit of Average + 3σ per RO Stage for each Conductivity Profiling Event.

The text labels (e.g., "PV # 33, 34") indicate the particular PV or PVs that were responsible for the exceedance.

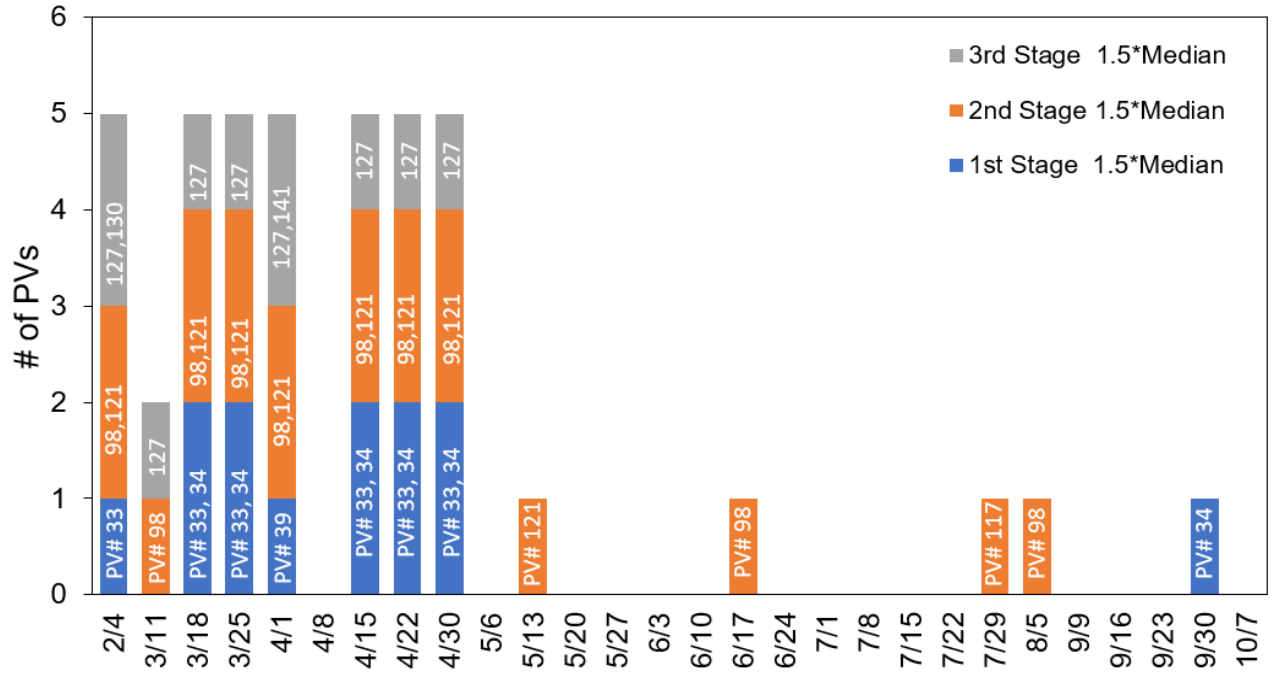

Figure 5-4. Number of RO Pressure Vessels Observed with Permeate EC 1.5*Median per RO Stage for each Conductivity Profiling Event.

The text labels (e.g., "PV # 98, 121") indicate the particular PV or PVs that were responsible for the exceedance.

Figure 5-5 shows the LRV calculated from EC profiling results (LRV_{defect}) based on the EC profiling data. To compare the significances of variations, the percent EC determined from the conductivity profiling results is also shown (Figure 5-6). All observed LRVs calculated from EC profile results were above 3.2-logs and ranged between 3.2 and 4.7-log. Here we note for reference that a ~0.5-log difference in removal (when it's above 3-log) is a *very* small difference in terms of percent removal, i.e., when transformed back to percent removal, a range of 3.2 to 4.7-log corresponds to a range of 99.937% (3.2-log) to 99.997% (4.5-log) removal which is just a 0.06% difference. Thus, an observation of 4.5-log compared to 3.2-log is actually not likely to be significantly different and is probably within the 5% error of the analytical accuracy of the EC instrument. Thus, for log crediting (regulatory) purposes, some consideration should be paid to uncertainty analysis to appropriately round measured log removal to a conservatively credited value.

On two dates (5/6/2021 and 7/8/2021), the LRV for the stage 1 and/or 2 were equal to or above 5-log, which, based on how the diffusion-adjusted LRV (LRV_{defect}) is calculated, indicates that the average permeate TDS was less than or equal to the median permeate TDS. In all cases, the percent EC removal calculated based on LRV_{defect} remains greater than 99.93%. The 2.5-week shutdown in August 2021 did not affect membrane performance, as percent rejection and LRVs remained at pre-shutdown values.

In summary, the statistical analysis of weekly EC profiles (over six months) from the full-scale 5 mgd RO unit yielded an average calculated LRV of ~3.6-log. This is a significant increase over the RO pathogen credit based on direct LRV for EC (bulk permeate compared to feed) of 1.9 to 2.0-log for the same dates. Beyond demonstration of a higher LRV using conductivity profiling analysis, the statistical analysis method can also help plant operators identify suspect PVs that have elevated permeate EC outside the alarm limits.

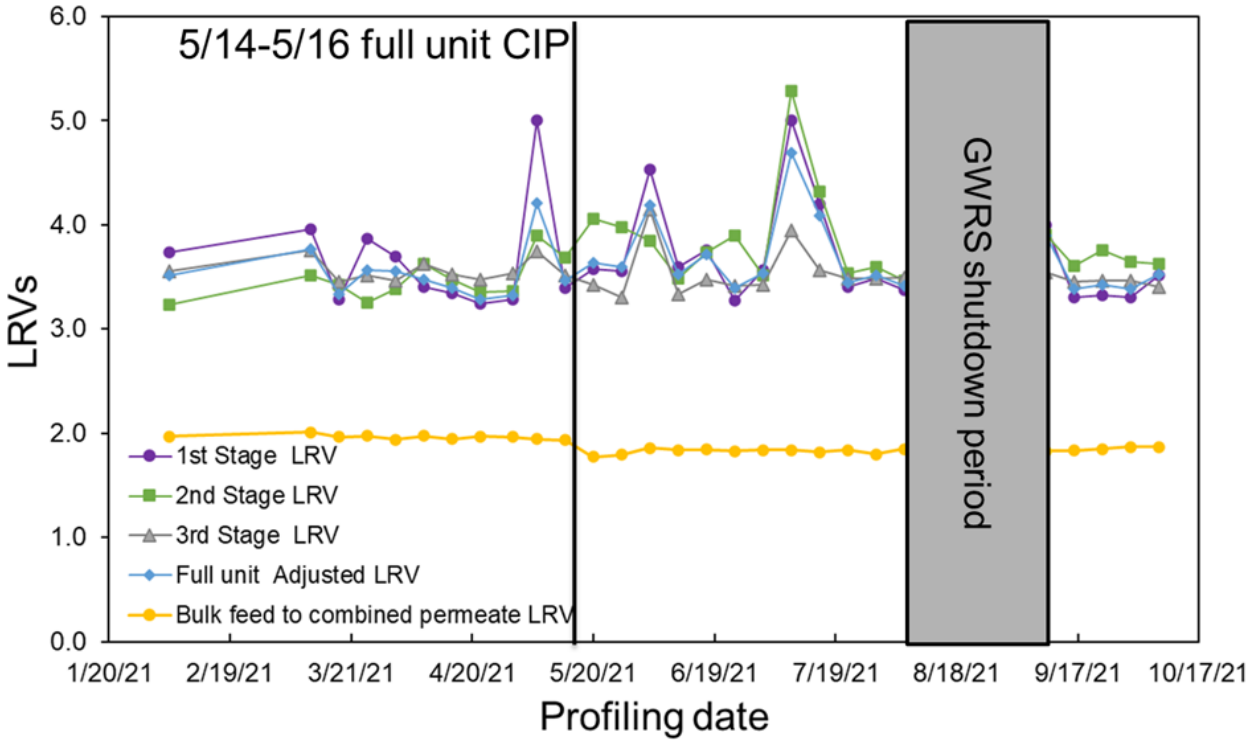

Figure 5-5. OCWD EC Profiling Calculated LRV for the 1st Stage, 2nd Stage, 3rd Stage, and Overall (Full Unit).
The grey shading indicates the approximate occurrence of a clean-in-place (CIP).

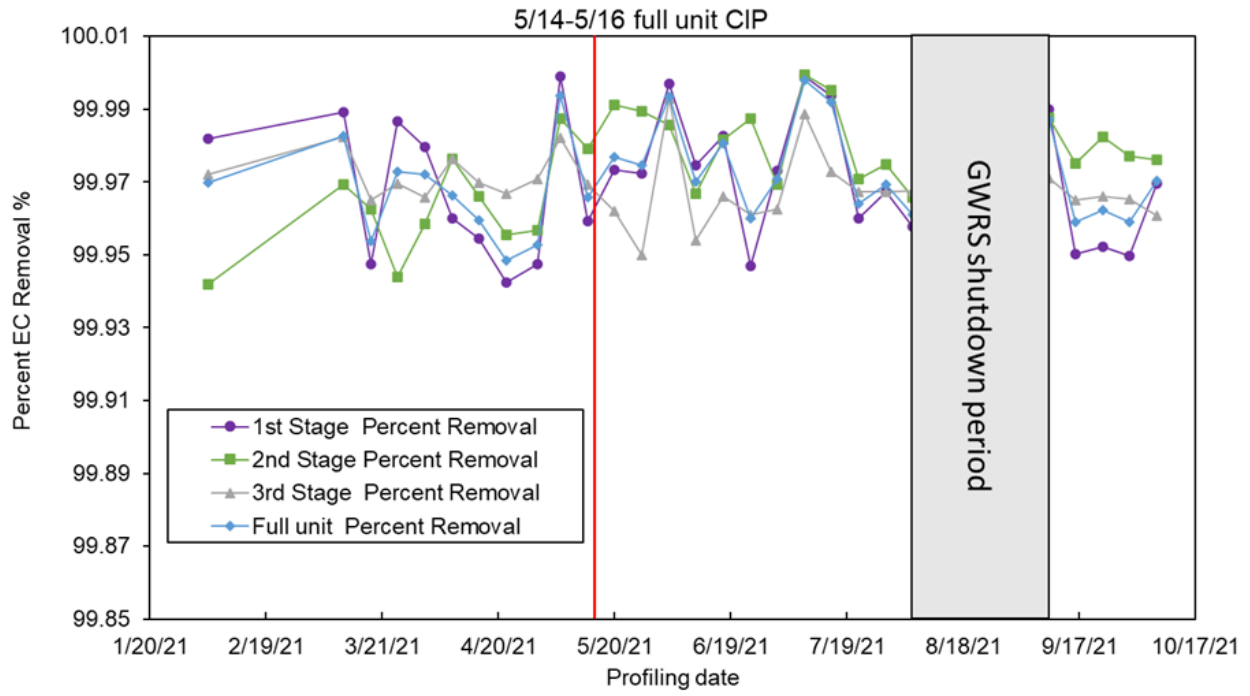

Figure 5-6. OCWD Percent EC Removal Calculated Based on EC Profile Results for the 1st Stage, 2nd Stage, 3rd Stage, and Overall (Full Unit).
The red line indicates the approximate CIP period.

5.3.1.1 Impact of Clean-in-Place on Conductivity Profiles

The weekly EC profiles can also be used to monitor the impact of membrane CIP on permeate conductivity. After a high pH clean, there will be a substantial recovery of permeate flux for the membrane element, but the polyamide layer of the RO membrane can be temporarily swollen by the high pH solution, resulting in a decrease in the salt rejection and increase in permeate EC (Ying et al. 2014). This condition can be reversed over time, but aggressive CIP conditions (pH >13 and high temperature) may permanently damage the polyamide layer and lower the salt rejection. CIP will also remove the fouling layer which effectively decreases the thickness of the selective layer (i.e., otherwise the salt has to diffuse through both the membrane and foulant).

A full RO unit membrane CIP was performed April 14-16, 2021. The CIP included a high pH (pH = 12) organic cleaner (AWC C-227, American Water Chemical, Plant City, FL) followed by citric acid wash (low pH cleaning, pH = 2). After the full unit CIP, the permeate EC increased in all stages with stage 3 having the highest increase (Figure 5-7). The EC values gradually returned to the pre-CIP levels after about 21 days (Figure 5-7), consistent with typical observations after CIP.

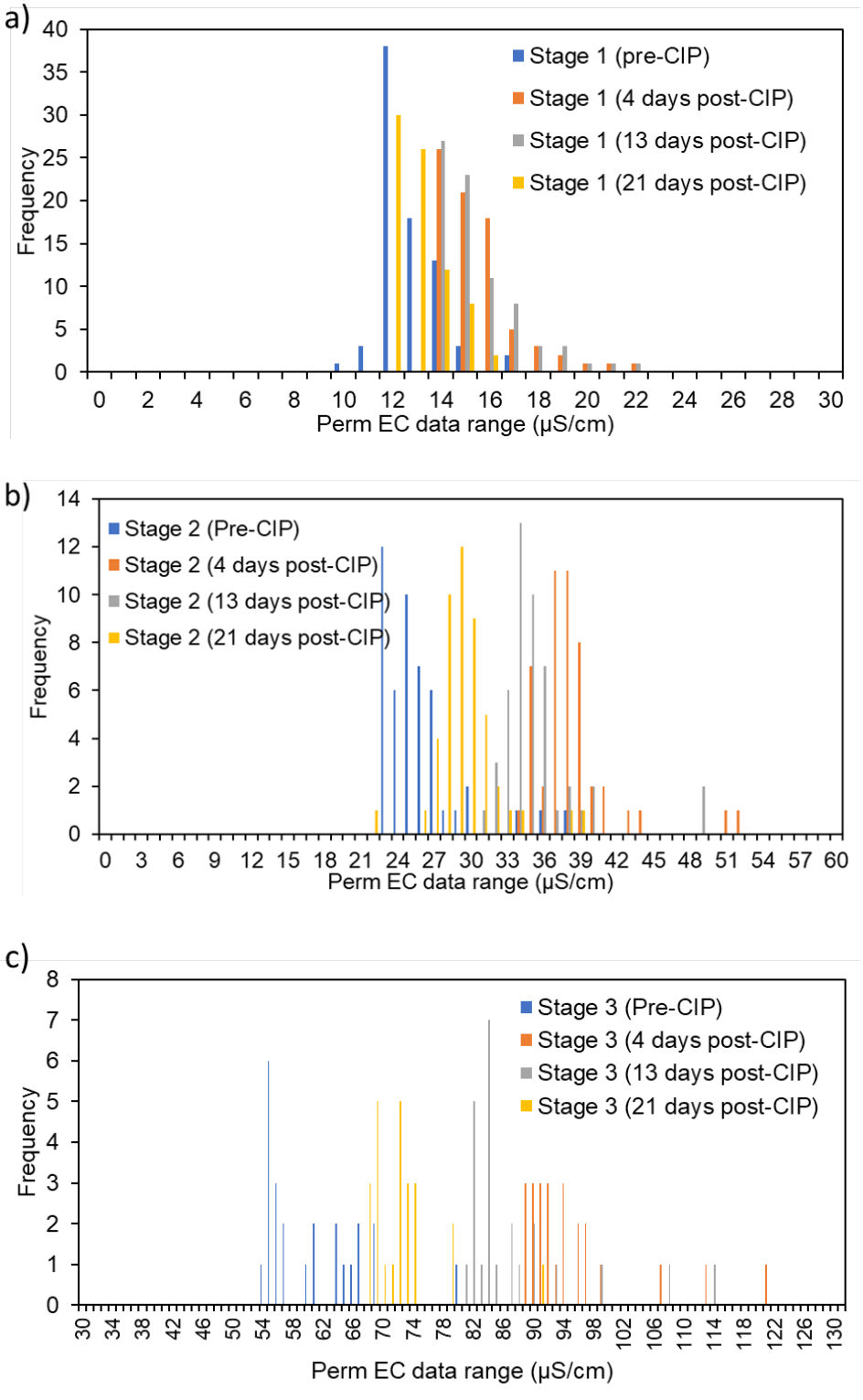

Figure 5-7. Impact of CIP on EC Profiling for a) Stage 1, b) Stage 2 and c) Stage 3 for the Array Studied at OCWD.
(Sampling completed on 5/13, 2021 [pre-CIP] and 5/16/2021 and 5/21/2021 [post-CIP]).
Tick marks (bin) on x-axis indicate the permeate EC and the y-axis ("frequency") indicates the number of pressure vessels that produce permeate with the corresponding EC.

5.3.2 TOC Removal

The RO system at the OCWD currently employs redundant (duty + duty) online TOC analyzers that continuously demonstrate 2+ log removal of TOC. The measured rejection of TOC across RO (feed to combined permeate) is applied as a pathogen credit value for GWRS based on the daily average TOC LRV. EC is used as a backup to demonstrate a 1.5+ log removal.

For the test array, portable TOC analyzers were used to monitor the treatment performance of this unit. TOC in ROF and ROP streams were measured continuously (every two minutes) in two periods 4/21/2021 to 5/14/2021 and 5/17/2021 to 6/4/2021. These periods cover the dates when the EC profiling, uranine testing, and sulfate and strontium sampling were performed at this site for the study, all for the same full-scale 5 mgd RO unit. Figure 5-8 shows the results from real time TOC monitoring. The resulting LRVs were consistently above 2.0-log during the monitoring periods. The average value of LRV was 2.2-log (equal to 99.2% TOC removal by RO). This dataset provided a reference dataset (control dataset) for LRV using the conventional approach (TOC monitoring) on the same full-scale RO unit that was utilized for the assessment of alternative LRV approaches.

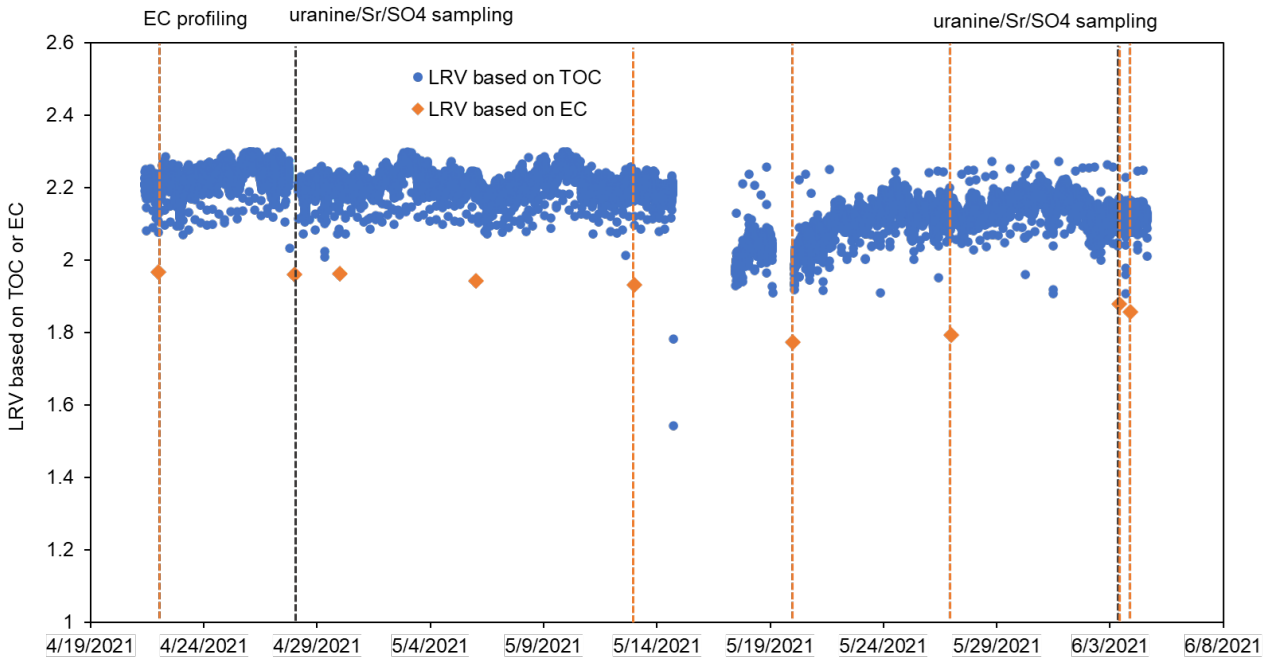

Figure 5-8. OCWD Full-Scale Test RO Unit LRV Based on Continuous Online TOC Measurement.
The orange dashed lines indicate the dates when EC profiling events were performed. The black dashed lines indicate the uranine, Sr, and SO_4 sampling events. A CIP was performed April 14-16, 2021, just before the time period on the chart.

5.3.3 Uranine Challenge Testing and Profiling

In Section 2.3, it was suggested that adjusting for contributions of diffusion versus defect flow may help to improve the sensitivity of marker-based testing. In this section, the individual vessel profiling approach and subsequent calculation of LRV (Section 4.3) was applied to results gathered during uranine challenge testing to quantify the potential sensitivity improvement.

5.3.3.1 Profiling of Uranine Compared to Conductivity

Two uranine spike challenge experiments (4/28/2021 and 6/3/21) were completed for the full-scale 5 mgd RO test unit along with full unit EC profiling and stage permeate sulfate and strontium sampling. Uranine sampling and analysis procedures are described in Section 5.2.3. The objective was to evaluate the observed LRV of uranine dosed into the RO feed for a full-scale RO unit to understand any benefit over the use of native markers, where the observed LRV refers to the "direct" LRV, i.e., from direct comparison of RO feed and permeate uranine concentration.

EC profiling (previously described) was completed prior to the injection of uranine to compare the LRV derived from statistical analysis (that accounts for EC diffusion, i.e., diffusion-adjusted LRV or LRV_{defect}) of a native marker. Sulfate and strontium profiling was considered but was cost prohibitive, thus more limited sampling from each stage was completed for exploratory purposes.

Figure 5-9. OCWD R&D Staff Await the Start of the Permeate Sampling for the GWRS 5 mgd RO Unit During the Uranine Dosing Experiment.

The 5 mgd RO unit (Figure 59) was in normal operation during the uranine challenge tests, as one of up to 21 AWPF RO units generating RO permeate at the plant; it is not possible for an AWPF RO unit to discharge its permeate to waste during normal operation. The dye stock solution was fed to the RO system upstream of the high-pressure feed pump to achieve an RO feed water uranine concentration of approximately 2.0 mg/L. The dye stock solution at high concentration exhibits a dark red color, but at more diluted concentrations, the solution appears with a bright yellowish-green color. Table 5-3 shows the uranine concentrations (mg/L) in the RO feed and stage concentrate streams. Figure 5-10 shows the color of the RO stage 1, 2, and 3 concentrate coming out of the sampling taps during one of the uranine dosing experiments. The water became darker (more concentrated) in yellow as it moved through the three RO stages.

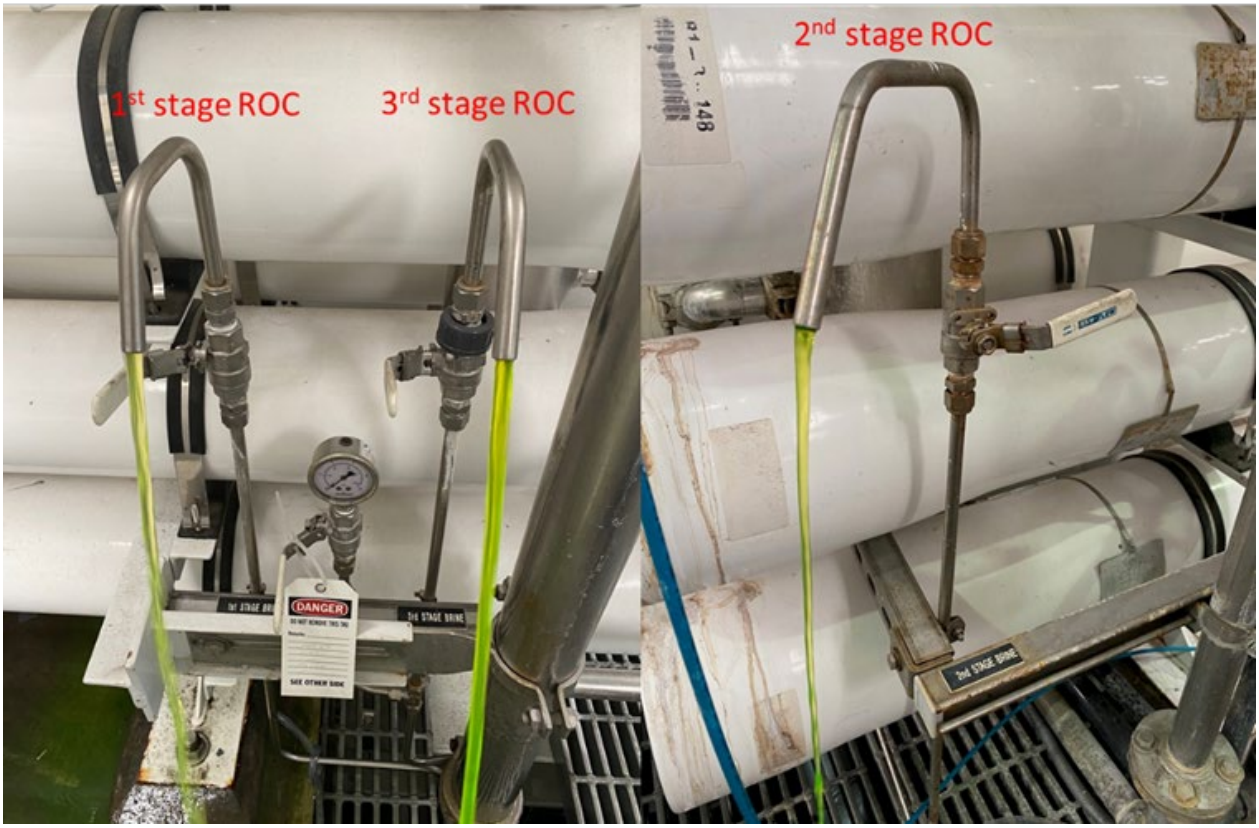

Figure 5-10. Photos of the RO Stage 1, 2, and 3 Concentrate Taps During Uranine Dosing Experiment.

Table 5-3. Uranine Concentrations (mg/L) in the RO Feed and ConcentrateSstreams.

Sampling date	RO feed	First stage RO concentrate	Second stage RO concentrate	Third stage RO concentrate
4/28/21	1.90	3.94	8.35	13.8
6/3/21	1.91	3.97	7.70	12.7

Table 5-4 shows uranine LRV results and Table 5-5 shows EC LRVs, both calculated using the profile data and approach from Section 4.3. The RO unit EC profile was collected before the uranine injection. In addition to LRV calculated based on profile data, the direct LRV (RO feed to combined permeate) is included to compare sensitivity enhancement. For stage 2 and 3, the feed concentration was the RO concentrate concentration from stage 1 and 2, respectively.

Table 5-4. Profile and Direct LRV from Uranine Challenge Testing.

Profile LRV, Uranine[1]					Direct LRV (Bulk ROP)
Test Dates	Stage 1	Stage 2	Stage 3	Full Unit	
4/28/2021	3.44	3.41	3.51	3.45	3.08
6/3/2021	3.47	3.62	3.53	3.51	3.18
Notes: 1. Profiling increased uranine LRV by approximately 0.5-log.					

Table 5-5. Conductivity Profile and Direct LRV Conducted Immediately Before Uranine Challenge Testing.

Profile LRV, Conductivity					Direct LRV (Bulk ROP)
Test Dates	Stage 1	Stage 2	Stage 3	Full Unit	
4/28/2021	3.48	3.50	3.50	3.49	1.96
6/3/2021	4.21	3.56	3.44	3.82	1.88

Comparison of the full unit LRV calculated from profiling to the direct LRV reveals the improvement in demonstrated LRV based on use of the statistical approach to account for diffusion. Significantly, it is approximately 1.5 to almost 2-logs greater, depending on the event, for conductivity. LRV calculated from uranine profile data (~3.5-log) increased by 0.4-log compared to the direct uranine LRV of ~3.1-log. The relatively small improvement in LRV due to profiling was not considered worth the significant additional effort to collect a much greater number of uranine samples required for the profile. In general, uranine is not expected to be practically useful from a regulatory perspective for RO LRV crediting since a DIT must be daily and dosing dye into RO feed would not be practical.

The traditional calculation of LRV (direct LRV in Table 5-3) for uranine (approximately 3.1) is much greater than for EC (approximately 1.9), as expected due to uranine's greater rejection by RO. However, the profiling LRVs (i.e., diffusion-adjusted LRVs) for uranine and EC were similar - within 0.3-log units. The greater increase in profiling LRV for EC compared to uranine may be explained by the greater diffusion of salt over uranine into RO permeate, i.e., EC LRV benefits more from a diffusion correction than does uranine. Good correlation between EC versus uranine was observed when the three RO stages were compared separately. Further, vessels that exhibited high EC permeate also showed high uranine concentration.

5.3.3.2 Uranine Profiling to Enhance Sensitivity of Defect Identification

As opposed to the use of spiked uranine to demonstrate credit for log removal, which per above is not likely to be practical, the study team explored whether uranine profiling in an RO unit may be useful for evaluating unit-wide membrane integrity to detect potential issues (i.e., outlier PVs), similar to how operators already utilize conductivity profiling, but with potentially more sensitivity due to the greater rejection of uranine. Figures 5-11, 5-12, 5-14, and 5-15 show color heat maps of the LRV values calculated from the individual PV's permeate uranine (or EC) concentration for both sampling events. The general observation is that the stage LRVs calculated from profiling data determined from the uranine dosing experiment were similar to the stage LRVs determined from the EC measurements. However, the range of individual PV's LRVs were wider for uranine (2.4 - 5.07) than the EC measurements (1.9 - 2.3) likely due to the greater sensitivity of uranine owing to its greater rejection. The linear regression correlation between EC and uranine for the three RO stages are shown in Figure 5-13 and Figure 5-16 for the two uranine experiment dates. The R-squared values for each stage show a good correlation between EC and uranine measurement.

Statistical analysis showed 7 of the 10 worst performing PVs (low uranine LRVs) were also the worst performing PVs for EC (low EC LRVs). This observation indicates both methods were able to identify membrane integrity issues, despite the wider variation in readings and more noticeable difference with uranine heat maps compared to EC. Thus, the use of uranine over EC provides limited value especially in light of the practical difficulties in spiking and offline measurement of uranine.

Overall, there is a wide range in observed LRV across the different PVs within this particular full-scale RO unit for both EC and uranine (i.e., observed permeate conductivity and permeate uranine concentration) which is in contrast to OCWD staff experience with other RO units and membranes for EC (from routine conductivity profiling). Because the RO membrane elements were fairly new (Dupont Filmtec BW30XFRLE-400i installed in October 2020), the current hypothesis is that there may be a membrane integrity issue for some of the elements that may be related to a manufacturing defect (e.g., leaking glue line). However, it should be noted that this variation does not translate into a lower-than-acceptable performance based on bulk (blended) permeate EC and TOC, where TOC is the current basis for OCWD pathogen credit for RO.

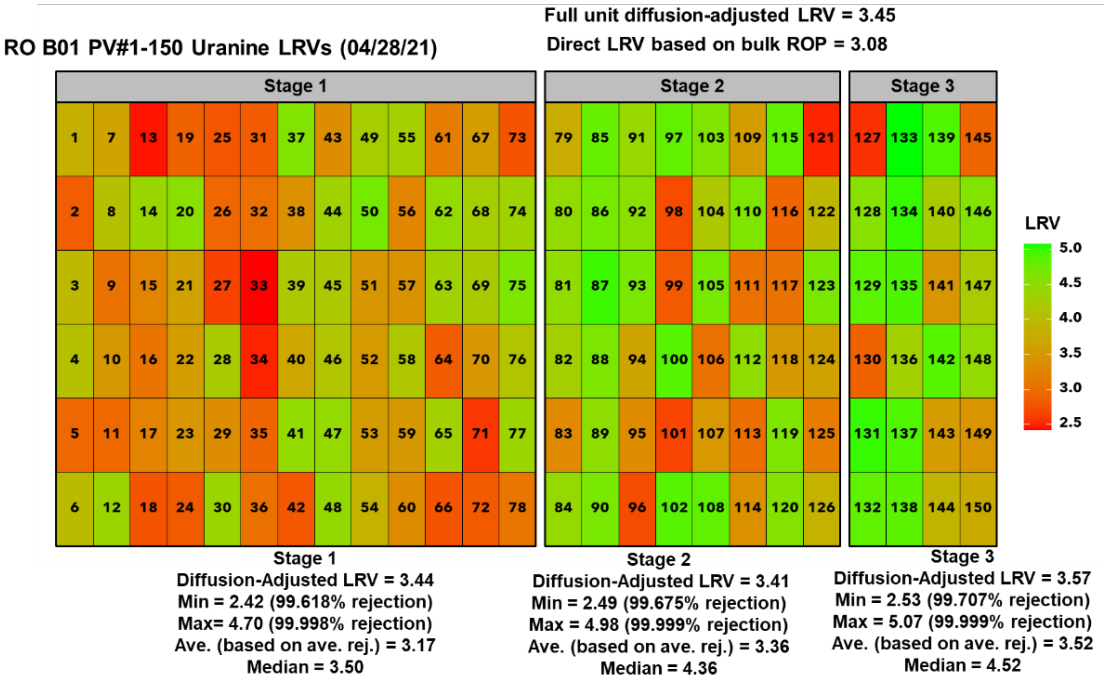

Figure 5-11. Color Heat Map of the LRVs Calculated from the Individual PV Permeate Uranine Concentrations. Sampling Completed on 4/28/2021.

The numbers in the boxes represent the pressure vessel number. Stage 2 and 3 LRVs were calculated using previous stage's concentrate as the feed value.

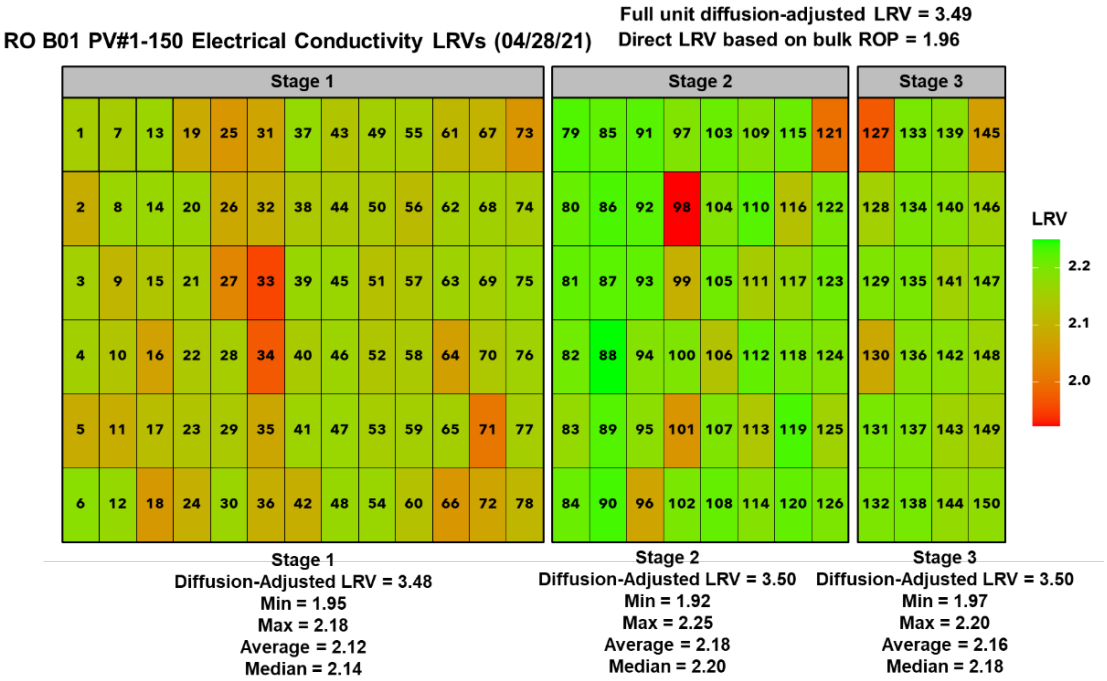

Figure 5-12. Color Heat Map of the LRVs Calculated from the Individual PV Permeate EC Concentrations. Sampling Completed on 4/28/2021.

The numbers in the boxes represent the pressure vessel number. Stage 2 and 3 LRVs were calculated using previous stage's concentrate as the feed value.

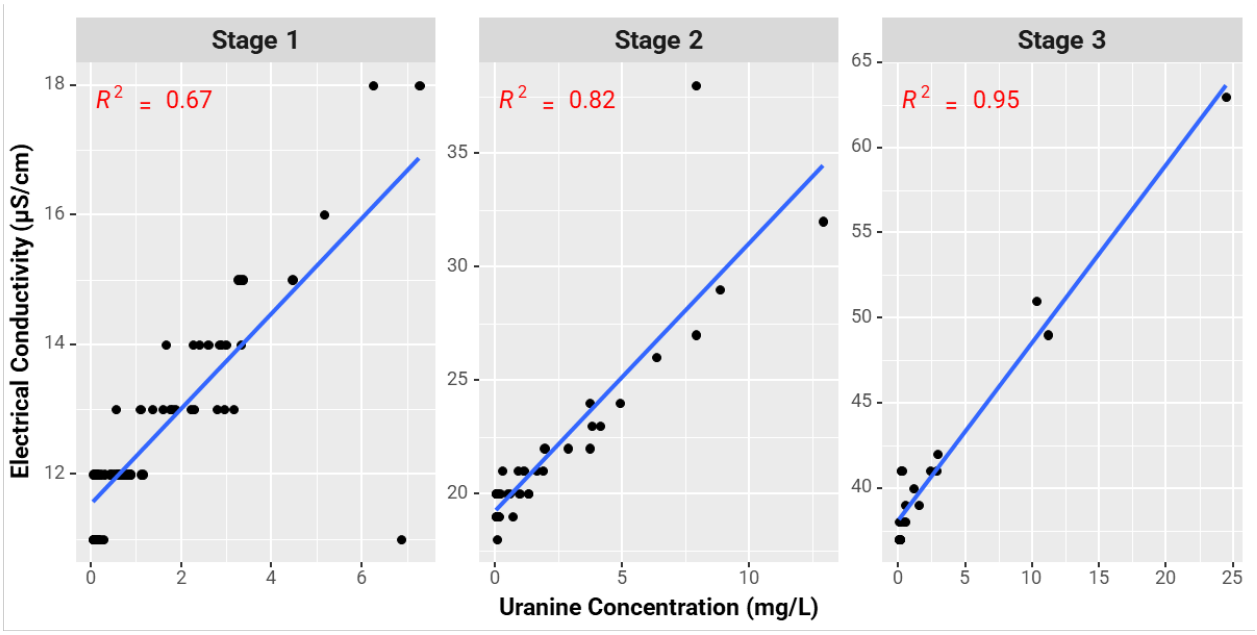

Figure 5-13. Correlation of EC Versus Uranine for each Individual Pressure Vessel's Permeate Sampled on 04/28/21.

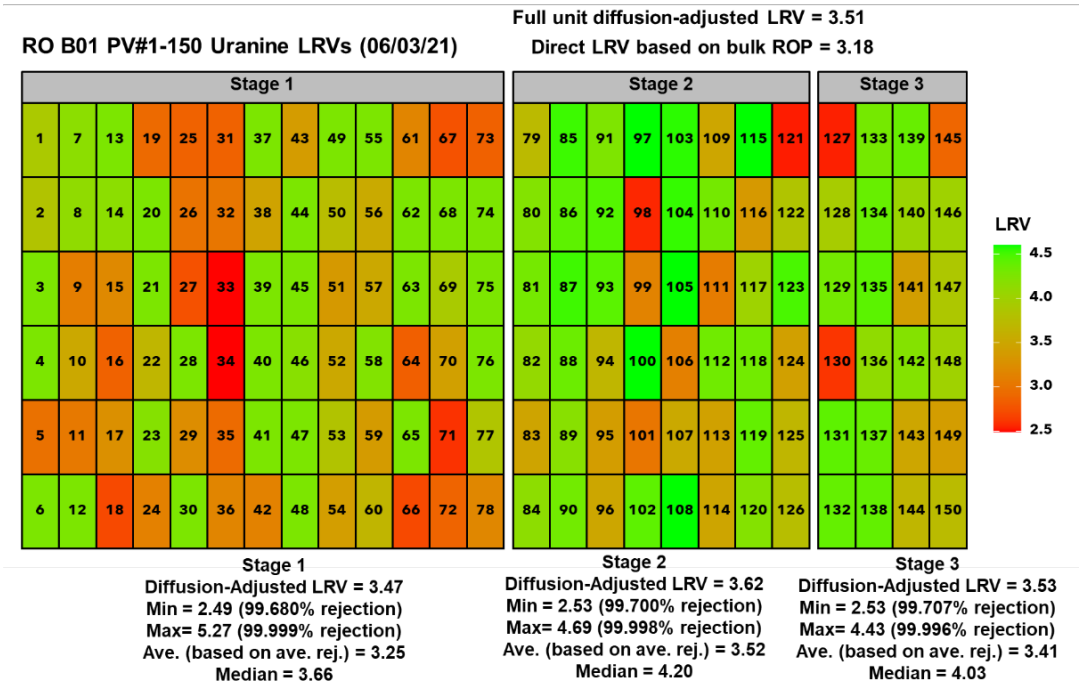

Figure 5-14. Color Heat Map of the LRVs Calculated from Individual PV Permeate Uranine Concentrations. Sampling Completed on 6/03/2021.

The numbers in the boxes represent the pressure vessel number. Stage 2 and 3 LRVs were calculated using previous stage's concentrate as the feed value.

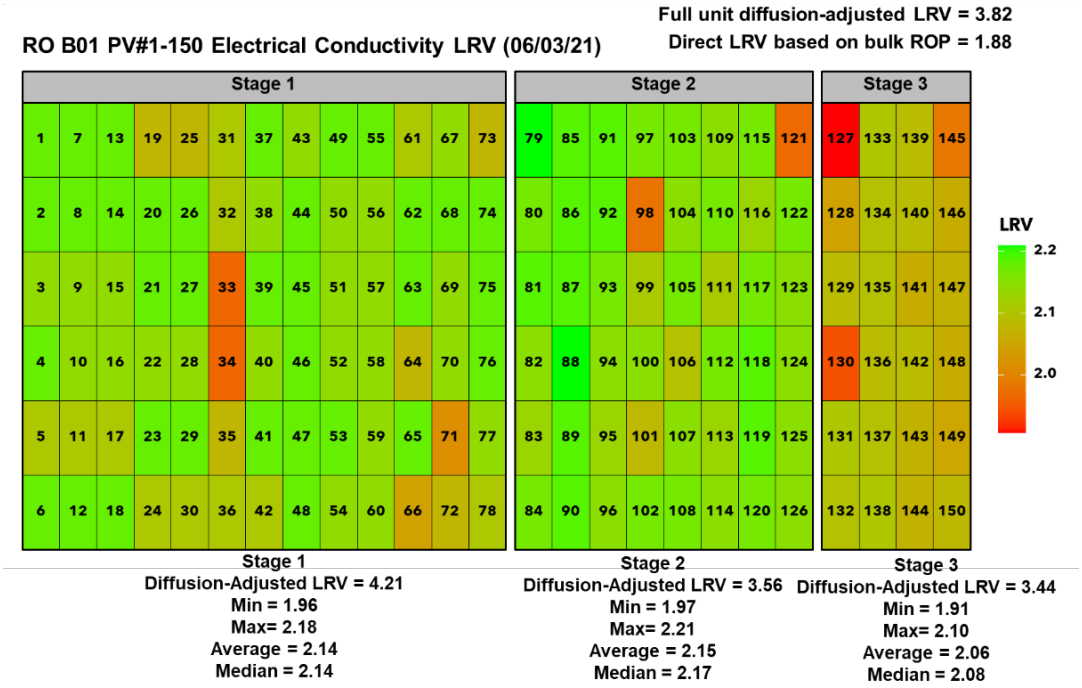

Figure 5-15. Color Heat Map of the LRVs Calculated from Individual PV Permeate EC Concentrations. Sampling Completed on 6/3/2021.

The numbers in the boxes represent the pressure vessel number. Stage 2 and 3 LRVs were calculated using previous stage's concentrate as the feed value.

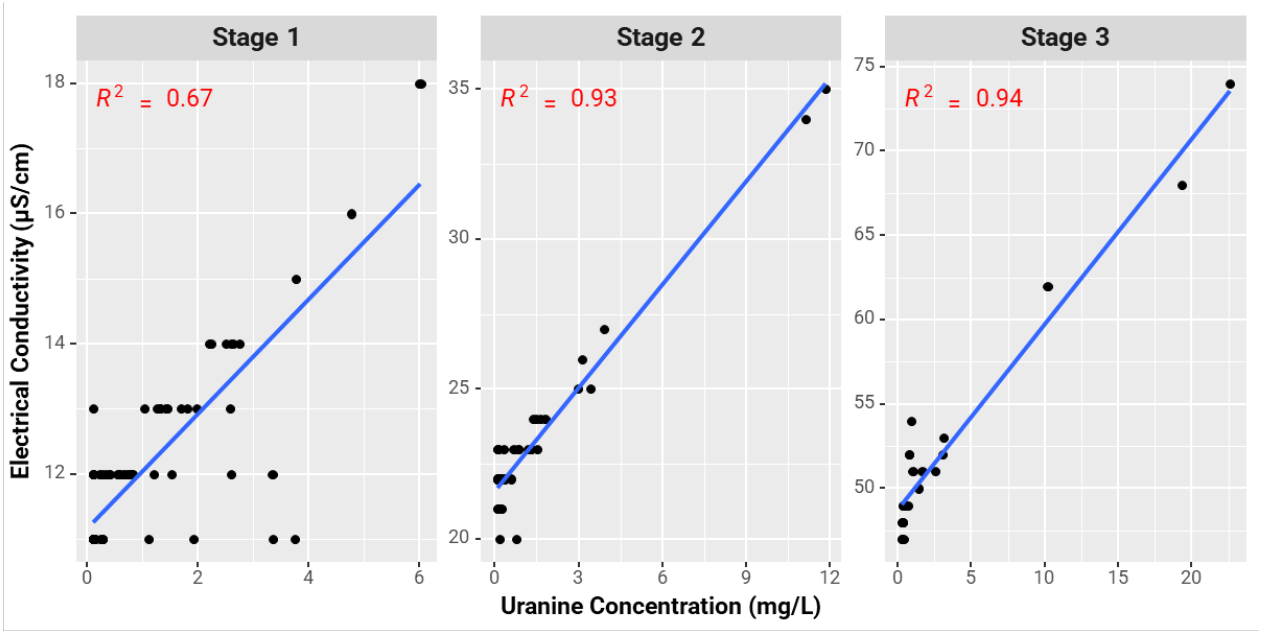

Figure 5-16. Correlation of EC Versus Uranine for each Individual Pressure Vessel's Permeate Sampled on 06/03/21.

5.3.3.3 Sensitivity Analysis of Membrane Repair on Array LRV

To evaluate the sensitivity of the uranine and EC integrity monitoring methods against changes in individual PVs' LRVs, hypothetical scenarios were studied and are presented in Figure 5-17.

The figure on the left shows the observed LRV calculated from uranine profiling (3.5), and the estimated full unit calculated LRV values for three hypothetical cases.

In hypothetical case #1, it was assumed the operators can fix 15% of the worst performing PVs (12 lowest ranked PVs for uranine, i.e., highest permeate concentrations) that have LRV less than 4 and convert them each to a new LRV of 4. An LRV of 4 was used as the nominal "replaced" membrane LRV value because it is close to the median LRVs for stages 1 to 3. A 15% repair to the median rejection was determined to increase the profiling LRV to 3.7 (from the original 3.5).

In hypothetical case #2, it was assumed that 30% of the worst performing PVs (23 PVs in this case) that have LRV less than 4 could be repaired or replaced such that they achieve an individual LRV of 4. A 30% repair was estimated to increase the calculated LRV based on profiling to 4.0 (from the original 3.5).

Hypothetical case #3 assumed all PVs that had LRV less than 4.0-log are fixed (76 PVs) to achieve an LRV of 4. The substantial repair or replacement of more than half of the PVs in the array was estimated to increase the LRV to 5.1 from the original 3.5.

The scenarios indicated that addressing and improving a fairly low number of PVs with apparent membrane integrity issues can improve the overall LRV based on uranine by 0.3 to 0.4-log; but this is not a dramatic improvement, and a significant number of PVs must be addressed to achieve much greater overall LRV (i.e., case #3). Further, it is unknown how likely the operators would achieve success in addressing the lower rejection of any number of PVs, since it may be a reflection of the quality of the element(s) as-received rather than a matter of addressing proper O-ring and permeate side interconnector installation.

A similar analysis was completed for the EC profiling data in Figure 5-17 (right). The bar chart on the right shows the observed full unit diffusion-adjusted LRV based on EC (3.8) and the estimated full unit diffusion-adjusted LRV for two hypothetical cases.

In hypothetical case #1, a 30% repair or replacement of the worst performing PVs (9 lowest ranked PVs for EC) was assumed. The impact of the repair was assumed to ensure that PVs with LRV less than 2.1 would achieve an LRV of 2.2 after repair. An LRV of 2.2 is equal to 99.4% EC removal and represents a nominal rejection number for a new RO element. The 30% repair was estimated to increase calculated conductivity profiling LRV to 4.2 (from original 3.8).

Hypothetical case #2 assumed all PVs that have LRV based on EC less than 2.1 are fixed (30 PVs with low LRV) to achieve an LRV of 2.2. A complete repair was estimated to increase the calculated full conductivity profiling LRV to 4.3 (from original 3.8).

Once again, addressing the high permeate conductivity of a fairly large number of PVs did not appear to significantly improve the overall profiling-based LRV (i.e., ~0.4 to 0.5-log improvement). On the other hand, if a potable reuse facility is basing their RO pathogen credit on the statistically determined, conductivity profiling-based method (diffusion-adjusted LRV or LRV_{defect}) and is operating fairly close to their required pathogen credits, identifying, and addressing problem PVs could provide a meaningful margin of safety. For instance, at the time of this report, OCWD GWRS demonstrates a total of approximately 12.2-log compared to required 12-log for virus from use of TOC for the RO process. However, it may not be straightforward to fix the issue. In the experience of this project, when the OCWD operators opened and inspected one of the more problematic PVs in the full-scale RO unit that served as the test unit for this study, no issues were observed, and it did not improve when the PV was put back into service (i.e., nominal inspection and replacement of O-rings upon reinstall did not appear to improve rejection).

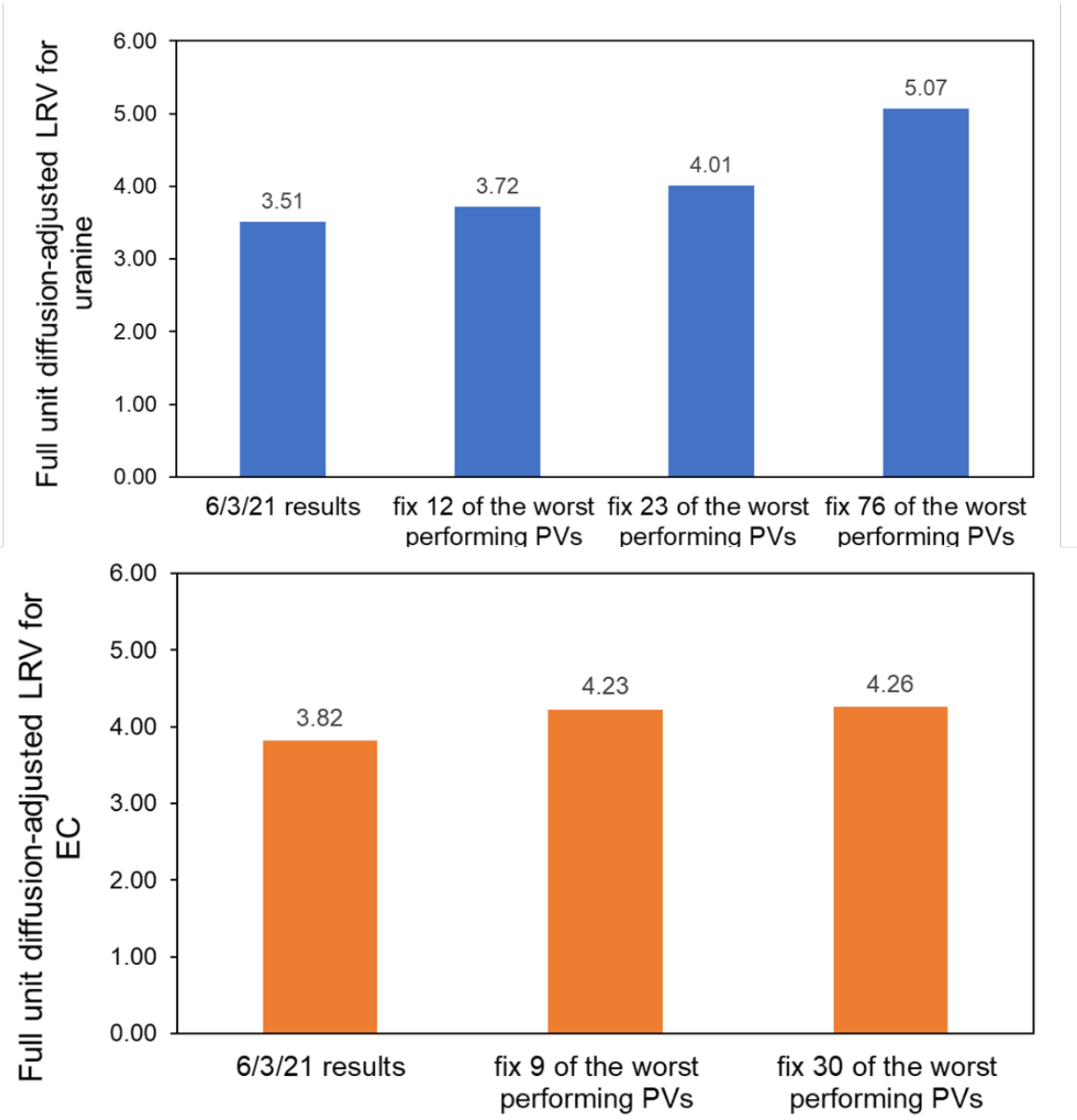

Figure 5-17. The Observed (6/3/21) and Theoretical (for Two or Three Hypothetical Cases) Full Unit LRV Calculated from Profiling Results for Uranine (Top Figure) and EC (Bottom Figure).

5.3.4 Marker Based Challenge Testing

Sulfate and strontium samples were collected during the 4/29/2021 and 6/3/2021 sampling events. The water samples were collected prior to the uranine dosing experiments. Figure 5-18 shows the LRV values based on sulfate and strontium concentrations measured in combined permeate samples and stage permeate sample mixtures. LRV was calculated directly by comparison of the feed and permeate concentrations in each case. The MRLs are 0.50 mg/L for sulfate and 0.3 µg/L for strontium in this study. Sulfate concentrations were below the MRL in all permeate samples, and thus the MRL value was used to substitute the non-detect value to conservatively calculate LRV.

The observed LRVs of strontium and sulfate were similar, ranging between >2.6 to >3.3-log units for sulfate and 3.1 to 3.9 for strontium. The observed sulfate LRV was limited as permeate concentrations were below the MRL of the analytical method. Past measurements in RO permeate at OCWD have successfully detected sulfate above a MRL of 0.25 mg/L using a more sensitive method (Safarik 2020).

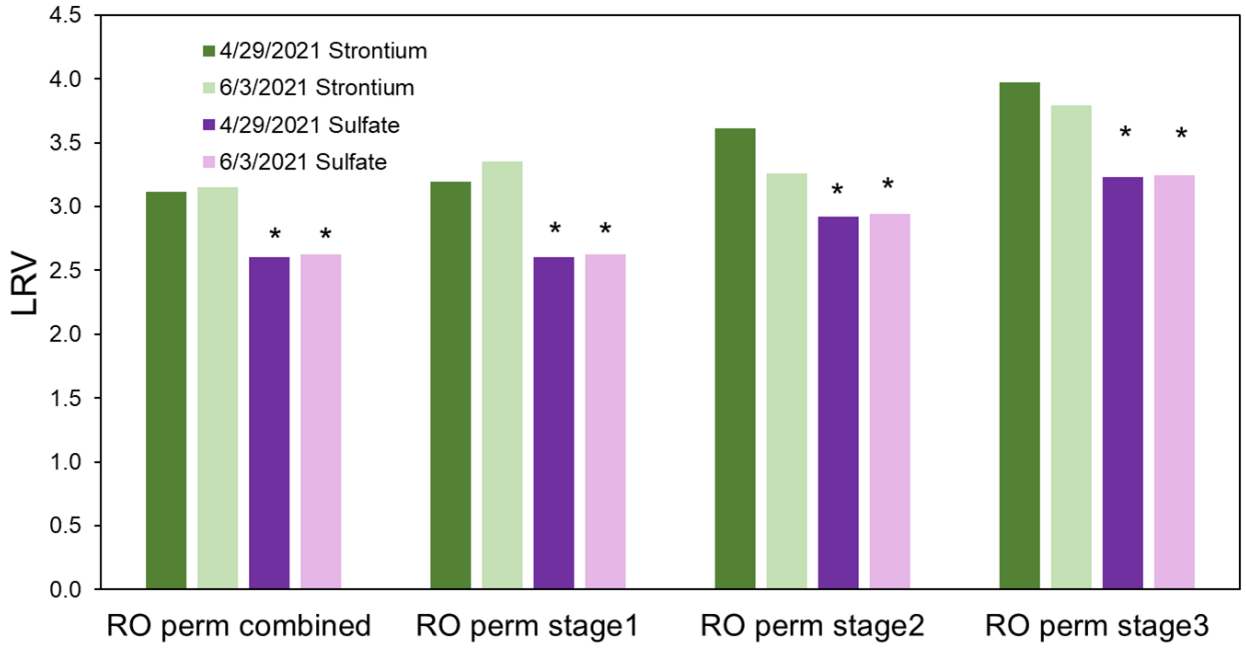

Figure 5-18. Sulfate and Strontium LRVs for the OCWD Full-Scale RO Test Unit.
* Indicate results lower than the MRL in permeate samples. LRVs calculated using the MRLs of 0.50 mg/L for sulfate.

Like conductivity, sulfate and strontium have the potential to pass through the membrane via diffusion (due to their small size) as well as through defects. Future work could perform complete pressure vessel profiling (similar to the EC and uranine profiling completed in this study as described above) in order to determine the impact of diffusion versus defects.

Results of sulfate and strontium LRV near or above 3.0-log show the potential to use naturally occurring ions to obtain higher LRVs than demonstrated with TOC, even without any enhanced LRV from calculation of LRV based on profiling results. However, specific LRVs are constrained by feed water (i.e., background) concentration which may limit the observed LRV if the native feed water concentration is not high enough. That is, surrogates are removed to below the analytical detection limits in the permeate and this limits the LRV able to be demonstrated.

5.4 Orange County Water District Summary

OCWD currently receives pathogen credit of approximately 2-logs for RO based on daily average LRV for TOC. From work completed prior to the present study, OCWD is pursuing enhanced credits based on a tiered framework for strontium, sulfate, and/or ATP monitoring (under regulatory review at time of this report).

For the present study, the project team conducted an investigation of RO integrity at full-scale at OCWD using marker-based and conductivity profiling-based approaches to investigate other alternative approaches for enhanced credit. The marker-based approaches included spiking of uranine into the RO feed to trace its passage into the permeate and to compare measurements to LRVs for native (naturally occurring in RO feed) sulfate and strontium. The conductivity profiling-based approach used a previously developed statistical analysis framework to demonstrate greater LRV credit than traditional EC LRV by isolating “diffusion” versus “defect” related measurements. This calculation method for calculating LRV based on profiling results was also extended to the uranine measurements. Table 5-6 summarizes the LRVs using these various monitoring techniques for the OCWD full-scale 5 mgd RO unit.

Table 5-6. Comparison of Calculated LRVs Using Various Monitoring Techniques for the OCWD Full-Scale 5 mgd RO Unit.

	LRV Calculated from Profiling[1]		Direct LRV[2]				
Test Dates	EC Profiling	Uranine Profiling	EC	Uranine	SO_4[3]	Strontium	TOC
4/28/2021	3.5	3.5	2.0	3.1	>2.9	3.1	2.1
6/3/2021	3.8	3.5	1.9	3.2	>2.9	3.2	2.1

Notes:
1. Profiling LRVs calculated using the approach outlined in Section 4.3.
2. LRV calculated from RO feed to array combined RO permeate.
3. Sulfate was not detected above the MRL of 0.5 mg/L and the MRL was substituted to calculate the LRV. Therefore, the true sulfate LRV is expected to be higher when available analytical method with lower MRL (more sensitivity) is used.

Sulfate and strontium concentrations were measured at the RO system level (i.e., bulk, or combined ROP) and stage level. Table 5-5 shows the direct LRV from bulk ROP for test dates corresponding to just before uranine was spiked into RO feed. The observed removals for strontium and sulfate were similar for the two test dates, 2.9 and 2.92 LRVs for sulfate and 3.13 and 3.15 for strontium. Future work could perform complete RO system profiling of sulfate and strontium (similar to the EC and uranine profiling that was conducted) in order to determine the impact of diffusion versus defects on calculated LRV to understand whether LRV may be significantly increased. This would require measuring sulfate or strontium in all PVs for a

given RO unit (e.g., 150 PVs in the case of a single 5 mgd RO unit at OCWD AWPF), i.e., sulfate/strontium profiling, which may be cost-prohibitive.

The statistical analysis method applied to weekly RO permeate conductivity profiles (over six months) from the full-scale 5 mgd RO unit demonstrated 3.6-log removal value on average. This was a significant increase or enhancement to potential RO pathogen credit based on direct LRV for EC (bulk permeate compared to feed) of 1.9 to 2.0-log for the same dates. On the other hand, a 3.6-log profile LRV from EC would represent a smaller relative enhancement over direct LRV from strontium or sulfate (~3-log; no profiling analysis required) for any facility considering basing their RO LRV regulatory credit on strontium or sulfate (pending sufficient native ions concentration in the feed water), but this more modest enhancement (~0.5-log) may be meaningful depending on the facility case or perspective.

Conductivity profiling can be performed manually daily or automated. It takes about one staff one hour to generate a conductivity profile using one conductivity meter. An automated profiling system, similar to that installed at YVWD (Vickers and Dummer 2020), would be necessary to meet the daily DIT monitoring requirement in order to claim an enhanced LRV credit.

Uranine spike tests in the same full-scale OCWD GWRS RO unit resulted in LRVs calculated from profile data up to 3.5-log for the full RO unit. However, due to the impracticality of dosing uranine into RO feed water every day, uranine is not expected to be useful for demonstration of RO LRV for regulatory credit (whether LRV is traditionally calculated as a direct LRV or using the statistical method as a profile LRV) but may be of interest for periodic RO unit integrity investigations. However, the present study revealed limited value of such investigatory uranine profiling over simple use of natively occurring EC for RO unit profiling because conductivity profiling revealed a similar number and identity of lower-performing PVs.

To evaluate the sensitivity of uranine and conductivity profiling LRVs to changes in individual PVs' LRVs, three hypothetical scenarios were mathematically considered to assess how much would the overall RO LRV calculated from profile data improve by improving some of the worst PVs. The results showed that although uranine testing is more sensitive to membrane defects, addressing some of the worst PVs' membrane integrity issues (here represented by artificially changing that PV's LRV to an ideal value in the profile dataset) will only slightly improve the overall LRV. A more notable LRV increase could only be achieved by addressing virtually all of the poorer performing PVs (which is unlikely to be achievable in reality). It was also true that from conductivity profiling data, the relative improvement to overall LRV from potentially fixing some of the worst PVs was modest, up to ~0.5-log. A 0.5-log difference in removal represents a relatively small change in potential regulatory credit (as well as an insignificant change in percent removal when the data is viewed as percent removal instead of log removal; especially at higher LRVs that is probably within the uncertainty of the analytical instruments). Thus, while it is probably prudent for RO facilities to periodically use conductivity profiling to identify poor-performing PVs, the benefit to LRV from addressing those defective membranes will likely be relatively small.

CHAPTER 6

Yucaipa Valley Water District

This chapter reports the full-scale testing conducted as part of this project that was completed at the YVWD WRWRF in Calimesa, California.

6.1 Site Description and Testing Objectives

6.1.1 Site Description

The WRWRF consists of primary, advanced biological secondary, and tertiary treatment with advanced total nitrogen reduction. The advanced tertiary purification consists of MF, RO, and UV disinfection. Treatment capacity of the wastewater treatment plant (WWTP) is 6.7 mgd and the tertiary treatment capacity is 2.4 mgd. Tertiary treatment meets the coliform bacteria reduction and turbidity requirements of California Title 22 for reclaimed water. The WRWRF uses chloramines as a disinfectant prior to the RO system.

The RO system at the WRWRF consists of a single two stage RO unit with a design flux of 11.8 gallons per square foot per day (gfd) (Figure 6-1). The first stage consists of 52 PVs and the second stage of 20 PVs with seven elements inside each PV. The membrane elements in the RO system are CSM RE-8040-Fen (each with 400 square feet (ft^2) of effective membrane area) which were installed in January 2013. This type of membrane is designed for brackish water and wastewater reuse applications, categorized as having a nominal salt rejection of 99.7% (minimum of 99.4% salt rejection when brand-new). Laboratory analysis from sodium and chloride testing in February of 2020 resulted in 98.4% rejection of chloride and 95% rejection of sodium.

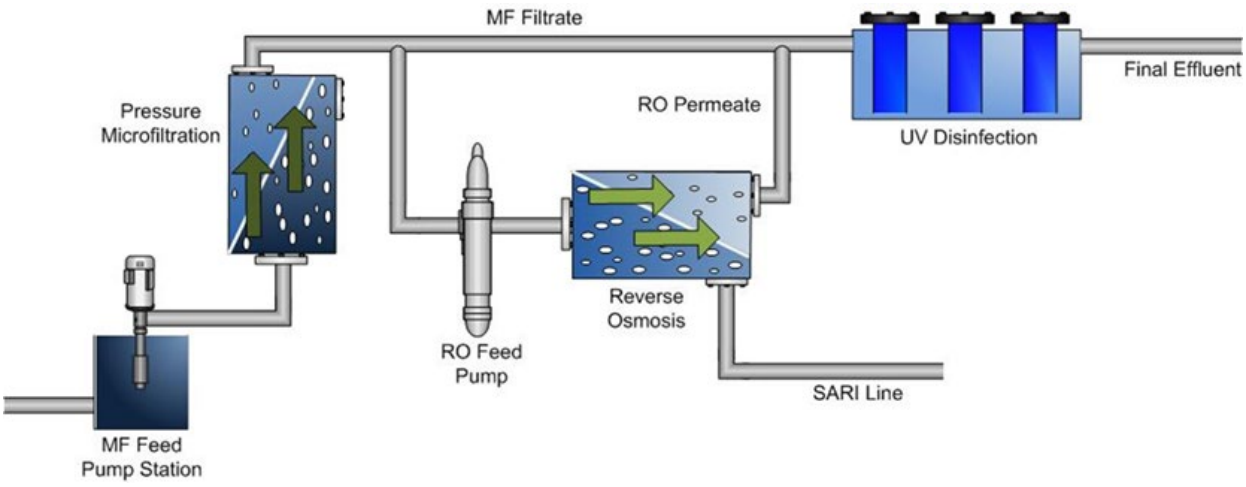

Figure 6-1. Schematic of the Wochholz Regional Water Recycling Facility.

The RO train is operated to produce water under California Article 5.1 (60320.100) regulations for Groundwater Replenishment Projects with Surface Application. Consequently, RO and UVAOP treatment is not required for the whole flow. Instead, the RO is operated in a side stream to reduce TDS of the finished product water. The compliance interval for salinity is 120 months. Thus, the RO train can be stopped and started as required by YVWD. This point aside, the design of the RO train itself is consistent with any train that would be typically associated with a groundwater replenishment facility. Details for the RO train are provided in Table 6-1.

Table 6-1. YVWD RO Train Design Parameters.

Parameter	Value
Design Permeate Capacity	1,650 gpm
Design Recovery	85%
Design Flux	11.8 gfd
Number of Stages	2
Interstage Pump / Energy Recovery Device	Stage 1-2
Array Configuration	52:20
Elements per Pressure Vessel	7
Total Number of RO Membranes	504
Area Per RO Membrane Element	400 ft^2
Membrane Element	Polyamide Thin Film Composite
Membrane Supplier and Model	CSM RE-8040-FE
Nominal Membrane Rejection	>99.4%

The design flow rates of the different streams in the RO system are shown in Table 6-2. Chemical addition to the RO feed includes sulfuric acid for pH adjustment to 6.5, chloramination for biofouling control, and a scaling inhibitor.

Table 6-2. RO Design Operating Conditions at YVWD.

Parameter	Units	Stage 1	Stage 2	Unit
Feed Flow	gpm	1,941	750	1,941
Concentrate Flow	gpm	750	291	291
Permeate Flow	gpm	1,192	458	1,650
Recovery	%	61%	61%	85%

The nomenclature of the pressure vessels in the YVWD RO system, as shown in Figure 6-2, are labelled as Stage-Column-Row (e.g., Vessel Number. 1-3-5 is in the first stage, the third column, and the fifth row).

1-1-7	1-2-7	1-3-7	1-4-7								
1-1-6	1-2-6	1-3-6	1-4-6	1-5-6	1-6-6	1-7-6	1-8-6				
1-1-5	1-2-5	1-3-5	1-4-5	1-5-5	1-6-5	1-7-5	1-8-5	2-1-5	2-2-5	2-3-5	2-4-5
1-1-4	1-2-4	1-3-4	1-4-4	1-5-4	1-6-4	1-7-4	1-8-4	2-1-4	2-2-4	2-3-4	2-4-4
1-1-3	1-2-3	1-3-3	1-4-3	1-5-3	1-6-3	1-7-3	1-8-3	2-1-3	2-2-3	2-3-3	2-4-3
1-1-2	1-2-2	1-3-2	1-4-2	1-5-2	1-6-2	1-7-2	1-8-2	2-1-2	2-2-2	2-3-2	2-4-2
1-1-1	1-2-1	1-3-1	1-4-1	1-5-1	1-6-1	1-7-1	1-8-1	2-1-1	2-2-1	2-3-1	2-4-1
FIRST STAGE								**SECOND STAGE**			

Figure 6-2. Nomenclature of Pressure Vessels for the YVWD RO.

6.1.2 YVWD RO Investigation Objectives

Recent results indicate that the reduction of MS2 bacteriophage by RO is generally above 5.0-log (Vickers 2018; Vickers et al. 2019). Building upon these initial results, the purpose of this investigation is as follows:

- Reproduce the initial results of MS2 testing to demonstrate their validity.
- Characterize the MS2 reduction of RO pressure vessels that exhibit high and low EC.
- Characterize the MS2 reduction before and after cleaning events.
- Characterize the MS2 reduction under artificial or naturally compromised conditions.
- Evaluate reduction performance of other surrogate indicators such as uranine and sulfate to further explain fecal virus reduction capabilities of RO systems.

Table 6-3 summarizes the purpose of each set of MS2 challenge testing results as they pertain to the above investigation goals.

Table 6-3. Targeted Variables for MS2 Challenge Testing at YVWD.

Test Conditions for MS2 Bacteriophage Challenge Tests	Purpose
Integral System or Normal Ops: The RO system is tested at its current state without any changes.	Establish a baseline for comparison when the same system is challenged under different conditions.
Before and after CIP Procedure (Pre- and Post-CIP).	To show that a defect is detectable while still maintaining a system feed to combined permeate LRV above the currently awarded value.
Intentionally Compromised System: End Adapter with a 1/16 of an inch orifice NF Element Substitutes RO Element.	To demonstrate the membrane's reduction properties immediately after cleaning when the membrane chemistry is temporarily changed.

6.2 YVWD Test Methods

6.2.1 Sampling and Monitoring Locations

Figure 6-3 provides an illustration of the sample locations associated with the YVWD RO unit. In addition, the specific unit studied is equipped with the automated conductivity profiling apparatus that is described in Section 4.4.

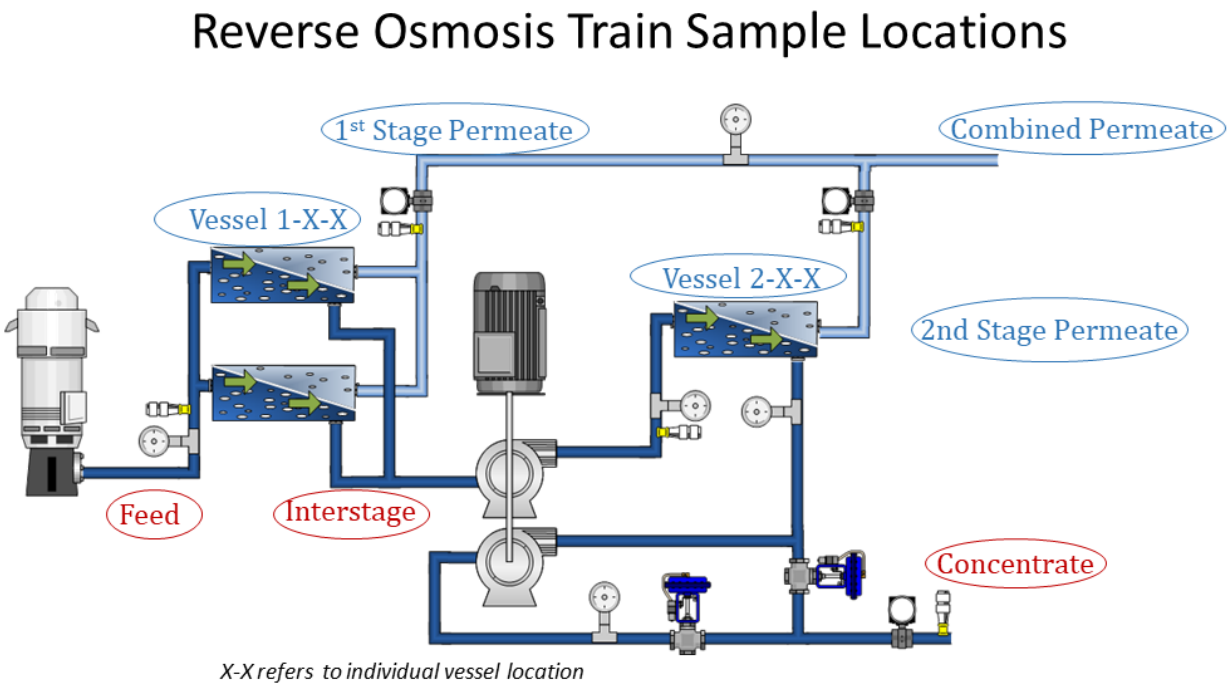

Figure 6-3. Sampling Locations of the YVWD RO System.

The traditional measurement of removal for compliance reporting is from the bulk feed to combined permeate. For the purposes of this study, additional data were collected to characterize the removal performance at intermediate locations (i.e., first stage permeate, interstage feed, and second stage permeate), as well as from selected pressure vessels to provide a better understanding and characterization of the removal performance across the system.

6.2.2 MS2 Challenge Testing

6.2.2.1 MS2 Stock Solution and Spiking

MS2 bacteriophage ATCC 15597-B1 was procured from GAP EnviroMicrobial Services (GAP, London, Ontario, Canada). The neat solution (shown in Figure 6-4) as received from the laboratory was immediately placed inside a cooler with ice cubes to maintain between 4-8 degrees Celsius (°C) if used the same day or stored in a refrigerator until testing occurred. The neat MS2 solution had a pH of 6.3 and an EC of 14.3 millisiemens per centimeter (mS/cm).

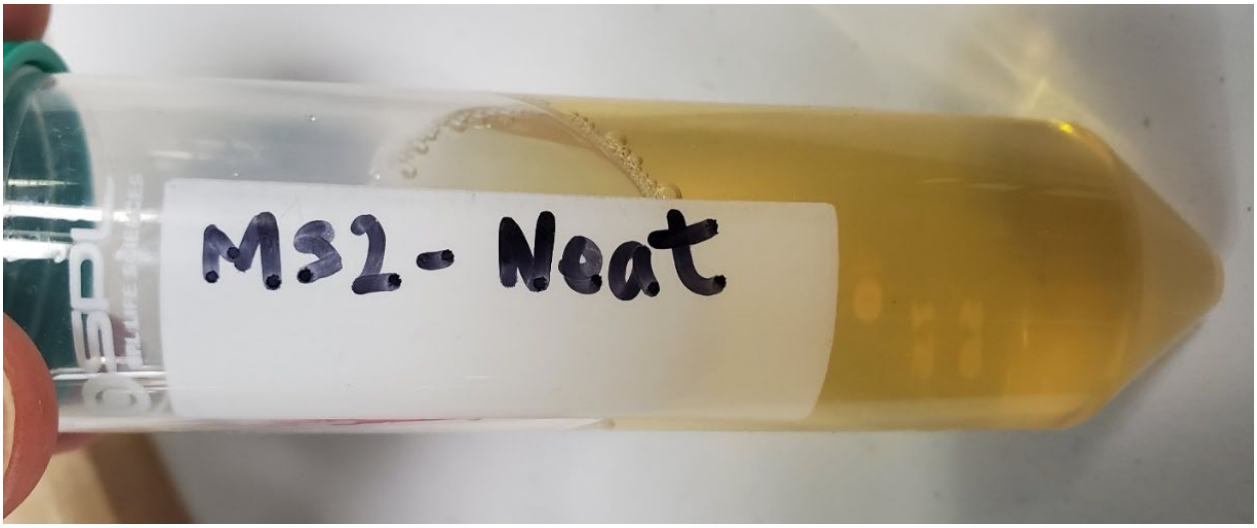

Figure 6-4. Sample of MS2 Bacteriophage Neat Solution as Sent from GAP EnviroMicrobial Services.

Immediately prior to dosing, the neat MS2 solution was diluted by adding 1.84 liters (L) of neat MS2 solution (~4-5 x 10^{11} PFU/mL) and 4.7 L of RO feedwater in a clean 5-gallon plastic pail. In order to induce further mixing, the stock solution was dosed into the RO feed immediately upstream of the high-pressure feed pump using a Cole Parmer MasterFlex Easy-Load L/S peristaltic pump (Vernon Hills, IL) at a flow rate of approximately 122 mL/min for a final target RO feed concentration of 3-5 x 10^{6} PFU/mL. MS2 bacteriophage was dosed for a total of 25 to 30 minutes per challenge test condition to allow sufficient time to mix through the RO skid.

As shown in Figure 6-5, a clean 1-gallon bottle of water (without stickers) was placed inside the pail to increase the top surface elevation of the solution in the pail and avoid having low levels of stock solution which could result in feeding lower than intended dosages.

Figure 6-5. MS2 Pumping Setup Upstream of RO High-Pressure Feed Pump at YVWD.

6.2.2.2 MS2 Testing Sample Controls

Blank samples of the RO system bulk flows were collected before the MS2 bacteriophage was dosed to confirm that values used in the LRV calculation were not based on indigenous background male-specific coliphage concentrations which can be detected by the MS2 assay. In addition, the blank samples serve to confirm that sampling practices were sufficient to avoid contamination. The following process locations were sampled prior to commencement of the challenge test:

- RO Feed.
- Interstage Feed (Stage 1 Concentrate).
- Concentrate.
- First Stage Permeate.
- Second Stage Permeate.
- Combined Permeate.

For this study, MS2 values are the result of triplicate plating, which means that from one 50-mL sample, three different values (PFU/mL) are obtained for the same stream (e.g., feed, min vessel, maximum, etc.). Creating triplicates of the microbial analysis assures the experimental validity of the MS2 testing results. To improve reliability in the experimental data, two separate triplicate samples were collected of the feed and the combined permeate streams. As a result, the LRV calculation was performed based on six data points for each stream (i.e., feed and combined permeate). The geometric mean approach was used to average the MS2 concentrations of the triplicates.

6.2.2.3 MS2 Sample Collection Protocol

It was important to carry out MS2 testing with practices that would not contaminate or degenerate the quality of the samples. Because RO permeate has a low electrical conductivity (~30 µS/cm) and a pH near 5.5, so precautions were taken to ensure that the MS2 would remain viable in the permeate matrix until sample analysis. Phosphate buffered saline (PBS) solution (ThermoFisher Scientific, Waltham, MA) was used to prepare a diluted PBS solution for addition to sample collection vials as a MS2 preservative. . Before sample collection, each permeate tube was dosed with 1 mL of the PBS solution to increase the pH from 5.2 to 5.9 standard units, and the conductivity from 29 to 410 µS/cm in an effort to avoid osmotic shock of MS2. Osmotic shock in the RO permeate was anticipated to result in inactivation of the MS2 an over estimation of LRV.

Once MS2 dosing began, no samples were collected for at least 10 minutes to turn over the volume in the RO system more than five times, allowing the MS2 spiked feedwater to reach steady state within the RO system. All sampling ports remained opened (Figure 6-6) for the first 10 minutes after dosing began to thoroughly flush the system. After 10 minutes of MS2 dosing, all sample lines were closed to minimize chances of incidental cross contamination, and sample collection began. Each sample collection involved: wiping down the sampling port with a paper towel, flushing the sampling line for one minute, and then collecting a 50 mL sample in a 50-mL sterile centrifuge tube.

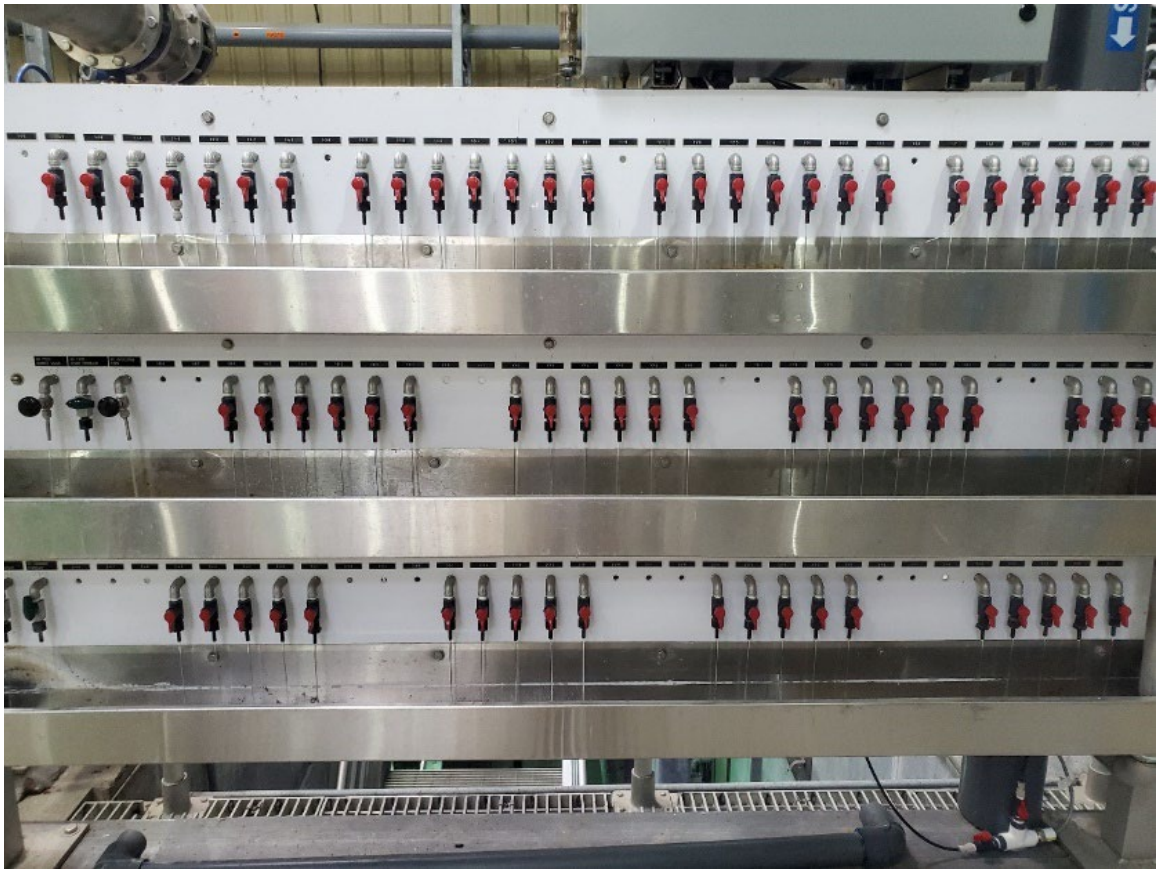

Figure 6-6. YVWD RO Sampling Ports Open for Flushing During Initial MS2 Dosing.

MS2 analysis of the collected samples was carried out by GAP EnviroMicrobial Services (London, ON Canada). Samples were shipped in a cooler with wet ice and ice packs for overnight delivery. MS2 samples collected at Wochholz were received by GAP typically at a temperature of 3°C, and their analysis commenced within 24 hours after sample collection.

6.2.3 Water Quality Parameters

All sampling of feed and permeate streams was performed using the RO vessel sampling ports (Figure 6-6).

Conductivity measurements were taken utilizing a Myron L TechPro II (Carlsbad, CA) which was calibrated prior to use with a 1413 µS/cm standard solution.

The pH of samples was measured with a Hach DR 900 (Loveland, CO) pH probe and calibrated prior to use with pH 4.0, 7.0, and 10.0 standard solutions.

The antiscalant and sulfuric acid for pH adjustment continued to be dosed during testing. The chloramination system was shut down prior to MS2 dosing to prevent inactivation of the dosed MS2 bacteriophage. The free chlorine and total chlorine concentrations were verified to be non-detect (<0.1 mg/L) using a Hach Pocket Colorimeter II (Loveland, CO).

Testing of sodium and chloride rejection by the RO unit was performed on February 19, 2020. Samples were shipped to Eurofins CalScience (Garden Grove, CA) and tested by Method 300.0 and Method 200.8 for chloride and sodium, respectively. The Method Detection Limit was 1.0 mg/L for chloride and 0.1 mg/L for sodium. Based on the laboratory results, the RO system rejected 98.3% of chloride and 95% of sodium.

6.2.4 Simulated Integrity Failure

Two methods were undertaken to artificially induce integrity failure and test the response of MS2 LRV as well as other surrogates of RO integrity. The two methods were 1) intentionally compromising an end adapter and 2) installation of an NF membrane in place of an RO membrane.

6.2.4.1 Compromised End Adapter

A pressure vessel end-adapter was drilled to yield a 1/16-inch orifice (Figure 6-7). The compromised end adapter substituted an integral end adapter in a randomly chosen vessel (Vessel Number 1-3-3). The end adapter was installed at the tail end of the pressure vessel so that it would be challenged with the more concentrated feed compared to the lead element.

The hole drilled in an end-adapter was considered to be representative of the type of damage that may result from a failure mode where complete bypass of the membrane barrier was possible. Examples of such failure modes include:

- A torn section of membrane (penetration of both selective polyamide layer and supporting polyethersulfone [PES] microfiltration layer) or
- a defective O-ring or interconnector seal.

Figure 6-7. End Adapter with an Intentionally Drilled 1/16-inch Orifice.

6.2.4.2 Substitution of an NF element

When membranes are exposed to certain disinfectants or cleaning solutions, the membrane chemistry may be affected, and the polyamide chains of the active layer could be cleaved (Antony et al. 2010). Such phenomenon would manifest itself as a decrease in rejection of ions. Previous studies intentionally oxidized membranes by exposing them to a solution containing free chlorine (Antony et al. 2016, 2010; Jacangelo et al. 2019). The effect is a reduction in salt rejection, which depending on the duration of exposure, an oxidized membrane could entirely lose its salt rejection properties.

The NF membrane was substituted with an RO membrane to simulate the rejection properties of an oxidized membrane without irreversibly damaging the full-scale assets at YVWD. The chosen NF element was the CSM NE8040-40 rated at 20 to 40% rejection of sodium chloride compared to the RO membrane rated at 99.7% nominal salt. According to the manufacturer, the permeate flow of the NF element is rated at 12,000 gpd compared to 10,500 gpd for the RO membrane element substituted during the challenge test. Given the increased nominal permeability of the NF membrane there may have been preferential flow through this element, relative to other RO elements in the same vessel, increasing the severity of simulated loss in salt rejection.

The brand-new NF element was acquired as donation from Toray in February 2020, and was stored dry, in sealed plastic, boxed in cardboard, and at room temperature until used for testing (Figure 6-8). The NF element was installed in the tail position of pressure Vessel Number 1-3-3. After installation of the NF element in Vessel Number 1-3-3, the RO system was operated for nearly 20 hours before the MS2 bacteriophage challenge test was conducted. This acclimation period was to allow the NF membrane element to equilibrate to the pressure and water flow conditions.

Figure 6-8. Nanofiltration Element Used to Substitute an RO Element at the Tail of Vessel 1-3-3.

The NF membrane is rated at 99% rejection of divalent ions (i.e., magnesium sulfate) and its free volume or pore size available for water transport ranges from 1 to 2 nm (Hosseini et al. 2016). Although the precise pore size of the NF membrane used in this study is proprietary, this type of membrane is targeted toward the larger pore size within the NF range (the pore size of RO membranes is estimated to be less than 1 nm). Given that MS2 bacteriophage is approximately 25 nm and larger than the sulfate ion, rejection of MS2 bacteriophage should be unchanged when comparing the RO to the NF membrane. Therefore, it was hypothesized for this testing that the salt rejection will decrease, and that the conductivity profile will recognize the change as a compromise (i.e., the rejection of salts will decrease but the virus LRVs should remain constant).

6.2.5 Uranine Challenge Testing

Uranine was also tested at YVWD as another potential integrity monitoring surrogate. The uranine challenge test on the RO system was conducted within a day of performing the MS2 challenge test and the sulfate indirect integrity test in order for flow conditions to be consistent across these three tests (i.e., "Normal Ops" tests with MS2, uranine, and sulfate had identical test conditions).

Studies have shown that uranine fluorescence is strongly dependent on pH with increasing fluorescence for more basic pH values (Blom et al. 2016; Gerke et al. 2013). The RO feed and permeate pH at Wochholz is 6.5 and 5.8, respectively, which would reduce the amount of fluorescence from the permeate samples relative to the feed. Therefore, pH of the samples collected at YVWD were adjusted in order to improve the representativeness of uranine challenge test results. Tris Base buffer was added to each sample in order to attain a pH of 8.2 for all samples collected at Wochholz (Tris Base molecular weight [MW] of 121.14 g/mol, Millipore-Sigma, St. Louis, MO). The pKa of the buffer is 8.0 and is stable from pH 7 to 9. Tris Base buffer solutions were prepared with ASTM Type II deionized water (Labchem, Zelienople, PA).

The effectiveness of a tracer test is rooted on the ability to conserve the tracer throughout the system. The uranine molecule can undergo photolysis, which is the breaking of its molecular structure by light. For this reason, precautions needed to be taken to preserve uranine solutions, including calibrating solutions, the feed solution, and the collected samples during uranine testing. Photolysis was prevented by preparing solutions immediately prior to their use and storing them in amber glass bottles. The feed bin and sample tubes were translucent; therefore, those containers were wrapped in aluminum foil to keep the solution away from light (Figure 6-9).

Figure 6-9. Dilutions, Uranine Stock Solution, and Sample Tubes.

Before uranine dosing, blank samples of every RO system stream were collected to determine any background fluorescent natural matter. The uranine stock solution was prepared in a clean 13-gallon bin wrapped in aluminum foil. The uranine solution was made by mixing 1.53 kilograms (kg) of uranine in powder form (MW of 376.27 g/mol, Millipore-Sigma, St. Louis, MO) with 4.4 gallons of bottled water (Crystal Geyser, Calistoga, CA). The pH of the bottled water was adjusted to 8.7 by adding aliquots of 1.0 M Tris Base buffer (Millipore-Sigma, St. Louis, MO) prior to adding the uranine. The uranine stock exhibits a dark red color at high concentrations and becomes a bright yellowish green color when diluted.

The uranine stock solution was prepared to have a concentration of 93 g/L and was dosed at 122 mL/min with a peristaltic pump (Cole Parmer MasterFlex Easy-Load L/S, Vernon Hills, IL) into the feed pipe ahead of the high-pressure feed pump (the pump and pumping conditions were identical to MS2 dosing experiments). The resulting uranine concentration in the RO feed was 2 mg/L - the same target used for testing at OCWD as discussed in Chapter 5.

The uranine spiking and sampling protocol was the same as for MS2 with respect to sample line flushing and system equilibration (See Section 6.2.2.3).

The fluorescence of uranine was analyzed onsite with a fluorometer TD-10AU (Turner Designs, Sunnyvale, CA). The instrument was calibrated prior to sample collection. The solutions that were used for calibration were (1) deionized water (LabChem, Zelienople, PA) to set a zero and (2) 40 µg/L of uranine to set the maximum reporting limit. This low range was chosen to read out accurately at low concentrations anticipated in the permeate, which also allowed the instrument to work in the range of linearity and avoid saturated readings.

Working with 40 µg/L as an upper limit in the reading ability of the fluorometer required that samples with known higher concentrations had to be diluted prior to sample analysis. The feed, interstage feed, and concentrate samples were diluted for their concentrations to be 40 parts per million (ppm). The pH of the dilution water was adjusted to 8.2 by adding three drops of 1.0 M Tris Base buffer.

Similar with the OCWD study, samples were collected for all pressure vessels and all bulk flows in order to generate a uranine concentration profile across the entire RO system. Flow and other operational conditions were identical to the "Normal Ops" test conditions used during the MS2 and sulfate tests carried out in November 2020.

6.2.6 Sulfate Challenge Testing

Samples for analysis of sulfate across the RO system at Wochholz were collected the same day as the MS2 challenge test in November of 2020 (this was part of the "Normal Ops" tests carried out consecutively to MS2 and uranine). A volume of 250 mL was collected for each sample. Samples were packaged with wet ice in a cooler and shipped overnight to the analytical laboratory. Sample analysis was carried out by Eurofins Calscience (Garden Grove, CA) following EPA Method 300.0 utilized to determine the concentration of inorganic ions by ion chromatography. The analytical instrument was a High-Performance Liquid Chromatography Ion Chromatography (HPLC/IC) with a method detection limit (MDL) of 0.24 mg/L and a reporting limit (RL) of 1.0 mg/L of sulfate.

Sulfate samples were collected for all bulk flows across the RO system as well as the selected pressure vessels that were sampled during the MS2 "Normal Ops" challenge test. The same pressure vessels were chosen with the purpose of establishing a methodical performance comparison across integrity indicators.

6.3 YVWD Results

The rejection properties of the full-scale RO system at YVWD were evaluated under the conditions shown in Table 6-4. In order to correlate marker-based surrogates to virus removal, the reduction of MS2 as a surrogate to enteric viruses was tested in a variety of conditions – before and after cleaning of the membranes, under normal operations, or under intentionally compromised conditions.

Table 6-4. Tests Performed on RO Unit at YVWD.

Test Condition or Variable	Indicator	Type of Test	Date
Test 1: Pre-CIP	MS2	Challenge Test	2/19/2020
Test 2: Intentionally Compromised End Adapter	MS2	Challenge Test	2/20/2020
Test 3: Post-CIP	MS2	Challenge Test	3/18/2020
Test 4: NF Element Substitutes RO Element	MS2	Challenge Test	9/16/2020
Test 5: Normal Ops	MS2	Challenge Test	11/18/2020
Test 6: Normal Ops	Uranine	Challenge Test	11/19/2020
Test 7: Normal Ops	Sulfate	Indirect Integrity Test	11/18/2020

6.3.1 Flow and Conductivity Data During Tests

The flow and conductivity of each online monitored point during the tests at YVWD is summarized in the tables below.

Table 6-5. System Flows and Recovery During each Testing Event.

Test	Parameter	Units	Stage 1	Stage 2	Unit
Test 1: Pre-CIP (2/19/2020)	Feed Flow	gpm	1,330	516	1,330
	Concentrate Flow	gpm	516	231	231
	Permeate Flow	gpm	814	285	1,099
	Recovery	%	61	55	82.6
Test 2: Compromised End Adapter (2/20/2020)	Feed Flow	gpm	1,330	516	1,330
	Concentrate Flow	gpm	516	231	231
	Permeate Flow	gpm	814	285	1,099
	Recovery	%	61	55	82.6
Test 3: Post-CIP (3/18/2020)	Feed Flow	gpm	1,894	750	1,894
	Concentrate Flow	gpm	750	290	290
	Permeate Flow	gpm	1,192	430	1,622
	Recovery	%	63	57	86
Test 4: NF Element Substituted (9/16/2020)	Feed Flow	gpm	1,941	720	1,941
	Concentrate Flow	gpm	720	291	291
	Permeate Flow	gpm	1,221	429	1,650
	Recovery	%	63	60	85
Test 5 and 7: Normal Ops (11/18/2020) Test 6: Normal Ops (11/19/2020)	Feed Flow	gpm	1,500	612	1,500
	Concentrate Flow	gpm	612	300	300
	Permeate Flow	gpm	888	312	1,200
	Recovery	%	59	51	80

The conductivity of bulk streams during each test, as well as temperature is summarized in the table below.

Table 6-6. Conductivity and Temperature During each Test Event.

	Conductivity (µS/cm)				
Sampling Location	Test 1	Test 2	Test 3	Test 4	Tests 5,6 and 7
Feed	776	758	754	744	770
Interstage Feed	1,881	1,673	1,859	1,644	1,671
Concentrate	4,247	3,179	4,204	3,105	3,164
First Stage Permeate	19.5	21	47	37.8	35.4
Second Stage Permeate	40	34.1	44.4	57.7	53.8
Combined Permeate	22.7	24.4	28.7	43.0	40.2
Feed Temperature (˚F)	69.9	68.1	63.2	83.7	76
Abbreviations: ˚F - degrees Fahrenheit.					

6.3.2 Test 1 Pre-Clean in Place

A challenge test of the RO system was carried out on February 19, 2020, at YVWD. Initially the test was intended to be performed at design parameters (as directed by the USEPA Membrane Filtration Guidance Manual). However, the second stage of the RO system was challenged by fouling, inhibiting the ability of the system to generate the design permeate flows without increasing the feed pressure to unsustainable levels. Thus, this test was useful as a Pre-CIP test in order to systematically compare against the MS2 reduction efficiency after cleaning. The flows and conductivity observed during Test 1 are summarized in Table 6-5 and Table 6-6. Total permeate flow was lower for subsequent tests.

To correlate virus reduction, salt passage, and system integrity, before conducting the MS2 challenge test, a conductivity profile was generated and is shown in Figure 6-10. The pressure vessels with median and highest conductivity values are shaded in orange and green (first and second stage, respectively).

SPI had been analyzing the automatic conductivity profile reports, and it was noted that Vessel Number 1-5-5 was triggering the Average + 3σ alarm in the first stage for several weeks prior to Test 1. For the second stage no alarms were being triggered, but Vessel Number 2-4-5 was consistently higher than the median of that stage by approximately 16%.

Besides sampling all bulk flows in the RO system for MS2 bacteriophage, the two pressure vessels with the maximum permeate conductivity were sampled during MS2 dosing to determine their virus rejection properties. Also, one pressure vessel with the median permeate conductivity was sampled for each stage.

Results from sample analysis indicated the RO feed was dosed with 5.5×10^6 PFU/mL during this MS2 challenge test. The pressure vessel with the highest permeate conductivity for the first stage (Vessel Number 1-5-5) attained 2.4-log of MS2 reduction (Figure 6-11). It was later discovered that Vessel Number 1-5-5 had a cracked element (see Section 4.4.5), which is attributed as the cause for higher MS2 passage. As a comparison, the pressure vessel with the median permeate conductivity (Vessel Number 1-3-5) attained 3.9-log MS2 reduction.

Although Vessel Number 1-3-5 attained higher reduction than Vessel Number 1-5-5, Vessel Number 1-3-5 attained lower LRVs compared to the overall first stage permeate, which leads to the conclusion that the MS2 concentration in Vessel Number 1-3-5 might have been the result of cross contamination during sample collection.

LRVs were calculated based on the results of MS2 concentration analysis. For regulatory purposes, the two-stage RO system is treated as a single unit with the LRV to be calculated as the log of the ratio of the feed concentration to the combined permeate concentration. For the Pre-CIP MS2 test, the RO system attained an LRV of 6-log, which means the RO system rejected 99.9999% of MS2 bacteriophage, even while having a cracked element in Vessel Number 1-5-5.

17.6	17.3	18.2	18.7								
19.5	19.8	18.1	17.8	17.7	19.0	19.2	20.5				
18.4	19.4	19.2	18.9	22.8	17.8	18.3	19.6	39.4	42.6	36.5	45.6
19.3	20.0	20.0	18.4	19.6	20.7	19.8	19.5	41.1	38.5	35.3	39.3
20.0	20.6	18.4	18.2	18.4	18.6	19.0	19.7	39.4	35.6	39.8	41.7
19.0	18.1	18.9	21.0	19.9	20.3	19.9	17.4	42.3	41.3	39.4	38.9
21.5	22.1	19.4	18.2	20.3	20.6	19.2	17.9	40.1	35.9	35.8	36.4
FIRST STAGE								**SECOND STAGE**			

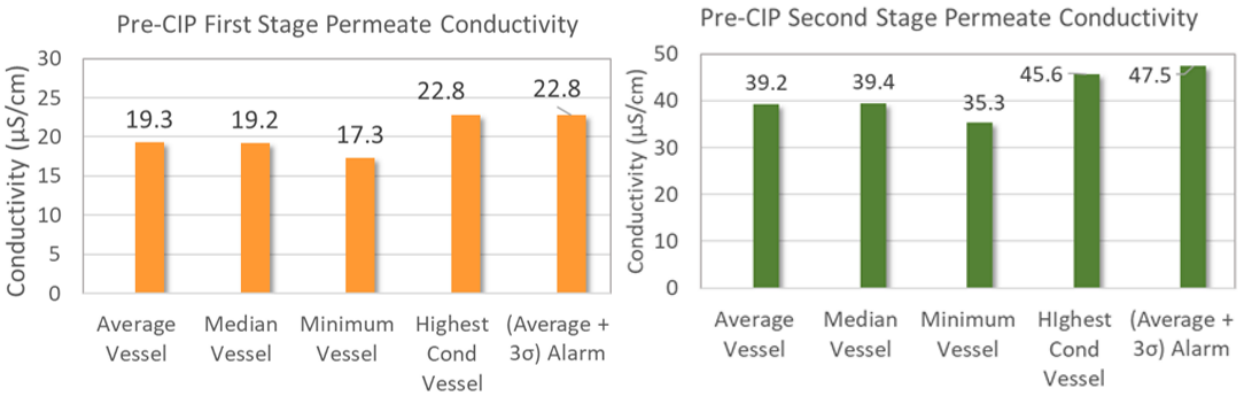

Figure 6-10. Manual Conductivity Profile Prior to MS2 Dosing for Test 1 - Pre-CIP.

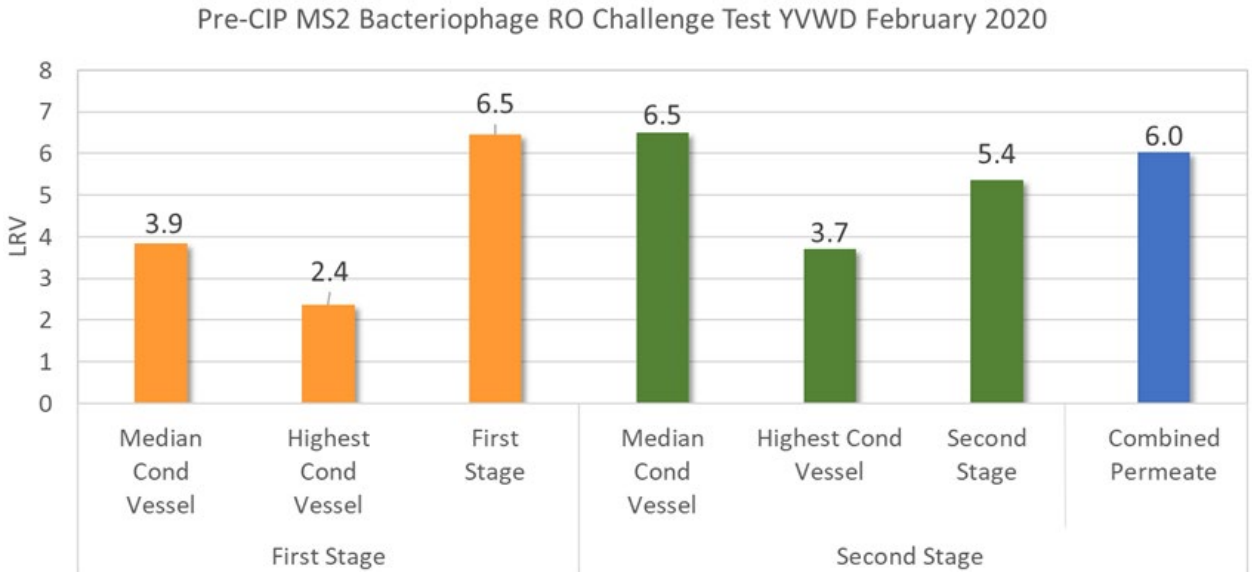

Figure 6-11. MS2 LRVs Attained by the YVWD RO System During Test 1 - Pre-CIP MS2 Test.

6.3.3 Test 2 Compromised End Adapter

An MS2 challenge test was carried out on the RO unit at Wochholz on February 20, 2020, after substituting a pressure vessel tail-end adapter with one that had a 1/16th inch orifice purposefully drilled (Vessel Number 1-3-3 was chosen randomly for this substitution).

This test took place the day after the Pre-CIP challenge test, thus the operating conditions were identical to the Pre-CIP test, as detailed in Table 6-5.

The results of the conductivity profile identified the compromised end adapter with higher permeate conductivity in that vessel (i.e., a detectable integrity issue). In Figure 6-12, shaded in orange and green are the pressure vessels that were flagged because they qualified as any of the following for the first and second stage:

- A pressure vessel with median permeate conductivity from each stage,
- The pressure vessel with the highest permeate conductivity from each stage, and
- The pressure vessel with the installed intentionally compromised end adapter (1/16th inch orifice).

As shown in Figure 6-12, the compromised end adapter vessel (Vessel Number 1-3-3) produced permeate with electrical conductivity of 45.6 µS/cm which was 70% higher than the average of that stage.

Figure 6-13 illustrates the calculated LRVs from this test. The RO feed had an MS2 concentration of 3.8×10^6 PFU/mL and the RO system attained 5.2-log of MS2 reduction even with the intentionally compromised end adapter in Vessel Number 1-3-3.

The median permeate conductivity vessel (Vessel Number 1-1-4) attained 7.4-log of MS2 reduction. Vessel Number 1-3-3 (vessel with compromised end adapter) attained 1.9-log of MS2 reduction, which is in the range of the typical LRV awarded to an integral RO system when calculated from bulk feed and permeate TOC or conductivity measurements. Besides the vessel with the compromised end adapter, Vessel Number 1-5-5 had shown to be potentially compromised in previous conductivity profiles, and its MS2-based LRV remained at 2.4-log for that individual vessel for this test condition and in agreement with the results of test 1.

None of the end adapters were compromised in the second stage, however Vessel Number 2-4-5 was recorded having the highest permeate conductivity of the second stage. As the highest permeate conductivity vessel in the second stage, Vessel Number 2-4-5 attained 4.9-log of MS2 reduction which was lower than the median vessel for the second stage which attained 6.7-log of MS2 reduction.

24.7	24.5	25.0	25.0								
26.0	26.4	25.5	25.0	24.8	26.1	26.3	27.8				
25.4	26.6	26.5	25.9	30.3	25.2	25.6	26.6	42	45.1	38.9	47.4
26.3	26.6	26.4	25.4	26.7	28.4	27.2	26.7	43.9	41.4	47.1	42.1
26.5	27.4	45.6	25.4	25.6	29.5	25.8	26.2	43.4	39	42.3	44.3
26.0	25.0	25.8	27.7	27.4	27.6	27.3	24.8	44.9	44	42.4	41.8
29.1	29.1	26.7	25.3	26.9	27.2	26.0	25.1	42.9	39	39.1	39.4
FIRST STAGE								**SECOND STAGE**			

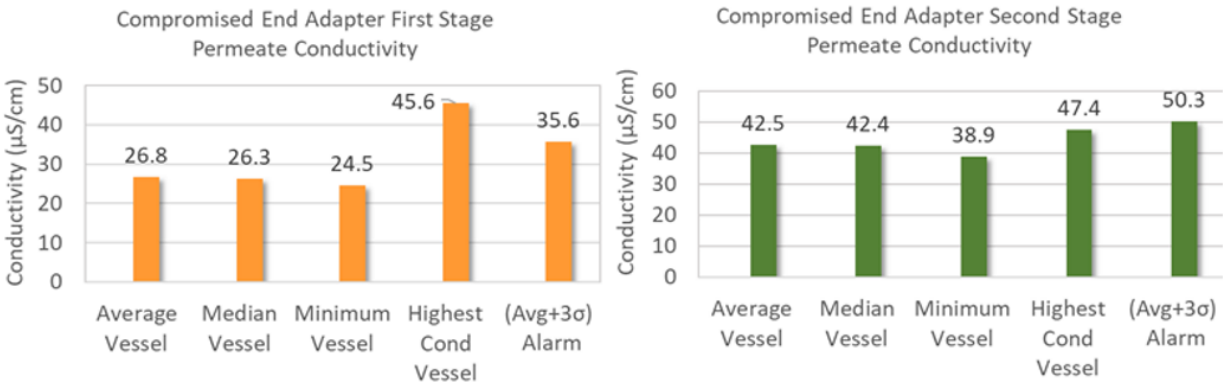

Figure 6-12. Manual Conductivity Profile Before MS2 Dosing for Test 2 - Compromised End Adapter.

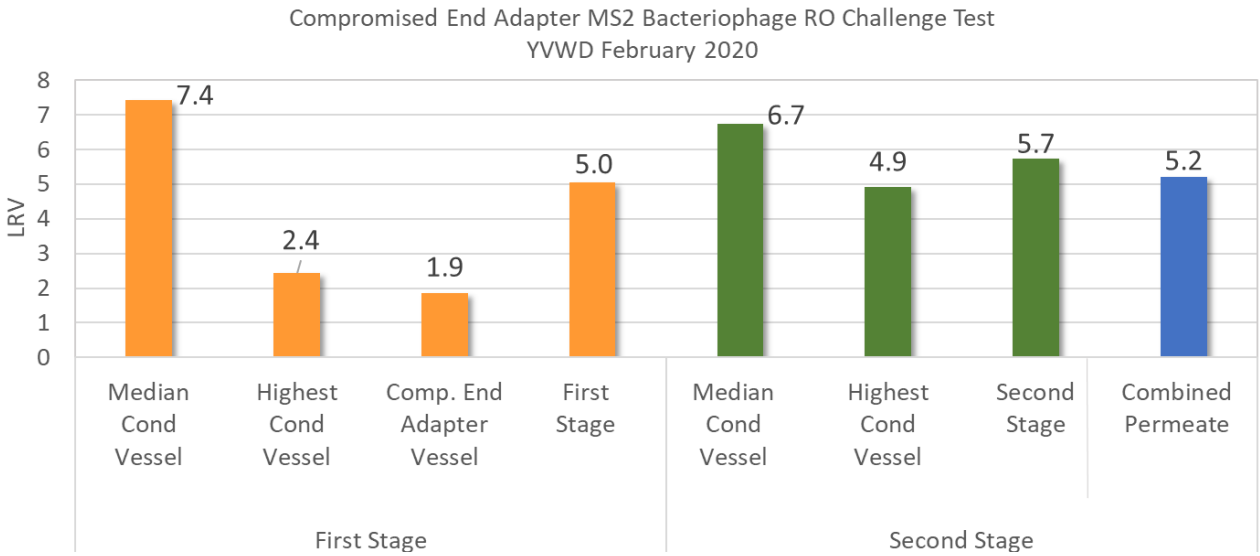

Figure 6-13. MS2 LRVs Attained by the RO System During Test 2 - Compromised End Adapter.

6.3.4 Test 3 Post-Clean in Place

On March 17, 2020, the RO system at YVWD underwent a CIP procedure. The CIP consisted of citric acid cleaning for two hours, followed by two hours of cleaning with a high pH solution (pH 10.5-11.0) at 105°F.

Prior to conducting the challenge test, the RO system was shut off and the cracked tail element in Vessel Number. 1-5-5 was replaced. The system was flushed with RO feed and operated at design conditions for at least 20 hours to reach steady state with the newly installed element prior to MS2 challenge testing.

The CIP procedure was successful in removing foulants and returning the permeate flow to design conditions; therefore, the flow conditions during the MS2 Post-CIP challenge test were higher for the Pre-CIP and the Compromised End-Adapter test, as listed in Table 6-5.

The Post-CIP MS2 challenge test took place on March 18, 2020, with a feed conductivity of 754 µS/cm and a feed temperature of 63.2°F. A manual conductivity profile was generated to determine the overall performance of the system. Figure 6-14 shows the permeate conductivity values of all pressure vessels Post-CIP, which are, in general, higher than Pre-CIP values. This is due to the fact that when the RO membrane is exposed to a high pH cleaning solution, the membrane material swells and salt rejection could potentially decline. Vessel Number 1-5-5, which had the tail element replaced, became the vessel with the minimum permeate conductivity. Vessel Number 1-1-1 became the pressure vessel with the highest permeate conductivity in the first stage. Pressure vessels sampled for MS2 analysis are shaded in orange and green in Figure 6-14 for the first and second stage, respectively. These pressure vessels were flagged because they qualified as any of the following:

- A pressure vessel with median permeate conductivity from each stage,
- The pressure vessel with the minimum permeate conductivity from each stage,
- The pressure vessel with the highest permeate conductivity, or
- The pressure vessel with the second highest permeate conductivity.

23.9	24.3	24.3	25.3								
26.6	27.6	25.2	23.9	22.5	24.1	24.2	27.2				
26.2	27.7	25.9	25.9	21.3	23.3	23.7	26.3	42.5	47.5	39.1	50.5
27.8	27.6	27.5	25.6	25.0	27.1	26.2	26.4	45.9	43.0	50.0	41.5
28.2	29.4	26.4	25.8	23.8	24.0	24.3	26.4	44.2	39.4	43.3	46.9
27.1	25.5	27.0	30.0	25.4	27.0	25.4	22.5	46.1	44.2	42.7	41.5
31.8	30.2	28.1	25.3	25.7	26.2	24.8	23.3	42.9	40.5	39.2	40.2
FIRST STAGE								**SECOND STAGE**			

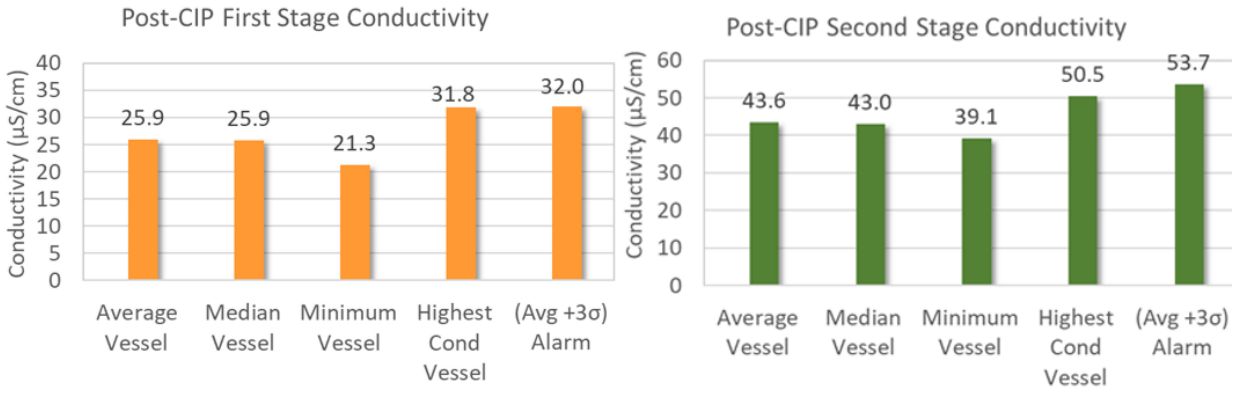

Figure 6-14. Manual Conductivity Profile Prior to MS2 Dosing for Test 3 - Post-CIP Test.

The RO feed had an MS2 concentration of 4 x 10^6 PFU/mL and a total of seven pressure vessels were sampled for their individual MS2 reduction. In the first stage, Vessel Number 1-1-1 and Vessel Number 1-2-1 were recorded as the pressure vessels having the highest and second highest permeate conductivity, respectively. Nonetheless, when considering MS2 reduction, Vessel Number 1-1-1 attained 5.2-log, which is significantly higher than the 3.4-log attained by Vessel Number 1-2-1. Even though Vessel Number 1-1-1 demonstrated slightly higher salt passage than Vessel Number 1-2-1, that did not translate to relatively high passage of virus.

In contrast to the LRVs attained by the pressure vessels with the highest permeate conductivity, Vessel Number 1-5-5, and Vessel Number 2-3-5, which exhibited the lowest permeate conductivity, also demonstrated a high capability for MS2 reduction achieving 7.1-log and 6.7-log for the first and second stage, respectively (Figure 6-15).

Although rejection of virus after a CIP was less than before the CIP, the results of MS2 analysis of the challenge test carried out Post-CIP demonstrated that the RO system at Wochholz was still capable of achieving greater than 5-log MS2 reduction (Figure 6-15) immediately after a CIP.

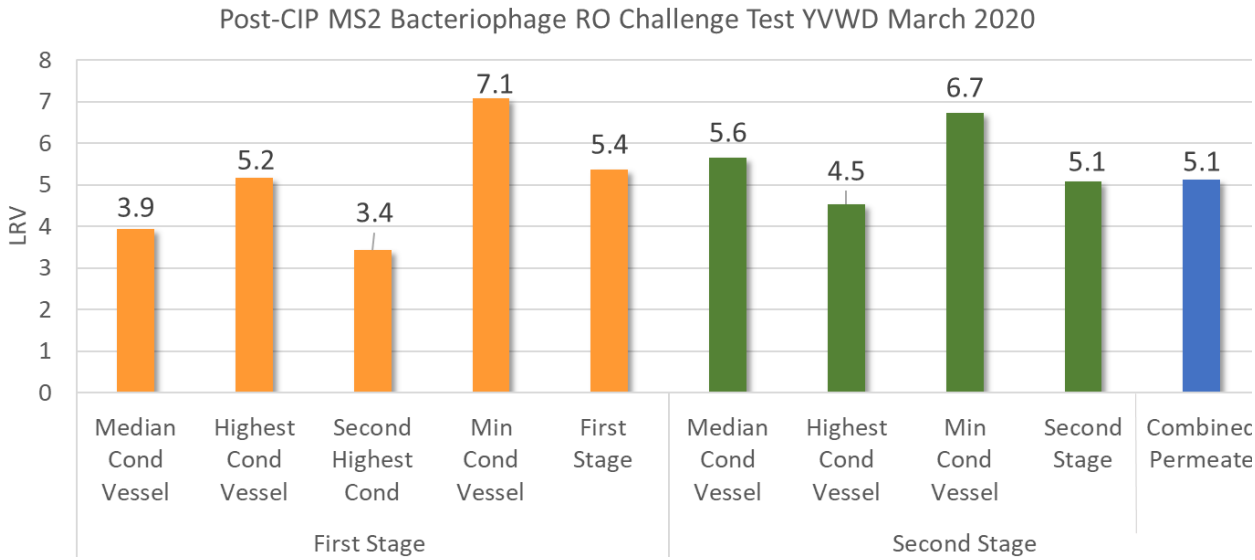

Figure 6-15. MS2 LRVs Attained by RO System During Test 3 - Post-CIP.

6.3.5 Test 4 Substitution of a NF Element to Simulate Oxidation

Test 4 was performed to demonstrate that it is possible for a "tight" RO membrane to lose rejection of total dissolved solids while maintaining a high rejection of virus. For this testing, the tail RO element from Vessel Number 1-3-3 was replaced with an NF element a day prior to the MS2 challenge test to allow the membrane to acclimate to the feedwater, pressure, and overall operating conditions of the RO system. The NF Element MS2 challenge test was carried out on September 14, 2020, with the flow rates shown in Table 6-5.

The manual conductivity profile collected after installing the NF element is shown in Figure 6-16. Vessels that were sampled for MS2 analysis are shaded in orange and green for the first and second stage, respectively, which were flagged because they qualified as any of the following for each stage:

- A pressure vessel with median permeate conductivity,
- The pressure vessel with the minimum permeate conductivity,
- The pressure vessel with the highest permeate conductivity,
- The pressure vessel with the second highest permeate conductivity, or
- The pressure vessel in the first stage that had the tail RO element substituted with an NF element.

26.9	26.9	27.1	27.4								
28.3	28.3	28.0	27.7	27.6	28.8	28.7	29.8				
27.7	29.3	28.7	28.1	26.2	28.2	28.8	29.0	47.2	50.2	44.6	54.4
28.3	28.6	28.7	27.7	29.1	30.6	28.3	29.1	49.6	47.0	54.5	46.8
29.1	29.8	192.6	28.2	28.5	28.7	28.7	28.4	48.6	44.5	47.7	50.3
29.5	28.0	28.8	30.2	29.8	30.0	29.6	28.0	50.0	48.6	47.7	47.0
31.6	31.2	29.1	28.3	29.7	29.8	28.9	28.4	47.5	45.0	44.5	45.3
FIRST STAGE								**SECOND STAGE**			

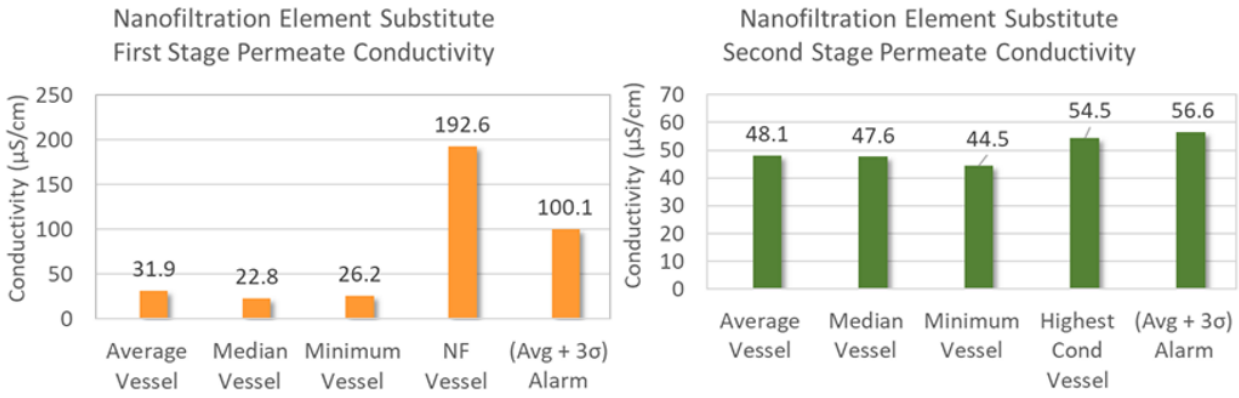

Figure 6-16. Manual Conductivity Profile Prior to MS2 Dosing for Test 4 - NF Element Substitution.

The dosed MS2 concentration in the RO feed was 5.7 x 10^6 PFU/mL. Figure 6-17 shows the LRVs at different points along the system to compare MS2 reduction by individual vessels.

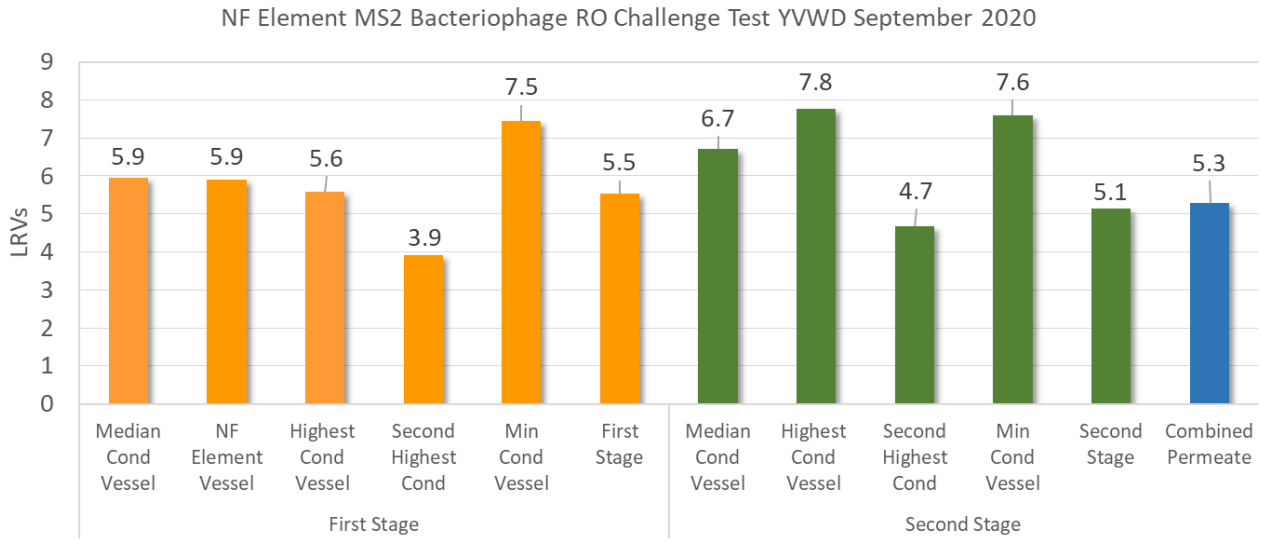

Figure 6-17. MS2 LRVs Attained by RO System During Test 4 - NF Element Substitution.

The vessel with the installed NF element (Vessel Number 1-3-3) had a permeate conductivity six times higher compared to the median of that stage. Even with such an increase in salt passage, Vessel Number 1-3-3 demonstrated an MS2 reduction of 5.9-log, which is equal to the pressure vessel with the median permeate conductivity, suggesting no change occurred in the MS2 reduction capabilities with the NF element substitute.

The second stage did not have any pressure vessels that exceeded the (Average +3*σ) alarm, although Vessel Number. 2-4-5 did exhibit a high permeate conductivity compared to the average of that stage. Vessel Number 2-4-5 also achieved 4.7-log of MS2 reduction which is lower than the 6.7-log attained by the pressure vessel with median permeate conductivity.

After substitution of a NF element for a RO element in Vessel Number 1-3-3 there was a noticeable spike in the conductivity for that vessel (six times the average permeate conductivity). Even with the intentionally added defect, the RO array still attained 5.3-log of MS2 reduction.

Vessel Number 1-2-1 was the second highest permeate conductivity for the first stage with a permeate conductivity of 31.2 µS/cm, which was relatively close to the 31.6 µS/cm of the maximum permeate conductivity (Vessel Number 1-1-1). However, when comparing the LRVs for Vessel Number 1-1-1 and Vessel Number 1-2-1, the latter attained 3.9-log compared to 5.6-log of virus reduction for Vessel Number 1-1-1.

A similar comparison between the two highest permeate conductivity vessels was observed in the second stage. Vessel Number 2-3-4 was recorded as having the highest permeate conductivity at 54.5 µS/cm and Vessel Number 2-4-5 had the second highest permeate conductivity at 54.4 µS/cm. Although their salt rejection was practically identical, Vessel Number 2-4-5 attained 4.7-log of MS2 reduction compared to the 7.8-log MS2 reduction of Vessel Number 2-4-5. In fact, Vessel Number 2-4-5, even with the highest permeate conductivity, did not pass much virus.

The pressure vessels with the minimum permeate conductivity for both the first and second stage (i.e., Vessel Number 1-5-5 and Vessel Number 2-2-3) did not pass much virus and attained 7.5-log and 7.6-log of MS2 reduction, respectively.

This test showed that an RO membrane that undergoes a loss in salt rejection does not necessarily represent a breach that will pass enteric virus. The RO system attained 5.3-log even though one RO element was replaced by a NF element that passed higher amounts of salt compared to the RO element. While conductivity profiling provides a means of fault detection, allowing the operators to pro-actively identify and correct system integrity issues, the increase in salt passage did not translate to a higher number of viruses in the permeate.

6.3.6 Tests 5, 6, and 7 Normal Operation with Sulfate and Uranine

In addition to the MS2 and conductivity profile testing under various operating conditions, three different indicators were utilized in November 2020 to measure the rejection performance and integrity of the RO system at Wochholz. The sampling campaign consisted of consecutive integrity tests analyzing the concentration in the permeate of (1) MS2 bacteriophage, (2) uranine, and (3) sulfate. All of these three tests were carried out at the same operating conditions (Shown in Table 6-5).

On November 18, 2020, a permeate conductivity profile was manually collected, illustrated in Figure 6-18. Shaded in orange and green (first and second stage, respectively) are the pressure vessels that were flagged because they qualified as any of the following for each stage:

- A pressure vessel with median permeate conductivity,
- The pressure vessel with the minimum permeate conductivity,
- The pressure vessel with the highest permeate conductivity, and
- The pressure vessel with the second highest permeate conductivity.

Figure 6-18 shows the results of calculating several statistics. In particular, the first stage had one pressure vessel that exceeded the Average +3σ alarm, whereas no pressure vessels in the second stage surpassed this set limit. Based on the results from this analysis, the shaded vessels (orange and green for the first and second stage, respectively) were sampled during the MS2, uranine, and sulfate tests to compare performance across indicators.

37.2	36.9	37.2	38.1								
39.2	38.6	38.1	38.0	35.7	35.8	36.3	37.8				
37.9	40.0	39.4	38.9	34.7	36.1	36.9	37.8	53.2	58.1	50.7	61.7
39.4	39.2	39.5	38.6	37.7	39.0	37.9	37.8	57.5	54.2	52.5	53.5
40.2	40.4	38.9	38.9	37.2	37.0	37.1	37.2	57.1	52.2	55.2	57.9
39.5	38.3	39.3	41.9	38.5	38.4	38.3	36.8	57.0	56.5	56.0	54.7
43.8	42.4	40.4	40.9	38.2	37.9	37.0	36.5	53.4	52.8	51.6	52.3
FIRST STAGE								**SECOND STAGE**			

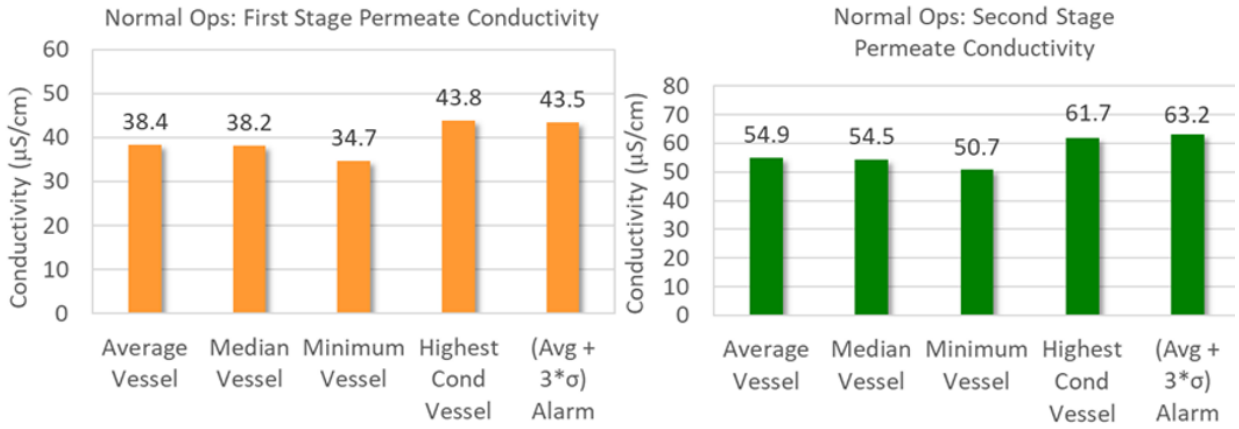

Figure 6-18. Conductivity Profile of the YVWD RO Prior to Normal Operation - Tests 5 (MS2), 6 (Uranine), and 7 (Sulfate).

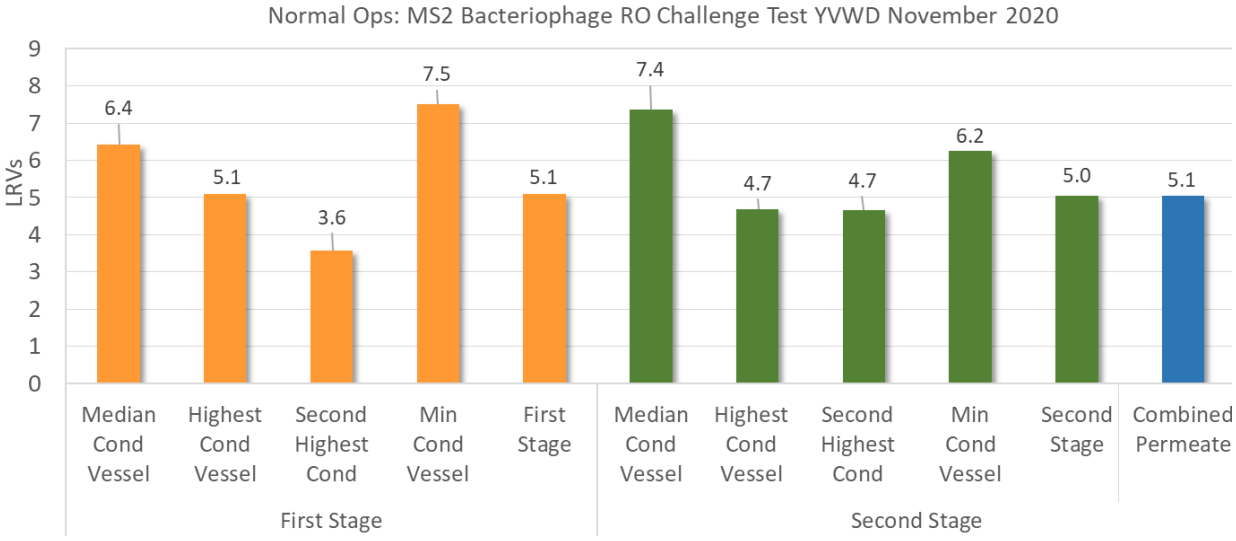

Figure 6-19. MS2 LRVs Attained by the YVWD RO System During Normal Operation - Test 5.

6.3.6.1 Test 5 - MS2 Challenge Testing Normal Operation

The feed conductivity was 770 µS/cm and the feed temperature was 76◦F. The MS2 concentration in the RO feed was 4 x 10^6 PFU/mL and resulted in the RO system attaining 5.1-log of MS2 reduction (Figure 6-19). Note, that Test 5 results are presumed to be comparable to sulfate and uranine results in Tests 6 and 7.

The pressure vessel in the first stage with the minimum permeate conductivity demonstrated the highest MS2 LRV, demonstrating 7.5-log. On the other hand, Vessel Number 1-2-1, which exhibited a high permeate conductivity, did suffer from higher passage of MS2, attaining only 3.6-log of MS2 reduction. Although Vessel Number 1-1-1 had the highest permeate conductivity for the first stage, Vessel Number 1-1-1 attained 5.1-log of MS2 reduction, reiterating that a higher permeate conductivity does not necessarily reflect a higher passage of virus. However, for the second stage, the two pressure vessels that exhibited the highest permeate conductivity also attained the lowest LRVs for MS2 reduction (both pressure vessels attained 4.7-log compared to the median of 7.4-log of MS2 reduction).

Overall, the RO unit attained greater than 5-log rejection of MS2 bacteriophage (i.e., 5.1-log of MS2 reduction) during normal operating conditions.

6.3.6.2 Test 6 - Uranine Challenge Testing and Profiling Normal Operation

The uranine challenge test was performed on November 19, 2020, with a feed conductivity of 813.6 µS/cm and a feed temperature of 76.4◦F. The RO feed pipe was injected with a stock solution to result in a final uranine concentration of 2 mg/L in the bulk RO feed.

Flow conditions were identical to the Normal Operation test as listed in Table 6-5 (i.e., permeate flow of 1,200 gpm operating at 80%t recovery). The dosed uranine concentration was visible to the naked eye as a light yellowish green solution which became a brighter color as was concentrated through the RO system (Figure 6-20).

Figure 6-20. Streams from Sampling Ports at YVWD During Uranine Dosing.

Permeate samples of every pressure vessel were collected to determine their uranine concentration (all bulk flows were sampled as well).

For consistency, LRV calculations were based on the measured fluorescence results rather than the targeted dosing feed concentration (i.e., LRV calculation was based on a feed concentration of 1.5 mg/L based on fluorometer reading, as opposed to 2.0 mg/L dosing target).

Shaded in orange and green for the first and second stage, respectively, are the pressure vessels that were flagged because they qualified during the Normal Operation permeate conductivity profile as any of the following:

- A pressure vessel with median permeate conductivity from each stage,
- The pressure vessel with the minimum permeate conductivity from each stage,
- The pressure vessel with the highest permeate conductivity, or
- The pressure vessel with the second highest permeate conductivity.

These vessels are based on the same conductivity profile collected the day prior to uranine testing. Table 6-7 lists the uranine concentrations of all bulk flows determined by fluorescence analysis.

Table 6-7. Uranine Concentrations Throughout the RO System.

Sampling Location	Uranine Concentration (µg/L)
Feed	1,473
Interstage Feed	3,479
Concentrate	7,309
First Stage Permeate	0.352
Second Stage Permeate	0.620
Unit Combined Permeate	0.419
Feed Temperature (˚F)	76

Figure 6-21 shows the profile of the uranine concentration across the two-stage RO system. In general, reduction of uranine by the RO system was high, with a uranine concentration in the combined permeate of only 0.419 ppb.

0.043	0.033	0.730	0.031								
0.026	0.028	0.620	0.553	0.039	0.079	0.085	0.071				
0.092	0.065	1.360	0.083	0.034	0.055	1.200	0.074	0.152	0.687	0.118	0.660
0.035	0.040	0.040	0.010	1.017	0.537	0.114	0.157	0.903	0.302	0.247	0.194
0.026	0.071	0.049	0.049	0.039	0.068	0.037	0.020	0.188	0.199	2.870	0.460
0.275	0.212	0.073	0.047	2.170	0.630	1.503	0.039	2.100	1.263	0.218	0.139
1.420	2.780	0.360	0.074	0.058	0.020	0.077	0.078	0.163	0.593	0.285	0.295
FIRST STAGE								**SECOND STAGE**			

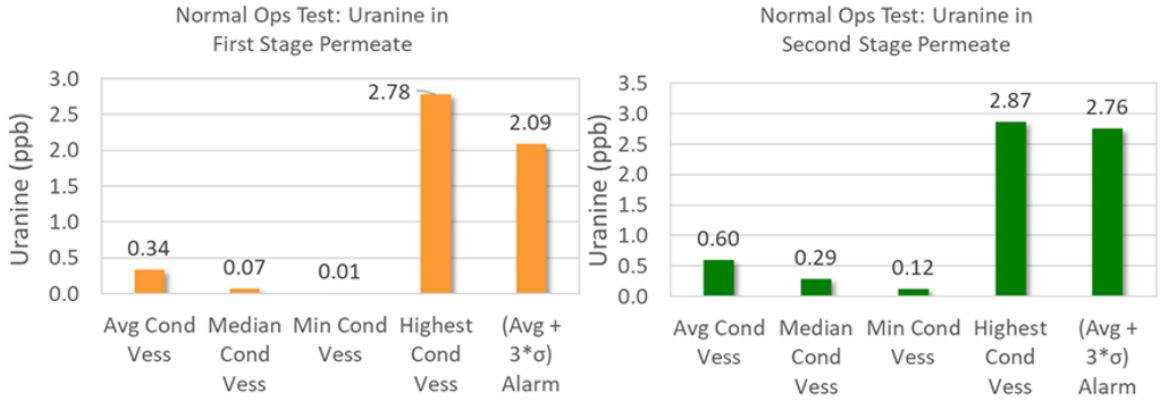

Figure 6-21. Uranine Profile of YVWD RO System During Normal Operation - Test 6.

The first stage had a few pressure vessels that were recorded as outliers with uranine concentrations higher than the median value by two orders of magnitude (minimum concentration of 0.010 ppb and the highest value was 2.78 ppb of uranine) as shown in Figure 6-21.

The uranine concentration in the interstage feed was 3.5 ppm, which as expected, made the average uranine concentration in the permeate of the second stage higher than the first stage (0.3 ppb for the first stage permeate compared to 0.6 ppb uranine for the second stage permeate).

In the first stage, Vessel Number 1-2-1 exhibited both the second highest permeate conductivity as well as the highest permeate uranine concentration. Vessel Number 1-1-1 had the highest permeate conductivity and a relatively high permeate uranine concentration compared to the median of that stage. Vessel Number 1-5-5 had the minimum permeate conductivity as well as one of the lowest permeate uranine concentrations. The permeate uranine concentration of Vessel Number 1-5-2 was unexpectedly high since the conductivity of that vessel is typically around the median.

The RO unit at Wochholz rejected 99.98% of the uranine fed into the system achieving a 3.5-log reduction of uranine (Figure 6-22). This information is relevant when considering other compounds or contaminants that have similar molecular weight and physical properties to predict rejection rate.

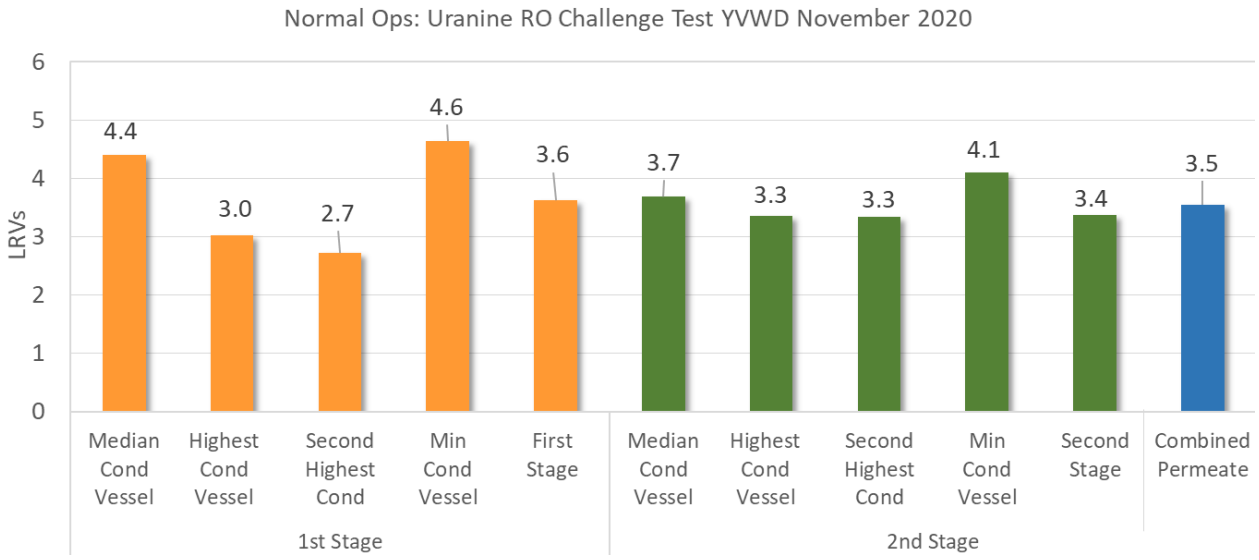

Figure 6-22. Uranine LRVs Attained by RO System During Normal Operation - Test 6.

Uranine has a molecular weight of 376 g/mol, which is relatively large considering that most molecules larger than 200 g/mol will be rejected by the RO membrane. The molecular weight of MS2 was estimated to be 3.6 x 10^6 g/mol (Kuzmanovic et al. 2003). Moreover, when comparing the size of uranine to that of a virus, uranine is much smaller than a virus by four orders of magnitude. Hence, uranine as a surrogate to virus is an extremely conservative approach because of their significant difference in size. In other words, the fact that a uranine molecule can pass the RO membrane does not necessarily imply that a virus can pass through that same

pathway or defect. Moreover, uranine membrane passage is more likely due to diffusion rather than bulk flow, in contrast to viruses which do not diffuse but only enter the permeate through a compromise (i.e., compromised membrane or faulty seals at O-rings).

A comparison of the conductivity profile versus the fluorescence profile for an integral RO system did not yield a direct correlation between the methods. Although there was a general trend associated with higher conductivity and higher fluorescence, the fluorescence profile yielded additional vessels with potential integrity defects. Such a small defect would result in higher passage of fluorescent compounds that would be minor and not associated with a significant change in overall conductivity that would represent an integrity concern. A similar observation was made during uranine profiling at OCWD (See Section 5.3.3.1).

Fluorescent compounds such as uranine have been historically described as being conservative indicators for virus removal. Because of the size differential of uranine and virus it would make sense that fluorescent compounds would appear before viruses.

Fluorescence testing for diagnostic purposes may be overly sensitive in terms of number of potential vessels that would be identified as a potential location of an integrity defect. From reviewing the other testing of vessels with high permeate conductivity, conductivity appears to be a sufficient “first pass” indicator of questionable integrity performance across a membrane unit for the purposes of diagnostic testing and is easier for utilities to manage compared to a spiked compound such as uranine.

6.3.6.3 Test 7 - Sulfate Rejection During Normal Operation

Sulfate (molecular weight 96 g/mol, charge of -2) monitoring is considered to be one of the most conservative indicators of membrane integrity because sulfate is orders of magnitude smaller than viruses. In order to be able to compare RO reduction of MS2, uranine, and sulfate, the same pressure vessels that were sampled for MS2 were sampled for sulfate immediately prior to carrying out the MS2 challenge test. Results of sampling and laboratory analysis determined sulfate concentrations at different locations within the RO system as shown in Table 6-8.

Table 6-8. Sulfate Concentrations Throughout the YVWD RO During Normal Operation - Test 7.

Sample Location	Sulfate Concentration (mg/L)
Feed	160
Interstage Feed	310
Concentrate	640
First Stage Permeate	0.26
Second Stage Permeate	0.30
Unit Combined Permeate	0.26
Feed Temperature (°F)	76

Part of the challenge of using sulfate as a quantitative marker is that RO membranes are particularly effective in removing sulfate. Therefore, the permeate concentrations are typically less than 1 mg/L, which can be a challenge to detect by less sensitive analytical probes. The MDL of the High-Performance Liquid Chromatography utilized in this study is 0.24 mg/L (RL is 1 mg/L). Results from the laboratory analysis determined the RO feed had a sulfate concentration of 160 mg/L, which was confirmed by having duplicate samples of the feed analyzed. The combined permeate samples were also collected in duplicates and averaged a sulfate concentration of 0.26 mg/L. Permeate sulfate concentrations are graphed in Figure 6-23 with the MDL of 0.24 mg/L plotted as a horizontal red line.

Figure 6-23. Sulfate LRVs Attained by the YVWD RO During Normal Operation - Test 7.

For the different pressure vessels tested for sulfate, a slight variation in the demonstrated LRVs was observed (Figure 6-23). Vessel Number 1-2-1 exhibited the second highest permeate conductivity and was recorded as having the highest permeate sulfate concentration. Vessel Number 1-2-1 exhibited the highest permeate conductivity and attained 2.6-log of sulfate reduction compared to 2.8-log attained by the median permeate conductivity vessel. Vessel Number 1-5-5, which was recorded as the minimum permeate conductivity, also had the lowest permeate sulfate concentration. However, the difference in sulfate concentration was so minor that it is not reflected in the LRV calculation when compared to the median vessel.

In the second stage, the vessel with the highest permeate conductivity exhibited the lowest sulfate LRV of 2.7-log compared to the 2.8-log of the pressure vessel with the minimum permeate conductivity.

Overall, the RO unit rejected 99.8% of the sulfate in the feed, achieving 2.8 LRVs of sulfate reduction.

6.4 YVWD Results

Table 6-9 lists the seven tests that were conducted at Wochholz during 2020 to demonstrate the integrity of the RO system and the number of attained LRVs (based on the log of the feed to the combined permeate). Table 6-9 also lists the LRVs that the system could demonstrate based on bulk feed and permeate conductivity as a surrogate indicator, based on current regulatory allowances in some states.

Table 6-9. Summary of Integrity Testing Conducted at YVWD.

Test Condition or Variable	Integrity Surrogate	LRV (Feed to Combined Permeate)	
		Integrity Surrogate	Conductivity
Test 1: Pre-CIP with unintentional cracked element	MS2	6	1.5
Test 2: Intentionally Compromised End Adapter with Unintentional Cracked Element	MS2	5.2	1.5
Test 3: Post-CIP	MS2	5.1	1.4
Test 4: NF Substituted for RO Element	MS2	5.3	1.2
Test 5: Normal Operation	MS2	5.1	1.3
Test 6: Normal Operation	Uranine	3.5	1.3
Test 7: Normal Operation	Sulfate	2.8	1.3

Based on the five MS2 bacteriophage integrity tests that were performed on the RO system at Wochholz, it was demonstrated that the RO unit attained at least 5.1-log of MS2 bacteriophage reduction.

When challenge tested with uranine, which has a molecular weight of 0.01% of the MS2 bacteriophage, the RO unit attained 3.5-log of uranine reduction. This is expected because the smaller the compound, the easier for it to permeate through small compromises (or, in the case of uranine, to diffuse across the membrane).

Ions are the smallest integrity surrogate indicators of an RO system and are typically grouped as conductivity which includes both monovalent (e.g., sodium and chloride) and divalent ions (e.g., sulfate and calcium). The RO system at Wochholz was tested for sulfate reduction and it was determined that the system attains 2.8-log of sulfate reduction.

In contrast, when considering conductivity as an integrity indicator, the RO system would only attain up to 1.5-log of conductivity reduction in these tests with the current regulations that are applied to facilities similar to Wochholz.

It is worth remembering that the role of an integrity indicator is to simulate the reduction of enteric viruses. This study demonstrated that viruses are removed to a much greater extent than a fluorescent marker (i.e., uranine) and sulfate. Similarly, it was also shown that viruses are removed much more than conductivity (i.e., MS2 test demonstrated an average of 5.3-log virus reduction compared to an average of 1.4-log of conductivity reduction).

When comparing sulfate to bulk conductivity, the RO unit attained 2.8-log of sulfate compared to 1.3-log of conductivity. This difference in sulfate LRVs to conductivity is due to the fact that reduction of monovalent ions in RO is lower than that of divalent ions. Therefore, conductivity is easily measured but it creates a disadvantage because it uses the smallest entities possible and uses them as proxy for enteric viruses which are much larger. Even in the case of sulfate or uranine, which the RO system attained 2.8-log and 3.5-log respectively, these values do not amount to the actual reduction of viruses. Fundamentally this gap is caused by comparing reduction of viruses with constituents that are of a different nature.

The uranine challenge test at Wochholz demonstrated that the RO unit rejected 99.98% of the uranine fed into the system (RO feed of 1.5 mg/L, attained 3.7-LRVs). This confirms that molecules larger than 376 g/mol (molecular weight of uranine) are rejected significantly by RO systems. Moreover, microbial pathogens that are orders of magnitude larger than uranine lack the ability to pass through the RO membrane and will be rejected at even higher rates.

Conductivity profiling appears to offer greater sensitivity when compared against the marker-based methods evaluated at the YVWD (described in other chapters) and elsewhere in literature. Although the technique relies on a relatively simple measurement, local defects that would pass a measurable amount of feed to permeate can be detected and subsequently identified as an outlier. The technique is applicable to typical full-scale systems that have a sufficient number of vessels in the stage to allow a meaningful statistical analysis.

The membranes evaluated for MS2 coliphage removal were commercially available products that were not challenge tested or certified for their ability to remove pathogens. Despite this, very high levels of virus removals were observed for arrays that were determined to be integral through statistical analysis of the conductivity profile of individual vessels.

The substitution of a NF for a RO membrane into an integral array did not result in a loss of virus removal. Thus, it appears that the composition of the underlying polysulfone membrane is an important consideration in the construction of the membrane element, as the membrane provides additional removal capability. Membrane cleaning and other periodic operational events do not appear to compromise the ability of the RO system to obtain MS2 LRVs exceeding 5-log.

A significant loss of integrity, either through a compromised element or defect in the sealing (e.g., interconnectors and/or end adapters) that results in passage of feed to permeate can be identified by conductivity profiling or other methods, such as probing to identify the location of the defect. Thus, sealing of the membranes is an important consideration in obtaining virus removal.

CHAPTER 7

Water Corporation of Western Australia

The Beenyup Groundwater Replenishment Scheme (BGRS) is Australia's only operating indirect potable reuse facility for groundwater replenishment. The results in this chapter were obtained from the Phase 1 BGRS facility which was commissioned in 2016. A second phase, doubling the facility's capacity, was commissioned in 2022. Prior to construction of phase 1, extensive testing was conducted between 2009 to 2012 at a purpose-built demonstration AWPF to evaluate treatment technologies and gather data on their performance and design requirements.

7.1 Testing Objectives

The testing objectives for BGRS included:

- Evaluate the removal of indigenous markers including sulfate, strontium, phosphate, and magnesium across a full-scale RO system over a significant time period (six months).
- Document bulk conductivity (feed to combined permeate) removal during the evaluation period.
- Evaluate the potential for spiked uranine challenge testing as a means to verify RO integrity on a pilot scale system.
- Compare the results with the performance demonstrated for other U.S. based systems measured as part of this project.

Testing at BGRS was completed between October 2020 to April 2021.

7.2 Site Description

The process flow diagram for the BGRS is shown in Figure 7-1 below. Nitrified/denitrified secondary effluent is sent from the Beenyup WWTP to the BGRS balance tank after coarse screening. After the balance tank, pre-formed chloramine is added inline to control biological growth and the influent then flows through fine screens and through the UF and RO systems. Following UF, the filtrate is sent to a filtrate tank. After the filtrate tank, sulfuric acid is used to adjust pH. Antiscalant can be added in the feed pump to the 2-Stage RO process. Design details for the full-scale RO are summarized in Table 7-1 below. The RO permeate is sent through a degasser and then to high dose UV disinfection. After UV, sodium hydroxide is added to control pH and the water is pumped from a product holding tank to the Leederville and Yarragadee aquifers. UF backwash residuals return to the wastewater treatment plant and RO concentrate is disposed via an ocean outfall.

The pilot RO unit located at the Beenyup AWPF is used for membrane performance testing on a feedwater representative of the BGRS. The pilot RO unit is fed from a side stream of the full-scale facility UF filtrate. The pilot feedwater is able to be fed different dosages of antiscalants, chloramine, and acid. Permeate and brine from the pilot RO skid are recombined and sent to the headworks. In this study, the pilot RO skid was used to evaluate uranine as a surrogate. The pilot skid was utilized due to regulatory concerns over addition of uranine to the main process line.

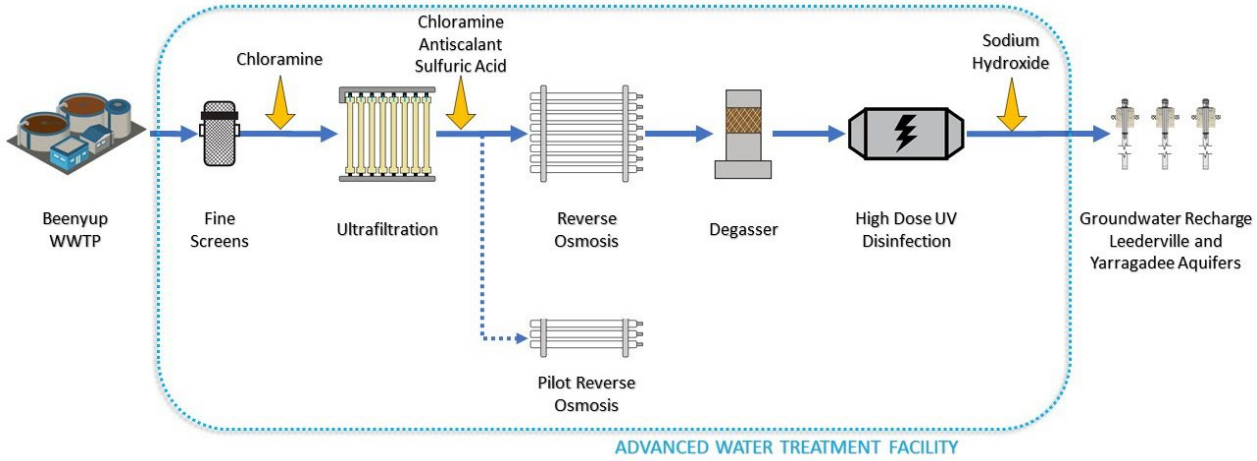

Figure 7-1. Beenyup Groundwater Replenishment Process Flow Diagram.
Testing was conducted on both the full-scale and pilot RO system.

Table 7-1. Full and Pilot Scale Design and Operational Parameters for the BGRS.

Parameter	Full Scale	Pilot
Membrane Modules	Hydranautics ESPA2-LD	Hydranautics ESPA2-LD
Membrane Age During Study (yr)	5 - 6	<1[2]
Array (each Train)	70:35	2:1
Modules per Vessel	7	7
Trains Onsite	4[1]	1
Average Flux (gfd)	11.3	11.3
Recovery (%)	75 - 80	75 - 80

Notes:
1. Only 2 of 4 Trains (Train 1 and Train 4) were the subject of this study.
2. Pilot membranes have been operated for a total of 6 months for intermittent project work. The membranes are approximately 3 years old and when not being used have been preserved in the pressure vessels.

7.2.1 Testing Protocol

Testing at Water Corporation was completed between October 2020 and June 2021. Naturally occurring sulfate, strontium, phosphate, and magnesium were analyzed weekly for six months from RO Trains 1 and 4 (RO1 and RO4). The specific sample locations included RO1 feed, RO1 stage 1 permeate, RO1 stage 2 permeate, RO4 feed, RO4 stage 1 permeate, RO4 stage 2 permeate, and RO combined permeate.

SCADA data for RO1 and RO4 including feed, permeate and concentrate flows from each stage. Pressures and conductivities were also provided at 30-minute intervals for the entire testing period. This data was first cleaned by filtering to ensure the RO unit was operational and not starting up or shutting down (i.e., feed flow > 170 L/s) and then the subset of data was expressed as a daily average of all operating data throughout the day.

Uranine testing was conducted as a one-time event on a separate 2-stage pilot system as testing required the injection of a prepared stock solution which was specifically approved for testing on the pilot system (by Australian regulators) for this project. Samples were collected every five minutes for a 45-minute period from feed stage 1, feed stage 2, permeate stage 1, permeate stage 2, combined permeate, and concentrate stage 2.

7.3 Water Corporation Results

7.3.1 Full Scale System Operation

The operational parameters for the sampled RO skids (RO1 and RO4) are included in table 7-2. Results are summarized as the minimum, median, and maximum daily average value (from sampling days) of the reported parameter. The results from the table show that over the course of the six-month testing period, both skids were operated with near identical set points and there was minimal operational variation on sample days. The exception was feedwater temperature, which changed seasonally.

Table 7-2. Operational Parameters of the RO Skids Studied at BGRS.
The results are daily averages on days when sampling for surrogates occurred.

	RO Skid					
	RO1 (n = 26)			RO4 (n = 26)		
Parameter	Min	Med	Max	Min	Med	Max
Feed Flow (gpm)	2954	2965	2972	2942	2959	2970
S1 Permeate Flow (gpm)	1524	1549	1567	1501	1539	1546
S2 Permeate Flow (gpm)	651	673	701	668	682	719
Temperature (°F)	74.5	83.3	85.0	74.5	83.3	85.0
Feed Conductivity (µS/cm)	1133	1197	1259	1138	1198	1254
S1 Permeate Conductivity (µS/cm)	36.5	47.3	54.1	37.2	46.8	52.4
S2 Permeate Conductivity (µS/cm)	72.4	100.4	123.7	69.3	101.1	116.3
Interstage Conductivity (µS/cm)	2212	2355	2472	2171	2324	2453
Concentrate Conductivity (µS/cm)	4092	4296	4539	4065	4257	4477
Notes: There were 26 days where markers were sampled on each skid.						

The normalized permeate flow (NPF) and normalized differential pressure (NDP) were calculated from the available operational data during the trial period. Calculations were performed using Hydranautics RODataXL which incorporates membrane specific constants to allow normalization of the performance data according to ASTM Standard D 4516-85 "Standard Practice for Standardizing Reverse Osmosis Performance Data". Analysis of the data shows when two CIPs were conducted (one for each test skid). RO1 was cleaned in mid-January 2021 and RO4 in mid-December 2020. There was a marginal increase in NDP across the first stage of both test skids suggesting slow development of an organic or biofouling layer, corresponding to a decrease in NPF for stage 1 and overall (Figure 7-2 and 7-3). The fouling was effectively managed by the CIPs and the overall variation in normalized data was less than 10%, demonstrating very stable system performance across the evaluation period.

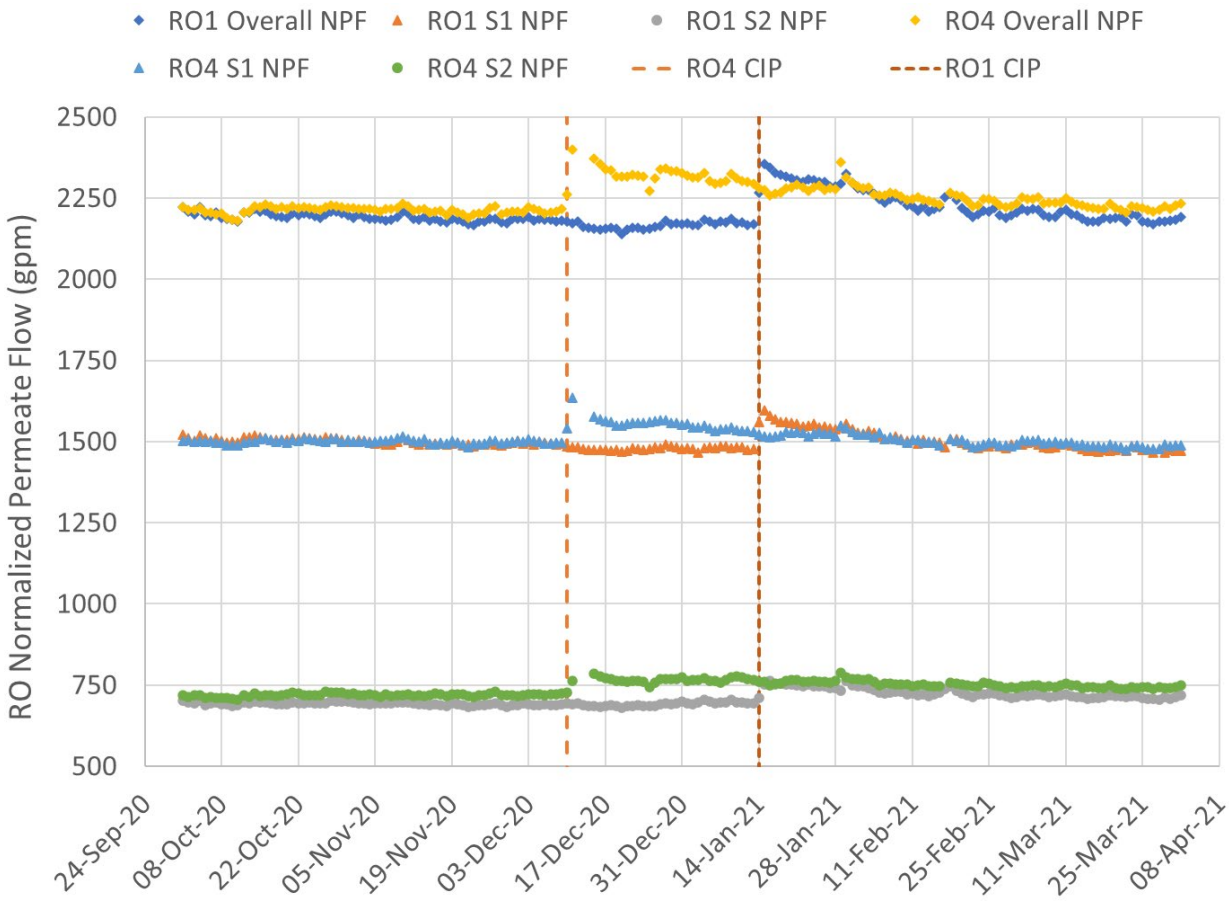

Figure 7-2. NPF for the RO Skids Evaluated During the Trial.
NPF increases following CIP indicating removal of foulants.
Overall performance was stable (< 10% variation across the trial).

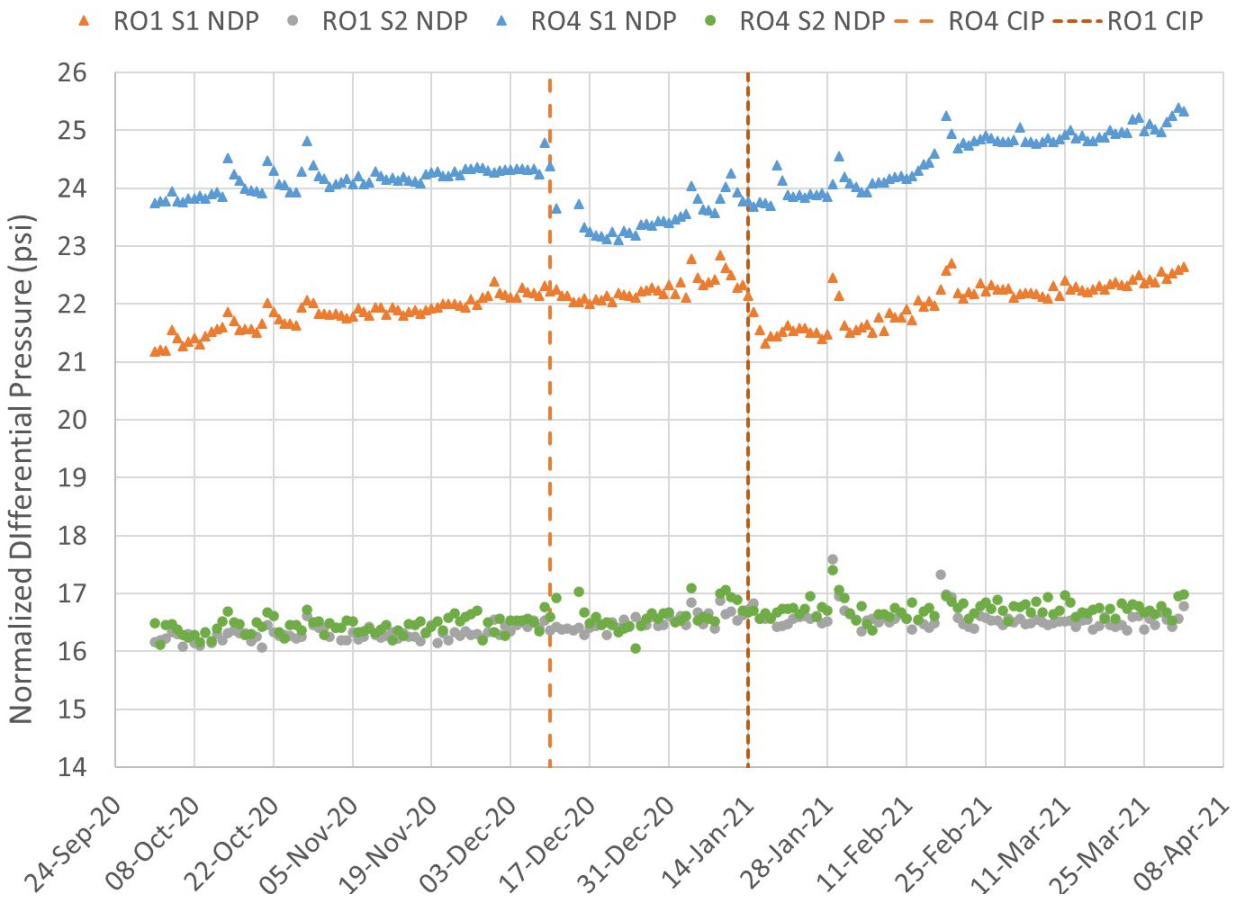

Figure 7-3. NDP for the RO Skids Evaluated During the Trial.
NDP Reduces for Stage 1 (S1) Following CIP Indicating Removal of Foulants**.**
NDP increases on the first stage a characteristic of biological or organic fouling common on RO in water reuse. Overall performance was stable (< 10% variation across the trial).

7.3.2 Full Scale Surrogate Results

The sampling results are summarized for all surrogates in Table 7-3. It is important to note that a majority of markers were not detected in the RO permeate. The proportion of values below the detection limit for each sample location is noted. Where markers were not detected, the detection limit was used to calculate LRV. Overall LRV for each skid was calculated using the flow weighted permeate quality (Table 7-4). A summary of the various native surrogate results is as follows:

- Sulfate detections were two to five times as common in stage 2 permeate, where the concentrate was approximately two times higher. Sulfate was detected at maximum concentration of 0.3 mg/L.
- Phosphate was the most frequently detected marker in the RO permeate with a detection frequency of between 20 to 60%. Similar to sulfate, detections were more frequent for stage 2, likely due to the higher concentration challenging the membrane. However, even when detected, phosphate concentrations in the permeate were very low, not exceeding 0.008 mg/L - close to the MRL of 0.005 mg/L.

- Strontium was not detected above the MRL of 0.002 mg/L in any permeate samples for either RO skid. The strontium feed water concentration was very low, between 0.13 to 0.17 mg/L.
- Magnesium was also not detected above its MRL of 0.1 mg/L in any permeate samples. Although the magnesium concentration in the RO feed was similar to phosphate, the detection limit is 20 times higher, limiting its usefulness as a surrogate marker to demonstrate removal.

Table 7-3. Surrogate Analytical Result Summary for the RO Skids Studied at BGRS.[1]

Surrogate (units)	Sample Location	RO Skid[4]							
		RO1 (n = 26)				RO4 (n = 26)			
		% Det	Min	Med	Max	% Det	Min	Med	Max
Sulfate (mg/L)	RO Feed[2]	100	83	101	125	100	85	100	127
	S1 Permeate	7.7	<0.1	<0.1	0.3	3.8	<0.1	<0.1	0.1
	S2 Permeate	15	<0.1	<0.1	0.1	15	<0.1	<0.1	0.2
	Combined Permeate[3]	19	<0.1	<0.1	0.1	19	<0.1	<0.1	0.1
Phosphate (mg/L)	RO Feed[2]	100	7.2	8.0	9.0	100	7.1	8.0	9.2
	S1 Permeate	23	<0.005	<0.005	0.008	23	<0.005	<0.005	0.008
	S2 Permeate	42	<0.005	<0.005	0.010	58	<0.005	0.006	0.009
	Combined Permeate[3]	54	<0.005	0.005	0.008	65	<0.005	0.005	0.008
Strontium (mg/L)	RO Feed[2]	100	0.13	0.15	0.17	100	0.13	0.15	0.17
	S1 Permeate	0.0	<0.002	<0.002	<0.002	0.0	<0.002	<0.002	<0.002
	S2 Permeate	0.0	<0.002	<0.002	<0.002	0.0	<0.002	<0.002	<0.002
	Combined Permeate[3]	0.0	<0.002	<0.002	<0.002	0.0	<0.002	<0.002	<0.002
Magnesium (mg/L)	RO Feed[2]	100	9.3	10.6	12.0	100	9.5	10.6	11.1
	S1 Permeate	0.0	<0.1	<0.1	<0.1	0.0	<0.1	<0.1	<0.1
	S2 Permeate	0.0	<0.1	<0.1	<0.1	0.0	<0.1	<0.1	<0.1
	Combined Permeate[3]	0.0	<0.1	<0.1	<0.1	0.0	<0.1	<0.1	<0.1

Notes:

1. All surrogates were detected in the RO feed but were rarely detected in RO permeate indicated by a low percent of detections (% Det).
2. RO feed for LRV calculations was the average of RO1 and RO4 feed samples taken on the same day
3. Combined permeate was the flow weighted average of Stage 1 and Stage 2. The detection limit was used for samples below the detection limit. If at least one stage was detected, then the combined permeate was listed as a detect.
4. Samples below the method reporting limit are represented as < the value of the MRL.

Table 7-4. Summary Statistics for the LRV Able to be Demonstrated Across each RO Skid and for Each Full-Scale Surrogate Tested at BGRS.[1]

Surrogate[2] (Stream Units)	Sample Location[3]	RO Skid LRV[4]					
		% Det	Min	5th Percentile	Med	95th Percentile	Max
Conductivity Profiling (µS/cm)[5]	RO1	31	3.4	-[5]	>5.0	-[5]	>5.0
	RO4	21	4.7	-[5]	>5.0	-[5]	>5.0
Direct Conductivity (µS/cm)[6]	RO1	100	1.2	1.2	1.3	1.4	1.4
	RO4	100	1.2	1.2	1.3	1.4	1.4
Sulfate (mg/L)	RO1	19	2.6	>2.9	>3.0	3.1	>3.1
	RO4	19	2.9	>2.9	>3.0	3.1	>3.1
Phosphate (mg/L)	RO1	54	3.0	3.1	>3.2	>3.2	>3.2
	RO4	65	3.0	3.1	3.2	>3.2	>3.2
Strontium (mg/L)	RO1	0.0	>1.8	>1.9	>1.9	>1.9	>1.9
	RO4	0.0	>1.8	>1.9	>1.9	>1.9	>1.9
Magnesium (mg/L)	RO1	0.0	>2.0	>2.0	>2.0	>2.0	>2.1
	RO4	0.0	>2.0	>2.0	>2.0	>2.0	>2.1

Notes:

1. Conductivity profiling, phosphate and sulfate were able to demonstrate the highest LRVs.
2. RO feed for LRV calculations was the average of RO1 and RO4 feed samples taken on the same day. N = 26 total samples per skid for markers unless otherwise specified.
3. RO feed for LRV calculations was the average of RO1 and RO4 feed samples taken on the same day except for conductivity LRV. Combined permeate was the flow weighted average of Stage 1 and Stage 2. The detection limit was used for samples below the detection limit. If at least one stage was detected, then the combined permeate was listed as a detect.
4. LRVs calculated with permeate results below the method reporting limit are represented as > LRV.
5. 13 conductivity profiles were available for RO1 and 14 were available for RO4. Due to the low sample numbers, calculation of a 5th and 95th percentile is not appropriate.
6. Direct conductivity LRV was calculated using the daily average flow weighted average permeate conductivity and feed for each skid. RO4 was offline for four days during the trial period. n = 183 for RO1 and n = 179 for RO4.

Summary statistics (i.e., min, max, median, and 5th and 95th percentiles) for LRV by direct conductivity across each train was identical. Direct conductivity also demonstrated the lowest LRVs of between 1.2 to 1.4. Strontium and magnesium underestimate LRVs as there was not sufficient sensitivity to detect either of these surrogates in the permeate relative to their feed water concentrations. Strontium LRVs were limited to demonstrating between 1.8 to 1.9-log. Magnesium was limited to demonstrating greater than 2.0-log.

Sulfate was detected sporadically at values close to the detection limit. When LRV results were sorted based on magnitude there were detects interspersed with non-detects. However, the minimum LRVs for sulfate when the compound was detected were 2.6 and 2.9 for RO1 and RO4, respectively. The maximum LRV demonstrated during this testing with sulfate was 3.1.

Phosphate was detected more frequently than sulfate (in approximately half of the permeate samples) and was able to demonstrate LRVs of 3.0 to > 3.2.

LRVs calculated from conductivity profiling were mostly capped at the maximum value of >5.0. For a majority of conductivity profiling results, the median was higher than the average and the difference between the two measurements was likely within the precision of the test. There were 4 events for RO1 and 3 events for RO4 where it was possible to calculate an LRV using conductivity profiling that was less than the maximum value. The high calculated LRV results (and very low standard deviations) from conductivity profiling are not surprising. Within the context of the surrogate sampling (i.e., a majority of non-detects), it would appear that although the RO system at BGRS has operated near continuously with the same membranes for 5 - 6 years, the membranes are still integral.

Two example conductivity profile plots of the WCWA data are shown in the figures below. The average, median, average + 3σ and the number of outliers more than 3 standard deviations higher than the stage mean are shown for reference. Note that for the lowest conductivity profile LRV of 3.4-log, there were 6 outlier vessels in stage 1 and one outlier vessel in stage 2. The average for stage 1 was also marginally greater than the mean (by 1 uS/cm) (Figure 7-4).

Overall Conductivity Profile LRV = 3.4

										Stage 1	
38	36	41	39	38	37	36	39	38	38	**Average**	**Median**
38	38	37	34	38	37	38	38	37	38	38	37
39	37	37	37	38	38	39	35	34	38	**Average + 3σ**	
36	38	37	34	36	35	35	37	36	35	44	
35	35	35	36	35	34	36	36	36	35	**n outliers**	
38	34	36	37	34	35	35	45	46	47	6	
43	45	45	43	43	45	41	40	39	39		

					Stage 2	
80	85	84	82	83	**Average**	**Median**
83	77	80	79	76	80	80
78	75	76	88	84	**Average + 3σ**	
81	85	82	77	80	88	
80	78	71	82	82	**n outliers**	
81	83	80	82	81	1	
78	80	79	72	68		

Figure 7-4. Example Conductivity Profile from WCWA where the LRV was 3.4.
Note there are a number of outlier vessels with performance beyond 3 standard deviations from the Stage average.

A more typical conductivity profile for the WCWA RO arrays is shown in Figure 7-5. For this scenario, the median is higher than the average, resulting in an LRV greater than 5.0-log. Also, there were no outliers in any vessels greater than 3 standard deviations from the mean, indicating very consistent array performance.

Overall Conductivity Profile LRV > 5.0

										Stage 1	
52	52	52	51	51	53	52	53	51	51	Average	Median
51	52	52	51	41	51	51	52	51	51	49	50
40	50	51	52	49	50	51	50	50	50	Average + 3σ	
48	50	51	48	50	50	48	48	51	49	54	
49	50	48	49	51	48	50	49	49	50	n outliers	
51	49	51	51	47	48	51	47	48	49	0	
48	49	50	47	48	47	46	46	44	44		

					Stage 2	
102	102	105	102	105	Average	Median
92	102	104	103	99	102	102
99	105	102	106	105	Average + 3σ	
104	104	104	100	102	108	
106	105	103	101	101	n outliers	
104	105	100	103	103	0	
100	101	101	99	96		

Figure 7-5. Example Conductivity Profile from WCWA where the LRV was >5.0.
Note there are zero outlier vessels, and the median is higher than or equal to the mean.

The direct conductivity LRVs were marginally lower (by 0.1 to 0.3-log) compared to facilities in the US. However, in general the flux was marginally lower at BGRS (ranging from 10 - 11 gfd on average) than the US installations analyzed in this study (average fluxes of 11 - 12 gfd). The ASTM normalized salt passage results (normalized for temperature, TDS, and flow) were converted to an LRV. Upon doing so, the overall system LRV increased to 1.5 to 1.7-log for both trains.

In an effort to try and explain the detection of surrogates and LRVs from conductivity profiling that were less than 5-log, the LRV results were plotted as a time series relative to the CIP events and are shown in the figures below.

Both phosphate and sulfate were detected in the permeate for three samples (two weeks) following a CIP on RO 1. In addition, 3 of the 4 conductivity profile LRVs that were not greater than 5.0-log coincided with detections of phosphate in the permeate. However, conductivity profiling did not appear to change in response to the CIP on RO 1. A marginal decrease in the LRV converted from normalized salt passage as well as the direct conductivity LRV was observed following the RO1 CIP.

RO4 results were similar to RO1, however, there was less certain response of surrogates to the CIP. In particular, in 2 of the 4 samples following a CIP, sulfate was detected. Phosphate was detected in 5 samples following a CIP but was also detected in 4 samples prior to the RO4 CIP and it was not clear if the CIP had influenced phosphate removal. There were only 3 conductivity profile LRVs that were not greater than 5-log for RO4 and these did not occur around the time of the CIP.

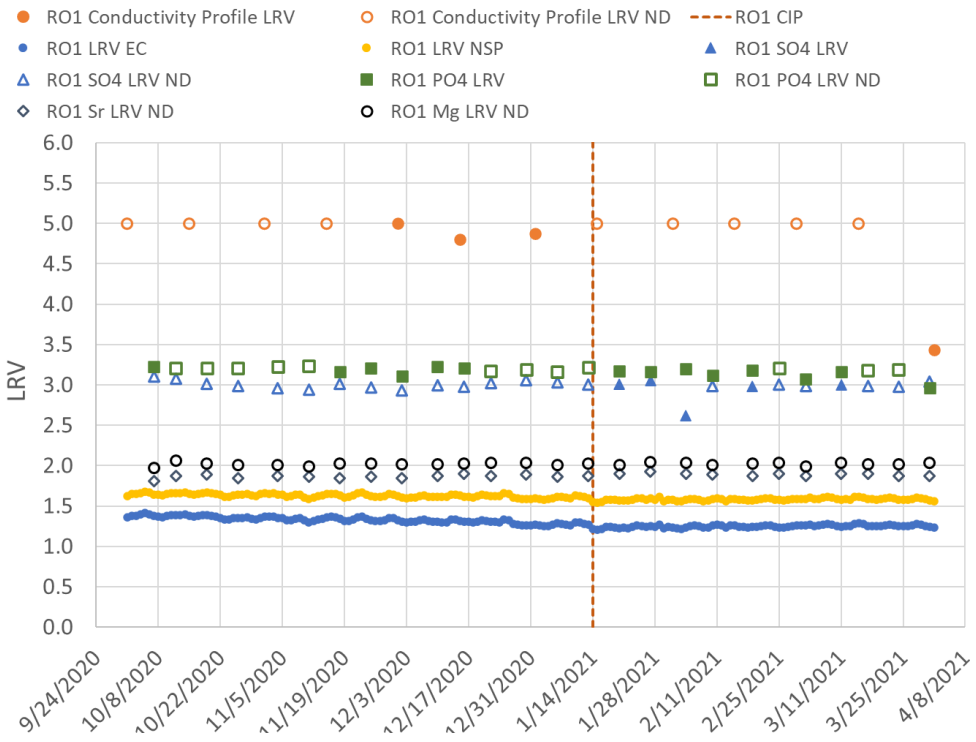

Figure 7-6. RO1 Surrogate LRV Results During the Test Period.
Open symbols indicate that the true value of the LRV is higher than shown.
Note the detections of sulfate and phosphate occur for the 3 samples following the CIP.

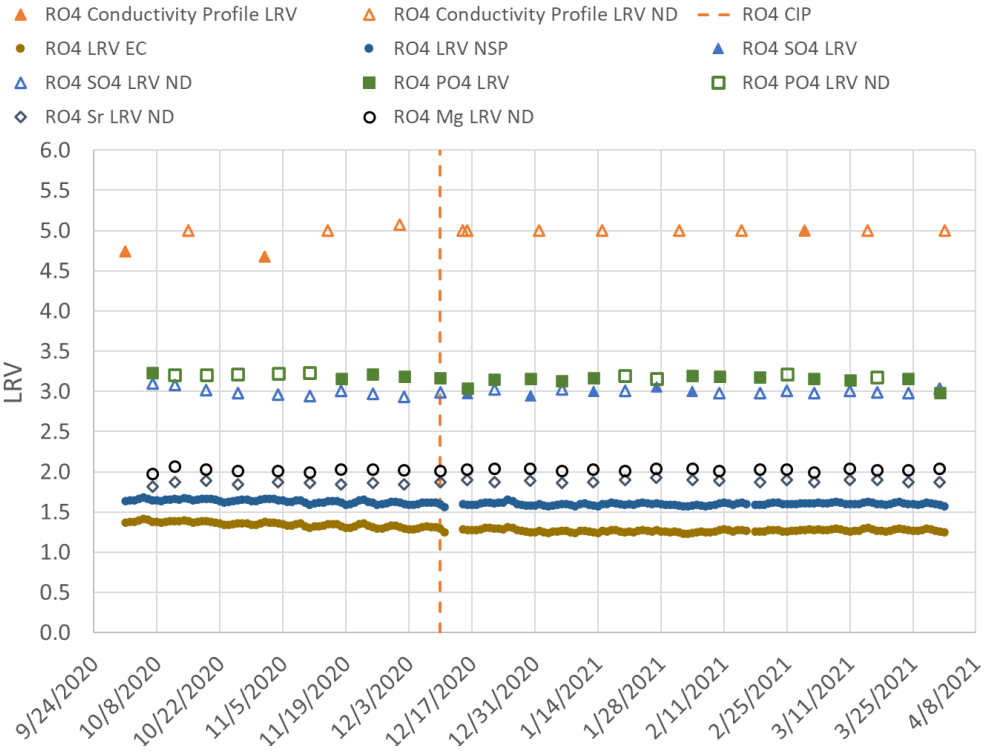

Figure 7-7. RO4 Surrogate LRV Results During the Test Period.
Open symbols indicate that the true value of the LRV is higher than shown.
Note the detections of sulfate for 2 of the 3 samples following the CIP. A trend with phosphate LRV is not clear.

7.3.3 Uranine Pilot Challenge Test Results

Uranine was evaluated on a pilot 2-stage RO unit that operates on a side stream of the full-scale system. The pilot was operated with the same model of membranes as the full-scale and at an average flux of 10.7 gfd and 75% recovery, representative of the full-scale system.

The uranine challenge test was conducted as a single event. Uranine was spiked to a target concentration of 3 mg/L and the effective MRL was determined to be 0.01 ug/L. All permeate samples were above the limit of reporting. The uranine concentration determined in the feed to both stage 1 and 2 as well as the concentrate was very stable for the duration of the test. However, all permeate streams appeared to require approximately 15 minutes to achieve stable values. The gradual increase and stabilization of permeate uranine concentrations is shown in the figure below.

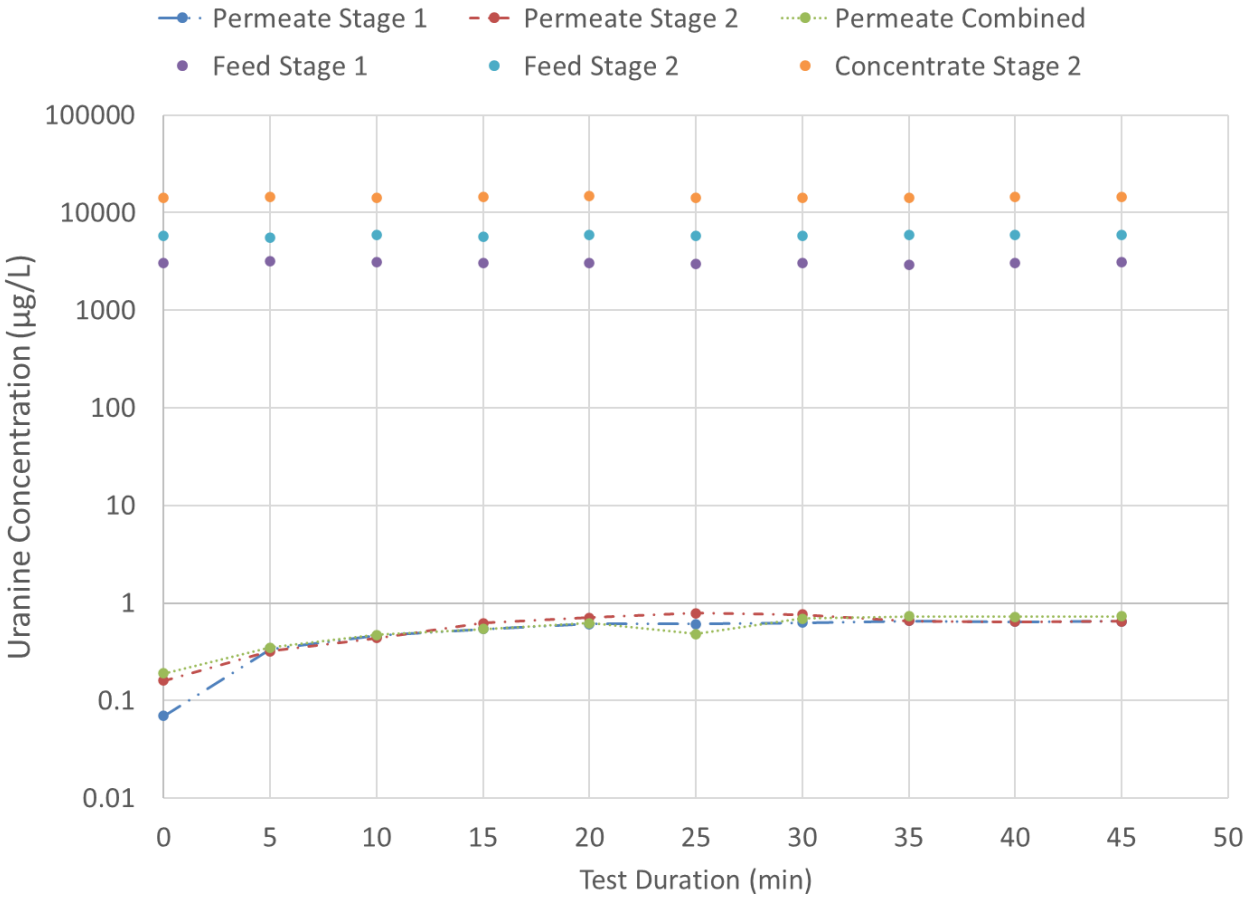

Figure 7-8. Uranine Concentrations During Challenge Testing of the WCWA Pilot RO.
Permeate values appeared to take at least 15 minutes to reach stable values.

It is unclear why uranine permeate concentrations required an equilibration time. At the experimental flows, the residence time based on dead volume of the system is estimated to be in the order of seconds. Also, the stability of the feed and concentrate results, where dead volume would have been the largest, suggest that the pilot was indeed equilibrated.

It was presumed that the initial lower removal may be due to adsorption of uranine to the membrane and system components. Once sites were saturated, the concentration at the membrane surface stabilized. Another potential reason for this change in LRV could be variations in sample collection and fluorometer reading (sensitivity to pH). Through this project, the testing teams have unanimously reported challenges with handling, dosing, and analyzing uranine. While it is a method that yields promising LRV results, consideration for the practical application of using this method on a routine basis at full scale installations must be considered.

A plot of the resulting LRV for each stage during the test run is shown in Figure 7-9. LRV decreased from approximately 4.2-log to 3.7-log after 15 minutes, after which the removal reached steady-state. Given the unsteady state observations for the first 15 minutes in the permeate, these readings were not included to determine an average and standard deviation for uranine LRV. The minimum and maximum overall uranine LRV (for samples after 15 minutes) was 3.6 - 3.8, the average LRV was 3.7 and the standard deviation was 0.07-log.

These uranine LRVs are very close to the 3.5-log removal reported for OCWD and YVWD and may be marginally higher due to the 3 mg/L dosed in this work compared to the 2 mg/L target dosed at OCWD and YVWD.

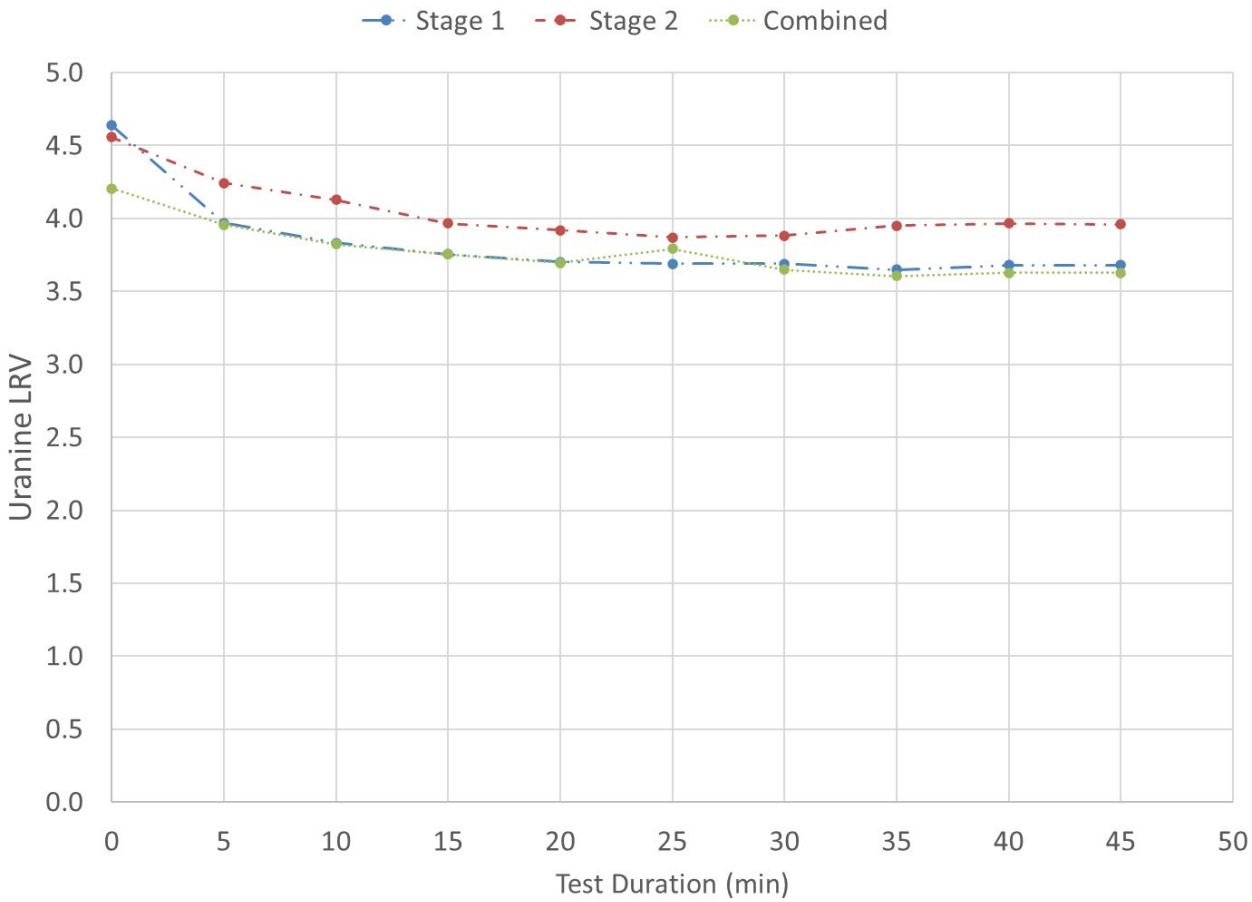

Figure 7-9. Uranine LRVs During Challenge Testing of the WCWA Pilot RO.
Permeate values appeared to take at least 15 minutes to reach stable values and are the cause of a 15-minute decline in observed LRV.

7.4 Water Corporation Testing Summary

An extensive sampling campaign for 2 full-scale RO skids was conducted over a 6-month period. From the surrogates analyzed, strontium and magnesium were not present at sufficient concentration in the RO feed and did not have low enough reporting limits to demonstrate improved LRVs. Sulfate and phosphate were able to demonstrate LRVs of 2.6 - >3.1 and 3.0 to >3.2, respectively. Phosphate was detected in almost half the permeate samples, whereas sulfate was only detected in approximately 30% of the samples.

The LRV calculated from conductivity profiling suggested that the RO membranes, which have been in operation 5-6 years have maintained very good integrity as most of the results were capped at 5-log due to the very small standard deviations and difference between average and median permeate conductivities of the arrays. However, there were events where the conductivity profiling LRV reduced to 3.4.

Surrogate result reproducibility was high at this site and when compared to other sites where the same markers were measured. In addition, the sensitivity demonstrated for sulfate was very similar to results from OCWD and YVWD. Strontium results did not compare well to other sites due to its low abundance in the RO feed water.

Phosphate and sulfate results appeared to show some responsiveness to CIP, with more frequent detections in the 2-week period following. Phosphate detections for RO1 appeared to coincide with 3 of the 4 conductivity profile results that were not greater than 5-log. However, the conductivity profiling at BGRS did not appear to change as a result of CIP, as observed at OCWD.

Based on the reporting limits and RO feed concentrations available at BGRS, phosphate may warrant further investigation at other facilities as it was the most frequently detected marker and demonstrated the highest and most reproducible surrogate LRV (with the exception of conductivity profiling).

Uranine was evaluated at pilot scale and was able to demonstrate steady state LRVs between 3.6 to 3.8-log units, close to the 3.5-log reported at OCWD and YVWD. However, permeate results appeared to take 15 minutes to stabilize. The stabilization may have been due to adsorption or pH correction issues with analysis. Although the uranine LRV is reproducible across sites and scale, this compound appears to have a number of practical challenges involved with spiking, stability, and analysis that may limit its widespread use.

CHAPTER 8

Southern Nevada Water Authority

This chapter details the experimental activities and results obtained from SNWA.

8.1 Testing Approach and Objectives

The goal of the testing at the SNWA was to evaluate the maximum sensitivity of marker-based integrity testing methods for RO and NF membranes. Three 4-inch RO membranes (Hydraunatics ESPA2-LD-4040, Dupont Filmtec BW30XFRLE 4040, Toray TMG10D) and one 4-inch NF membrane (CSM NE4040-40) were tested in both new condition and after chlorine oxidation. The membranes evaluated were selected for investigation as they are commonly used in potable reuse applications. The testing conducted at SNWA with a pilot scale system offered the flexibility to examine a variety of operating conditions as well as the ability to oxidize the membranes.

Prior to evaluation, each membrane element was characterized for permeability and salt rejection by testing under standard conditions similar to membrane certification testing conditions stated in the membrane data sheets. Then, a novel approach using a single-element pilot system was employed to characterize marker rejection under operational conditions relevant to a full-scale RO array. The single-element pilot tests were operated to represent full-scale potable reuse high-pressure membrane systems operated at 85% recovery. Within the pilot apparatus, hydraulic conditions were adjusted to provide a representative operational environment expected to occur for both lead and tail elements within a full-scale array. Sulfate, conductivity, uranine, sucralose, and MS2 rejection were quantified to enable comparison of marker sensitivity between membranes, relative level of integrity failure, and full-scale results presented in other chapters.

8.2 Experimental Methods

8.2.1 RO Pilot Apparatus

All experiments in this chapter were conducted with a single element RO pilot skid shown in Figure 8-1. The single-element pilot skid has a design flow range of 5-10 gpm using a 4-inch diameter membrane element. Pilot-testing was conducted in batch-recirculation mode in which all streams (feed, concentrate, permeate) continuously returned to the feed tank. Temperature was kept constant at a standard reference temperature of 25°C. The challenge testing steps are described in detail in Section 8.2.6. Briefly, a synthetic feedwater was made, and markers were spiked into a 150 L feed tank. Feed and recycle flows were adjusted to match either lead or tail element conditions. Markers were then sampled after an equilibration and mixing period of 30 minutes. A process flow diagram detailing sampling locations (feed, permeate, and concentrate) is shown in Figure 8-2.

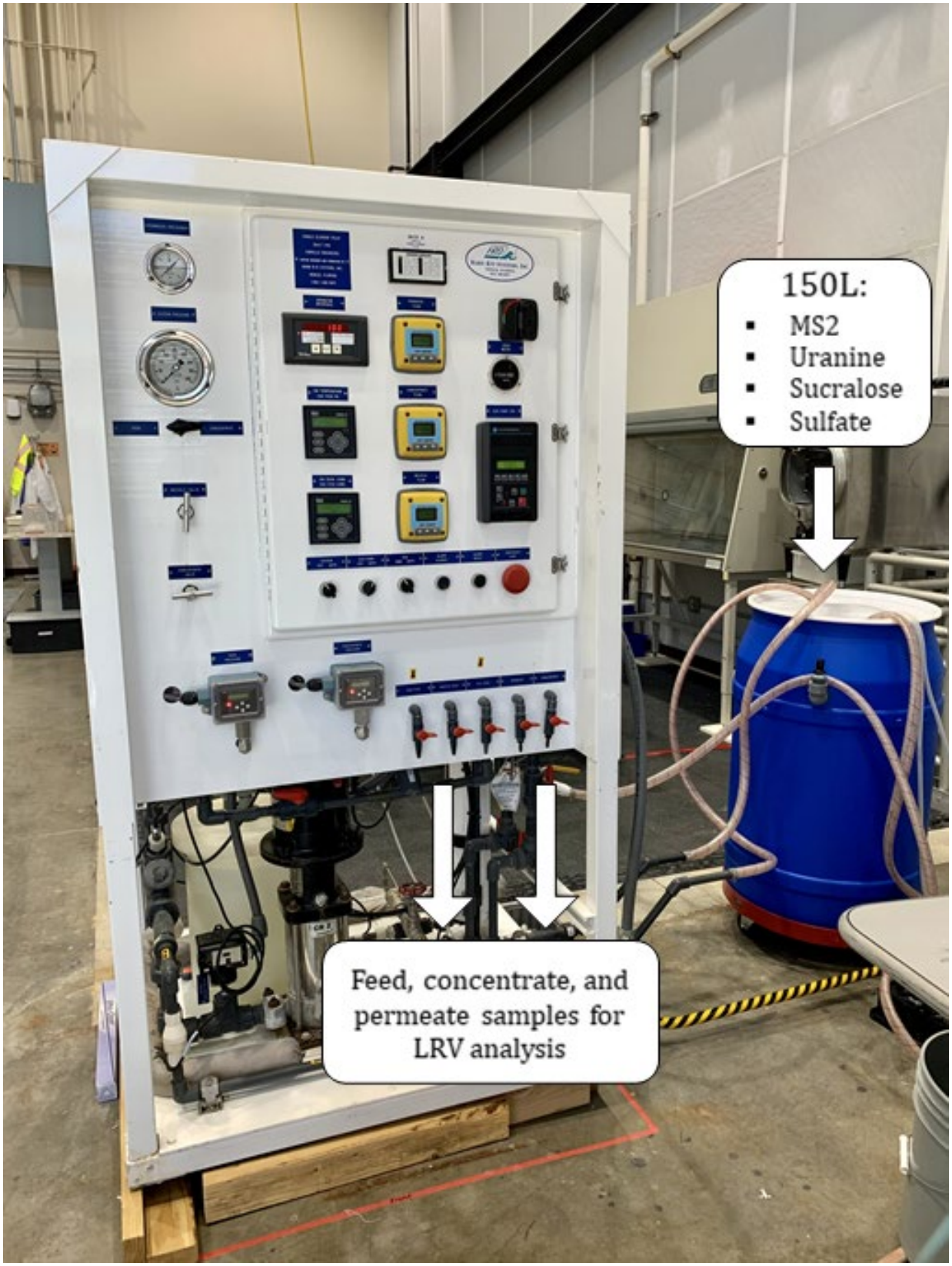

Figure 8-1. The Single-Element Pilot Skid and 150L Synthetic Feed Reservoir Used During Challenge Testing Experiments at SNWA.

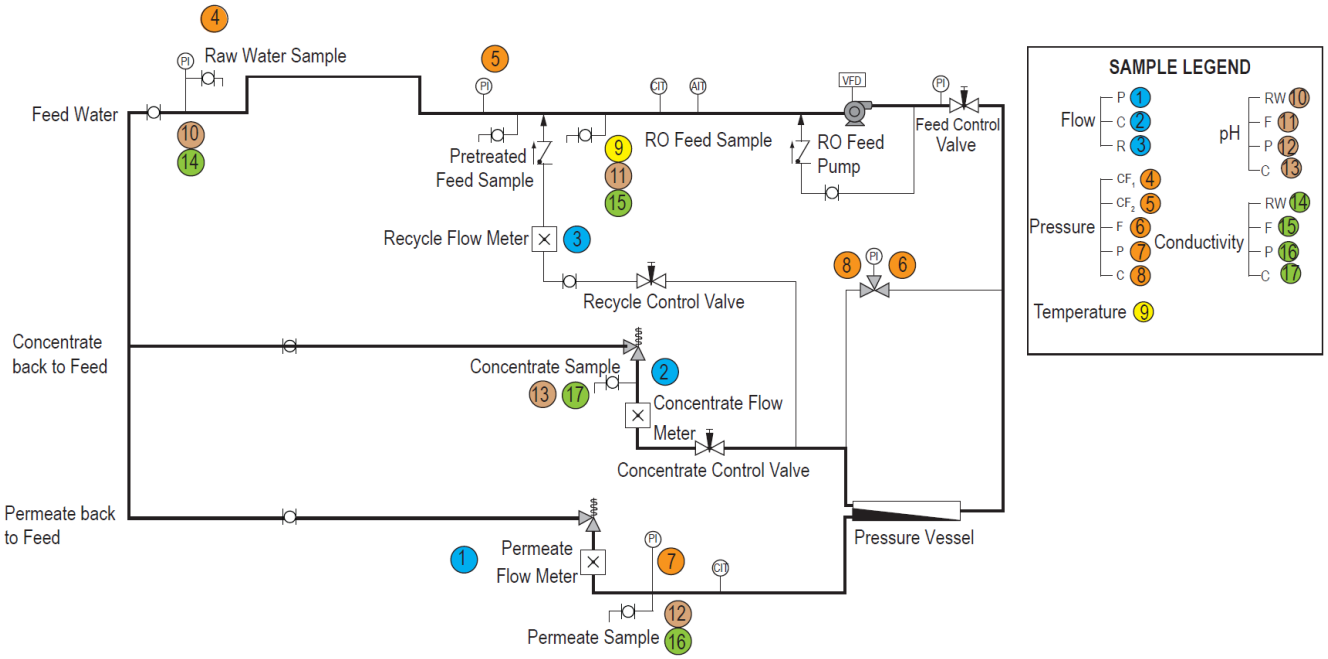

Figure 8-2. Process Flow Diagram of the Single-Element Pilot Skid Used at SNWA.

8.2.2 RO Pilot Operational Setpoints

During operation, full-scale RO systems produce a permeate, leaving higher concentrations of constituents in the brine. In addition, as permeate is produced, the flowrate of the brine decreases longitudinally along a pressure vessel. The resulting velocity reduction, combined with the increasing concentration of rejected salts means that an RO element in the lead position is exposed to different salt concentrations and cross flow velocities than an RO element in the tail position. The increase in salt concentration and a lower crossflow velocity in the tail means there is a higher transfer rate of dissolved constituents across the RO membrane due to diffusion. As a result, a lesser quality of permeate is expected to be observed from tail elements compared to lead elements. Full-scale RO systems are comprised of multiple stages, with several individual pressure vessels per stage, and a series of elements in each vessel. A full-scale system design must consider the saturation of dissolved constituents and the cross flow to ensure permeate quality goals are met and to reduce the chance of membrane scaling. In this chapter, RO projection software was used for each membrane type tested to establish operational conditions for the single element pilot so that the rejection performance of both lead and tail elements in a full-scale array could be accurately simulated.

Flux, recovery, feed pressure, and flow (feed, permeate, and concentrate) were adjusted to simulate the performance of the lead and tail elements for a system operating at 85% recovery. The four membranes tested for this project were:

- Filmtec BW30XFRLE 4040 RO membrane.
- Hydranautics ESPA2-LD-4040 RO membrane.
- Toray TMG10D RO membrane.
- CSM NE-4040 NF membrane.

The RO membranes tested were selected based on use in existing facilities to provide a basis for analysis of widely used products. The membranes provided were commercially available products not specifically manufactured or certified for their virus removal capabilities.

Proprietary RO design software from each vendor was used to determine the operating conditions for the selected membranes. The basis of design for each membrane system considered conditions to represent a full-scale system comprising a 2 stage, 20 by 10 array with each pressure vessel containing seven elements, a feed flow of 197.5 gpm, and 85% recovery. An illustration showing an example 20:10 array and the locations of the membrane elements that were simulated in this study is shown in the Figure below.

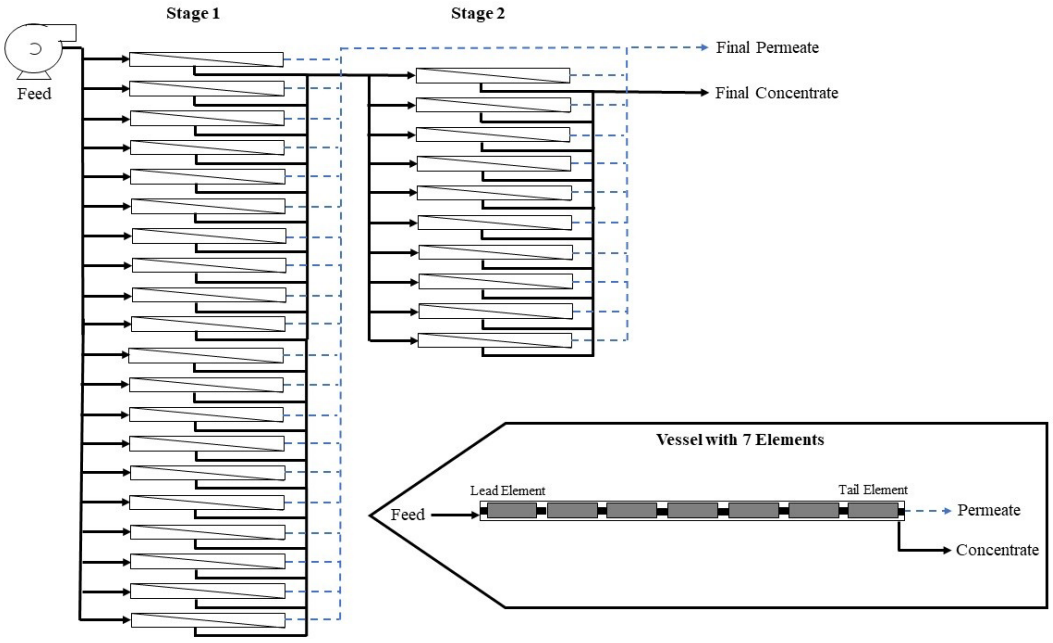

Figure 8-3. Illustration of an Example 20:10 Array Showing the Positions of the Membrane Elements That Were Simulated in this Study.

The first element in the first stage was used for the lead element operating conditions. The last element in the second stage was used for the tail element design. The recycle flow for the tail element was determined by using the software to simulate a single stage, one element system that operated at 85% recovery. The recycle flow was changed until the flow conditions with the recycle matched the full-scale tail element conditions. Filmtec WAVE software was used to determine the operating conditions for the BW30XFRLE 4040 RO membrane, Toray software was used to determine the operating conditions for the TMG10D RO membrane, IMS Design software was used to determine the operating conditions for the ESPA2-LD-4040 RO membrane, and CSMPRO software was used to determine the operating conditions for the CSM NE-4040 NF membrane. Since the BW30XFRLE 4040 element is not included in the Filmtec WAVE software, the LC LE-4040 element was used to determine flow conditions with special adjustments made by a Filmtec software specialist. Similarly, the flow conditions for the CSM NE4040-40 element were determined using the NE4040-70 element because the NE4040-40 was also not contained in the CSMPRO software. The operating conditions for each membrane are summarized in Table 8-1.

Table 8-1. Operational Conditions for the Lead and Tail Element Testing for each Membrane Based on their Respective Design Software.

Operating Parameter	Operating Conditions for the Lead Element Test	Operating Conditions for the Tail Element Test
Filmtec BW30XFRLE-4040 - Filmtec WAVE Software		
Feed Flow (FF), gpm	9.9	3.5
Recycle Flow (RF), gpm	0	2.98
Permeate Flow (PF), gpm	1.0	0.42
Concentrate Flow (CF), gpm	8.7	3.06
Recovery (PF/FF)	12.1%	12.1%
Flux, gfd	18.40	7.80
Max Feed Pressure, psi	97.6	225
Hydranautics ESPA2-LD-4040 - IMS Design Software		
FF, gpm	9.59	3.76
RF, gpm	0	3.12
PF, gpm	1.0	0.41
CF, gpm	8.59	0.23
Recovery (PF/FF)	10.4%	10.9%
Flux, gfd	18.0	7.38
Max Feed Pressure, psi	109	216
Toray TMG10D - Toray Software		
FF, gpm	9.40	3.53
RF, gpm	0	3.0
PF, gpm	1.12	0.27
CF, gpm	8.28	3.26
Recovery (PF/FF)	11.9%	8.2%
Flux, gfd	18.54	3.97
Max Feed Pressure, psi	102	199
CSM NE4040-40 - CSM PRO Software		
FF, gpm	7.12	3.67
RF, gpm	0	2.99
PF, gpm	0.92	0.47
CF, gpm	6.20	0.21
Recovery (PF/FF)	14.8%	12.8%
Flux, gfd	15.59	11.35
Max Feed Pressure, psi	59	228

8.2.3 Direct Integrity Test Methods

Considering the findings of the literature review and regulatory stakeholder input, the following molecular-based markers were evaluated at SNWA before and after membrane oxidation: uranine, sulfate, sucralose, and MS2. Water quality was monitored with online and handheld sensors (i.e., pH, conductivity, and temperature) and grab samples were collected for uranine, sulfate, sucralose, and MS2 during each lead/tail element test from the feed, permeate, and concentrate lines to enable the calculation of constituent rejection and assess mass balance (i.e., loss to sorption to the membrane). Prior to any sample collection, the pilot was operated until steady-state conditions were achieved (i.e., stable flux, pressure, temperature, and conductivity rejection). Establishment of steady state conditions typically occurred after 30 minutes of run time. Tests (integrity/permeability, lead, and tail) were conducted with 25% replication for membranes with one sample for sucralose, duplicate samples for uranine and sulfate, and triplicate samples for MS2 to ensure precise results. To assess precision, replicate testing and analysis was conducted for each membrane according to the schedule outlined in Table 8-2.

Table 8-2. Experiment, Sample, and Analysis Replicates for each Marker and Experimental Condition (Intact and Oxidized Membrane) to Provide the Total Data Points Produced for each Marker (n Total).

Marker	Replicate Type[1,3]	Intact Membrane (Lead and Tail)	Oxidized Membrane (Lead and Tail)
Sucralose	Experimental Run	12	11
	Sample Replicates	1	1
	Analysis Replicates	1	1
	n Total	12	11
Sulfate	Experimental Run	13	11
	Sample Replicates	2	2
	Analysis Replicates	1	1
	n Total	26	22
MS2	Experimental Run	12	10
	Sample Replicates	3	3
	Analysis Replicates[2]	1, 2	1, 2
	n Total[2]	54	45
Uranine	Experimental Run	13	11
	Sample Replicates	2	2
	Analysis Replicates	1	1
	n Total	26	22

Notes:

1. Experimental runs = how many times each scenario was tested for each membrane product. Sampling replicate = how many samples were taken during each experimental run. Analysis replicate = how many times each sample was analyzed e.g., 1 experimental run with MS2, sampled in triplicate, plated on 3 plates = 9 total data points.
2. Halfway through the project, the number of analysis replicates for MS2 was increased from 1 to 2.
3. Not included in this table were the SNWA lab analysis replicates. The SNWA lab conducted 33% duplication in that for each experiment, either the feed, concentrate, or permeate sample was replicated for uranine and sucralose. Thus, for each experiment, there was one sample type (i.e., feed, concentrate, permeate) that had 2 analysis replicates for sucralose and 3 analysis replicates for uranine.

The single-element pilot tests (not including vacuum tests) for each membrane occurred in the following order (eight tests per membrane, not including randomized duplicates, four tests with water quality sampling):

- New membrane:
  - Vacuum decay test.
  - Integrity/permeability tests.
  - Lead element test (DIT sampling).
  - Integrity/permeability tests.
  - Tail element test (DIT sampling).
- Oxidized membrane:
  - Vacuum decay test.
  - Integrity/permeability tests.
  - Lead element test (DIT sampling).
  - Integrity/permeability tests.
  - Tail element test (DIT sampling).
  - Vacuum decay test.
- Duplicate test (randomized condition which occurred directly after the first test for that particular condition):
  - Integrity/permeability tests.
  - Duplicate test (DIT sampling).

8.2.4 Feed Water

Synthetic water was used as the feed for pilot testing to better control test conditions and minimize performance effects due to organic fouling, biofouling, and inorganic scaling. Deionized water was spiked with the selected markers at concentrations that demonstrate up to 4-log removal for sucralose, sulfate, and uranine and 6-log removal for MS2 (Table 8-3). The synthetic feed water consisted of only sucralose, MS2, uranine, and sodium sulfate. The average pH of the synthetic feed water for the lead tests was pH = 6.33 (n=14) and pH = 6.79 (n=10) for the tail tests.

Table 8-3. Target Concentrations of Spiked Constituents in the Feed Waters for Lead and Tail Element Tests with their MRL for RO Permeate.

Marker	MRL for RO Permeate	Spiked Feed Water Concentrations in the Lead Element Test	Spiked Feed Water Concentrations in the Tail Element Test
Sucralose (ng/L)	5[1]	50,000	300,000
Sulfate (mg/L)	0.25[2]	2,500	15,000
MS2 (pfu/mL)	0.1[3]	4×10^6	2.4×10^7
Uranine (mg/L)	0.1[1]	2	12

Notes:
1. MRL set by SNWA R&D Lab.
2. MRL set by Eurofins Eaton Analytical LLC.
3. MRL set by GAP EnviroMicrobial Services Ltd analysis and dependent on sample volume.

8.2.5 Experimental Procedure

This section describes the testing procedure for each element, including preliminary characterization of integrity and permeability at standard test conditions and quantification of marker removal under representative full-scale conditions.

8.2.5.1 Standard Module Integrity and Permeability Characterization

Prior to conducting the experimental testing, integrity tests were performed with each membrane according to membrane specifications (i.e., 2,000 mg/L sodium chloride (NaCl) feed, 150 psi for RO, 75 psi for NF, 15% recovery, etc.). Concurrently, permeability tests were conducted by incrementally increasing the pressure and recording the permeate flux, temperatures, and pressures. These brief tests ensured membrane integrity and established baseline flux, rejection, and permeability for comparison of membranes and test conditions. The standardized integrity and permeability characterization involved the following steps:

- Installed new membrane into the pilot apparatus and flushed preservatives out with 150 L of DI water and confirmed by a permeate conductivity < 5 µS/cm.
- Prepared the standard salt solution detailed in the membrane data sheet (2,000 mg/L NaCl for the TMG10D, CSM NE4040-40, and BW30XFRLE 4040 and 1,500 mg/L NaCl for the ESPA2 LD-4040) with DI into 55-gallon tank.
- Increased the pressure/flux stepwise 4 times at intervals of ~15 psi and allowed to reach steady state before recording flux, pressure, and temperature:
  - Calculated the permeability.
- Circulated the solution at the specified operating conditions in the membrane data sheet and recorded flux, temperature, conductivity, etc.
- Verified whether the salt rejection was within the acceptable range for a new membrane.
- Ended operation and flushed the skid with deionized water.
- In addition to characterizing salt rejection and permeability under standard conditions, a vacuum decay test was conducted on each new membrane to directly measure integrity and ensure there was no physical damage. The vacuum test was conducted according to the following procedure.
  - Rinsed the element by circulating DI water through the system.
  - Drained the membrane element, and then sealed the internal permeate tube.
  - Connected the permeate tube to a vacuum pump and created a vacuum inside the tube until it reaches steady state (~22.5 in. Hg).
  - By using a valve, the permeate tube was isolated from the vacuum pump and a vacuum gauge was used to monitor the vacuum decay over time.
  - Recorded the initial vacuum pressure. Took readings every 60 seconds over a 5-minute period. The acceptable decay rate is 6.0 in. Hg/min as stated in the ASTM D3923-94.

A schematic of the vacuum decay test apparatus can be seen in Figure 8-4 and the setup used at SNWA can be seen in Figure 8-5.

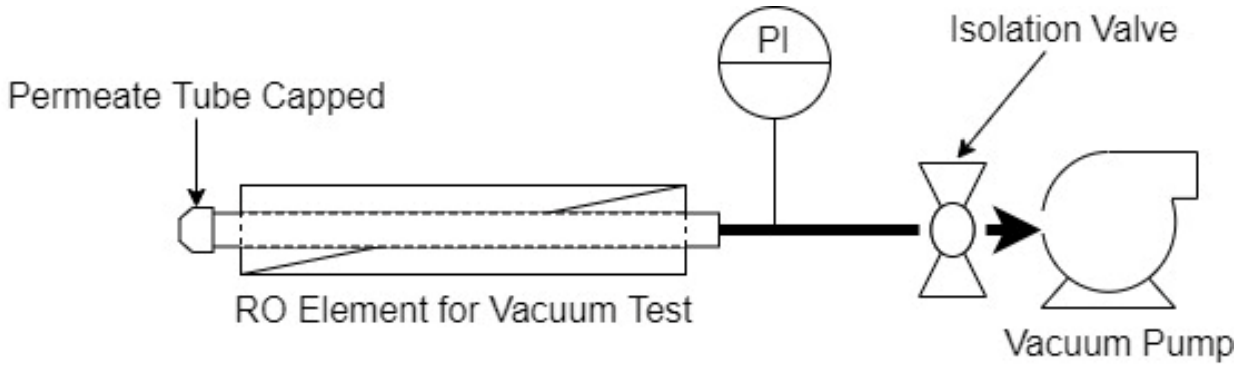

Figure 8-4. Schematic of a Vacuum Decay Test for an RO or NF Membrane Element.

Figure 8-5. Picture of the Vacuum Hold Test Performed at SNWA.

8.2.6 Marker Challenge Testing

8.2.6.1 Lead Element Testing

The following steps were followed during challenge testing:

- Filled the feed tank with 150 L of DI water.
- Added the sulfate and sucralose at the specified concentrations (Table 8-3).
- Ran the system at the specified operating conditions for the lead element tests (Table 8-1).
- When operating conditions were stable (after approximately 30 min), collected a feed, concentrate, and permeate sample to serve as the blank samples for the uranine analysis.
- Covered all clear tubing in tin foil to reduce light exposure since uranine is photosensitive.
- Added the uranine and MS2 at the specified concentrations (Table 8-3).
- Began the test by circulating the synthetic feed water at the specified operating conditions for lead element tests (Table 8-1) until conductivity, pressure, temperature, and flux were stable.
- When all parameters were steady, collected samples and noted test parameters: temperature, flux, feed/permeate/concentrate conductivity, feed pressure, and backpressure.
- Flushed the system with 300 L of DI.
- Shut off the skid.

8.2.6.2 Tail Element Testing

- Repeated integrity and permeability tests with NaCl solution.
- Repeated steps for lead element test except with a new feed batch at the specified constituent concentrations (Table 8-3) and operating conditions (Table 8-1) for tail element tests.
- When the tests were completed, high pH cleaning was conducted according to the following steps:
  - Filled the tank with DI water and adjusted the pH to 11 with sodium hydroxide.
  - Circulated the solution for one hour and then let the solution sit for ~4 hours
  - Flushed the skid with 300 L of DI water.

8.2.6.3 Oxidized Element Testing

RO membranes are typically thin film composites with an outer very thin layer of polyamide over a polysulfone support. The polyamide layer is responsible for salt rejection. Free chlorine is known to react with and oxidize the polyamide layer, resulting in reduced salt rejection. During oxidized element testing, the membrane was exposed to high concentrations of sodium hypochlorite to damage and remove the polyamide layer (Antony et al. 2016; Lawler et al. 2013). The element oxidation procedure and testing followed the steps below:

- Oxidized element test for a RO membrane:
  - Flushed the skid with DI water.
  - Conducted a vacuum test to confirm no membrane damage was present.
  - Circulated DI water with 500 ppm NaCl and sodium hypochlorite (1000 mg/L chlorine [Cl_2]) until conductivity rejection decreased to 80-85%.

- Disposed of chlorinated water and quenched any potentially remaining chlorine within the skid with sodium thiosulfate. Tested the concentrate and permeate for chlorine residual using the DPD method (HACH Method 8021).
- Conducted a vacuum test to ensure that the mechanical integrity was still intact after oxidation.
- Repeated integrity, permeability, lead, and tail element tests as described above with the oxidized membrane.
- Conducted a vacuum test at the end of the oxidized element testing for mechanical integrity assurance.

- Oxidized element test for the NF membrane:
  - Flushed the skid with DI water.
  - Conducted a vacuum test to confirm no membrane damage was present.
  - Circulated DI water and bleach (1000 mg/L Cl_2) for the same contact time as the oxidized element test for the RO membranes. The contact time for the Toray TMG10D membrane was used as the set contact time for the NF membrane as time varied slightly for each RO membrane. The contact time was 10.5 hours of active circulation through the pilot and 15.1 hours in the pilot with no flow with the bleach solution.
  - Disposed of chlorinated water and quenched the remaining chlorine out of the skid with sodium thiosulfate. Tested the concentrate and permeate for chlorine residual with chlorine Hach pillow packets.
  - Conducted a vacuum test to ensure that the mechanical integrity was still intact after oxidation.
  - Repeated integrity, permeability, lead, and tail element tests as described above with the oxidized membrane.
  - Conducted a vacuum test at the end of the oxidized element testing for mechanical integrity assurance.

The value of 85% salt rejection as a basis for chlorine exposure represents an extreme condition that would be unlikely to occur in the operation of an RO system. Prior to reaching this low level salt rejection, permeate conductivity alarms would trigger diversion or shutdown and the defective membranes identified and replaced. This value was selected as a worse case condition and to test the hypothesis that the underlying polysulfone membrane serves as an additional barrier to virus.

8.3 SNWA Test Results

8.3.1 Nominal and Oxidized Salt Rejection Results

The standard sodium chloride rejection results for each test are summarized in Table 8-4 below. The approximate level of chlorine exposure required to decrease salt passage, due to cumulative oxidation of each membrane product is summarized in Table 8-5.

Table 8-4. Sodium Chloride Rejection Observed During the Integrity Tests Which Were Performed Prior to each Experiment to Assess Membrane Integrity.

Membrane Type	Test Condition	Test Type	NaCl Rejection (%)
Hydranautics ESPA2-LD-4040 (RO Membrane)	New Membrane	Lead	99.6
		Lead Duplicate	99.6
		Tail	99.5
	Oxidized Membrane	Lead	85.1
		Lead Duplicate[1]	85.8
		Tail	78.9
Toray TMG10D (RO Membrane)	New Membrane	Lead	99.5
		Lead Duplicate	99.5
		Tail	99.5
		Tail Duplicate	99.5
	Oxidized Membrane	Lead	87.9
		Tail	84.6
CSM NE4040-40 (NF Membrane)	New Membrane	Lead	24.0
		Lead Duplicate[2]	24.9
		Tail	24.3
	Oxidized Membrane	Lead	38.9
		Lead Duplicate	25.8
		Tail	27.8
Filmtec BW30XFRLE 4040 (RO Membrane)	New Membrane	Lead	99.4
		Lead Duplicate	99.3
		Tail	99.4
	Oxidized Membrane	Lead	87.8
		Tail	91.3
		Tail Duplicate	64.6

Notes:

1. The duplicate test was performed on a different membrane than the other Hydranautics ESPA2-LD-4040 tests.
2. The duplicate test was performed on a different membrane than the other CSMNE4040-40 tests that did not pass the pressure decay test.

Table 8-5. The Chlorine Exposure Needed to Oxidize each Membrane to the Salt Rejection in Table 8-4.[1]

Membrane Type	Chlorine Exposure (mg-h/L)
Hydranautics ESPA2-LD-4040	21,920 - 29,820[2]
Toray TMG10D	25,580
CSM NE4040-40	25,580[3]
Filmtec BW30XFRLE 4040	60,270

Notes:
1. Calculated from the estimated chlorine concentration used at the beginning of each test and the contact time.
2. The Hydranautics oxidized lead and tail tests were performed with a membrane that had a chlorine exposure of 29,820 mg-h/L while the oxidized lead duplicate test was performed with a different oxidized membrane that had an estimated chlorine exposure of 21,920 mg-h/L.
3. The CSM NE4040-40 membrane has a salt rejection below the desired sodium chlorine rejection threshold established for the oxidized membrane condition. Thus, the membrane was exposed to the same estimated amount of chlorine as the Toray TMG10D.

Abbreviations: mg-h/L - milligrams per liter times hours.

The Hydranautics ESPA2-LD-4040 estimated chlorine exposure was 29,820 mg-h/L for the oxidized lead and tail tests while the oxidized lead duplicate membrane had an estimated chlorine exposure of 21,920 mg-h/L. The chlorine exposure resulted in a NaCl rejection after oxidation of 85.1% for the lead, 85.8% for the lead duplicate, and 78.1% for the tail.

The Toray TMG10D had an estimated chlorine exposure that falls within the range of the two chlorine exposures for the Hydranautics ESPA2-LD4040, 25,580 mg-h/L, which resulted in a NaCl rejection of 87.9% for the lead and 84.6% for the tail.

The CSM NE4040-40 membrane has a salt rejection below the desired NaCl rejection threshold established for the oxidized membrane condition. Thus, the membrane was exposed to the same estimated amount of chlorine as the Toray TMG10D. The chlorine exposure did not reduce the NaCl rejection of the NF membrane but rather resulted in a slight increase in rejection going from ~24% for the new membrane tests to 25-38% for the oxidized tests. The high chlorine exposure and little effect seen by the NF membrane points to a possible increased oxidation resistance of the CSM NE4040-40 membrane.

The BW30XFRLE 4040 required an estimated chlorine exposure of 60,270mg-h/L to achieve a NaCl rejection of 87.8% for the lead, 91.3% for the tail, and 64.6% for the tail duplicate tests. The BW30XFRLE 4040 membrane required more than two times the estimated amount of chlorine compared to the two other RO membranes required to achieve a NaCl rejection within the desired threshold. Thus, it can be inferred that the BW30XFRLE 4040 had increased oxidation resistance. A drop in NaCl rejection was seen between the lead and tail tests for the oxidized conditions for the three RO membranes inferring a drop in performance. The Hydranautics ESPA2-LD-4040 and Toray TMG10D had a 3.3-6.9% drop in NaCl rejection between oxidized lead and tail tests. The BW30XFRLE 4040 had a 23.2% drop between the oxidized lead and duplicate tail test. The extensive chlorine exposure required for this particular membrane likely caused severe damage to the membrane surface. Over time as the membrane was tested for the lead, tail, and then tail duplicate test, the membrane deteriorated more causing the performance and NaCl rejection to drop.

8.3.2 Marker Rejection

Both new and oxidized membranes were tested at conditions simulating the lead and tail elements of an RO treatment train. Each membrane and experimental condition were completed in single replicates with randomized duplicate tests performed throughout the project. Twenty-four experiments were completed for four different membranes. For each experiment, the LRVs for sulfate, sucralose, uranine, MS2, and electrical conductivity (EC) were calculated and a summary of the LRV data is in Table 8-6.

Table 8-6. LRV for each Membrane in New Condition and Oxidized by Chlorine for Tests Corresponding to Lead and Tail Positions.

Membrane Type and Condition		Test Type	Marker LRV				
			Sulfate	Sucralose	Uranine	MS2	EC
Hydranautics ESPA2-LD-4040 RO Membrane	New	Lead	3.4	3.1	3.4	5.9 ± 1	3.2
		Lead Duplicate	3.5	3.2	3.4	5.8 ± 2	3.2
		Tail	2.7	2.7	2.9	5.0 ± 0.5	2.4
	Oxidized	Lead	1.9	1.1	1.4	N/A	1.8
		Lead Duplicate[1]	1.4	1.2	1.4	5.4 ± 0.4	1.3
		Tail	1.3	1.1	1.0	5.9 ± 0.2	1.2
Toray TMG10D RO Membrane	New	Lead	3.5	N/A	3.5	N/A	3.2
		Lead Duplicate	3.9	3.3	3.6	6.6 ± 0.2	3.2
		Tail	2.5	2.7	2.7	7.5 ± 0.1	2.3
		Tail Duplicate	2.5	2.7	2.8	>7.8 ± 0.3	2.3
	Oxidized	Lead	2.4	1.9	2.2	6.2 ± 0.2	2.2
		Tail	1.2	1.2	1.4	7.0 ± 1	1.1
CSM NE4040-40 NF Membrane	New	Lead	2.3	1.9	2.3	6.0 ± 0.5	1.5
		Lead Duplicate[2]	2.2	1.9	2.1	5.8 ± 0.5	1.6
		Tail	1.8	1.4	2.0	6.2 ± 0.2	1.6
	Oxidized	Lead	2.5	1.9	2.4	2.5 ± 2.6	1.8
		Lead Duplicate	2.5	1.9	2.4	7.1 ± 0.0	2.2
		Tail	1.8	1.3	2.0	7.0 ± 0.5	1.6
Filmtec BW30XFRLE 4040 RO Membrane	New	Lead	2.9	2.7	2.7	2.5 ± 0.7	2.6
		Lead Duplicate	3.1	3.1	3.1	3.6 ± 0.2	2.7
		Tail	2.9	3.0	3.1	4.4 ± 0.1	2.7
	Oxidized	Lead	1.4	1.1	1.5	5.1 ± 0.6	1.3
		Tail	0.52	0.39	0.79	5.3 ± 0.3	0.43
		Tail Duplicate	0.26	0.19	0.55	5.7 ± 0.3	0.23

Notes:

The duplicate test was performed on a different membrane than the other Hydranautics ESPA2-LD-4040 tests.

The duplicate test was performed on a different membrane than the other CSMNE4040-40 tests that did not pass the vacuum decay test.

Table 8-7. Permeate Concentrations of the Spiked Constituents for the New Lead, Lead Duplicate, and Tail Tests for the Three RO Membranes Tested.

Membrane Type	Test Type	Sulfate (mg/L)	Uranine (µg/L)	Sucralose (ng/L)	EC (µS/cm)
BW30XFRLE 4040 RO Membrane	Lead	3.5	3.7	84	12
	Lead Duplicate	1.8	1.6	40	9
	Tail	17	9.0	300	48
ESPA2-LD-4040 RO Membrane	Lead	0.91	0.91	34	3
	Lead Duplicate	0.81	0.82	33	3
	Tail	27	16	560	86
Toray TMG10D RO Membrane	Lead	0.79	0.57	26	3
	Lead Duplicate	0.27	0.51	N/A	3
	Tail	42	23	500	117

8.3.2.1 Sulfate Rejection

The LRVs between the Hydranautics ESPA2-LD-4040 and Toray TMG10D membranes were very similar with little discrepancy between the two. The new membrane lead and tail test sulfate-based LRVs for both membrane types were 3.4-3.5-log and 2.5-2.7-log, respectively. The difference of ~1-log LRV between the lead and tail tests can be explained by the increased concentration of sulfate in the tail tests compared to the lead tests. The increased sulfate concentration during the tail test increases the overall solution concentration gradient causing a greater amount of sulfate to diffuse into the permeate subsequently decreasing the observed LRV (Abdullah et al. 2018). The CSM NE4040-40 NF membrane had lower LRVs of 2.2-2.5-log for the new membrane lead tests and 1.8-log for the new membrane tail test due to the larger pore sizes of the NF membrane allowing greater diffusion of divalent ions. The CSM NE4040-40 membrane data sheet reports 99.5% removal of magnesium sulfate which is equivalent to an LRV of 2.3-log for sulfate. Thus, the results observed for sulfate for the lead test are within the expected range for the commercial membrane.

The Filmtec BW30XFRLE 4040 RO membrane had lower LRVs for the new membrane lead tests, 2.9-3.1-log, in comparison to the other two RO membranes, 3.4-3.5-log. However, the BW30XFRLE 4040 new membrane tail test LRV, 2.9-log, was consistent with the observed LRVs for the other RO membranes. Interestingly, there was little difference between the LRVs for the new membrane lead and tail test conditions. This might be explained by the BW30XFRLE 4040 manufacturing and/or membrane stabilization time. The 4-inch BW30XFRLE 4040 membrane is not commercially available and must be specialty made. This may have led to a manufacturing error, which could have affected the membrane's performance. Note, the other three membranes tested were able to reach stabilization and achieve the minimum salt rejection given in the membrane data sheet within 30-60 minutes of operation. However, the BW30XFRLE 4040 membrane required much longer operation (1-3 days) before reaching the minimum salt rejection. This agrees with the Filmtec's (BW30XFRLE 4040) technical manual stating that after installation, stabilization may be achieved within hours or multiple days (DuPont 2021). Therefore, it is probable that even after reaching the minimum salt rejection, the membrane had yet to achieve optimal performance for the first two tests (lead and lead

duplicate tests) and thus had lower LRVs compared to the other two RO membranes. By the time of the tail test, the membrane may have had enough operation time to achieve the optimal performance. This is supported by the permeate concentrations given in Table 8-7, which shows a notable decrease in the permeate concentrations for the spiked constituents between the lead and lead duplicate tests for the BW30XFRLE 4040 membrane. The ESPA2-LD-4040 and Toray TMG10D membranes had lower concentrations along with little concentration discrepancies between the duplicate tests. Interestingly, despite having higher permeate concentrations for the lead tests, the BW30XFRLE 4040 membrane permeate concentrations for the tail test were lower as compared to the ESPA2-LD-4040 and Toray TMG10D membranes, also suggesting stabilization may have not occurred during the lead tests but occurred by the tail test.

For all three RO membranes, oxidation of the membrane decreased the LRVs between the lead and tail tests. A difference in LRVs of 1.0 - 2.6-log was observed between the new and oxidized membranes for both the lead and tail tests. The oxidation of the membrane breaks down the top layer of the membrane surface allowing for greater diffusion of sulfate (Surawanvijit et al. 2016). In contrast, very little change was seen in the LRVs for the new vs. oxidized membrane conditions for the CSM NE4040-40 membrane, possibly indicating an increased oxidation resistance of the NF membrane. The oxidized Filmtec BW30XFRLE 4040 testing indicated a ~1 LRV drop for the lead test (LRV = 1.4) and a significant drop in LRVs of 2.5-2.6 for the tail tests (LRVs = 0.3-0.5). As previously discussed in the membrane oxidation section above, the Filmtec BW30XFRLE 4040 membrane had over two times the chlorine exposure as the two other RO membranes. A decrease in membrane LRV performance was observed for each subsequent oxidized membrane test (lead, tail and then tail duplicate). The duplicate tail tests were not consistent in LRV performance inferring that even after oxidation had occurred and was quenched from the system; the membrane performance was worsening over time. Thus, it can be assumed that the membrane surface was damaged so severely due to oxidation that it was deteriorating over time shown by the decreasing LRVs for each subsequent test.

8.3.2.2 Conductivity

The conductivity in the synthetic feed water is solely contributed to by the sodium sulfate. Therefore, the conductivity LRVs are not indicative of the LRVs that a typical potable reuse system would achieve. If more monovalent ions were present, such as in a typical potable reuse matrix (i.e., treated wastewater), the difference in LRVs observed between sulfate and conductivity would have likely been greater. The LRVs between the Hydranautics ESPA2-LD-4040 and TMG10D membranes were very similar with little discrepancy between the two. The new membrane lead and tail test LRVs for both membrane types were 3.2-log and 2.3–2.4-log, respectively. The oxidized membrane lead and tail test LRVs for both membrane types were 1.8–2.2-log and 1.1-1.2-log, respectively. The observed trend for sulfate was also observed for conductivity where the CSM NE4040-40 NF membrane had lower LRVs compared to the RO membranes due to the larger pore structure of the NF membrane. The LRVs were also consistent between the new and oxidized conditions as reported for sulfate. The LRVs ranged from 1.5-2.2-log for the lead tests and were 1.6-log for the tail tests.

LRVs for the Filmtec BW30XFRLE 4040 membrane were 2.6-2.7-log for new membrane lead tests and 2.7-log for the new membrane tail test. Thus, for the new membrane condition, the Filmtec BW30XFRLE 4040 did not exhibit the ~1 LRV decrease between the lead and tail tests as observed for the other RO membranes for both sulfate and conductivity due to the lowered rejection of the BW30XFRLE 4040 membrane during the lead tests as the membrane likely did not reach optimal performance. The LRVs for the Filmtec BW30XFRLE 4040 oxidized membrane condition were 1.3 for the lead test and 0.2-0.4 for the tail tests. As seen for the sulfate results, the BW30XFRLE 4040 membrane experienced the most extensive drop in LRVs for the oxidized tail tests. The conductivity LRVs correlate very closely with the LRVs observed for sulfate. This is due to the sulfate contributing most of the measurable electrical conductance in the synthetic test solution. The sulfate LRVs were slightly higher compared to conductivity due to the presence of sodium from the sodium sulfate used to dose sulfate which is a smaller compound and can diffuse into the permeate at a higher rate than sulfate thus lowering the conductivity LRV.

8.3.2.3 Sucralose

For the new membrane conditions, the LRVs between the Hydranautics ESPA2-LD-4040 and TMG10D were very similar with little discrepancy between the two. The new membrane lead and tail test LRVs for both membrane types were 3.1-3.2 and 2.7, respectively. The difference of ~0.5 LRV between the lead and tail tests, like sulfate, can be explained by the increased concentration of sucralose in the tail tests compared to the lead tests. The sucralose LRVs for the Filmtec BW30XFRLE 4040 followed a similar trend as seen for sulfate and conductivity for this membrane, where the new membrane lead and tail LRVs (2.7-3.1) had little discrepancy between the two types of tests as discussed above due to the lowered membrane performance during the lead and lead duplicate tests.

For the oxidation tests, a difference in LRVs was observed between the lead tests of the Hydranautics ESPA2-LD-4040 and the Toray TMG10D. The Hydranautics ESPA2-LD-4040 had an LRV of 1.1 for both the lead and tail tests, while the Toray TMG10D had an LRV of 1.9 for the lead and 1.2 for the tail. The lower LRV for the Hydranautics ESPA2-LD-4040 lead test can be explained by the greater extent of oxidization of the Hydranautics ESPA2-LD-4040 observed in the NaCl rejection after oxidation compared to the Toray TMG10D which was discussed above in the salt characterization section and can be seen in Table 8-6. The greater extent of oxidation of the Hydranautics ESPA2-LD-4040 membrane would allow a greater amount of sucralose to diffuse into the permeate lowering the LRV. The LRVs observed for the sucralose LRVs for the CSM NE4040-40 membrane were consistent between the new and oxidized conditions with 1.9-2.2-log for lead tests and 1.3-1.4-log for tail tests. As seen for conductivity and sulfate, oxidation caused a significant drop in LRVs (1.1-0.2) for the Filmtec BW30XFRLE 4040 membrane which lowered for each oxidation test (lead and tail).

8.3.2.4 Uranine

For the new membrane conditions, as seen by sulfate and sucralose, the LRVs between the Hydranautics ESPA2-LD-4040 and Toray TMG10D membranes were very similar with little discrepancy between the two. The new membrane lead and tail test LRVs for both membrane types were 3.4–3.6-log and 2.7–2.9-log, respectively. The difference in LRVs between the lead and tail tests, like sulfate and sucralose, can be explained by the increased concentration of uranine in the tail tests compared to the lead tests. The CSM NE4040-40 had very similar LRVs for uranine as it did for sulfate. In addition, as observed for sulfate, conductivity, and sucralose, there was very little change in LRVs between the new and oxidized condition with LRVs of 2.1—2.4-log for lead test and 2.0-log for tail tests. The Filmtec BW30XFRLE 4040 had LRVs of 2.7—3.1 for new membrane lead tests and 3.1-log for the tail test.

For the oxidation tests, the same trend was observed as was noted for sucralose with the Hydranautics ESPA2-LD-4040 having lower LRVs compared to the Toray TMG10D which further points to the greater extent of oxidation as the cause for difference in LRVs. The Hydranautics ESPA2-LD-4040 had a difference in LRVs of ~2-log for both the lead and tail tests while the Toray TMG10D had a difference of ~1.3-log. For the BW30XFRLE 4040, the oxidized membrane conditions experienced a significant drop in rejection with LRVs of 1.5-log for the lead test and 0.6—0.8-log for tail tests, a trend which was also observed for sulfate, conductivity, and sucralose.

A mass balance was performed for each experiment to understand if uranine adsorbed to the membrane surface. For all lead experiments (new and oxidized conditions), the percent differences in uranine loading between the feed and the combined permeate and concentrate were all 10.5% or less. It is reasonable to say that 10% or less is not significant enough to link to adsorption to the membrane surface as both human and analytical error could contribute to this small difference in uranine loading. However, for all tail tests, percent differences ranging from 27-72% were observed. This loss in uranine loading from the feed to the combined concentrate and permeate is hypothesized to be absorption of the uranine to the membrane surface. The greater concentration of uranine dosed during the tail tests (six times higher, see Figure 8-6) compared to the lead tests facilitated the adsorption to the membrane surface. Hydrophobic interactions between the RO and NF membrane surfaces and hydrophobic compounds have been reported to facilitate adsorption of these compounds to the membrane surface (Braeken et al. 2005; Kim et al. 2018; Kimura et al. 2003; Van Der Bruggen et al. 2002; Xie et al. 2012). Therefore, since uranine is a hydrophobic compound, it is reasonable, that at the higher concentrations used during the tail tests, uranine loading may have been lost to adsorption to the membrane surface.

Figure 8-6. Photo of the Feed Bucket Containing the Uranine Dosed for the Tail Test Condition.

8.3.2.5 MS2

It is important to note that both LRV values as well as MS2 concentrations should be considered to evaluate performance as varying concentrations of MS2 in the feed can alter the LRV while the concentrations of MS2 in the permeate remain the same. A difference in LRVs between the Hydranautics ESPA2-LD-4040 and Toray TMG10D membranes were observed for MS2 with the Toray TMG10D having consistently higher LRVs as well as lower permeate concentrations (see Table 8-8) compared to the Hydranautics ESPA2-LD-4040. The Hydranautics ESPA2-LD-4040 had LRVs of 5.8–5.9 and 5 for the lead and tail tests, respectively, while the Toray TMG10D had LRVs of 6.6 and 7.5–>7.8 for the lead and tail tests, respectively. Thus, the Toray TMG10D is more effective for MS2 rejection compared to the Hydranautics ESPA2-LD-4040. The CSM NE4040-40 membrane had high LRVs and low permeate values for both the new membrane lead and tail tests. This is consistent with the Hydranautics ESPA2-LD-4040. The LRVs for lead tests were 5.8—6-log and 6.2-log for the tail test. The Filmtec BW30XFRLE 4040 membrane had a reduced performance for MS2 rejection compared to the other three membranes observed by both the lower LRVs and higher permeate values. The new membrane lead tests had LRVs of 2.5—3.6-log and 4.4-log for the tail test. The steady increase in LRVs observed for the new membrane tests supports the hypothesis that the BW30XFRLE 4040, while having met the minimum salt rejection, had not yet reached optimal performance and thus an increase in LRVs was seen for each constituent, including MS2, between the lead, lead duplicate, and tail tests.

For the oxidation tests, The Toray TMG10D had decreased LRVs compared to the new membrane, but they still achieved LRVs above 6. The Hydranautics ESPA2-LD-4040 had a decrease in LRV for the lead test and an increase in LRV for the tail test. The decrease in LRV for the lead test can be explained by greater passage of the MS2 into the permeate due to the oxidization of the membrane surface, reducing rejection performance. This is supported by the higher permeate values for the oxidized conditions compared to the new membrane conditions. The oxidization had less of an impact on the Toray TMG10D and CSM NE4040-40 membranes with the Toray TMG10D still achieving LRVs above 6 and the CSM NE4040-40 achieving LRVs of 5.8 or higher except for one CSM NE4040-40 lead test. In addition, the concentration of MS2 in the permeate remained low. The steady increase in MS2 rejection observed for the new membrane tests for the BW30XFRLE4040 was also observed during the oxidation tests. The LRVs increased from 5.1 for the oxidized lead test to 5.7 for the oxidized tail duplicate test. Thus, further supporting that the extended run time necessary for optimal membrane performance had not been met until the oxidized lead tests were performed.

Discrepancies in MS2 theoretical dosing concentrations and the lab reported concentrations for the lead tests was an observed challenge. Kinetic tests and varied timed feed samples (samples taken directly after dosing before running the feed solution through the pilot and samples taken after ~30 min of operation when steady-state had been reached) determined the discrepancy to most likely be due to accumulation of MS2 on the membrane surface due to increased hydrophobic interactions provided by the high concentration of sulfate (Farrah, 1982) which reduced the LRVs for MS2 because the system was run in batch mode and not pass through. Absorption to the membrane surface reduced the overall concentration of MS2 in the system that can be removed between the feed and permeate. Increased flushing volume between tests and base cleanings after tail tests were determined to help remove residual MS2

and uranine from the membrane surface reducing adsorption sites to the membrane surface. In addition, due to the experimental variability of MS2 dosing and sampling at such high concentrations, a mass balance of the MS2 loading did not allow for accurate differentiation between losses of MS2 to adsorption on the membrane surface and experimental variability.

Table 8-8. The Average MS2 Feed and Triplicate Permeate Data Points with the Calculated Standard Deviations and LRVs.

Membrane Type and Condition		Test Type	MS2 Feed (PFU/mL)	MS2 Permeate (PFU/mL)[1]				LRV
				No. 1	No. 2	No. 3	σ	
Hydranautics ESPA2-LD-4040 RO Membrane	New	Lead	2.1×10^5	0.2	0.4	0.1	0.2	5.9 ± 1.0
		Lead Duplicate	3.2×10^5	<0.1	0.1	1.6	0.8	5.8 ± 2.0
		Tail	5.2×10^6	33	58	57	14	5.0 ± 0.5
	Oxidized	Lead	9.2×10^5	2.9	3.3	4.5	0.8	5.4 ± 0.4
		Lead Duplicate[2]	N/A	N/A	N/A	N/A	N/A	N/A
		Tail	2.1×10^6	2.6	2.3	2.7	0.2	5.9 ± 0.2
Toray TMG10D RO Membrane	New	Lead	4.1×10^5	<0.1	0.1	<0.1	0.0	6.6 ± 0.2
		Lead Duplicate	N/A	N/A	N/A	N/A	N/A	6.6 ± 0.2
		Tail	2.8×10^6	0.1	<0.1	<0.1	0.0	7.5 ± 0.1
		Tail Duplicate	6.5×10^6	<0.1	<0.1	<0.1	0.0	>7.8 ± 0.3
	Oxidized	Lead	1.7×10^5	0.1	<0.1	<0.1	0.0	6.2 ± 0.2
		Tail	6.4×10^6	0.8	0.8	0.1	0.4	7.0 ± 1.0
CSM NE4040-40 NF Membrane	New	Lead	1.1×10^6	1.0	1.6	0.7	0.4	6.0 ± 0.5
		Lead Duplicate[3]	1.5×10^6	2.8	2.0	1.8	0.6	5.8 ± 0.5
		Tail	9.6×10^6	6.6	5.4	6.8	0.7	6.2 ± 0.2
	Oxidized	Lead	4.9×10^5	0.3	0.1	4,446[4]	2,567	2.5 ± 3.0
		Lead Duplicate	1.5×10^6	0.1	0.1	<0.1	0.0	7.1 ± 0.0
		Tail	8.4×10^6	0.9	1.0	0.5	0.3	7.0 ± 0.5
Filmtec BW30XFRLE 4040 RO Membrane	New	Lead	3.7×10^4	171	159	50	66	2.5 ± 0.7
		Lead Duplicate [5]	1.2×10^6	360	267	354	52	3.6 ± 0.2
		Tail	1.1×10^7	437	452	478	21	4.4 ± 0.1
	Oxidized	Lead	1.0×10^6	8.2	10	4.5	2.8	5.1 ± 0.6
		Tail	1.2×10^7	63	73	50	12	5.3 ± 0.3
		Tail Duplicate	1.1×10^7	22	19	17	2.5	5.7 ± 0.3

Notes:

1. The triplicate permeate samples were taken with less than 1 min in between samples.
2. The duplicate test was performed on a different membrane than the other Hydranautics ESPA2-LD-4040 tests.
3. The duplicate test was performed on a different membrane than the other CSMNE4040-40 tests that did not pass the vacuum decay test.
4. Lab confirmed the sample was correct by separate plate testing. It is likely contamination occurred during the sampling event.
5. The duplicate test was performed due to the low MS2 feed concentration in the original lead test.

8.4 SNWA Testing Summary

The maximum observed LRV for the Hydranautics ESPA2-LD-4040 and Toray TMG10D new membrane tests for sulfate, sucralose, uranine, and conductivity were very comparable, 3.1-3.5-log for the lead tests and 2.7-2.9-log for the tail tests, with sulfate and uranine being consistently the highest of the four parameters. While having lower LRVs, the CSM NE4040-40 and Filmtec BW30XFRLE 4040 exhibited similar trends with sulfate and uranine LRVs being consistently higher. It is important to note, that the BW30XFRLE 4040 membrane may not be the same membrane used for full-scale applications as the 4-inch element was custom made and issues with membrane operation stabilization were experienced. However, consistent data was observed for the randomized duplicate tests throughout the project.

The challenges associated with using uranine as a molecular marker, such as high chemical dosing, disposal, and difficult chemical handling (see Figure 8-6), point to sulfate being a preferred molecular marker.

While the difference in LRVs observed between sulfate and conductivity were minimal, 0.1-0.3-log, it is important to note the synthetic test solution comprised mostly of a divalent ion. If more monovalent ions were present, such as in a typical potable reuse matrix (i.e., treated wastewater), the difference in LRVs observed between sulfate and conductivity in a real feedwater would likely have been greater. Assuming typical wastewater has the necessary background sulfate concentrations, the higher concentrations of monovalent ions would decrease conductivity's ability to sustain LRVs as high as sulfate due to monovalent ion diffusion. Therefore, in a typical potable reuse matrix, the gap between sulfate and conductivity LRVs is anticipated to increase.

The greatest challenges for sulfate to be used as a molecular marker will be its indigenous wastewater concentration as well as finding a suitable analytical instrument with a low enough detection limit to allow demonstration of high LRV.

A key finding is the limitation of the tested membranes to demonstrate LRVs of 4 or greater for the tested molecular markers due to diffusion limitations. Each molecular marker was dosed and tested so that a 4-log removal could be demonstrated by the analytical instruments. However, with nominal diffusion, sulfate, conductivity, sucralose, and uranine were unable to reach LRVs of 4-log. The necessary concentrations for this to be demonstrated caused the concentration gradient to increase to the point that diffusion of the molecular markers occurred, so the LRVs were below 4. It can also be concluded that other similar compounds would be expected to be unable to reach LRVs of 4-log as well.

Nevertheless, all markers tested responded to membrane oxidation and behaved as anticipated, with lower observed rejections of tail, relative to lead elements. Further, due to the ability of the molecular markers to move through the RO membrane by diffusion, they yielded more conservative LRVs than MS2 and were much more practical for ongoing system verification.

The testing appears to suggest that there may be some differences in manufacturing between membrane elements and some elements such as the DuPont BW30-XFRLE may require a conditioning period to fully achieve the ultimate virus removal capabilities. Again, it is noted that the RO membranes evaluated were not specifically developed, marketed, or sold for their virus removal ability. In the case of the DuPont membrane, the element was supplied for the testing as a non-commercially available product.

CHAPTER 9

Approaches for Crediting High Pressure Membranes

This chapter compiles the results of the tests conducted as part of this project with the focus of outlining considerations on how to implement pathogen crediting in RO and NF systems used in potable reuse.

9.1 Evaluation of Surrogate Performance

9.1.1 Sensitivity

The sensitivity of a surrogate is defined by the LRV able to be demonstrated (USEPA 2005). The markers evaluated in this report were primarily limited by diffusion as to the upper bound of LRV sensitivity (as demonstrated in Chapter 8). Ultimately, the fact that diffusion caps the maximum sensitivity for surrogates means that implementation would inherently provide a level of conservatism when compared to actual virus removal. For example, the LRV calculated from conductivity profiling was conservatively capped to demonstrate a maximum of 5-log and by virtue of the selected calculation upper limit, the maximum sensitivity of this technique has been limited.

In practice, additional site-specific limitations exist in the typical concentrations in RO feedwater as well as analytical detection limits for each marker in the permeate. Of the markers tested, strontium was very sensitive to feedwater concentrations (as noted when comparing OCWD to WCWA results). Sulfate concentrations were generally higher and able to demonstrate consistent results across multiple sites. In part, this is because sulfate ions are added when sulfuric acid is used for pH correction in the RO feed which enhances the baseline concentration. The range of LRVs demonstrated for each technique evaluated in this report is summarized below in Table 9-1.

Table 9-1. Summary of Surrogate Sensitivity Observed for High Pressure Membranes During Field and Pilot Tests.

Surrogate	Sensitivity Verified (LRV Range)	Sites Demonstrated
Direct Conductivity	1.2 - 2.1	WCWA, YVWD, OCWD[1]
Conductivity Profiling	2.7[2] - >5.0	WCWA, YVWD, OCWD, Sites 1 - 4
Sulfate	2.6 - 3.9	WCWA, YVWD, OCWD, SNWA[3]
Strontium	>1.8[4] - 3.9	WCWA, OCWD
Sucralose	2.7 - 3.3	SNWA[3]
Phosphate	3.0 - >3.2	WCWA
Magnesium[4]	>2.0->2.1	WCWA
Uranine (Direct)	2.7 - 3.8	WCWA[5], YVWD, OCWD, SNWA[3]
Uranine Profiling LRV	3.4 - 3.5	OCWD

Notes:

1. SNWA conductivity results were omitted as they were due to the presence of divalent sulfate only and were able to demonstrate higher than typical LRVs with indigenous conductivity.
2. Conductivity profiling LRV of 2.7 was from an array suspected of containing integrity failure and was the lowest value observed. Typical conductivity profiling LRVs were >3.5
3. Oxidized and nanofiltration membrane results were excluded from sensitivity reporting. The NF membrane tested at SNWA generally achieved lower LRVs of surrogates but was resistant to oxidation. MS2 rejection by the NF membrane was more than 5.
4. Low range strontium LRV limited by low feedwater abundance at WCWA. Magnesium LRVs were all removed to below the permeate limit of detection and are underestimates.
5. Maximum direct uranine LRV of 4.2 was observed during WCWA pilot challenge testing but the permeate concentration was anticipated to be lower than steady state for this sample. LRVs stabilized between 3.6 to 3.8.

9.1.2 Resolution

The resolution of a DIT is defined by the smallest defect able to be detected (USEPA 2005). A proposed approach to evaluate resolution for each technique is to compare the reported molecular weight as well as hydrodynamic radii for surrogates evaluated in this report. The assumption with reporting this data is that provided the molecular weight and hydrodynamic radii, if available, of a surrogate is smaller than a virus, then the surrogate should have sufficient resolution to verify a virus sized breach.

Based on hydrodynamic radii, ions anticipated to make up conductivity in RO feedwater are approximately 100 to 1,000 times smaller than MS2. The molecular weight difference is even larger with common ions likely to be at least 16,820 times lighter than the molecular weight of MS2. Consequently, the surrogates listed in Table 9-2 have more than sufficient resolution to also verify protozoa sized breaches.

For the individual marker species, sulfate, strontium, phosphate, and magnesium, the hydrodynamic radii are 140 to 250 times smaller than MS2 and the molecular weight was between 37,500 to 150,000 times lower.

Hydrodynamic radii for sucralose and uranine molecules were not available, but their molecular weights are 9,050 and 9,570 times lighter than MS2 respectively, suggesting they should be sufficiently conservative to detect virus sized breaches.

Cryptosporidium oocysts are 3 µm (i.e., 120 times larger than MS2) and *Giardia* cysts are even larger. Consequently, the surrogates listed in the table have more than sufficient resolution to also verify protozoa sized breaches.

Table 9-2. Relative Size and Molecular Weight of Surrogates Compared to MS2.

Surrogate	Surrogate MW (Da)	MS2 MW[1] / Surrogate MW	Surrogate Hydrodynamic Radii (nm)[2]	MS2 Size/ Surrogate Radii
Direct Conductivity and Conductivity Profiling[3]	1 - 214	>16,820	0.027 – 0.27	92 - 930
Sulfate	96	37,500	0.12	220
Strontium	88	40,900	0.16	160
Sucralose	398	9,050	-[4]	-[4]
Phosphate	95	37,900	0.10	250
Magnesium[4]	24	150,000	0.17	140
Uranine	376	9,570	-[4]	-[4]

Notes:

1. MS2 MW reported to be 3.6 x 10^6 g/mol (Kuzmanovic et al. 2003) and size of 25 nm (Antony et al. 2012).
2. Hydrodynamic radii of ions reported from (Kadhim and Gamaj 2020) unless otherwise specified.
3. Range of molecular weights and sizes for common ions reported in (Kadhim and Gamaj 2020) used to define minimum and maximum for conductivity.
4. The hydrodynamic radii for uranine or sucralose was not able to be found.

9.1.3 Frequency

To qualify as a DIT per the MFGM, daily integrity verification must occur (USEPA 2005). All of the surrogates investigated in this report could be analyzed daily. However, the level of effort and cost to achieve such high frequency and representative measurement differs for each of the surrogates.

For example, automated conductivity profiling provides an assessment of rejection at an individual pressure vessel level and relies on indigenous surrogates. Other indigenous DIT surrogates include sulfate, strontium, and phosphate. Measuring these indigenous surrogates at a vessel level requires expensive analytical equipment and trained lab staff. Measurement at a vessel level could be achieved with online monitors for the indigenous surrogates, especially if a single analyzer could be used for a number of arrays and analysis time was sufficient to allow testing of the permeate from the multiple arrays within a single day.

Non-indigenous (i.e., spiked or continuously fed) surrogates, like uranine, could be analyzed by relatively simple equipment with rapid onsite analysis, but would require additional equipment to feed the surrogate. Pulsed non-indigenous surrogates have been commercially provided in the past to allow confirmation of RO integrity at a frequency of at least daily, but it is anticipated that the practical limitations of uranine including pH sensitivity, difficult stock

solution preparation, and the propensity to degrade with UV exposure. These factors inhibit the practical nature of using this compound for routine array challenge testing.

9.1.4 Assurance of Risk Mitigation

RO systems, and more generally membrane systems, do not suffer the same magnitude of potential treatment upsets compared to other disinfection processes where LRV can reduce to 0-log if dose delivery shuts off. For example, a power outage or ballast failure would result in UV lamps shutting off and having water transfer through a UV reactor without being exposed to photolysis. In this instance, the high LRVs attributed to a UV system could realistically drop from greater than four to zero. There are no documented hazardous events where a membrane system has experienced undetected damage sufficient to bypass 100% of the flow in an untreated state. Rather, some small proportion of flow may pass untreated through physical defects such as an O-ring breach. In the hypothetical example below, an RO system would not be anticipated to reduce to a LRV of less than 1.0-log without the permeate conductivity reaching values 300% higher than normal. Under typical operations, high pressure membrane elements would be replaced well before reaching this condition. Permeate conductivity measurement is instantaneous and this level of gross failure could be detected and quickly rectified by diversion based on a maximum conductivity limit.

The unlikely event of gross integrity failure aside, damage and decline of LRV in membrane systems is anticipated to be minimal. Trending of periodic marker tests or conductivity profiles should be adequate to characterize changes in integrity. The gradual change in integrity was highlighted in the conductivity profiling results that identified a cracked module at YVWD. This defect resulted in an identifiable and prolonged exceedance of profile results. It is important to note, that at the time the defect was identified by conductivity profiling, the RO skid was challenge tested and was still verified to be achieving an MS2 LRV greater than 5-log since the proportion of flow through he cracked module was very low (<<0.001%). As a test methodology, conductivity profiling appears to be more sensitive than marker-based methods.

For the LRV of a membrane system to reduce from a nominal level (5-log MS2 rejection is considered typical for RO) to levels at which RO is credited on conductivity (e.g., 1.5- log), a significant proportion (1 - 10%) of elements would have to be completely bypassed (e.g., a total failure of the polyamide and supporting PES membrane sheet). A hypothetical scenario is outlined below and illustrated in Figure 9-1. The hypothetical scenario is subject to the following assumptions:

- An RO system operates with a moderate feedwater conductivity of 1200 µS/cm and a typical permeate conductivity of 40 µS/cm (e.g., 1.5-log).
  - If the feedwater conductivity were to increase (noting that a number of plants in Southern California have an RO feed conductivity closer to 2000 µS/cm), then the LRV able to be demonstrated by conductivity would also increase and the impact of blending into the permeate via defects would also be magnified.
- The maximum LRV of 5-log MS2 is conservatively assigned. If higher LRVs are assigned, MS2 LRVs in the figure below would be transformed up the Y axis by 1-log.

- Markers are assumed to be present at concentrations in the feedwater and have permeate detection limits to demonstrate up to a maximum LRV of 3.0-log.

The figure below illustrates the impact to MS2 LRV and the corresponding response of direct conductivity as well as marker LRVs in response to set proportions of flow that are bypassed un-treated. The typical credit of 1.5-log based on conductivity monitoring is marked for reference. In addition, an arbitrary alarm limit of 30% higher than typical values (assumed to be 40 µS/cm) is set for the permeate conductivity. Projected permeate concentrations based on the bypassed flow are shown on a secondary Y-axis.

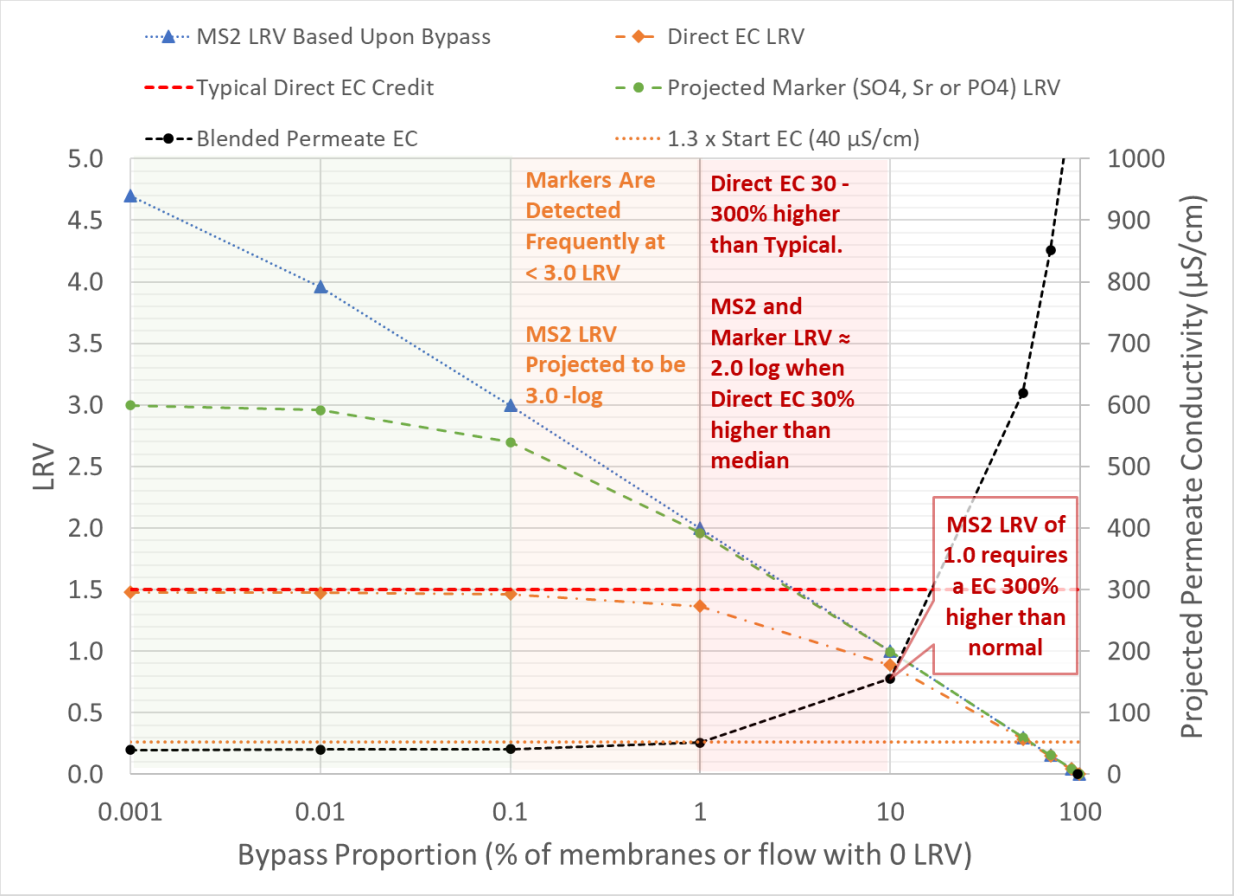

Figure 9-1. Hypothetical Response of MS2, Marker and Conductivity LRV to Increasing Proportions of Bypass Flow.

Note that values of permeate conductivity 300% higher than normal would be required to reduce below 1-log reduction of MS2 rejection (subject to the system assumptions above).

Based on the illustration of the scenario in Figure 9-1, the following comments can be made with respect to RO integrity monitoring:

- Assuming regular monitoring of surrogates, a gradual decline and more frequent detections of the surrogates should occur prior to sufficient bypass flow to allow MS2 LRV to reduce below 3.0 (conservatively assuming a 5-log maximum LRV of MS2).
- Direct permeate conductivity exceedances of more than 30% higher than typical values should be readily observed at flow bypass levels of 1%.

    - If the conductivity values are due to complete bypass (i.e., not just polyamide layer oxidation). At this level of bypass, both surrogate and MS2 LRVs would be anticipated to be close to 2-log.
    - If the conductivity exceedance was due to polyamide layer damage, but the PES support layer was intact, then MS2 LRV would still be anticipated to be high (see oxidized membrane test results in Chapter 8) but surrogate LRVs may have reduced.
- For an RO system to reduce to MS2 LRVs of less than 1-log, approximately 10% of flow would need to bypass untreated. At this level, permeate conductivities would be anticipated to be 300% higher than nominal. A permeate conductivity 300% higher than normal would be rapidly detected and would rapidly identify the breach and take corrective action (i.e., diversion) would automatically occur prior to providing less than 1-log treatment.
- Although not shown on Figure 9-1, conductivity profiling could enhance the sensitivity to breaches. The reason for this is that the profiling conductivity would identify specific vessels which are already responsible for a fraction of possible flow (i.e., a maximum possible bypass proportion which is subsequently diluted by remaining nominal vessels) with conductivity exceedances. This means that if one of one hundred vessels was identified and rectified by conductivity profiling then the same limits as direct EC would apply on the chart above, but the sensitivity would be shifted to the left by 2 orders of magnitude. In other words, a 30% exceedance corresponding to an MS2 LRV of 2-log, would become an MS2 LRV of 4-log, provided there was only 1 vessel exceeding the 30% limit.

The example in Figure 9-1 highlights a number of themes including:

- Direct Conductivity Monitoring should be sufficient to verify LRV of at least 1.0 to 1.5-log.
- Use of surrogates at a sufficient frequency to meet MFGM DIT requirements should be able to verify a minimum of 2.0-log to a probable diffusion and abundance limited maximum of 3.0-log.
- Depending on the number of vessels in a total array relative to the number identified to be above 30% higher than typical values, conductivity profiling may have the ability to improve sensitivity of direct EC measurement by at least 2.0-log. As long as individual vessel limits are carefully selected relative to the total array size and maximum defect flow possible from a single vessel, then conductivity profiling should be able to verify a minimum LRV of 3.0-log to 3.5-log. The maximum conductivity profiling LRV is conservatively limited to 5.0-log by the constant selected for use in the equation:
    - Periodic verification of the minimum sensitivity of conductivity profiling could be achieved by grab sampling of markers.
    - The maximum sensitivity of conductivity profiling could be increased by conducting sufficient MS2 testing to relate median conductivity of a system to a typical MS2 LRV. However, the absolute limitation on this would be 6-log if USEPA MFGM challenge testing guidance was followed. In addition, the number of samples that may be necessary to increase the maximum cap from 5 to 6-log may be cost prohibitive.

9.2 Regulatory Perspectives on RO and NF LRV Credit

Two workshops with health regulators from around the US and Australia were conducted as part of this project. The first workshop was held in August 2021, prior to starting the testing at the various project testing locations. The second workshop was held in March of 2022, after the completion of testing and final project results were presented. Regulators from eight states as well as the USEPA and the Association of State Drinking Water Administrators (ASDWA) attended this workshop. Attendees were also sent an optional survey after the workshop to gauge their input on aspects of RO and NF LRV crediting. The focus questions and responses received from the survey are synthesized below in an effort to capture current perspectives.

1. MS2 Bacteriophage (25 nm diameter) is commonly used as a virus indicator for spiked challenge testing of membrane products and intact RO systems generally achieve MS2 LRVs greater than 5-log. *Giardia* and *Cryptosporidium* are approximately 100 times larger than MS2. If a surrogate (e.g., strontium, sulfate, or conductivity) is suitable to demonstrate virus LRV across RO then should an equal LRV of *Giardia* and *Cryptosporidium* also be claimed based on the same monitoring protocol?

   The general response to this question was yes. It was noted that if research could demonstrate a good correlation between a surrogate for MS2 and MS2 LRVs, then future challenge test results for demonstrating MS2 removals would be accepted. If a given virus removal credit was granted based on MS2 challenge testing, the same LRV could be granted to *Giardia* and *Cryptosporidium*. However, it was noted that Oregon, Virginia, and Texas do not award virus or protozoa credit to RO and consistent with the MFGM does not award virus LRV to MF/UF.

2. The LRV of conductivity (bulk feed to permeate), total organic carbon (TOC), sulfate, strontium, uranine, and a diffusion-adjusted conductivity profiling approach were investigated in WRF4958. In your jurisdiction (i.e., Utility, State, etc.):
   a. Have any of the above listed surrogates been approved to demonstrate RO LRV?
   b. Would any of the surrogates listed above not be suitable for approval, if so, why?
   c. For the surrogates that are approved, what LRV is credited to the RO system?
   d. What monitoring and reporting frequency is required for the approved surrogates?

   It was noted that the Australian WaterVal protocol for RO validation (WaterSecure 2017) outlines approaches for RO validation using conductivity, TOC, dissolved organic matter, sulfate, and rhodamine. During the meeting, it was noted that in California, the use of direct LRV or TOC is presently accepted to claim a 1:1 virus and protozoa credit. In addition, strontium has been approved on a site-specific basis to claim LRVs of 2.5-log. Colorado has allowed the use of sulfate and TOC for surface water compliance indicators and grants 3.0-log of protozoa removal to RO in Bin 1 surface water conditional on downstream chlorination sufficient to achieve 4.0-log virus removal. In Alaska, conductivity is accepted for a continuous indirect integrity monitoring technique. Oregon does not grant RO credit and has not approved the use of the surrogates proposed in this work. Virginia has also not adopted any of the surrogates proposed in this report for crediting RO.

Concerns were raised about spiked surrogates and suggestions were noted that indigenous surrogates (i.e., sulfate and strontium) would be more appropriate provided that analytical detection limits and feedwater concentrations were sufficient to claim a meaningful LRV. Concerns were noted about the use of TOC and conductivity to verify RO removal in locations where the concentrations of those surrogates were low in the feedwater.

For Virginia, although not specifically established for RO systems in our regulations or policy, there was a preference to adopt a crediting approach for RO along the lines specified in the MFGM. That is, allowing *Cryptosporidium* LRV credit for MF and UF membrane filtration if supported by ongoing direct integrity testing. For RO, it was suggested that the credit would be the calculated LRV daily based on ongoing membrane integrity tests rather than awarding a fixed minimum LRV. The overall plant LRV achieved would need to be sufficient to meet the required, *Cryptosporidium, Giardia,* and virus inactivation, including other disinfection unit operations.

With respect to frequency of monitoring, daily integrity testing in line with the MFGM was accepted. That is, if it were an approved surrogate, the frequency would be equivalent to the Direct Integrity Test frequency in LT2ESWTR (once per day of operation, where "day" is any amount of water production). Allowances for less frequent system verification would require clear guidance from the USEPA.

3. For potable reuse, RO systems are always installed after pretreatment (most commonly microfiltration or ultrafiltration) to remove suspended solids and other contributors to RO fouling. RO permeate is then typically treated by high dose UV and potentially chlorination. In drinking water, specific treatment techniques have been attributed with a treatment technology pathogen removal credit conditional on standard operational and design requirements.

   RO is never installed in potable reuse without upstream and downstream treatment trains (i.e., there will be redundant barriers). Additionally, RO will be installed for the purpose of removing TDS (i.e., conductivity). Accordingly, would it be suitable to grant RO a treatment technology pathogen removal credit subject to certain design constraints and permeate conductivity limits?

   There was some receptiveness to a combined treatment technology approach for potable reuse. That is, a crediting framework similar to how LT2ESWTR specifies operation of coagulation and filtration to obtain virus and protozoa removal. However, such an approach would need sufficient caveats, design constraints, and monitoring would need to be defined as part of the approach to ensure that suboptimal pathogen removal, below the credit, was not possible with the RO treatment.

4. If a treatment technology credit could be provided to RO, could there be capacity to increase the level of credit based on monitoring of individual stages or vessels (i.e., using the diffusion-adjusted conductivity profiling approach described in WRF4958)?

   In general, the response was yes, noting that increases in credit would need to be verified by ongoing integrity monitoring. In addition, it was noted that the default treatment

technology credit would be conservatively lowered relative to what could be verified by integrity monitoring and challenge testing. Also, enhanced credit was possible, but maximum LRVs would need to be capped at 6-log per process, consistent with current paradigms.

5. RO systems have been credited assuming a 1:1 relationship between LRV of conductivity (typically 1.0 - 1.5) and LRV of virus (typically > 5.0). In your jurisdiction (State/Utility) would it be possible to claim at a ratio that is not 1:1. For example, if sulfate (or conductivity) could be shown to hold a 1:2 relationship with virus LRV, could a virus LRV of 2 be claimed provided sulfate or conductivity LRV always remained above 1?
   a. If the answer to the question above was no, what additional information or testing might demonstrate that a ratio approach could be used?

   The general answer to question 5 was yes. However, most responders stipulated a requirement for site specific demonstration of such a ratio during commissioning to ensure no site-specific interferences. In addition, peer reviewed robust data sets across multiple facilities would help to justify acceptance of such an approach.

   There were concerns that such an approach would be too complex and that regardless of evidence the ratio would not be accepted because of perceptions about resulting monitoring complexity. Grandfathering a relationship from alternate sites without site specific demonstration was also noted as a barrier.

   In addition, testing suggests that membrane element integrity and sealing of the membrane into the vessel plays an important role in the ability of the RO train to achieve virus removal. In the case of a sealing defect, a significant breach would be measurable by conductivity and would likely pass larger pathogens.

6. Please add any additional feedback or questions to the project team here.

   The final question was proposed in an effort to capture more general concerns or needs for information.

   The project was seen as a means to help regulators credit RO systems. There was a preference that any framework for crediting RO should fit within a LT2ESWTR compliance. There were concerns that the crediting approach may change depending on the source water Bin. In other words, assuming the water you are treating is Bin 4 - would the USEPA have a problem with the surrogate method?

   Interest was noted in a protocol for validation testing using the proposed new techniques/surrogates such that an independent laboratory (e.g., NSF, UL, WQA) could use to develop a validation process for RO and NF. A membrane that has undergone validation testing by an independent third-party laboratory was considered easier to approve (from the point of view of State regulators in a small state). It would also help if there was a checklist developed for regulators to assist in evaluating validation studies that use the proposed new techniques/surrogates; similar to that available in the MFGM or what was

done in WRF project 4376 dealing with UV disinfection or in EPA publication EPA/600/R-20/094 from 2020 for innovative approaches for UV validation.

Some concern was raised about crediting approaches for RO systems with recycle streams. Newer high recovery cyclic RO processes where permeate concentration changes throughout a cycle (i.e., CCRO or PFRO) would potentially require different sampling protocols if the surrogates above were suitable.

9.3 Proposed Approach for Crediting RO Membranes

There are a number of methods to verify the virus removal performance of RO trains and these have been explored in a number of research programs. The approaches proposed below are limited to the surrogates evaluated and their sensitivity demonstrated within this report. In addition, recommendations are made with respect to monitoring to more closely adhere to the frequency requirements and paradigms established for UF in the MFGM.

As noted in the survey responses, there is no formal pre-validation testing for RO membranes, but such testing could provide a benefit for regulators when considering approval of an RO unit. A number of MF/UF products are not used in drinking water systems unless they are independently certified according to NSF 419 for protozoa removal, established via microsphere testing. Some UF systems also report NSF 419 virus LRV results established via MS2 testing at maximum anticipated flows.

The MFGM requires product certification and non-destructive performance testing (NDPT) with an associated Quality Control Release Value (QCRV) as part of the membrane manufacturing process. Individual element testing for salt passage is routinely performed by many but not all membrane element suppliers. When higher LRV's are being sought, individual element testing is strongly recommended.

Under the current regulatory framework of the MFGM, product certification (NSF 419 or equivalent) is required for membrane products seeking to obtain 3-log removal or higher *Cryptosporidium* credit. To date in their application in potable reuse, RO membranes have been awarded 2-log removal which is less than 3-log requirements contained in the MFGM. The applicability of NF and RO as a means to achieve virus removal was briefly mentioned in Appendix E but did not detail the techniques or approach such as vessel conductivity profiling described herein.

It is anticipated that the MS2 challenge test results will be high, likely exceeding 5 or even 6-log. However, verification of a product lines performance would provide useful information and an administrative control on the quality and potential formulation variations which might occur for typical RO systems without such a requirement. In other research it has been noted that there is presently no commercial incentive for suppliers of brackish water RO membranes (commonly used in potable reuse) to assure that the membranes don't contain defects to an extent that could reduce LRV below 4-log as it is difficult to verify such removal at scale with currently available surrogates and anticipated salt passage due to diffusion (Trussell et al., 2020). Independent RO product challenge testing, like that performed by the NSF for MF/UF

membranes, could be incorporated into a RO crediting framework. This testing could help as an administrative control to ensure a minimum expected level RO membrane integrity.

Uranine is not included in the approaches proposed below due to the practical difficulties consistently experienced with its storage, feed, and analysis. However, uranine was more sensitive when profiled than other surrogates and can be relatively easily measured onsite. As such, periodic profiling of uranine or other low molecular weight fluorophores may be useful for periodic confirmation of array integrity.

9.3.1 Tier 1 RO Crediting

Tier 1 proposes simply assuming a 1:1 pathogen relationship between direct conductivity or TOC LRV and pathogen LRV (virus, *Cryptosporidium*, or *Giardia*).

As noted in the hypothetical example above, significant changes to baseline conductivity are anticipated before the probable pathogen LRV reduces below 1-log (See Figure 9-1).

Tier 1 credits are anticipated to be in the 1.0 to 2.0-log unit range based on conductivity or TOC depending on the feedwater concentration and operational conditions of the RO system.

Tier 1 has the advantage of being continuously measured online with robust and cost-effective conductivity probes which will lead to a simple approach with minimal investment or complexity.

Tier 1 is generally accepted in most (but not all) US states as a means to verify a conservative RO removal credit.

9.3.2 Tier 2 RO Crediting - Use of Surrogates

Tier 2 assumes a 1:1 pathogen relationship between direct LRV of surrogates measured in the RO feed and combined permeate of each array and pathogen LRV (virus, *Cryptosporidium,* or *Giardia*).

As noted in the hypothetical example above, significant changes to a surrogate are anticipated before the probable pathogen LRV reduces below 2.0-log (i.e., Tier 1, see Figure 9-1).

Diffusion, feedwater abundance, and permeate detection limits will likely limit the maximum LRV able to be demonstrated by surrogates to 3.0-3.2 log range depending on the method used. Either sulfate or strontium are recommended based upon this report. Phosphate may also be promising based upon the WCWA results. Prior to selection of a surrogate, it is recommended that the proposed source water is characterized, with grab sample data, to determine the surrogate concentration relative to the analytical detection limit is confirmed to be acceptable. It should be noted that the sensitivity of the marker test may be improved by permeate monitoring of the individual stages due to the feedwater concentration effects and combining of permeate.

To meet the requirement as a DIT per the USEPA MFGM, surrogates would have to be measured daily from each RO array. The results and verified LRVs would also need to be calculated and acted upon within each 24-hour period. Given the level of sampling and the

response time needed, application of Tier 2 would entail enhancement of in-house laboratories with the equipment and experience to process multiple samples on a routine basis. Alternatively, online instrumentation could be integrated into the arrays (in a manner similar to automated conductivity profiling). This would provide more uniform and efficient coverage. The limit of detection of the instrumentation would need to be sufficient to verify the desired LRV. Also, different online instrumentation may be needed to account for the 3 orders of magnitude difference expected in the RO feed compared to the RO permeate. However, the expense of such instruments, which exist for strontium and sulfate, may be a challenge for some utilities.

Tier 2 credits are anticipated to be in the 2.0 to 3.0-log unit range based on surrogate monitoring depending on the feedwater concentration, permeate detection limit and operational conditions of the RO system.

Monitoring of permeate conductivity and ideally feed to permeate direct LRV would be required to serve as a continuous indirect integrity monitor and allow for corrective actions in the event of gross membrane integrity failures. Corrective actions are expected to include diversion and expedited sampling of surrogates on suspect arrays to confirm that LRV is adequate prior to returning the unit to service.

9.3.3 Tier 3 RO Crediting - Conductivity Profiling

Tier 3 maintains the 1:1 pathogen relationship between LRV calculated from conductivity profiling each individual vessel in each array and pathogen LRV (*virus*, *Cryptosporidium*, or *Giardia*).

The maximum LRV able to be claimed would be 5.0-log due to the cap inherent in the conductivity profiling calculation method described in Section 4.3 of this report. In practice, the minimum LRV able to be claimed will be related to the tolerance for deviations from median conductivity. At the level of 1.3*median suggested within this report, it is anticipated that LRVs of more than 3.5-log should be able to be verified by conductivity profiling.

If alarm tolerance is increased, then a theoretical model accounting for the maximum number of off specification vessels and the magnitude of off specification performance should be calculated to specify a new minimum credit.

Although daily conductivity profiling could be performed manually every day for each array – meeting the daily DIT requirement, automating the sampling process would allow for the analysis to occur routinely without overburdening operations staff. Automation of conductivity profiling is anticipated to lead to results that are more reproducible and can be directly incorporated into plant SCADA systems. This will allow for automated and consistent LRV calculations, ease of reporting, and timely alarms on performance outliers.

Conductivity profiling compares the relative difference of vessels under the same operational conditions, including diffusion rates, feedwater abundance and operational conditions. This is advantageous as a monitoring technique as of the seasonal variation of these uncontrolled variables is cancelled out prior to calculation of an operating LRV.

This conductivity profiling approach was not tested for NF systems and further research and evaluation of full-scale NF arrays may help to further understand if the passage of monovalent ions reduces the sensitivity of conductivity profiling for NF.

The calculation model for conductivity profiling requires a sufficient number of arrays to calculate a median and average. At least six vessels per stage is recommended to adequately calculate these metrics. For a system with 8-inch elements, this may mean that conductivity profiling is only able to be applied for 3 stage arrays with a capacity close to 1 mgd. Smaller total array flows could be verified for 2 stage arrays or for 4-inch element RO systems.

It is recommended that the minimum sensitivity of conductivity profiling is confirmed by the use of periodic checks with a Tier 2 surrogate (either for each stage or the system as a whole). A monthly frequency should be considered sufficient to confirm that each array is indeed achieving near or above a LRV of 3-log. Establishing long term trends of conductivity profiling results between different arrays at the same site could be used to further verify that subtle integrity failure is not occurring. Grab sampling of the RO feed and combined array permeate is considered sufficient for the purpose of verification sampling.

Tier 3 credits are anticipated to be in the 3.0 to 5.0-log range based on conductivity profiling monitoring depending on the alarm limits set and maintained across the array (i.e., distance from the median permitted).

Monitoring of permeate conductivity as well as feed to permeate direct LRV (i.e., Tier 1) would be required to serve as a continuous indirect integrity monitor and allow for corrective actions in the event of gross membrane integrity failures. Corrective actions are expected to include diversion and expedited conductivity profiling and/or surrogate sampling of suspect arrays to confirm that LRVs are achieved as the system was intended to operate (and permitted) prior to returning the unit to service. Table 9-3 summarizes the three tiers for RO crediting.

Table 9-3. Summary of Options for Crediting RO.

Parameter	Tier 1	Tier 2	Tier 3
Validation Type	1:1 pathogen relationship between direct conductivity LRV (Combined Feed to Combined Permeate) and pathogen LRV.	LRV of surrogates (e.g., SO_4^{2-}, Sr) measured in RO Feed and Combined Permeate of each array adopted for pathogen LRV.	LRV calculated from conductivity profiling each vessel in each array adopted for pathogen LRV.
Anticipated LRV Range	1.0 – 2.0	2.0 – 3.0	3.0 – 5.0
Requirements	Continuous online measurements of Conductivity. Optional online monitoring of TOC LRV for the purpose of enhancing credit in low conductivity waters.	Enhancement of in-house laboratories and/or addition of complex online instrumentation for RO arrays. Must verify surrogate LRV daily for each array. Indirect continuous conductivity monitoring to detect and respond to gross failures.	Daily conductivity profiling every day for each array. Indirect continuous conductivity monitoring to detect and respond to gross failures. Periodic checks with Tier 2 surrogates recommended as a secondary verification. May not be suitable for NF or for very small systems (<<1 mgd)

CHAPTER 10

Conclusions and Recommendations

Information and test results for the integrity of RO and NF systems were developed in this study. Although the regulatory compliance methodology has focused on RO systems and the reduction of specific parameters from feed to permeate, characterization of performance within the unit itself by conductivity profiling was investigated to provide a better understanding as how to identify and characterize the location of potential integrity breaches that would be of regulatory significance.

Findings for the measurement of overall removal performance (feed to permeate) can be summarized in the following points:

- At a system level, LRVs are attained for regulatory compliance as RO feed to combined permeate.
- The use of sulfate as a surrogate indicator (nominally 2.8-log) appears to allow for higher LRVs compared to EC due to EC (nominally 1.3-1.5-log) encompassing ions that have lower rejection rates than sulfate. As evidence from data available with operating facilities, sulfate removal is higher than TOC removal. Thus, higher LRVs are obtainable if sulfate is used as a surrogate indicator.
- Fluorescent compounds, such as uranine, provide a higher level of demonstrated removal (nominally 3.5-log). From a practical perspective, the use of fluorescent compounds poses operational challenges due to the burden of feeding a non-indigenous surrogate.

Findings of the testing as related to the underlying principles associated with the mechanisms that are associated with integrity are as follows:

- The hypothesis that the membrane itself is not a source of virus passage appears to be consistent with the testing data collected in this study. As described in the introduction, the composite structure (polyamide membrane on top of an ultrafiltration membrane) used for spiral wound NF/RO elements provides a multiple barrier approach for removal. Results of testing suggest the integrity issues arise from compromised sealing within the element or external O-rings. These types of issues would pass a significant amount of conductivity that could be pro-actively identified in operation with appropriate membrane element integrity testing methods.
- The salt rejection of the membrane element used should not affect virus reduction. It can be inferred from the testing of oxidized RO elements as well as NF elements that integrity for virus removal is not a function of the salt rejection characteristics of the membrane.
- Furthermore, membrane cleaning, which temporarily changes conductivity removal performance does not appear to substantially change virus removal.
- It is noted that the membranes tested at the YVWD were approximately seven years old. The membranes tested at WCWA were five to six years old, which is at least half the typical design life of 8-10 years. Even though these membranes have been continually operated for

a significant portion of their life, loss of virus and surrogate rejection through membrane aging and cleaning does not appear to be substantial.
- Natural variation in permeate conductivity is caused by temperature increases which drive higher diffusion of ions across the membrane. This is a phenomenon associated with the removal of all ions.

Findings for the subsequent identification of the location of potential integrity breaches are as follows:

- Based upon the results of the testing campaign, it appears that there is a correlation between high permeate conductivity vessels (outliers) and virus passage demonstrating that conductivity profiling is a useful diagnostic tool to determine potential integrity issues. It is logical to further investigate those vessels that are associated with the highest permeate conductivity as potential integrity issues.
- Sulfate, uranine, or other surrogate indicators of larger molecular weight can also be used for profiling.; however, their implementation is more involved and costly and should be reserved for non-routine monitoring purposes.
- Data seems to be consistent with the literature that the defects (i.e., integrity compromises) have to be significant in size to drastically lower the LRV.
- Historically used practices such as vessel probing and vacuum testing are useful and effective tools in the identification of membrane elements with questionable integrity.

An approach where different RO credit can be sought based upon choice of surrogate was proposed, that is:

- Tier 1 - an LRV of 1.0 - 2.0-log can be obtained based on verifying the equivalent direct LRV of conductivity:
  - This approach is conservative with respect to the actual level of virus removal anticipated across a RO system.
- Tier 2 - an LRV of 2.0 - 3.0-log can be obtained based on daily measurement and analysis of results of surrogates such as sulfate, strontium, or phosphate across an RO array:
  - Limits for conductivity reduction or absolute permeate conductivity would need to be established as a continuous indirect integrity monitor performance between surrogate sampling events.
  - Daily measurement and analysis of results will either require grab sampling and an in-house lab or investment in multiplexing online surrogate meters that have sufficiently low detection limits to demonstrate the proposed LRV.
- Tier 3 - an LRV of 3.0 - 5.0-log can be obtained based on the LRV calculated from conductivity profiling:
  - Limits for conductivity reduction or absolute permeate conductivity would need to be established as a continuous indirect integrity monitor to verify performance between conductivity profiles.
  - Use of an automated profiler capable of testing all operating skids at least once per day.

- Periodic (i.e., monthly) grab samples of a selected surrogate are recommended to ensure that LRV always remains above 3.0.
- Further research may be needed on interpretation of conductivity profile results and conversion to an LRV for full scale nanofiltration arrays. At this time, and with available information, Tier 3 is proposed to only apply to RO membranes.

Although it would be a challenging undertaking, an approach to achieve widespread acceptance of a framework for RO crediting would be to revise and amend the USEPA MFGM. Amendments could include details on new approaches to verify virus removal of RO and clarification on requirements that have potentially been narrowly interpreted as a basis to avoid granting RO credit. Tier 1 testing falls below the 3-log *Cryptosporidium* removal requirement contained in the MFGM but is consistent with a treatment technique or continuous indirect integrity monitoring credit. Tier 2 is consistent with the MFGM description for marker-based testing. Tier 3 testing is a suggested approach that was developed specifically for the application of RO and will require regulatory concurrence. The MFGM allows for the adaptation and use of alternative DIT methods provided that they satisfy the criteria of resolution, sensitivity, and frequency. Sensitivity of conductivity testing can be improved/adapted for use with RO by the subtraction of naturally occurring diffusion and by individual vessel sampling to identify outliers. As noted previously, product certification and non-destructive performance testing are elements of the MFGM approach that should be incorporated into the Tier 3 compliance strategy.

The necessity to implement potable reuse projects continues to become more prevalent as the availability of water resources diminishes. Establishing pragmatic RO and NF integrity monitoring techniques with a sensitivity that correlates and is equivalent to the results from MS2 challenge testing could play a vital role in supporting the implementation of potable reuse. This project has identified and proposes a tiered approach for surrogates that can achieve 3 – 5-log LRV, which would be an improvement over the current (and inconsistent) implementation of pathogen credits for high pressure membranes which are fundamental advanced treatment processes.

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