Proposed Methodology for Determining Reverse Osmosis Unit Integrity for Pathogen Removal Credit

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It is intuitive to suggest that reverse osmosis (RO), which is used for the removal of dissolved solutes (salts), can also be used to achieve the removal of virus, *Giardia*, and *Cryptosporidium*. The challenge is that the use of conductivity for water quality monitoring of the RO process lacks sufficient sensitivity, and a direct integrity test that translates water quality performance to regulatory compliance for pathogen removal has not been established or accepted as a practice. The basis of this article is as follows: It is proposed for a productionlevel (e.g., 1 mgd or larger) RO system that the sensitivity can be increased by using the results of a conductivity profile, which is an existing diagnostic tool used to identify integrity defects within an RO unit. The proposed direct integrity test methodology uses the results of a conductivity profile to (1) determine that RO unit integrity exists within statistical limits, (2) isolate and differentiate between the conductivity associated with diffusion and with a defect, and (3) calculate the log removal value (LRV) that would be associated with an RO membrane defect. The resulting calculation approach significantly increases the sensitivity of the LRV calculation and is supported by full-scale testing data using MS2 coliphage as the challenge organism.

Keywords: log removal value, pathogen, regulatory, reverse osmosis

From a historical regulatory perspective, the efficacy of the reverse osmosis (RO) process was simply assumed as an effective barrier for Giardia and virus in the Surface Water Treatment Rule (AWWA 1991). Publication of the Membrane Filtration Guidance Manual (MFGM) (USEPA 2005) formalized direct integrity testing concepts and facilitated the implementation of membrane processes for drinking water and recycled water facilities. Although the MFGM was applicable to any type of membrane filter and specifically written for Cryptosporidium compliance for systems that would require 3-log or greater removal, drinking water facilities that used hollow fiber microfiltration and ultrafiltration processes with pressure-based integrity testing benefited from the guidance provided. As a result of the MFGM, regulatory authorities have linked direct integrity testing to log removal value (LRV) credits for all types of membrane processes and the other regulated contaminants (virus, Giardia).

Recent activity in the indirect potable reuse field and the direct potable reuse initiative will include membrane processes designed to provide pathogen removal. California has implemented a 12-10-10 log removal

requirement for virus, Giardia, and Cryptosporidium for proposed groundwater recharge projects (California Code of Regulations 60320.208 2014). Regulations that specify a specific treatment technique (i.e., RO) and establish log removal for treatment trains are being implemented. Guidelines provided by various recycled water organizations suggest LRVs of 1.5 for virus, Giardia, and Cryptosporidium because of the lack of a direct integrity testing methodology for RO systems (WateReuse Research Foundation 2015). Actual fullscale data of pathogen removal by RO is limited (Kitis et al. 2003, Adham et al. 1998). However, the results of more recent pilot studies suggest higher removal typically greater than 4 log. Virus challenges with an oxidized RO membrane suggest that removal of viruses is possible, and defects in the form of O-ring failure are most likely the mechanism of significant integrity loss (Jacangelo & Gray 2015). Thus, there is a specific need to address the RO process in the context of regulatory compliance for pathogen removal.

The ability of an RO unit to act as a pathogen barrier has been the subject of recent work, particularly in the area of water recycling, where compliance with existing and new regulatory concepts for pathogen removal is required. Much of the research to date has focused on low-molecular-weight fluorescence indicator compounds, which can be used as part of a spiked challenge or a continuous monitoring technique (Surawanvijit et al. 2015, WateReuse Research Foundation 2014). Some have used specific constituent(s), with periodic sulfate or fluorescence monitoring being the most common surrogate for microbial removal. Concentrationbased methods (including conductivity) have the same underlying issue; they are far smaller than viruses, and the sensitivity of the measurement generally is less than what would be observed through a challenge test. A more practical issue with using indicators other than conductivity is associated with the specialized equipment and analytical instrumentation necessary to perform the analysis, whereas a conductivity profile can be obtained in typically 15-30 min using common and inexpensive equipment.

In section 4.3.1.3 of the MFGM, there is a framework for establishing removal credits for membranes using a direct integrity test (USEPA 2005). Within the framework for air pressure-based direct integrity testing, there is language that recognizes air diffusion as an intrinsic property of a microfiltration or ultrafiltration membrane and that results of integrity testing may be adjusted to account for the air diffusion across the membrane (USEPA 2005). The net effect is that the inclusion of the diffusion component increases the sensitivity of the direct integrity test.

In appendix A of the MFGM, there is a discussion regarding the application of an integrity verification program (IVP) for a membrane system (USEPA 2005). Key objectives of an IVP are as follows:

- Verifying integrity on an ongoing basis
- Identifying and correcting any integrity problems
- Recording and analyzing integrity test data
- Preparing any compliance reporting
- An IVP includes the following key elements:
- Direct integrity testing
- Continuous indirect integrity monitoring
- Diagnostic testing
- Membrane repair and replacement
- Data collection and analysis
- Reporting

When an RO system is considered, the MFGM provides general guidelines for the development of the IVP. RO is an established process, with the fundamental concepts developed in the 1960s and refined over the past 50 years.

It should be noted that the MFGM contains additional language regarding the production of membranes for *Cryptosporidium* removal that require challenge testing and the development of a nondestructive performance test for validation of the product in order to determine the maximum LRV of manufactured products.

BACKGROUND

Historically, RO systems have been used to achieve removal of dissolved salts and other dissolved constituents in water. The mechanism of removal is typically described using the solution-diffusion model (Lonsdale et al. 1965). An RO membrane is most commonly made by casting a thin polymeric film onto an underlying ultrafiltration membrane and nonwoven backing material that supports the membrane. RO membranes do not have pores in the conventional sense, as solutes pass through interstitial spaces between molecular structures based on molecular charge and size, which are controlled by principles of diffusion. Pathogen removal by size exclusion is theoretically infinite; however, differences in manufacturing may limit the overall removal performance of the RO membrane element.

To establish the LRV of an RO unit, it is necessary to understand the underlying principles of RO. RO is a pressure-driven process by which water passes from the feed to permeate side under pressure. Net driving pressure is the parameter that controls the passage of water from feed to concentrate (Porter & Sudak 1990). Solutes (dissolved ions and organic compounds) diffuse from the feed to permeate based on a concentration difference and the mass transport coefficient that is based on the specific constituent's mass, structure, and ionic charge, as well as interaction with the membrane and the water temperature (Lonsdale et al. 1972). Dissolved gases simply pass through the membrane (AWWA 2007).

Conductivity is a measure of the electrical property of water proportional to the electrical charge (valence) of the ion. At elevated concentrations, the relationship between conductivity and concentration can become nonlinear in nature as the excess ions in solution can interfere with the linearity of the conductivity measurement (Miller et al. 1986). Typically, RO removes 94–99% (1.2–2.0 log) of the conductivity or total dissolved solids (TDS) from a water supply, and the removal is dependent on the membrane used, array configuration, operating conditions, and water temperature.

RO membranes are characterized by their salt rejection characteristics and generally tested for rejection during the manufacturing process using a known solute such as sodium chloride (AWWA 2007). Wet test data can be normalized to a standard set of conditions established by the manufacturer, and software projection programs can be used to determine the removal of other constituents for a specific membrane (AWWA 2007).

In an operating RO system, flow, pressure, conductivity, and temperature are normally monitored. Diffusion across the membrane is based on a number of design and operating variables, and data normalization is used to approximate performance at a set of standard conditions. Parameters such as percent rejection, salt passage, normalized permeate conductivity, normalized salt rejection, and/or normalized permeate flow are used to monitor system performance and account for variations in feedwater quality and temperature changes (ASTM International 2010).

A significant increase in permeate conductivity can signal the loss of water quality resulting from the failure of a membrane module or O-ring. Normalized data may be used to determine whether the breach was associated with a gross change in feedwater quality or operational event, such as membrane cleaning or failure of an O-ring. However, for small defects, the incremental increase in conductivity in the bulk permeate is not significant and can be obscured by the normal variations in operating conditions. Routine conductivity profiling of the pressure vessels is used to identify individual vessels with higher conductivity that may be associated with an integrity defect.

R0 systems. For drinking and recycled water systems, the typical RO unit contains a series of pressure vessels that can be arranged in a variety of configurations. Vessels are arranged in parallel or series fashion in order to satisfy the production requirements. This is commonly referred to as the array with a nomenclature 78:48:24, where 78 represents the number of vessels in the first stage, 48 represents the number of vessels in the second stage, and 24 represents the number of vessels in the third stage. Each pressure vessel contains multiple elements, but for the overwhelming majority of systems, the number of elements is somewhere between four and eight, with six and seven element configurations being the most popular.

In a typical staged array, the feedwater is concentrated as permeate is produced. This directly affects the permeate quality as water quality from the first stage has the lowest concentration, while the third stage has the highest. A mass balance can be developed for the removal across the individual stage or the unit as a whole. Figure 1 illustrates the mass balance around a typical three-stage RO system.

Most RO systems are instrumented such that the flow, pressure, and conductivity are measured and/or can be calculated through addition and subtraction.

Each pressure vessel that operates in parallel should have permeate flow and water quality that is comparative in conductivity. Individual pressure vessels have a permeate sample port that is used to periodically monitor the conductivity of permeate from each vessel and to assist in the identification of conductivity excursions or defects in integrity. Figure 2 shows a permeate sampling location. It is a common practice of RO operation to periodically monitor the conductivity from each pressure vessel as a form of diagnostic testing in order to confirm that the vessels are performing similarly (AWWA 2007).

In the event of a question regarding the integrity of a full-scale unit, the first step would be to obtain a conductivity profile to identify the most probable location of the defect.

Vessel probing is another diagnostic tool used by membrane operators to determine the performance of each membrane element if the conductivity from a vessel is questionable (AWWA 2007). In this method, a sample line is introduced into the permeate carrier tube and indexed along the length of the RO pressure vessel. Normally, the length of each index is 20 in., the length needed to identify conductivity associated with the RO membrane and the interconnectors between membrane elements. This approach is used to locate single elements that may be defective or to identify O-ring failures. Some utilities prefer to remove and individually test RO elements to identify problems.

To summarize, in an RO system of a given staged arrangement, flow and water quality (conductivity) for





each stage is monitored, and performance can be calculated and/or normalized to a set of standard conditions. Permeate conductivity from individual pressure vessels or membrane elements can also be used to isolate performance and initiate corrective measures. Thus, three of the criteria of an IVP—continuous indirect integrity monitoring, diagnostic testing and membrane repair, and replacement practices—exist within existing plant operations.

US Environmental Protection Agency (USEPA) methodology for determining LRVs. An IVP requires a periodic direct integrity test. The MFGM established criteria for three different parameters: resolution, sensitivity, and frequency, to qualify as a direct integrity test (USEPA 2005).

 Resolution is defined as the size of the smallest integrity breach that contributes to a response from a direct integrity test. Any direct integrity test that is applied to meet the requirements of USEPA's Long Term 2 Enhanced Surface Water Treatment Rule is required to have a resolution of 3μ or less for *Cryptosporidium* removal.

- Sensitivity is the maximum LRV that can be reliably verified with the direct integrity test.
- Frequency indicates that a direct integrity test be conducted on each membrane unit at least once each day that the membrane unit is in operation for rule compliance, unless the state approves less frequent testing. The rule also has requirements for the establishment

of lower and upper control limits (USEPA 2005).

METHODOLOGY

For the purposes of illustration, Figure 3 shows an example of a typical three-stage RO unit.

The system includes the following:

- Combined permeate flow (Q_p)
- Flow measurement for stage 2 and stage 3 permeate (Q₂, Q₃)
- Individual stage permeate conductivity (C_1, C_2, C_3)
- Combined permeate and concentrate conductivity analyzers (*C*_p, *C*_c [not used])
- Feed conductivity (C_f)
- Concentrate flow (*Q*_c)

The MFGM provides a methodology for the determination of removal for a marker-based test based on the difference from sampling of combined feed to permeate in section 4.3.2. A volumetric concentration factor (VCF) is not required for a marker-based test, as the reference point is the feed by definition.

Step 1: Validate integrity through conductivity profiling and statistical analysis. In this step, it is necessary to determine whether all RO vessels are producing water of acceptable quality. The conductivity profile can be used to obtain information that validates the performance of the RO unit as a whole. Periodic routine monitoring is important to the overall implementation, as membrane performance changes over time as a result of



water quality, temperature, and membrane fouling/ cleaning. Thus, it is important to have a baseline and/or historical reference to assess the overall performance of the system. Conductivity samples are taken to establish that vessels are performing similarly, with no defective membranes or O-ring seals. Operators perform conductivity profiling to identify vessels that have high conductivity, commonly called outliers, through simple observation of data without statistical means. However, the data obtained should also fit within a statistical distribution. Membranes that are performing outside of acceptable limits can be identified through statistical analysis, which formalizes the operational practice.

A handwritten conductivity profile from the Orange County Water District (OCWD), Fountain Valley, Calif., with membranes located in a 78:48:24 array, which had been in service for approximately eight years, is shown in Figure 4.

The conductivity profile includes overall unit performance and the operating information. The data indicate the basic elements of RO operation—that membranes within a stage perform similarly; that there is an increase in the average permeate conductivity from the first to third stage; and that despite the membrane age, the data were remarkably consistent. The data were entered in a spreadsheet to facilitate further examination.

The data set fostered thought about the basic assumptions regarding membrane integrity and whether data of

this type could be used as the basis for a direct integrity test that would satisfy regulatory requirements. The unique aspect of this approach, and one that was not contemplated in the development of the MFGM, is that the overall performance is being characterized by the individual sampling of permeate quality at multiple locations-i.e., a diagnostic test with a variety of numeric results and not the result of a single unit test, and subsequent visual observation. Figure 5 illustrates the functional difference between the air pressure-based pressure decay test that is performed on a group of membrane modules and the marker-based conductivity profile that uses the individual pressure vessels. One observation about the difference is that information regarding individual module integrity in a pressure decay test is obscured, while the larger data are associated with the vessel performance conductivity profile, which provides a basis for comparison and provides additional context.

Questionable performance is associated with vessels that have high conductivity, commonly called outliers. Conceptually, the performance of an integral stage should fit a statistical distribution; thus, statistical tests within a spreadsheet could be used 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 OCWD, the following approaches were developed and appear to correlate to the operational practice to statistical methods.

IGURE 4	Normal v	essel cor	nductivi	ty profile	e with o	perating c	lata					
						Stage 1						
25	24	26	25	20	31	31	21	25	29	24	30	27 Avera
23	22	30	25	22	29	25	22	26	23	26	21	21 24.5
24	20	20	27	21	21	23	26	24	24	24	26	25
22	26	24	26	24	22	19	23	25	27	30	24	23
24	25	25	27	22	25	22	23	32	22	25	24	22
22	28	25	27	23	22	28	27	27	24	28	21	22
			Stage 2									
71	53	73	57	71	56	85	75	Average				
58	47	75	87	65	67	65	47	63.4				
66	72	79	80	65	69	52	66					
54	61	80	76	57	58	51	56					
56	56	55	60	57	60	48	58					
67	60	57	72	67	74	51	53					
	Stage 3											
302	319	260	250	Average								
274	318	272	205	275								
313	300	216	215									
315	338	253	252									
291	351	213	230									
307	306	255	241									
rain Calculatio	ns											
arameter		Units	6	Stage 1	Stage 2	Stage 3						
eed Flow		gpm		4085	2045	926						
Conc Flow		gpm	1	2045	926	613						
Permeate Flow		gpm		2040	1119	313						
eed Conductiv	ity	µS/cr	n	1764								
factor		mg/L/µS	S/cm	0.55								
eed TDS		mg/L	-	970	1923	4201						
Peremate Cond		µS/cr	n	24.6	63.4	275						
Kfactor		mg/L/µS	S/cm	0.6	0.6	0.6						
ermeate TDS		mg/L	-	14.7	38	165						



- One approach is to apply the central limit theorem and the "three-sigma rule" (i.e., in any normal distribution of data, 99.73% of the samples should fall between three standard deviations of the mean) regarding the sample in order to test the validity of the population (Kazmier 2003). The central limit theorem is valid for an RO unit, as there are differences in membrane flow and salt rejection properties that yield small differences in permeate conductivity (and flow) 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. This type of test is commonly applied to situations such as a new membrane installation, which may have multiple questionable vessels. This concept may be more applicable for use in smaller systems that have fewer pressure vessels available for conductivity monitoring by statistical analysis.
- 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 in which a defect may be present (Joanes & Gill 1998). In an RO system, outliers associated with defects will cause skew in the positive direction.

Figure 6 illustrates the characteristics of normal and outlier data groups. In a data set that exhibits a normal distribution, the following statements can be made:

- All data fit between -3σ and $+3\sigma$ standard deviations.
- The median of the data is the same as the average of the data.
- The skew is equal to zero; i.e., data above and below the median, or the average, are symmetrical.

The following statements are associated with outlier data groups:

- There are data that exceed $+3\sigma$ standard deviations (outliers).
- The average of the data becomes greater than the median.
- There is a positive skew in the data.

Table 1 provides a summary of the statistical analysis for a conductivity profile. Conditional formatting (green) has been applied to indicate that the results from profiling are less than statistical limits or that the test was successful.

Figure 7 illustrates the change in stage performance with a single outlier value (in yellow) in each stage of the RO unit and includes the revised statistical parameters described earlier.

Table 2 provides the revised statistical calculations, which indicate that a single vessel in each stage operating with higher conductivity does not pass the three



tests (three-sigma, median + 50%, skew) proposed as integrity indicators. In this case, the conditional formatting (red) indicates that the maximum profiling test results are higher than the acceptable statistical test result.

Statistical tests, such as the ones described, can be used to identify integral as well as nonintegral membrane systems. It is noted that the relative change from a "pass" to "fail" result in the first stage was a change in one of 78 vessels from a conductivity value of 32 to 40. Thus, the periodic monitoring of vessel permeate conductivity profiles with subsequent statistical tests can be used to validate the integrity of a stage within an RO system. Subsequently, if all stages are determined to be integral, the corresponding unit would be stated to be integral.

Step 2: Calculate the LRV associated with the defect (LRV $_{defect}$). Although the calculation of LRV that is

TABLE 1	of a typ	ical		
	Conductivity C	Calculatio	ns	
Pa	rameter	Stage 1	Stage 2	Stage 3
Average—µS/	'cm	24.55	63.4	275
Standard dev	iation—µS/cm	2.9	10.2	42.5
Median—µS/	cm	24.0	60.5	273.0
Maximum—	uS/cm	32	87	351
Average + 3 x	≺ standard— <i>µS/cm</i>	33	94	402
Median + 50	%—µS/cm	36	91	410
Skew		0.52	0.43	-0.05

Green formatting indicates that the results from profiling are less than statistical limits or that the test was successful.

associated with overall unit performance is not a requirement, the basic principles associated with the MFGM are explained in part 1, with modification to the approach described in part 2.

Part 1: Calculate the LRV for the RO unit. The LRV for an RO unit can be calculated in the following relationships for flow (*Q*, Eqs 1–4). Eq 5 can be used to calculate the mass balance around an RO unit, where *C* represents concentration of TDS (mg/L). The overall mass balance around an RO unit is provided in Eq 5.

$$Q_{\rm f} = Q_{\rm p} + Q_{\rm c} \tag{1}$$

$$Q_{1-2} = Q_p - Q_1 + Q_c \tag{2}$$

$$Q_{2-3} = Q_p - Q_2 - Q_3 + Q_c \tag{3}$$

$$Q_1 = Q_p - Q_2 - Q_3 \tag{4}$$

$$Q_{\rm f}C_{\rm f} = Q_{\rm p}C_{\rm p} + Q_{\rm c}C_{\rm c} \tag{5}$$

Conductivity (μ S/cm) may be used as a surrogate measurement for TDS (mg/L) using the following relationships:

$$C_{\rm f} = {\rm Cond}_f K_f \tag{6}$$

$$C_{P1} = \operatorname{Cond}_{P1} K_1 \tag{7}$$

$$C_{P2} = \operatorname{Cond}_{P2} K_2 \tag{8}$$

$$C_{P3} = \operatorname{Cond}_{P3} K_3 \tag{9}$$

where factors K_f , K_1 , K_2 , and K_3 are empirically measured and can be established for the system through water quality (anion and cation) measurement. Feed conductivity is used as the basis of the calculation to minimize discrepancies associated with the nonlinear property of the conductivity measurement in concentrated solutions. Permeate-related factors K_1 , K_2 , and K_3 and the overall permeate constant K_p are likely to be similar in magnitude and can be obtained from the

FIGURE 7 Example of an outlier conductivity profile

		,				Stage 1		,	,	,			
25	24	26	25	20	31	31	21	25	29	24	30	27	Average
23	22	30	25	22	29	25	22	26	23	26	21	21	24.62
24	20	20	27	21	21	23	26	22	24	24	26	25	
22	26	24	26	24	22	19	23	24	27	30	24	23	
24	25	25	27	22	25	22	23	40	22	25	24	22	
22	28	25	27	23	22	28	27	27	24	28	21	22	
			Stage	2									
71	53	73	57	71	56	85	75	Average					
58	47	75	87	65	67	65	47	64.4					
66	72	79	80	110	69	52	66						
54	61	80	76	57	58	51	56						
56	56	55	60	57	60	48	58						
67	60	57	72	67	74	51	53						
	Stage	3											
302	319	260	250	Average									
274	318	480	205	284									
313	300	216	215										
315	338	253	252										
291	351	213	230										
307	306	255	241										
Train Calcula	ations												
Parameter		Uni	its	Stage 1	Stage 2	Stage 3							
Feed Flow		gp	m	4085	2045	926							
Conc Flow		gp	m	2045	926	613							
Permeate F	low	gp	m	2040	1119	313							
Feed Cond		μS/	cm	1764									
Kfactor		mg/L/J	JS/cm	0.55									
Feed TDS		mg	/L	970									
Peremate C	ond	μS/	cm	24.6	64.4	284							
Kfactor		mg/L/J	JS/cm	0.6	0.6	0.6							
Permeate T	DS	mg	/L	14.8	38.6	170							
DS—total o	dissolved so	lids											
ellow highli	ighting deno	tes single o	outlier valu	ues in each	n stage of	the RO un	it.						

TABLE 2	Statistical	tests	for	outlier	scenario
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Parameter	Stage 1	Stage 2	Stage 3
Average—µS/cm	24.62	64.4	284
Standard deviation—µS/cm	3.3	12.2	59.6
Median—µS/cm	24.0	60.5	282.5
Maximum—µS/cm	40	110	480
Average + 3 × standard— μ S/cm	34	101	462
Median + 50%— <i>µS/cm</i>	36	91	424
Skew	1.54	1.24	1.43

water quality analysis of permeate, along with a measurement of conductivity.

$$C_P = \operatorname{Cond}_P K_P \tag{10}$$

From the MFGM, the calculation for the removal is provided by the following general expression and is based on concentration:

$$LRV = \log C_f - \log C_p \tag{11}$$

where the permeate concentration can be calculated by using the overall permeate conductivity or, more accurately, by using the flow-weighted average of the individual-stage permeate concentrations.

$$C_{p} = \frac{C_{1}Q_{1}}{Q_{p}} + \frac{C_{2}Q_{2}}{Q_{p}} + \frac{C_{3}Q_{3}}{Q_{p}}$$
(12)

Table 3 provides the calculation for the overall log removal using the flow weighted by stage method.

Part 2: Calculate the LRV_{defect} associated with the defect. The underlying approach is to isolate the conductivity associated with diffusion that is not of pathogenic concern from conductivity associated with a defect that would be associated with a pathogen.

In order to calculate the LRV that would be associated with a defect of pathogenic concern, the mass flow rate for the permeate $(Q_p C_p)$ can be rewritten in the following manner. It comprises two components—one related to the diffusion (expressed as subscript "diff") of conductivity associated with non-pathogenic salts (solutes) and the other related to the flow of untreated feed through a defect that would also pass salts and would be of regulatory concern.

$$Q_p C_p = Q_{diff} C_{diff} + Q_{defect} C_{defect}$$
(13)

This equation separates the overall mass flow rate into its component-related (diffusion and size exclusion) terms. To calculate the LRV, the dilution model approach that is described in the MFGM as equation 4.3 can be used. It is shown here as Eq 14, where the Q_{breach} from the MFGM is the same as Q_{defect} in Eq 13.

$$LRV_{DIT} = log\left(\frac{Q_{p}}{VCF \cdot Q_{defect}}\right)$$
(14)

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.

Eq 13 can be rewritten and solved for any given stage using the relationship that permeate flow is equal to the sum of diffusive flow and breach flow and would apply to the vessel outlier data points. Eq 15 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.

$$\sum_{i=1}^{n} Q_{\text{defect}n} = \frac{Q_{\text{p}n} C_{\text{p}n} - Q_{\text{p}n} C_{\text{diff}n}}{C_{\text{defect}n} - C_{\text{diff}n}}$$
(15)

The defect flow, Q_{defect} , can be calculated using the outlier vessel(s) and the feedwater conductivity for a given stage. Ultimately this method requires an approach regarding 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 +2 σ , could be used on the basis of the premise that RO membranes that would fit within the normal distribution would also be integral. A value of +3 σ is not believed to be appropriate, as all the data do 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 compared with the feed conductivity C_{defectn} .

In the dilution model, VCF, as discussed in Section 2.5 of the MFGM, is used to describe the increase in suspended solids when integrity is determined at the membrane. In the case of RO, the mass quantity (Q_{defect} C_{defect}) inherently accounts for the concentration effects, such that the VCF is equal to 1. Stated in a different manner, the reference point for a marker based test is the feed by definition, and the VCF term would not apply.

A simpler approximation is also possible. As previously stated, and using Figure 6 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 to provide an LRV calculation. The approach is logical as the conductivity that would be associated with an outlier defect $Q_{defect}C_{defect}$ (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. The net effect is that Eq 12 can be rewritten for the flow-weighted concentration by the stage that is associated with a defect.

$$C_{\text{defect}} = \frac{(C_{p1} - C_{\text{diff1}})Q_1}{Q_p} + \frac{(C_{p2} - C_{\text{diff2}})Q_2}{Q_p} + \frac{(C_{p3} - C_{\text{diff3}})Q_3}{Q_p}$$
(16)

Parameter	Symbol	Stage 1	Stage 2	Stage 3	Train
Feed TDS— <i>mg/L</i>	Cf				970
Log feed TDS	$\log C_{\rm f}$				2.99
Average permeate conductivity—µS/cm		24.6	63.4	275	59.6
Adjusted permeate TDS—mg/L	C_{p}	14.7	38.1	165	35.8
Log permeate TDS	$\log C$	1.17	1.58	2.22	1.40
LRV					1.59

TABLE 3	Initial I RV	calculation
		calculation

where the average stage values are used for C_{pn} , the median stage value is used for C_{diffn} , and *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, 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 LRV:

$$LRV_{defect} = \log C_{f} - \log C_{defect}$$
(17)

For this calculation, the median conductivity of the stage is being used as the basis for C_{diff} . 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 value 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_{diff} , as the resulting LRV would be infinity. In the subsequent example, train values are flow-weighted by stage.

In Table 4, there are no conditions in which the median is greater than the average; such a condition would result in the maximum LRV assignment for the stage. The author's experience from working with the model suggests that the method has a practical upper limit for sensitivity of 4.0 log, using the median conductivity approach. A higher LRV can be obtained using the individual vessel calculation. Comparing Table 3 with Table 4, the LRV of the RO unit increases from 1.59 to 3.19. The increase in LRV is a result of removing the median diffusion-related conductivity to calculate the defect-related conductivity.

FULL-SCALE FIELD VALIDATION OF THE APPROACH

The Yucaipa Valley Water District (YVWD), Yucaipa, Calif., operates an RO system for the reduction of TDS for its recycled water supply. A simplified schematic of the system is shown in Figure 8. Details associated with the RO system are provided in Table 5.

In August 2017, YVWD performed testing of its RO system using MS2 coliphage (ATCC 15597-B1) as a surrogate indicator for viruses. MS2 coliphage was dosed for a minimum of 10 min prior to sampling to allow for stabilization through the unit. Two tests were performed. The first test was an evaluation of MS2 removal using an integral system. The second test was performed using a defect (1/16 in.-diameter hole) drilled into the end adaptor that isolates the membrane feed from the membrane permeate as shown in Figure 9. The end adaptor was placed into the system at the permeate collector of pressure vessel 1-2-3. In operation, the interstage feed would pass into permeate (Figure 9).

Results of the conductivity profile with the associated statistical calculations are provided in Figure 10. The defect acted as an orifice and allowed untreated feed to pass into permeate at an amount significant enough to change the conductivity, resulting in a profile that indicated a compromised condition existed as indicated by the shading. The difference in individual vessel performance from the two sampling events is associated with the normal variation in feed conductivity, and the vessels move together as a group. It is noted that the compromised vessel induced a significant change in the skew of the data, 0.32 versus 4.95, even though the vessel conductivity was approximately doubled and similar to the stage conductivity associated with the second stage.

Table 6 suggests that the actual removal of MS2 coliphage is higher than the calculated LRV_{defect} values (5.38 versus 3.56) associated with the integral array. Thus, use of the median is viewed as a conservative approximation of the overall removal obtained. Under compromised conditions, the LRV associated with the individual pressure vessel 1-2-3 was reduced from 5.04 to 1.7. The interstage feed conductivity was measured and used to estimate the size associated with the

Parameter	Symbol	Stage 1	Stage 2	Stage 3	Train
Feed TDS—mg/L	Cf				970
Log feed TDS	$\log C_{\rm f}$				2.99
Average permeate conductivity—µS/cm		24.6	63.4	275	59.6
Average permeate TDS— <i>mg/L</i>		14.7	38.1	165	35.8
Median TDS— <i>mg/L</i>		14.4	36.3	163.8	34.9
Adjusted permeate TDS—mg/L		0.33	1.76	1.1	
Log permeate TDS— <i>mg/L</i>	C_{defect}	-0.48	0.25	0.0	-0.20
LRV (defect)	LRV _{defect}				3.19

TABLE 4 Calculation of LRV_{defect} using median-stage TDS

defect and was calculated at 0.483 gpm. The resulting bypass would lower the LRV to 3.53 using the individual vessel approach. Actual unit removal under compromised conditions was measured at 4.16 and was higher than the calculated value of 3.09. Thus, the data suggest that a defect that is associated with an outlier conductivity increase within a vessel has a measurable response in terms of actual performance and the calculated LRV.

CONSIDERATIONS IN THE USE OF THE METHODOLOGY

In operation, there are various issues that may affect the LRV calculation. The following list provides examples of where erroneous results may be obtained.

- Changes in feedwater conductivity: For some systems, feedwater conductivity can be variable. Although the calculation of percent removal addresses conductivity changes, the underlying assumption that the water quality matrix increases or decreases with conductivity—resulting in K_f , K_1 , K_2 , and K_3 being a constant—may not be true under all circumstances.
- Location of integrity breach: A typical pressure vessel contains six to eight RO elements. If the integrity breach is located toward the feed end of the stage, the feed conductivity is lower than the average, and conversely, if the breach is located near the concentrate, the conductivity will be higher than anticipated. In some systems, the location of the permeate sample (i.e., feed or concentrate end) may make identification of an integrity breach more difficult as permeate is collected from multiple elements. This may necessitate individual element testing.

TABLE 5Yucaipa Valley Water District RO system
design parameters

Parameter	Value
Design permeate capacity—gpm	1,650
Design recovery—%	85
Design flux—gfd	11.8
Number of stages	2
Interstage pump/energy recovery device	Stages 1–2
Array configuration	52:20
Elements per pressure vessel	7
Area per RO membrane element— ft^2	400
Membrane element	Polyamide thin film composite
Membrane supplier and model	CSM RE-8040-FE
Nominal membrane rejection—%	>99.4
RO—reverse osmosis	

- Installation of a replacement membrane element: If a single, nonintegral membrane is replaced, the replacement element will likely have different salt rejection and water flux characteristics. This may result in a temporary excursion until the element acclimates to the operating conditions.
- Changes in water temperature: Salt passage across an RO membrane increases with water temperature. The membrane supplier should be able to provide water and salt passage coefficients, commonly called temperature correction factors, to account for changes in water temperature.





- Water flow, pressure, and system recovery: In any membrane system, the operational performance is dependent on overall system performance monitored at the stage level. Changes in performance that are associated with fouling or scaling events may skew results of the test.
- Unit start-up and shut-down: During normal startup and shut-down processes, it is common practice to reduce the recovery and flush the feed/concentrate side of the membrane with permeate or

feedwater. Conductivity may not be stable enough to perform on-line, real-time calculations for log removal. Alarms and calculations should be disabled during these sequences. The time required to obtain stable operation varies but generally is within 10-15 min after the unit reaches its production set point.

- Instrument calibration: Faulty or out-of-calibration flow, pressure, temperature, or conductivity instrumentation may yield errors in the calculations. Because the methodology is sensitive to the conductivity analyzer, more frequent calibration may be required. Differences in calibration between handheld conductivity meters and process instrumentation may result in inaccurate calculations.
- Membrane cleaning: Membrane cleaning removes foulants and scalants that have accumulated on the membrane. Normally, after the membrane has been cleaned, some temporary loss of rejection may be observed, resulting in higher permeate conductivity. Thus, it is normally recommended to perform profiling after cleaning in order to re-establish performance.
- Membrane type: The proposed methodology is suitable for RO systems with membrane elements that have salt rejection characteristics above 99% using sodium chloride as the indicator. The methodology and statistical validity may not apply for membranes with lower rejection (i.e., nanofiltration).

			Stag	ge 1							Stag	ge 1			
21.36	18.05	19.17	18.46	17.63	18.56	17.17	Average	17.46	14.93	15.82	15.51	14.70	15.50	14.67	Average
20.85	17.59	20.43	19.58	18.93	19.35	16.93	18.76	17.30	14.63	33.80	16.29	15.81	16.08	14.52	16.12
18.88	18.56	18.08	19.82	18.42	18.40	17.60		15.73	15.43	15.21	16.42	15.58	15.67	14.91	
17.72	20.62	17.77	18.19	18.84	17.13	18.23		14.84	17.20	15.67	15.13	16.06	14.72	15.36	
19.90	19.30	17.81	17.69	20.19	17.22			16.50	16.62	15.17	15.92	17.61	14.74		
20.33	20.16	18.30	20.57	17.56	18.44			16.73	16.83	15.67	17.40	15.04	15.60		
19.20	19.02	18.87	19.59	17.57	18.76			16.21	16.12	15.93	15.47	15.10	15.83		
17.33	16.97	19.41	19.15	19.40	20.31			14.60	14.60	16.22	16.05	16.40	17.03		
		Star	1e 2							Sta	ne 2				
42 63	45.52	44 89	44 89	43 07	Average			31.83	35.57	34.94	34.88	33.07	Average		
39.79	45.82	39.31	42.59	47.85	44.0			29.96	35.66	30.23	32.61	36.95	33.7		
38.84	45.04	41.88	53.49	42.22				28.89	34.89	32.26	32.61	36.95			
39.16	43.2	45.63	42.91	51.36				30.25	33.66	35.61	33.31	40.38			
onductivit	y Calcula	tions						Conductivit	y Calcula	tions					
arameter		Units	Stage 1	Stage 2				Parameter		Units	Stage 1	Stage 2			
verage		µS/cm	18.76	44.0				Average		µS/cm	16.12	33.7			
td Deviatio	on	µS/cm	1.1	3.8				Std Deviation	on	µS/cm	3.5	2.8			
ledian		µS/cm	18.7	43.1				Median		µS/cm	15.6	33.5			
laximum		µS/cm	21	53				Maximum		µS/cm	34	40			
vg +3*Std		µS/cm	22	55				Avg +3*Std		µS/cm	27	42			
ledian +50	1%	µS/cm	28	65				Median +50)%	µS/cm	23	50			
kew			0.32	0.93				Skew			4.95	0.32			

FIGURE 10 Conductivity profile and statistical calculation: the Yucaipa Valley Water District (YVWD)

	Test Condition	Feed Concentration—pfu/mL	Feed to Vessel 1-2-3	Feed to Combined Permeate
MS-2 virus reduction	Integral	1.21×10^{6}	5.04	5.38
Calculated LRV _{defect} (median)	Integral	NA	NA	3.56
MS-2 virus reduction	Compromised	1.45×10^{6}	1.70	4.16
Calculated LRV _{defect} (median)	Compromised	NA	NA	3.09

• Number of vessels/vessel flow: Ideally, there should be a minimum number of vessels used to provide a basis for sampling in order to develop a statistically valid result. In addition to a statistically valid result, the results need to be random and not specifically associated with vessel location (top to bottom, left to right). Flow from individual vessels is assumed to be within statistical limits.

CASE STUDY RESULTS

OCWD operates the Groundwater Replenishment System for the production of advanced treated recycled water for indirect potable reuse that is used as a seawater intrusion barrier and to augment groundwater supplies through surface spreading. The 100 mgd facility consists of 21 RO units, each with a nominal capacity of 5 mgd. Of the units, 15 were commissioned in 2007, and six additional units were placed in operation in 2015. The configuration of 15 RO units is 78:48:24, and six of the units are configured as 77:49:24. Membranes installed in the facility range from zero to nine years in age. There are a total of 3,150 pressure vessels in operation.

OCWD routinely monitors conductivity from its system and has its own operational criteria for maintenance. A unit permeate conductivity limit of 110 μ S/cm for any unit and a system limit of 95 μ S/cm have been established as operational standards based on the internal practices and input from an independent advisory panel. Conductivity profile results from August–September 2016 were used to assess the condition of the facility using the statistical methods described earlier. A summary of the results is provided here.

- The conductivity profile indicates an average of $47 \,\mu$ S/cm, with a minimum of 20 μ S/cm and maximum of 83 μ S/cm, and is well below the unit operational limit of 110 μ S/cm.
- Using the flow-weighted median TDS as a basis for C_{diff}, the overall LRV for the units ranged from a

minimum of 3.18 to a maximum of 4.11, with an arithmetic average of 3.55.

- No individual vessel was found to be operating with a conductivity of greater than 100% of the average or median stage conductivity.
- Using the three-sigma rule, 26 pressure vessels (0.8%) were identified as having possible integrity issues. The three-sigma rule appeared to be the most sensitive to newer membrane installation with a very low standard deviation of the conductivity profile distribution. Further observation indicated that only 12 of the 26 vessels exhibited issues with three-sigma and skew higher than 1.0.
- Using the median + 50% criterion, 21 vessels (0.7%) were identified as having potential integrity issues. Of those 21 vessels, seven were associated with an older unit with a mixed membrane installation. It was noted that the three-sigma test did not indicate an integrity issue with this unit.
- Of the 3,150 vessels evaluated, seven did not pass the three-sigma and median + 50% criteria (0.2%) and were scheduled for further evaluation; three of the vessels involved were associated with a recent membrane installation that involved a less stringent criterion for initial acceptance.
- Membrane age did not appear to be a determining factor in the identification of integrity issues.
- One unit stage was observed with a negative skew outside of the limit, a result of a prior operational occurrence that necessitated membrane replacement in that vessel.

Thus, aside from potential issues associated with routine operation, the overall approach appears to identify correctable issues necessary to ensure operational performance as integral units. The use of both the threesigma and median + 50% tests to identify potential issues was subjectively viewed as the most practical approach to identify integrity issues. Skew was more difficult to assess as an indicator of overall integrity; however, it was useful for identifying vessels that did not pass the three-sigma test as a result of new membrane installation.

SUMMARY

As stated at the beginning of this article, the intent of this discussion was to develop a compliance methodology that would satisfy the USEPA's MFGM IVP for an RO system using a statistical analysis of a conductivity profile for a typical production unit. The use of individual vessels provides a basis for the evaluation of comparative vessel performance that does not exist with pilot equipment.

The proposed approach is analogous to that which is used for air pressure–based integrity tests. In a pressure decay test, change in air pressure applied to one or more membranes is converted to a flow and correlated to an LRV through the use of a model. For RO systems, a statistical analysis of the results of a conductivity profile can be used to increase the sensitivity of the integrity test. Overall conductivity can be described in terms of its diffusion or defect-related components, and it is the defectrelated conductivity which would pass a pathogen.

Because the removal across the RO unit is a function of feedwater quality, each stage of the unit has to be examined individually. Unit integrity is determined by vessel conductivity profiling by stage and analyzed using statistical methods. The calculation of LRV for an RO unit is modified to account for the naturally occurring diffusion of solutes across the membrane, such that the result provided indicate the LRV_{defect} that is defect (pathogen) related. Although the method can be applied to the combined permeate, it is necessary to perform the calculation using the individual stage to address the underlying change in concentration that occurs in an RO unit. If a defect is identified, individual element testing can be performed to identify nonintegral elements that may be replaced or have defective O-rings replaced. The concept that sampling of permeate quality from a group of integral RO pressure vessels should fit a normal statistical distribution represents an alternative approach to the determination of integrity. While this method used conductivity, it may be applied to chemical or fluorescent markers for enhancement of those techniques.

ACKNOWLEDGMENT

The author acknowledges OCWD, Bill Dunivin, Mehul Patel, and Tyson Neely for allowing information from the Groundwater Replenishment System system to be used as the inspiration in the preparation of this concept, and Doug Owen of Doug Owen Water for reviewing the concept and providing comments before submittal. YVWD supported the MS2 testing of their RO system. The author acknowledges Joseph Zoba, Kevin King, Thaxton VanBelle, and Kevin Lee. Vic Moreland, Steve McDermitt of Trojan Technologies, and Shawn Verhoeven of GAP EnviroMicrobial Services Ltd., who assisted with the MS2 testing at YVWD, are also acknowledged. In addition, the mentoring of Vincent P. Oliveri and Richard G. Sudak was appreciated and has been missed.

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https://doi.org/10.1002/awwa.1087

PEER REVIEW

Date of submission: 11/15/17 Date of acceptance: 04/12/18

REFERENCES

- Adham, S.; Trussel, S.; Gagliardo, P.; & Trussel, R., 1998. Rejection of MS-2 Virus by RO Membranes. *Journal AWWA*, 90:135. https://doi. org/10.1002/j.1551-8833.1998.tb08505.x.
- ASTM International, 2010. *Standard Practice for Standardizing Reverse* Osmosis Performance Data. Designation D4516-00. ASTM International, West Conshohocken, Pa.
- AWWA, 2007. Reverse Osmosis and Nanofiltration. *Manual of Water* Supply Practices M46. AWWA, Denver.
- AWWA, 1991. Guidance Manual for Compliance With the Filtration and Disinfection Requirements for Public Water Systems Using Surface Water Sources. AWWA, Denver.
- California Code of Regulations 60320.208 revised. 2014.
- Jacangelo, J. & Gray, S., 2015. Assessment of Selected Methodologies for Monitoring the Integrity of Reverse Osmosis Membranes for Water Recycling. Webinar. WateReuse Research Foundation, Alexandria, Va.
- Joanes, D.N. & Gill, C.A., 1998. Comparing Measures of Sample Skewness and Kurtosis. *Journal of the Royal Statistical Society: Series D (The Statistician)*, 47:1:183. https://doi.org/10. 1111/1467-9884.00122.
- Kazmier, L., 2003. *Schaum's Outlines: Business Statistics*. McGraw Hill Professional.
- Kitis, M.; Lozier, J.C.; Kim, J.H.; Mi, B.; & Marinas, B., 2003. Microbial Removal and Integrity of RO and NF Membranes. *Journal* AWWA, 95:12:105. https://doi.org/10.1002/j.1551-8833.2003.tb10515.x.

Lonsdale, H.K.; Lacey, R.E.; & Loeb, S., editors, 1972. Chapter 8: Theory and Practice of Reverse Osmosis and Ultrafiltration. *Industrial Processing with Membranes*. John Wiley, New York.

Lonsdale, H.K.; Merten, U.; & Riley, R.L., 1965. Transport Properties of Cellulose Acetate Osmotic Membranes. *Journal of Applied Polymer Science*, 9:4:1341. https://doi.org/10.1002/app.1965.070090413.

- Miller, R.L.; Bradford, W.L.; & Peters, N.E., 1986. Specific Conductance: Theoretical Considerations and Application to Analytical Quality Control. USGS Water Supply Paper No 2311. US Government Printing Office, Denver, Colo.
- Porter, M. & Sudak, R., 1990. Reverse Osmosis. *Handbook of Industrial Membrane Technology*. Noyes Publications, Park Ridge, N.J.
- Surawanvijit, S.; Thompson, J.; Rahardianto, A.; Frankel, V.; & Cohen, Y., 2015. Pulsed Marker Method for Real-Time Detection of Reverse Osmosis Membrane Integrity Loss. *Desalination*, 370:25. https://doi.org/10.1016/j.desal.2015.05.003.
- USEPA (US Environmental Protection Agency), 2005. *Membrane Filtration Guidance Manual*. EPA-815-R-06-009. USEPA, Cincinnati, Ohio.
- WateReuse Research Foundation, 2015. *Framework for Direct Potable Reuse.* Project 14-20. WateReuse, Alexandria, Va.
- WateReuse Research Foundation, 2014. *New Techniques for Real Time Monitoring of Membrane Integrity for Virus Removal*. Project 09-06. WateReuse, Alexandria, Va.