

Technologies To Meet Numeric Nutrient Criteria

Erica Stone, Rod Reardon

The US Environmental Protection Agency (EPA) adopted numeric nutrient criteria (NNC) for total nitrogen (TN) and total phosphorus (TP) in inland lakes and flowing waters for the state of Florida in November 2010. Fifteen months were given for cities, businesses, utilities, and other stakeholders to develop strategies for implementing and meeting these criteria. In addition, EPA is working to develop similar criteria for coastal and estuarine waters and canals by 2012, potentially affecting nearly all wastewater dischargers in the state. This leaves many utilities with unanswered questions, including those about available treatment technologies that can reliably meet these low nitrogen and phosphorus concentrations.

The best performance for nutrient removal across the state has historically been accepted as advanced wastewater treatment (AWT) limits. In Florida, these limits are 5 mg/L carbonaceous five-day biochemical oxygen demand (cBOD₅), 5 mg/L total suspended solids (TSS), 3 mg/L TN, and 1 mg/L TP. Typ-

ically, biological nutrient removal (BNR) facilities with tertiary filters are able to consistently and reliably meet the AWT standards. EPA's numeric nutrient criteria (NNC) limits fall below these levels at 0.67 to 1.87 mg/L TN and 0.06 to 0.49 mg/L TP for flowing waters, depending on the nutrient watershed region (EPA 2010). Should the EPA's NNC survive legal challenges, some wastewater utilities may ultimately need to plan for the potential implementation of technologies capable of meeting these new standards.

Technologies

To reduce TN and TP concentrations in wastewater effluents below those that can be achieved with conventional AWT, the characteristics of the residual TN and TP must be considered. TN is made up of inorganic fractions including ammonia, nitrate, and nitrite, which are typically reduced to low levels in a biological process, as well as organic nitrogen fractions, the most challenging of which is re-

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fractive dissolved organic nitrogen (rDON). TP is made up of orthophosphates and also a refractive dissolved organic phosphorus (rDOP) fraction.

Both rDON and rDOP have received much attention recently due to the difficulty in reducing effluent nutrient concentrations lower than the rDON or rDOP fraction. One study evaluated the fractions of effluent phosphorus from six different tertiary treatment technologies. The rDOP was the dominant fraction of phosphorus in all cases and only showed signs of removal with adsorptive media processes; chemical treatment and filtration were not effective methods of removal (Liu et al. 2011). Another study evaluated the effluent rDON in several advanced wastewater treatment plants and found rDON to make up more than 80 percent of the NNC for TN (Bratby et al. 2008; Matthews et al. 2011). There is currently little known about the various species that make up rDON and rDOP, which adds to the complications of removal in a treatment plant. Most of the rDON and rDOP in wastewater effluents are from the influent that has passed through the treatment process, contributed by industrial sources or even the potable water supply. However, a small portion of rDON and rDOP are also produced in the biological processes with cell decay.

In Florida, and in other areas of the country, technologies are in use that can meet the NNC limits for TP. These include tertiary coagulation, clarification and filtration, tertiary membrane filters, membrane bioreactors (MBRs), reactive filtration, and wetlands, with innovative forms of ion exchange also available. Removal of residual TN will likely be considerably more challenging, but technologies exist that may be able to meet the new NNC. Technologies with the potential to meet very low TN criteria, potentially at lower cost than the traditional gold standard of microfiltration/reverse osmosis (MF/RO), include separate stage biological treatment, granular activated carbon (GAC), advanced oxidation processes possibly combined with biological treatment and membrane filtration, and again,

Facility	Process	Effluent N	Effluent P
Lab-scale study ¹	Enhanced coagulation-MF	0.3 mg/L rDON	0.1 mg/L rDOP
Rock Creek ²	Classic tertiary treatment	No data	0.04 mg/L TP
Concord ³	CoMag™	No data	<0.05 mg/L TP
Gwinnett County ⁴	Coagulation-MF/UF	No data	0.04 mg/L TP

Notes:
1.0 (Aarnaldos and Pagilla 2010)
2.0 (Pagilla and Urgan-Demirtas 2009)
3.0 (Ellis and Cathcart 2008)
4.0 (Neethling et al. 2009)

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land-based systems like soil aquifer treatment (SAT) and wetlands.

Optimizing BNR Processes

Optimizing conventional BNR processes is the first step toward reliably achieving lower effluent nutrient concentrations. Some options for increasing nutrient removal in conventional BNR processes include increasing biomass concentrations, finding optimum solids retention times (SRTs), adding supplemental carbon, making use of reactors in series, managing recycle streams, improving monitoring and control of the process, and providing increased redundancy or multiple barriers. Increased biomass concentrations mean longer SRTs and increased nitrification efficiency. Supplemental carbon increases denitrification rates and phosphorus removal efficiency. Reactors in series help processes approach plug flow reactor performance, which provides higher reaction rates that translate into better removal within existing tank volumes. Managing recycle streams may include treatment of sidestreams to remove nutrients prior to recycling back into the biological process.

The BioMag™ process is an example of an innovative method that can enhance the

performance of existing BNR processes by simultaneously increasing the biomass concentration and chemically precipitating phosphorus (Woodard and Andryszak 2011). BioMag™ continuously adds magnetite (Fe₃O₄) ballast to the mixed liquor of the activated sludge process. In the secondary clarifier, the ballasted solids rapidly settle and thicken. Waste activated sludge is passed through a shearing device to recover the magnetite for recirculation to the biological process. The ballast allows for increased mixed liquor suspended solids (MLSS) concentrations in the bioreactors and improved settling in the secondary clarifiers. The improved settling characteristics of the sludge may also eliminate the need for tertiary filters. Selected effluent water quality from two facilities that piloted BioMag™ is presented in Table 1.

Tertiary Coagulation and Sedimentation

Tertiary coagulation, flocculation, and sedimentation processes help to aggregate smaller particles into larger ones to facilitate enhanced solids removal by sedimentation and more efficient filtration. This is accomplished with the addition of a metal salt coagulant with rapid mixing followed by

flocculation, sedimentation, and filtration. This conventional process may be a valid alternative, depending on the target TP. Addition of sand ballast (Actiflo™), magnetite ballast (CoMag™), or sludge recirculation (DensaDeg) can be used with tertiary sedimentation processes to enhance coagulation and improve removals. These processes also provide more rapid settling, thus helping to minimize process land requirements.

Filtration

The type of filtration technology that follows coagulation/flocculation/sedimentation processes can greatly affect effluent quality. Filtration improves as the filter medium pore sizes get smaller. For example, granular media filtration has larger pores than microfiltration membranes, which have larger pore sizes than ultrafiltration membranes. An example particle size distribution is shown in Figure 1, illustrating the difference between the filtered secondary effluent, filtration following coagulation/flocculation/sedimentation, and microfiltration membranes. Table 2 shows the effluent quality of some select tertiary coagulation processes with different effluent filters.

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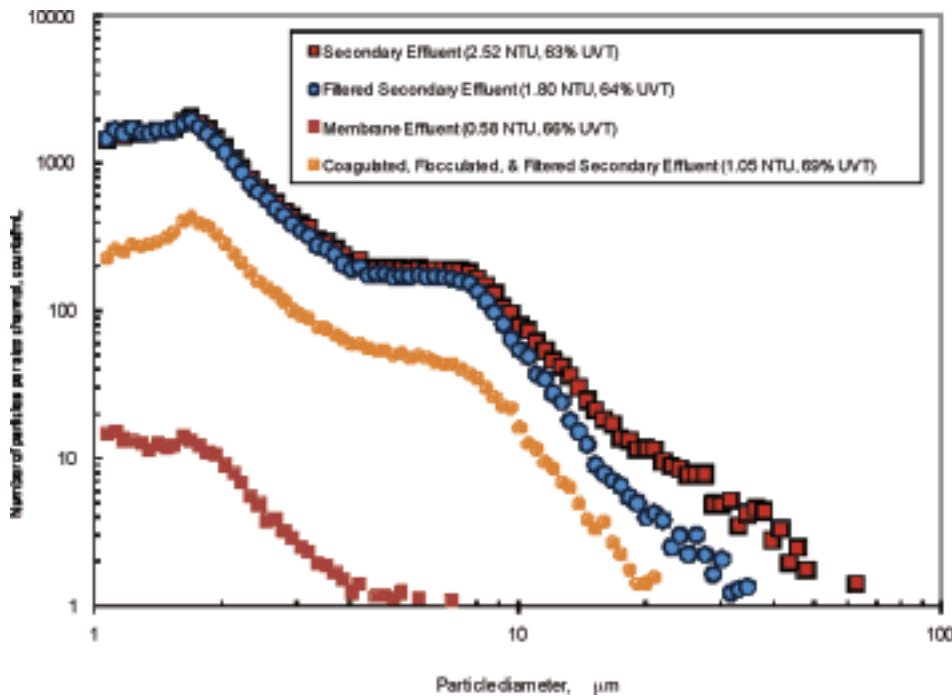


Figure 1 Particle size distribution for various filtration technologies (Bourgeois 2010)

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Land-Based Systems

Land-based systems like wetlands and SAT (Fox et al. 2001) have been used in some places across the country to offer a barrier to organics, nutrients, and pathogens for treated wastewater effluent discharging to both surface water and groundwater. Wetlands contain decomposing microorganisms that remove dissolved biodegradable material and aquatic plants that release oxygen into the water and take up nutrients. Much of the nitrate in wastewater is denitrified in wetlands. Wetlands can typically achieve effluent quality of 1-2 mg/L TN, less than 0.05 mg/L NO_x-N, less than 0.2 mg/L NH₃-N, and less than 0.05 mg/L TP (Knight 2011).

The SAT is often accomplished by aquifer recharge with reclaimed wastewater via spreading basins. Water percolates through the unsaturated zone to groundwater where it is mixed, transported, stored, and polished. Ammonia is adsorbed onto the upper parts of the soil when water is distributed to spreading basins. As the soil dries, the air enters the soil and ammonia is oxidized to nitrate. Nitrate is more mobile and is transported deep to the vadose zone where anoxic conditions are present and is converted to nitrogen gas if organic carbon is present. Nitrogen removal of 25 to 90 percent has been observed by this mechanism; however, most SAT systems do not have enough organic carbon, so anammox bacteria

have been the proposed mechanism. Cycling between wet and dry periods forces both ammonia and nitrate to reach the deeper anoxic and anaerobic zones that support anammox microorganisms. Typically, 50 percent or more nitrogen removal is achieved (Fox et al. 2001; Fox et al. 2006).

Adsorption

Adsorption is another mechanism by which nutrients in wastewater effluent can be reduced. The most well-known adsorption process is GAC; however, proprietary adsorption processes are under development that remove phosphorus by adsorption onto media.

The primary use of GAC has been to adsorb soluble organics from wastewater effluent (Culp et al. 1978). Adsorption occurs when molecules adhere to the internal walls of pores in carbon particles produced by thermal activation. Regeneration of the carbon is accomplished by thermal means and organics in the pores are oxidized and removed. GAC adsorption is typically a tertiary process after filtration and before disinfection. The Niagara Falls Wastewater Treatment Plant has carbon beds that provide filtration of solids, adsorption of chemical compounds, and biological degradation from anaerobic activity in the beds. The facility reduces 800 pounds of pollutants to 12 pounds in the effluent at 35 mgd of influent flow. The Niagara Falls plant was originally designed as a physical/chemical plant in 1985 due to the high industrial wastes it received without intentions of nutrient removal. Today,

industrial flow is about 17 percent of the total flow and the discharge permit includes limits on volatile compounds, acid compounds, base/neutral compounds, pesticides, metals, and cyanide (EPA 2000). The Upper Occoquan Sewage Authority (UOSA) Millard H. Robbins Reclamation Facility treats 24 mgd, with a VPDES permit that limits chemical oxygen demand (COD) to 10 mg/L. GAC is used to reduce non-biodegradable soluble organics to a COD concentration of 6-7 mg/L, which is a 50 percent removal of COD in the GAC process. The beds are regenerated when removal is reduced to 25 percent (EPA 2000). This facility uses GAC to meet the COD limit. By extension, GAC should remove rDON and rDOP from secondary effluents, but this remains to be demonstrated.

Bayoxide® E33, a product sold by Severn Trent Services, is a granular synthetic ferric oxide media with more than 70 percent Fe₂O₃ content. The media can be regenerated with potassium hydroxide eight to 10 times with a weak caustic solution, and can also be land-filled when exhausted. Since 1999, the Bayoxide® media has primarily been used for arsenic removal from drinking water. There are more than 70 installations in the U.S., 30 in the United Kingdom, and others in South America. A rapid small-scale column test in Telford, Pa., on tertiary-treated wastewater was shown to adsorb phosphorus for 62,903 bed volumes without achieving breakthrough. The test resulted in removal of 90 percent of phosphorus at start-up, and then declined to 61 percent removal about half way through the test. Effluent water quality averaged 0.22 mg/L TP (Dennis and Clark 2010).

The Blue PRO® process provides phosphorus removal by adsorption onto hydrous ferric oxide-coated sand media in continuous backwash moving bed filters with chemical (FeCl₃) addition. The system can operate in a single- or double-pass configuration, depending on effluent TP requirements. The waste stream of phosphorus and iron removed from the sand media can be recycled in the plant, added to sludge handling, or dewatered and handled separately. Effluent from a single-pass system at a plant in Hayden, Idaho, contained 0.192 mg/L TP, while effluent from a dual-pass system was 0.014 mg/L TP. Second-pass effluent concentrations as low as 0.009 mg/L TP were achieved as a monthly average (Leaf and Johnson 2006).

A new synthetic adsorbent material created by Asahi Kasei Chemicals contains a metal oxide with ion exchange properties mixed with polymer. A pilot plant treating secondary effluent showed that effluent phosphorus concentrations consistently below 0.03 mg/L were possible. An alkaline solution (5

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percent sodium hydroxide) was able to desorb 90-97 percent of the phosphorus, with the media regenerated by acid neutralization. The phosphorus can then be recovered by precipitation with calcium hydroxide, and the alkaline solution can be reused for desorption. The recovered phosphate is similar to phosphate ore and citric-soluble phosphorus, so there is the potential for use as fertilizer (Fitzpatrick et al. 2009). Table 3 shows the reported effluent water quality for a few of the processes described above that use adsorption mechanisms for phosphorus removal.

Ion Exchange

Ion exchange resins have long been used for ammonia removal from many water sources and nitrate removal in drinking water applications (Belser Baykal et al. 2011; Severn Trent Services 2007). Ion exchange resins range from naturally occurring zeolites to manufactured synthetic media. Typical drinking water applications can reduce nitrate concentrations to 0.8 mg/L NO₃-N. Since the drinking water standard for nitrate is 10 mg/L NO₃-N, little effort has been made to reduce concentrations much lower. The ability to recover ammonia from spent regenerate and to recycle it as fertilizer is once again gaining interest.

Clinoptilolite is a natural zeolite that can remove ammonia from most sources via ion

exchange, including domestic wastewater (Gisvold et al. 2000; Hedstrom and Rastas Amofah 2008; Mercer et al. 1970). Clinoptilolite has been successfully used to remove ammonia from water for decades, albeit with much higher target effluent concentrations. Studies have also shown that spent clinoptilolite can be used as fertilizer and release 90 to 96 percent of the accumulated ammonia over several months of irrigation to soils (Belser Baykal et al. 2011).

It remains to be demonstrated that ion exchange can be effective and economical in removing ammonia and nitrate to very low concentrations to be considered a tool for meeting NNC.

Tertiary Biological Process

Studies have shown that tertiary biological processes, including tertiary nitrifying filters, fluidized bed reactors, MBRs, two-sludge processes, and membrane biofilm reactors (MBfRs), offer the potential to provide incrementally lower effluent concentrations of TN (Parker et al. 2011). Two-sludge systems make use of tertiary biological process by separating the nitrification and denitrification processes into separate reactors with their own clarifiers (thus, producing two sludge streams). Typically, supplemental carbon (like methanol) is added to the second stage due to low influent carbon from the first stage. MBRs have tradi-

tionally been used as the principal biological treatment process for applications requiring high quality reclaimed water; however, the potential exists to apply MBRs as tertiary biological treatment systems for both nitrification and denitrification, where their ability to completely capture suspended solids could provide a strong advantage over traditional two-sludge processes.

MBfRs use gas permeable fabric as attachment media and to deliver hydrogen gas to the biomass. Bacteria grow on the fabric media using the hydrogen as the electron donor for denitrification; the hydrogen gas is highly efficient due to low solubility. The bacteria are naturally occurring and use bicarbonate as a carbon source. Table 4 shows a summary of systems that make use of tertiary biological processes and their typical effluent water quality.

Oxidation

Oxidation has been proposed as another method for reducing organic nitrogen and phosphorus compounds. Very few oxidation processes have been used for this purpose to date. Ideally, large, recalcitrant organic substances containing nitrogen and phosphorus would be broken down into small, biodegradable compounds by the oxidant that a subsequent biological system can remove. However, the biological process may itself create additional recalcitrant organics.

One strong oxidant for which treatability studies in wastewater treatment plants have been done is ferrate. Ferrate ion (Fe⁺⁶) is a very strong oxidant that in the past could not be economically produced in commercial quantities. Recently, Ferrate Treatment Technologies has developed a proprietary treatment system for making Fe⁺⁶, and is marketing its system for odor control, metal removal, enhanced coagulation, biosolids stabilization, disinfection, and nutrient reduction. Two treatability studies are summarized in Table 5. The first treatability study had an influent phosphorus concentration of 1.4 mg/L in the aeration basin effluent channel, 0.79 mg/L downstream of alum addition, 0.60 mg/L before the denitrification filters, and 0.86 mg/L after the denitrification filters. Samples from each location were dosed and analyzed in the laboratory. In the second treatability study, water quality was 1.2 mg/L total phosphorus, 0.76 mg/L orthophosphorus, and 7.5 mg/L NH₃-N in the chlorinated disc filter effluent. Ferrate does not create disinfection byproducts and also reduces some micropollutants including endocrine-disrupting compounds (EDCs) and pharmaceuticals and personal care products (PPCPs).

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Table 3 Reported Effluent TP Concentrations from Several Adsorption Processes

Facility	Process	Effluent TP
Telford, PA ¹	Bayoxide®, Column Test	0.017 mg/L
Hayden, ID ²	Blue PRO®, Oxidation Ditch	0.014 mg/L
Japan – pilot ³	Asahi synthetic media	0.02-0.04 mg/L
Notes:		
1. (Dennis and Clark 2010)		
2. (Leaf and Johnson 2006)		
3. (Fitzpatrick et al. 2009)		

Table 4 Tertiary Biological Processes Water Quality

Facility	Process	Effluent N	Effluent P
River Oaks, FL ¹	Two-sludge	0.17 mg/L NH ₃ -N 0.49 mg/L NO ₃ -N 0.73 mg/L N _{org} 1.49 mg/L TN	0.26 mg/L TP
California, pilot ²	MBfR	<0.5 mg/L NO ₃ -N <0.1 mg/L NO ₂ -N	0.88 mg/L PO ₄ -P
Hamilton, pilot ³	Tertiary MBR	< 2 mg/L NH ₃ -N	0.15 mg/L TP
Notes:			
1. (Jimenez et al. 2007)			
2. (LeBrun 2011)			
3. (Constantine et al. 2007)			

Location	Ferrate Dose	Effluent N	Effluent P
Treatability Study 1¹:			
Aeration basin effluent channel	1 mg/L	No data	0.27 mg/L TP
	3 mg/L		0.19 mg/L TP
	5 mg/L		0.08 mg/L TP
Downstream of alum addition	1 mg/L	No data	0.50 mg/L TP
	3 mg/L		0.15 mg/L TP
	5 mg/L		0.064 mg/L TP
Before denitrification filters	1 mg/L	No data	0.14 mg/L TP
	3 mg/L		<0.025 mg/L TP
After denitrification filters	1 mg/L	No data	0.20 mg/L TP
	3 mg/L		0.12 mg/L TP
Treatability Study 2²:			
Chlorinated disk filter effluent	2 mg/L	3.25 mg/L NH ₃ -N	0.14 mg/L TP
	4 mg/L	2.6 mg/L NH ₃ -N	0.12 mg/L TP
Notes:			
1. (Ferrate Treatment Technologies LLC 2010b)			
2. (Ferrate Treatment Technologies LLC 2010a)			

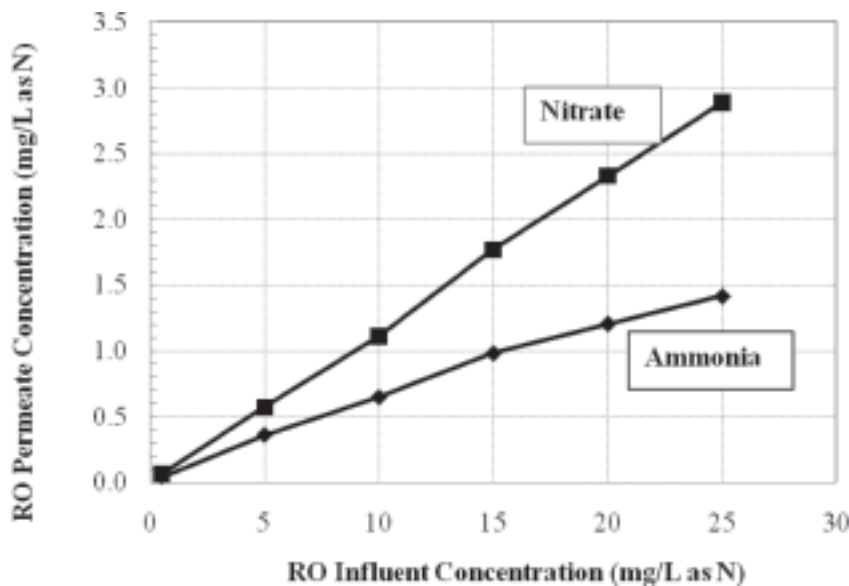


Figure 2: Estimated Rejection of Ammonia and Nitrate for One Brackish Water RO Membrane Based on RO Projection Software

Parameter	Year	Average Effluent	Removal
Ammonia	2003	0.16 mg/L-N	95%
	2007	0.32 mg/L-N	39%
Nitrate	2003	0.93 mg/L-N	91%
	2007	2.19 mg/L-N	66%
Nitrite	2003	<0.01 mg/L-N	100%
	2007	<0.01 mg/L-N	100%
TP	2003	0.06 mg/L	99%
	2007	0.06 mg/L	99%

High-Pressure Membranes

Reverse osmosis (RO) and nanofiltration (NF) membranes are high-pressure membranes noted for their ability to remove a wide range of organic and inorganic substances from water. Both NF and RO have been used for treatment of water and wastewater for applications requiring very high quality water. Typically, MF or ultrafiltration (UF) membranes are used as pretreatment to the RO membranes.

The largest application for RO membranes in wastewater treatment is to reclaim water for indirect potable reuse. In this application, nutrient removal has not been a concern and little information has been published on the ability of high-pressure membranes to remove nitrogen and phosphorus from wastewater. A recent study (Merlo et al. 2011) collected data from RO pilots and full-scale plants. This data showed 68-94 percent removal of organic N, which suggests that organic nitrogen removal with RO is not as efficient as for most inorganic ions. Projections by manufacturers' software for one brackish water RO membrane commonly used for wastewater treatment also show reduced rejection of ammonia and especially nitrate, as shown in Figure 2. Additionally, the performance of membranes has in some cases decreased as the membranes age. A full-scale RO plant has reported a drop in ammonia removal from 95 percent in 2003 to 39 percent in 2007, and for nitrate, a drop from 91 percent to 66 percent. A summary of this data is shown in Table 6.

Conclusions

Numeric nutrient criteria are still new – their ultimate form awaits the promulgation of the criteria for coastal and estuarine waters due in August 2012 and the resolution of recent court challenges. The specific methods for implementation of NNC are likewise still being developed. However, review of the current status of potential technologies for meeting more stringent nutrient limits can help in understanding the challenges in producing reclaimed water with very low concentrations of nutrients.

Table 7 presents a summary of the reported nutrient removal efficiency for the technologies described above. For many of the technologies, only limited data are available from pilot plants, short-term trials, or applications of the technologies for purposes other than nutrient removal. Such data is not comparable to the long-term data required to show compliance with geometric annual means for

periods of three years as required by EPA's NNC. As shown in Table 7, there are technologies other than MF/RO potentially capable of achieving NNC as end-of-pipe limits under the right circumstances. Due to the inherent limitations of biological kinetics in allowing biological processes to meet low substrate concentrations (Grady et al. 1999; Rittmann and McCarty 2001), a combination of biological and physical/chemical technologies will likely be required to meet ultra-low nutrient standards.

Capital and operating costs and long-term performance of these technologies for this application are largely unknown. Their use will represent a significant economic and political risk for utilities until the technologies are more fully developed. However, the development of new, more effective, and more economical technologies could ease the pain for any facilities that ultimately are required to produce reclaimed water meeting NNC.

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Table 7 Summary of Reported Effluent Nutrient Concentrations for Potential Innovative or Emerging Technologies for Meeting NNC

Category	Range TN (mg/L)	Range TP (mg/L)
EPA NNC	0.67-1.87	0.06-0.49
Optimize BNR	1.2-1.4	0.05-0.11
Tertiary Coagulation/ Sedimentation/Filtration	No data	0.04-0.10
Land Based Systems	1-2	<0.05
Adsorption	3	0.014-0.040
Ion Exchange	0.8	0.1
Tertiary Biological Processes	0.6-2.0	0.15-0.88
Oxidation	2.6-3.25	0.025-0.500
RO	1.10-2.25	0.06
<u>Notes:</u>		
1. See previous tables for references.		

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