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Proposed nutrient criteria for water supply lakes and reservoirs

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Nutrient enrichment of lakes and reservoirs used for a potable water supply can lead to a broad array of adverse effects ranging from operational problems to potential increases in certain human health–related risks. Human health risks that may be exacerbated by nutrient enrichment stem from increases in disinfection by-products, cyanotoxins, and arsenic. New York state is developing numeric nutrient criteria for the protection of water supply lakes and reservoirs by establishing relationships between nutrients, algal abundance, dissolved organic carbon, and trihalomethanes, and then targeting established regulatory endpoints to set appropriate numeric nutrient criteria thresholds. This approach represents a much-needed bridge between the Safe Drinking Water Act and the Clean Water Act by defining source water protection goals. Findings to date from the investigation of 21 systems indicate that a mean chlorophyll *a* threshold of 4–6 µg/L would likely be protective of potable water supply lakes and reservoirs.

KEYWORDS: *algae, DBPs, CWA, eutrophication, nutrients, SDWA*

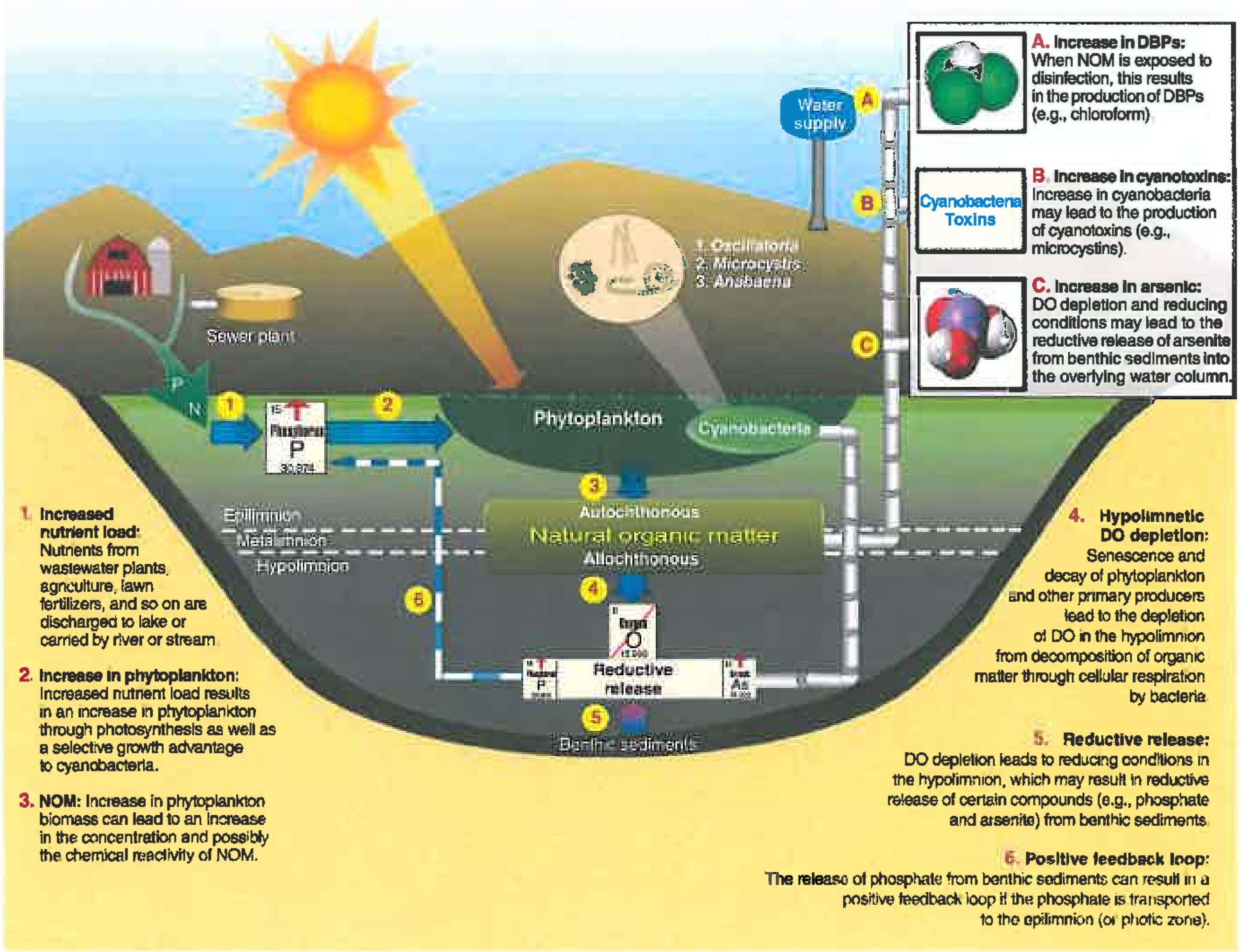
The US Environmental Protection Agency (USEPA) initiated a National Nutrient Strategy in 1998 (USEPA, 1998) that calls on states to establish numeric nutrient criteria (NNC) in an effort to address the adverse effects on designated uses originating from nutrient enrichment. Although many states are in the process of developing NNC, New York is one of the few states that have identified potable water supply (PWS) use as a specific target for NNC. Nutrient enrichment of lakes and reservoirs used for PWS can lead to a wide range of adverse effects ranging from operational problems (e.g., filter clogging) to customer nuisance complaints (e.g., taste and odor issues) to potential increases in certain human health–related risks. Human health–risk factors that may be exacerbated by nutrient enrichment include increased generation of disinfection by-products (DBPs), increased production of cyanotoxins by certain types of cyanobacteria, and increased arsenic concentrations—each of which has shown evidence of carcinogenicity, although to differing degrees. Figure 1 shows a conceptual model of the theoretical linkages between nutrient enrichment and potential effects to PWS. This article reports on New York state's efforts to develop such criteria for lakes and reservoirs with a designated use classification of PWS. This approach involves linking nutrient-related metrics to existing regulatory targets for one of the major classes of DBPs—total trihalomethanes (TTHMs). Although the scientific investigations presented are largely complete, the regulatory rulemaking process is ongoing and may affect how the findings and conclusions from the investigations are used and implemented.

The advent of disinfection for drinking water supplies represents a seminal event in the protection of public health from waterborne disease and remains a cornerstone of public health protection (Calderon, 2000). However, the disinfection process itself can bring about certain unintended and undesirable consequences, most significant of which is the production of DBPs, which can form when an oxidizing agent (e.g., chlorine) reacts with natural organic matter (NOM). The link between disinfection and the formation of DBPs was first recognized by Rook (1974) as well as Bellar and Lichtenberg (1974), who observed increases in chloroform and other DBPs associated with the chlorination of source waters. Subsequent studies have identified more than 600 DBPs (Richardson, 2002).

Research on the chemical pathways and mechanisms of formation for DBPs initially focused on the allochthonous (external or terrestrial) portion of the NOM precursor pool and what are termed humic and fulvic acids, which consist largely of plant and animal materials originating in the contributory watershed of the lake or reservoir. However, subsequent studies identified the autochthonous (in situ) portion of NOM—consisting of algae and other sources of organic matter generated within the water body itself—as an important portion of the NOM precursor pool. Evidence for the importance of autochthonous precursors in the production of DBPs has come from both field investigations and laboratory studies of algal cultures. Hoehn et al (1977) observed seasonal variations in THM concentrations in a Virginia reservoir, in addition to a direct correlation between chlorophyll and THM levels. Morris and Baum (1978) provide evidence of the importance of algae in the generation of DBPs in experiments with chlorophyll, whereas work by Arruda and Fromm (1989) and Cooke and Kennedy (2001) show direct correlations between trophic state and THM formation potential (THMFP) levels. Furthermore, Hoehn et al (1980) demonstrated that algal cells, both whole cells and extracellular products, readily form THMs when exposed to chlorine and that chloroform yield from extracellular materials was at least as high on a per-unit carbon basis as observed with humic and fulvic acids. Wardlaw et al (1991) also concluded that algae can be equally potent generators of DBPs as are humic and fulvic acids, and determined that algae could contribute THM concentrations of up to 150 µg/L. Palmstrom et al (1988) provide an informative synopsis of the autochthonous DBP precursor pool and offer evidence for the involvement of aquatic macrophytes in DBP production. These findings provide evidence for steps 3 and A in Figure 1.

There are also several important distinctions between allochthonous and autochthonous precursor pools that are relevant to management considerations. First, with respect to source water management, the potential to control ambient concentrations of the two precursor pools varies substantially in that the allochthonous component generally comes from inputs of terrestrial vegetation (e.g., leaves) that are difficult to control, whereas the autochthonous component is more amenable to mitigation through the management of nutrient inputs to the lake or reservoir.

FIGURE 1 Conceptual model of theoretical linkages between nutrient enrichment and drinking water human health concerns



A. Increase in DBPs: When NOM is exposed to disinfection, this results in the production of DBPs (e.g., chloroform).

B. Increase in cyanotoxins: Increase in cyanobacteria may lead to the production of cyanotoxins (e.g., microcystins).

C. Increase in arsenic: DO depletion and reducing conditions may lead to the reductive release of arsenite from benthic sediments into the overlying water column.

1. Increased nutrient load: Nutrients from wastewater plants, agriculture, lawn fertilizers, and so on are discharged to lake or carried by river or stream.

2. Increase in phytoplankton: Increased nutrient load results in an increase in phytoplankton through photosynthesis as well as a selective growth advantage to cyanobacteria.

3. NOM: Increase in phytoplankton biomass can lead to an increase in the concentration and possibly the chemical reactivity of NOM.

4. Hypolimnetic DO depletion: Senescence and decay of phytoplankton and other primary producers lead to the depletion of DO in the hypolimnion from decomposition of organic matter through cellular respiration by bacteria.

5. Reductive release: DO depletion leads to reducing conditions in the hypolimnion, which may result in reductive release of certain compounds (e.g., phosphate and arsenite) from benthic sediments.

6. Positive feedback loop: The release of phosphate from benthic sediments can result in a positive feedback loop if the phosphate is transported to the epilimnion (or photic zone).

Source: Clifford W. Callinan, Rachel E. Holt, and Frank S. Herec

DBP—disinfection by-product, DO—dissolved oxygen, N—nitrogen, NOM—natural organic matter

Second, with respect to water treatment, the allochthonous precursor pool is generally more amenable to removal by water treatment processes than is the autochthonous precursor pool. For example, Cheng and Chi (2003) observed an inverse relationship between degree of eutrophication and removal efficiency, and Takaara et al (2007) observed that dissolved organic carbon (DOC) removal efficiency declined as cyanobacteria levels increased.

Third, autochthonous precursors have been shown to generate greater concentrations of certain unregulated DBPs (e.g., nitrogenous DBPs), which may have greater toxicity than regulated DBPs. For example, Fang et al (2010) found that cultures of *Microcystis aeruginosa* generated more than double the level of dichloroacetonitrile than did a humic and fulvic acid standard when normalized for TOC.

Finally, there are several other human health–related concerns that can be exacerbated by nutrient enrichment, including the production of cyanobacterial toxins and the reductive release of arsenic (Figure 1, steps 4–6, B and C). In summary, there are several reasons to focus on nutrient controls, including the ability to control the autochthonous precursor pool, the limited ability to remove this portion of the precursor pool through conventional water treatment processes, the increased propensity to produce certain unregulated DBPs of concern, and the ability to mitigate other related concerns such as cyanotoxins and arsenic mobilization. Furthermore, other operational issues and customer-related concerns can be mitigated through the effective control of nutrient enrichment.

As the scientific understanding of the chemistry and toxicology of DBPs has expanded, the regulatory community has attempted to keep pace, as evidenced by the multistage process of formulating and implementing maximum contaminant levels (MCLs) for several classes of DBPs. However, most efforts to respond to these increasingly stringent regulatory endpoints have targeted treatment-based approaches rather than source water protection. Also, the treatment-based approaches can have certain unintended consequences. For example, the current regulatory focus on only certain classes of DBPs can lead to a "chase-the-MCL" approach through which treatment plant operations are modified (e.g., use of alternative disinfectant). Such modifications may address regulated DBP concerns but also lead to an increase in unregulated DBPs of potentially greater concern (e.g., greater toxicity). In a review of emerging unregulated DBPs, Richardson et al (2007) concluded that the levels of many emerging DBPs—a number of which are believed to be more toxic than regulated DBPs—are increased substantially by alternative disinfectants.

In contrast to these treatment-based measures, source water nutrient controls can mitigate the generation of DBPs more broadly, while simultaneously addressing several other human health–related PWS concerns (e.g., cyanotoxins and arsenic). This approach of limiting the production of autochthonous precursors through nutrient controls represents a bridge between the Clean Water Act (CWA) and the Safe Drinking Water Act (SDWA) by establishing a nexus between ambient water quality that falls largely within the CWA and drinking water regulatory limits as defined by MCLs that fall under the SDWA. In summary, the implementation of management measures to control nutrient enrichment of source waters represents a clear example of source water protection (a principal component of the multibarrier approach) would significantly reduce the burden placed on the treatment component of the multibarrier approach, and, most important, would substantially enhance the quality of water delivered to the consumer.

The purpose of this study is to respond to USEPA's charge to develop statewide NNC and specifically to derive NNC for water supply lakes and reservoirs based on relationships between algal biomass, NOM, and DBP formation. These criteria are expected to prevent exceeding regulated DBPs from autochthonous sources, assist in addressing several other human health–related concerns, and help mitigate a number of operational difficulties associated with the cultural eutrophication of surface water sources.

METHODS

Sampling. Sampling activities for the study involved instrument measurements and collection of water samples from 21 lakes and/or reservoirs within the state of New York (Figure 2). The systems represent a broad range of trophic conditions (oligotrophic to eutrophic) and include waters from several of the USEPA Level III ecoregions in New York state. In general, sampling was conducted monthly between May and October 2004 and/or 2007, and epilimnetic samples were collected using a 2-m polyvinyl chloride pipe equipped with a one-way flap valve.

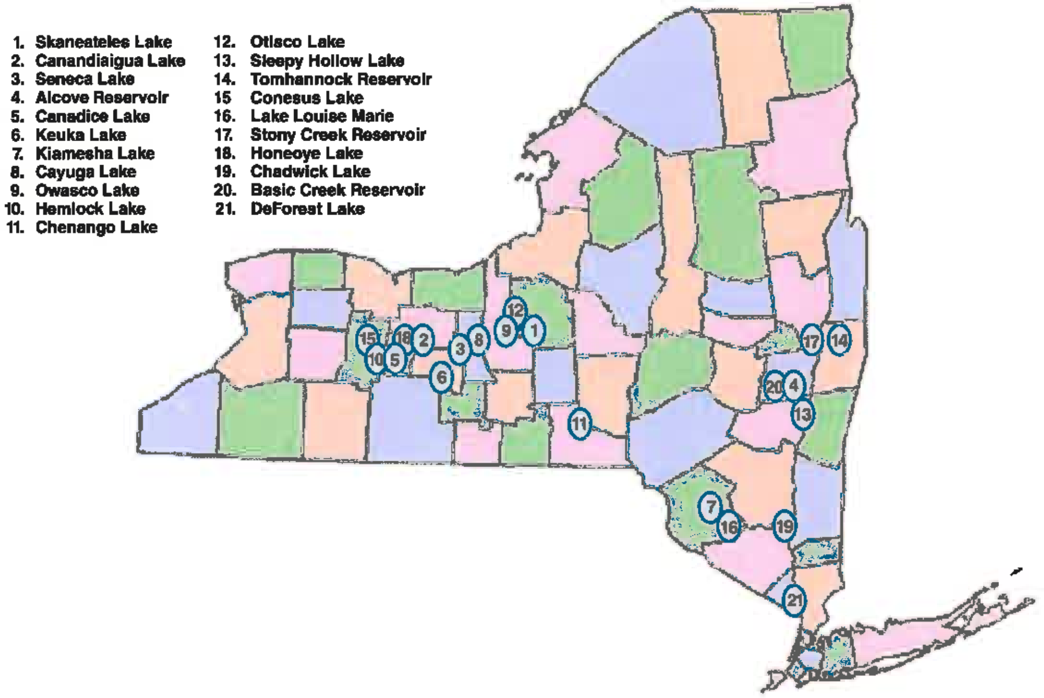
Chemical and biological analysis. Water samples collected during the study were analyzed for both conventional parameters (e.g., trophic state variables) and THMFP.

Conventional parameters. Total phosphorus (TP) was analyzed according to method 4500-PE (*Standard Methods*, 1998) with a limit of detection of 0.7 µg/L, and chlorophyll *a* (Chl*a*) was analyzed per methods outlined in Parsons et al (1984) with a limit of detection of 0.2 µg/L. DOC was analyzed according to method 5310C (*Standard Methods*, 1998) with a limit of detection of 0.3 mg/L.

THMFP. THMFP was determined by method 5710B (*Standard Methods*, 1998) modified to use 71-mL serum bottles with THM determination using method 6230C (*Standard Methods*, 1998) modified to use an electron capture detector. THMs quantified were trichloromethane (chloroform), dichlorobromomethane, chlorodibromomethane, and tribromomethane (bromoform). This method is designed to yield the maximum amount of THMs that can be produced from a sample by using a large excess of free chlorine. It does not necessarily represent the amount produced during actual water treatment and distribution conditions.

THMFP Quality control. All THMFP samples were analyzed in duplicate. Field blanks, consisting of boiled deionized water placed in sample bottles and sent to the field with sample bottles, were processed with each set of samples, as were field duplicate samples. Compounds (chloroform, dichlorobromomethane, chlorodibromomethane, bromoform) were identified by retention time and quantified by external standards. External standard solutions were prepared from a standard solution of 2,000 µg/mL of each component in methanol.1 Working gas chromatography standards were prepared by serial dilution of the stock to 2-, 4-, 6-, 8-, 10-, and 12-µg/L solutions. Standard curves were

FIGURE 2 Study sites



determined with each set of samples. A check standard (6 µg/L) was analyzed after every tenth sample.

RESULTS AND DISCUSSION

Several introductory comments about the study results are in order. A substantial portion of the study's results are given using bivariate regressions based on system-specific means rather than individual paired measurements. This is standard practice in limnology (Carlson, 1977; Dillon & Rigler, 1974) and is used here for several reasons. With respect to ecological considerations (see steps 1–3 in Figure 1), there are inherent temporal lags in nutrient–phytoplankton–NOM linkages (e.g., nutrient transformations, phytoplankton growth, reproduction). Furthermore, the autochthonous precursor pool is composed of living, senescing, and dead cells as well as extracellular components and thus, by definition, is best represented by an aggregate matrix. With respect to the human health indexes (Figure 1, steps A–C), although there are acute toxicity concerns related to DBPs (e.g., developmental concerns) and cyanotoxins (e.g., neurotoxicity), the criteria developed here are premised on carcinogenic endpoints (regulatory MCL for TTHMs) and thus fall within the realm of chronic toxicity, which are best represented by metrics of central tendency.

As indicated previously, the gas chromatograph used here for THM analysis incorporated an electron capture detector, and although this detector allows very low detection limits, it can become saturated at high THM concentrations. With the 43-fold dilution used in this study, THM concentrations > 430 µg/L could not be measured. Some samples, particularly summer samples from lakes with high Chl*a* concentrations, exceeded the upper limit of the analysis. Consequently, THMFP means and maximums for those lakes are artificially low because they are based on censored data (i.e., we included these data in the analysis, but the limitations should be understood). These systems are shown in Table 1, specified in various figures, and identified in the text.

Finally, there are two lakes in the current study—Kiamesha Lake and Sleepy Hollow Lake—that are believed to be heavily influenced by allochthonous precursor sources during the growing season as evidenced by relatively high color measurements (unpublished data) indicative of humic and/or fulvic acids. Thus, several of the relevant analyses and plots will present findings from the entire dataset as well as without these two systems (shown in Table 1, specified in various figures, and identified throughout the text).

Linking nutrient enrichment to THMs. Mean epilimnetic (i.e., surface water) concentrations for two primary trophic indexes—TP and Chl*a*—as well as for DOC and THMFP are shown in Table 1. The systems span a relatively broad trophic range with mean phosphorus and Chl*a* concentrations varying by approximately 10- and 40-fold, respectively.

The regression relationship between mean Chl*a* and TP is shown in Figure 3 and indicates that 78% of the variability in mean phytoplankton biomass (as measured by Chl*a*) is accounted for by changes in mean TP concentration. This is consistent with the premise that phytoplankton biomass within these systems is controlled by phosphorus concentrations during the growing season; see Figure 1, step 2.

TABLE 1 System-specific epilimnetic means arrayed by increasing Chla concentration

System Name	Chla—*µg/L*	TP—*µg/L*	DOC—*mg/L*	THMFP—*µg/L*
Skaneateles Lake	0.71 (5)	4.14 (5)	1.4 (5)	61.39 (4)
Canandaigua Lake	2.32 (5)	7.69 (5)	2.8 (5)	96.29 (4)
Seneca Lake	2.36 (4)	9.01 (5)	2.4 (5)	147.11 (5)
Alcove Reservoir	2.43 (5)	5.69 (6)	3.0 (6)	212.93 (4)
Canadice Lake	2.53 (5)	11.37 (5)	2.5 (5)	162.37 (4)
Keuka Lake	2.62 (5)	5.70 (5)	2.7 (5)	138.39 (4)
Kiamesha Lake (1, 2)	3.84 (5)	12.71 (6)	4.1 (6)	350.57 (4)
Cayuga Lake	4.07 (4)	15.82 (5)	2.5 (5)	143.89 (5)
Owasco Lake	4.55 (4)	13.04 (5)	2.5 (5)	118.89 (5)
Hemlock Lake	5.16 (5)	10.22 (5)	2.6 (5)	151.84 (5)
Chenango Lake	5.59 (3)	8.65 (2)	2.9 (4)	207.04 (4)
Otisco Lake	6.34 (5)	16.21 (5)	2.2 (5)	179.35 (5)
Sleepy Hollow Lake (1, 2)	6.74 (5)	20.97 (6)	5.2 (6)	289.41 (5)
Tomhannock Reservoir	6.90 (7)	14.30 (7)	2.9 (7)	193.80 (7)
Conesus Lake	7.37 (6)	28.31 (5)	3.1 (6)	220.95 (6)
Lake Louise Marie	8.50 (5)	19.51 (6)	4.1 (6)	213.54 (5)
Stony Creek Reservoir	9.00 (6)	18.60 (6)	4.6 (6)	338.00 (6)
Honeoye Lake	11.36 (5)	26.90 (5)	3.7 (5)	288.02 (5)
Chadwick Lake (2)	20.96 (5)	27.60 (6)	4.8 (6)	384.63 (5)
Basic Creek Reservoir (2)	22.75 (5)	35.34 (6)	4.5 (6)	361.96 (4)
DeForest Lake (2)	28.49 (5)	47.14 (5)	4.5 (5)	371.05 (4)

1—systems with substantial allochthonous precursor contribution, 2—systems with at least one THMFP measurement exceeding the detector, Chla—chlorophyll a, DOC—dissolved organic carbon, THMFP—trihalomethane formation potential, TP—total phosphorus

Number of samples for each system/parameter is in parentheses, not including replicates. Some THMFP means (see 2) are artificially low because of analytical issues.

The relationship between mean epilimnetic DOC and Chl*a* is shown in Figure 4, and although the relationship is not as predictive as the relationship between Chl*a* and TP, it does indicate a trend of increasing DOC concentrations with increasing trophic state. The relationship between Chl*a* and DOC is not expected to be as predictive as for Chl*a* and TP given that a portion of the DOC is of allochthonous origin and that the autochthonous portion is composed of living as well as senescing and dead phytoplankton material. There are two fairly significant outliers—the two data points in the upper left quadrant representing the two systems discussed earlier (Sleepy Hollow Lake and Kiamesha Lake)—that diminish the predictive strength of the relationship. When the regression is run without these systems, the r^2 improves from 0.44 to 0.66, again suggesting that these systems may warrant exclusion from the relationship. Thus the relationship between DOC and Chl*a* indicates that DOC levels are to some degree governed by phytoplankton abundance within many of the study systems during the growing season; however, there also appears to be a substantial qualitative change in the nature of the DOC with respect to reactivity as reflected in THMFP findings. In summary, taken collectively, findings between phytoplankton biomass and DOC provide evidence in support of step 3 of the conceptual model depicted in Figure 1.

Study findings generally support the premise that THMFP levels are substantially influenced by primary productivity within these systems during the growing season (Figure 1, steps 3 and A). The importance of the autochthonous precursor pool in the generation of THMs is supported by several lines of evidence, including: (1) increased THMFP concentrations with increased trophic state of the study systems (Figure 5), (2) observed correlations between mean concentrations of THMFP and trophic indexes (Figures 6 and 7), and (3) a tendency for THMFP concentrations to increase during the growing season in most of the study systems (Figure 8).

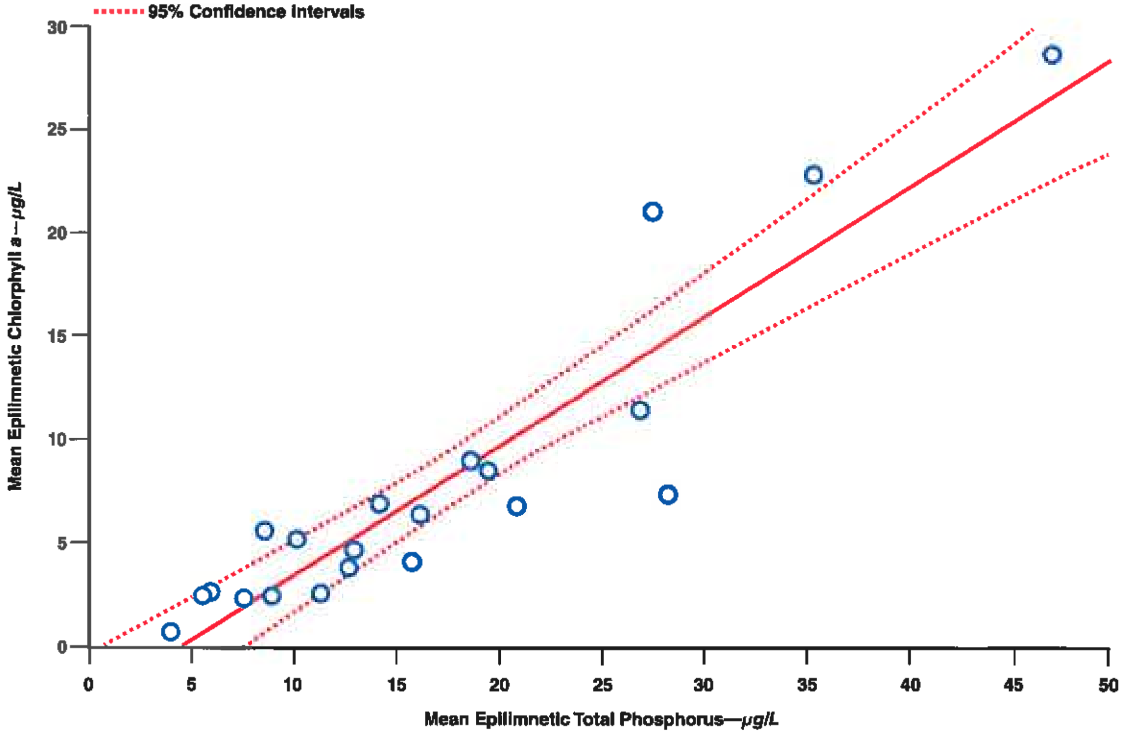
The first line of evidence supporting the premise that autochthonous processes are playing a significant role in the production of TTHMs within these systems is that there is a general tendency toward increasing THMFP levels with increasing trophic state. Figure 5 shows a box-and-whisker plot of epilimnetic THMFP results for the study systems arrayed in order of increasing mean Chl*a* concentration (note both the trend of increasing median concentrations as well as increases in system maxima with increasing Chl*a* concentration). Although there are clearly some exceptions (e.g., Alcove Reservoir, Kiamesha Lake, Sleepy Hollow Lake), there is a relatively strong upward trend in THMFP concentrations (both medians and maxima) with increasing trophic state.

A second line of evidence in support of the THMFP–autochthonous production nexus can be seen in regression relationships between central tendencies for THMFP concentration and major tropic state metrics of phytoplankton abundance (as measured and represented by Chl*a*) and total phosphorus. Figure 6 shows regression relationships between system-specific mean epilimnetic THMFP and Chl*a*, inclusive of both a logarithmic relationship for the entire dataset as well as a linear relationship for a censored dataset. The logarithmic relationship indicates that 62% of the variation in mean THMFP is attributable to changes in mean Chl*a*. Once again, the two systems in the upper left quadrant (representing Kiamesha and Sleepy Hollow Lakes) diminish the predictive strength of the relationship, and exclusion of these two systems might be justified given the substantial allochthonous precursor contribution; removing these systems from the relationship improves the r^2 of the relationship from 0.62 to 0.76.

A second regression (linear regression) is also shown in Figure 6, but this relationship is drawn from a censored dataset that excludes Kiamesha and Sleepy Hollow Lakes as well as the three data points in the upper right quadrant (because of exceeding the limit of the electron capture detector). There are a couple of reasons for presenting this second regression. For example, the logarithmic regression may not accurately represent the true relationship because of the reasons cited previously. The linear relationship is similar to findings from a subsequent companion study (Callinan, 2010) that addressed the issue of exceeding detector limits and therefore is believed to better represent the actual relationship between THMFP and phytoplankton abundance. It is also worth noting that the linear relationship is reasonably similar to the logarithmic curve in the area of the plot where the criteria are likely to be set. Another reason for presenting the linear regression is that the *y*-intercept is suggestive of a possible residual allochthonous precursor pool of approximately 80 µg/L, which is consistent with some other evidence to be discussed later. The relationship between mean THMFP and TP concentrations shown in Figure 7 suggests that 55% of the change in THMFP is attributable to changes in TP.

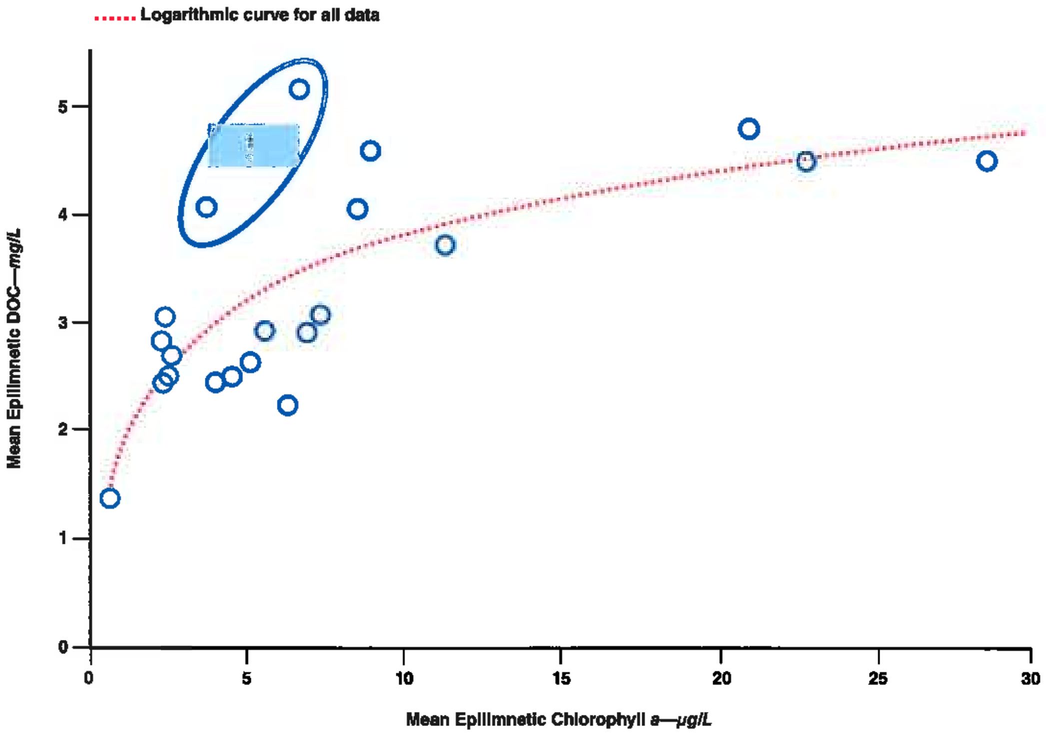
The third line of evidence in support of the importance of autochthonous precursor sources is the observed seasonal increases in THMFP concentrations. Findings indicate that 17 of the 21 systems in the study exhibit a pronounced increase in epilimnetic THMFP levels during the growing season (data not shown). Figure 8, part A, illustrates this point by comparing the late spring (May or June) minimum to the mid-to-late summer maximum THMFP concentrations for each system. Spring minima may represent an allochthonous residual, whereas summer maxima represent the combination of allochthonous residual plus autochthonous contribution with the differential representing the potential autochthonous contribution. In certain systems, mid-to-late summer maxima were three- to fourfold greater than the

FIGURE 3 System-specific mean epilimnetic chlorophyll *a* and total phosphorus concentrations



Mean epilimnetic chlorophyll a = 0.62 (mean epilimnetic total phosphorus) – 2.84; r^2 = 0.78; *P* = 9.29E-08.

FIGURE 4 System-specific mean epilimnetic DOC and chlorophyll *a* concentrations



1—systems with substantial allochthonous contribution, DOC—dissolved organic carbon

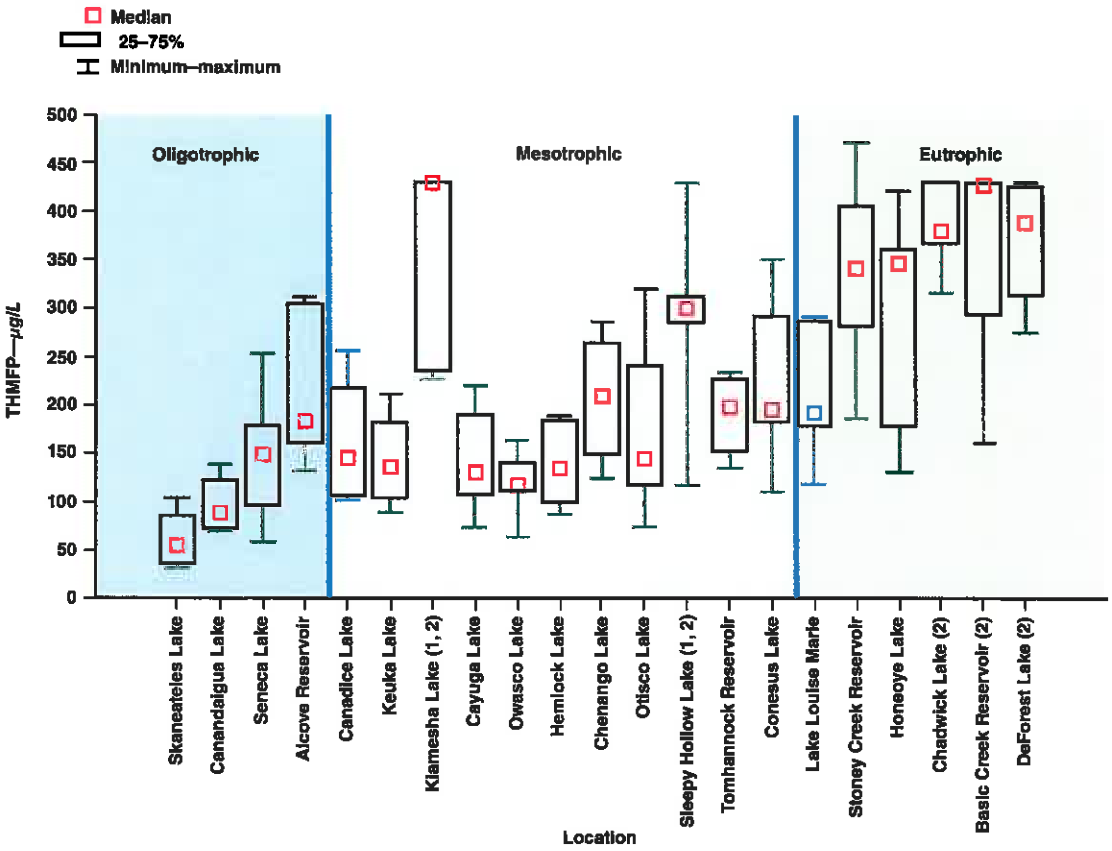
Mean epilimnetic DOC = 2.03 × $\log_{10}$ (mean epilimnetic chlorophyll *a*) + 1.79; $r^2 = 0.44$; $P = 0.0014$

late-spring minima, and all systems showed some degree of increase. This trend in THMFP levels was not merely due to increases in water temperatures (at least directly) because the THMFP method is standardized at 25°C, thus negating direct effects of water temperature on formation potential. Finally, this phenomenon of seasonal increases in DBPs has been reported by other researchers (e.g., Hoehn et al, 1977; Chen et al, 2008) and is a common occurrence at many water treatment plants (WTPs), although this latter situation may be due in part to the direct effects of temperature on the rate of DBP formation.

Figure 8, part A, is also consistent with the notion of a nonautochthonous residual, as indicated by the y-intercept of the linear relationship in Figure 6, in that the spring minimum for most of these systems is anchored around 50–150 µg/L, with only four systems significantly exceeding 150 µg/L. Figure 8, part B, shows a similar spring-minima and summer-maxima plot for DOC. A comparison of these two figures reinforces the notion that (1) there is a marked qualitative change in the nature of DOC in a number of the systems, as evidenced by the more pronounced increase in summer-maxima THMFP than in DOC and (2) that the linkage between THMFP and DOC during the growing season is not simply a reflection of a quantitative change in DOC concentrations, but rather the result of a combination of quantitative and qualitative (e.g., increase in its reactivity) changes in DOC. These findings are consistent with the findings of Kraus et al (2011), who found that "Despite only moderate variation in bulk DOC concentration (3.0–3.6 mg C/L), changes in DOM composition indicated that terrestrial-derived material entering the reservoir was being degraded and replaced by aquatic-derived DOM produced within the reservoir. Substantial changes in the propensity of the DOM pool to form THMs and haloacetic acids . . . illustrate that the DBP precursor pool was not directly coupled to bulk DOC concentration and indicate that algal production is an important source of DBP precursors."

In addition to reasonably robust relationships observed between THMFP and common trophic indexes, results also demonstrate a sound relationship between mean epilimnetic THMFP and DOC within the study systems (Figure 9) and indicate that approximately 80% of the variation in mean THMFP is attributable to DOC. However, as shown in Figure 8, parts A and B, there also appears to be a marked seasonal increase in the relative "potency" of DOC within a number of the systems with respect to the ability to generate THMFP. This is further underscored by

FIGURE 5 System-specific box-and-whisker plot of THMFP arrayed by increasing mean chlorophyll *a* concentration



1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, THMFP—trihalomethane formation potential

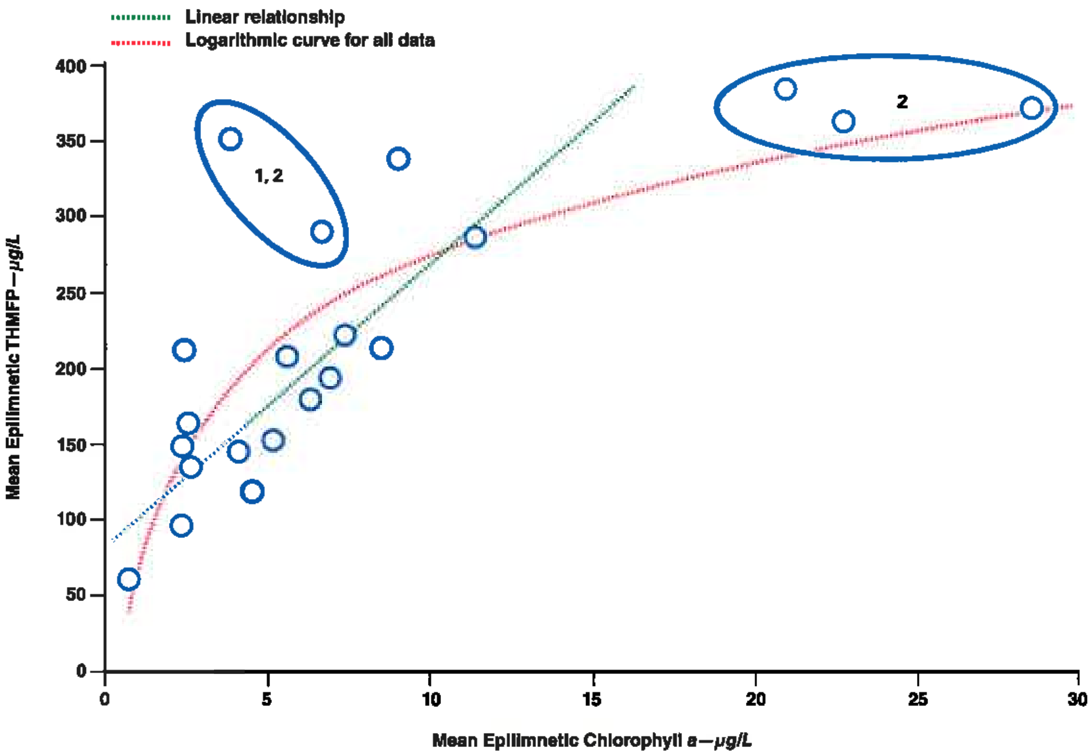
looking at the seasonal change in specific THMFP yield (derived by dividing THMFP by DOC). Figure 10, which compares spring minimum to summer maximum yields, demonstrates a marked increase in yield within a number of the systems as the growing season progresses. Other investigations have also observed large seasonal increases in DBP concentrations, and several have evaluated the relative potency of the autochthonous precursor pool. For example, Graham et al (1998) found THM yields from two species of phytoplankton to be comparable to yields from humic and fulvic acids. Wardlaw et al (1991) also concluded that THM yields from algae were comparable to those from humic and fulvic acids. Nguyen et al (2005) found THMFP-specific yields for three species of phytoplankton that ranged from 30 to 64 µg/mg-C. Finally, Klake (2008) observed an average chloroform yield of 95 µg/mg-C in cultures of 28 algal species. A rough comparison from the present study can be made by taking the difference between the summer/fall maximum and the spring minimum, which results in a THMFP-specific yield range of 14–87 µg/mg-C.

In summary, the relatively strong relationships observed between THMFP and trophic indexes provide a sound basis for the derivation of nutrient thresholds protective of autochthonous-based THM production in these ponded systems. Those threshold levels and the approach taken to derive these numbers are provided in the following section.

Derivation of preliminary NNC. The general approach taken to establish NNC consists of identifying an algal biomass threshold as well as companion TP threshold(s) that would likely prevent exceeding the established regulatory limits for TTHMs from autochthonous precursors; the MCL for TTHMs is 80 µg/L (USEPA, 2006). Recall that DBP precursors can arise from both autochthonous and allochthonous precursors; it is only the former category that can be mitigated by establishment of nutrient-related criteria.

One significant issue inherent in the interpretation of this study's findings is the need to extrapolate the THMFP results into what would be expected under actual treatment/distribution conditions. Although the THMFP procedure is valuable because it provides a uniform set of conditions that enables an objective source-to-source comparison, the procedure is more extreme than conditions present in most PWS treatment/distribution

FIGURE 6 System-specific mean epilimnetic THMFP and chlorophyll *a*



1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, THMFP—trihalomethane formation potential

Mean epilimnetic THMFP = 206.97 × $\log_{10}$ (mean epilimnetic chlorophyll *a*) + 67.46; $r^2 = 0.62$; $P = 2.24\text{E-}05$

Linear relationship excludes both circled sets of data.

systems and thereby overestimates expected TTHM concentrations in actual PWS systems. Therefore, it is necessary to establish a plausible approach to extrapolate the THMFP findings to what would be reasonable to expect under real-world conditions. The approach taken to accomplish this extrapolation involves two main steps.

First step. The first step is to fit the observed data to an off-the-shelf TTHM simulation model. The simulation model used for this exercise is from Rodriguez et al (2000) and was applied without modification

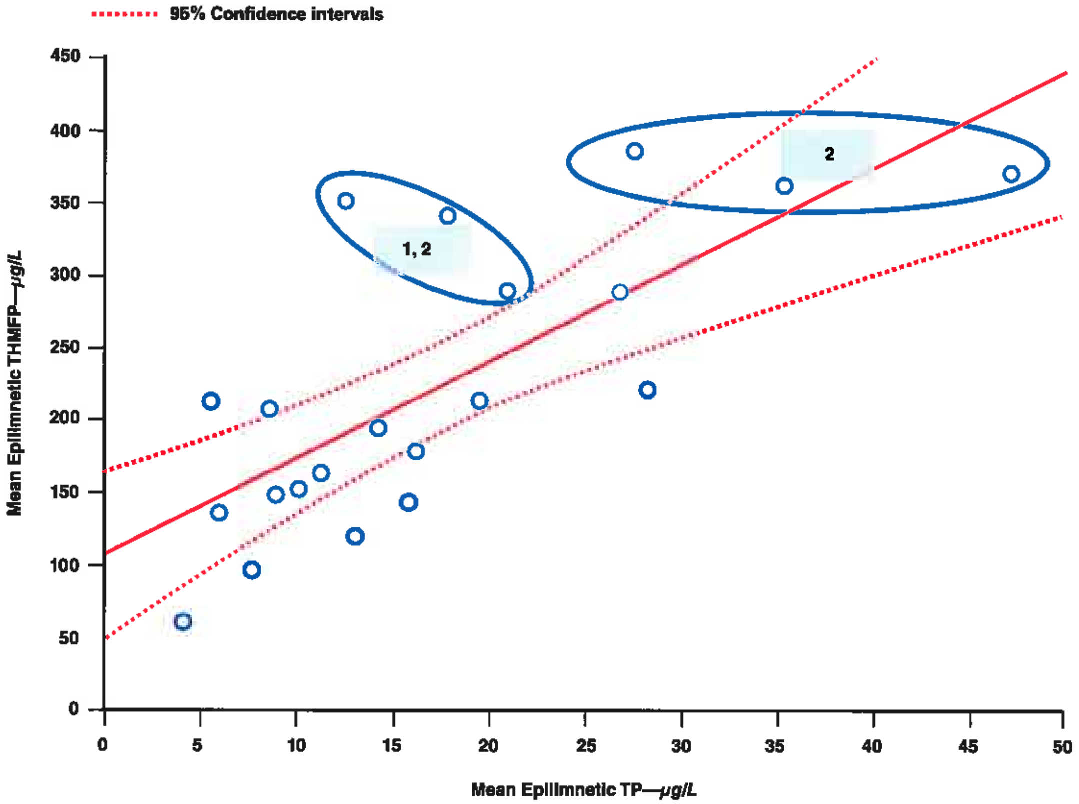
$$[\mathrm{TTHM}] = 0.044\,(\mathrm{DOC})^{1.030} \times (\mathrm{Time})^{0.262} \times (\mathrm{pH})^{1.149} \times (\mathrm{Dose})^{0.277} \times (\mathrm{Temp})^{0.968} \qquad (1)$$

in which [TTHM] is the concentration of TTHMs (µg/L), DOC is dissolved organic carbon concentration (mg/L), time is disinfection contact time (h), pH is designated hydrogen ion concentration, dose is designated disinfectant dose (mg/L), and temp is water temperature (°C). The model provides a reasonably good fit to observed data when parameterized per the THMFP method specifications as follows: time of 168 h (seven days), pH of 7.0, chlorine dose of 5.0 mg/L, water temperature of 25°C, and observed system-specific mean epilimnetic DOC concentrations. A comparison of the modeled THMFP values to the actual observed THMFP values results in an r^2 of 0.79 with no modification of the underlying model algorithm.

Second step. The second step involves running the simulation model under more representative treatment/distribution system conditions. Residence time is set at 72 h (three days) based on a USEPA report (2002). Source water pH is set at 7.8, chlorine dose is set at 1.0 mg/L, and temperature is set at 20°C based on Summers et al (1996). These values are believed to represent realistic but protective conditions. The model is then run using the MCL of 80 µg/L, and the equation is solved for DOC. This exercise results in a DOC = 3.0 mg/L and could be thought of as the "critical" DOC concentration.

The critical DOC threshold is then used to establish a "critical" THMFP ($\mathrm{THMFP}_{\mathrm{cr}}$) concentration based on the observed

FIGURE 7 System-specific mean epilimnetic THMFP and total phosphorus



1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, THMFP—trihalomethane formation potential, TP—total phosphorus

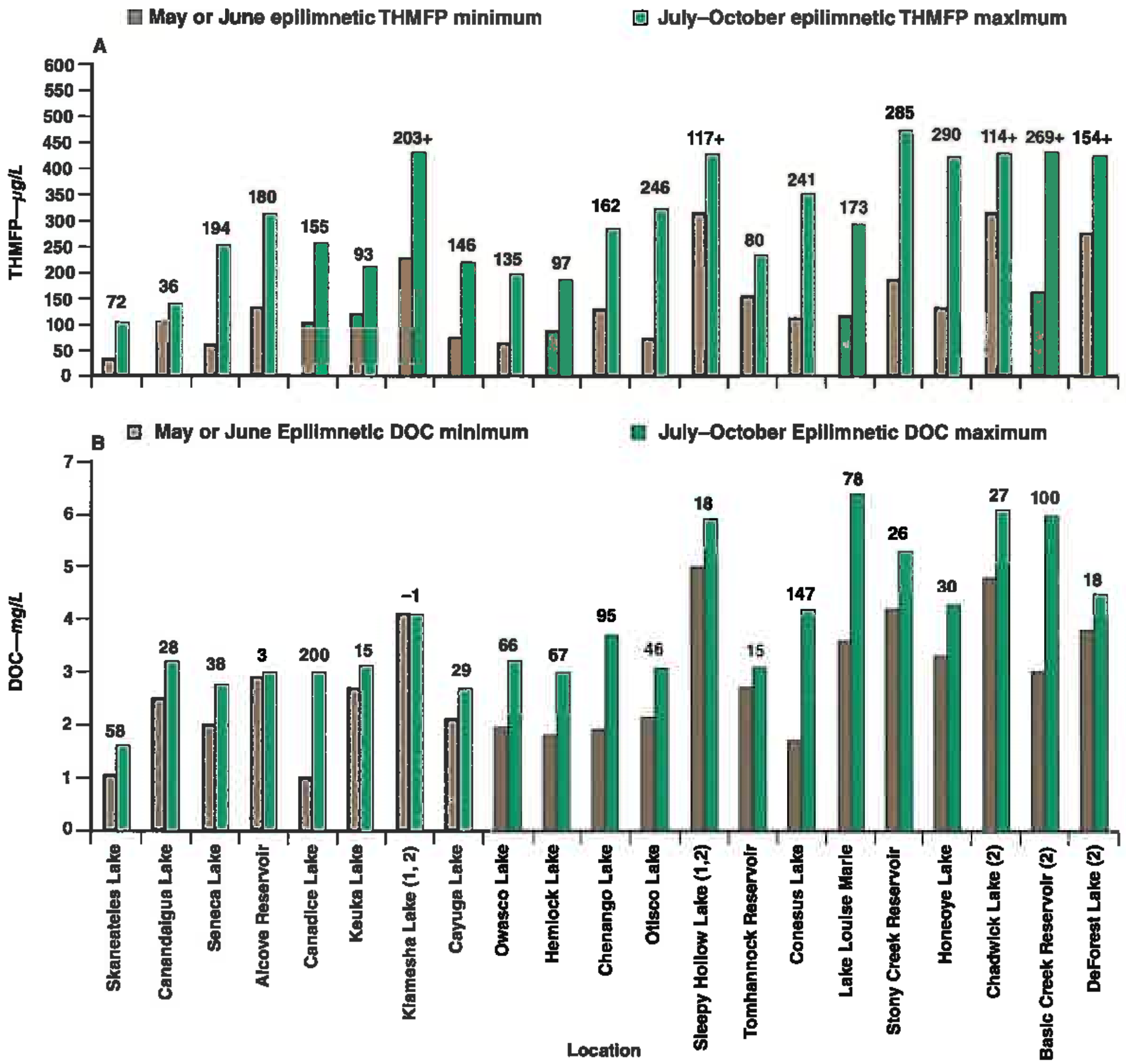
Mean epilimnetic THMFP = 5.67 (mean epilimnetic total phosphorus) + 106.34; *r*2 = 0.55; *P* = 0.00013.

relationship between THMFP and DOC (Figure 9). This results in $THMFP_{cr}$ of 190 µg/L. The $THMPF_{cr}$ is then used together with the respective relationships between mean THMFP and mean trophic metrics (Chl*a* and TP; Figures 6 and 7, respectively) to establish trophic metric thresholds by solving the requisite regression equations for $THMFP_{cr}$. This results in the following thresholds: Chl*a* = 4.0 µg/L (based on the logarithmic relationship shown in Figure 6) and TP = 12 µg/L. Returning to Figure 5, it can be see that the thresholds for THMFP and Chl*a* are quite consistent with the figure (absent the exceptions previously noted for Kiamesha and Sleepy Hollow Lakes) in that median THMFP levels generally exceed 180–200 µg/L beginning with Chenango Lake and that this corresponds with a mean Chl*a* of approximately 4–5 µg/L.

The Chl*a* criterion of 4 µg/L is considered appropriate for New York state waters carrying an AA classification, given that these waters are required to be acceptable for consumption following disinfection only. Somewhat less restrictive criteria are deemed appropriate for class A waters because of the expectation that they should meet appropriate drinking water standards after conventional water treatment defined as "treatment equal to coagulation, sedimentation, filtration, and disinfection." The approach used to derive these less restrictive criteria was to adjust the more restrictive class AA criteria by a percentage representative of the DOC removal efficiency of a conventional WTP. A review of available literature suggests that conventional WTPs can attain DOC removal efficiencies in the range of 10–79% (Volk et al, 2000; Randtke, 1988). However, cultural eutrophication will tend to diminish the DOC removal efficacy of WTPs. For example, Cheng and Chi (2003) observed an inverse relationship between degree of eutrophication and removal efficiency, and Takaara et al (2007) observed that DOC removal efficiency declined as cyanobacteria levels increased. Given these findings, a conservative DOC removal efficiency of 10% was assumed. This is a reduction in DOC, rather than phosphorus or chlorophyll. Target criteria for TP and Chl*a* are derived by using the adjusted target source water DOC of 3.3 mg/L, which equates to THMFP of 220 µg/L. This in turn equates to a total phosphorus concentration of 17 µg/L and a Chl*a* concentration of 6.0 µg/L.

It could be contended that crafting the criteria to address summer peaks may be overly protective given the chronic nature of the concern; however, there are several factors that counter this line of

FIGURE 8 System-specific epilimnetic spring minimum and summer maximum THMFP (part A) and spring minimum and summer maximum DOC (part B)



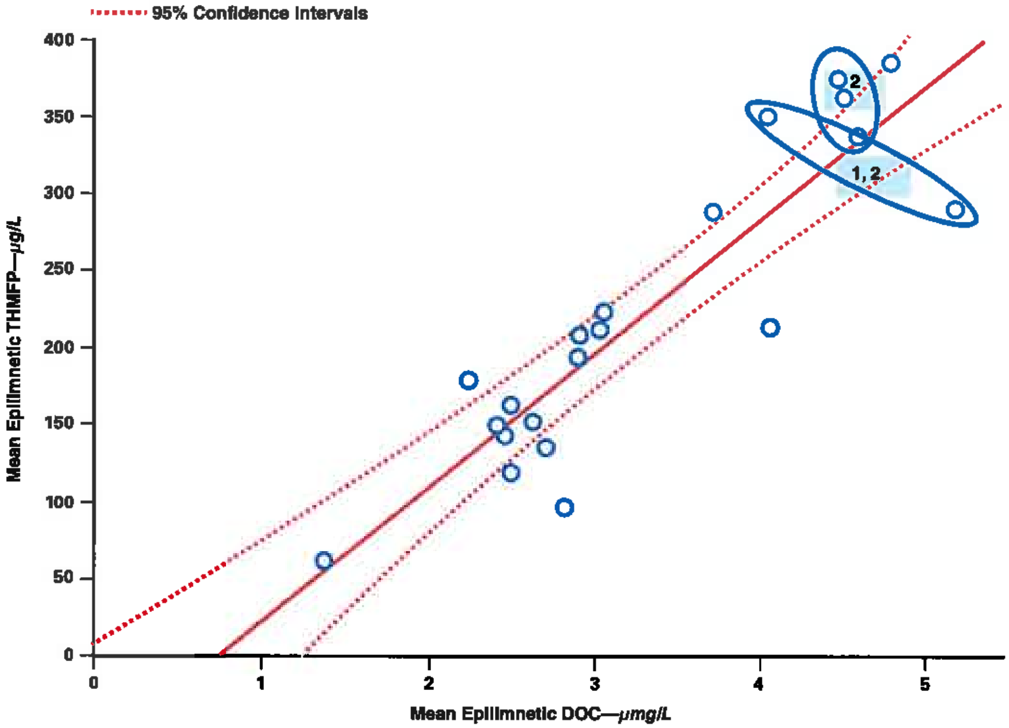
1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, DOC—dissolved organic carbon, THMFP—trihalomethane formation potential

Inset numbers represent percent increase relative to spring minimum.

reasoning. For example, findings from the present study indicate that the seasonal peaks in THMFP can linger well into the fall, suggesting that the effects of the autochthonous precursor pool extend beyond the conventional growing season. In addition, climate change is expected to extend the growing season substantially. Finally, there is also an evolving body of literature concerning possible links between DBP exposure and risks of adverse reproductive and developmental effects, that fit more appropriately in the realm of acute toxicity effects. For example, a recent study by Hwang and Jaakkola (2012) assesses possible links between TTHMs and the risk of stillbirth and concludes that the risk is related to prenatal exposure to TTHMs. On a related note, cyanotoxins include both chronic and acute risks; thus, there is a need to remain cognizant of shorter-term exposures to TTHMs as well.

Although specific criteria thresholds for both the stressor variable (TP) and the response variable (Chl*a*) are presented, New York is considering an implementation approach that would place primacy on the response threshold. The approach would establish a fixed Chl*a* criteria but would consider a range of TP criteria dependent on the stressor-response relationship

FIGURE 9 System-specific mean epilimnetic THMFP and DOC



1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, DOC—dissolved organic carbon, THMFP—trihalomethane formation potential

Mean epilimnetic THMFP = 85.95 × (mean epilimnetic DOC) – 62.20; r^2 = 0.81; P = 4.13E-08

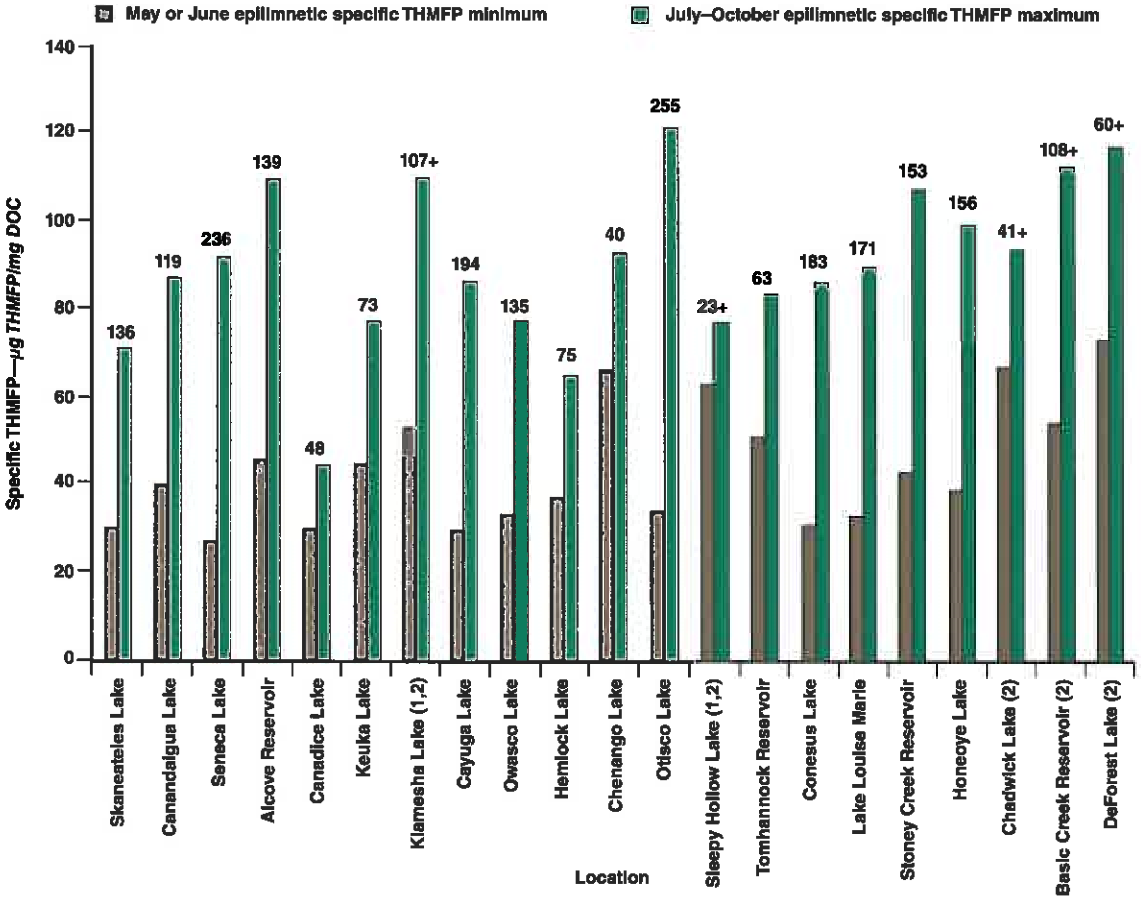
and associated uncertainty bands as well as lake- or reservoir-specific water quality conditions. The reasoning for this response variable–centric approach is that the designated use (water supply) is more closely linked to the response variable (algal biomass) and that there is substantial natural variability in the stressor-response relationship. The approach being considered is somewhat similar to the USEPA proposal for the derivation of NNC for Florida lakes (USEPA, 2009).

Ground-truth and corroboration. Several preliminary ground-truth exercises were also undertaken to investigate whether the proposed criteria struck the appropriate balance between source water protection and reasonable allowances for nutrient load. The exercises involve review of historical TTHM data for individual WTPs coupled with available trophic state data for the applicable source waters.

The first case involves a PWS with source water that has a mean Chl*a* concentration in the 7–9 μg/L range, which is slightly above the proposed class A criteria and moderately above the class AA criteria. This PWS includes treatment beyond disinfection and serves several adjacent communities (or "purchase" systems). Figure 11 shows the TTHM rolling annual average as reported in the systems' annual consumer confidence reports for the primary community as well as two purchase systems. The plot indicates that both the primary and purchase systems have at times struggled with meeting the current MCL for TTHMs, although to differing degrees.

The second case study involves a PWS (which includes treatment beyond disinfection) with source water that has experienced a substantial increase in Chl*a* over the past several years, moving from levels very near the proposed criteria to levels moderately above the proposed criteria (4–6 μg/L). Figure 12 shows temporal trends in both mean epilimnetic growing season Chl*a* and third-quarter TTHMs (these values would not constitute a violation of the MCL on their own because they are third-quarter values and not annual averages). There are several noteworthy elements in this figure, such as the fairly synchronous trends in the two curves, most significantly the marked increase in Chl*a* and TTHM from 2005–06. In addition, this same "event" represents a substantial departure relative to both the proposed Chl*a* criteria and the TTHM MCL (although, again, the MCL does not directly apply). In summary, these preliminary ground-truth exercises show that the proposed criteria strike the appropriate balance between source water protection and acceptable nutrient loadings, and that the proposed criteria should provide adequate protection of source water supplies with respect to autochthonous precursors.

FIGURE 10 System-specific epilimnetic May or June minimal versus July–October maximal specific THMFP yield



1—systems with substantial allochthonous contribution, 2—systems in which some THMFP samples exceeded detector, DOC—dissolved organic carbon, THMFP—trihalomethane formation potential

Inset numbers represent percent increase relative to spring minimum.

Although the current study has compiled a substantial body of information with which to establish appropriate NNC for PWS lakes and reservoirs, it is important to compare these findings with other independent investigations. Thus, several other studies were reviewed to assess the relative comparability of the present study to the findings of other researchers.

A study by Arruda and Fromm (1989) of 180 lakes in Kansas investigated the relationship between trophic indexes and DBPs and arrived at remarkably consistent conclusions to the present study. Findings from the Arruda and Fromm investigation indicate that a trophic state index of 45, corresponding to a Chl*a* threshold concentration of 5 μg/L, would be required to meet 0.1 mg/L (100 μg/L) TTHM, which was the standard in place at the time of their study.

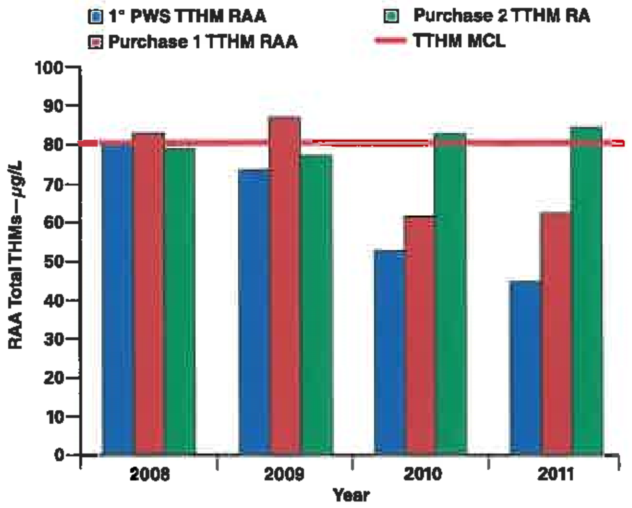
Colorado conducted a subsequent study similar in nature to the New York study and determined that a Chl*a* concentration of 5 μg/L would be an appropriate threshold for direct-use public water supply source water reservoirs (Colorado DPHE, 2011). The Colorado study did include a number of enhancements and somewhat different approaches to the derivation of threshold criteria, yet the two studies arrive at very similar endpoints. Of particular note is that the Colorado study used a more directly applicable analytical method termed Uniform Formation Condition developed by Summers et al (1996) that eliminated the need to extrapolate THMFP findings to TTHMs.

CONCLUSIONS AND RECOMMENDATIONS

Although findings and recommendations from the current study remain a work in progress, initial findings suggest that autochthonous precursors contribute substantially to the DBP precursor pool in lakes and reservoirs and that the establishment of NNC for the protection of PWS source waters is warranted and feasible. In addition, findings suggest that an algal biomass threshold (as measured by mean Chl*a*) on the order of 4–6 µg/L would be sufficient to attain the existing TTHM MCL of 80 µg/L in systems in which autochthonous precursors predominate. Furthermore, the relative ability to both control ambient autochthonous precursor levels (e.g., through nutrient controls and implementation of NNC) and the relative inability to effectively remove this same pool of precursors suggest that nutrient management may be one of the most efficacious approaches for the control of DBPs that arise from the use of lake and reservoir source waters. Implementation of these criteria could also be expected to pay additional dividends in terms of limiting the production of certain unregulated DBPs, cyanotoxins, and the mobilization of sediment-bound arsenic. Finally, although the proposed criteria are intended to enable attainment of existing TTHM regulatory criteria, it is important to stress that even lower levels of nutrients would provide additional protection.

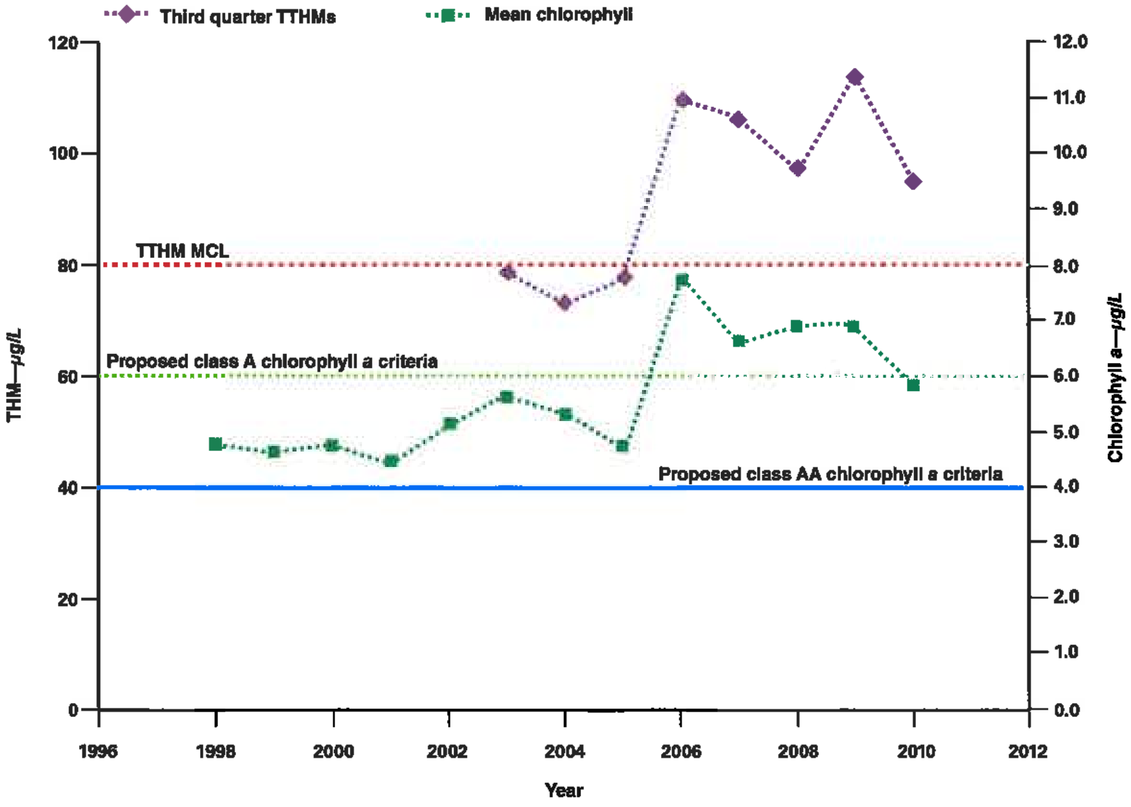
Findings from the present study provide a sound basis for establishing NNC targeted at the protection of lakes and reservoirs used for public water supply. These findings are also consistent with findings from other studies (Colorado DPHE, 2011; Arruda & Fromm, 1989). However, given the importance of these issues to the protection of public health, additional study is likely warranted, and it is recommended that a comparable nationwide study of both ponded and flowing PWS source waters be undertaken. Further-

FIGURE 11 Running annual average TTHMs for PWS and purchase systems for ground-truth case 1



MCL—maximum contaminant level, PWS—potable water supply, RAA—running annual average, TTHM—total trihalomethanes

FIGURE 12 Third-quarter TTHMs and mean epilimnetic growing season chlorophyll *a* (number of chlorophyll *a* samples between 8 and 10 per year)



MCL—maximum contaminant level, PWS—potable water supply, RAA—running annual average, TTHM—total trihalomethane more, this effort should draw on existing ambient water quality and PWS datasets at the local, state, and national level. Collectively, these studies would help advance a long overdue bridge between SDWA and CWA programs in an effort to better protect public water supply sources.

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FOOTNOTE

[1] Supelco #48140-U methanol, Sigma-Aldrich, St. Louis, Mo.

PEER REVIEW

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