Environmental Science & Technology
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Prediction of Reference Phosphorus Concentrations in Swedish Lakes

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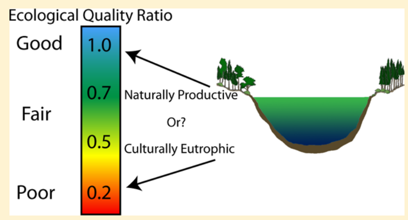
S *Supporting Information*

ABSTRACT: The relationship between total phosphorus (TP) and chemical, climatic, morphological, and geographic variables was examined for over a thousand reference lakes across Sweden. A significant relationship was found between TP and both absorbance of irradiance (at 420 nm, filtered) and altitude for all lakes. These two variables alone, however, were not able to adequately predict TP concentrations in naturally turbid lakes. A natural particulate matter factor (PM_n) was developed as part of this study to incorporate the effect of natural suspended matter on lake phosphorus concentration. Variability in TP concentration was well explained with the addition of PM_n to the model (R^2 = 0.71) even though conditions external and internal to the lakes varied greatly. The ability of the three parameter model to identify culturally eutrophic systems was then successfully tested using a data set of lakes with known anthropogenic phosphorus loads. Thus, the model allows for estimation of reference TP concentration and by extension can be used to identify when a reference concentration has been exceeded due to anthropogenic phosphorus loading. The model output also provides a realistic end point to which phosphorus concentrations should be reduced to achieve a natural trophic state in both clear water and naturally turbid lakes.



INTRODUCTION

Development of water quality standards, based on reference conditions, is an ongoing process but the prediction of reference values for lakes is problematic due to the limited number of natural lakes and inadequate modeling techniques currently available. The European water framework directive (EUWFD) requires member countries to set environmental objectives for the ecological quality of surface water bodies. Surface water quality is quantified as the ratio (ecological quality ratio, EQR) of a reference value to an observed value for a given metric. Thus, a baseline or reference condition is essential but predictions of reference condition for lakes generally have high variance or apply only to specific lake types. Total phosphorus (TP) concentration is commonly used as an indicator of water quality for lakes because it correlates well with chlorophyll (i.e., phytoplankton biomass) and water clarity.[1,2] While TP is a simple and valid indicator of trophic status, adopting rigid and subjective trophic boundaries are not likely to provide realistic goals for management. Instead, a predicted reference TP concentration, based on quantifiable characteristics of a given lake, should be used to determine the ecological quality of a lake.

Reference concentrations for TP in natural or minimally disturbed lakes have been estimated by a number of different methods. Vighi and Chiaudani[3] used a morphoedaphic index (MEI) to predict background TP concentrations in lakes. The MEI is estimated from a ratio of conductance or alkalinity to mean depth (Z_m). This method is sound conceptually because phosphorus (P) in natural lakes is generally controlled by P sources form natural weathering, which correlate with the concentration of ionic dissolved solids.[4,5] The data set used to develop the relationship between MEI and TP, however, was composed of mostly North American, deep water lakes that are likely low in natural particulate matter. No information was provided on humic status or other variables (except TP, alkalinity, conductivity, and Z_m) that may influence natural levels of TP in lakes. Cardoso[6] used altitude, alkalinity, and Z_m (both independently and as a MEI) to predict reference TP concentrations for humic (elevated concentration of humic substances and organic acids) and nonhumic lakes. Although both shallow and deep lakes were included in the model data set, lakes with TP concentrations greater than 35 μg/L were excluded. The authors acknowledged some of the study lakes may have been naturally eutrophic but used the OECD[7] classification scheme as a subjective definition of low ecological quality. The Swedish environmental protection agency (SEPA) used two variables (absorbance of irradiance and altitude) to develop a simplified predictive model for TP reference concentrations in Swedish lakes[8] based on the relationship between TP and absorbance in natural lakes (Meili).[9] The model, developed using 148 reference lakes, explains a substantial portion of the variability in TP between lakes but tends to underestimate reference concentrations in lakes with TP concentrations greater than 15 μg/L. The authors attributed this difference to potential effects from unknown anthropogenic nutrient sources.

Received: October 8, 2012
Revised: January 28, 2013
Accepted: January 28, 2013
Published: January 29, 2013

None of the predictive models in previous studies incorporate variables that can directly account for the influence of natural particulate matter on P concentration in lakes. Because particulate matter in water contains P,[9] use of such models increases the likelihood for erroneous determination of degraded state. Application of such models to data sets including shallow lakes, which are more likely to be naturally turbid, has resulted in poor explanation of TP variability (29%[10] and 51%[6]). Locating natural lakes with which to develop a model capable of predicting a reference state is also problematic. In Sweden, however, there are nearly 100,000 lakes (greater than 1 ha in size) with adequate water chemistry, climatic, and land use data for modeling purposes. Many of these lakes are not directly affected by anthropogenic P loading. In this study, we derive reference conditions for TP based on a large data set of randomly sampled lakes (2402) across Sweden. We hypothesize that inclusion of naturally derived particulate matter will improve TP concentration prediction, especially for shallow lakes that are common throughout Sweden and other regions of the world. Thirty seven variables including chemical, climatic, morphologic, and geographic data were used to develop a model describing TP variation in the reference lakes. The model was then applied to lakes with known anthropogenic P loads to determine the potential for identifying culturally eutrophied lakes.

METHODS

Data Collection and Handling. All lake data were collected as part of the national monitoring program in Sweden and downloaded from the Department of Aquatic Sciences and Assessment at the Swedish University of Agricultural Sciences (Supporting Information, Table S1, http://www.slu.se/vatten-miljo). One surface sample was collected for each lake and sampling occurred during autumn circulation between September and November (temperatures generally ≤8 °C). The samples were taken from the center of the lake during autumn circulation because a surface sample is generally representative for the lake as a whole during this period.[11] Lake water chemistry data were analyzed at the same laboratory using analytical methods accredited by the Swedish Board for Accreditation and Conformity Assessment (SWEDAC). Absorbance of irradiance at 420 nm, both filterd (AbsF) and unfiltered (AbsUF), were used because these measurements have been standard in national monitoring since the 1960s. Elevation, also included the data set, is hereafter referred to as altitude (Alt) because it is designated as such in the database and other studies detailing the prediction of reference TP concentrations.[6,8] Further details on methods, detection limits, quality control, and other information can be found at the Web site referenced above. Temperature and precipitation data were collected from the Swedish Meteorological and Hydrological Institute (see Huser et al.[12] for details), and 10-year means for temperature, precipitation, and runoff were calculated using these data (Supporting Information, Table S2).

2410 sampling events were available from randomized national lake surveys conducted from 2007 through 2009. Because reference conditions were the focus of this study, only sites with no or minimal anthropgenic disturbance were included in model development. To satisfy this requirement, lakes were only included if they had: no urban areas, less than 1% agricultural land, and no point source pollution within the watershed. Fifteen lakes were excluded because TP was less than soluble reactive P. This discrepancy was likely due to measurement error when P values were near the reporting limit (1 μg/L). 458 lakes were excluded from analysis because they are affected by acid deposition or liming activities. An additional 181 lakes were excluded because they have anthropogenic nutrient sources within the watershed. These lakes were used as a test data set to determine the ability of the model to detect degraded lakes. After removal of the above sites and values with probable measurement error, 1100 different lakes remained in the reference data set (Supporting Information, Figure S1). One of the 1100 sites was considered an outlier due to elevated TP concentration (385 μg/L) relative to other lakes in the ecoregion.

Previously defined ecoregions were used to divide the country into seven regions (Supporting Information, Figure S1) based on natural geographic differences, the highest coastline elevation, and elevations greater than 200 m above sea level.[13] All ecoregions, except for the alpine ecoregion 7, are dominated by forest (>50%) with some wetland and open water areas (approximately 20%). The total number of sites within each region varied (Supporting Information, Table S3) and ecoregion 5 contained only five reference sites due to the prevalence of agriculture in this region. Data from the national lake inventory program from 2000 and 2005 were then matched with the reference lake data set to determine if mean TP concentrations from multiple years (N = 3) were more useful in model development compared to a single data point for each lake.

Calculated Variables. A number of calculated variables were used in the analysis (Supporting Information, Table S2) including Z_m and lake volume,[14] residence time, P retention (Rpred = 15/(18 + annual water load)),[15] MEI_{alk} (Alkalinity/Z_m),[3] and the morphometric Osgood Index (mean depth/(lake area)$^{0.5}$).[16] Marine corrected calcium (Ca*), magnesium (Mg*), and base cations (BC*) were also estimated.[17] A natural particulate matter factor (PM_n) was developed as part of this study using the difference between filtered absorbance (AbsF) and unfiltered absorbance (AbsUF) at 420 nm, also designated as the absorbance differential at 420 nm.

Statistical Analysis. Most data were log10 transformed, except for pH and variables with binomial distribution, to normalize distributions and reduce skewness. Thirty seven variables were initially included in the data set for analysis using partial least-squares regression (PLS) with SIMCA+ statistical software (Umetrics V. 12.0.1.0). A set of common variables, most important to the PLS projection, was then used in multiple linear regression (MLR) analysis. Further description of PLS analysis is included in the Supporting Information (Methods). Before stepwise MLR was conducted, however, four variables with high bivariate correlation (>0.7) were excluded from analysis to reduce instability in the matrix inversion (Supporting Information, Table S1). Stepwise MLR analysis was performed by ecoregion to determine variables best suited for TP prediction. Variables were added to individual ecoregion models until p > 0.05. A general model was then developed for all lakes in the reference lake data set using the variables common to the ecoregion models. The common model was then compared to randomly generated data (N = 100) from the reference data set for model verification.

RESULTS

General differences detected between ecoregions in the reference data set (Supporting Information, Table S3) included lower TOC in lakes in northern Sweden (ecoregions 1−3), higher pH, Alk, and Alt, and lower AbsF when comparing regions 1 and 2 with southern Sweden (ecoregions 4−7). Total phosphorus ranged from 1 to 55 μg/L for all lakes and varied by ecoregion (Figure 1), ranging from a median of 3 μg/L in

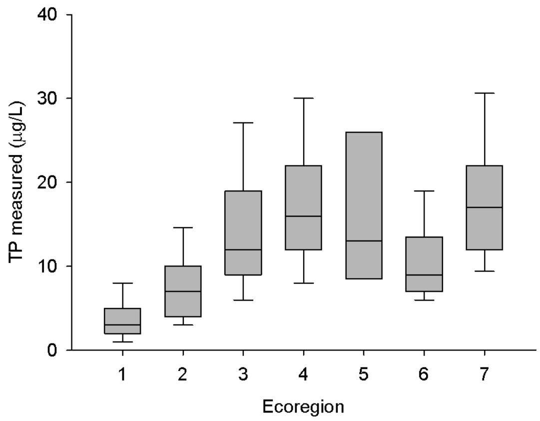


Figure 1. Median TP concentration, 25th and 75th percentiles, and 10th and 90th percentiles (whiskers) by ecoregion in Sweden.

alpine areas (ecoregion 1) to 17 μg/L in southern Sweden (ecoregion 7). Median TP concentrations were pairwise tested by ecoregion using the nonparametric Wilcoxan method on ranks. Statistical differences ($p < 0.05$) were detected for TP between most ecoregions except between ecoregions 3 and 7 and ecoregions 4 and 7.

Model Development. Analysis of the reference lake data set ($N = 1099$) using PLS revealed a set of important predictor variables for TP. Dominant factors were determined by summing occurrence as a strong predictor (VIP > 0.8) in each ecoregion, excluding ecoregion 5 (only five lakes). The most common variables were PM_n, AbsF, AbsUF, Fe, TOC, and Z_m (Supporting Information, Tables S1 and S2). Twenty-one variables were included in stepwise MLR. Four of these variables, however, were removed due to multicollinearlity, leaving 17 variables. Stepwise MLR for all lakes resulted in a model with 10 significant ($p \leq 0.05$) variables that explained 74% of variability in TP for the entire data set (Table 1). Stepwise models developed for each individual ecoregion varied with the most dominant and strongest predictors being PM_n, AbsF, and Alt (Table 1).

A simplified, general model was then developed for all lakes using the most common factors from the individual ecoregion MLR analysis and a variable inclusion threshold limit of at least 2% variance explained. The resulting model (Table 2) included PM_n, AbsF, and Alt and explained 71% of the variability in the

Table 2. Model Parameter Estimates (Based on Log10 Values) and R^2 Values for Models Developed for All Lakes in the Reference Lake Data Set

model	R^2	intercept	AbsF	Alt	PM_n	FePart
model 1 (APA model)	0.71	1.91	0.15	−0.11	0.35	
model 2	0.61	1.63	0.31	−0.16		
model 3	0.62	1.27	0.22	−0.15		0.10
model 3b	0.57	1.13	0.17	−0.15		0.10

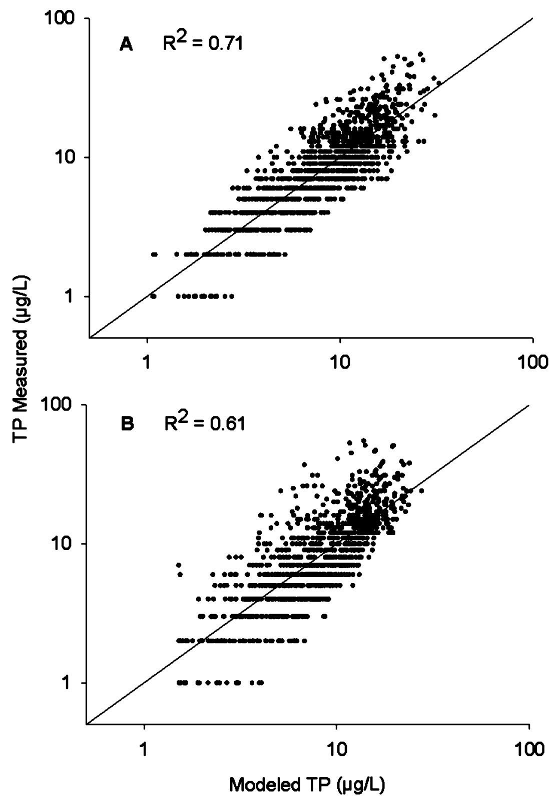


Figure 2. Relationships between measured TP and modeled background TP using Model 1 (the APA model, A) and Model 2 (B).

reference lake data set (Figure 2A). For comparison purposes, we applied a previously developed model (Model 2)[18] with

Table 1. Models Developed Using All Available Variables[a]

ecoregion	R^2	PM_n	AbsF	Y	X	Alt	OI	Mg	Na	Fe	SO_4	F	Alk	R^2 (APA model)
1	0.75	+	+	+			+							0.70
2	0.61	+	+		+		+		−					0.59
3	0.54	+	+			−					−			0.52
4	0.65	+	+			−		+	+					0.55
5	NA													0.98
6	0.58	+	+	+		−								0.47
7	0.42	+	+											0.43
all lakes	0.74	+	+	+	−	−	−		+	−		−	+	0.71

[a] + or − indicates sign of estimate. Significance for inclusion of variables was $p \leq 0.05$. APA model R^2 for each ecoregion is shown as well.

dx.doi.org/10.1021/es3040413 | Environ. Sci. Technol. 2013, 47, 1809−1815

AbsF and Alt as the only variables (Figure 2B). Because Models 1 and 2 differed only by the natural particulate matter factor developed herein, we were able to assess the impact of this variable on predictive capability. Model 1, or the *A*bsorbance, *P*articulate matter, *A*ltitude (APA) model, was then applied to each individual ecoregion in Sweden (Supporting Information, Figure S2).

Model Improvement Due to Inclusion of Natural Particulate Matter. To determine where predictive capability improved within the data set, three particulate matter levels were developed using the median and 10% and 90% quantiles for PM_n. These levels were used to classify lakes as moderately turbid, nonturbid, or turbid (Supporting Information, Table S4). TP concentration varied between each of these levels, increasing as natural particulate matter increased. A comparison between residuals for the APA model (with PM_n) and Model 2 (without PM_n), using the Mann−Whitney rank sum test, revealed a statistically significant shift in median toward zero ($p < 0.001$), and 25% and 75% quartiles were more evenly distributed around zero in the turbid lakes using the APA model (Figure 3). There was little difference between the APA model and Model 2 for the moderately turbid lakes but the median of residuals shifted significantly toward zero ($p < 0.001$) and quartiles were more centered around zero for the nonturbid lakes. Inclusion of PM_n improved model predictability for both turbid (reducing model underestimates) and nonturbid lakes (reducing model overestimates).

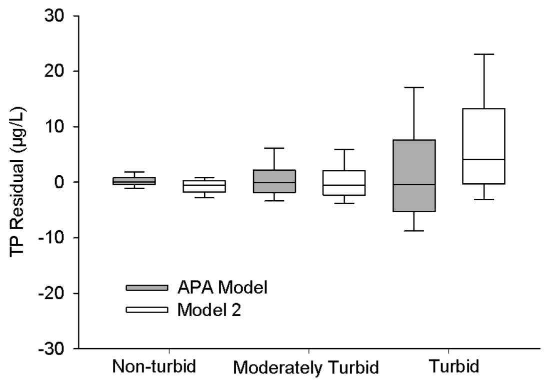


Figure 3. TP residuals for the APA model (Model 1) and Model 2 by natural particulate matter (turbidity) level.

Model Validation and Application. To validate the APA model, simulations were conducted on randomly generated data sets (100 sets of 100 lakes) created from the original data set. The simulation results were good, with a standard deviation for R^2 of 0.043. In order to determine if a single TP measurement from each lake was suitable for model development, we compared modeling results using single measurements with mean concentrations for lakes that had three values for TP. Additional data came from national lake inventories (2000 and 2005). Because AbsUF was not measured in the earlier sampling periods, PM_n could not be calculated and it was necessary to use particulate Fe to compare sample size effects on modeling. Particulate Fe was used because it correlates well with naturally formed particles in natural systems,[19] is related to particulate P content,[20] and facilitates P complexation with dissolved organic carbon.[21] The empirical relationship developed by Köhler[22] was used to estimate dissolved Fe as a function of pH and total Fe (r^2 of relationship = 0.94). Particulate Fe was then calculated as the difference between dissolved total Fe. Particulate Fe correlated with PM_n ($r^2 = 0.62$) and was substituted for PM_n in the APA model (Model 3, Table 2). A comparison of TP models using PM_n (APA model) and particulate Fe (Model 3) revealed a strong 1:1 relationship (Supporting Information, Figure S3). Similar results were found (Table 2) when comparing models based on only one sampling period (Model 3) and the mean of three sampling periods (Model 3b). Thus, inclusion of additional data did not improve model development in this case.

The APA model was applied to the portion of the data set with known point sources of P within the tributary watershed (Figure 4). Ecological quality ratios (EQR, background TP/measured TP) were then used to categorize the level of degradation in quality caused by anthropogenic P loading. Of the 181 sites, 36 sites were elevated above model predictions with an EQR less than 0.5 and 42 sites had EQRs between 0.5 and 0.75. 100 sites were near predicted TP concentrations (i.e., EQR > 0.75),

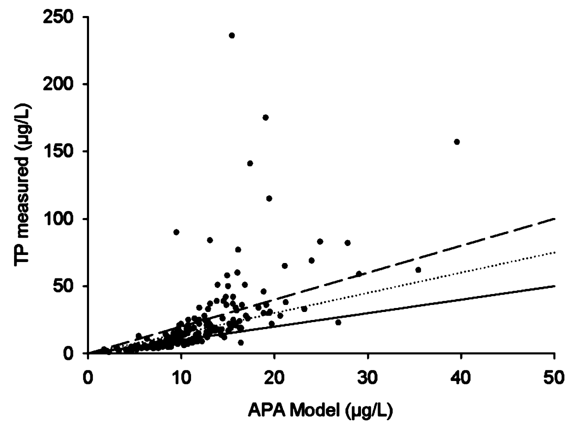


Figure 4. The APA Model applied to lakes with known point sources of P in the tributary watershed. Solid, dotted, and dashed lines represent 1:1 slope (estimated background concentrations) and TP levels 50% and 100% (representing an EQR of 0.5) greater than background concentrations, respectively.

■ DISCUSSION

The variables PM_n, AbsF, and Alt together explain 71% of the variation for TP concentration in the reference lake data set used in this study. The results are in line with those documented by other studies that show shallow lakes with higher suspended particulate matter,[23] greater humic content,[24,6,8] and lower altitude[6,8] generally have higher TP concentrations. TP has been shown to correlate well with humic matter in northern, boreal lakes,[9] and thus, its importance in a predictive model for TP is expected. Altitude, on the other hand, is somewhat unexpected as a predictor variable for TP. Differing weathering rates, controlled by temperature, may be one explanation for the importance of this variable. Another possible explanation is that bioavailable P is likely to be greater as one moves from high to lower areas within the landscape due to the increased time for transformation of P contained in larger, nonsuspended particles to more bioavailable forms.

The inclusion of PM_n improved the predictive ability of the model developed herein, especially for lakes with either low or high particulates. To the best of our knowledge, this is the first

 dx.doi.org/10.1021/es3040413 | *Environ. Sci. Technol.* 2013, 47, 1809−1815

study to include such a factor for predicting TP concentration in a large set of lakes. The ability of previous models[3,6,18] to predict reference TP concentrations in naturally turbid systems is limited due to either the lack of shallow lakes or parameters representing naturally derived particulate matter. Inclusion of Z_m in the MEI developed by Vighi and Chiaudani[3] and applied by Cardoso et al.[6] indirectly links natural particulate matter to these models because particle settling rates are generally dependent on lake morphology and residence time.[25] The lakes studied by Vighi and Chiaudani, however, are generally deep, stratified lakes that are likely low in natural particulate matter (no information was provided in the study). Although Cardoso et al. included both shallow and deep, stratified lakes, they removed any lakes with TP concentrations >35 μg/L before model development and were still only able to explain 51% of TP variation in their reference data set.

Model Predictability. Inclusion of PM_n increased variability explained in the data set by 10% when compared to a similar model developed previous to this study[18] using only AbsF and Alt. Improvement was seen in lakes with both high and low particulate matter. The improvement at both ends of the particulate range is due to the nature of Model 2 (without PM_n) whereby the prediction of TP concentration is based on an average condition relative to particulate matter. Thus, one would expect a bimodal effect when adding PM_n to the model, a decrease in both the overprediction of low values and underprediction of high values. This is an important distinction between the APA model developed herein and other models that do not include lakes with elevated particulate matter for model development. Most improvement in prediction using the APA model occurs for lakes that have relatively low or elevated particulate matter (Figures 2 and 3); however, the number of these lakes in relation to the total data set is low (Supporting Information, Table S4).

In contrast to Cardoso et al.,[6] grouping lakes by humic status did not improve model prediction capability. Separating the reference lake data set into nonhumic (AbsF < 0.1) and humic (AbsF $\geq$ 0.1) lakes resulted in no improvement in model predictability. Z_m (as either Z_m or in MEI_{alk}) was also not as important as expected for prediction. Z_m was estimated for the reference lakes using the relationship developed by Sobek et al.[14] wherein maximum shore slope and lake area were used to develop an empirical model. There is considerable error in estimating Z_m using this relationship, which likely explains the limited importance of the variable in our modeling.

The reference data set did, however, include 70 lakes (out of 1099) where Z_m was measured. Analysis of these 70 sites, in a similar manner used to develop the APA model, resulted in a model with three factors including PM_n, Alt, and Z_m (R^2 = 0.70). Z_m was the strongest variable for prediction (53% explained TP variation) whereas PM_n and Alt explained 10% and 7% of total variability, respectively. Interestingly, Cardoso et al.[6] found that Z_m explained a similar amount of variability in their reference data set (51%). AbsF was the fourth most important factor in MLR analysis but was not significant (p = 0.054) and explained slightly less than 2% of total variability in the data set. Thus, Z_m was an important predictor variable when only measured values were used in modeling. Measured Z_m values, however, are only available for a small percentage of lakes in Sweden. Accurately measuring Z_m is both costly and time prohibitive when considering Sweden alone has approximately 100,000 lakes.

One restriction on modeling, imposed by data availability, was the use of absorbance of irradiance (420 nm) to estimate particulate matter in the study lakes. Absorbance has been a standard of measurement in the Swedish national monitoring program since the 1960s. The origin of particles (e.g., mineral or organic) can affect how light is absorbed, as well as P content. The use of PM_n in the APA model improved the prediction of reference TP concentrations, but there is an inherent variability in this measurement. Use of turbidity, or some other measurement for particulate matter, could further improve variance explained by the APA model. It should also be noted that only round values (i.e., integers) were reported at low TP concentrations ($\leq$10 μg/L) in our study. The integer only values result in a deviation from a bivariate normal distribution and thus may bias the resulting R^2 values.

Regional Models. The APA model was applied to individual ecoregions in Sweden. A major portion of TP variability was explained in each ecoregion, and model predictability was generally similar to individual models developed using all possible predictor variables (Table 2). Using the APA model did result in a reduction of 10% and 11% of variance explained in ecoregions 4 and 6, respectively. Ecoregion 4, in southeast Sweden, is an area with high calcium (Ca) and magnesium (Mg) carbonates in the surrounding watershed soils that are known to correlate to P concentrations in surface waters.[26] Because these carbonate minerals contain P, their concentrations in surface waters can represent P released through weathering processes. Na was also a significant factor in ecoregion 4, possibly indicating the influence of the Baltic Sea on deposition and P concentrations in this area. TP concentrations in ecoregion 6, located in southwest Sweden, correlated to longitude in addition to PM_n, AbsF, and Alt. Longitude may indicate the influence of strong deposition gradients from coastal to inland areas. Thus, region specific models may provide additional predictive power in areas where local differences in deposition or watershed geology substantially influence TP concentration.

Model Application to Sites with Anthropogenic P Loads. The importance of a scientifically sound definition of reference state to determine ecological status, especially in relation to other pillars within the EUWFD, is stressed by a number of studies (e.g., Valinia et al.[27]). The APA model identified 81 lakes in the anthropogenic P load data set as degraded (i.e., EQR less than 0.75). Approximately half of these lakes had an EQR less than 0.5. The 100 sites with EQR values greater than 0.75 appeared to be minimally affected by anthropogenic loading. Nutrient loads within a watershed, however, do not necessarily mean that all or even a portion of the load reaches the lake. The distance between source and lake was great (up to 90 km) for some lakes, providing opportunity for uptake and/or sedimentation. For example, severely degraded lakes (EQR < 0.5) had a mean ratio of P load (kg) to distance between source and lake (km) of 46, whereas the ratio for lakes with an EQR > 0.75 was only 12. Thus, as distance from source to lake increased relative to the total anthropogenic P load, the measured TP concentration of a lake was more likely to be closer to the predicted background concentration. Supporting this were weak, negative relationships between EQR and both P load/source distance and P load/watershed area (data not shown).

Three sites with anthropgenic P loads had substantially lower measured TP concentrations compared to background concentrations (EQR > 2) predicted using the APA model.

dx.doi.org/10.1021/es3040413 | Environ. Sci. Technol. 2013, 47, 1809−1815

Two of the sites are located downstream from water bodies that are limed to increase surface water pH. This may artificially reduce P availability in the system due to increased input of P binding metals to the limed water body.[28,29] The measured TP concentration (1 μg/L) for the remaining lake with an EQR > 2 is at the analytical detection limit, increasing the chance for analytical error.

Inclusion of a natural particulate matter factor in the APA model overcomes a key weakness in previously developed models, allowing for prediction of reference TP concentrations in not only deep lakes but also shallow lakes with naturally elevated particulate matter. The results of this study indicate that models incorporating particulate matter can be useful for future standard development (e.g., EUWFD), especially for lakes with naturally elevated P concentrations due to suspended particles. Further work should include investigation into the relationship between particle origin and P content and the affect this has on predicting reference TP concentrations in lakes.

■ ASSOCIATED CONTENT

Supporting Information

Figures S1−S3, Tables S1−S4, and indicated portions of the Methods section. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This study was supported by funding from the Swedish Environmental Protection Agency. We are grateful for thoughtful comments and advice on the manuscript from three anonymous reviewers.

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