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Forms and Variability of Phosphorus in the Baltic Sea—A Challenge to Ecosystem Modeling

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Abstract: Knowledge of the partitioning between the dissolved and particulate phases of nutrient elements is a key factor in aquatic ecosystem modeling since partitioning regulates the availability to demand ratio of the nutrient in question. This is seldom taken into account in environmental monitoring programs. In this paper, the occurrence and variability of particulate and dissolved phosphorus were studied in the coastal zone of the Baltic Sea. The particulate fraction (PF) of total phosphorus (TP) concentration in coastal waters from some forty stations along the east coast of Sweden was, on average, 0.33. Dissolved inorganic phosphorus (DIP) was a poor predictor of total dissolved phosphorus (DP) representing only 20%–30% of this fraction. Sensitivity analyses showed that the value of PF had a significant impact on modeled predictions of TP concentration in the water on a Baltic sub-basin scale, whereas an applied coastal model was insensitive to variations in PF. Hence, this study encourages further sampling efforts on the partitioning of phosphorus in the open waters of the Baltic Sea.

Keywords: phosphorus, particulate fraction, ecosystem modeling, Baltic Sea

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Introduction

Phosphorus is probably the most studied plant nutrient in aquatic sciences. It is often described as the nutrient that limits the growth and biomass of algae.1 Numerous regressions have been constructed to link phosphorus, especially total phosphorus (TP), to variables such as algal chlorophyll, algal weight and productivity, and Secchi depth. $2-7$ Phosphorus in natural waters can be divided into three component parts: dissolved inorganic phosphorus (DIP), dissolved organic phosphorus (DOP) and particulate phosphorus (PP). The sum of DIP and DOP is termed dissolved phosphorus (DP), and the sum of all phosphorus components combine to give TP. Soluble and particulate phosphorus are operationally differentiated by whether they pass through a $0.45 \mu m$ filter.⁸ The "soluble" fraction does not necessarily contain only dissolved phosphorus forms. The filter excludes most particles, but colloidal phosphorus may be present in the filtered fraction.

DIP largely consists of orthophosphate $(PO₄)$. Orthophosphate can be taken up directly by algae, and the concentration of this fraction indicates the amount of phosphorus that is immediately available for algal growth. DIP is quickly regenerated and is therefore a poor predictor of nutrient status in aquatic systems.⁹ A number of organic phosphorus molecules have been identified that would fall within the DOP category but two main classes seem to predominate DOP in natural waters.⁸ The first category includes low molecular weight compounds, which are apparently derived from algal and bacterial metabolism, and are not considered as being directly biologically available. The second group of molecules includes colored large molecular weight compounds, eg, phosphorus bound in humid complexes, which release orthophosphate in the presence of ultraviolet light. These compounds may form a pool of phosphorus available for uptake by biota.10–12

The Baltic Sea has been suffering from eutrophication for decades. Remedial measures have, until recently, been focused on reducing the nitrogen load. However, recent findings using both modeling $13,14$ and empirical approaches^{15,16} have led to a shift in the abatement strategy, whereas the phosphorus input to the Baltic proper is nowadays recognized as important to reduce.^{17,18} Empirical data on the differentiation between PP and DP in the Baltic Sea are scarce,

since most environmental monitoring programs only measure TP and DIP.14 This fact potentially limits the predictive power of dynamic mass balance models when applied in the Baltic Sea. Comprehensive studies in boreal glacial lakes $19,20$ have found that DP and PP roughly represent 50% each of TP. To use this order of magnitude when differentiating between DP and PP has been successful in modeling phosphorus turnover in the different sub-basins of the Baltic Sea.¹⁴ Nevertheless, since data from the outer Oslo fjord in vicinity of the Baltic Sea, $2¹$ for example, suggests that as much as 90% of TP is DP, it is warranted to test the hypothesis that the PF value for phosphorus found in lakes also prevails in the brackish Baltic Sea. It is well known that salinity affects the aggregation and sedimentation of particles in estuaries, eg, in relation to the concept of the zone of maximum turbidity.²² It also possible that salinity influences the partitioning between dissolved and particulate phases of phosphorus, as has been found for trace metals.23

This study was designed to investigate the occurrence and variability of different physical fractions of phosphorus in Swedish Baltic coastal waters and to investigate the sensitivity associated with the PF value when modeling TP concentrations at different ecosystem scales.

Materials and Methods Study area

Between 2008 and 2010, 88 samples from surface and bottom water (depth ranging from 5 to 50 meters) were collected from 32 stations (App. 1) situated in the northwestern Baltic proper and the south-western Bothnian Sea (Fig. 1). Samples were collected from all seasons but samples from the production period (May–September) predominated. The study area contains a gradient in water salinity because of the limited exchange of water between the Baltic Sea and the Atlantic, with values ranging from 6 PSU in the south towards 4 PSU in the north.²⁴ Moreover, a gradient in nutrient concentrations is also present because of differences in population densities, with TP levels in the south averaging around 20 μ g L⁻¹ decreasing to 10 µg L^{-1} in the northern part of the study area.¹⁴ In general, river mouths create an east–west gradient in salinity and nutrient levels. As reference material, water samples were collected from the oligotrophic Lake Vättern in central Sweden

Figure 1. Map of the study area with sampling stations marked with dots.

and the Särö archipelago on the Swedish west coast where water salinity is considerably higher (around 20 PSU).

Water sampling and analysis

Water samples were collected with a Ruttner sampler and stored cold in two 1-L plastic bottles. When returned to the laboratory, part of the sampling volume (normally 1 L) was filtered through a 0.45 µm Whatman GF/C glass microfiber filter for later analysis of suspended particulate matter (SPM) and suspended organic matter (SOM). Filters used

for suspended matter analysis had been dried at 550 °C and pre-weighed. Another part of the sampling volume (normally 500 mL) was filtered through a 0.45 µm Whatman cellulose acetate filter for later analysis of particulate phosphorus. The filtrate was saved as well as the remaining unfiltered water volume. The chemical analyses were performed at Erken Laboratory, Uppsala University. The analyses were done according to Swedish standard methods.²⁵

The water salinity and temperature were measured online in the field using an YSI INC Model 30 M handheld salinity, conductivity and temperature

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device. The Secchi depth was measured using a standard Secchi disk.

Data Treatment

The definition of PF is:

$$
PF = \frac{C_{part}}{C_{tot}}\tag{1}
$$

where C_{tot} is the total (unfiltered) concentration and C_{part} is the particulate concentration.²⁶

 \overline{C}_{part} was calculated in two ways: (1) as the measured PP concentration on the filters and (2) as the difference between measured TP concentration in unfiltered water (TP) and filtered water (DP). The DP concentration was directly measured as TP in the filtrate (DP1). DP was also calculated as TP-PP (DP2). Correspondingly, PF was calculated directly from PP/TP (PF1) and from (TP-DP)/TP (PF2).

The variability within the coastal area was studied through analyses of ten replicates each from three areas. The variation was expressed using the coefficient of variation (CV), defined as the ratio between the standard deviation and the arithmetic mean value of a sample.

To investigate how different values of PF for phosphorus may affect the mass balance for TP, and thereby the water concentration of TP in coastal areas and sub-basins of the Baltic Sea, a sensitivity analysis was performed where different values of PF were used and the resulting prediction of the target variable the TP-concentration in the surface water was simulated. This was done using the simulation software Stella®. Two types of model for TP turnover were applied on a coastal ecosystem and a sub-basin of the Baltic Sea, respectively. First, a simplified version (Fig. 2a) of a validated mass-balance model for TP-turnover in Baltic coastal areas⁶ was applied in one semi-enclosed coastal area, the Kallrigafjärden Bay. This area has previously been modeled using a similar approach.²⁷ It has an extensive background dataset and typical characteristics of Swedish Baltic coastal areas concerning, for example, water turnover. The other applied model for sensitivity analysis was a crude phosphorus budget for the whole Baltic Sea divided into sub-basins (Fig. 2b) based on nutrient budget calculations presented in Wulff et al²⁸ and Håkanson and Bryhn.14

To put the results from the sensitivity analysis of PF into perspective, a similar analysis was performed with the riverine input of TP as the sensitivity parameter. The variability in the yearly riverine load of TP was estimated using a dataset (App. 2**)** covering 35 rivers during the period 1997–2006 from the national Swedish monitoring program of estuaries provided by the Sveriges Lantbruksuniversite (SLU; Swedish University of Agriculture).

Results

Basic statistics for the measured water variables are presented in Table 1. Most measured variables had a wide range. For example, TP varied between 9 and 170 μg L⁻¹ with a mean value of 25 μg L⁻¹, and PP varied between 2 and 108 μ g L⁻¹ with a mean value of 9 µg L[−]¹ . The average value of PF calculated from Equation 1 for each sample was 0.33.

A summary of the different DP and corresponding PF values are given in Table 2. The results in Table 2 show that the mean DP concentration was lower (10 μ g L⁻¹) when measured in the filtrate (DP1) compared to the calculated DP2 (average 15 μ g L⁻¹). Similarly, the calculated PF values differed. The mean PF1 was 0.33, while the mean PF2 was 0.51. The DIP concentration was considerably lower $(\text{mean} = 3 \mu g L^{-1})$ compared to the DP concentration, indicating that DOP and/or colloidal forms of phosphorus constituted a major part of total DP-concentration in the analyzed samples.

The measured average CV for different water variables are presented in Table 3. DIP had the largest inherent variability, with a CV of 0.28, followed by PP with a CV of 0.16, whereas TP and DP had a rather low CV of 0.07.

Table 4 shows a correlation matrix between different phosphorus fractions and other studied physical and chemical characteristics. A strong positive correlation was found between PP and TP $(r^2 = 0.94)$. PP was also positively correlated with SPM and SOM $(r = 0.96$ and 0.93, respectively). A strong positive correlation $(r = 0.99)$ was also found between the SPM and SOM of suspended matter (Table 4). The correlations between TP and PP and SPM and SOM, respectively, after logarithmic transformation are presented in Figure 3.

Considering the mean PF values of 0.33 and 0.51 found in this study depending on the analytical

Figure 2. Outlines of the applied simplified versions of models for **a**) Baltic coastal areas and **b**) the sub-basins of the Baltic Sea. Comprehensive descriptions of the models are given in Håkanson and Eklund (2007), Håkanson and Bryhn (2008) and Karlsson et al (2010). **Abbrevitations:** BP, Baltic proper; BS, Bothnian Sea; BB, Bothnian Bay.

technique applied, the global average PF value of $0.56¹⁴$ and the reported value of 0.10 from the Oslo fjord, 21 it was found reasonable to run the sensitivity analysis with the following values of PF: 0.5, 0.25 and 0.75 respectively. The choice of PF value had

a relatively large impact on the prediction of the TP concentration in the surface water of the Baltic basin proper whereas the sensitivity in the modeled coastal area was insignificant $(< 0.5 \mu g L^{-1})$ (Fig. 4). The steady-state value (100 years) of the

	Water temperature $(^{\circ}C)$	Salinity (psu)	Secchi depth (m)	SPM $(mg L-1)$	LOI $(mg L-1)$	ΤP (μ g L ⁻¹)	PP $(\mu g L^{-1})$	PO4-P $(\mu g L^{-1})$	PF
Mean	12	5.4	4.8	4.6	2.4	25	9	2.4	0.33
Median	13	5.2	4.5	3.2	1.4	22	6	1.4	0.33
Sta. Dev.	6	2.4	2.1	8.1	6.3	20	14	6.3	0.17
Max	22	17.2	10.5	65	50	170	108	50	0.66
Min	-0.1	2.9	1.8	1.2	0.5	9		0.5	0.06

Table 1. Basic statistics of measured variables, $n = 60$.

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Table 2. Comparison between different DP- and PFvalues, $DP1 = measured TP-concentration$ in the filtrate, $DP2 = TP-PP, PF1 = PP/TP, PF2 = (TP-DP1)/TP, n = 63.$

Variable	Mean
TP (μ g L ⁻¹)	23
PP (μ g L ⁻¹)	8
DP1 (μ g L ⁻¹)	10
DP2 (μ g L ⁻¹)	15
DIP (μ g L ⁻¹)	3
PF ₁	0.33
PF ₂	0.51

TP concentration in the Baltic basin proper was calibrated to 19 µg L^{-1} applying a PF value of 0.5. Changing PF to 0.25 increased the predicted TP concentration to 23 μ g L⁻¹, whereas a PF value of 0.75 resulted in a lower TP concentration of 16 μ g L⁻¹ (Fig. 4). In a simulation of Kallrigafjärden Bay, the predicted steady-state (60 months) TP concentration was around 13 μ g L⁻¹ regardless of which PF value that was applied (Fig. 4**)**.

Analysis of the SLU dataset (App. 2) regarding 35 Swedish river estuaries showed that a typical CV for the yearly TP transport is 0.40. A sensitivity simulation with the Baltic sub-basin model using this value to generate 300 normally distributed values of the total riverine TP load within the Baltic Sea resulted in a distribution in the TP concentration of the surface water presented in Figure 5. The confidence interval, calculated as $\pm 1.96 \times$ the standard deviation (SD), for the predicted TP concentration was wide (6, 30). This means that setting up a phosphorus budget in the Baltic Sea using input data from one single year, as often is done in international assessments of the ecological status of the Baltic Sea,²⁹ would contain large uncertainties.

Table 3. Coefficients of variation (CV) for SPM, LOI and different phosphorus fractions based on 10 replicates from three areas.

Variable	CV
SPM	0.13
LOI	0.13
TP	0.07
PP	0.16
DP	0.07
DIP	0.28

Discussion

The mean value of PF for phosphorus found in this study, 0.33, was higher than previously reported from the outer Oslo fjord in the north-east Atlantic, 21 but was lower than what has been reported from European boreal glacial lakes^{19,20} and the productive Chesapeake Bay on the east coast of the United States.¹⁴ This could possibly be interpreted as an indication that there is a general gradient in PF, with higher values recorded in shallower, nutrient-rich and productive lakes and coastal areas, following a decreasing trend towards the open sea where the SPM concentration generally is lower because of lower primary production and less resuspension.30 It should be mentioned though, that the data set presented in this study is limited and that the values reported should be considered as indicative and seen as a first step towards empirical quantification of phosphorus partitioning in the Baltic Sea.

Water samples were collected all year round but there was a bias in the sampling matrix, with more samples from the production period (May–September) compared to the winter period. When the dataset was disaggregated by season, the PF value was higher during the production period (0.4) compared to the period October to February (0.2). This may be one of the causes of the deviation from the PF value reported by Håkanson and Bryhn,¹⁴ which reflects summer conditions. Moreover, the DIP concentration was higher during the winter months, probably reflecting a mineralization of phosphate from seston that not was directly incorporated into new algae biomasses. This pattern is typical of the seasonal dynamics of nutrients in the Baltic Sea.³¹ No significant differences appeared when comparing samples from surface waters with bottom water samples. This can be explained by the fact that the water mass was relatively uniform in the sampled areas, without any marked thermo- or haloclines. Salinity does not seem to be an important factor for the partitioning of phosphorus, as seen for trace metals.²³

The organic content of SPM was surprisingly uniform, with an average value for SOM of 47% ww and a CV of 24%. This number is very close to what has been reported from fine sedimentary matter caught in traps in lakes³² but higher than found in recent coastal accumulation sediments, which typically have an organic content (SOM) of 15%.³³ The lower

	Sampling depth(m)	Water temperature (°C)	Salinity (psu)	Secchi depth (m)	SPM $(mg L-1)$	SOM $(mg L^{-1})$	ΤP $(\mu g L^{-1})$	PP $(\mu g L^{-1})$	DIP $(\mu g L^{-1})$	DP $(\mu g L^{-1})$
Sampling depth(m)	1.00	0.00	-0.06	-0.08	-0.15	-0.13	-0.14	-0.18	0.14	0.01
Water temperature $(^{\circ}C)$		1.00	0.13	-0.06	0.24	0.07	-0.31	-0.01	-0.76	-0.17
Salinity (psu) Secchi depth (m) SPM (mg L^{-1}) LOI (mg L^{-1}) TP (μ g L ⁻¹) PP (μ g L ⁻¹) DIP (μ g L ⁻¹) DP (μ g L ⁻¹)			1.00	0.07 1.00	0.49 -0.12 1.00	0.18 -0.33 0.99 1.00	-0.12 -0.66 0.91 0.94 1.00	-0.14 -0.45 0.93 0.96 0.94 1.00	0.07 -0.03 -0.10 -0.08 0.12 -0.09 1.00	0.00 -0.62 -0.22 -0.23 0.70 -0.07 0.49 1.00

Table 4. Kendall rank correlation coefficients. *r*-values marked bold are significant at $P < 0.05$, n = 60.

Figure 3. Observed correlations between log TP and log PP and log SPM and log LOI, respectively, (n = 60).

Figure 4. Predicted TP-concentrations (mg m[−]³) in surface waters when varying the PF-values in **a**) the Kallrigafjärden Bay and **b**) the Baltic proper.

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Figure 5. Sensitivity analysis using the Baltic Sea sub-basin model. The target variable is TP (µg/l) in the Baltic Proper and the TP-loading has been varied (CV = 0.40 , n = 300). The box-and-whisker plot shows the mean, ±1 standard deviation and ±1.96 * standard deviation.

organic content in recent surface sediments compared to the suspended matter in the overlying water column can partly be explained by mineralization of easy degradable organic matter and partly by dilution with minerogenic matter from resuspension processes occurring during strong wind events.34,35 The average value of the SPM concentration found in this study $(4 \text{ mg } L^{-1})$ is of the same order of magnitude as earlier reported in Pustelnikov³⁶ (average 3.3 mg L^{-1}) and Håkanson³⁰ (average 2.5 mg L^{-1}).

As seen from Table2, a difference appeared between DP measured directly in the filtrate and DP calculated from TP-PP, making it possible two calculate PF in two ways. So the question is: which analytical method is best for determining PF? Measuring the particulate fraction as the matter collected on the filter is the traditional method of determining the particulate fraction.37,38,25,19,20 It has the advantage that a rather large volume of water can be filtered, which reduces the risk of getting samples with concentrations of the actual substance that are below the detection limit of the analytical method. This can be a problem when performing analyses on the filtrate, especially with substances with high affinity with particles. On the other hand, the analytical methods are consistent when identical analyses are performed on unfiltered and filtered water, respectively. The results from this study also indicate that the inherent variability in phosphorus analyses may be lower water samples compared to filter analyses (Table 3).

DIP, which is a standard parameter in environmental monitoring programs, was a poor predictor of DP since, on average, it only represented 20%–30% of total DP (Table 2). Hence, the data presented in this study suggests that DIP cannot be used as an estimator of the non-settling phase of phosphorus in mass balance modeling. Similar conclusions have been drawn by Håkanson and Bryhn¹⁴ and Håkanson and Stenström-Khalili.³⁹

The rather strong correlations found in this study (eg, between TP and PP and SPM and LOI, respectively (Table 4, Fig. 3)) is, to a large extent, explained by the wide range in the dataset, which, in turn, depends on the fact that one sampling excursion was undertaken during a significant algae bloom. If this data point was removed from the dataset, the *r*-values in Table 4 dropped markedly but the correlation was still significant $(P < 0.05)$. Nevertheless, these regressions should be treated as indicative and they need to be further elaborated.

Varying the PF value had a marked impact on the prediction of TP concentration in the Baltic basin proper but not on the prediction of the TP concentration in the coastal estuary of Kallrigafjärden Bay (Fig. 4). The main reason for this is the generally fast water turnover in coastal areas, which gives internal processes, eg, sediment–water exchange, little importance for the nutrient mass balance. On the other hand, in the basin of the Baltic proper and the other large basins of the Baltic Sea where the water turnover is slow (within the range of decades), the settling of particles, although also a slow process, is important to the overall mass balance. A similar sensitivity to PF value when modeling on a Baltic Sea sub-basin scale is indicated in Häkanson and Bryhn,¹⁴ whereas other Baltic Sea models (eg, 13,40,41) have a different parameterization of the sedimentation processes, making model comparisons irrelevant.

Not surprisingly, the variation in the riverine load of TP had a large impact on the modeled TP concentration in the Baltic basin proper (Fig. 5). A similar sensitivity to the allochthonous input has also been recognized in various lake ecosystem models.32,42 The calculated CV for the yearly riverine load of TP to the Baltic Sea of 0.40 infers that an average of 16 years, transport data, at least, a required to establish a typical mean value for the riverine load if one accepts an error of 20%.⁴³ Wulff et al⁴⁴ advocated that the conditions in the Baltic Sea are slowly getting less eutrophic, based on calculations from riverine load data for the years 1994–2006. The calculations above show that such a conclusion, drawn from a rather short period, may be premature.

Conclusions

The indicative PF value obtained in this study is lower but of the same magnitude as that reported earlier in boreal lakes. PP measured as the remains on the filter after filtering is not equivalent with PP measured as the difference between TP in unfiltered and filtered water. DIP was a poor predictor of DP, representing only a small fraction of the total dissolved phase. DIP also had the largest inherent variability of measured phosphorus fractions. A strong correlation was found between TP and particulate phosphorus (PP) in the sampling matrix. This study demonstrates that an accurate PF value is necessary for obtaining high predictive power in phosphorus mass balance models of the Baltic Sea. Since the empirical basis for this is weak, more attention should be given to determining of the particulate fraction of phosphorus in future sampling programs of open waters of the Baltic Sea.

Disclosures

This paper is unique and not under consideration by any other publication, and has not been published elsewhere. The author and peer reviewers report no conflicts of interest. The author confirms that he has permission to reproduce any copyrighted material.

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Note: *Average of 10 replicates. **Note:** *Average of 10 replicates.

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Replicate sampling at three stations.

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TP-transport (tonnes yr-1) in 35 Swedish rivers from the SLU-database, calculated standard deviations and CV-values for selected periods.

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