Estimating the amount of mobile phosphorus in Baltic coastal soft sediments of central Sweden

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A new dataset based on 102 sediment cores was examined to estimate the amount of phosphorus (P) that will eventually be released to the water column from the Baltic coastal sediments along the Swedish coast between Öregrund and Oxelösund. Total P (P_{tot}) concentration in the surface sediments varied between 840 and 7100 µg g⁻¹ dry weight (dw) with an average of 1650 µg g⁻¹ dw. In deep sediments, the P_{tot} concentration was around 1000 µg g⁻¹ with small variation. The difference between surface concentration and the stable, deeper, concentration represents P to be released, i.e. the mobile P. Pools of mobile P varied between 1.5 and 18.2 g m⁻². Correlations between surface P_{tot} concentrations and amounts of mobile P were strong (r² = 0.88). We estimate the amount of mobile P in the coastal sediments of the investigated region to be between 1000 and 4000 tonnes. Assuming a turnover time of the mobile P between three and ten years gives an average annual P release of 100–1300 tonnes yr⁻¹.

Introduction

The trophic status of the Baltic Sea is determined by the availability of phosphorus (P) and nitrogen (N) in the water column (e.g. Larsson et al. 1985, Granéli et al. 1990, O’Melia et al. 2006). The role of the sediments in the cycling of P is often highlighted (e.g. Eilola et al. 2009, HELCOM 2009). In a number of studies, P fluxes between the sediments and the water column are indirectly quantified by mass-balance calculations or by monitoring changes in water-column concentrations. Such calculations indicate that internal P loads are of the same order of magnitude as the external ones (Emeis et al. 2000, Conley et al. 2002, Savchuk 2005). Redistribution of sediments through wind/wave action and land uplift may mobilize even larger amounts (Jonsson et al. 1990, Blomqvist and Larsson 1994, Eckhäll et al. 2000, Håkanson and Bryhn 2008). From an ecological perspective, however, release of molecular phosphate from the sediments represents a paramount source since it is all bioavailable. In contrast, resuspension of particulate P forms and external P load consist of a mixture of labile and inert P forms.
Total phosphorus (P_{tot}) concentrations in the Baltic Sea sediments range between 300 and 2500 µg g^{-1} dw (Jonsson et al. 1990, Virtasalo et al. 2005). According to Carman and Cederwall (2001), P_{tot} concentrations in surface sediments (0–1 cm) of different parts of the Baltic Sea and in different bottom types are 700–2500 µg g^{-1} dw (median 1300 µg g^{-1} dw). The highest concentrations were found in accumulation areas and the lowest in erosion areas. Emelyanov (2001) collected at least 386 samples of surface sediments (0–5 cm) from different parts of the Baltic Sea and found average P concentrations in different bottom types to be between 600 and 1400 µg g^{-1} dw. Deep sediments (below 5 cm) in accumulation areas typically contain around 1000 µg P g^{-1} dw (Lehtoranta and Heiskanen 2003, Lukkari 2008).

Knowledge of permanent immobilization processes for P in brackish environments is poor (Conley et al. 2009). Settling P is associated with either organic or inorganic particles. Some fractions of inorganic P are inert and are consequently not affected by chemical processes in the sediment. These stable P forms, including apatite and P bound to aluminium, will therefore stay and be buried in the sediment after settling from the water column, as observed in lake sediments (e.g. Boström et al. 1982, Rydin 2000, Reitzel et al. 2005). Other forms of P are influenced by chemical conditions in the sediment. Redox-sensitive P fractions are mostly associated with iron compounds, although e.g. redox-sensitive manganese compounds may also bind P under oxic conditions (Mortimer 1941, 1942, Boström 1984, Ingri et al. 1991, Gunnars and Blomqvist 1997).

Even if surface sediments are oxidized, the redox potential a few centimeters down in fine sediments is always low and consequently the concentration of iron-bound P in deep sediments is generally low. Organic matter may be more or less degraded which also includes P present in this material. The amount of easily degradable material decreases with sediment depth. At greater sediment depths, non-degradable or refractory material dominates the organic fraction. The amount of organically-bound P generally decreases with sediment depth together with the organic content (Gächter et al. 1988, Ahlgren et al. 2006).

Dissolved P in sediment pore-water can diffuse upward in the sediment and reach the water column or become trapped by sediment particles, in particular Fe(III) compounds, in oxidized surface sediment. Dissolved iron may be scarce in marine sediments due to formation of insoluble iron sulphides (e.g. Matthiesen et al. 1998, Blomqvist et al. 2004). P which is potentially released to the water column is mostly present in the top 2–3 centimeters of sediment (Carman and Jonsson 1991, Lukkari 2008).

Carman and Jonsson (1991) attempted to calculate total amounts of P in Baltic Sea sediments and concluded that a major proportion (~95%) of the potentially mobile P fractions is present in near-shore and archipelago areas. One reason for this is the higher P binding capacity of the permanently oxidized surface sediments in shallow areas. Their estimate, however, was based on the top 2 cm only. Lukkari (2008) studied the P composition in 20 sediment cores from the Gulf of Finland and the Archipelago Sea and quantified mobile pools and burial of P. Rydin et al. (2011) estimated long-term average of gross and net loading, including the release of sediment P at four stations in the Stockholm Archipelago. The difference between the generally higher P_{tot} concentrations found in surface sediments and the lower concentrations found in older, deeper layers indicates the magnitude of the pool of P to be eventually released (Rydin et al. 2011). The stock of sediment P that in the future will eventually be released as dissolved P has been referred to as “reactive P” (Lukkari et al. 2009 and references therein) or “potentially mobile P” (Rydin 2000). It may also be referred to as bioavailable P although it is only bioavailable after it is released from the sediment. It is the potentially-mobile P (Rydin 2000) that we attempted to estimate in this study, and we will hereafter refer to it as “mobile P”.

Given the key role of the sediments for the P cycling and the nutrient status in the Baltic Sea there is a need to quantify the pool of mobile P in the Baltic Sea sediments, and to improve the understanding of the mechanisms determining the fate of different P forms in sediments and the response to environmental forcings in terms of, e.g. external P load or climate change.

In this study, we present a new set of data on P composition and P_{tot} concentrations in the Swed-
ish east coast around the Stockholm Archipelago. The aim was to estimate the pool of mobile P, and to improve the understanding of the P dynamics including quantification of different P forms in the sediments of the studied area.

**Methods**

**Sediment sampling**

In total, 102 cores of sediments were collected with a Gemini double corer (inner diameter 80 mm) or with a Willner sampler (inner diameter 63 mm) from 33 coastal areas of the southern Bothnian Sea and the northwestern Baltic Proper from Öregrund to Oxelösund including the Stockholm Archipelago (Fig. 1) between November 2008 and February 2010. Core samples were sliced in the field in 2-cm-thick layers (a few 1-cm layers from surface sediments were also prepared, see Appendix). Surface sediments (0–2 cm) and deep sediments (between 2- and 5-cm-thick layers located between 10 and 70 cm below the sediment surface depending on the sediment core structure) were analyzed for water content, organic content and P\(_{\text{tot}}\). Twelve sediment cores were selected for P fractionation at several depth levels (1–2 cm slices) with close intervals above 10 cm sediment depth and more sparsely at deeper levels (Table 1). In addition to the 12 fractionated sediment cores, 90 surface samples and 73 deep sediments from our collected cores were used in this work.

**Chemical analyses**

Chemical analyses were performed at the Erken laboratory (Uppsala University). All the cores were stored in darkness at 4 °C until preparation in the laboratory. Water content was determined after freeze drying, and organic content (loss on ignition, LOI) after ignition at 550 °C for two hours.

**Table 1.** Labels (location), positions (WGS-84) and characteristics of sediment cores collected for P fractionation.

<table>
<thead>
<tr>
<th>Location</th>
<th>Latitude (N)</th>
<th>Longitude (E)</th>
<th>Depth (m)</th>
<th>Water content (%)</th>
<th>Loss on ignition (%)</th>
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<td>34</td>
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<td>28</td>
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<tr>
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<td>6009</td>
<td>1841</td>
<td>41</td>
<td>91</td>
<td>17</td>
</tr>
<tr>
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<td>5911</td>
<td>1845</td>
<td>58</td>
<td>91</td>
<td>23</td>
</tr>
<tr>
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<td>1849</td>
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<td>19</td>
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<td>19</td>
</tr>
<tr>
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<td>1845</td>
<td>27</td>
<td>88</td>
<td>16</td>
</tr>
<tr>
<td>8 Gål Q (Gålman)</td>
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<td>1846</td>
<td>31</td>
<td>88</td>
<td>17</td>
</tr>
<tr>
<td>9 Tor C (Torsbyfjärden)</td>
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<td>1828</td>
<td>31</td>
<td>88</td>
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<tr>
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<tr>
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</table>
Content of P_{tot} in sediments was analyzed as PO_{4}^{3–} after acid hydrolysis at high temperature (340 °C) according to Murphy and Riley (1962). Phosphorus forms were separated into NH_{4}Cl-rP, BD-rP, NaOH-rP, NaOH-nrP, HCl-rP and residual P following, in principle, the sequential extraction scheme suggested by Psenner et al. (1988). These fractions are defined by the extraction method, but ideally each fraction corresponds to a specific P containing substance in the sediment. Generally, NH_{4}Cl-rP is regarded as loosely bound P, BD-rP as P associated with iron hydroxides, NaOH-rP as P bound to aluminium, NaOH-nrP as organic P forms, and HCl-rP as calcium bound P compounds. Residual P is given by subtracting extracted and identified P from P_{tot}. P fractionations were started within a week after sampling.

Quantifying the pool of mobile P

To compute the amount of mobile P per unit area from the sediment core data, the burial concentration, i.e. the P concentration in deep sediment in each core where sediment diagenesis has ceased, was subtracted from the higher concentrations in more shallow layers. Burial concentrations were taken as the average concentrations below the stabilization depth. Concentration differences (in µg g–1 dw) were multiplied by dry matter content to obtain the amount of mobile P in each layer. Dry matter content was determined by calculating the bulk density in each layer adjusting for water content and organic content according to Håkanson and Jansson (1983). The surpluses of P in each sediment layer were taken as mobile P and integrated over the depth profile to obtain the amount of mobile P per square meter accumulation bottom area (missing layers were filled by linear interpolation). This procedure was followed for P_{tot} as well as for the different P fractions.

Statistical analyses

A Mann-Whitney U-test was used to determine if average P concentrations differed in different subsamples. A least-square linear regression analysis was also carried out.

Results

Basic sediment parameters

Basic information about our sampling sites is given in the Appendix. In our dataset, all surface samples were categorized as accumulation sediments according to Håkanson and Jansson (1983). Water content varied between 75.0% and 97.9% (mean = 86.6%) and LOI (≈ organic content) varied between 9.1% and 54.7% (mean = 16.5%).

P_{tot} in surface and deep sediments

In the deep sediments, the P_{tot} concentration was typically around 1000 µg g–1 and the variation was relatively small (Fig. 2). Its standard deviation was 266 µg g–1 but excluding one outlier (3000 µg g–1; the second highest value was 1700 µg g–1) the standard deviation was reduced to 154 µg g–1.

In the surface sediments, variation in P_{tot} concentration was considerably greater. The dataset included 11 coastal areas with 4–20 samples, making statistics meaningful within these areas. The within-site coefficient of variation was on average 0.28. P_{tot} concentration was not correlated with water depth, depth below wave base (calculated according to Håkanson and Eklund 2007), water content or organic matter content (r^2 < 0.25, n = 102).

P fractions in sediment profiles

Concentrations of P_{tot}, iron-bound P and organic P decreased with depth (Fig. 3). It is apparent that the loosely bound P fraction generally constitutes a quantitatively minor fraction (Fig. 4). The iron-bound P decreased with depth towards a well defined burial concentration of around 100 µg g–1 dw. The relatively constant concentrations with depth of P associated with calcium and aluminium indicate that these fractions are more or less inert. The aluminium-bound P, however, declined slightly with depth close to the surface which is probably due to imperfections in the chemical extraction rather than sediment dynamics (Rydin et al. 2011). Organic P declined with
depth, and in some profiles the decline continued throughout the cores indicating that degradation of organic material in some cases may proceed decades after deposition. However, P_{tot} concentrations generally stabilized below 5 cm or even shallower depths, hence most mobile P occurs above this level. Deeper sediment layers typically contained around 1000 µg P g⁻¹ dw. In one sediment core — Norr A — the concentration profile deviated from the general pattern found in the other cores. Some exceptional P_{tot} concentrations were found in layers Pil A (8–10 cm), Bull J (6–8 cm) and Tor P (48–50 cm). The P_{tot} decline with depth is larger than the combined decline of iron-bound and organic P, indicating that some of the mobile P was present in the residual fraction.

**Mobile P in sediment profiles**

Burial concentrations used to estimate the mobile P (see Methods) were obtained from average concentrations below 10 cm depth for each fraction. In Norr A, the concentrations at 28–30 cm depth were taken as the burial concentrations. In Tor P, the value at 48–50 cm depth was excluded due to the fact that it was three SD above the mean for the rest of the core below 10 cm depth. When summarizing amounts of mobile P, negative values (i.e., actual concentrations lower than burial concentrations) were excluded.

The total pools of mobile P varied between 1.5 and 18.2 g m⁻² (Fig. 5). Mobile P constituted 19%–79% (mean = 47%) and 8%–51% (mean = 26%) of P_{tot} in the top 2 and 10 cm of the sediment, respectively. Typically the concentration of mobile P decreased rapidly with depth. In four cores, the 0–1-cm and the 1–2-cm layers were analyzed separately, and the P_{tot} concentration was between 1.3 and 2.9 times higher in the 0–1 cm layer. On average 57% of the mobile P occurs in the top 2 cm of the sediment (n = 12). Correlations between surplus P_{tot} concentrations and the sums of mobile fractions (Fig. 6a) and between surface P_{tot} concentrations and amounts of mobile P_{tot} (Fig. 6b) were strong ($r^2 = 0.90$, n = 12; and $r^2 = 0.88$, n = 12; respectively).

**A dataset for comparison**

We had the opportunity to compare our data with 83 surface samples (0–2 cm) collected by the Swedish Geological Survey (SGU) between July 1996 and October 2002 from the same area as ours. The samples in the SGU database were a sub-selection from a larger data set rejecting data from sediments which were considered to be from non-accumulation areas (based on water depth and sediment water content). The mean P_{tot} concentration in the SGU data was 1550 µg g⁻¹ dw. The average P_{tot} concentration in the surface sediments found by us did not differ statistically (Mann-Whitney U-test) from the values in the SGU database.

**Discussion**

This study estimates mobile P in accumulation sediments. Hence, the pool of mobile P in ero-
sion and transportation areas is not considered although it could be substantial. However, sediment dynamics above the wave base are different than in accumulation areas, since sediment resuspension interrupts P diagenesis and creates a different chemical environment. For this reason, in erosion and transportation areas all P is “mobile” (but only partly bioavailable). As stated in the

Fig. 3. Vertical concentration profiles of different P fractions in 12 sediment cores.
introduction, typical P concentrations are lower in erosion and transportation sediments than in accumulation sediments.

Total amounts of P differ considerably between locations, which was largely explained by the iron-bound fraction. In fact, variation in the iron-bound fraction explained 95% of the variation in $P_{\text{tot}}$. For example in Tor P, concentration of iron-bound P in the surface sediment (0–2 cm) was very high (3600 µg g$^{-1}$ dw), while the iron-bound P concentration in Pil A was very low (170 µg g$^{-1}$ dw). The extremely high surface concentrations of iron-bound P may either be upward migrating P trapped in the oxidized sediment surface or phosphate precipitating from the deep water column at the sediment surface.

In this work, temporal variability in sediment P storage was effectively disregarded. Deposition of organic material is higher during the growing season which enhances P accumulation in surface sediments. In lakes, the accumulation of organic material in surface sediments typically decreases the redox potential and causes release of iron-bound P. It is reasonable to expect that temporal dynamics also affects the P dynamics in the coastal sediments of this study, although seasonality is less predictable than in lakes due to e.g. events of water exchange.
Amounts of mobile P in different sediment cores reflect spatial variability in sediment accumulation, P content in settling matter and diagenesis/tturnover rates in the sediments. Sediment cores contain valuable information about these processes, but the quantification of different rates requires additional information. Average and median P concentrations in our sediment data are consistent with values in the Baltic Sea reported in the literature, however, in the upper part of the range given in e.g. Jonsson et al. (1990) and Emelyanov (2001). The reason for this is probably that our dataset includes samples from accumulation sediments only. Our median deep-sediment P concentration of 1000 µg g⁻¹ dw is consistent with that of Lehtoranta and Heiskanen (2003) and Lukkari (2008). The average Pᵣₒₒ concentration in surface sediments found by us did not differ from the values in the SGU database.

The sampling stations used in this study included both oxic and anoxic areas. In some cases it is likely that during recent years the sediments had undergone regime shifts moving from anoxic towards oxic conditions (Karlsson et al. 2010). This clearly affects the pool of mobile P in the surface layers of the sediments. However, we did not find any significant differences (Mann-Whitney U-test) when comparing deep-sediment Pᵣₒₒ concentrations from sampling stations with anoxic surface sediments with those from stations with oxic surface sediments. The average deep-sediment Pᵣₒₒ concentrations in the anoxic and oxic sediments were 1001 and 1010 µg g⁻¹ dw, respectively. Studies of coastal sediments in the southern Baltic Sea have shown that precipitation of dissolved P with iron in oxidized surface sediment layers is a quantitatively important but transitory immobilization pathway of dissolved sediment P (Jensen and Thamdrup 1993, Jensen et al. 1995).

Persson et al. (1993) and Jonsson et al. (2003) used results from seafloor mapping by means of side scan sonar, sediment echo sounder and common echo sounder in combination with sediment sampling of cores to estimate sediment accumulation areas along the Swedish east coast. Using the results from Persson et al. (1993) and
Jonsson et al. (2003) the area of accumulation bottoms in the 33 areas where surface sediment were collected for this study was estimated to equal 253 km². Using hypsographic information (SMHI 2003) in combination with algorithms for predicting the depth of the wave base (Håkanson and Eklund 2007), we estimated the coastal sediment accumulation area between Öregrund and Oxelösund to be 983 km².

The strong correlations between the surface $P_{\text{tot}}$ concentrations and amounts of mobile P probably reflect the fact that surface conditions typically determine how much P can be retained in the profile. We may use the regressions given in Fig. 6 to estimate amounts of mobile P at the sampled sites. The outcomes of both regressions when the deep-sediment data ($n = 33$) were used in calculations were comparable (Fig. 7). Our extensive dataset allowed us to tentatively extrapolate our estimate of potentially mobile P in accumulation areas in our study area. Multiplying the average mobile P concentration in our 33 recently sampled areas (2.5 g m⁻²) by the sediment accumulation area as mapped by Jonsson et al. (2003) we found that this area contained ~630 tonnes of mobile P. For the entire area, the corresponding amount of mobile P would be 2460 tonnes.

Hence, it is fair to say that the amount of mobile P in the coastal accumulation sediments of the investigated region is probably between 1000 and 4000 tonnes.

Typical sediment accumulation in this region is between 5 and 20 mm yr⁻¹ (Jonsson et al. 2003) and mobile P concentrations below 5 cm are generally close to zero (see Fig. 2). We may thus conclude that the turnover time for mobile P is between 3 and 10 years (5 cm divided by 5 to 20 mm). This in turn means that the release of P from the accumulation sediments in our region will be between 100 and 1300 tonnes yr⁻¹ (or between 0.1 and 1.3 g m⁻² yr⁻¹ which is comparable to experimental data from various sites in the Baltic Proper; see e.g. Matthesen et al. 1998, Emeis et al. 2000, Hille et al. 2005, Mort et al. 2010). This is admittedly a very rough calculation but a reasonable order-of-magnitude long-term estimate that allows us to assess the quantitative importance of the mobile P pool. These values are comparable to the P transport of around 150 tonnes yr⁻¹ in the Norrström River — the outflow of Lake Mälaren with a catchment area of 22 650 km² constituting the major terrestrial water inflow to the Stockholm Archipelago. For comparison, the P release estimates from the Gotland Basin sediments and the anoxic parts of the Baltic Proper are 14 000 tonnes yr⁻¹ (Emeis et al. 2000) and 33 000 tonnes yr⁻¹ (Ahlgren et al. 2006), respectively. In any case the sediment P release from the investigated region has some quantitative importance.

The storage of potentially mobile P in the Baltic Sea accumulation sediments is comparable to the P amount in the water column, and the output from the sediments is in the same order of
magnitude as the input from land (Carman and Jonsson 1991, Conley et al. 2002). This study is an effort to quantify the mobile amount of P in the sediments in one Baltic coastal region. Further efforts to improve the knowledge of the P storage in other parts of the Baltic Sea are motivated by the quantitative importance of the P fluxes between sediment and water. Improved knowledge about factors and processes regulating burial and release of sediment P is crucial for the management of the Baltic Sea ecosystem.

Conclusions

Based on 102 coastal sediment surface samples from the Swedish east coast between Öregrund and Oxelösund we found that surface P$_{\text{tot}}$ concentrations varied between 840 and 7100 µg g$^{-1}$ dw with an average concentration of 1650 µg g$^{-1}$ dw and a median concentration of 1400 µg g$^{-1}$ dw. The standard deviation for the surface P$_{\text{tot}}$ concentrations was around 1250 µg g$^{-1}$ dw. In deep sediments the P$_{\text{tot}}$ concentration was typically around 1000 µg g$^{-1}$ (n = 85) and the variation was remarkably small. Excluding one outlier from the dataset, the standard deviation in the burial P$_{\text{tot}}$ concentration was 154 µg g$^{-1}$.

Phosphorus fractions were analyzed in detail in 12 sediment cores. On average 57% of the mobile P occurred in the top 2 cm of sediment. Total pools of mobile P varied between 1.5 and 18.2 g m$^{-2}$. Correlations between surface P$_{\text{tot}}$ concentrations and amounts of mobile P were strong ($r^2 = 0.88$). We estimate the amount of mobile P in the coastal sediments of the investigated region to between 1000 and 4000 tonnes. Assuming a turnover time of the mobile P between three and ten years gives an average annual release of between 100 and 1300 tonnes yr$^{-1}$, which is in the same order of magnitude as the regional P input from land.

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References


Emeljanov E.M. 2001. Biogenic components and elements...
Appendix. Information about sampling stations.

<table>
<thead>
<tr>
<th>Area</th>
<th>Lat. WGS-84</th>
<th>Long. WGS-84</th>
<th>Depth (m)</th>
<th>Wave base (m)</th>
<th>Oxic status</th>
<th>Water content 0–2 cm (%)</th>
<th>Loss on ignition 0–2 cm (%)</th>
<th>( P_{\text{tot}} ) 0–2 cm (µg g(^{-1}) dw)</th>
<th>( P_{\text{tot}} ) deep (µg g(^{-1}) dw)</th>
<th>Number of samples</th>
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