Global Biogeochemical Cycles

RESEARCH ARTICLE
10.1029/2018GB005914

A Century of Legacy Phosphorus Dynamics in a Large Drainage Basin

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Key Points:
- A three-parameter, two-box model describes phosphorus dynamics in the Baltic Sea drainage basin since 1900
- Transfers from "legacy" phosphorus pools that accumulated in previous years contribute about half of current waterborne loads to the sea
- After decades of accumulation, the mobile legacy pool appears to be depleting, suggesting that future loads to the sea could decrease

Supporting Information:
- Supporting Information S1
- Data Set S1
- Data Set S2
- Table S1
- Table S2
- Table S3

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Citation:
McCrackin, M. L., Muller-Karulis, B., Gustafsson, B. G., Howarth, R. W., Humborg, C., Svanbäck, A., & Swaney, D. P. (2018). A century of legacy phosphorus dynamics in a large drainage basin. *Global Biogeochemical Cycles*, *32*, 1107–1122. https://doi.org/10.1029/2018GB005914

Received 21 FEB 2018
Accepted 16 JUN 2018
Accepted article online 27 JUN 2018
Published online 20 JUL 2018

Abstract

There is growing evidence that the release of phosphorus (P) from "legacy" stores can frustrate efforts to reduce P loading to surface water from sources such as agriculture and human sewage. Less is known, however, about the magnitude and residence times of these legacy pools. Here we constructed a budget of net anthropogenic P inputs to the Baltic Sea drainage basin and developed a three-parameter, two-box model to describe the movement of anthropogenic P though temporary (mobile) and long-term (stable) storage pools. Phosphorus entered the sea as direct coastal effluent discharge and via rapid transport and slow, legacy pathways. The model reproduced past waterborne P loads and suggested an ~30-year residence time in the mobile pool. Between 1900 and 2013, 17 and 27 Mt P has accumulated in the mobile and stable pools, respectively. Phosphorus inputs to the sea have halved since the 1980s due to improvements in coastal sewage treatment and reductions associated with the rapid transport pathway. After decades of accumulation, the system appears to have shifted to a depletion phase; absent further reductions in net anthropogenic P input, future waterborne loads could decrease. Presently, losses from the mobile pool contribute nearly half of P loads, suggesting that it will be difficult to achieve substantial near-term reductions. However, there is still potential to make progress toward eutrophication management goals by addressing rapid transport pathways, such as overland flow, as well as mobile stores, such as cropland with large soil-P reserves.

Plain Language Summary

All life depends on phosphorus (P), which is why it is an important crop fertilizer. Humans generally consume more P than needed and the excess ends up in sewage systems. Past management of P in fertilizer and human sewage has led to the accumulation of P in soils and sediments of lakes and streams. This accumulation is called "legacy" P because it can leak for decades to downstream lakes and coastal areas where it contributes to environmental problems. We developed a model to understand P dynamics for the entire drainage basin of the Baltic Sea since 1900. This model included a rapid transport pathway that represented sources such as runoff from cropland and a slow pathway that represented leakage from mobile legacy sources. The model suggests that loss from the mobile pool contributes about half of current waterborne inputs to the sea; as a result, it could be difficult to make substantial near-term reductions. However, there are opportunities to meet environmental goals by slowing the accumulation of P in the landscape and by implementing measures that address the rapid transport pathway, such runoff from cropland, and the mobile stores, such as cropland with large soil-P reserves.

1. Introduction

Over the past century, human activities have substantially altered the global phosphorus (P) cycle. Phosphorus inputs to the biosphere have increased at least fourfold through the use of fertilizers produced from mined phosphate rock (Chen & Graedel, 2016; Falkowski et al., 2000). Over the same period, P fluxes to coastal oceans have increased at least threefold (Howarth et al., 1996; Seitzinger et al., 2010). Despite the potential for scarcity (Cordell et al., 2009), P is not used efficiently in the agri-food system. Less than half of P applied to cropland in mineral fertilizers, and manure is converted to harvested crop (Smil, 2000) and only 17–25% of P inputs are converted to food for human consumption (Ott & Rechberger, 2012; van Dijk et al., 2015). The remainder accumulates in agricultural soils, groundwater, lakes, and rivers along the land-to-sea continuum. These "legacy" sources can leak for decades, leading to time lags between the implementation of nutrient abatement measures and reductions in P loads to downstream water bodies (Haygarth et al., 2014; Powers et al., 2016; Sharpley et al., 2014). Such time lags are generally longer than political cycles and lead to questions about the efficacy of the measures.

A number of watershed models have improved our understanding of the effects of agricultural practices on P leaching and river nutrient loads (Radcliffe et al., 2009), but can be highly parameterized, have large data input requirements, and assume that nutrient balances are in steady state. Such models are often applied to small agricultural drainage basins and do not describe P dynamics at broader spatial scales or for areas with diverse land uses. Other approaches, such as P-flow analyses (van Dijk et al., 2015) and input-output budgets (Asmala et al., 2011; Hong et al., 2017), provide evidence that P is accumulating in the landscape, but do not give insight into the fate of this P.

Indeed, it is difficult to simulate P fluxes to receiving waterbodies when the accumulated P pool in the drainage basin is orders-of-magnitude larger than the annual loss from this pool. However, it is possible to build on existing data to estimate net P imports to the drainage basin and use historical reconstructions of waterborne P inputs to the sea to estimate the accumulation of P in the landscape (e.g., Powers et al., 2016). Understanding the "leakiness" of accumulated P poses a greater challenge. Phosphorus in soils is found along a continuum of availability, ranging from being dependent on slow weathering processes to being highly labile (Withers, Sylvester-Bradley, et al., 2014). Despite this uncertainty, it is critical to understand how past practices could affect future P loads to the sea in order to inform eutrophication management decisions.

Eutrophication is a pervasive and serious environmental problem in the Baltic Sea. Anthropogenic land-based nutrients have accumulated in the sediments and water column of the sea owing to long water residence times and limited exchange with the North Sea. This is especially true for the pool of P, which is nearly 3 times larger than preindustrial conditions (Gustafsson et al., 2012). The Baltic Sea is home to the world's largest anthropogenic hypoxic area (Conley et al., 2002). In what has been termed a "vicious circle," bottom-water hypoxia releases P into the water column and can fuel cyanobacteria blooms that, in turn, support anoxic conditions and high rates of P release from sediments (Funkey et al., 2014; Vahtera et al., 2007).

Currently, agriculture and municipal sewage together account for more than half of waterborne nutrient loading (total of riverine inputs and sewage effluent from coastal cities) to the Baltic Sea (HELCOM, 2011). Policies such as the European Union (EU) Urban Wastewater Directive and the Helsinki Commission's (HELCOM) Baltic Sea Action Plan (BSAP) have aimed to reduce nutrient inputs to the sea (Iho et al., 2015). Indeed, P loads have halved since the 1980s (to 31 kt P/year for 2012–2014), but remain above nonanthropogenic, background loads (11 kt P/year) and BSAP targets (22 kt P/year; Gustafsson et al., 2012; HELCOM, 2016; Savchuk et al., 2008, 2012).

In 2010, only about 4% of net anthropogenic P inputs (NAPI) to the Baltic Sea drainage basin were lost to the sea and the remainder was retained on land or in lakes and streams (Hong et al., 2017). Unlike the case of nitrogen, for which much "retention" is accounted for by denitrification losses to the atmosphere, the accumulation of P in the region could be substantial. Farmers have been advised to build up soil-P fertility in order to maximize crop production, resulting in high soil-P concentrations that persist in some areas today (Tóth et al., 2014). Reconstructed, global nutrient budgets of crop and livestock production systems suggest that about 50 Mt P has accumulated in agricultural soils in the Baltic Sea region between 1900 and 2010 (Bouwman et al., 2011). In addition, a substantial portion of NAPI is converted to human excreta (30% in 2010); P-rich wastewater was discharged to coastal and inland waters prior to the implementation of tertiary treatment in centralized wastewater treatment plants (WWTP; Naturvårdsverket, 2014; Swinarski, 1999).

The release of P from anthropogenic legacy pools could mask the effects of management actions taken to reduce agricultural losses and to improve sewage-treatment capabilities. For effective adaptive management of eutrophication in the Baltic Sea—and other human-dominated regional lakes and seas—we must better understand how past management practices could affect future P loads. Here we present an admittedly simple three-parameter, two-box model that describes P movement though mobile and stable storage pools for the entire Baltic Sea drainage basin and reproduces waterborne inputs to the sea. We investigate P dynamics in the Baltic Sea drainage basin over the past century using historical records and reconstructions of fertilizer imports, commodity trade, riverine fluxes, and discharge of sewage effluent from coastal cities. Our objectives are to (1) quantify annual NAPI for the entire drainage basin for years 1900 to 2013; (2) estimate the

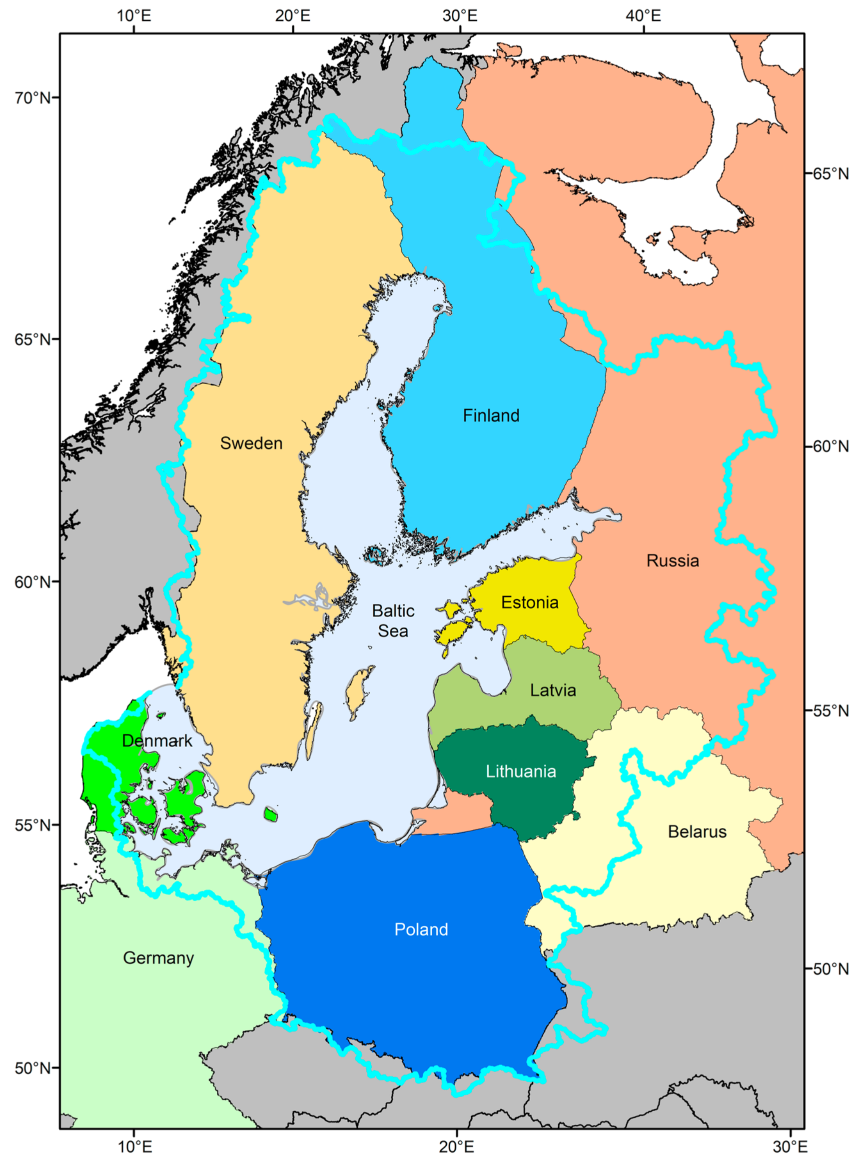

Figure 1. Map of the Baltic Sea drainage basin. Countries included in our analysis are shown in different colors; other countries are shown in gray. The blue line delineates the drainage basin border.

sizes of the mobile and stable P pools, the residence time of the mobile pool, and the extent to which loss from the mobile pool contributes to waterborne inputs to the sea; and (3) explore implications of mobile, legacy sources on future P loads to the Baltic Sea.

2. Methods

2.1. The Baltic Sea Drainage Basin

The Baltic Sea drains an area of 1.6 million km^2 that includes all or part of 14 countries (Figure 1). We do not further discuss the Czech Republic, Norway, Slovakia, or Ukraine because these countries occupy $<$3% of the basin area. There is a north-south gradient of land cover across the basin, with boreal forests and wetlands in the north and agricultural land in the south. There is a long history of human settlement in the region, which is currently home to about 90 million people. Agriculture is the dominant land use in Poland, northern Germany, Denmark, and southern Sweden.

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2.2. Net Anthropogenic Phosphorus Inputs

We estimated the accumulation of P in the drainage basin using a modified NAPI approach that accounts for "new" sources of P and not "recycled" sources, such as human excreta or animal manure. Traditionally, NAPI is estimated as the sum of net import P in fertilizer, detergent, food, and feed (Hong et al., 2017). The net food and feed component is derived as the difference between P in locally produced crops and dietary P needs of both people and livestock. Given the lack of historical data for such an approach, here we estimated net food and feed as the difference between P embedded in imports and exports of food, feed, and live animal commodities, similar to the approach used by Lassaletta et al. (2013) for nitrogen.

$$\mathrm{NAPI} = P_{fert} + P_{det} + P_{imp} – P_{exp} \tag{1}$$

where P_{fert} is imported fertilizer, P_{det} is P in detergent consumption, and P_{imp} and P_{exp} are P embedded in the import and export, respectively, of traded commodities.

We assumed that atmospheric P deposition is negligible, consistent with previous NAPI assumptions for the region (Hong et al., 2017). Components of NAPI for years 1900–2013 are given in Data Set S1 in the supporting information.

2.3. Data Sources

Due to data limitations, we used different approaches to estimate components of NAPI before and after 1961, the first year for which country-scale fertilizer and commodity trade data were available. For the years 1961 to 2013, our general approach was to use country-scale data from Food and Agriculture Organization Statistics (FAOSTAT, 2016) for P fertilizer consumption and quantities of traded food, feed, and live animal commodities. Phosphorus (P) embedded in trade was estimated as the product of a commodity-specific coefficient of P content and the mass of the commodity as reported in FAOSTAT (2016). Data for P coefficients were obtained from online databases (www.feedphosphates.com) and scientific literature (Text S1 and Table S1; El-Faer et al., 1991; Hong et al., 2011; Maertens et al., 2005; Quynh et al., 2005; Soltner, 1979; Swaney & Lassaletta, 2015).

We included country-scale data for Denmark, Estonia, Finland, Latvia, Lithuania, Poland, and Sweden in the NAPI budgets and only portions of data for Belarus, Germany, and Russia (Table S2 and Figure S1). Subcountry fertilizer data were available for 1995 to 2013, depending on country, from Hong et al. (2017). For years when such data were not available, the proportion of country-scale fertilizer that was consumed in the drainage basin was considered to be the same as that for more recent years; for Belarus, Germany, and Russia these proportions were about 42%, 18%, and 1.6%, respectively (Text S2 and Table S3). The portion of traded commodities in the drainage basin was estimated as the proportion of the total population living in the basin (Belarus 52%, Germany 9%, and Russia 9%; Text S3). Population data were obtained from FAOSTAT (2016) and https://www.clio-infra.eu/Indicators/TotalPopulation.html (Table S4).

Between 1961 and 1991, data for Belarus, Estonia, Latvia, Lithuania, and Russia were included with the former Union of Soviet Socialist Republics (USSR). Fertilizer consumption for years 1977 to 1991 for Estonia, Latvia, and Lithonia were based on Yurkovskis (2004) and the relative differences in utilized agricultural areas between these countries (Text S2; Eurostat, 2015). Otherwise, we back-calculated P fertilizer consumption and commodity trade based on the year-to-year percentage changes in those components for USSR. For example, year 1990 fertilizer consumption in the USSR was 188% of that for 1991; accordingly, we estimated year 1990 fertilizer consumption in Belarus as 188% of that for 1991 (Table S3).

To estimate P in imported detergents, we used a coefficient of 0.225 kg P $\cdot$ person^{-1} $\cdot$ year^{-1} (Wind, 2007). This coefficient was applied starting in the mid-1950s for Denmark, Finland, Germany, and Sweden and starting in the mid-1990's for Belarus, Estonia, Latvia, Lithuania, Poland, and Russia to reflect economic differences between these groups of countries (Text S3 and Table S4).

Data to support the calculation of NAPI as in equation (1) are lacking for years 1900 to 1960. Our overall approach was to back-calculate P in fertilizer and net commodity imports from 1961 to 1900 based on historical records of global phosphate rock consumption and global trade of agricultural products (Text S4). For example, Kelly and Matos (2016) showed that year 1960 global phosphate rock production was 90% of that for 1961; accordingly, we estimated year 1960 P fertilizer consumption in the Baltic Sea region as 90% of that for 1961. We repeated this process year by year to 1900 in order to create a trajectory in reverse. We compared our estimates of year-to-year changes in phosphate rock consumption to fertilizer-related data for the few years for which archival data were available (FAO, 1949, 1951, 1952, 1955) (Table S5). Overall, the changes between years for our estimates and archival sources were comparable, which we believe lends support to our approach. We used a similar back-calculation method with historical estimates reported by Federico (2005) to estimate P embedded in net imports of agricultural products from 1960 to 1900 (Text S4).

2.4. Breakpoint Analysis

The fraction of NAPI exported to the sea can be estimated as the slope of the linear relationship between NAPI and waterborne P loads (Swaney et al., 2012). To understand how this relationship changed over time, we conducted breakpoint analysis in *r* with *strucchange* (Zeileis et al., 2002, 2003). The minimum number of data points in each segment (h) was set to the default, 15% of the sample size ($n = 114$ for years 1900–2013). The algorithm minimizes the Bayesian Information Criterion, which penalizes models with more parameters (i.e., including more breakpoints) to determine the optimal number of breaks and minimizes the residual sum of squares using an iterative procedure to estimate the optimal break positions. The resulting segments have different slopes and intercepts.

2.5. Model of Anthropogenic Phosphorus Accumulation and Release

At the whole drainage-basin scale, NAPI has a number of fates (Figure 2a). In the form of mineral fertilizer, NAPI is converted to plant biomass and, if not taken up by plants, can accumulate in soils or be lost via overland flow or move through the soil profile and be transported further through drainage systems. Plants that are harvested and consumed, together with imported food and feed, are converted to human and animal biomass and excreta. Animal manure can be recycled in crop production as a fertilizer. Phosphorus in human excreta and detergents enters the waste stream where it can be discharged into surface waters or septic system drain fields as effluent. Phosphorus removed as sludge by WWTP can be recycled in agriculture, similar to livestock manure, or used for landfill. Phosphorus from sewage and agricultural sources that enters upstream surface waters can be converted to organic matter in the water column, buried in sediments, or transported downstream. Legacy P can be stored in relatively mobile pools, with timescales ranging from months to decades, and in relatively stable pools for centuries and longer. For example, loosely bound forms in soils and surface sediments and dissolved or particulate forms in surface waters that are mobile can be transported across the landscape to receiving waterbodies. In contrast, forms that are tightly bound in organic and mineral soil complexes and that are buried in deep sediments are retained in stable pools (Sharpley et al., 2014).

Building on this conceptual approach, we constructed a model that accounts for the accumulation and release of anthropogenic P in a temporary storage pool and reproduces waterborne P loads to the sea (Figure 2b). The model is described by three equations:

$$\frac{dP_m}{dt} = \left[\left(1 - a\frac{Q(t)}{\overline{Q}}\right)\cdot \mathrm{NAPI}(t) - F_{pc}\right] - b\frac{Q(t)}{\overline{Q}}P_m - cP_m \quad (2)$$

$$\frac{dP_s}{dt} = cP_m \quad (3)$$

$$F_{\mathrm{pwater}} = a\frac{Q(t)}{\overline{Q}}\cdot \mathrm{NAPI}(t) + b\frac{Q(t)}{\overline{Q}}P_m + F_{pc} + F_{pb} \quad (4)$$

Recognizing this to be a simplification, P is stored in a temporary, mobile pool and in a stable pool from which there are no losses (i.e., an unlimited P "sink" for the timescales we consider here). P_m and P_s are the size of the mobile and stable P pools, respectively, NAPI is net anthropogenic P input (equation (1)). The first term in brackets represents NAPI that is not transported to the sea via rapid transport (parameter *a*) or direct loading from coastal sewage effluent (F_{pc}) and which enters the mobile pool, P_m. In concept, transfers to the mobile pool includes, for example, P that accumulates in agricultural soils and NAPI that is converted to human sewage and enters waste treatment systems that do not discharge directly to the sea. The mobile pool is depleted by transfers to the sea (parameter *b*) and to the stable pool (P_s, parameter *c*) in proportion to the

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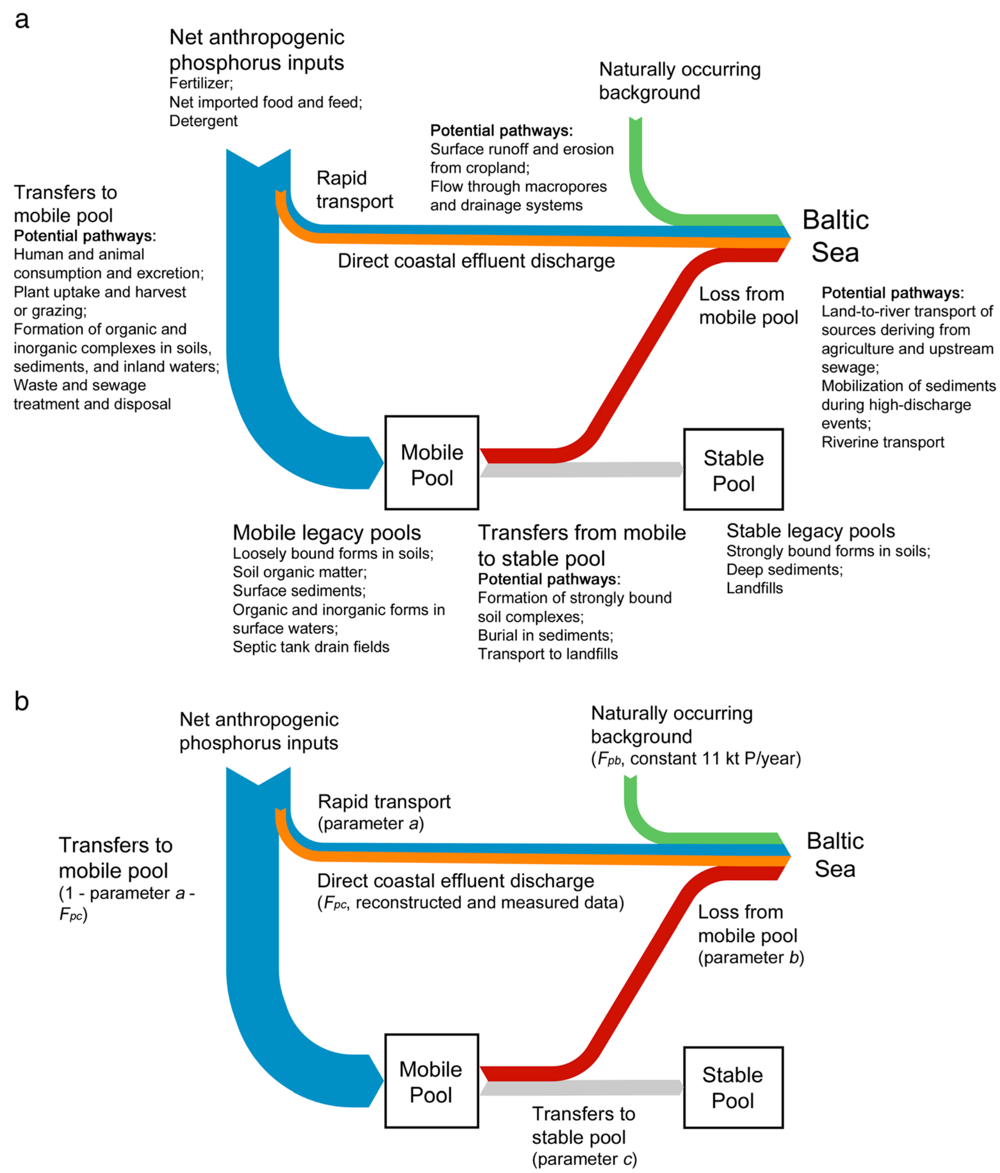

Figure 2. (a) Conceptual representation of anthropogenic phosphorus dynamics at the whole drainage-basin scale. (b) Conceptual diagram of the model structure. The arrows illustrate flux direction and connection between mobile and stable pools; the arrow widths are not in proportion to flux sizes.

accumulated storage of P in the mobile pool. We explicitly account for coastal effluent (F_{pc}) because it is known from reconstructions and HELCOM reports; other sewage-related fluxes are implicitly represented by parameters b and c. The stable P pool does not affect P loads to the sea but accumulates transfers from the mobile pool (equation (3)). Because P_m and P_s represent anthropogenic P only, we assume initial conditions of approximately zero and recognize that they represent storage since 1900. Waterborne load of total phosphorus to the sea (F_{pwater}; equation (4)) is the sum of (1) a portion of NAPI that is rapidly transported, (2) a portion of the mobile, legacy pool that is transported by slow loss pathways, (3) sewage effluent discharged from coastal cities (F_{pc}), and (4) a constant, preindustrial background load (F_{pb}) of 11 kt P/year (Savchuk et al., 2008).

Data for F_{pc} for years 1900–1994 were from Savchuk et al. (2012). Here coastal effluent discharge was reconstructed by piecewise, linear interpolation between estimates for a few years that were based on the

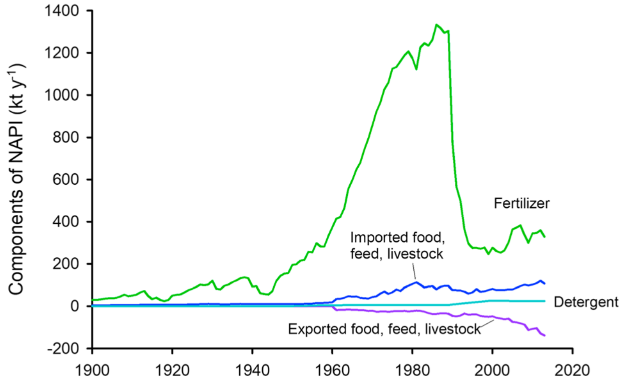

Figure 3. Annual components of net anthropogenic phosphorus inputs (NAPI) for years 1900 to 2013.

population of large cities or were obtained from periodic HELCOM reports and other technical papers; the reconstructions reflected improvements in P-removal capabilities by centralized WWTP over time (e.g., Naturvårdsverket, 2014). Data for years 1995–2013 were reported by coastal counties in HELCOM (2015).

Both rapid transport and loss from the mobile pool depend on riverine discharge (Q). $\overline{Q}$, the long-term historical average discharge (years 1900–2013), is used as a scaling parameter to express yearly discharge in relative terms. Units for pools and fluxes are Mt P and kt P/year, respectively.

2.6. Model Calibration

Coefficients a, b, and c were fit to the calibration data set by minimizing root-mean-square error (RMSE). We ran the model in *r* using *ode* for model integration and *optim* for parameter fitting (R Core Team, 2014; Soetaert et al., 2010).

The calibration data set included both reconstructed and country-reported data for riverine P loads and riverine discharge (Q; Data Set S2). Data for years 1900–1994 were from Savchuk et al. (2012). Here riverine P loads for years 1900–1970 were reconstructed by piecewise, linear interpolation between estimates for a few fixed years, due to a lack of measurement data. Data for 1971–1994 were based on measurement-derived discharge and concentration data that were collected with high coverage and resolution. For years 1995–2013, we used country-reported data from HELCOM (2015); P was total phosphorus in unfiltered water as estimated by peroxodisulfate oxidation, which included dissolved, particulate, organic, and inorganic forms (HELCOM, 2017).

We calculated Nash-Sutcliffe efficiency, a measure of the goodness of fit of model-derived waterborne P loads and the calibration data set to the 1:1 line (Moriasi et al., 2007).

2.7. Sensitivity and Uncertainty Analyses

2.7.1. Sensitivity Analysis

We conducted sensitivity analysis through a number of model “experiments.” First, we increased and decreased each best fit parameter a, b, and c individually by a factor of 2 and then ran the model to fit the remaining two parameters. Next, we fit the model with only parameter a (parameters b and c set to 0) to represent conditions where no mobile pool existed. Lastly, to represent conditions with no rapid transport component (all P routed to the mobile pool), we fit the model using only parameters b and c (parameter a set to 0). To estimate steady state anthropogenic P loads, each model was run to year 2500 with future NAPI held constant at the average of 2010–2013 levels.

2.7.2. Uncertainty Analysis of Net Anthropogenic Phosphorus Inputs

To reflect potential uncertainty in NAPI estimates, we fit the model using high and low estimates of NAPI and compared RMSE, parameters, and anthropogenic waterborne P loads to the best fit model. The fitting procedure was the same as described previously. The high and low estimates were calculated by increasing or decreasing, respectively, fertilizer by 5%, detergent by 16%, and imports and exports by 12% based on uncertainty ranges used by van Dijk et al. (2015) for P-flow analyses of European countries. To reflect greater uncertainty for years before 1961, we doubled the uncertainty value for each NAPI component (Text S5). Each model was run to year 2500 as described above.

2.8. Future Scenarios

To understand the potential effects of different P management strategies on future P loads, we constructed three scenarios for the 2013–2050 period. The “business as usual” (BAU) scenario held future NAPI constant at average rates for years 2010–2013 (359 kt P/year). The other two scenarios reflected reductions in NAPI through improved agronomic practices. Past reductions in P inputs to the Baltic Sea have been largely attributed to improved P-removal capabilities in WWTP (HELCOM, 2011); meeting the BSAP targets will require addressing diffuse sources from agriculture. One way to reduce diffuse losses from agriculture is by reducing over-fertilization and better using recycled sources, such as manure.

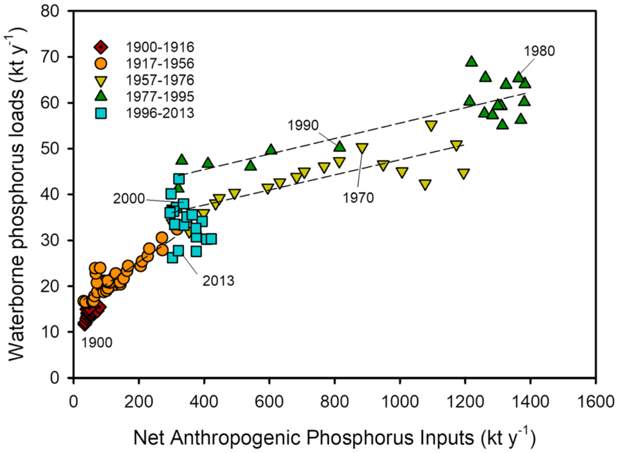

Figure 4. Relationship between net anthropogenic phosphorus inputs (NAPI) and waterborne loads to the Baltic Sea (from the calibration data set) for five time periods determined by breakpoint analysis. Regression relationships for all time segments are shown (broken lines) except for period 1996–2013 (blue squares), which was not significant ($p > 0.05$; Table 1).

Previous work found that P use-efficiency (PUE, the ratio of P removed in crop harvest to the sum of mineral P fertilizer and P in livestock manure) in crop production in the Baltic Sea region averaged 60% and ranged between 40 and 120% depending on country, suggesting that there is room for improvement (McCrackin et al., 2018). Farms with large amounts of manure in relation to agricultural area often apply more P than what crops need (resulting in low PUE), while crop farms with few or no animals must import fertilizers. Thus, there is potential to redistribute excess manure nutrients and, ultimately, reduce fertilizer imports. Such actions would reduce NAPI (equation (1)) and increase PUE.

Phosphorus in livestock manure and crop production are not explicit terms in the modified NAPI approach we use here (equation (1)). However, Hong et al. (2017) specifically calculated these terms to explore agricultural nutrient use efficiency in the Baltic Sea drainage basin and we used them for our scenarios. Hong et al. (2017) estimated P in livestock manure as the product of numbers of different types of animals and an excretion coefficient. Phosphorus in crop production was estimated as the product of harvested crop (either in tons or area) and a P-content coefficient.

For the scenarios, we reduced the P_{fert} component of NAPI such that basin-wide PUE improved from 60% to 80% and 90% (scenarios $PUE_{80\%}$ and $PUE_{90\%}$, respectively). Based on McCrackin et al., 2018, these scenarios are plausible because several countries in the region achieved PUE >90% in 2010. The scenarios reduced NAPI by 74 kt P/year (23% to 247 kt P/year) and 112 kt P/year (35% to 209 kt P/year) compared to 2011–2013 levels, respectively. We ran the scenarios to year 2050 to capture a long-term management window.

3. Results

3.1. Trajectories of Net Anthropogenic Phosphorus Inputs

Annual NAPI varied widely over the past century, reflecting economic and political changes across the region (Figure 3). Between 1900 and 1940, NAPI was relatively low and ranged between 30 and 145 kt P/year. After World War II, NAPI increased dramatically from 80 kt P/year in 1945 to 1,400 kt P/year in 1986 (average increase 6%/year). For the period 1989 to 1992, NAPI decreased sharply, coinciding with severe economic recessions in Poland, the Baltic States, Russia, and Belarus. Since then, NAPI increased at an average rate of <1%/year to about 360 kt P/year in the most recent years. Between 1900 and 2013, cumulative NAPI totaled 46.4 Mt P. Over the same period, waterborne loading transferred 2.4 Mt of anthropogenic P from land to the sea, leaving 44 Mt P in the drainage basin.

When examining the relationship between NAPI and P loads between 1900 and 2013, we found breakpoints at years 1916, 1956, 1976, and 1995 (Figure 4). There were strong, positive linear relationships for all time segments except for the period 1996–2013 (Table 1), where the slope of the relationship was negative but neither physically meaningful nor statistically different from zero ($p > 0.05$). The proportion of NAPI transferred to the Baltic Sea (slope of regression line) decreased from about 0.06 for years 1900–1916 to

Table 1
Linear Regression Equations for Segments Identified by Breakpoint Analysis

Time segment	Regression equation	r^2	p
1900–1916	P_{water} = 10.8 kt P/year + 0.062 × NAPI kt P/year	0.42	0.003
1917–1956	P_{water} = 15.5 kt P/year + 0.049 × NAPI kt P/year	0.80	0.0001
1957–1976	P_{water} = 31.1 kt P/year + 0.016 × NAPI kt P/year	0.68	0.0001
1977–1995	P_{water} = 38.7 kt P/year + 0.017 × NAPI kt P/year	0.80	0.0001
1996–2013	P_{water} = 48.5 kt P/year − 0.043 × NAPI kt P/year	0.14	0.1

Note. (P_{water}) is waterborne phosphorus loads from the calibration data set; NAPI is net anthropogenic phosphorus inputs.

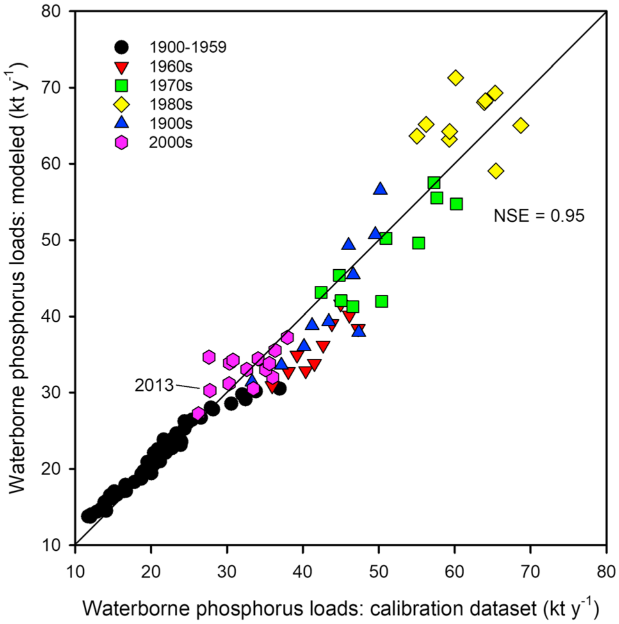

Figure 5. Comparison of model-derived waterborne total phosphorus loads with the calibration data set. Points are data for the years 1900 to 2013, with different symbols and colors to identify different time periods. NSE is Nash-Sutcliffe efficiency; diagonal line is 1:1.

about 0.02 for years 1957–1976 and 1977–1995. The intercept of the regression equations increased from 11 kt P/year for years 1900–1916, the background load, to nearly 40 kt P/year for the time segment ending in 1995.

3.2. Model Calibration

The best fit parameters were 0.0127, 0.0008/year, and 0.0317/year for *a*, *b*, and *c*, respectively, with RMSE of waterborne P loads of 0.17 kt P/year. The model underestimated loads between 1955 and 1971; for these years, the percent bias ranged from −7 to −19% (median = −13%; Figure 5). The model overestimated loads in the 1980s, when loads reached their maximum, with percent bias between 6 and 19% (median = 8%). For all years, median percent bias was 3%, suggesting a slight tendency to overestimate. Nash-Sutcliffe efficiency was 0.95.

3.3. Phosphorus Accumulation and Release

The model suggests that P is strongly retained in the landscape. Between 1900 and 2013, only 6% of cumulative NAPI entered the sea through one of three pathways: rapid transport (1% of NAPI), sewage effluent from coastal cities (3.5%), or losses from the mobile pool (1.5%). Over the same period, about 60% of NAPI was transferred to the stable pool. The balance remained in the mobile pool, which had a 27-year residence time.

Fluxes in and out of the mobile pool and to the stable pool varied substantially over time, reflecting changes in the magnitude of NAPI (Figure 6a), which was driven primarily by P fertilizer usage. Through the 1980s, inputs to the mobile pool exceeded transfers out, resulting in a net accumulation. The mobile pool peaked in 1991 at 23 Mt P and declined to 17 Mt P by 2013 because transfers out exceeded inputs. Since 1900, 27 Mt P accumulated in the stable pool (Figure 6b).

Sources of waterborne P loads similarly reflected the dynamics of changing NAPI and P-removal efficiency of coastal WWTP (Figure 7). Between the 1920s and 1960s, the rapid transport component increased from 6% to nearly 30% of loads and then decreased to 14% in recent years. The contribution of coastal sewage-related sources peaked at 42% in the 1940s, remained above 30% through the 1980s, and decreased to 8% in the 2000s. The contribution of loss from the mobile pool steadily increased to 46% of loads in the 2000s, when it became the single-largest source.

3.4. Sensitivity and Uncertainty Analyses

3.4.1. Sensitivity Analysis

Compared to the base model (RMSE 0.17 kt P/year), RMSE increased for all sensitivity experiments that we considered (RMSE between 0.18 and 0.33 kt P/year, depending on model) whereas residence times and steady state P loads both increased and decreased (Table 2 and Figures 8a and S2). Across all sensitivity experiments that included the mobile pool, the year 2013 pool ranged from 5 to 42 Mt P and steady state loads were between 22 and 58 kt P/year (*b* and *c* only and $a \times 2$, respectively) with corresponding residence times of 10 to >3,000 years. One model, $a \times 2$, did not reach steady state by year 2500. For all sensitivity experiments, the size of the mobile pool and magnitude of steady state anthropogenic loads increased with increasing residence times.

Cumulative P loading during the calibration period (1900 to 2013) remained within 5% of the base model, except if high rates of rapid transport were assumed ($a \times 2$, *a* only). Total storage in mobile and stable pools by 2013 was about 30 times the cumulative P load, with less than 1% difference between all sensitivity experiments that included a stable pool. RMSE stayed mostly within 8% of the base model but increased by 25–96% when the model structure was simplified (*a* only, *b* and *c* only) or a high rapid transport rate ($a \times 2$) was prescribed.

Two sensitivity experiments resulted in increases in future P loads compared to 2013 (which was about 31 kt P/year; Figures 8b and S3). When we increased rapid transport ($a \times 2$) or reduced the transfer from

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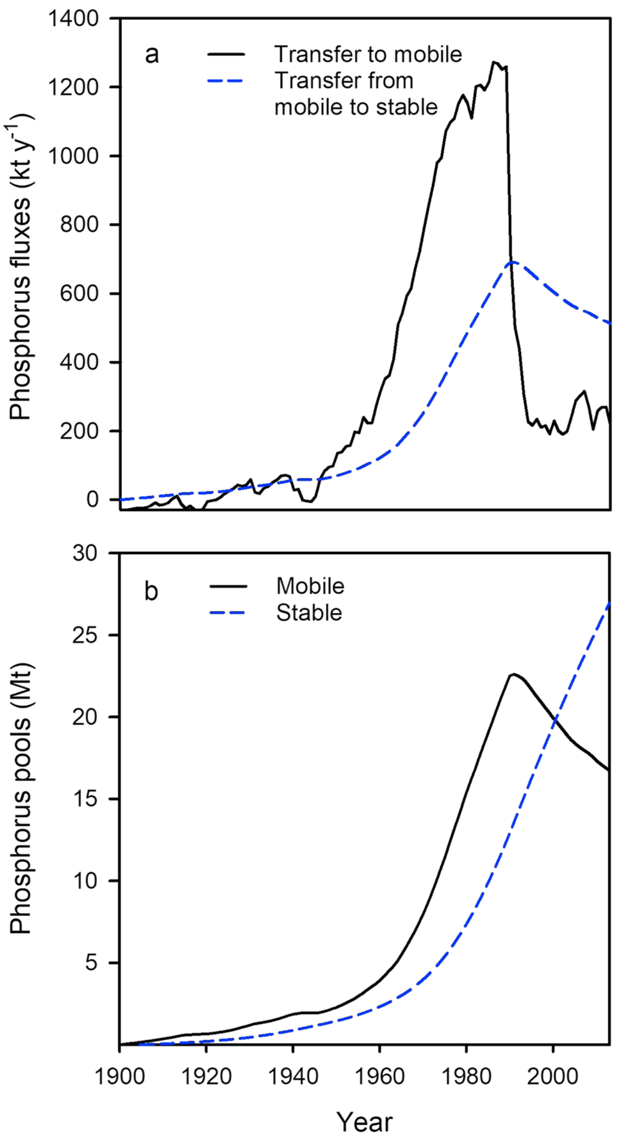

Figure 6. (a) Modeled fluxes of phosphorus to the mobile pool and from the mobile to the stable pool. (b) Modeled balances in the anthropogenic mobile and stable pools.

the mobile pool to the sea ($b \div 2$), more P accumulated in the mobile pool; steady state anthropogenic P loads increased 32% and 124%, respectively, compared to 2013. The other sensitivity experiments decreased steady state loads by a range of 13 to 54% ($c \div 2$ and *a* only, respectively) compared to 2013.

3.4.2. Uncertainty Analysis

High and low estimates of NAPI yielded cumulative values of 49.6 Mt P and 43.4 Mt P, respectively, for the 1900–2013 period (Figure S4). These estimates were within ±6% of base NAPI (46.4 Mt P). RMSEs for model runs using high and low NAPI were 0.169 and 0.172 kt P/year, respectively, compared to 0.170 kt P/year for the base model (Table S6). Using the high NAPI estimate, the proportion of NAPI that is lost via rapid transport (parameter *a*) decreased by 7% compared to the base NAPI model, the proportion of the mobile pool transferred to the sea (parameter *b*) did not change, and the proportion of the mobile pool transferred to the stable pool (parameter *c*) increased by 3%. Using the low NAPI estimate, parameter *a* increased by 9%, parameter *b* increased by 20%, and parameter *c* decreased 4% compared to the base NAPI model. The residence time of the mobile pool only changed under the low NAPI estimate, decreasing by one year. Waterborne P loads using high and low estimates of NAPI were nearly identical to that of the base model, both for the 1900–2013 period and future years (Figures S5 and S6).

3.5. Future Scenarios

We considered three scenarios of NAPI to explore the potential effects of P management. When NAPI was held constant in future years at 2011–2013 levels (BAU), P inputs to the sea decreased by 18% in 2050, because of net draw-downs of the mobile pool (Figure 9a). Scenario $NUE_{80\%}$ reduced NAPI by 23% compared to BAU; as a result, loads decreased by about 26% between 2013 and 2050. Under scenario $NUE_{90\%}$, NAPI was reduced by 35% compared to BAU. Corresponding loads decreased by 30% between 2013 and 2050.

The relative contributions of different P sources varied somewhat between the BAU and reduced NAPI scenarios (Figure 9b). Across all scenarios, however, transfers from the mobile pool were the largest anthropogenic source, followed by rapid transport and costal effluent sources.

4. Discussion

The scaling exercise we present here builds on previous studies that have shown the potential for anthropogenic P that has accumulated in the landscape to remain mobile for decades and contribute to eutrophication of surface waters (Haygarth et al., 2014; Powers et al., 2016). We estimated NAPI for the entire Baltic Sea drainage basin and developed a model to describe pools and fluxes of anthropogenic P over the past century. Model fit deteriorated the most in sensitivity experiments that manipulated rapid transport or removed the mobile pool, suggesting that waterborne P loads are best described by a model that includes both rapid transport and temporary storage components.

Since 1900, human activities have led to the net accumulation of 44 Mt P in the landscape, 17 and 27 Mt P in mobile and stable legacy pools, respectively (Figure 6b), virtually all of which originated as imported mineral fertilizer over the same period. For comparison, Bouwman et al. (2011) used a global nutrient budgeting approach and found that about 50 Mt P has accumulated in arable land and grasslands in the Baltic Sea region between 1900 and 2010. The amount of legacy anthropogenic P in the landscape is small compared to the naturally occurring, background pool. Mapped, global estimates suggest that background P in the top 50 cm of all soils in the drainage basin is 630 Mt P (Yang et al., 2013), 14 times larger than the legacy pools in 2013.

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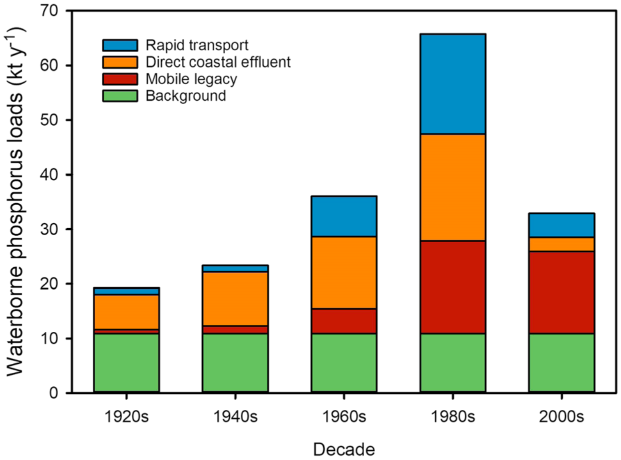

Figure 7. Model-estimated sources of waterborne phosphorus loads to the Baltic Sea over time.

Our approach assumes "one-way" transfers from the mobile to the stable pool, but accumulated P from mineral fertilizer and manure are retained in soils in a continuum of availabilities (Figure 2a). Field studies show that when fertilizer is withheld and the readily available labile pool has been depleted to a critical level, diffusion from the pool of more strongly bound soil-P can satisfy crop needs (Withers, Sylvester-Bradley, et al., 2014). Using agricultural soils as an analog, we speculate that at some point in the future there is the potential for net transfers from the stable to the mobile pool when the mobile pool becomes more depleted. At the whole-catchment scale, however, P will continue to accumulate even under the scenarios of reduced NAPI. By 2050, net drawdowns could reduce the mobile pool to between 8 and 12 Mt P under $PUE_{90\%}$ and BAU-levels of NAPI, respectively, which is about 50% to 70% of the year 2013 pool. It is unknown if the concept of a critical threshold for net transfers from the stable pool is relevant for spatial and temporal scales that we consider here; however, this is an interesting question to explore in future modeling efforts.

Previous studies have found that for a number of regions, NAPI is correlated to riverine P loads across watersheds and that the slope of the linear relationship is the proportion of NAPI delivered to coastal areas (Swaney et al., 2012). Hong et al. (2017) found, in addition, that the change over time in P loads to Baltic Sea subbasins was directly related to the corresponding change in NAPI. Here for the Baltic Sea region as a whole, we found temporal correlations between NAPI and P loads in several periods; Interestingly, the segment for years 1977 to 1995 captured the substantial increase and decrease in both NAPI and P loads. The breakpoint segments showed how relationships changed over time and ultimately decoupled due to legacy effects (Figure 4), which the model was able to reproduce (Figure 5). For example, between 1900 and 1995, P loads showed strong, positive relationships with NAPI during which the proportion of annual NAPI transported to the sea decreased from 0.06 for years 1900–1916 to less than 0.02 for years 1957–1995. The lack of a relationship between NAPI and waterborne P loads for years 1996–2013 resulted from mobile legacy sources that completely masked input-output relationships. Another factor contributing to the observed response was the dramatic reduction in direct effluent discharge from coastal cities, which decreased 55% between 1996 and 2013 due to improvements in sewage treatment, while NAPI decreased about 5% over the same period.

Caution should be used in interpreting the intercept term of the regression equations over time because it includes both nonanthropogenic and legacy-P effects. For example, while the intercept for the segment 1900–1916 was the same as preindustrial, background loads of 11 kt P/year, the intercept for years 1977–

Table 2
Results of Sensitivity Experiments

Sensitivity experiment	RMSE [1] (kt P/year)	Residence time of mobile pool (y)	Steady state waterborne phosphorus loads (kt P/year)	Mobile phosphorus pool in 2013 (Mt P)	Stable phosphorus pool in 2013 (Mt P)	Parameter *a*	Parameter *b* (per year)	Parameter *c* (per year)
Best fit	0.170	27	24.6	16.7	27.0	0.0127	0.00085	0.03150
$c \div 2$	0.173	54	28.3	25.8	17.9	0.0145	0.00057	0.01583
$b \div 2$	0.177	162	37.2	36.0	7.72	0.0161	0.00042	0.00669
$c \times 2$	0.179	14	22.6	8.11	35.6	0.0093	0.00154	0.06330
$b \times 2$	0.181	13	22.7	7.62	36.1	0.0080	0.00169	0.06503
$a \div 2$	0.184	14	22.8	8.50	35.2	0.0064	0.00170	0.06084
b and *c* only (no *a*)	0.213	10	22.0	4.99	38.8	0	0.00296	0.09170
$a \times 2$[2]	0.266	> 3460	55.7	42.1	1.48	0.0255	0.00018	0.00002
a only (no *b* and *c*)	0.335	n.a.	20.2	0	0	0.0260	0	0

[1]RMSE is root-mean-square error; sensitivity experiments are sorted in ascending order of RMSE. [2]The $a \times 2$ experiment never reached steady state, and values for year 2500 are shown, the final year of the model run period.

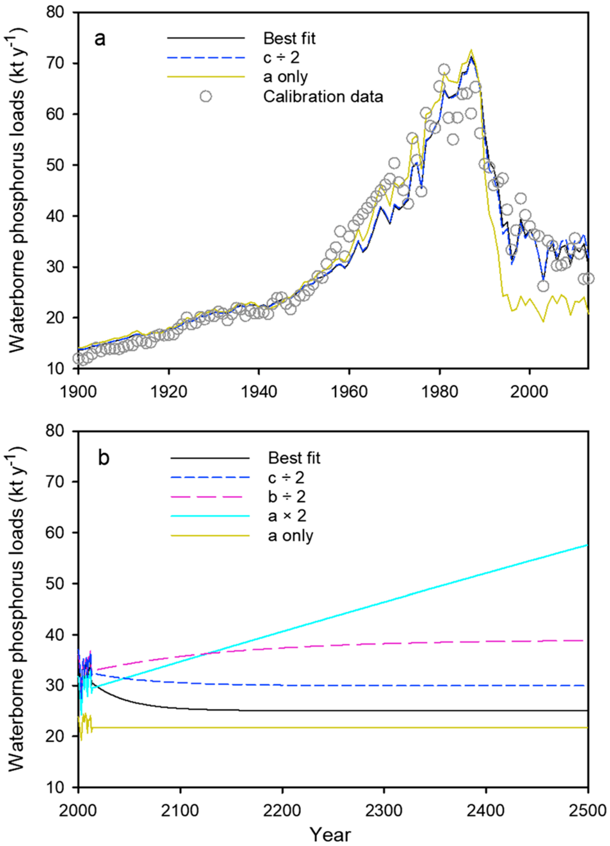

Figure 8. Comparison of phosphorus loads for best fit model and sensitivity experiments with lowest ($c \div 2$) and highest (a only) root-mean-square error (Table 2; all sensitivity runs shown in Figures S2 and S3). (a) For years 1900–2013, calibration data are also shown. (b) For years 2000–2050, results for $b \div 2$ and $a \times 2$ are also shown, the two cases where future waterborne loads increase over year 2013 levels.

1995 increased to about 40 kt P/year (Table 1). The increase in the intercept suggests that legacy sources are 29 kt P/year in 1995 (i.e., 40 kt P/year minus 11 kt P/year); these legacy sources represent 45 to 65% of total waterborne loads between 1977 and 1995, depending on year.

4.1. Implications for Eutrophication Management

The pulse of fertilizer imports between the 1960s and 1980s (Figure 3) appears to have been felt already. About 80% of the dramatic increase in waterborne P loads over this period can be attributed to rapid transport and loss from the mobile pool (Figure 7). Since then, modeled loads decreased by half, returning to 1960s levels. Over 40% of this decrease was due to reduced rapid transport, which resulted from the sharp drop in fertilizer consumption in Poland and the former Soviet republics. Improvements in coastal sewage treatment capabilities in the 1980s and 1990s had an even greater impact, accounting for more than half of the decrease in waterborne loads. Losses from the mobile pool also decreased between the 1980s and 2000s, showing that after years of accumulation the mobile pool has shifted to a depletion stage (Figure 6; Haygarth et al., 2014; Powers et al., 2016).

To meet the goals of the BSAP, P inputs to the sea must decrease by 30% compared to 2012–2014 levels by 2021 (HELCOM, 2016). While current loads are dominated by losses from the mobile pool, there is potential for future loads to decrease as drawdown of the mobile pool continues. The model suggests that waterborne loads could decrease by as much as 10% by 2021 and 18% by 2050, even if there are no further reductions in NAPI. The scenarios showed the potential benefit of reducing NAPI below current levels. Reductions in fertilizer imports (and NAPI) from 2013 levels could decrease loads by up to 15% to 17% between 2013 and 2021 and 23% to 30% between 2013 and 2050 (depending on scenario; Figure 9).

While sensitivity experiments found that future P loads could decrease from current levels, there were two cases when future loads increased ($b \div 2$, $a \times 2$; Table 2 and Figure 8b). The $b \div 2$ case performed nearly as well as the best fit model (RMSE: 0.177 kt P/year compared to 0.17 kt P/year, respectively) while $a \times 2$ performed poorly (RMSE: 0.266 kt P/year); hence, we only further explored $b \div 2$. When we halved b compared to the best fit model, rapid transport (parameter a) increased by 25% and transfers to the stable pool (parameter c) decreased by 80%. Together, these changes had the effect of retaining a greater proportion of NAPI in the mobile pool with a residence time of 160 years. Under these conditions and with BAU levels of NAPI, the mobile pool would continue to accumulate and not reach a depletion phase for centuries. Thus, while the best fit model and the $b \div 2$ model both reproduce historical loads (Figure 8a), they describe completely different futures. This work highlights the need for further investigation into the size, locations, and residence times of legacy P stores in order to inform nutrient reduction strategies and reduce uncertainty in the timescales over which mobile legacy sources could affect water quality.

4.1.1. Phosphorus in Agriculture

Our results suggest that P loading to the Baltic Sea is currently dominated by mobile legacy sources. While it will be difficult to achieve substantial near-term load reductions, nutrient abatement strategies should target both rapid transport (parameter a) and mobile legacy pathways (parameters b and c). Currently, 14% of waterborne P loads are attributable to the rapid transport pathway (Figure 7). In the model, rapid transport reaches the sea without passing through the legacy pool and is estimated as 1% of NAPI. Field and experimental studies have found that 0.7–46% of P fertilizer could be lost in runoff (Hart et al., 2004; Smith et al., 2016), although these results are not strictly comparable to ours due to obvious differences in spatial scales and methods. Both overland flow and vertical leaching combined with transport through tile

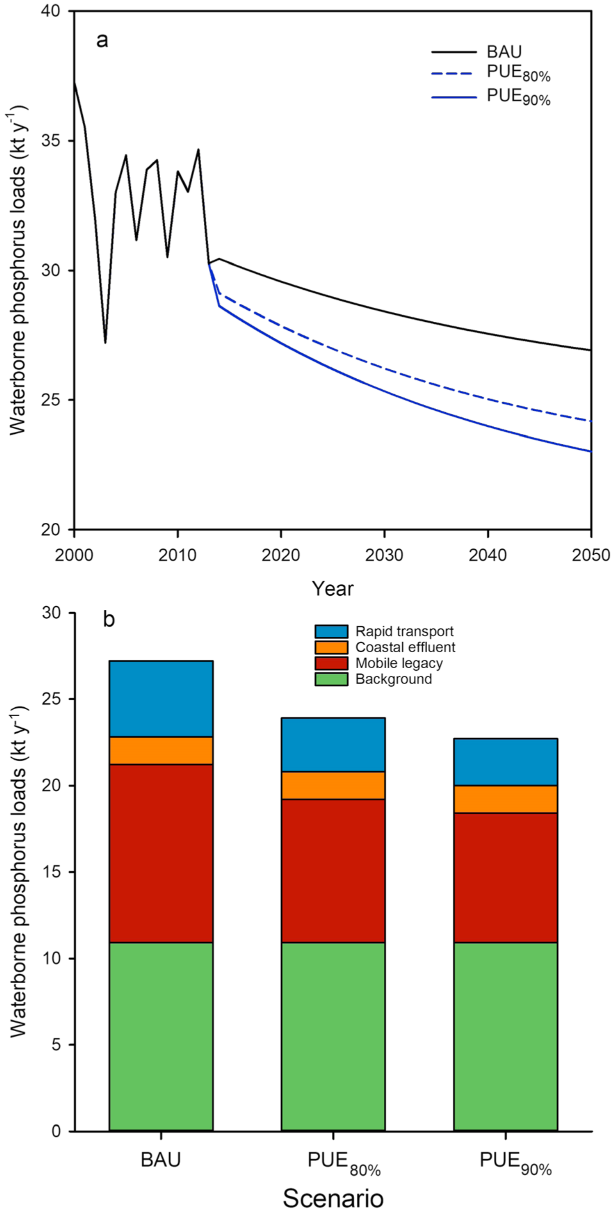

Figure 9. Comparison of scenario results. Under BAU (business as usual), net anthropogenic phosphorus inputs were held constant at 2010–2013 levels. Phosphorus use efficiency in crop production (PUE) was increased to 80% in $PUE_{80\%}$ and 90% in $PUE_{90\%}$. (a) Future estimated waterborne phosphorus loads for years 2013–2050. (b) Sources of waterborne phosphorus loads for year 2050.

drains are important pathways for rapid P transport from agricultural fields. More than half the arable land in the drainage basin is tile-drained (Feick et al., 2005). Rapid transport pathways could be managed, however, through the timing of manure and fertilizer application, manure and fertilizer application methods, choice of tillage, and crop rotations that reduce transport (Schoumans et al., 2014; Sharpley, 2016).

There also is potential to better utilize existing soil reserves, which would draw down the mobile pool and reduce losses to the sea. The 17 Mt P stored in the mobile pool (Figure 6) is nearly the same magnitude as 30 years of P in crop harvest (based on year 2010 crop harvest of 0.4 Mt P/year per Hong et al., 2017). This estimate falls within the range of 20 to 54 years reported by Rowe et al. (2015), who made similar estimates for western Europe. Not all mobile P is in cropland, but field-scale testing and soil mapping provide information that could be used to determine site-specific crop P needs. Once identified, soil-P reserves could be utilized by actively reducing the labile pool to agronomically critical levels. Rowe et al. (2015) also argued that "full exploitation" of legacy P requires a more holistic approach to agriculture that involves technological advances, such as breeding more P-efficient crops and engineering microbes to better mobilize soil P. Buffer strips and sedimentation ponds are examples of common practices to intercept nutrient loss and reduce transport to surface water (Schoumans et al., 2014; Sharpley, 2016). To be effective, these measures must be adapted to local conditions and informed by research that links farming practices with nutrient loss pathways (Withers, Neal, et al., 2014).

4.1.2. Phosphorus in Sewage

Improved sewage treatment in coastal cities has played an important role in reducing waterborne P loads to the sea (Figure 7). The contribution of direct effluent discharge decreased from 40% to 8% of loads between the 1980s and 2000s and accounted for 40% of the total decrease in loading between these decades. While WWTP capabilities have improved substantially in accordance with the EU Waste Water Directive, HELCOM has recommended even stricter standards for coastal areas (HELCOM, 2007), although this is not legally enforceable. Phosphorus-removal efficiencies in WWTP vary greatly in the region, averaging 63% for Latvia and 97% for Finland, Germany, and Sweden (Hautakangas et al., 2014). HELCOM estimated the potential for further WWTP upgrades to reduce inputs to the sea by 4.5–6.5 kt P/year (HELCOM, 2011), which is about 15% to 20% of current waterborne loads (31 kt P/year). Longer term strategies should focus on improved recycling; NAPI could be reduced by replacing imported mineral fertilizer with sewage-derived P and, ultimately, reduce waterborne P loads.

5. Conclusion

There is widespread recognition that landscape-scale nutrient legacies pose a challenge to setting eutrophication goals and implementing abatement strategies, yet appropriate models that can capture the storage and release of P over large areas and long timescales are scarce. The problem is exacerbated by the general lack of long-term data sets that are essential for identifying the role of legacy nutrients and properly parameterizing models to identify them (Haygarth et al., 2014; Zhang et al., 2017). Using a relatively simple approach that accounts for temporary storage in a mobile pool, we reproduced the long-term dynamics of waterborne P inputs to the Baltic Sea, which lends support to our model. Our work suggests that since the 1980s, P loads have decreased, due largely to reductions in rapidly transported anthropogenic sources of P and coastal sewage effluent. In the Baltic Sea drainage basin, P loads are currently dominated by legacy sources, which illustrate the need for a long-term perspective in eutrophication management. The potential to assess responses of the mobile pool to changes in NAPI offers important insight not only for the Baltic Sea but also for other regional waterbodies, such as Chesapeake Bay and the Laurentian Great Lakes.

Continued losses from the mobile pool pose a challenge to meeting the targets of the BSAP. The model suggests that the majority of fertilizer imported to the region since 1900 has ended up in a stable landscape pool; thus, the issue of the true long-term stability of this pool is a concern. There are opportunities to prevent further accumulation of P in the landscape, primarily by reducing fertilizer imports, by employing more efficient recycling of P in manure and human sewage, and by utilizing existing soil legacies. Thus, we must better implement current best management strategies and identify new approaches that improve water quality while sustaining or increasing food production.

Acknowledgments
This work was financially supported by Baltic Eye, the Baltic Nest Institute, and the Baltic Sea 2020 Foundation. We thank Oleg Savchuk, Geneviève Metson, and two anonymous reviewers for helpful comments that greatly improved this paper. The data used are listed in the references, tables, and supporting information.

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