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Nutrient retention and export to surface waters in Lithuanian and Estonian river basins

Arvydas Povilaitis, Per Stålnacke and Anatoly Vassiljev

ABSTRACT

The statistical model MESAW was applied to simultaneously estimate export coefficients and retention of nutrients in four Lithuanian and three Estonian river basins (range 946–8,388 km^2). This modelling approach uses non-linear regression to yield the export coefficients for total nitrogen and total phosphorus losses to surface waters, considering different land-use classes and retention of nutrients in the river network. The model was applied to data from 40 water quality monitoring sites and corresponding sub-basin data on land use, point sources and atmospheric deposition. The results showed that the studied river basins had a high nutrient retention capacity: 67–78 and 24–63% of total nitrogen and total phosphorus inputs remained in surface waters. The estimated retention was large in lakes: 27–59% for nitrogen and 11–31% for phosphorus. In-stream retention was apparently much lower, in the range 11–15% for total N and 3–12% for total P. Retention in lakes was lower in Estonia than in Lithuania due to the locations and smaller areas of the lakes in the Estonian basins. In Estonia, the highest relative retention in lakes was 46% for nitrogen and 35% for phosphorus. In-stream retention was also somewhat lower than in Lithuania, possibly because of the lower temperature in Estonian rivers in summer.

Key words | Estonia, export coefficients, Lithuania, MESAW, nutrients, retention

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INTRODUCTION

The Water Framework Directive (WFD) 2000/60/EC introduced new criteria and plans of action for managing Europe's water resources based on an integrated ecosystem-based approach. The aim of the WFD is to achieve good ecological and chemical quality status for all surface, ground and coastal waters by applying pollution-control measures that must be planned at the hydrological river basin level. For point and diffuse sources of pollution, the WFD requires identification of human pressures and their impact on water quality. To be able to analyse the current basin status and obtain estimates of the relative significance of the various sources of pollution, it is first necessary to characterize the water status, describe the effects of pollution sources, establish monitoring programs and implement river basin management plans (Kronvang *et al.* 2004).

Analysis of nutrient exports from river basins (i.e. from the sources to the mouths of rivers) requires quantitative knowledge of retention, especially if the goal is to determine the export from unmonitored diffuse sources of nutrients (Hejzlar *et al.* 2009). Nitrogen and phosphorus discharges from anthropogenic and natural sources are affected by temporary and more permanent sinks, and by cyclic and removal processes (e.g. denitrification and sedimentation in streams, lakes, reservoirs and flooded riparian areas). These internal retention processes in a river system should be taken into account when assessing the relative importance of nitrogen and phosphorus emissions from different sources. If this is not done when quantifying riverine load and source apportionment, the results regarding nitrogen and phosphorus losses from diffuse sources will be biased.

In earlier studies, only data on lakes were included in estimations of nutrient retention in river basins. However, various investigators (Billen *et al.* 1995; Behrendt 1996) have pointed out that it may be wrong to assume that the

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retention in river systems can be excluded from such calculations. Indeed, many studies have shown that there are also substantial losses of nitrogen in rivers (Behrendt & Opitz 2000; Saunders & Kalff 2001; Garnier et al. 2002; Wagenschein & Rode 2008; Rücker & Schrautzer 2010). Furthermore, there is evidence that measured denitrification rates may be higher in rivers than in lakes (Howarth et al. 1996), and that the sum of phosphorus inputs to a river system is larger then the observed transport (Probst 1985). Based on the cited studies, it can be concluded that both lakes and river networks influence nutrient retention, although such information is scarce for the Baltic States.

Nutrient source apportionment is generally achieved by inventories of point and diffuse sources. If all the sources and the gross emissions are known, total nutrient retention can be estimated as the difference between the total emissions and the load measured at the river outlet. This methodology can be divided into two regression categories: regression analysis between observed concentration and water discharge, and regression analysis between observed load and basin characteristics (Behrendt 1996, 1999; Grimvall & Stålnacke 1996). The development of dynamic process-based models has opened up new possibilities for nutrient source apportionment in large river basins.

Many water quality models have been developed to estimate pollution loadings to water bodies, and these range from simple regression-based to conceptual and physically-based approaches. Process-based models allow forecasting and a better understanding of processes, but they require a sizeable amount of detailed information on river basins, which is seldom available. This situation means that sophisticated models cannot be used to evaluate the large number of water bodies in which no water quality monitoring is conducted. Therefore, for decision makers and scientists who are faced with a specific water management problem, it is essential to choose a method that performs satisfactorily and provides the desired results even if input data are limited, and is also economically feasible (Lidén et al. 1999).

Simplified models that are based on the export-coefficient approach that addresses water quality issues have also been proposed in the literature (Johnes 1996; Worrall & Burt 1999; Grizzetti et al. 2005; Shrestha et al. 2008). These methods emanate from the idea that the nutrient load exported from a basin is the sum of the losses from individual sources, and the assumption that specific land use will result in emission of characteristic quantities of organic matter and nutrients to a receiving water body. However, reports have also indicated large differences in export coefficients for the same land-use categories (Smith et al. 1997; McFarland & Hauck 1999; Lepistö et al. 2006), and, in Lithuania, export coefficients have been shown to vary in different regions (Šileika et al. 2006; Šmitienė 2008). Hence, it is clear that export coefficients must be estimated for each region on the basis of available measurements.

The objectives of the present study were as follows: (i) to estimate export coefficients for different types of land use; (ii) to assess retention relative to nitrogen and phosphorus inputs at a river basin scale; and (iii) to distinguish between the retention in lakes (including reservoirs) and that in the stream network. To achieve those goals, the statistical model MESAW was used to evaluate nitrogen and phosphorus export coefficients and retentions in surface waters of four basins in Lithuania and three basins in Estonia.

STUDY AREAS AND DATA

Lithuania

In Lithuania, the study included the Merkys, Mūša, Nevėžis and Žeimena River Basins (here designated MER, MUS, NEV and ZEI, respectively; Figure 1). These areas are part of the Baltic Sea drainage area, and they cover 28% of the total area of Lithuania and represent diverse soil types, land use, hydrology and nutrient load conditions. Characteristics of the river basins during the period of modelling 1995–2006 (2000–2006 for ZEI) are summarized in Table 1.

The climate in these basins is transitional between maritime and continental. The mean annual air temperature is about +6 °C. Mean annual amounts of precipitation are similar in the northern and central river basins (MUS and NEV) but slightly higher in the south-eastern basins (MER and ZEI). In addition to the increased precipitation, the south-eastern basins show a spread of highly water-permeable sand and sandy loam soils that absorb snow and rainwater, and thus the values for specific runoff and base flow index (ratio of mean annual 30-day minimum runoff to mean annual runoff) are highest at MER and ZEI and much lower at MUS and

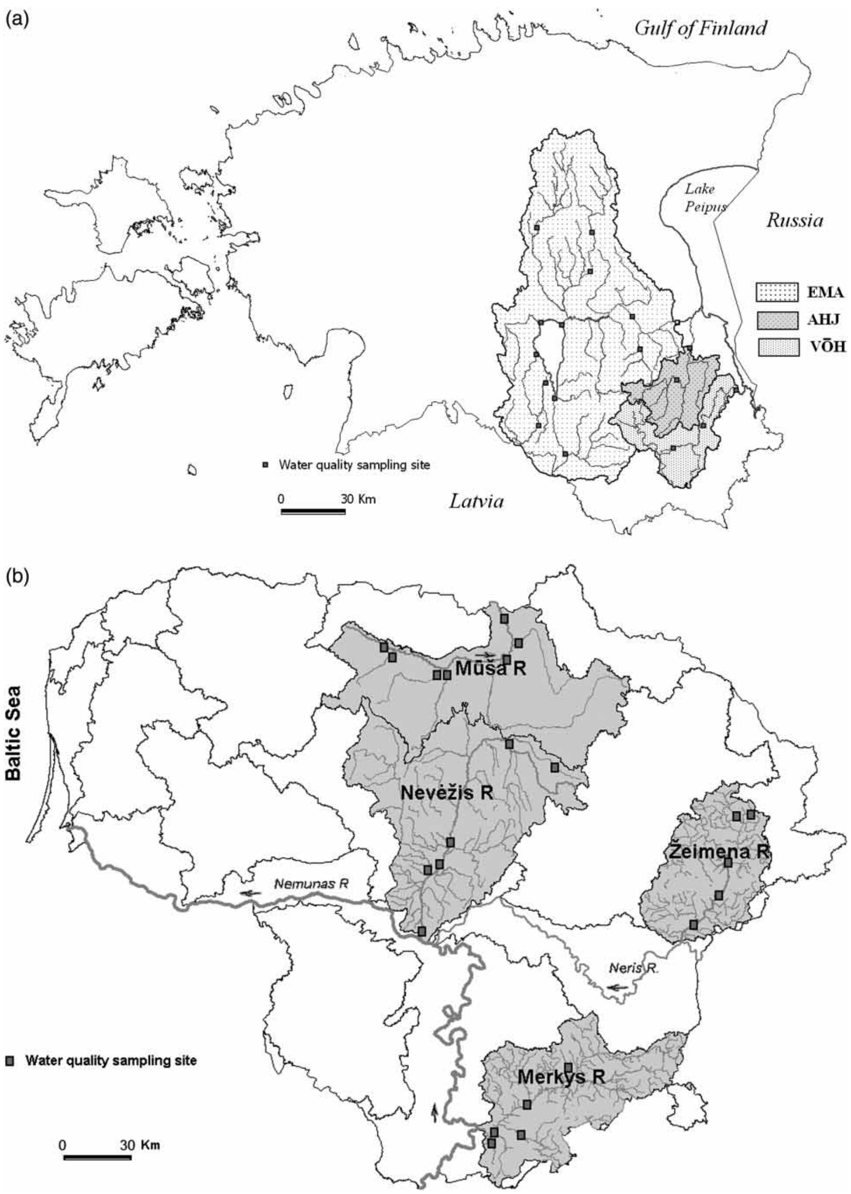

Figure 1 | Maps showing the locations of the river basins studied in Estonia (upper) and Lithuania (lower).

NEV. The basins also differ with regard to the presence of standing water bodies: ZEI has a large system of interconnected lakes (surface area >0.5 ha, $n = 479$); MER has numerous individual lakes ($n = 175$); NEV has artificial reservoirs (surface area >5 ha, $n = 107$); MUS has the smallest number of lakes and reservoirs (38 and 54, respectively). ZEI is one of the largest lake-dominated river basins in Lithuania, and that aspect, along with the runoff characteristics in the area, strongly influences the hydraulic load (defined as the annual runoff divided by the water surface area) in this basin.

MUS and NEV are dominated by agricultural land, whereas MER and ZEI are largely forested and also include 17–21% intensively farmed land. MUS and NEV represent typical fertile lowlands that have river systems with small differences in altitude and low flow velocities. During the last century, large-scale land reclamation measures were put in place in Lithuania to provide areas for agriculture. Consequently, up to 63 and 72% of the total areas of NEV and MUS, respectively, are drained artificially (mainly tile drainage). By comparison, ZEI and MER have waters that flow across hilly

Table 1 | River basin characteristics

Characteristic	MER^{LT}	MUS^{LT}	NEV^{LT}	ZEI^{LT}	EMA^{EST}	AHJ^{EST}	VÕH^{EST}
Total area, km^2	4.416	5.462	6.140	2.793	9.740	1.116	1.373
Artificially drained area, %	14.4	71.3	62.5	12.6	38.7	60.0	55.6
Area above the lowest downstream water quality sampling site, km^2	4.259	5.296	6.140	2.634	8.388	946.0	1,074
Mean annual precipitation, mm	720	630	580	730	646	638	640
Mean specific runoff, l/s km^{-2}	7.2	3.6	4.8	7.9	7.15	6.40	7.17
Hydraulic load of surface waters, m yr^{-1}	24.3	14.7	27.0	3.7	5.33	47.5	12.4
Base-flow index	0.66	0.13	0.05	0.63	0.56	0.53	0.47
Land cover, %							
Agricultural land	17.0	66.5	59.9	20.5	18.9	13.5	9.5
Forest	56.9	21.7	28.6	54.2	43.4	43.0	43.0
Wetlands	8.3	0.61	0.64	1.31	4.49	3.30	3.1
Pastures and meadows	13.7	5.4	7.3	15.4	9.6	11.3	9.3
Lakes and reservoirs/streams	0.82/0.21	0.94/0.20	0.43/0.20	6.8/0.19	3.7/0.53	0.12/0.35	1.26/0.37
Population density, inhabitants km^{-2}	16	36	25	20	26	18	25
Soils, %							
Sand	60.1	12.8	11.1	49.4	14.4	8.9	1.7
Loam	8.8	7.9	16,6	28.6	3.7	2.9	14.8
Clay	0.00	12.8	0.54	0.42	66.6	79.0	71.3
Peat	11.4	3.6	3.6	8.7	15	7.6	9.6
Mean slope of the main watercourse/ mean slope of tributaries, m km^{-1}	0.67/1.86	0.48/1.08	0.35/1.14	0.62/1.43	–/–	0.92/–	0.62/–
Flow-weighted concentrations at the lowest sampling site:							
N_{tot}, mg L^{-1}	1.525	4.471	4.211	0.962	2.072	1.578	1.363
P_{tot}, mg L^{-1}	0.101	0.134	0.196	0.053	0.079	0.098	0.087

forested areas that are less affected by human activity, and only the upper and middle reaches of these two basins are influenced by agriculture and urbanization. In all four basins, discharges from municipal wastewater treatment facilities are the largest point source contributors of nutrients.

Total nitrogen (N_{tot}) and total phosphorus (P_{tot}) were evaluated using monthly water quality sampling data obtained at 23 sites during the above-mentioned period. The actual sampling and chemical analyses were performed by the Lithuanian Environment Protection Agency. The same institution also provided the digital information that was used to delineate sub-basins and a database on the atmospheric deposition and point source emissions from 368 sites (32, 124, 167 and 45 in MER, MUS, NEV and ZEI, respectively). The load from atmospheric deposition was set to 9.5–10.0 kg ha^{-1} for N and 1.0–1.2 kg ha^{-1} for P. The digital CORINE land cover map was used to derive land use statistics for each of the 23 sub-basins where the water quality data had been collected.

The load of each water quality constituent was calculated as a function of daily concentration of the constituent and the stream discharge. Daily concentrations were estimated by linear interpolation between the values measured at two sampling events. Annual loads were obtained by summing the daily load values. Average annual loads for the period 1995–2006 were used in the MESAW model.

Daily data on continuous measurements of water discharge were provided by the Lithuanian

Hydrometeorological Service. Information on daily discharge at the sites that lacked measurements was obtained by linear regression using the data from the most adjacent sites with flow measurements.

The mean annual N_{tot} and P_{tot} concentrations at the lowest sampling sites were related to runoff in different ways (Figure 2). A negative correlation ($p < 0.05$) between P_{tot} and runoff at MUS and ZEI suggests that the importance of point sources decreased with increasing flow, whereas the values for the other basins were scattered and showed no significant correlations, which indicates the complexity of the processes that are involved in nutrient losses from the basins. In addition, the flow-weighted riverine concentrations listed in Table 1 reveal that the loads of nutrients entering the waters were much higher in MUS and NEV than in the other basins.

Estonia

The data from Estonia represent the Emajõgi, Ahja and Võhandu River Basins (here designated EMA, AHJ and VÕH, respectively; Figure 1), which cover 23% of the total area of the country. Characteristics of these basins during the modelling period 1993–2000 are summarized in Table 1. The mean annual air temperature is about $+5\,^{\circ}$C, and the mean annual precipitation is essentially the same in all three basins. Specific runoff is slightly lower in AHJ, and the base-flow index is highest for EMA and lowest for VÕH. EMA is also distinguished by having the largest lake (270 km^2), although VÕH has the largest total area of lakes.

The land in all three of the Estonian basins is dominated by forests. Artificially drained land represents around 60% of the total area in VÕH and AHJ, and about 40% in EMA. Drained peat soils occupy fairly large parts of all basins, and they predominate in the northern part of EMA. In all the basins, discharges from municipal wastewater treatment facilities constitute the largest point source contributors of nutrients.

Water quality sampling data from 17 sites were used to analyse N_{tot} and P_{tot}. The sampling and chemical analyses were conducted by the Estonian Ministry of the Environment. The same agency also provided digital information for delineation of sub-basins and data on atmospheric deposition and point source emissions. The digital CORINE land cover map was used to derive land-use statistics for each of the 17 sub-basins where the water quality data were collected. Considering the lowest sampling sites, the mean annual N_{tot} and P_{tot} concentrations were stable at VÕH

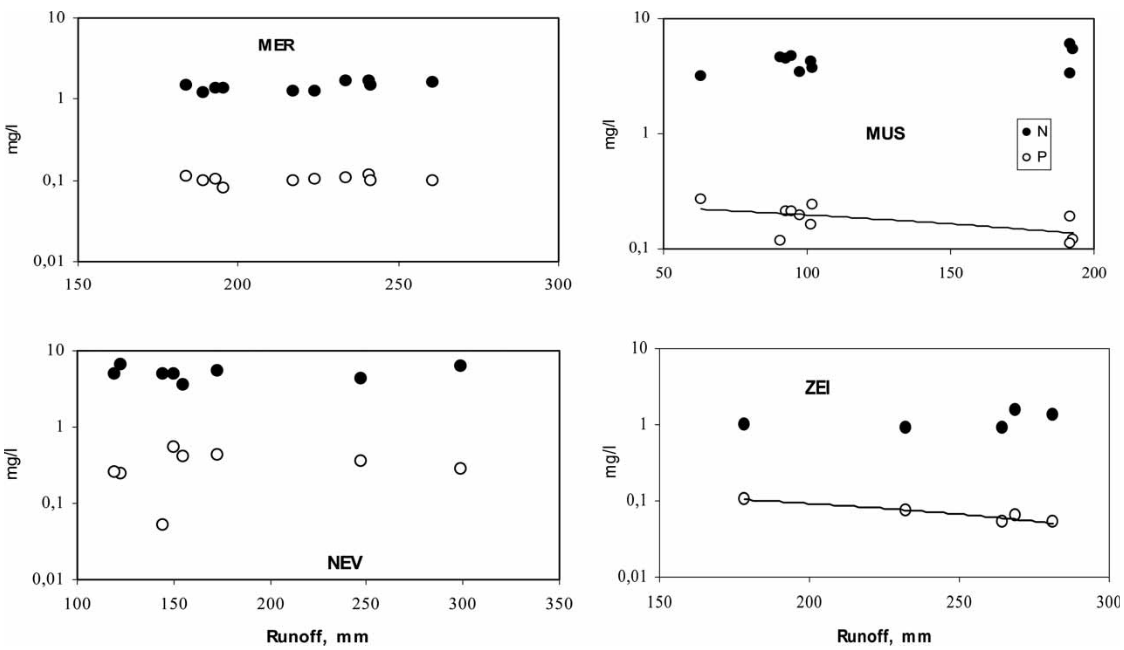

Figure 2 | Scatter plots of measured mean annual N_{tot} and P_{tot} concentrations (log scale) against runoff depth at the lowest sampling points of the Merkys (MER), Mūša (MUS), Nevėžis (NEV) and Žeimena (ZEI) basins. Trend lines show significant ($p < 0.05$) relationships.

and showed a pattern involving a slight decrease with increasing water flow at EMA and AHJ.

The annual load and daily concentrations of each water quality constituent were estimated using the same methods as described for the Lithuanian river basins. Average annual loads for the period 1993–2000 and for the year 2007 were used in the MESAW model. Daily data on continuous measurements of water discharge were provided by the Estonian Meteorological and Hydrological Institute.

METHODOLOGY

MESAW model

MESAW is a statistical model for source apportionment of the riverine transport of pollutants (Grimvall & Stålnacke 1996). This model uses non-linear regression for simultaneous estimation of source strength (i.e. export coefficients for loadings to surface waters) for different land-use or soil categories and retention coefficients for pollutants in a river basin. The basic principles and major steps in the procedure are as follows: (1) estimation of riverine loads at each water quality monitoring site; (2) subdivision of the entire drainage basin into sub-basins, defined by the water quality monitoring sites and their upstream–downstream relationships (describing the river system); (3) derivation of statistics on, for example, land use, lake area, point source emissions and other relevant data for each sub-basin; and (4) application of a general non-linear regression expression with loads at each sub-basin as the dependent/response variable and sub-basin characteristics as covariates/explanatory variables (French et al. 2003).

The load at the outlet of an arbitrary sub-basin is estimated using the following general expression (Vassiljev & Stålnacke 2005; Vassiljev et al. 2008):

$$L_i = \sum_{j=1}^{n} (1 - R_{i,j})L_j + (1 - R)S_i + (1 - R)P_i + (1 - R)D_i + \varepsilon_i \quad (1)$$

where L_i is the load at the outlet of sub-basin i; L_j is the load at the outlet of the nearest upstream sub-basin j; $R_{i,j}$ is retention occurring from the outlet of sub-basin j to the outlet of sub-basin i; n is the number of sub-basins located nearest upstream; S_i denotes total losses from soil to water in sub-basin i; P_i represents point source discharges to waters in sub-basin i; D_i stands for atmospheric deposition on surface waters in sub-basin i; R is retention in sub-basin i; ε_i is the statistical error term.

The load at each sub-basin is decomposed into contributions originating from sources located in sub-basins further upstream (the first term in Equation (1)) and those coming from sources located within the sub-basin under consideration (the S_i, P_i and D_i terms). The parameterization of the model is flexible and can be specific to the study area. The model is fitted by minimizing the sum of squares for the difference between observed and estimated load. In this study, it was assumed that P_i and D_i were known, and that S_i was a simple function of land use according to:

$$S_i = \beta_1 a_{1i} + \beta_2 a_{2i} + \beta_3 a_{3i} \quad (2)$$

where a_{1i}, a_{2i} and a_{3i}, respectively, denote the area of agricultural (arable) land, forests and wetlands (combined area because most wetlands are located in forested land), and pastures and meadows (combined) in sub-basin i, and β_{1-3} are unknown emission/export coefficients for the land-use categories. The point source emission (P_i) and atmospheric deposition (D_i) are allocated to the respective sub-basin.

Nutrients are normally retained temporarily or permanently in watercourses. Therefore, retention in the model is given as a summary expression for all hydrological and biogeochemical processes that may decrease the transport or losses of nutrients. It can be parameterized by any empirical function. In this study, the retention was best estimated according to the following equation:

$$R = 1 - \frac{1}{1 + (PAR \cdot X)} \quad (3)$$

where PAR is an unknown parameter estimated by the model, and X is a suitable covariate (i.e. explanatory variable) such as water surface area, lake surface area, specific runoff, hydraulic load or drainage area of the sub-basin.

The water surface area (A_s, km^2) in a river basin can be calculated using the total area of all lakes and reservoirs

(A_{Lake}, km^2) shown in the CORINE land cover map, and the stream (river network) surface area obtained by applying the equation proposed by Behrendt & Opitz (2000):

$$A_s = A_{\mathrm{Lake}} + 0.001 \cdot A^c \tag{4}$$

where A is the total area of the basin (km^2), and c is the power coefficient.

The mentioned equation was adopted to estimate water surface areas in the Estonian and the Lithuanian river basins which were given as $c = 1.185$ and 1.100, respectively.

In the MESAW model, the retention in lakes and in the river network can be parameterized separately according to Equation (3), and thus the user can select up to two covariates for retention. In addition to distribution of different land-use classes, these covariates serve to explain the observed riverine loads. Table 2 shows the covariates that best distinguish between retention in lakes and reservoirs and retention in the river network. The PAR values corresponding to appropriate cases vary from 6.80×10^{-4} to 1.98×10^{-1} ($p < 0.05$).

Retention from an arbitrary sub-basin m to the river mouth R_{mouth} is derived from:

$$R_{m,\mathrm{mouth}} = 1 - \prod_{j=1}^{kk} (1 - R_j) \tag{5}$$

where $R_{m,\mathrm{mouth}}$ is retention occurring from the outlet of the sub-basin m to the mouth of the river; kk is the number of sub-basins downstream of sub-basin m; R_j are the values of retention within the sub-basins downstream of sub-basin m.

Lastly, the estimated export coefficients β_{1-3} and the retention parameters are used to calculate the contribution from each source and sub-basin to the riverine load at the outlet. The advantage of the MESAW model is that the export coefficients and retention are evaluated simultaneously.

Nutrient retention descriptors

Three variables were used to describe N and P retention in surface waters: (i) relative retention (R_N^r and R_P^r, %), i.e. the proportion of nutrients from all basin sources that is retained in surface waters; (ii) specific retention per area of surface waters in the basin (R_N^{Sp} and R_P^{Sp}, kg ha^{-1} yr^{-1}), which expresses the intensity of retention processes in water bodies in relation to hydraulic conditions and nutrient loss processes; and (iii) allocated relative retention of nutrients from all basin sources in lakes and reservoirs (R_N^{LR} and R_P^{LR}, %) and the river network (R_N^{RR} and R_P^{RR}, %). The latter variable was presented as a covariate-weighted average for each river basin derived from retention in sub-basins under covariates.

RESULTS

Estimation of riverine loads

Over the study period, the riverine nutrient loads at the sampling sites varied with the inputs from the basins and the runoff volumes. Figure 3 presents a comparison of the observed and calculated mean annual loads of N_{tot} and P_{tot} for all sampling sites (outlets of sub-basins).

Table 2 | Covariates[a] used to estimate nutrient retention

		River basin						
Type of hydrographic network	**Constituent**	**MER^{LT}**	**MUS^{LT}**	**NEV^{LT}**	**ZEI^{LT}**	**EMA^{EST}**	**AHJ^{EST}**	**VÕH^{EST}**
Lakes and reservoirs	N_{tot}	LS	HL	HL	WA	LS/WA	LS/WA	LS/WA
	P_{tot}	LS/WA	LS	HL	A	LS/WA	LS/WA	LS/WA
Rivers and streams (river network)	N_{tot}	A	RS	RS	RS/WA	WA$^{0.5}$	WA$^{0.5}$	WA$^{0.5}$
	P_{tot}	HL	HL	WS	WS	WA$^{0.5}$	WA$^{0.5}$	WA$^{0.5}$

[a]LS/WA = lake and reservoir surface area divided by the total water surface area; RS/WA = river and stream surface area divided by the total water surface area; HL = hydraulic load; LS = lake and reservoir surface area; RS = river and stream surface area; WA = water surface area.

In Lithuania, the riverine loads of nitrogen were found to be lowest in the ZEI and MER sub-basins (from 30×10^3 to 630×10^3 and from 50×10^3 to $1{,}450 \times 10^3$ kg yr^{-1}, respectively) and highest in the MUS sub-basins (from 360×10^3 to $5{,}600 \times 10^3$ kg yr^{-1}). The highest transport of phosphorus occurred at the outlets of the MER and NEV sub-basins (from 2×10^3 to 100×10^3 and from 2×10^3 to 150×10^3 kg yr^{-1}, respectively), whereas the lowest riverine P_{tot} load was measured at the outlets of the ZEI sub-basins (from 2×10^3 to 35×10^3 kg yr^{-1}).

The observed nitrogen loads in Estonian rivers varied from 59×10^3 kg yr^{-1} for the smallest sub-basin in the year with the lowest load to $4{,}874 \times 10^3$ kg yr^{-1} for the largest river (EMA) in the year with highest load. The corresponding values for phosphorus are 2×10^3 and 182×10^3 kg yr^{-1}, respectively. The loads at AHJ and VÕH varied much less and were, respectively for these two basins, 96×10^3 to 398×10^3 and 62×10^3 to 360×10^3 for nitrogen, and 3×10^3 to 26×10^3 and 3×10^3 to 37×10^3 kg yr^{-1} for phosphorus.

The MESAW model performed well in estimating the loads. Absolute values of the deviation between observed and calculated loads varied by 1–15% from the 1:1 line (Figure 3). The Nash–Sutcliffe coefficient showed 90–99% modelling efficiency. In turn, the loads of each water quality constituent from each sub-basin were set as a dependent variable to derive source strength (i.e. export coefficients) and retention.

Export coefficients

The multiple regression algorithms in the MESAW methodology were used to estimate the loading from each land-use type. The dependent variables were the annual loads of constituents, and the independent variable was the proportions of different land-use categories in each sub-basin. The results of the analysis are summarized in Table 3 as estimated export coefficients for diffuse sources under the average conditions of three land-use classes in the basins. All the coefficients are significant at $p < 0.05$, which indicates that the land-use categories used as independent variables explained a large proportion of the differences in loadings.

For the river basins in Lithuania, the results showed that the losses of N_{tot} from agricultural land were four to six times higher than the corresponding losses from forested land and pastures and meadows. The same pattern was not found for P_{tot}; in short, the export of phosphorus was only slightly larger from agricultural land than from pastures and meadows or forested land. Forested areas with average losses of 2.6 kg ha^{-1} yr^{-1} N_{tot} and 0.15 kg ha^{-1} yr^{-1} P_{tot} represented the least diffuse source contributions of nutrients. The results also showed that the P_{tot} export coefficients for all land-use classes were significantly lower than the atmospheric deposition rate, and this was probably due to phosphorus being subject to a high adsorption capacity, plant uptake and limited release from the soil. For N_{tot}, significantly higher export from agricultural land compared with the rates of atmospheric deposition reflects the effect of applied fertilizers.

The highest estimates of N_{tot} emissions from agricultural land (19–20 kg ha^{-1} yr^{-1}) and from pastures and meadows (4.9–5.1 kg ha^{-1} yr^{-1}) were noted for the NEV and MUS, where there is intense crop and cattle farming, and nitrogen retention processes in the soil are probably restricted by the relatively large area of tile drainage.

The estimated rates of N_{tot} losses from arable land and from perennial grass areas were much lower (40% on average) for MER and ZEI than for NEV and MUS. Although the mean annual precipitation is higher in MER and ZEI, these two basins have substantially lower proportions of artificial drainage, and they overlie sandy aquifer outcrops. Therefore, a larger amount of the precipitation can infiltrate the soil, and consequently greater quantities of nitrate–nitrogen can be temporarily stored or removed by denitrification in these basins.

The finding that the rates of total phosphorus emissions from agricultural land were comparatively (1.4–2.0 times) higher in MER can be explained by the steeper slopes and greater precipitation in those areas, conditions that increase the washout of soil particles and loss of particulate phosphorus caused by erosion.

Considering nutrient emissions in the Estonian river basins, estimates of nitrogen exports from agricultural land were much higher for EMA than for AHJ or VÕH. This can be explained by greater runoff from the part of EMA that has a larger portion of agricultural land. On the other hand, the larger proportion of drained peat soils in EMA might be a significant source of nitrogen in that basin. As noted for

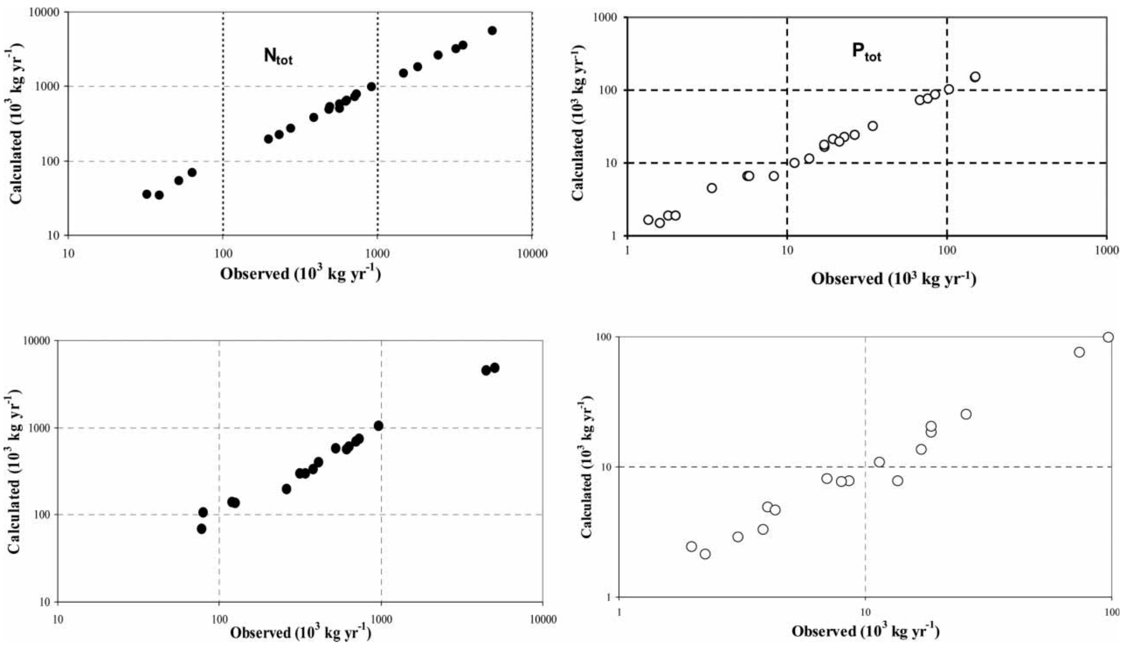

Figure 3 | Observed total nitrogen and total phosphorus loads versus loads calculated using the MESAW model (log scale), shown for the Lithuanian (upper) and Estonian (lower) sub-basins.

Table 3 | Average export coefficient estimates (kg ha^{-1} yr^{-1})

		River basin						
Land-use class	**Constituent**	**MER^LT**	**MUS^LT**	**NEV^LT**	**ZEI^LT**	**EMA^EST**	**AHJ^EST**	**VÕH^EST**
Agricultural land (arable)	N_{tot}	13.7	19.2	19.9	10.9	16.2	9.3	9.4
	P_{tot}	0.30	0.14	0.22	0.17	0.4	0.41	0.39
Forest and wetlands	N_{tot}	2.53	2.85	3.1	1.9	3.1	2.7	2.9
	P_{tot}	0.20	0.13	0.14	0.13	0.04	0.04	0.04
Pastures and meadows	N_{tot}	3.00	4.9	5.1	2.1	6.2	3.8	3.7
	P_{tot}	0.18	0.14	0.19	0.17	0.38	0.39	0.36

the Lithuania sub-basins, the export of nitrogen in Estonia was much greater from arable land than from pastures and forested lands. Furthermore, the rate of phosphorus release in Estonia was almost the same for arable land and pastures, but was much lower for forests, possibly due to very limited erosion from soil in the forested areas.

In general, there were no significant differences between the Lithuanian and Estonian river basins with respect to the estimates of nitrogen export, whereas the corresponding values for phosphorus varied considerably. Phosphorus exports from agricultural land were as much as two times lower in Lithuania, and in Estonia P losses were three to five times lower from forested land and more than two times higher from pastures and meadows. These differences can be attributed to the features of land use in a basin. It is believed that export of phosphorus depends largely on the areal distribution of land use within a basin. For example, if there is a much larger proportion of agricultural land than forests, in an analysis this should be considered as a case of forest located within an agricultural area. However, if the opposite is true, i.e. there is much more forested than agricultural land, it should be regarded as a case of agricultural areas situated within forested areas. In the latter case, the forests will retain much of the nutrients that are transported towards the river network. This situation is more common in Estonian river basins, which have a larger portion of forests surrounding agricultural areas. However, the Estonian basins have much hillier topography and more intense soil erosion, whereas there are more farming and grass-covered areas in Lithuania, and thus phosphorus emissions are larger in the latter country.

Although average annual export values are presented in Table 3, analysis of the data showed that nutrient losses depend on the runoff and vary from year to year. Therefore, the relationship between the export coefficients of nutrients and the runoff was explored according to the following:

$$EC = EC_{av} + k \cdot (h_w - h_{w-av}) \qquad (6)$$

where EC is the export coefficient for runoff depth h_w (kg ha^{-1} yr^{-1}); EC_{av} is the average annual export coefficient (see Table 3); $h_{w\text{-av}}$ is the average annual runoff depth (181 and 209 mm for Lithuania and Estonia, respectively); and k is the empirical coefficient (see Table 4).

Equation (6) can be applied within the limits of 63–299 mm in Lithuania and 119–300 mm in Estonia.

Nutrient retention

The N_{tot} and P_{tot} retention estimates obtained for the studied basins are presented in Table 5. The relative retention of nitrogen, R_N^r, ranged from 67 to 78% and did not differ significantly between the basins, which clearly illustrates the high nitrogen retention capacities in the hydrographic network. In contrast, the relative retention of phosphorus, R_P^r, differed significantly (9–64%) among the basins. The highest R_P^r value was found for ZEI, apparently due to the large area of the lakes and reservoirs in that basin. The lowest R_P^r was noted for MER, probably because streambed altitudes in that basin (compared with the other basins) decreased more markedly along the river network (Table 1), which implies higher flow velocities and consequently a weaker capacity to retain sediment and phosphorus. It is assumed that estimated R_P^r values for the NEV and MUS represent typical P retention in Lithuanian lowland river basins.

The lake and reservoir area is small in MUS and NEV, and both those basins were found to have much higher specific retention of nitrogen, R_N^{Sp}, compared with the basins with large lake/reservoir areas (e.g. ZEI). Essentially the same pattern, albeit not as pronounced, was found for the R_P^{Sp} values: the estimates of specific phosphorus retention were highest for NEV. These regularities were presumably caused by the conditions comprising a relatively small water surface area and low flow velocity that are prevalent in NEV.

As mentioned above, the intention here was to distinguish between the retention in lakes and reservoirs and the retention in the stream network in each basin. The results expressed as R_N^{LR} and R_P^{LR} indicated that there was substantial retention of both nitrogen (27–59%) and phosphorus (11–31%) in lakes and reservoirs. By comparison, the in-stream retention, R_N^{RR} and R_P^{RR}, appeared to be much lower and varied from 11 to 15% for total N and from 3 to 12% for total P.

As expected, the highest R_N^{LR} estimates were obtained for ZEI (large proportion of lakes and reservoirs) and MER (numerous scattered small lakes), where the input of N is relatively small. The R_P^{LR} value was also highest for ZEI. These results show that the large proportion of lakes and reservoirs had a marked impact on N and P retention in this basin. The R_P^{LR} values for the other basins were much more variable (ranging from 11.3% for MER and 12.5% for NEV to 27.2% for MUS) due to uneven inputs from P sources and disparate lake fractions (including reservoirs). The relatively high R_P^{LR} value for MUS can probably be explained by the capacity to retain point source phosphorus in Lakes Rėkyva (11.8 km^2), Talkša (0.56 km^2) and Ginkūnai (0.175 km^2) located near the large urbanized town of Šiauliai, which has a population of 126,000 inhabitants.

The relative retention of nitrogen and phosphorus in river networks (R_N^{RR} and R_P^{RR}) did not differ significantly among the basins, except for the comparatively low R_P^{RR}

Table 4 | Values of k used in Equation (6)

Constituent	Agricultural land	Forest and wetlands	Pastures ad meadows
Nitrogen			
Lithuania	0.0729	0.0176	0.0160
Estonia	0.0368	0.0095	0.0368
Phosphorus			
Lithuania	0.0014	0.000781	0.00077
Estonia	0.0019	0.000120	0.00190

Table 5 | Estimated nutrient retention values

Variable	River basin						
	MER^{LT}	MUS^{LT}	NEV^{LT}	ZEI^{LT}	EMA^{EST}	AHJ^{EST}	VÕH^{EST}
R_N^r, %	77.8	72.2	66.7	72.9	46.4	10.6	28.4
R_P^r, %	23.5	40.9	44.1	62.8	35.0	5.0	18.0
R_N^{Sp}, kg ha^{-1} yr^{-1}	1,174	1,249	1,885	92	116	78	62
R_P^{Sp}, kg ha^{-1} yr^{-1}	7.2	8.8	31.8	3.0	1.5	1.3	1.6
R_N^{LR}, %	59.0	27.6	27.2	48.6	49.1	2.0	19.6
R_P^{LR}, %	11.3	27.2	12.5	31.1	43.3	1.5	15.9
R_N^{RR}, %	14.6	11.8	14.5	11.3	10.0	6.2	5.7
R_P^{RR}, %	3.3	11.5	10.9	9.8	3.8	3.0	2.7

value of 3.3% for MER. This low capacity for phosphorus retention in MER is presumably related to the specific hydraulic conditions mentioned above.

The estimates presented in Table 5 show that relative nutrient retention varies substantially in the Estonian river basins. The higher relative retention in EMA can probably be explained by the large Lake Võrtsjärv situated in the middle of this basin, assuming that the location of large lakes within a basin has an impact in this context. For example, relative retention was found to be 64% in the sub-basin at the outlet of Lake Võrtsjärv in EMA. Considering the other two basins of interest in Estonia, there are lakes in the upper part of VÕH, whereas there are no large lakes in AHJ, and thus relative retention is much lower there. The specific retention of nitrogen and phosphorus was quite low in Estonia and was comparable with corresponding values noted for ZEI in Lithuania. In most cases separate estimation of relative retention in lakes and rivers gave values close to those obtained for Lithuanian rivers, and only the relative retention of phosphorus was lower in Estonia.

DISCUSSION

This study used the MESAW model based on multiple regression methodology to estimate nutrient export coefficients and retention in Lithuanian and Estonian river basins. The export coefficients determined for various types of land cover and retention are discussed below.

Export coefficients

The estimated export coefficients presented here for N_{tot} in various land-use categories (i.e. agriculture, forest, and pasture and meadow) corroborate with values obtained in other investigations assessing the effects of similar climatic conditions (Kronvang *et al.* 2003; Grizzetti *et al.* 2005; Šileika *et al.* 2006; Shrestha *et al.* 2008; Šmitienė 2008; Vassiljev *et al.* 2008). In all those studies the export coefficients were highest for basins dominated by intensive agriculture and lowest for forested areas.

In lowland river basins that are used predominantly for agricultural purposes, estimations indicating higher diffuse N emissions can be explained by the widespread artificial drainage in such areas, which increases nitrate leaching. Tile draining is often employed in agriculture to improve the soil by creating better moisture and aeration conditions and shortening the water residence time, and hence this represents an important pathway for nutrients to reach adjacent water bodies. Nitrogen losses are always larger under drained rather than under undrained soil conditions (Povilaitis 2000; Tiemeyer *et al.* 2006). Behrendt & Bachor (1998) estimated that 47% of the nitrogen and 12% of the phosphorus emissions from the state of Mecklenburg-Vorpommern in north-eastern Germany to the Baltic Sea originated from tile drainage. At the field scale, annual nitrate-nitrogen losses of up to 105 kg ha^{-1} have been measured (Vinten *et al.* 1994; Kladivko *et al.* 1999), and annual leaching rates of 25–101 kg ha^{-1} via drainage systems have been observed in Lithuania (Povilaitis 1998;

Bučienė 2003; Šmitienė 2008). On the other hand, the significantly higher N export coefficient for agricultural land also reflects the effect of fertilizer use and agricultural management practices.

The results of the current study also suggest that the impact of nitrogen leaching may be more limited in river basins overlying sandy aquifers, because larger quantities of nitrate–nitrogen can be removed by denitrification in such areas. For example, it has been estimated that 48% of the nitrogen losses from arable land in southern Sweden occurs during the transport to surface waters (Arheimer & Brandt 2000). Hetling *et al.* (1999) have concluded that, once fertilizers have been spread on a field, the loss of nitrogen through denitrification and volatilization can be about 10–30% of the agricultural input.

The estimated diffuse P_{tot} emissions of 0.13–0.30 kg ha^{-1} yr^{-1} in the present study correspond well with field-scale measurements made under different land-use conditions in Lithuania (Bučienė 2003). However, the annual exports from the basins in that study were lower than values found in other countries: 0.2–0.8 kg ha^{-1} in various basins in the northern temperate zone (Svendsen *et al.* 1995), 0.5–2 kg ha^{-1} in England (Haygarth *et al.* 1998) and up to 2.5 kg ha^{-1} in Ireland (Tunney *et al.* 2000). The differences can be attributed to the lower specific runoff and other characteristics of loading and storage of phosphorus in Lithuanian basins. The soils in Lithuania are generally low in phosphorus, and, according to Mažvila (1998), only small areas of the country are dominated by soils rich in labile phosphorus (i.e. >100 mg kg^{-1}). Therefore, there is little risk of phosphorus leaching, even in the basins with intense drainage. Nevertheless, loss of particulate phosphorus through erosion may be an important component of the phosphorus loads in the rivers located in the parts of the basins that have steeper slopes and higher precipitation. Most export coefficients for Estonian rivers are very close to values noted for Lithuanian rivers. Greater losses of nutrients from pastures and meadows may be explained by the fact that many of the pastures are situated on drained peat soils, which represent a source of nutrients. Low phosphorus output from forested areas in Estonia agrees fairly well with the values reported in the literature; e.g. Wickham & Wade (2002) found evidence that phosphorus loads from forest land can range from 0.01 to 0.83 kg ha^{-1} yr^{-1}.

The P_{tot} export values from forested areas can be attributed to the background losses. Although the estimated P_{tot} export from forest land in Lithuanian river basins is relatively high (0.13–0.20 kg ha^{-1} yr^{-1}) this does not contradict the observed values. The long-term monitoring data from the Skroblus River (forest area covers 94% of the total basin area) in south Lithuania indicate that riverine transport of P_{tot} varies between 0.157 and 0.398 kg ha^{-1} yr^{-1}. In the Buka River (forest covers 60% of the basin's area in east Lithuania) the average P_{tot} transport is 0.138 (0.062–0.396) kg ha^{-1} yr^{-1}. The background losses of P_{tot} in Danish streams are in a similar range (Kronvang *et al.* 2005). Relatively high P export from forest covered areas ranging from 0.117 to 0.710 kg ha^{-1} yr^{-1} were also reported by Dillon & Kirchner (1975), McFarland & Hauck (1999), Haggard *et al.* (2003), Salvia-Castellvi *et al.* (2005) and Shrestha *et al.* (2008).

Retention

Earlier results have indicated greater retention of nitrogen than phosphorus in surface waters, which is confirmed by the current finding that the retention of total nitrogen in the Lithuanian and Estonian river basins (67–78% relative to the input) was higher than values obtained by other researchers in basins with similar climatic conditions. Arheimer & Brandt (1998) have estimated that 45% of the gross annual nitrogen load in southern Sweden is reduced during transport, and Howarth *et al.* (1996) reported nitrogen retention values of 0–45% for different European catchments. Furthermore, Lepistö *et al.* (2006) have determined that, of the total N input to Finnish river systems, 0–68% is retained in surface waters (mean 22%), and the highest retention (36–61%) occurs in the basins with the largest proportion of lakes, whereas the lowest retention (0–10%) is in the basins with practically no lakes. However, Vassiljev & Stålnacke (2005) contend that up to 80% of nitrogen input can be retained in river basins in the Nordic–Baltic region. Moreover, a study conducted by Trepel & Palmeri (2002) in Germany showed that the efficiency of nitrogen removal in river basins varies from 22 to 77%. Together, these observations imply that nitrogen retention varies greatly and is site specific; it depends on the size of the water body and flow conditions; and it is greatly affected by several biogeochemical and physical processes, including plant uptake, denitrifcation and sedimentation.

The retention of total phosphorus (24–63%) found in the current study falls within the range reported in the literature (Gelbrecht *et al.* 2005; Vassiljev & Stålnacke 2005; Withers & Jarvie 2008). Little information is available regarding the proportions of phosphorus retained in lakes and river networks in the Baltic countries. However, Taminskas *et al.* (2007) have reported that retention of total phosphorus in the lakes of the Dovinė River Basin in southern Lithuania varies between 27 and 56%. Also, in Estonia, research has indicated that 33% of nitrogen and 35% of phosphorus is retained in lakes, whereas in-stream retention is lower, with values of 11 and 14%, respectively (Vassiljev & Stålnacke 2005).

Considering other countries and regions, Howarth *et al.* (1996) found nitrogen retention in the range of 20–80% in lakes in areas around the North Atlantic Ocean. Jansson *et al.* (1994) have proposed that productive lakes might remove up to 50% of total N inputs, and a study in Sweden showed 50% retention of total nitrogen in two eutrophic lakes (Ahlgren *et al.* 1994). Results regarding in-stream retention indicate rate fluctuations from 2 to 30% for nitrogen and from negative values (due to desorption and resuspension processes in streams) to 60% for phosphorus (Billen *et al.* 1991; Hill 1997; Withers & Jarvie 2008). These levels are within the variation range of retention noted in the present study (Table 4).

The current estimations also indicate that much larger amounts of nitrogen and phosphorus are retained in lakes and reservoirs than in river networks. There is no doubt that the lakes in the studied basins act as nutrient sinks. In addition, the results concerning in-stream retention suggest that flow conditions constitute the most critical factor controlling nutrient removal, which has been emphasized by other investigators as well. Grizzetti *et al.* (2003) pointed out that increases in nutrient removal occur primarily during summer, when low flow and higher temperatures allow more substantial sedimentation and acceleration of biological processes. Those researchers also observed that nitrogen removal by denitrification and settling decreases in deeper channels, where exchange between stream waters and benthic sediments is reduced. Withers & Jarvie (2008) examined in-stream retention and cycling of phosphorus and discovered that, during very high flows, P inputs to streams were flushed through without entering the biogeochemical pathways in the investigated watercourses. House (2003) found that in-stream P retention rates varied from 10 to over 30% under a wide range of flow conditions, whereas Jarvie *et al.* (2002) recorded up to 60% net retention during low flow in the River Kennet in England.

CONCLUSIONS

Due to its uncomplicated structure, the MESAW model proved to be a simple but reliable tool for simultaneous estimation of nutrient sources and retention in river basins. Moreover, the approach based on implementing export coefficients in MESAW turned out to be very useful for estimating the total annual loads of nutrients from diffuse sources to a water body, and hence it can be used to estimate the relative contribution of each N and P source to riverine export.

For all of the studied river basins, the export coefficients for N and P were much higher for agricultural land than for forested land and pastures and meadows. Even though the method used in the MESAW model does not take into account the specific mechanisms underlying surface runoff and nutrient transport, the derived coefficients can be used to estimate the diffuse source pollution loadings from the major land-use classes.

The present study also showed that the river basins investigated in Lithuania had a substantial capacity to retain nitrogen and phosphorus. More precisely, 67–78% of total nitrogen and 24–63% of total phosphorus relative to the input are retained in the surface waters in those regions. Estimates of retention in lakes were larger, 27–59% for nitrogen and 11–31% for phosphorus, whereas in-stream retention appeared to be much lower, varying from 11 to 15% for total N and from 3 to 12% for total P.

Lower nutrient retention values were found for Estonia (as compared with Lithuania) due to the smaller lake areas in the basins that were studied in that country. The highest rates of relative retention of nitrogen and phosphorus in Estonian lakes were 46 and 35%, respectively. In-stream retention was also somewhat lower in Estonia than in

Lithuania, which might be explained by low temperatures in Estonian rivers in summer.

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