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Journal of Marine Systems 74 (2008) 485-494

Contents lists available at ScienceDirect



## Journal of Marine Systems



journal homepage: www.elsevier.com/locate/jmarsys

# The Baltic Sea a century ago – a reconstruction from model simulations, verified by observations

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#### ARTICLE INFO

Article history: Received 24 September 2007 Received in revised form 28 December 2007 Accepted 19 March 2008 Available online 31 March 2008

Keywords: Nitrogen Phosphorus Reference conditions Coastal point loads Biogeochemical fluxes Trophic state Baltic Sea

#### 1. Introduction

A common statement in environmental management objectives worldwide has been to 'restore ecosystems'. Although appealing, this rather vague statement has to be made more precise in order to be useful as a management objective. In Europe this is attempted by the Water Framework Directive (WFD), established by the European Union in 2001 (Anonymous, 2000). The WFD provides authorities in the EU member states with a legal binding basis for the maintenance and recovery of water quality to achieve good ecological and chemical status for all surface waters and good chemical status for groundwater. Open marine waters are not covered, but the WFD is likely to influence management of all marine ecosystems because all land-based inputs of pollutants pass through the coastal zone to the open waters (Andersen et al., 2004).

A cornerstone of the WFD is the definition of good ecological status of the waters or 'reference conditions'.

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#### ABSTRACT

"Pre-industrial" trophic conditions in the Baltic Sea were simulated with SANBALTS (Simple As Necessary BAltic Long-Term large Scale) model. External nutrient inputs to the major basins of the Baltic Sea a century ago were reconstructed from various literature and data sources. The reconstructed input of total nitrogen was less than a half and that of total phosphorus was about a third of their contemporary values. The simulated "pre-industrial" conditions are validated by comparison to actual historical data on the water transparency, oxygen concentration, primary production, and net sediment accumulation. The "pre-industrial" trophic state could have been more phosphorus limited than today because simulated basin-wide annual averages of dissolved inorganic phosphorus concentrations of  $0.06-0.3 \mu$ M P are about 40–80% of their present day values, while dissolved inorganic nitrogen concentrations of  $2-4 \mu$ M N are almost the same as today or even slightly higher.

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Practically it is defined as conditions that occurred prior to the intensification of agriculture 100–150 yr ago (Andersen et al., 2004). The reference conditions vary around Europe due to regional variation in climatic, morphometric, hydrographic and biogeochemical conditions. Several methods for the establishment of reference conditions can be used. The methods described in the legislation for the WFD includes the use of historical data, modeling (empirical or dynamic), and a combinations of these approaches, and finally, expert judgment.

For the Baltic Sea, several of these methods have been tried with variable success (HELCOM, 2006). A very ambitious attempt to analyze long-term coastal data sets from the Baltic was made in several EU funded projects but with limited success due to highly variable data quality from different regions and short time coverage. Only for a few Baltic regions are good ecological data sets available to actually calculate reference conditions (i.e. Frederiksen et al., 2004). Another approach is to develop transfer functions using sediments core as archives to reconstruct past conditions. Quantitative paleoecological methods, using relationships between fossil remains of organisms have been applied to some coastal

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waters in the Baltic (Clarke et al., 2003; Weckstrom et al., 2004) as well as geochemical proxies employing nutrients (Emeis et al., 1998, 2000), organic structural compounds (Miltner et al., 2005), and isotope signatures of sediments (Struck et al., 1998, 2000).

The unique features of the Baltic, with decadal residence times of water and many years for nutrients, mean that all coastal regions with residence time of days and a few weeks (Schernewski and Wielgat, 2004) have been affected by anthropogenic sources, not only from land but also by offshore-coastal exchange. This is in contrast to many other European seas where offshore conditions are still pristine and variations in coastal conditions reflect only variations in local inputs from land.

Schernewski and Neumann (2005) used a modeling approach by shifting the focus from coastal waters, where only little early data are available towards the river basins for which better long-term data and information are available. By estimating the riverine and atmospheric nutrient inputs a century ago and using these as forcing to a coupled physicalbiogeochemical 3D model, they calculated nutrient, phytoplankton and chlorophyll concentrations a century ago. However, the model results are difficult to validate since good measurements of nutrients and plankton are only available from the last four decades.

In this paper, we use the same approach for reconstruction of the "pre-industrial" conditions as Schernewski and Neumann (2005) but we validate our results against data that are actually available from observations 100 yr ago. Moreover, we have included a more complete coverage of the various nutrient sources, i.e., riverine and diffuse sources, coastal point inputs, and atmospheric deposition. To reconstruct the conditions in the Baltic Sea basins that existed a century ago, we use an aggregated biogeochemical model calibrated for contemporary conditions (Savchuk and Wulff, 2007). Key features of the Baltic Sea are the long residence times and the intimate coupling between the major basins through large advective flows of water and nutrients (Wulff et al., 2001b; Savchuk, 2005) that are well represented in this model. Emphasis is given not only to the external nutrient load but also to internal source and sink terms for nitrogen and phosphorus that are crucial for controlling nutrient levels in the Baltic Sea.

#### 2. Methods and data

#### 2.1. The model

SANBALTS (Simple As Necessary BAltic Long-Term large Scale) model simulates annually averaged coupled nitrogen and phosphorus cycles in the major basins of the Baltic Sea (Fig. 1). The dynamical balance between nutrient sources and sinks in all the boxes is described by a system of ordinary differential equations, where different terms represent nutrient inputs from land and atmosphere, the water transports between boxes, including exchange with the Skagerrak, and the internal biogeochemical processes (Fig. 2). In order to obtain a quantitative solution of this system, we must prescribe the driving forces representing external nutrient inputs and water flows between boxes. With invariable forcing implemented in this study, the system of



**Fig. 1.** Major basins of the Baltic Sea and their watersheds: BB – the Bothnia Bay, BS – the Bothnia Sea, BP – the Baltic Proper, GF – the Gulf of Finland, GR – the Gulf of Riga, DS – the Danish Straits, KT – the Kattegat. Ska – indicate the "boundary" waters of the Skagerrak strait. The Baltic Proper is split into surface (BPs, 0–60 m) and deep (BPd, >60 m) waters. Stars show locations of the cities, whose population a century ago exceeded 100,000 persons.

equations is numerically integrated until a steady state is reached. In this state, all the simulated concentrations, transport flows and biogeochemical fluxes are interrelated, mutually consistent and describe some conditions at equilibrium with the applied forcing. The full description of SANBALTS, including details of parameterization, tuning, and successful testing for the contemporary conditions is given elsewhere (Savchuk and Wulff, 2007, in press).

Secchi depth, a measure of water transparency, is important for this study since observations are actually available from measurements made over 100 yr ago (Aarup, 2002; Laamanen et al., 2004). We have developed empirical relationships between annual means of water transparency and basin-wide nutrient concentration based on simultaneous measurements of Secchi depth, total N, and total P (Savchuk et al., 2005; Savchuk and Wulff, 2007).

#### 2.2. Forcing – contemporary and pristine

#### 2.2.1. Land loads

Contemporary land loads are given as averages for 1997–2003 based on the data on riverine and coastal point sources provided by HELCOM (Table 1). In the model, organic



**Fig. 2.** State variables and internal biogeochemical processes in SANBALTS boxes comprising water and sediments in the major basins of the Baltic Sea. ON and OP – organic nitrogen and phosphorus, consisting of labile and refractory fractions, DIN and DIP – dissolved inorganic nitrogen and phosphorus, BEN and BEP – bioavailable nitrogen and phosphorus in sediments,  $O_2$  – oxygen in the Baltic Proper deep box.

nitrogen (N) and phosphorus (P) inputs are split into labile and refractory fractions, the labile fraction is assumed being smaller in the nitrogen input from north-eastern forested watersheds than in the input from south-western agricultural and urbanized areas, while the labile organic phosphorus fraction is assumed basin-invariant (Savchuk and Wulff, 2007, in press).

Nutrient loads in the past are more difficult to reconstruct due to a lack of direct measurements. Schernewski and Neumann (2005) have carefully estimated background concentrations of inorganic nutrients in the fifteen major rivers, together comprising about 90% of the total freshwater discharges into the Baltic Sea. According to these estimates, the total riverine input of dissolved inorganic nitrogen (DIN) and phosphorus (DIP) to the Baltic Sea in 'pre-industrial' conditions was about a third and a quarter of the contemporary one, respectively, albeit with clear differences between nutrients and basins. We aggregated their river-wise scenario inputs by basins and used such basin-wise estimates to reduce contemporary DIN and DIP inputs to 'pre-industrial' levels. Assuming labile organic fraction of nutrients being closely related to inorganic fraction by the sources, transports and catchment retentions, the proportions between riverine labile organic and inorganic concentrations in the 'preindustrial' conditions were taken the same as in the contemporary ones. The inputs of refractory organic matter into the sea a century ago were assumed the same as in contemporary conditions, since these substances originate from the huge stocks of soil organic matter that have been built up over many centuries in the northern forested and southern cultivated watersheds.

These assumptions appear justified since land-use patterns and nutrient leakage from agricultural soils have not changed dramatically over the last century. A case study by Hoffmann et al. (2000) shows that both specific leaching rates and gross load of nitrogen in the middle of 19th century were approximately the same as they are today for the whole of south and central Sweden. These regions represent two characteristic types of agricultural practices in the Baltic Sea area, the southern part dominated by intensive agriculture and the central part with less intensive agriculture. Agriculture is not significant in northern Sweden and in major parts of northern Finland and the area covered by cultivated land is

#### Table 1

Watershed characteristics, contemporary and "pre-industrial" (in brackets) inputs of nutrients to different basins prescribed in the model according to data and assumptions explained in the text

Basin	BB	BS	BP	GF	GR	DS	KT	Total
Watershed properties								
% forest	74	81	30	64	39	6	60	
% arable land	4	5	57	20	51	79	23	
Person km <sup>-2</sup>	5	11	96	29	30	181	37	48
Nitrogen land loads (10 <sup>3</sup> to	nnes yr <sup>-1</sup> )							
River	48.1	51.2	320.0	99.4	76.8	41.2	61.6	698.4
	(36.1)	(33.1)	(163.4)	(58.1)	(32.9)	(10.9)	(19.1)	(353.7)
Coastal	3.3	5.5	7.2	13.3	1.6	4.7	2.6	38.3
	(0.3)	(0.8)	(2.2)	(4.0)	(0.5)	(1.4)	(0.8)	(9.9)
Phosphorus land loads (ton	nes $yr^{-1}$ )							
River	2477	2144	18440	5550	1985	1037	1399	33031
	(982)	(937)	(3994)	(2234)	(668)	(546)	(743)	(10 104)
Coastal	108	313	439	1310	196	372	174	2913
	(9)	(44)	(132)	(393)	(59)	(111)	(52)	(800)
Atmospheric deposition								
Nitrogen	10.6	32.6	154.9	15.4	12.0	28.5	24.4	278.4
$(10^3 \text{ tonnes yr}^{-1})$	(1.1)	(3.3)	(15.5)	(1.5)	(1.2)	(2.8)	(2.4)	(27.8)
Phosphorus	562	1178	3205	445	270	318	336	6315
(tonnes yr <sup>-1</sup> )	(20)	(60)	(286)	(28)	(22)	(53)	(45)	(514)

Basins with their full names are presented in Fig. 1.

less than 5% for most major river catchments (cf. Table 1). Nutrient land-sea fluxes in these boreal watersheds are affected by forestry, however, draining activities and clear cuts increasing nutrient leaching from forested soils (Grip, 1982) were assumed to be constant over the last century. The agriculture in the southeastern part of the Baltic Sea watersheds and especially for Poland (Tonderski, 1997; Eriksson et al., 2007) and Estonia, Latvia, and Lithuania (Löfgren et al., 1999) can be described as low productive using relative low amounts of fertilizers compared to western countries (Tonderski, 1997). Nutrient leaching from these agricultural soils has probably not changed dramatically (Eriksson et al., 2007) similar to the situation in Sweden (Hoffmann et al., 2000). Eriksson et al. (2007) conclude that even in almost 10 yr after the drop in fertilizer use the riverine nutrient loads of the Oder and Vistula did not show any trend downwards. This lack of a response pattern might be related to leakage thresholds (Oenema et al., 1998), i.e. agricultural soils start to leak significantly more amounts of nutrients when nutrient surpluses become higher than a certain amount agricultural soils can buffer due to storage and denitrification.

Riverine loads describe nutrient contributions from point and diffuse sources upstream from monitoring stations, which are usually some distance away from the coast. Typically, there are also municipalities located at the river mouths and their contributions to the input to the sea can be quite substantial. Around the Baltic, about 20% of the total population of 84 million lives within 10 km from the coast (Hannerz and Destouni, 2006), and about 5% of total nitrogen and 8% of total phosphorus input from the land to the entire Baltic Sea come today from coastal point sources (cf. Table 1). Note though, that contribution of point sources differs between basins and nutrients, spanning the ranges 4% (BP) to 30% (DS) for nitrogen and 10% (BP) to 41% (DS) for phosphorus.

It is indeed not easy to estimate the magnitude of coastal point sources in "pre-industrial" times. Quantitative information, useful for indirect calculations of nutrient loads is difficult to find for this region that has suffered from two world wars during the last century. Only one of the fourteen countries within the Baltic Sea watershed still has the same national borders. Within the uncertainty arising from different administrative borders and the years of reference, the population of the Baltic Sea watershed in the late 1890s to the early 1900s can be estimated from a number of articles in the Entciklopeditcheskii slovar' (1890-1907) to about 38 million people. This is less than half of the present day population. The city-wise comparison of information from the Entciklopeditcheskii slovar' (1890-1907) with the present day cities' Internet sites shows that the population of 26 major cities along the Bothnian Bay, the Bothnian Sea, and the rest of the Baltic coasts was about 8%, 14%, and 30% of the contemporary population, respectively. Thus, one estimate of the "preindustrial" point sources can be obtained by reducing the contemporary values proportionally to the changes of population occurred in the coastal cities. This would result in reduced pre-industrial nutrient loads of 3% for total nitrogen and 7% for total phosphorus (cf. Table 1). This scenario can be considered as a low estimate since these reductions are applied to the contemporary inputs that are already substantially affected by sewage treatment. The forcing with such point sources will be referred to as the "lower load" or LL scenario in the simulations described below.

The history of the urban water management in the Baltic region has been studied mostly in a qualitative sense and the results have been published in a series of papers (Laakkonen and Laurila, 1999, 2001). A century ago, cities around the Baltic had grown to such a degree that human and industrial wastes created major problems. Constructions of water supply and sewage systems diminished the occurrence of cholera and typhus but increased the output of waste to inland and coastal waters. The construction of sewage plants were in most cases initiated after the Second World War. However, a century ago all transports were horse drawn, most food processing (slaughter, fish markets, bakery, brewery, etc.) was done within the cities. All these activities abundantly added nutrient effluents. Assuming that the urban population growth has been fully compensated by changes in the ways of life, including sewage treatment, the municipal and industrial nutrient inputs a century ago can be prescribed the same as today. In this scenario, coastal point sources contribute about 10% of total nitrogen and 22% of total phosphorus input from the land (cf. Table 1). Apparently, such inputs can be considered as an estimate from above and will be referred to as the "higher load" (HL) scenario.

#### 2.2.2. Atmospheric depositions

Contemporary basin-wise estimates of annual DIN atmospheric deposition have been obtained from the Co-operative programme for monitoring and evaluation of the long range transmission of air pollutants in Europe (EMEP) and averaged for 1997-2002 (see, e.g. HELCOM, 2004). Additionally, input of organic nitrogen from atmosphere was prescribed as contributing 15% of total nitrogen atmospheric deposition (cf. Table 1). This estimate is within the ranges of both global generalizations (e.g. Cornell et al., 2003; Jickells, 2005) and regional studies (Nehring and Wilde, 1982; Falkowska, 1985; Rahm et al., 1995). However, there is still not enough knowledge about how much of deposited organic nitrogen is bioavailable (Seitzinger and Sanders, 1999; Cornell et al., 2003). On the other hand, this assumption may not be critical, especially in reconstruction of the "pre-industrial" loads. In the contemporary conditions this component constitutes less than 5% of the total inputs of the bioavailable nitrogen to the entire Baltic Sea. Correspondingly, its exclusion from the contemporary forcing in sensitivity numerical experiment resulted in less than 5% perturbations of the simulated system (Savchuk and Wulff, 2007).

The contemporary atmosphere is also assumed to deliver 15 kg P km<sup>-2</sup> yr<sup>-1</sup> evenly distributed over all basins (Savchuk, 2005 and references therein), which is in the middle of the reported range of 4–33 kg P km<sup>-2</sup> yr<sup>-1</sup> observed in the Northern Europe (Anttila et al., 1995; Pollman et al., 2002; Ellermann et al., 2003) and is an important source of phosphorus, as was shown by another numerical experiment (Savchuk and Wulff, 2007).

For the "pre-industrial" conditions we adopted assumptions of Schernewski and Neumann (2005) and prescribed DIN atmospheric deposition as being 10% of the contemporary, while DIP atmospheric depositions was given as 1% of the molar DIN deposition.

**Table 2** Contemporary and "pre-industrial" concentrations of nitrogen and phosphorus in the Baltic Sea basins ( $\mu$ M L<sup>-1</sup>)

BB	BS	BPs	BPd	GF	GR	DS	KT		
rogen									
20.5±	18.9±	19.7±	23.1±	23.2±	36.3±	21.8±	18.9±		
0.9	0.8	1.0	1.4	2.8	3.3	1.4	1.2		
21.3	18.7	19.4	22.1	24.5	33.2	20.1	17.7		
17.5/	16.5/	16.5/	18.3/	19.8/	25.7/	15.6/	13.9/		
18.3	17.0	17.0	18.9	20.9	26.0	16.1	14.2		
Dissolved inorganic nitrogen									
7.0±	3.5±	2.6±	7.4±	4.7±	7.6±	3.4±	4.7±		
0.9	0.5	0.4	1.3	1.4	1.8	0.5	1.3		
7.0	3.2	2.5	7.4	5.6	7.0	3.1	4.6		
4.9/	4.5/	3.1/	6.0/	5.0/	7.0/	3.0/	3.9/		
5.2	4.4	2.9	6.0	5.0	6.6	3.0	3.8		
osphorus									
0.22±	0.46±	0.67±	2.30±	0.78±	1.05±	0.83±	0.71±		
0.03	0.04	0.07	0.44	0.14	0.08	0.11	0.07		
0.20	0.50	0.80	2.18	0.81	1.01	0.78	0.68		
0.12/	0.25/	0.33/	0.74/	0.38/	0.40/	0.38/	0.46/		
0.13	0.27	0.36	0.83	0.44	0.44	0.41	0.47		
Dissolved inorganic phosphorus									
0.05±	0.23±	0.41±	2.04±	0.49±	0.54±	0.42±	0.41±		
0.01	0.04	0.05	0.41	0.12	0.08	0.12	0.08		
0.05	0.25	0.50	1.95	0.52	0.49	0.40	0.37		
0.06/	0.19/	0.22/	0.65/	0.29/	0.32/	0.23/	0.29/		
0.06	0.19	0.23	0.73	0.33	0.35	0.25	0.30		
	BB rogen 20.5± 0.9 21.3 17.5/ 18.3 d inorgan 7.0± 0.9 7.0 4.9/ 5.2 0.03 0.22± 0.03 0.20 0.12/ 0.12/ 0.13 d inorgan 0.55± 0.01 0.05 0.06/ 0.06	BB         BS           rogen $20.5\pm$ $18.9\pm$ $0.9$ $0.8$ $21.3$ $18.7$ $17.5/$ $16.5/$ $18.3$ $17.0$ d inorganic nitroge $7.0\pm$ $3.5\pm$ $0.9$ $0.5$ $7.0\pm$ $3.5\pm$ $0.9$ $0.5$ $7.0\pm$ $3.2$ $4.9/$ $4.5/$ $5.2$ $4.4$ osphorus $0.22\pm$ $0.46\pm$ $0.03$ $0.04$ $0.20$ $0.50$ $0.12/$ $0.25/$ $0.13$ $0.27$ d inorganic phosph $0.23\pm$ $0.01$ $0.04$ $0.05\pm$ $0.23\pm$ $0.01$ $0.04$ $0.05\pm$ $0.23\pm$ $0.01$ $0.04$ $0.05\pm$ $0.23\pm$ $0.01$ $0.04$ $0.05 = 0.25$ $0.06/$ $0.19/$	BB         BS         BPs           rogen         19.7±           20.5±         18.9±         19.7±           0.9         0.8         1.0           21.3         18.7         19.4           17.5/         16.5/         16.5/           18.3         17.0         17.0           d inorganic nitrogen         7.0         3.5±         2.6±           0.9         0.5         0.4         7.0           7.0         3.2         2.5         4.4         2.9           osphorus         2.5         4.4         2.9           osphorus         0.44.4         2.9         3.1/           5.2         4.4         2.9         3.1/           0.22±         0.46±         0.67±         0.03           0.12/         0.25/         0.33/         0.13           0.13         0.27         0.36         3.1/           d inorganic phosphorus         0.41±         0.01         0.04           0.05±         0.23±         0.41±         0.01         0.04           0.055         0.25         0.50         0.06/         0.09/         0.22/           0.06/         0.19/	BB         BS         BPs         BPd $20.5\pm$ $18.9\pm$ $19.7\pm$ $23.1\pm$ $0.9$ $0.8$ $1.0$ $1.4$ $21.3$ $18.7$ $19.4$ $22.1$ $17.5/$ $16.5/$ $18.3/$ $18.3/$ $18.3$ $17.0$ $17.0$ $18.9$ $d$ inorganic nitrogen $7.0\pm$ $3.5\pm$ $2.6\pm$ $7.4\pm$ $0.9$ $0.5$ $0.4$ $1.3$ $7.0$ $3.2$ $2.5$ $7.4$ $0.9$ $0.5$ $0.4$ $1.3$ $7.0$ $3.2$ $2.5$ $7.4$ $0.9$ $0.5$ $0.4$ $1.3$ $7.0$ $3.2$ $2.5$ $7.4$ $4.9/$ $4.5/$ $3.1/$ $6.0/$ $5.2$ $4.4$ $2.9$ $6.0$ osphorus $0.22\pm$ $0.46\pm$ $0.67\pm$ $2.30\pm$ $0.74/$ $0.20$ $0.50$ $0.80$ $2.18$ $0.12/$ $0.63$ $0.12/$ $0.25/$	BB         BS         BPs         BPd         GF           20.5±         18.9±         19.7±         23.1±         23.2±           0.9         0.8         1.0         1.4         2.8           21.3         18.7         19.4         22.1         24.5           17.5/         16.5/         16.5/         18.3/         19.8/           18.3         17.0         17.0         18.9         20.9           d inorganic nitrogen         -         -         4.7±         0.9           0.9         0.5         0.4         1.3         1.4           7.0         3.5±         2.6±         7.4±         4.7±           0.9         0.5         0.4         1.3         1.4           7.0         3.2         2.5         7.4         5.6           4.9/         4.5/         3.1/         6.0/         5.0/           5.2         4.4         2.9         6.0         5.0           osphorus         -         -         -         -           0.22±         0.46±         0.67±         2.30±         0.78±           0.3         0.04         0.07         0.44         0.14 <td>BB         BS         BPs         BPd         GF         GR           20.5±         18.9±         19.7±         23.1±         23.2±         36.3±           0.9         0.8         1.0         1.4         2.8         3.3           21.3         18.7         19.4         22.1         24.5         33.2           17.5/         16.5/         16.5/         18.3/         19.8/         25.7/           18.3         17.0         17.0         18.9         20.9         26.0           d inorganic nitrogen         -         -         7.4±         4.7±         7.6±           0.9         0.5         0.4         1.3         1.4         1.8           7.0         3.5±         2.6±         7.4±         4.7±         7.6±           0.9         0.5         0.4         1.3         1.4         1.8           7.0         3.2         2.5         7.4         5.6         7.0           4.9/         4.5/         3.1/         6.0/         5.0/         7.0/           5.2         4.4         2.9         6.0         5.0         7.6           0.33         0.04         0.07         0.44</td> <td>BB         BS         BPs         BPd         GF         GR         DS           rogen         20.5±         18.9±         19.7±         23.1±         23.2±         36.3±         21.8±           0.9         0.8         1.0         1.4         2.8         3.3         1.4           21.3         18.7         19.4         22.1         24.5         33.2         20.1           17.5/         16.5/         16.5/         18.3/         19.8/         25.7/         15.6/           18.3         17.0         17.0         18.9         20.9         26.0         16.1           dinorgation introget           7.0±         3.5±         2.6±         7.4±         4.7±         7.6±         3.4±           0.9         0.5         0.4         1.3         1.4         1.8         0.5           7.0         3.2         2.5         7.4         5.6         7.0         3.1           4.9/         4.5/         3.1/         6.0/         5.0/         7.0/         3.0/           5.2         4.4         2.9         6.0         5.0         6.6         3.0            0.22±         0.60</td>	BB         BS         BPs         BPd         GF         GR           20.5±         18.9±         19.7±         23.1±         23.2±         36.3±           0.9         0.8         1.0         1.4         2.8         3.3           21.3         18.7         19.4         22.1         24.5         33.2           17.5/         16.5/         16.5/         18.3/         19.8/         25.7/           18.3         17.0         17.0         18.9         20.9         26.0           d inorganic nitrogen         -         -         7.4±         4.7±         7.6±           0.9         0.5         0.4         1.3         1.4         1.8           7.0         3.5±         2.6±         7.4±         4.7±         7.6±           0.9         0.5         0.4         1.3         1.4         1.8           7.0         3.2         2.5         7.4         5.6         7.0           4.9/         4.5/         3.1/         6.0/         5.0/         7.0/           5.2         4.4         2.9         6.0         5.0         7.6           0.33         0.04         0.07         0.44	BB         BS         BPs         BPd         GF         GR         DS           rogen         20.5±         18.9±         19.7±         23.1±         23.2±         36.3±         21.8±           0.9         0.8         1.0         1.4         2.8         3.3         1.4           21.3         18.7         19.4         22.1         24.5         33.2         20.1           17.5/         16.5/         16.5/         18.3/         19.8/         25.7/         15.6/           18.3         17.0         17.0         18.9         20.9         26.0         16.1           dinorgation introget           7.0±         3.5±         2.6±         7.4±         4.7±         7.6±         3.4±           0.9         0.5         0.4         1.3         1.4         1.8         0.5           7.0         3.2         2.5         7.4         5.6         7.0         3.1           4.9/         4.5/         3.1/         6.0/         5.0/         7.0/         3.0/           5.2         4.4         2.9         6.0         5.0         6.6         3.0            0.22±         0.60		

Data — mean±standard deviation of annual basin-wide averages of observations in 1991–2002, Now — simulated with the contemporary forcing, Past — simulated with the "pre-industrial" LL/HL scenario.

#### 2.2.3. Water exchange

The steady state water flows between basins including outflow to- and inflow from the Skagerrak were taken from (Savchuk, 2005) as long-term means for 1991–1999, the time interval covering quite different physical states of the Baltic Sea. As shown by Meier (2005), the impact of changing freshwater supply or sea level in Kattegat on the average age of the Baltic Sea water masses is rather small, suggesting invariable stability and ventilation in steady state. According to analysis of the long-term oceanographic data (Fonselius and Valderrama, 2003), no important changes in the hydrographic structure of the Baltic Sea occurred during the 20th century. Taken together, these conclusions justify implementation of the same transports both for the present and past conditions.

#### 2.2.4. Imports from Skagerrak

Advective nutrient inputs from the Skagerrak to the Kattegat, totaling annually up to 419 10<sup>3</sup> tonnes N and 34 10<sup>3</sup> tonnes P, were calculated from water inflow and average volume-weighted annual concentrations estimated for the years 1997–2002 with the SwingStations tool from data in the Baltic Environmental Database (BED; Sokolov et al., 1997; Sokolov and Wulff, 1999; Savchuk and Wulff, 2007).

For the pristine conditions, we reduced the present day concentrations of nitrogen and phosphorus in Skagerrak by 15%. The nutrient concentrations in the eutrophied German Bight waters, which constitute about a quarter of the mixture at the entrance to the Baltic Sea (Aure et al., 1998) have

doubled or tripled during the past 100 yr (e.g. Radach and Pätsch, 1997; OSPAR, 2003; van Beusekom, 2005). This relatively minor reduction is corroborated by results from several studies. Andersson (1996) did not find long-term changes of nutrient concentration over 1971-1990 in the Eastern Skagerrak, at the entrance to the Baltic Sea. The nutrient status of this area is to a great extent determined by the Jutland Coastal Current considered containing 75% of water from the southern and central parts of the North Sea (Aure et al., 1998), where no trend-like changes were found in the data since the 1950s (Radach and Pätsch, 1997). The Northern Atlantic waters deliver to the North Sea about 70% of total nitrogen input and 90% of total phosphorus input (Brion et al., 2004) without any significant trends in concentrations (Laane et al., 2005). No long-term increase in oxygen depletion rate was observed between 1955 and 1991 in the Skagerrak deep layers (Aure and Dahl, 1994), while even drastic 50% reductions of riverine nutrient input to the North Sea resulted in only about 5% decrease of the simulated primary production in the Skagerrak that was far less than the natural inter-annual variations (Skogen et al., 2004).

#### 2.3. Data for comparison

Time series of annual basin-wide volume-weighted averages of nutrient concentrations were calculated from 3D fields reconstructed with the Data Assimilation System (DAS, Sokolov et al., 1997) from the data in BED for 1991–2002. Basin-wide oxygen concentrations and hypoxic areas (areas of bottom confined by the oxygen isosurface of 2 ml/l, see also Conley et al., 2002; Vahtera et al., 2007; Savchuk and Wulff, in press) were calculated with the DAS tool from 3D fields of oxygen concentrations reconstructed as annual time series for 1991–2002 (on thousands of samples from hundreds of oceanographic stations for every year), and as averages for the 1905–1906 (on 412 samples from 93 stations) and 1930– 1932 (on 466 samples from 112 stations) periods.

Secchi depth measurements available from ICES for all the Baltic Sea basins (e.g. Aarup, 2002; Laamanen et al., 2004)



**Fig. 3.** Modeled "pre-industrial" (LL – sparse stripes, HL – dense stripes) and contemporary (shaded) distributions of the water transparency (Secchi depth) compared to statistics (mean±SD) of observations before 1940 (left) and after 1985 (right) in the different basins of the Baltic Sea presented in Fig. 1.

#### Table 3

Comparison between observed and simulated contemporary and "preindustrial" LL/HL scenario oxygen concentration (annual average below 60 m) and hypoxic area ( $O_2 < 2 \text{ ml/l}$ ) in the Baltic Proper deep layers for the different time periods

Period	Oxygen below 60	) m (ml/l)	Hypoxic area (10 <sup>3</sup> km <sup>2</sup> )		
	observed	simulated	observed	simulated	
1991-2002	3.4±1.0	3.5	44.4±14.1	42.0	
1930–1932	4.8	5.9/5.7	18.5	6.5/8.5	
1905–1906	6.0		3.3		

were separated into coastal and offshore subsets. The offshore subset of measurements made since 1985 was updated with data from BED to represent the contemporary environment, while measurements between the 1900s and 1940 were considered to represent less eutrophic conditions.

Basin-wide average nutrient concentrations in the uppermost centimeter of sediments for all the major Baltic Sea basins eastwards of the Danish Straits have been estimated from an extended data set on nitrogen and phosphorus concentrations in the different bottom types occurring within certain depth intervals (Carman and Cederwall, 2001). Rates of benthic nutrient transformation processes were estimated from 51 surface sediment samples and 9 dated sediment cores collected in a comprehensive study made in 2003 over the Eastern Gotland basin (Hille, 2005; Hille et al., 2005).

#### 3. Results and discussion

#### 3.1. Simulated and observed concentrations and fluxes

The steady state basin-wise distribution of annual concentrations, simulated under contemporary forcing, matches observations indicating the model's good performance in reproducing of both inter-basin and vertical (BP) gradients (Table 2). The water transparency calculated from the contemporary simulated concentrations is also in a good agreement with observed distribution (Fig. 3). Secchi depth calculated from simulated "pre-industrial" concentrations shows larger deviations from the averaged observations for the beginning of the past century, comparing to the contemporary conditions (Fig. 3). However, even these deviating values are still well within the observed ranges. This comparability can also be considered as an independent indirect validation of the reconstructed "pre-industrial" concentrations in Table 2.

A direct validation of the model capability to reasonably respond on a centennial change in forcing comes from a good match between observed and simulated oxygen situation in the Baltic Proper deep layers (Table 3, Fig. 4). Deep-water oxygen concentration is determined by a balance between ventilation due to both saltwater inflow and mixing across halocline vs. oxygen consumption in the water column and by the sediments for mineralization of organic matter. However, such a balance can be reached with different combinations of oxygen sources and sinks, i.e. with transport and consumption fluxes being simultaneously either higher or lower. Therefore, independent estimates of mineralization that, in turn is determined by the overall intensity of nutrient cycles, are important for validation.

As shown for the contemporary Baltic (Savchuk and Wulff, 2007), total mineralization of 5.5 mol C  $m^{-2}$  yr<sup>-1</sup> simulated in the Baltic Proper deep box (>60 m) is in a good agreement with 4.4–4.7 mol C  $m^{-2}$  yr<sup>-1</sup> estimated from measurements below 70 m in the Eastern Gotland basin (Schneider et al., 2002), if the differences in water volumes and sediment areas between the studies are taken into account. Unfortunately, we do not have field data on the mineralization of organic matter in the oligotrophic Baltic a century ago. However, there are estimates of a long-term change in primary production of organic matter, whose contemporary basin-wise annual integrals simulated by SANBALTS (Table 4) correspond well to the existing estimates (Savchuk and Wulff, 2007, in press). From analysis of pCO<sub>2</sub> data Schneider and Kuss (2004) inferred "an increase in the net biomass productivity of the Baltic Proper by a factor of 2.5 (maximum uncertainty 1.8–3.8) since the beginning of the last century". This estimate envelopes our simulated increase by a factor of 3.0-3.4, if we compare the pre-industrial and the contemporary simulations (Table 4). In the 1950s, the annual primary production in the open waters of the Kattegat and Danish



**Fig. 4.** Area of hypoxic bottoms of the Baltic Proper underlying waters of  $<2 \text{ ml L}^{-1}$  oxygen reconstructed from observations averaged over 1905–1906 (left), 1930–1932 (middle) and 1996 (right). The year 1996 with a hypoxic area of 44,530 km<sup>2</sup> illustrates the contemporary expansion of hypoxic conditions with average (1991–2002) coverage of 44,376 km<sup>3</sup>.

#### Table 4

Major biogeochemical fluxes simulated under contemporary (Savchuk and Wulff, 2007) and "pre-industrial" (LL/HL scenario) conditions: primary production (PP, g C  $m^{-2}$  yr<sup>-1</sup>), dinitrogen fixation (NF, 10<sup>3</sup> tonnes N yr<sup>-1</sup>), and dinitrogen release (NR, 10<sup>3</sup> tonnes N yr<sup>-1</sup>)

Basin	BB	BS	BP	GF	GR	DS	KT
PP	25	124	188	141	259	216	223
	(8/9)	(19/25)	(55/63)	(19/28)	(20/27)	(72/77)	(113/115)
NF	0	18	366	18	1	6	3
	(0)	(0)	(14/44)	(0/1)	(0)	(1/2)	(2/2)
NR	17	88	858 <sup>*</sup>	64	46	42	88
	(5/6)	(14/18)	(196/233)	(9/13)	(4/5)	(14/15)	(46/47)

Note: \* - sum of fluxes in the surface sediments, deep sediments, and deep water layers.

Straits was roughly a half and a third of the contemporary values, respectively (Richardson and Heilmann, 1995; Rydberg et al., 2006). These estimates correspond well to the decrements between simulated contemporary and "pre-industrial" scenarios, assumingly separated by a 100 yr span (cf. Table 4). Therefore, such a correspondence implies that the trophic status of the Entrance area has undergone only insignificant changes until the 1950s.

Table 4 also presents simulated basin-wise distributions of such important biogeochemical fluxes as the fixation and release of dinitrogen, nitrogen gas. In the shallow brackish Baltic Sea both processes are to a large extent determined by the feedback loops, involving redox mediated sediment reactions (e.g. Wulff et al., 2001a; Conley et al., 2002; Vahtera et al., 2007). The lower primary production and sedimentation in a less eutrophied state would require less oxygen and other electron acceptors for the mineralization of organic matter. This reduced demand in electron acceptors results in reduced dinitrogen release and conservation of inorganic nitrogen in the water column. In contrast, improved oxygen conditions enhance phosphorus removal from the water column and its retention in the sediments. Both phenomena lead to higher N:P ratio, less favorable for dinitrogen fixation by cyanobacteria. These feedbacks are clearly seen in Table 4, where the decrements of the dinitrogen release in sediments are almost identical to changes of primary production everywhere except for the deep layers in the Baltic Proper. Here, a larger decrement of the dinitrogen release appears to be inflicted by the shrinkage of the hypoxic zone (cf. Tables 3 and 4). A hundred years ago, this lesser extension of hypoxia was also the reason for higher phosphorus retention in sediments that, together with smaller release of dinitrogen and higher N:P ratio in external inputs (cf. Table 1), resulted in much weaker nitrogen limitation and a considerably smaller dinitrogen fixation.

In addition to such verification of the internal consistency of biogeochemistry in the model, we also attempted to compare simulated sediment variables with published data (Table 5). While analyzing these distributions, one should bear in mind that model variables represent the entire pools of bioavailable nitrogen (BEN, Table 5) and phosphorus (BEP, Table 5) homogeneously distributed over bottoms of the model boxes. Therefore, the model variables are not directly comparable to those average concentrations (TN and TP in Table 5) that can be estimated from scarce sediment samples sparsely distributed over mosaics of different bottom types (Carman and Cederwall, 2001). Within this uncertainty, the simulated values resemble the actual measurements in two important aspects. The contemporary ranges of concentrations span the same order of magnitude; hence, the sizes of sediment nutrient pools are realistic. Even more importantly, as can be calculated from Table 5, the basin-wise distribution of N:P ratio in simulated sediments is rather similar to the distribution in the sampled sediments, with a coefficient of linear correlation between them being equal to 0.78, if the Gulf of Riga sediments are ignored. Since the sediment N:P ratio reflects the degree of N depletion vs. P enrichment, such correlation indicates that large-scale internal biogeochemical mechanisms in the model are geographically realistic as well. Too low phosphorus content in the simulated Gulf of Riga sediments is caused by underestimated phosphorus input from the land as already discussed by Savchuk and Wulff (2007).

Further validation of the simulated sedimentary processes can be found in a very detail study of deep-water sediments in the Eastern Gotland basin (Hille, 2005). Here, the statistics (mean ± sd) of net nutrient accumulation in the surface sediments of  $114\pm115$  mmol N m<sup>-2</sup> yr<sup>-1</sup> and  $6.4\pm$ 6.1 mmol P m<sup>-2</sup> yr<sup>-1</sup> match the 107 mmol N m<sup>-2</sup> yr<sup>-1</sup> and 7.5 mmol P m<sup>-2</sup> yr<sup>-1</sup>, calculated as a difference between the simulated contemporary fluxes of sedimentation from the water column and release from the sediments in the Baltic Proper deep box. A simulated phosphate release of 14.7 mmol P m<sup>-2</sup> yr<sup>-1</sup> also reasonably corresponds to the release of  $11.9 \pm 11.0 \text{ mmol P} \text{ m}^{-2} \text{ yr}^{-1}$  estimated from the sediment cores (Hille et al., 2005). In the Gulf of Finland, simulated annual DIP efflux of 16 10<sup>3</sup> tonnes P yr<sup>-1</sup> is close to 18 10<sup>3</sup> tonnes P yr<sup>-1</sup> estimated from measurements by Lehtoranta (2003). DIP effluxes of 1.2 and 0.9 tonnes km<sup>-2</sup> yr<sup>-1</sup> simulated for the Danish Straits and Kattegat, respectively, reasonably correspond to 1 tonne km<sup>-2</sup> yr<sup>-1</sup> estimated for the Entrance area (Rasmussen et al., 2003 and references therein). The simulated net accumulations of 50-56 mmol N  $m^{-2} yr^{-1}$  and 4.8–5.2 mmol P  $m^{-2} yr^{-1}$  in the "pre-industrial" surface sediments are predictably higher than 44±31 mmol N m<sup>-2</sup> yr<sup>-1</sup> and 4.0±3.1 mmol P m<sup>-2</sup> yr<sup>-1</sup> estimated for a 100 year old layer in the dated sediment cores, where nutrient pools have been for a century affected by diagenesis.

Table 5
imulated and estimated concentration of nitrogen and phosphorus in the
Baltic Sea sediments (g N (P) $m^{-2}$ )

Basin	BB	BS	BPs	BPd	GF	GR	DS	KT
TN	6.8	7.8	12.9	10.2	12.5	9.2		
BEN	7.8	16.8	18.7	11.4	16.7	11.7	22.1	17.6
	(2.5/	(2.7/	(5.4/	(3.3/	(2.4/	(0.9/	(7.4/	(9.2)
	2.8)	3.5)	6.2)	3.8)	3.4)	1.2)	8.0)	9.4)
TP	4.2	4.5	3.9	2.5	5.3	5.2		
BEP	8.0	13.3	9.7	2.8	6.9	1.6	5.2	3.4
	(2.5/	(2.2/	(2.8/	(1.1/	(1.0/	(0.1/	(1.8/	(1.8/
	2.8)	2.8)	3.2)	1.2)	1.5)	0.2)	1.9)	1.8)

Note: TN and TP — basin-wide means of total nitrogen and phosphorus, respectively, in the uppermost centimeter of sediments estimated from data in Carman and Cederwall (2001); BEN and BEP — model variables representing contemporary and pre-industrial (LL/HL) pools of bioavailable nitrogen and phosphorus, respectively, in the surface "active" sediment layer of undefined thickness.

As these comparisons show, several important variables and fluxes simulated for "pre-industrial" conditions are rather similar to the values independently estimated from measurements corresponding to the conditions that existed a century ago. In other words, the SANBALTS model, calibrated and validated for the contemporary nutrient inputs (Savchuk and Wulff, 2007), also realistically responds to quite different, "pre-industrial" nutrient inputs. Thus, the model can with a reasonable confidence be used in scenarios requiring drastic changes of forcing (e.g. Wulff et al., 2007), including reconstruction of the "pre-industrial conditions".

#### 3.2. Nutrient limitation in pre-industrial conditions

Apparently, the simulated basin-wide annual averages, representing total nutrient pools, are not directly comparable to the winter surface fields and local concentrations of inorganic nutrients often considered as indicators for "reference conditions" (e.g. Schernewski and Neumann, 2005; HELCOM, 2006). Since the euphotic layer is to a large extent depleted of nutrients during the productive season, annual basin-integrated average should not generally exceed the winter surface concentration. Such constraint is largely fulfilled for the DIP indicator, as can be seen in a comparison of simulated "pre-industrial" annual average concentrations of 0.06–0.3  $\mu$ M P (cf. Table 2) with reference values for the winter surface concentrations of 0.1–0.35 µM P provisionally compiled for the "open waters" within the HELCOM EUTRO project (HELCOM, 2006). This comparison also shows similar increases of concentrations from the Bothnia Bay towards the Kattegat. However, for DIN the simulated annual means of 3- $5 \,\mu\text{M}$  ( $7 \,\mu\text{M}$  N in the Gulf of Riga) are significantly higher than the reference winter values of 2-3 (4) µM N from HELCOM (2006). Consequently, comparing to the Redfield molar ratio of 16, the derived range of reference N:P ratios of 5-11 indicate clear nitrogen limitation of the primary production everywhere except the Bothnia Bay, which has always been strongly limited by phosphorus according to both the estimated reference ratio of 35 and the simulated "preindustrial" ratio of 90. In contrast, simulated "pre-industrial" basin-wide N:P ratios of 12–24 imply that a hundred years ago the primary production in the Baltic Sea could be less limited by nitrogen and more limited by phosphorus than it is today. Similar relaxation of the nitrogen limitation relatively to contemporary measurements have been simulated with 3D model for the Bornholm and Eastern Gotland basins, where model results were considered most reliable by Schernewski and Neumann (2005).

The main difference between expert estimates of the reference conditions and simulated "pre-industrial" trophic state can be formulated as a question — whether a basin-wise distribution of nutrient limitation of the primary productivity in the past was similar to the contemporary situation or had it been shifted towards the phosphorus limitation? In the model this shift results from a reduced dinitrogen production and higher phosphorus retention by the sediments caused by a lesser extension of hypoxia. Such effects of hypoxia have recently been confirmed by a large-scale analysis of the actual field measurements spanning the latest three–four decades (Conley et al., 2002; Vahtera et al., 2007). In addition to these internal feedbacks, the numerical experiment run without any

nutrient loads from the land and atmosphere have shown that the pre-industrial nutrient import from the Skagerrak alone could sustain in the entire Baltic Sea over 600 thousand tonnes of DIN, which is about a half of the pools simulated for contemporary or pre-industrial conditions. Remarkably, even with such unrealistically reduced nutrient input the simulated DIN concentration in the surface Baltic Proper would be equal to 2.1 µM that is not much lower than in scenarios (cf. Table 2). The inorganic N:P ratio in the Skagerrak "oceanic" waters of 15.2 drives the integral inorganic N:P ratio in the entire Baltic Sea towards 15.7, i.e. practically to the same value as obtained in the pre-industrial scenarios. Although these considerations qualitatively support our hypothesis about the more extensive phosphorus limitation in the past than today, a confident quantification of the pre-industrial limiting conditions would require extensive paleoceanographic reconstructions and evidence that are far beyond the scope of this paper.

#### 4. Conclusions

A mechanistic model of the large-scale nutrient cycles SANBALTS has been tuned and validated for the contemporary trophic state of the Baltic Sea (Savchuk and Wulff, 2007, in press). In the present study, we exposed the model to two scenarios with rather drastically reduced loads corresponding to "pre-industrial" conditions. The scenarios are identical except of the nutrient inputs from the coastal point sources that were either assumed the same as they are today or were reduced proportionally to the changes of population occurred in the coastal cities during last century. The external input of total nitrogen has doubled and that of total phosphorus tripled since the beginning of the 20th century, according to these reconstructions.

The simulated "pre-industrial" conditions are validated by a comparison to actual historical measurements and reconstructions of such integral indicators of the trophic state as water transparency, oxygen concentration, primary production and net sediment accumulation. This validation gives certain creditability to other simulated values as well since all the simulated variables and fluxes are mutually interdependent. For the entire Baltic Sea, the simulated "pre-industrial" biogeochemical fluxes were about a quarter to a third of the contemporary fluxes, albeit with marked differences between basins corresponding to different changes of total nutrient loads.

Generally, the simulated "pre-industrial" trophic state is more phosphorus limited than today because DIP concentrations could be about 40–80% of their present day values, while DIN concentrations could be almost the same or even higher. The biogeochemical mechanism causing this shift towards phosphorus limitation in the model combines higher N:P ratios of the external nutrient inputs with feedbacks in the nutrient cycles. Reduced primary production leads to reduced sedimentation of organic matter and reduced oxygen consumption that results in reduced dinitrogen release and conservation of nitrogen in the water column. In contrast, improved oxygen conditions increase phosphorus removal from the water column and its retention in the sediments.

Independent validation of whether the Baltic Sea "preindustrial" trophic state was indeed more phosphorus limited than today would require additional paleo-ocenographic evidence.

#### Acknowledgments

We thank Pekka Kotilainen from the Finnish Environment Institute for assistance with the HELCOM data on the contemporary land loads and Jerzy Bartnicki from EMEP for the data on atmospheric nitrogen deposition. Miguel Rodriguez Medina was very helpful with the Secchi depth data. This work was supported by the Swedish foundation for Strategic Environment Research (MISTRA) within MARE programme, by the EU Sixth Framework Programme through ELME project, and by the Swedish Environmental Protection Agency funds for the Baltic Nest Institute.

#### References

- Aarup, T., 2002. Transparency of the North Sea and Baltic Sea a Secchi depth data mining study. Oceanologia 44 (3), 323–337.
- Andersen, J.H., Conley, D.J., Hedal, S., 2004. Palaeoecology, reference conditions and classification of ecological status: the EU Water Framework Directive in practice. Mar. Pollut. Bull. 49 (4), 283–290.
- Andersson, L., 1996. Trends in nutrient and oxygen concentrations in the Skagerrak-Kattegat. J. Sea Res. 35, 63–71.
- Anonymous, 2000. Directive 200/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for Community action in the field of water policy. Official J. L 327/1.
- Anttila, P., Paatero, P., Tapper, U., Järvinen, O., 1995. Source identification of bulk wet deposition in Finland by positive matrix factorization. Atmos. Environ. 29, 1705–1718.
- Aure, J., Dahl, E., 1994. The oxygen, nutrients, carbon and water exchange in the Skagerrak basin. Cont. Shelf Res. 14, 965–977.
- Aure, J., Danielsson, D., Svendsen, E., 1998. The origin of Skagerrak coastal water off Arendal in relation to variations in nutrient concentrations. ICES J. Mar. Sci. 55, 610–619.
- Brion, N., Baeyens, W., De Galan, S., Elskens, M., Laane, R.W.P.M., 2004. The North Sea: source or sink for nitrogen and phosphorus to the Atlantic Ocean. Biogeochemistry 68, 277–296.
- Carman, R., Cederwall, H., 2001. Sediments and macrofauna in the Baltic Sea – characteristics, nutrient contents and distribution. In: Wulff, F., Rahm, L., Larsson, P. (Eds.), A Systems Analysis of the Baltic Sea. Springer-Verlag, Berlin, pp. 289–327.
- Clarke, A., Juggins, S., Conley, D.J., 2003. A 150-year reconstruction of the history of coastal eutrophication in Roskilde Fjord, Denmark. Mar. Pollut. Bull. 46 (12), 1615–1618.
- Conley, D.J., Humborg, C., Rahm, L., Savchuk, O.P., Wulff, F., 2002. Hypoxia in the Baltic Sea and basin-scale changes in phosphorus biogeochemistry. Environ. Sci. Technol. 36, 5315–5320.
- Cornell, S.E., Jickells, T.D., Cape, J.N., Rowlans, A.P., Duce, R.A., 2003. Organic nitrogen deposition on land and coastal environments: a review of methods and data. Atmos. Environ. 37, 2173–2191.
- Ellermann, T., Hertel, O., Skjøth, C.A., Kemp, K., Monies, C., 2003. Atmosfærisk deposition 2002. NOVA 2003. Danmarks Miljøundersøgelser. 90s. – Faglig rapport fra DMU, nr. 466.
- Emeis, K.-C., Neumann, T., Endler, R., Struck, U., Kunzendorf, H., Christiansen, C., 1998. Geochemical records of sediments in the Gotland Basin—products of sediment dynamics in a not-so-stagnant anoxic basin? Appl. Geochem. 13, 349–358.
- Emeis, K.-C., Struck, U., Leipe, T., Pollehne, F., Kunzendorf, H., Christiansen, C., 2000. Changes in the burial rates and C:N:P ratios in Baltic Sea sediments over the last 150 years — relevance to P regeneration rates and the phosphorus cycle. Mar. Geol. 167, 43–59.
- Entciklopeditcheskii slovar' (Encyclopedic Handbook). 1890–1907. Publishing House of F.-A. Brockhaus and I.A. Efron, 86 volumes, Sankt-Petersburg (in Russian).
- Eriksson, H., Pastuszak, M., Löfgren, S., Mörth, C.-M., Humborg, C., 2007. Nitrogen budgets of the Polish agriculture 1960–2000: implications for riverine nitrogen loads to the Baltic Sea from transitional countries. Biogeochemistry 85, 153–168.
- Falkowska, L., 1985. Doplyw zwiazkow azotu i fosforu z atmosfery do Baltyku na podstawie badan w 1981 i 1983r. Stud. Mater. Oceanol. 48, 5–28.
- Fonselius, S., Valderrama, J., 2003. One hundred years of hydrographic measurements in the Baltic Sea. J. Sea Res. 49, 229–241.
- Frederiksen, M., Krause-Jensen, D., Holmer, M., Laursen, J.S., 2004. Long-term changes in area distribution of eelgrass (*Zostera marina*) in Danish coastal waters. Aquat. Bot. 78, 167–181.

- Grip, H., 1982. Water chemistry and runoff in forest streams at Kloten. . UNGI Rapport, 58. Uppsala University, Dept. Phys. Geography. 144 pp.
- Hannerz, F., Destouni, G., 2006. Spatial characterization of the Baltic Sea drainage basin and its unmonitored catchments. Ambio 35, 214–219.
- HELCOM, 2004. Nutrient pollution to the Baltic Sea in 2000. Baltic Sea Environ. Proc. 100 22 pp.
- HELCOM, 2006. Development of tools for assessment of eutrophication in the Baltic Sea. Baltic Sea Environ. Proc. 104, 62 pp.
- Hille, S., 2005. New aspects of sediment accumulation and reflux of nutrients in the Eastern Gotland basin (Baltic Sea) and its impact on nutrient cycling. PhD thesis, Rostock University, 120 pp.
- Hille, S., Nausch, G., Leipe, T., 2005. Sedimentary deposition and reflux of phosphorus (P) in the Eastern Gotland basin and their coupling with P concentrations in the water column. Oceanologia 47 (4), 663–679.
- Hoffmann, M., Johnsson, H., Gustafson, A., Grimvall, A., 2000. Leaching of nitrogen in Swedish agriculture – a historical perspective. Agric. Ecosyst. Environ. 80, 277–290.
- Jickells, T., 2005. External inputs as a contributor to eutrophication problems. 2005. J. Sea Res. 54, 58–69.
- Laakkonen, S., Laurila, S. (Eds.), 1999. The history of urban water management in the Baltic Sea Region. European Water Management, vol. 2, pp. 29–76.
- Laakkonen, S., Laurila, S. (Eds.), 2001. The sea and the cities. Ambio, vol. 30, pp. 263–326.
- Laamanen, M., Fleming, V., Olsonen, R., 2004. Water transparency in the Baltic Sea between 1903 and 2004. HELCOM indicator fact sheets 2004. http://www.helcom.fi/environment2.
- Laane, R.W.P.M., Brockmann, U., Van Liere, L., Bovelander, R., 2005. Omission targets for nutrients (N and P) in catchments and coastal zonez: a North Sea assessment. Estuar. Coast. Shelf Sci. 62, 495–595.
- Lehtoranta, J., 2003. Dynamics of sediment phosphorus in the brackish Gulf of Finland. Monogr. Boreal Environ. Res. 24, 1–58.
- Löfgren, S., Gustafson, A., Steineck, S., Stålnacke, P., 1999. Agricultural development and nutrient flows in the Baltic states and Sweden after 1988. Ambio 28, 320–327.
- Meier, H.E.M., 2005. Modeling the age of Baltic Seawater masses: quantification and steady state sensitivity experiments. J. Geophys. Res. 110, C02006. doi:10.1029/2004JC002607.
- Miltner, A., Emeis, K.-C., Struck, U., Leipe, T., Voss, M., 2005. Terrigenous organic matter in Holocene sediments from the Central Baltic Sea, NW Europe. Chem. Geol. 216 (3–4), 313–328.
- Nehring, D., Wilde, A., 1982. Untersuchungen über den atmosphärischen Nährstoffleintrag in die Ostsee. Acta Hydrochim. Hydrobiol. 10, 89–100.
- Oenema, O., Boers, P.C.M., van Eerdt, M.M., Fraters, B., van der Meer, H.G., Roest, C.W.J., Schröder, J.J., Willems, W.J., 1998. Leaching of nitrate from agriculture to groundwater: the effect of policies and measures in the Netherlands. Environ. Pollut. 102, 471–478.
- OSPAR Commission, 2003. OSPAR integrated report 2003 on the eutrophication status. 59 pp.
- Pollman, C.D., Landing, W.M., Perry Jr., J.J., Fitzpatrik, T., 2002. Wet deposition of phosphorus in Florida. Atmos. Environ. 36, 2309–2318.
- Radach, G., Pätsch, J., 1997. Climatological annual cycles of nutrients and chlorophyll in the North Sea. J. Sea Res. 38, 231–248.
- Rahm, L., Håkansson, B., Larsson, P., Fogelquist, E., Bremle, G., Valderrama, J., 1995. Nutrient and persistent pollutant deposition on the Bothnian Bay ice and snow field. Water Air Soil Pollut. 84, 187–201.
- Rasmussen, B., Gustafsson, B.G., Stockenberg, A., Ærtebjerg, G., 2003. Nutrient loads, advection and turnover at the entrance to the Baltic Sea. J. Mar. Syst. 39, 43–56.
- Richardson, K., Heilmann, J.P., 1995. Primary production in the Kattegat past and present. Ophelia 41, 317–328.
- Rydberg, L., Ærtebjerg, G., Edler, L., 2006. Fifty years of primary production measurements in the Baltic entrance region, trends and variability in relation to land-based input of nutrients. J. Sea Res. 56, 1–16.
- Savchuk, O.P., 2005. Resolving the Baltic Sea into seven subbasins: N and P budgets for 1991–1999. J. Mar. Syst. 56 (1–2), 1–15.
- Savchuk, O.P., Wulff, F., 2007. Modeling the Baltic Sea eutrophication in a decision support system. Ambio 36, 141–148.
- Savchuk, O.P., Wulff, F., in press. Long-term modelling of large-scale nutrient cycles in the entire Baltic Sea. Hydrobiologia.
- Savchuk, O.P., Elmgren, R., Larsson, U., Rodriguez Medina, M., 2005. Secchi depth and nutrient concentrations in the Baltic Sea: model regressions for the MARE's NEST. manuscript at http://nest.su.se.
- Schernewski, G., Neumann, T., 2005. The trophic state of the Baltic Sea a century ago: a model simulation study. J. Mar. Syst. 53 (1–4), 109–124.
- Schernewski, G., Wielgat, M., 2004. Towards a typology for the Baltic Sea. In: Schernewski, G., Löser, N. (Eds.), Managing the Baltic Sea. Coastline Reports, 0928-2734, 2, pp. 35–52 (2004).
- Schneider, B., Kuss, J., 2004. Past and present productivity of the Baltic Sea as inferred from pCO<sub>2</sub> data. Cont. Shelf Res. 24, 1611–1622.

- Schneider, B., Nausch, G., Kubsch, H., Peterson, I., 2002. Accumulation of total CO<sub>2</sub> during stagnation in the Baltic Sea deep water and its relationship to nutrient and oxygen concentrations. Mar. Chem. 77, 277–291.
- Seitzinger, S.P., Sanders, R.W., 1999. Atmospheric inputs of dissolved organic nitrogen stimulate estuarine bacteria and phytoplankton. Limnol. Oceanogr. 44, 721–730.
- Skogen, M.D., Soiland, H., Svendsen, E., 2004. Effects of changing nutrient loads to the North Sea. J. Mar. Syst. 46, 23–38.
- Sokolov, A., Wulff, F., 1999. SwingStations: a web-based client tool for the Baltic environmental database. Comput. Geosci. 25, 863–871.
- Sokolov, A., Andrejev, O., Wulff, F., Rodriguez Medina, M., 1997. The Data Assimilation System for data analysis in the Baltic Sea. . Systems Ecol. Contr., vol. 3. Stockholm Univ., 66 pp.
- Struck, U., Voss, M., von Bodungen, B., Mumm, N., 1998. Stable isotopes of nitrogen in fossile Cladoceran Exoskeletons: implications for nitrogen sources in the Baltic Sea during the past century. Naturwissenschaften 85, 597–603.
- Struck, U., Emeis, K.C., Voss, M., Christiansen, C.C., Kunzendorf, H., 2000. Records of Baltic Sea eutrophication in d<sup>13</sup>C and d<sup>15</sup>N of sedimentary organic matter. Mar. Geol. 164, 157–171.

- Tonderski, A., 1997. Control of nutrient fluxes in large river basins. PhD thesis Linköping University, Linköping Studies in Arts and Science 157.
- Vahtera, E., Conley, D.J., Gustafsson, B.G., Kuosa, H., Pitkänen, H., Savchuk, O.P., Tamminen, T., Viitasalo, M., Voss, M., Wasmund, N., Wulff, F., 2007. Internal ecosystem feedbacks enhance nitrogen-fixing cyanobacteria blooms and complicate management in the Baltic Sea. Ambio 36, 186–194.
- van Beusekom, J.E.E., 2005. A historic perspective on Wadden Sea eutrophication. Helgol. Mar. Res. 59, 45–54.
- Weckstrom, K., Juggins, S., Korhola, A., 2004. Quantifying background nutrient concentrations in coastal waters. A case study from an urban embayment of the Baltic Sea. Ambio 33, 320–327.
- Wulff, F., Rahm, L., Larsson, P., 2001a. Introduction. In: Wulff, F., Rahm, L., Larsson, P. (Eds.), A Systems Analysis of the Baltic Sea. Springer-Verlag, Berlin, pp. 1–17.
- Wulff, F., Rahm, L., Hallin, A.-K., Sandberg, J., 2001b. A nutrient budget model of the Baltic Sea. In: Wulff, F., Rahm, L., Larsson, P. (Eds.), A Systems Analysis of the Baltic Sea. Springer-Verlag, Berlin, pp. 353–372.
- Wulff, F., Savchuk, O.P., Sokolov, A., Humborg, C., Mörth, C.-M., 2007. Management options and effects on a marine ecosystem: assessing the future of the Baltic Sea. Ambio 36, 243–249.