**Baltic Earth Assessment Report on the biogeochemistry of the Baltic Sea**

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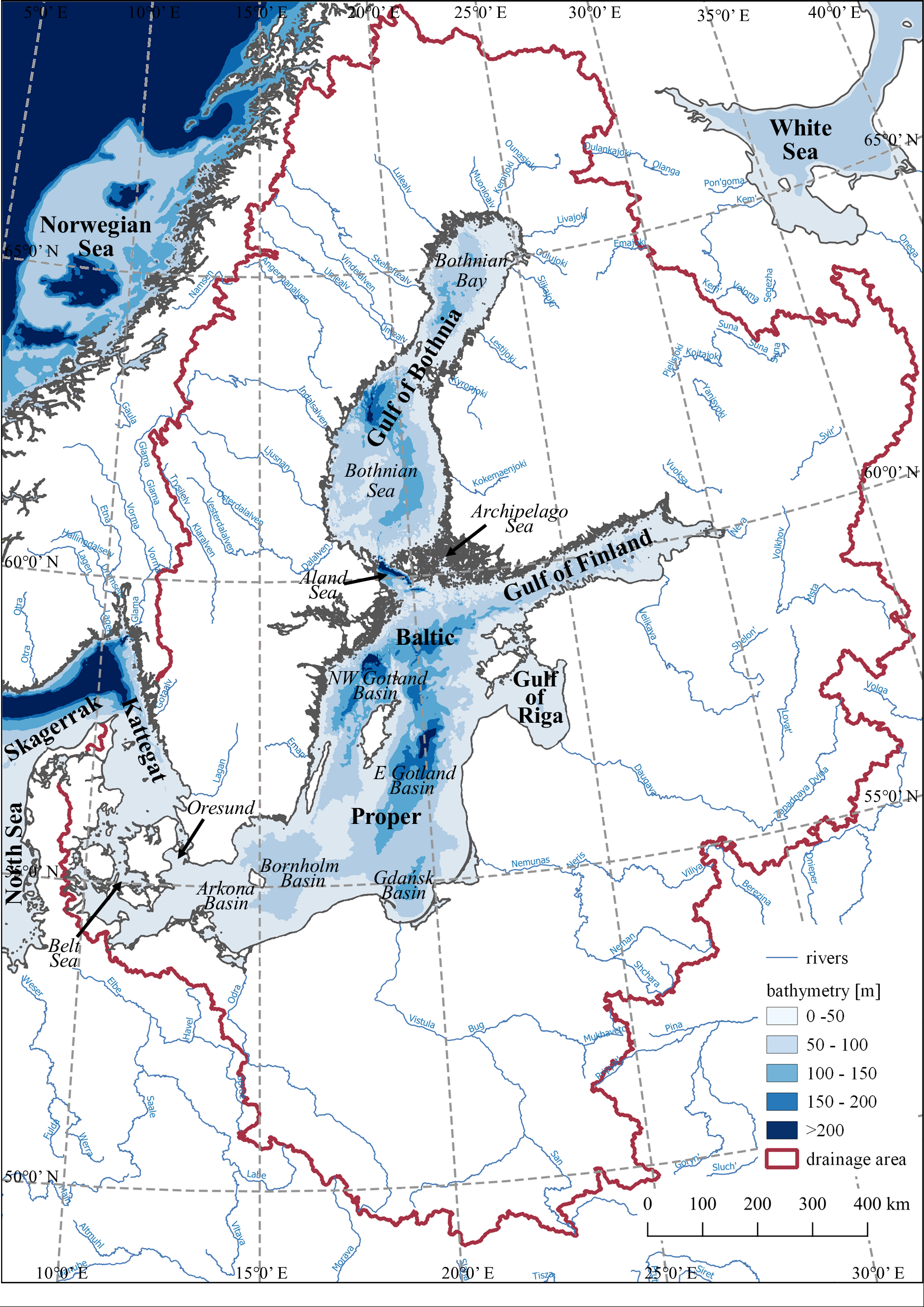
**1. Introduction**

The Baltic Sea is one of the most deeply studied marine ecosystems in the world. The long tradition of marine research is continued here within the framework of statutory activities of research institutes located around the Baltic Sea as well as in a number of national and international scientific projects. In recent years important contributions to the understanding of the Baltic Sea ecosystem has been made by the projects funded by BONUS - a funding mechanism dedicated for the Baltic Sea region. In addition to that, regular monitoring of physical, chemical, and biological variables is continuing under the auspices of HELCOM (Baltic Marine Environment Protection Commission) by the states surrounding the Baltic Sea. HELCOM has also been evaluating the contemporary state of the Baltic Sea ecosystem for almost 40 years (e.g. Melvasalo et al., 1981; HELCOM, 2018). Since more than two decades the knowledge on the Baltic Sea ecosystem has also been systematically assessed, initially by BALTEX and since 2013 by its successor Baltic Earth. As a result two comprehensive assessments have been already released: BACC I (2008) and BACC II (2015). The present study is one of the thematic Baltic Earth Assessment Reports (BEARs), which comprises a series of review papers that summarize and assess the available published scientific knowledge on climatic, environmental and human-induced changes in the Baltic Sea region (including its catchment). As such, the series of BEARs constitutes a follow-up of previous assessments: BACC I (2008) and BACC II (2015). BEARs are constructed around the major scientific topics (so called Grand Challenges) Baltic Earth deals with. One of those topics, summarized in this study, is related to the biogeochemical functioning of the Baltic Sea.

Marine biogeochemistry is a relatively new discipline, which deals with transport and transformations of chemical elements that are crucial for marine ecosystems, in particular: carbon, nitrogen, phosphorus, silicon and oxygen. As such, it takes into account all the physical, chemical, biological, and geological processes, which control cycling of those elements in the marine environment. On the top of that there are two overarching processes, namely: organic matter production and remineralization, which link inorganic and organic pools of substances and drive the matter cycling in the ocean. In addition to natural functioning of marine ecosystems with its periodicity, the anthropogenic pressure has to be taken into account as it has a potential to change the biogeochemical cycles on both global and regional scales. This directly links marine biogeochemistry with climate change and its consequences as well as with human-induced nutrients inputs, while the issues like deoxygenation, eutrophication or ocean acidification and their development in the future warmer and high CO2 world are in the center of interests of the present-day marine biogeochemistry. The case of the Baltic Sea shows that biogeochemical functioning of the marine ecosystems is especially complex in the coastal and shelf seas, which are interlinkages between land, open ocean and the atmosphere and where the anthropogenic drivers are manifested the most.

The biogeochemical functioning of the Baltic Sea ecosystem is directly related to its location, specific topography and hydrographic settings (Schneider et al., 2015, 2017). The Baltic Sea is a semi-enclosed shelf sea located in northern Europe (Fig. 1). Its catchment, being about four times larger than the sea surface itself and inhabited with about 85 million people, is highly diverse. The Scandinavian Peninsula being drained by many smaller rivers is not that densely populated and covered widely with boreal forests. This is in opposite to the continental part, which is to a large extent used for agriculture, while freshwater enters the Baltic Sea from that densely inhabited region through large riverine systems (Elken and Matthäus, 2008; Snoeijs-Leijonmalm and Andrén, 2017). The Baltic Sea is connected to the North Sea (and thus to the North Atlantic Ocean) through the narrow and shallow Danish Straits. Only sporadic inflows of saline waters from the North Sea and large riverine runoff cause that the Baltic Sea is one of the largest brackish water bodies on Earth. The salinity gradient on the surface extends along the southwest to northeast direction from more than 20 in the Kattegat through about S=7 in the central Baltic Sea (so called Baltic Proper) to almost freshwater conditions (S=2) in the highly separated northern part of the Gulf of Bothnia. In addition to horizontal salinity gradients, the water column in the Baltic Sea is stratified with the permanent halocline located in the Baltic Proper at the depth of about 60-80 m. It separates the surface brackish water layer (including euphotic zone) from more saline bottom waters and limits ventilation of deeper waters (Elken and Matthäus, 2008). This, together with eutrophication the Baltic Sea suffers from, leads to oxygen deficiency or even anoxia and/or H2S presence in the bottom waters in the vast part of the Baltic Proper and large parts of the Gulf of Finland. Consequently, the redox alterations of N and P biogeochemical cycles give rise to the “vicious circle” (see also 2.3), the positive feedback self-supporting the Baltic Sea eutrophication (Vahtera et al., 2007; Schneider et al., 2015, 2017; Savchuk, 2018).

Although the Baltic Sea can still be considered as the mesotrophic ecosystem, significant biogeochemical changes occurred here in the last decades. The increasing nutrients loads from rivers and atmospheric deposition having their maxima in 1980s lead to the increase in the ecosystem productivity. According to the conclusions made by Schneider et al. (2015) being the contribution to BACC II (2015) based on data and scientific literature available at that time, the net ecosystem production has increased in the Baltic Sea by factor 2.5 since 1920s/1930s, which corresponds well to the 3-fold increase in winter nutrients concentrations. As a consequence, the hypoxic and anoxic areas in the Baltic Sea have also expanded. Since the 1980s the nutrients loads to the Baltic Sea have been gradually decreasing. However, due to the long residence time of phosphorus in the system, extended by its liberation from sediments under anoxic conditions in the bottom waters, the decrease in ecosystem productivity and reduction in the extent of anoxic/hypoxic areas have not been observed. In 2007 the Baltic Sea Action Plan (BSAP) was adopted by HELCOM member states, in which the target loads of N and P have been set. They assumed controlled further reduction by 19% for N and 42% for P loads by 2021 compared to the period 1997-2003 (HELCOM, 2007). However, shortly after BSAP was published, Eriksson Hägg et al. (2010) reported that while there is space for P loads reduction, problems can occur with reducing N loads. This was linked mostly with an expected increase in livestock, protein consumption and agriculture development, especially in some eastern European countries.



*Figure 1: The Baltic Sea and its catchment.*

In 2013, when the previous assessment report (BACC II, 2015) was being prepared, there was too short duration of BSAP to conclude anything about its effectiveness based on observations, especially taking into account long response time of the system. Thus, most of the studies referred to model simulations assuming different scenarios of nutrients loads and climate change (e.g. Meier et al., 2011 and 2012; Neumann, 2010; Neumann et al., 2012; Omstedt et al., 2012). They showed that climate change will strengthen the effects of eutrophication, while the scale of those changes will depend largely on the nutrients loads scenario. According to those reports, keeping nutrients loads unchanged (business-as-usual scenario) will significantly increase the anoxic and hypoxic areas. On the other hand the implementation of BSAP has a potential to slightly decrease the extent of hypoxic and anoxic areas despite counteracting influence of climate change. Another feature shown in those studies was that rising atmospheric CO2 will lead to a pH decrease in the Baltic Sea surface waters, while the changes in ecosystem productivity will amplify the seasonal variability of pH without affecting the mean pH value.

Since the work on the last assessment (BACC II, 2015) was carried out, intensive research on the biogeochemical cycling in the Baltic Sea has been conducted including studies on past, present and future changes. This paper not only summarizes results of these recent studies but comprehensively assesses currently available, published knowledge on the biogeochemical functioning of the Baltic Sea, while pointing out knowledge gaps and future research needs. The scope of this study extends from changes occurring in the catchment and their influence on C, N and P loads to the Baltic Sea, through biogeochemical transformations of those elements in the coastal zone and changes in organic matter production (eutrophication) and remineralization (oxygen availability) to burial and turnover of C, N, P in sediments. Additionally, the paper also directly addresses the changes in the marine CO2 system (including ocean acidification), role of microorganisms in the biogeochemical functioning of the Baltic Sea and interactions between biogeochemical processes and chemical contaminants. Although the main focus in this study is on the cycling of C, N, P and O in the Baltic Sea, the attention is paid also to other substances, which take part as electron acceptors in redox processes playing important roles in organic matter remineralization under hypoxic and anoxic conditions.

This paper, apart from being a comprehensive assessment of the biogeochemical functioning of the Baltic Sea, is also a timely contribution and an important baseline to the discussion on future actions towards reaching good environmental status of the Baltic Sea. As of 2020, when this assessment is concluded, the first time frame for BSAP is coming to an end, while the discussion is continuing on the selection of new measures and actions for the updated BSAP.

**2. The current state of knowledge**

**2.1 Changes in the catchment and in the inputs to the Baltic Sea**

**2.1.1 Changes in external drivers**

The biogeochemistry of the Baltic Sea is largely fuelled by external loads of nutrients. Changes in nutrient loads are driven by human activities in the catchment and modified by climatic conditions (primarily temperature and precipitation). There has been dramatic changes in these factors and in their drivers over the past decades, and these are expected to change further into the future as affected by socioeconomic and climatic change (Pihlainen et al., 2020). For the factors affecting N and P loads to the Baltic Sea basin, modelling shows that changes in societal factors have a potential to outweigh effects of changes in the climate (Bartosova et al., 2019; Pihlainen et al., 2020).

Anthropogenic activities drive much of the nutrient inputs to the Baltic Sea, either through stream discharges or atmospheric deposition. Stream discharges of N and P are mainly driven by land use activities and wastewater discharges from urban areas. These wastewater discharges depend on the population, their diets and the efficiency of wastewater treatments (van Puijenbroek et al., 2015). The land use activities that drive N and P loads are mainly associated with agricultural activities (Reusch et al., 2018), and these activities vary greatly across the Baltic Sea Basin with much greater N inputs and losses in the southern compared to the northern part of the basin (Andersen et al., 2016). Therefore, regulating these activities may have the greatest impact on the N loads to the Baltic Sea (Olesen et al., 2019), although there may also be effects of changes in land use in other parts of the basin (Bartosova et al., 2019). The Baltic Sea is also affected by N (and S) deposition from the atmosphere, and these originate from many different sources, including ammonia from primarily agricultural activities in the region and beyond, and NOx emissions from on-land combustion and from shipping in the Baltic Sea (Karl et al., 2020).

An updated architecture for future scenarios related to climate change was developed in support of the IPCC process (Ebi et al., 2014). This approach distinguishes scenarios of climate change from those of socioeconomic developments. The climate change scenarios were simplified into four Representative Concentration Pathways (RCP), representing typical developments in radiative forcing during the 21st century and beyond. There are four core RCPs (RCP2.6, RCP4.5, RCP6.5 and RCP8.5) that represent key pathways for global warming, and where the numbers refer to the additional radiative forcing in 2100 in W m-2. These RCPs have been applied with global climate models (GCM) to project changes in future climate, and results of the CMIP5 study that contain projections of many different GCMs have been widely used in impact studies (Knutti and Sedlacek, 2012). These GCM projections need to be downscaled for use in impact models, in particular to resolve regional biases in the climate models. The results generally show a greater variation among climate models than between RCPs for projections until the mid-21st century, but greater variation among RCPs towards the end of the century.

Projections of climate change in the Baltic Sea region by 2050 compared to the late 20th century shows annual temperature increases of 1 to 5C with an average of 2.5C and annual precipitation increases of 0 to 20% with an average of 10% (Bartosova et al., 2019). The projected changes show greater temperature increases in the northern parts of the Baltic Sea Basin than in southern parts, in particular during winter (EEA, 2017). There are no clear spatial patterns of changes in projected precipitation across the area.

The revised IPCC scenario approach also includes Shared Socio-economic Pathways (SSPs) that reflect how different policies within climate change mitigation and adaptation interact with other sustainable development policies and pathways (Ebi et al., 2014). There are five core SSPs (SSP1-SSP5) that span a matrix of challenges for adapting to climate change and challenges for mitigating climate change. Zandersen et al. (2019) adapted this concept to the environmental problems for the Baltic Sea Basin, so that the SSPs span a matrix of challenges for adapting respectively mitigating Baltic Sea environmental problems. This resulted in narratives that allow quantification of changes in the drivers of emissions of N and P to the Baltic Sea through modelling (Bartosova et al., 2019; Pihalainen et al., 2020). Of the SSPs, the most contrasting in terms of nutrient loads to the Baltic Sea are SSP1 (sustainable development) and SSP5 (fossil-fueled development). The policies in SSP1 focus greatly on mitigating environmental issues leading to reductions in agricultural land use of technologies that lowers all emissions, whereas agricultural land use expands in SSP5 with some adoption of more efficient technologies. Results of scenario analyses with this approach shows that the targets of the Baltic Sea Action Plan can only be achieved following the trajectories of SSP1 (Pihlainen et al., 2020). Other scenario analyses have focused on impact of existing policies, and this has for instance shown that the EU Agricultural Policy does not contribute to lowering nutrient emissions from agricultural activities (Jansson et al., 2019).

**2.1.2 Hydrological regime**

A large amount of matter enters the Baltic Sea with riverine flows, playing a significant role in the biogeochemical conditions of the marine ecosystem. Changes in runoff can thus significantly impact inflows of nutrients and organic matter into the Baltic Sea. Several studies indicate an overall increase in mean discharge (Donnelly et al 2014, Bartosova et al 2019, Hesse et al 2015) to the Baltic Sea projected for future climate scenarios. Although the studies agreed on the significance and the direction of the change, the magnitude of the increase varied with the selected climate model (GCM/RCM), hydrological simulation model, or bias adjustment method.

Both the projected magnitude of the increase and the confidence in the change vary spatially across the drainage basin. The largest increase was projected for the northern part of the Baltic Sea Drainage Basin (BSDB). The projected change in the southern part was more uncertain and likely of lower magnitude than in the northern part. The projected increases in freshwater inflow to the Baltic Sea can affect the surface sea salinity with potentially negative effects on biotic communities in the Baltic Sea (Kniebusch et al 2019).

Most studies again agree on decrease in flows associated with snowmelt and increase of winter flows. The change of summer flows is then uncertain, varying from decrease (Donnelly et al 2014) to a smaller increase (Hesse et al 2015). The difference may be associated with a larger uncertainty in future evapotranspiration as calculated by the different models with different underlying assumptions.

Aside from the changes in magnitude, spatial, and temporal distribution of discharge, other characteristics are also affected. Klavins et al (2009) pointed out e.g. the ongoing reduction in ice cover and time shift in the ice break-up to earlier periods in all rivers in the Baltic Region except the most southern and most northern rivers. The ice cover duration was declining by 2.8 to 6.3 days per 10 years during the past 30 years, having been strongly influenced by the North Atlantic Oscillation index.

# **2.1.3 Nutrient legacy pools**

Over the last century, developments in agriculture have added significant amounts of N and P to agricultural land in the form of fertilizer and manure. This considerably exceeds the amounts removed by the harvest, and the current nutrient use efficiency is only about 50% for both N and P integrated over all agricultural land of the Baltic Sea catchment (McCrackin 2017). Therefore, large amounts of N and P have accumulated in the agricultural land. This led to hotspots of agricultural nutrient losses especially in the southern part of the catchment where N root zone leakage is significant (Andersen et al. 2016) and constitutes the major pathway (> 50%) of N emissions into the Baltic Sea. However, whereas catchment wide dynamic modelling of agricultural losses do exists for N, for P mainly direct observations (Pengrud et al. 2015) and empirical approaches have been developed addressing hot spots and risk areas (Djodjic and Markensten 2019) by means of empirical relationships. Further, the long-term legacy of these inputs may have greater impacts through their effects on the land-based nutrient pools than the accumulated legacy nutrient pools in the Baltic Sea (McCrackin et al. 2018). In this study the authors developed a three-parameter box model approach and estimated that more than 44 Tg P has accumulated in agricultural soils in the entire catchment over the last century of which 17 and 27 Tg P has accumulated in a mobile and stable storage pool, respectively. Presently, losses from this mobile pool contribute nearly half of the riverine P loads. The model suggests an overall residence time of P in the mobile pool of some 30 years and that riverine loads could decrease by as much as 10% by 2021 and 15% as a result of recent measures by 2050, even if there were no further reductions in P inputs.

# **2.1.4 Weathering and trends in alkalinity and TOC**

Carbon is entering the Baltic Sea mainly as total inorganic (TIC) and organic (TOC) C. Riverine input C amounts to 10.9 Tg C yr−1 of which 37.5 % has been estimated as TOC (Kulinski and Pempkowiak 2011). Most terrestrial derived TOC is respired in the Baltic Sea (Fransner et al. 2016; Fransner et al. 2019) and therefore exerts a positive feedback to atmospheric CO2 concentrations. In contrast, dissolved inorganic carbon (DIC) and alkalinity production via silicate and carbonate weathering exerts a CO2 sink, because atmospheric CO2 is consumed during the various weathering reactions between minerals and carbonic acid supplied by precipitation that form DIC and alkalinity (Berner 1991). In general, it is assumed that both TOC mobilization and weathering increase with temperature rise due to increased biomass turnover and faster chemical reaction rates. In fact, TOC concentrations have increased from 12 to 15.1 mg l-1 corresponding to an increased riverine input of 0.28 Tg C yr−1 between 1993-2017 in the northern boreal watersheds of the Baltic Sea catchment (Asmala et al. 2019). Similarly, weathering fluxes (as expressed as total dissolved solids, TDS and containing DIC and alkalinity) have increased by 10-20% over the last 40 years (Sun et al. 2017). The increase in TOC can be related to increasing trends in water discharge and pH whereas weathering fluxes could be related to precipitation only. However, CO2 consumption rates by weathering are estimated with about 3 g C m-2 yr-1 which corresponds to 3-30% of the net ecosystem carbon exchange in the boreal part of the Baltic Sea catchment (Sun et al. 2017). Overall, river chemistry data are more available and reliable for the boreal part of the Baltic catchment compared to the southern river catchments and therefore overall carbon trends in river loads and related climate feedback processes are still uncertain. This is also the reason why the potential dampening effect of increased alkalinity loads on Baltic Sea acidification still needs to be better quantified (Gustafson et al. 2019).

**2.1.5 Nutrient loads under changing climate**

Changes in nutrient loads from the Baltic Sea Drainage Basin (BSDB) to the Baltic Sea due to changing climate or anthropogenic influences were studied using a number of different approaches in recent years, including modeling framework, trends analyses, and functional relationships.

Bartosova et al. (2019) projected an increase in nitrogen and phosphorus loads to the Baltic Sea from a mini-ensemble of climate projections using the hydrological model E-HYPE. Hesse et al. (2015) reported decreasing trends for nitrate, ammonia, and phosphate loads on average in the Vistula lagoon basin using a hydrological model (SWIM). However, a wide range of impact projections were reported for individual ensemble members. Hägg et al. (2014) also projected an increase in nutrient loads using a split model approach to project changes in TN and TP loads into Baltic Sea sub-basins. Discharges modeled with CSIM for a climate projection ensemble were combined with a statistical approach for nutrients using population and projected population changes. Huttunen et al. (2015) in a study of Finnish basins draining to the BSB used a national nutrient load model (VEMALA) with a mini-ensemble for climate impacts. Even here were total nitrogen and total phosphorus loads found increasing.

Øygarden et al. (2014) used measurements in a number of small agricultural catchments to establish functional relationships between precipitation, runoff, and N losses from agricultural land, and qualitatively related their findings to projected precipitation change pattern across the BSDB under climate change scenarios, as well as mitigation measures to counter the climate-driven effects. While such data-driven approaches avoid uncertainties related to impact model chains, it is limited spatially by the availability of measurements and by assuming these relationships will stay unaffected in changing conditions.

Several studies compared the relative importance of the changing climate and changing socioeconomic conditions or adaptations scenarios, agreeing that the socioeconomic factors play a significant role (Pihlainen et al 2020, Bartosova et al 2019, Huttunen et al 2015, Hägg et al 2014) and may in some cases outweigh or even reverse the climate impacts (Pihlainen et al 2020, Bartosova et al 2019, Hägg 2015). Impacts of socioeconomic adaptation choices on nutrient loads to the BSB in the same magnitude range as climate impacts indicate the importance of effective mitigation strategies in the region.

**2.1.6 Atmospheric pathway of the nutrient input to the Baltic Sea**

The driving mechanisms of the atmospheric input are the dry deposition and scavenging with precipitation of a variety of gaseous and particulate nutrient species. Among the considered nutrients, the most-significant input from the atmosphere is for the nitrogen compounds (~220 kt or >20% of the total inflow), which is related to the strong atmospheric emissions of both oxidized and reduced nitrogen in Europe. Phosphorus input via the atmospheric pathway is utterly uncertain, but found to be roughly at the level of 2 kt or 5% of the total load. (Svendsen et al., 2015).

During the last two decades, the nitrogen supply via the atmospheric pathway has been noticeably reduced, owing to overall reductions of European emissions (Gauss et al., 2017). Largest reductions have been for oxidized nitrogen – since 1995 it has lowered by over 40%, whereas reduced nitrogen (ammonium and ammonia) has reduced with ~10%. The total nitrogen deposition on the Baltic Sea surface has therefore been reduced by about 30%.

The nitrogen supply to the Baltic Sea has strong geographical variation and a south-to-north decreasing tendency because the majority of the nitrogen sources are located south of the sea. There are also several mechanisms that control the deposition patterns. Oxidized nitrogen comes into the atmosphere mainly in the form of NO and NO2, which are very poorly soluble gases. Formation of secondary pollutants, such as nitric acid and nitrate aerosols requires certain time and favorable environmental conditions (for ammonium nitrate formation – also the presence of ammonia in the air). As a result, near-source deposition is quite limited and the long-range transport of the pollutants plays the key role in the final deposition pattern (Hongisto, 2011). Episodic character of the transport and deposition events leads to high variability of the load, even at annual level (Bartnicki et al., 2011). As a way to reduce the variability, a “normalized” deposition was introduced using the EMEP source-receptor matrices for reducing the meteorology-induced inter-annual variability – see e.g. Annex D in (Bartnicki et al., 2017). The above trends have been estimated using this noise-reduction approach. Interestingly, over 50% of these reductions were achieved in recent years (Gauss et al., 2017), (Bartnicki et al., 2011), despite the lowering pace of emission reduction. However, analysis of (Gauss et al., 2017) rather shows that the driving factors for the faster decrease in recent years was of meteorological origin – and the normalized deposition exhibits practically constant decreasing trend.

Nitrogen deposition pattern, apart from the south-north gradient, also reflects regional differences and different trends in Western and Eastern parts of Europe. In particular, the transformation of economy and environmental practices in Eastern Europe resulted in growth of NOx and NHx emission in several countries during the 1990s with subsequent reduction in the 2000s (Bartnicki et al., 2018).

Shipping in Baltic Sea is a significant contributor to the nitrogen deposition: in some regions and seasons, ships can contribute to more than 50 % to the total load (Stipa et al., 2008). In-average, about 17% of the total NOx load, according to (Jonson et al., 2015), originate from ship exhausts. The study also pointed out that this contribution has a clear tendency to grow. If no measures are taken, by 2030 it is estimated to reach 25%. With the currently planned Nitrogen Emission Control Area (NECA) in Baltic and North Seas, the growth is still expected but confined within < 20% (Jonson et al., 2015; Karl et al., 2019).

# **2.1.7 Nutrient inputs from the catchments**

Humans have for a very long period of time impacted nutrient inputs to the Baltic Sea through agriculture and deforestation. Potentially already during the Medieval era anthropogenic nutrient inputs caused significant eutrophication effects in the Baltic Sea (Zillen et al., 2008). With the industrialization, human and industrial waste sources also started to influence the nutrient inputs to the sea.

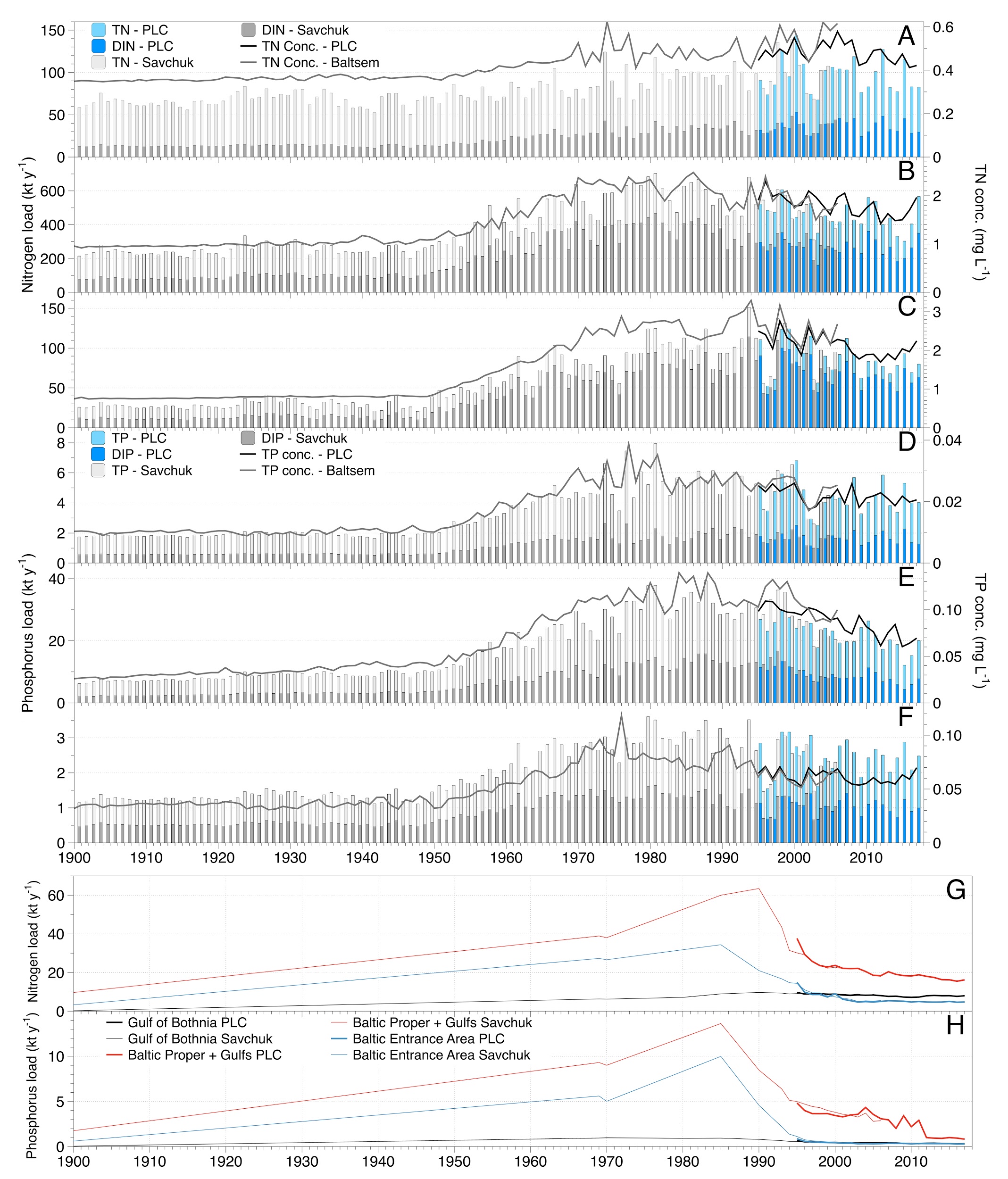
Several studies have investigated nutrient inputs at about 1900. Partly these studies been limited to certain countries as driven by the requirement of the WFD to evaluate the state of environment in relation to a reference state (e.g. Hirt et al., 2013; Rosenstrand Poulsen et al., 2017), but there are a few studies on pan-Baltic scale (e.g., Schernewski and Neumann, 2005; Savchuk et al., 2008).

First comprehensive time-series of riverine nutrient inputs to the Baltic Sea was constructed by Stålnacke et al. (1999). This study compiled data to a complete time-series covering the period 1970-1993, although data from several major rivers were lacking for the first decade, e.g. the major Polish rivers, and had to be reconstructed. This data set was later extended to 2006 by Savchuk et al. (2012a) using among other sources data from HELCOM PLC. Savchuk et al. (2012a) also added estimates of coastal direct point source loads. Savchuk et al. (2012b) reconstructed nutrient loads 1850-1970. For riverine loads they used established loads at 1900 as one fix point (Savchuk et al., 2008) and two step linear increase with a breakpoint at 1950. Coastal direct point source changes were estimated using population changes in major cities as a driver, but also here linear changes between a few specific years was assumed. Relatively recently HELCOM PLC started to annually publish comprehensively quality controlled time-series of nutrient loads, both riverine and coastal direct point sources loads (HELCOM, 2019). These time-series begin in 1995.

In Figure 2, the data sets from Savchuk et al., (2012a, 2012b) are shown together with the data from HELCOM PLC. It is evident that we know very little about the temporal development of nutrient inputs prior 1970, but there is clear evidence that much of the riverine load increase happened after 1950 as was assumed in the reconstruction. Differences between the loads from Savchuk et al. (2012a) and the HELCOM PLC time-series are relatively small in the overlapping period and some of the difference in concentrations might be due to differences in river runoff data used. In the Gulf of Bothnia riverine nitrogen loads (Fig 2-A) have been relatively stable during the monitored period (1970-2017), but for phosphorus (Fig 2-B) it seems that loads dropped to a lower level after about year 2000. Still, average flow weighted concentrations in the Gulf of Bothnia rivers are about 25% (nitrogen) and 100% (phosphorus) higher than pre-industrial concentrations. In the Baltic Proper (including Gulfs of Finland and Riga), riverine nitrogen loads (Fig 2-C) are weakly declining after about year 2000 from a long period (1970-2000) of relatively constant loads. However, for the Baltic Proper we have to bear in mind the lack of data from major rivers already before 1980. The riverine loads of phosphorus (Fig 2-D) are very much lower today than during maximum period (ca. 1975-2000) and concentrations dropped with close to 40% or so. This is a case where the two data sets differ quite a bit in the period 1995-1999. In the Baltic Entrance area, both riverine nitrogen and phosphorus loads decreased (Fig 2-E and 2-F). Flow weighted concentrations are at times misleading in this basin since a large proportion of the water flow is supplied with the Göta River that drains an area quite to the northeast of the remaining catchment of the Baltic Entrance area.

The temporal development of the nitrogen and phosphorus coastal point source loads (Figure 2-G and 2-H) have had a significant influence on the temporal development of the total loads, in particular for phosphorus.

In summary, riverine nutrient loads to the Baltic Sea generally decrease, in particular since about 2000. Today, coastal point sources contribute with relatively small amounts of nutrients compared to the rivers, but have been very large contributors to eutrophication in the past. Both in Baltic Proper and the Entrance area, it is estimated that today’s coastal point sources contribute with less nutrients than they did in 1900.



*Figure 2: Nitrogen (panels A-C and G) and phosphorus (panels D-F and H) nutrient load time-series. Total and inorganic riverine loads of nitrogen and phosphorus, respectively, are shown as bars and flow weighted concentrations sums of total nutrients calculated from the annual load and flow are shown as lines. The riverine loads to Gulf of Bothnia are shown in panels A and D, Baltic Proper plus Gulfs of Riga and Finland in panels B and E, and the Baltic Entrance Area (Kattegat, the Sound and Belt Sea) in panels C and F. In panels G and H are the loads from coastal direct point sources drawn for total nitrogen and total phosphorus, respectively. Loads are assessed from two sources (Savchuk et al., 2012a and 2012b) and from the HELCOM PLC (HELCOM, 2019 for total nutrients and directly from the HELCOM PLC-water database for the inorganic nutrients).*

**2.2 Transformations of C, N, P in the coastal zone**

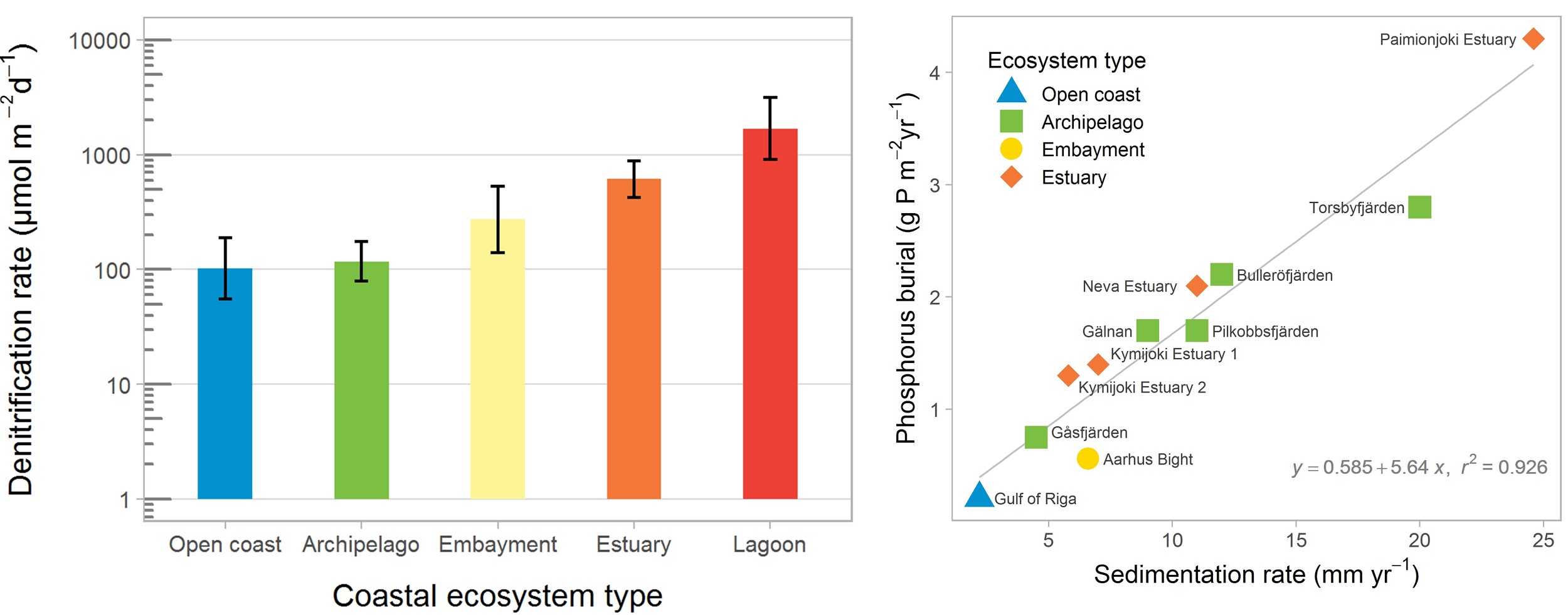
**2.2.1 Functioning of the coastal filter**

The coastal zone is the link between land and the open sea with a diverse range of habitats. These complex coastal ecosystems are important for the cycling of elements at global, regional and local scale. Inputs of organic matter (OM) and nutrients (N and P) from land are bypassed, transformed, retained and removed on their passage to the open sea through the coastal zone, removal being the only process permanently directing nutrients and OM outside the aquatic ecosystems (Asmala et al. 2017). The efficiency of the coastal filter is highly variable, depending on its hydromorphology and biological configuration (McGlathery et al. 2007; Carstensen et al. 2020). Coastal ecosystems harbor diverse biological communities and the strong benthic-pelagic coupling in shallow coastal ecosystems plays an important role for the functioning of the coastal filter. However, reduced functional biodiversity from nutrient over-enrichment and hypoxia, particularly the loss of deep-burrowing benthic macrofauna, hampers the coastal filter significantly (Conley et al. 2009, Norkko et al. 2012, Carstensen et al. 2020).

The most important processes for permanent nutrient removal in the coastal zone are denitrification and phosphorus burial (Fig. 3), and both these processes are strongly modulated by oxygen conditions. The Baltic Sea coastal zone removes approximately 16% of total N and 53% of total P inputs from land (Asmala et al. 2017), whereas less is known about the removal of OM in the coastal filter.

Denitrification removes N by reducing nitrate to dinitrogen that escapes to the atmosphere, and this process is regulated by availability of nitrate and labile organic carbon as well as temperature and oxygen concentrations (Piña-Ochoa and Álvarez-Cobelas, 2006). Denitrification typically occurs at the oxic-anoxic boundary in sediments, with nitrate supplied through nitrification of ammonia to nitrate or diffusive transport from the nitrate-rich overlying water. However, denitrification can be limited by nitrate availability, particularly with reduced nitrification under hypoxic conditions. Nitrification of ammonia to nitrate is inhibited by low oxygen in the sediments and loss of bio-irrigating macrofauna reduces transport of nitrate across the sediment-water interface. Denitrification rates vary by two orders of magnitude across Baltic coastal ecosystems (Fig. 3; Asmala et al. 2017).

Phosphorus burial occurs in fine-grained sediments in three general forms: 1) organic P, 2) Fe-oxide bound P, and 3) authigenic minerals. The latter form is generally thought to be dominated by apatite (Ca-P form), but in low salinity areas such as the Baltic Sea vivianite (Fe(II)-P form) can constitute a major sink for P (Slomp 2011). However, these burial forms have different stability and phosphate is liberated from Fe-oxides under anoxic conditions. On the other hand, increasing Fe inputs from land and decreasing salinity promote vivianite formation. P burial rates are tightly coupled to sedimentation rates and vary by an order of magnitude across coastal ecosystems (Fig. 3; Asmala et al. 2017).



*Figure 3: Mean denitrification rates coastal ecosystem types in the Baltic Sea (left panel). Error bars show the 95% confidence intervals for the mean estimates. Relationship between sedimentation accumulation rate and phosphorus burial in 11 study sites across the Baltic Sea (right panel). Redrawn from Asmala et al. (2017).*

Organic matter is removed through heterotrophic consumption, photochemical degradation, flocculation and burial. More than half of the organic carbon consumed by bacteria is respired as CO2 (Asmala et al. 2013), suggesting that this pathway is an important process for OM removal. Bacterial consumption is further stimulated by photodegradation, transforming biologically recalcitrant OM into more labile constituents (Moran et al. 2000). OM inputs from land are characterized by large molecules with humic properties, which are susceptible to flocculation when freshwater mixes with saltwater (Asmala et al. 2014). Flocculation contributes to OM burial, together with the autochthonous production of particulate OM. The biogeochemical processing of organic carbon is complex and variable across coastal ecosystems, with profound changes in the quantity and quality of OM inputs from land to sea.

Coastal ecosystems of the Baltic Sea are hydromorphologically diverse, ranging from lagoons, archipelagos, river-dominated estuaries, embayments and open coastal stretches, but they also vary broadly in their physical-chemical conditions (Carstensen et al., 2020). Particularly, hypoxia is widespread in the coastal zone due to stratification and low ventilation of bottom waters combined with high inputs of nutrients and organic matter (Conley et al. 2011). The increase in coastal hypoxia over the last century has therefore reduced the “filter function” of the coastal zone, enhancing nutrient enrichment of the open Baltic Sea. Despite that nutrient inputs to the Baltic Sea have been curbed, hypoxia remains prevalent in many coastal ecosystems and even continues to increase in some because of warming (Conley et al., 2011). Increasing temperature promotes hypoxia through reducing solubility of oxygen in surface water and enhancing respiration. However, warming also prolongs the productive period, when nutrient and organic matter inputs from land are intercepted and processed by coastal organisms, thereby enhancing the filter function. The diversity of coastal ecosystems and watersheds around the Baltic Sea has resulted in different trends in the past regarding biogeochemical functioning, and will likely experience different trajectories in the future.

**2.2.2 Coastal filter processes across different coastal ecosystem**

**2.2.2.1 Archipelagos**

Archipelagos are found mainly along the Swedish east coast and in southern Finland. Nutrient inputs are generally low from the boreal watersheds, dominated by forests, draining into the archipelagos. With the exception of archipelagos receiving large inputs of nutrients and OM from point sources (e.g. urban areas and pulp and paper industry), these coastal ecosystems are not severely affected by eutrophication. However, due to the complex bathymetry and restricted ventilation of bottom waters, some archipelagos are naturally prone to locally low oxygen conditions. Increasing inputs of nutrients and OM have disrupted the subtle balance between oxygen supply and consumption, causing hypoxia to develop in many locations (Conley et al. 2011). Archipelagos also have sheltered sedimentation basins due to the complex bathymetry, which promote particle trapping and subsequent elevated sedimentation. Consequently, archipelagos are important for burial of P and POM (Jilbert et al. 2018). However, due to the low terrestrial inputs of nitrate and labile organic matter, denitrification rates are low and primarily fuelled by autochthonous carbon from the spring bloom (Hellemann et al. 2017). In contrast, archipelagos with longer residence times that receive large inputs of N and OM constitute efficient filters for C, N and P. For example, Almroth-Rossell et al (2016) estimated that 72% and 65% of N and P inputs from land were removed in the Stockholm Archipelago.

**2.2.2.2 Estuaries**

Estuaries are found mainly in the south-western Baltic Sea, where population density in the catchments is higher and land use dominated by agriculture. Consequently, most of these estuaries have suffered from eutrophication for long time after receiving substantial inputs of nutrients and organic matter, although efforts to reduce these inputs have been successful over the past 2-3 decades (Riemann et al. 2016). Estuaries are typically stratified in the deeper parts, but the renewal of bottom water is dynamic, driven by changes in freshwater inputs and winds. Many estuaries experience seasonal hypoxia in summer and early autumn, when oxygen consumption outpaces oxygen supply. Despite nutrient reductions oxygen conditions have not improved, as these efforts have been counteracted by increasing temperatures (Conley et al. 2007, Carstensen and Conley 2019). Estuaries harbour rich biological communities, stimulating removal processes and thereby increasing the coastal filter efficiency (Carstensen et al. 2020). Estuaries with longer residence times have higher sedimentation rates, enhancing burial of phosphorus and OM as well as denitrification (Seitzinger et al. 2006). Due to the high productivity and degradation/removal of OM inputs from land, the characteristic of the OM pool rapidly changes from terrestrial to marine (Asmala et al. 2018).

**2.2.2.3 Lagoons**

Coastal erosion and sediment transport have formed lagoons along the southern coastline. These lagoons receive nutrients and OM through rivers of variable sizes, draining a watershed dominated by agriculture. Despite significant freshwater input, residence times can be long in these lagoons due to the restricted connection with the open Baltic Sea. The lagoons are mostly shallow with low burial rates, but the high inputs of nitrate and labile OM result in high rates of denitrification (Asmala et al. 2017). Remineralisation and denitrification rates are further stimulated by abundant benthic microalgae and chironomids (Benelli et al. 2018). However, the relatively high N removal in lagoons is compensated by nitrogen fixation during summers favourable to cyanobacteria blooms (Vybernaite-Lubiene et al. 2017).

**2.2.2.4 Large river plumes**

A number of large rivers discharge directly into the open Baltic Sea, with physical mixing of river and seawater dominating conditions in the plume. In the southern Baltic Sea, the large rivers deliver high inorganic nutrient inputs that sustain high productivity and sedimentation in the plume. The sedimenting particles are partly buried, partly remineralised and partly shuttled towards the deeper Baltic Sea (Hall et al. subm.). In contrast, the large rivers in the northern Baltic Sea have low nutrient but high DOM concentrations, and the considerable OM load partially flocculates in the plume and settles onto the seafloor. Overall, the large river plumes are mostly conduits of nutrients and OM and the filter efficiency is low.

**2.2.2.5 Open coasts**

Large stretches of the southeastern Baltic Sea coastline is dominated by open, sandy shores, where local inputs from land are negligible and exchange with the open sea is significant. Due to the strong and variable hydrodynamics sedimentation and resuspension occurs intermittently, whereas permanent burial is small. Denitrification rates are also small relative to estuaries and lagoon (Sundbäck et al. 2006). Consequently, open coasts are primarily conveyors of C, N and P, i.e. having a low coastal filter effect. The role of open coasts in biogeochemical processing of nutrients and OM will most likely not be altered substantially with the expected climate changes.

**2.2.3 Efficiency of the coastal filter in the future**

The Baltic Sea region is expected to warm even further and precipitation is expected to increase in the future. This will have large consequences for the biogeochemical processing of nutrients and organic matter in the coastal zone. Hypoxia is an important factor governing the efficiency of the coastal filter, and hypoxia will increase in a warmer climate due to reduced oxygen solubility and enhanced respiration, and this will consequently reduce coupled nitrification-denitrification and iron-bound P burial, which are two important processes of the coastal filter. The negative effects of developing hypoxia on the coastal filter will be most pronounced in archipelagos and estuaries.

Higher temperatures enhance remineralization processes and thereby ammonia production in sediments, which could stimulate coupled nitrification-denitrification, as long as nitrification is not inhibited by low oxygen. However, higher remineralisation under oxic conditions also reduces the availability of labile organic matter for denitrification. Most likely, denitrification will be stimulated in lagoons and estuaries where the availability of labile OM is high, whereas denitrification will be reduced in archipelagos that receive low inputs of nutrients and OM. Thus, the resulting outcome of temperature increase on denitrification varies among coastal ecosystem and the seasonality in nutrient and organic matter inputs and processing (Bartl et al. 2019).

The productive period is prolonged with increasing temperatures, already signified by the occurrence of earlier spring and later autumn phytoplankton blooms (Wasmund et al. 2019). This implies that more inorganic nutrients are intercepted, thereby enhancing the coastal filter efficiency. This effect is particularly pronounced in estuaries, lagoons and large river plumes, where most of the nutrients are discharged during winter and spring. Warming also stimulates nitrogen-fixing cyanobacteria, particularly in brackish lagoons, which counters the coastal filter effect by adding nitrogen to the coastal ecosystem.

Increasing precipitation may also involve increasing freshwater discharge, which reduces estuarine residence time and increases stratification. The resulting effect on oxygen supply below the pycnocline is highly site-specific and thus, oxygen condition may improve, deteriorate or remain unaltered with associated consequences for removal of nutrients and OM. Moreover, increasing freshwater discharge will extend the large river plume zone further and enhance the direct transport of nutrients and OM into the open Baltic Sea. Enhanced export of Fe from land and decreasing salinity from freshening in the coastal zone can promote burial of P in more stable forms, such as vivianite, in low-salinity archipelagos in the Gulf of Bothnia (Lenstra et al. 2018).

Inputs of organic matter from land are also expected to increase in the future in response to increasing precipitation and warming. These changes are expected to be most pronounced in boreal watersheds. It is therefore possible that this expected increase can alleviate carbon limitation of denitrification, which occurs in northern coastal ecosystems (Hellemann et al. 2017). However, the lability of this organic carbon source is considered low and may not significantly enhance denitrification.

In summary, there is no uniform response of the coastal filter to climate change for either C, N or P removal. The complexity of the biogeochemical processes as well as how these are modulated by coastal organisms mean that the future outcome of the coastal filter in a given ecosystem depends on several oppositely directed processes. Consequently, predicted changes in the coastal filter can only be resolved through coupled system-specific hydrodynamic-biogeochemical-biological coupled models.

**2.3 Changes in organic matter production (eutrophication)**

This section focuses mostly on the effects of nitrogen and phosphorus on organic matter production, as the primary reason for increasing organic matter production in the Baltic Sea is a relatively fast enrichment of its ecosystem with limiting nutrients, i.e. man-made eutrophication (e.g. Hutchinson, 1973; Smith et al., 2006; Chislock et al., 2013).

**2.3.1 Baltic Sea nitrogen and phosphorus budgets**

Rapid load changes, in particular between the 1950s and 1980s, have disturbed the balance between nutrient inputs, their biogeochemical sinks, and their export from the Baltic Sea. In result, combined nitrogen and phosphorus loads from land and atmosphere during the 1980s were about 3 and 4 times higher than in 1900 (Gustafsson et al., 2012). Pelagic and sediment nutrient pools followed the load increase with a delay and, after loads declined significantly starting from the mid 1980s, are close to a balance with present day nutrient loads (Gustafsson et al., 2012). Because denitrification provides an efficient sink for nitrogen, 87% of the annual nitrogen load is removed by biogeochemical processes, compared to only 69% of the annual phosphorus load (Tab. 1, Fig. 4). In total, about 99% of the annual nitrogen and 96% of the annual phosphorus load are lost by exports and biogeochemical processes such as denitrification and burial, the remaining 1% and 4% accumulate in water column and sediments. For comparison, the pelagic pools of nitrogen and phosphorus alone hold about 5 times the annual nitrogen and 11 times the annual phosphorus load (Savchuk, 2018). Between 1950-1980, during the peak increase in Baltic nutrient loads, about 1% of the annual nitrogen inputs and 12% of the phosphorus load accumulated in the water column alone; adding sediment storage, as much as 12% and 43% of the annual inputs accumulated in total (calculated from Gustafsson et al., 2012).

Organic compounds make up 81% of the pelagic nitrogen, but only 30% of the pelagic phosphorus pool in the Baltic Sea (Savchuk, 2018). Their long-term dynamics are poorly understood and over the past three decades (1970-2016, calculated from Savchuk (2018)) organic and inorganic nitrogen pools correlated weakly with each other (Bothnian Sea and Bay, Gulf of Finland, deep Baltic Proper, p<0.05, R2adj 0.08-0.20). Organic and inorganic phosphorus pools correlated only in the Baltic Proper above the halocline (p<0.05, R2adj 0.11). The share of labile compounds that are degradable by bacteria within weeks, is generally higher for dissolved organic nitrogen and, especially, phosphorus than for dissolved organic carbon (Hoikkala et al., 2015). DON and DOP feed into biogeochemical processes via direct phytoplankton uptake, or mineralization to ammonium and phosphate via photodegradation, bacterial mineralization, or excretion by heterotrophs within the microbial loop pool (Hoikkala et al., 2015 and sources therein). A modelling study estimates that 71% (89%) of phytoplankton nitrogen and phosphorus uptake in the Baltic Proper were channeled through dissolved organic pools (Kreus et al., 2015).

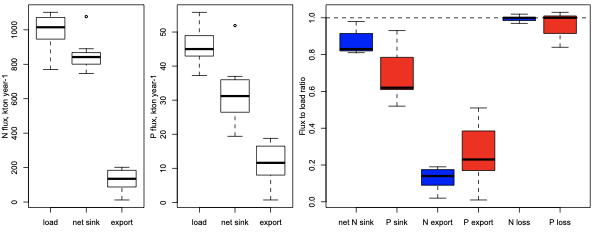
Nitrogen and phosphorus are exchanged intensely between different parts of the Baltic Sea. Both nutrients are transported westward from the Baltic Proper towards the Danish Straits, while phosphorus is also transported northwards, because its concentrations are successively and significantly declining from the Baltic Proper to the Bothnian Sea to the Bothnian Bay (e.g. Savchuk, 2018 and references therein). In fact, most budgets estimate that highest phosphorus removal takes place in the Bothnian Sea. Nitrogen, in contrast, is transported southward from the Bothnian Bay and Bothnian Sea into the Baltic Proper, where most of the nitrogen removal takes place (see Table 1 and references therein). Net nutrient exchange between the Baltic Proper and the Gulf of Finland and the Baltic Proper and the Gulf of Riga is directed towards or away from the Baltic Proper, depending on budget calculation method and period covered.

Nutrient pools in the Baltic Proper, which contains 61% of the entire pelagic nitrogen and 78% of the phosphorus pool (Savchuk, 2018), depend on deep water oxygen conditions. The pelagic nitrogen pool declines when deep water hypoxia expands, whereas the phosphorus pool increases (Conley et al., 2009, 2002; Savchuk, 2018; Vahtera et al., 2007). Therefore, both declining loads as well as fluctuating bottom water oxygen conditions have contributed to the decrease in Baltic Proper winter DIN concentrations since the 1980s with stable, high winter DIP concentrations (Andersen et al., 2017; HELCOM, 2018).

*Table 1: Nitrogen and phosphorus budgets for the entire Baltic Sea, both based on observations with water fluxes reconstructed from salt budgets, as well as on model results. Fluxes show loads from land and atmosphere, the net export to the North Sea, and the biogeochemical sinks within the Baltic Sea, indicating the basin with the largest sink in brackets (BP=Baltic Proper, BS=Bothnian Sea). The sink for nitrogen is calculated as the net result of nitrogen fixation and biogeochemical sinks.*

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Source** | **Period** | **Method** | **N flux, ktons year-1** | | | **P flux, ktons year-1** | | |
|  |  |  | **load** | **net sink** | **export** | **load** | **sink** | **export** |
| Liu et al. 2017a | 1971-1999 | simulated | 1102 | 1077 (BP) | 42 | 56 | 52 (BP) | 0.7 |
| Gustafsson et al. 2017 | 1980-2014 | simulated | 1099 | 890 (BP) | 199 | 51 | 31 (BP) | 11.6 |
| Savchuk 2005 | 1991-1999 | empirical | 1045 | 842 (BP) | 202 | 44 | 27 (BS) | 17 |
| Schneider et al. 2017 | 1985-2005 | empirical | 938 | 806 | 133 | 47 | 37 | 11 |
| Savchuk and Wulff 2007b | 1997-2003 | simulated | 1015 | 846 (BP) | 169 | 42 | 26 (BP) | 16 |
| Gustafsson et al. 2012c | 1997-2006 | simulated | 770 | 746 | 12 | 45 | 35 | 5 |
| Savchuk 2018 | 2005-2018 | empirical | 954 | 795 (BP) | 135 | 37 | 19 (BS) | 18.8 |

a) simulated with data assimilation; excludes Kattegat and transports are calculated as net flux between model grid points along the system boundary; b) simulated steady state budget; c) includes only bioavailable nitrogen fraction.

*Figure 4: Nitrogen (left) and phosphorus (middle) fluxes corresponding to nutrient budgets published for 1971-2018 (B.G. Gustafsson et al., 2012, E. Gustafsson et al., 2017; Liu et al., 2017; Savchuk and Wulff, 2007; Savchuk, 2018, 2005; Schneider et al., 2017). Right panel shows ratio between loads, sinks, export and net losses to external inputs. Box and whiskers show the distribution of the seven flux estimates.*

**2.3.2 Primary production and nutrient limitation**

Phytoplankton primary production is the main source of organic matter in Baltic Sea food-webs. Despite its importance, temporal and spatial dynamics are difficult to describe and data coverage is poor.

Currently, primary production is excluded from the coordinated HELCOM monitoring programme (<https://helcom.fi/action-areas/monitoring-and-assessment/monitoring-manual/introduction/>), but measurements are included in some national programs and are used as a research tool. Most commonly measured by 14C incubations, data reflect a continuum between gross and net primary production, i.e. between the carbon uptake rate into phytoplankton cells and the net effect of carbon uptake and release due to cellular respiration and exudation (Milligan et al., 2015; Sakshaug et al., 1997; Spilling et al., 2019). Primary production is rarely measured in-situ but mostly by incubating samples at selected light levels and temperatures, which introduces. methodological problems (ICES Working Group on Primary Production, 1987; Platt and Sathyendranath, 1993; Sakshaug et al., 1997). Further, water samples also contain bacteria, microzooplankton and detrital organic carbon, which all affect the fate of 14C in water samples (Spilling et al., 2019). Attempts to automate primary production measurements have focused on Fast Repetition Rate fluorometry (FREF), an automated in-situ technique based on light absorption by photosystem II (Houliez et al., 2017). Deriving carbon fixation from FREF is hampered by conversion efficiencies that depend on species and growth conditions (Hughes et al., 2018; Lawrenz et al., 2013), and by wavelength adaptations to capture cyanobacteria (Houliez et al., 2017; Simis et al., 2012). Other automated measurements based on the air-sea CO2 exchange describe the changes in total inorganic carbon, i.e. depict the net effect of autotroph and heterotroph processes.

The sparse primary production measurements that cover the entire seasonal cycle show that productivity declines northward from the Baltic Proper to the Bothnian Sea and Bothnian Bay, and that productivity is somewhat higher in shallow areas like the Danish Straits and the Gulf of Riga (Tab. 2). Temporal trends are best described for the Kattegat and the Danish Straits, where Rydberg et al. (2006) found that primary production doubled between 1950 and 1980 and its seasonality had changed from uniform rates throughout the growth season to pronounced spring and autumn blooms.

*Table 2: Annual primary production (g C m-2 year-1) in different areas of the Baltic Sea. Updated from Savchuk et al. (2012)*

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Period** | **Kattegat** | **Danish Straits** | **Baltic Proper** | **Bothnian Sea** | **Bothnian Bay** | **Gulf of Finland** | **Gulf of Riga** |
| 1970-1982 | 90-125a | 100-195a | 91-135f | 50-70d | 12-20d | 70-100d | 90-125a |
| 1991 |  |  | 100-200c |  |  |  |  |
| 1994-2006 | 116b -165a | 185-200a | 65e-200d  172j | 32e-52d | 16-17d | 80d-130g | 200h-250d |
| 2011-2012 |  |  |  |  |  |  | 353-376i |

a) Rydberg et al. (2006), b) Carstensen et al. (2003), c) Kazmarek et al. (1997), d) Wasmund et al. (2001) and references therein, e) Larsson et al. (2010), f) Renk (1990), g) Raateoja et al. (2004), h) Savchuk (2002) and references therein, i) Purina et al. (2018), oxygen method, j) recalculated from Gustafsson et al. (2013), Landsort Deep

The phytoplankton spring bloom is nitrogen limited in the Kattegat, Danish Straits, Baltic Proper and the Gulf of Finland, N/P co-limited in the Gulf of Riga, and phosphorus limited in the Bothnian Bay (see Schneider et al., 2015, 2017). The Bothnian Sea has gone through a change from alternating N/P limitation to N limitation of the spring bloom since the 1990s, in particular in its southern part, because inflows of phosphorus rich water from the Baltic Proper shifted the nutrient balance and probably also increased production and sedimentation (Ahlgren et al., 2017; Rolff and Elfwing, 2015). The increasing phosphorus concentrations without a matching nitrogen increase have also led to higher cyanobacteria abundance in the Bothnian Sea (Kahru and Elmgren, 2014; Kuosa et al., 2017), and might have induced nitrogen fixation in the range of external N inputs (Olofsson et al., 2020).

Since the late 1980s, phytoplankton seasonality changed, with an earlier spring bloom and a delayed autumn bloom in coastal (Wasmund et al., 2019) and open areas of the Baltic Proper (Kahru et al., 2016). The length of the growing season roughly doubled between 1998 and 2014 and the biomass maximum shifted from spring to summer (Kahru et al., 2016). Spring bloom intensity correlated with an index of winter DIN and DIP concentrations (Baltic Proper, Fleming and Kaitala, 2006; Groetsch et al., 2016), or winter DIN concentrations alone (Arkona and Bornholm basins, Raateoja et al., 2018). Declining nutrient levels since the end of the 1990s led to a slight drop in peak spring biomass (Groetsch et al., 2016), while higher water temperature, more intense solar radiation and low wind speed caused longer blooms and a faster transition between spring and summer communities (Groetsch et al., 2016).

However, it is not clear whether changes in phytoplankton seasonality also affect phytoplankton nutrient uptake and total primary production. In the global ocean increased stratification is expected to lead to a decrease in primary production because of a reduced nutrient supply from deeper water layers (*IPCC Special Report on the Ocean and Cryosphere in a Changing Climate*, 2019). In the Baltic Sea both temperature and salinity stratification have strengthened since the early 1980s (Liblik and Lips, 2019). However, in the Baltic Sea increased stratification reduces bottom oxygen concentrations (Meier et al., 2018a) and will thus affect nitrogen and phosphorus turnover.

**2.3.3 Nitrogen fixation**

Nitrogen fixation by cyanobacteria is a substantial source of nitrogen to the Baltic Sea, with inputs comparable to riverine and atmospheric loads (see e.g. Schneider et al., 2015, 2017). Warm (Jaanus et al., 2011; Kaiser et al., 2020; Kanoshina et al., 2003; Laamanen and Kuosa, 2005; Lips and Lips, 2008; Mazur-Marzcev et al., 2006) and calm conditions (Kanoshina et al., 2003; Mazur-Marzcev et al., 2006) seem to favor cyanobacteria blooms. At time-scales longer than five years, surface accumulations are related to the total amount of phosphorus in the Baltic Sea, bottom water hypoxia and temperature (Kahru et al., 2020), at shorter time-scales cyanobacteria blooms follow pronounced three-year oscillations, probably caused by biological feedback mechanisms (Kahru et al., 2018).

The dinitrogen fixed during cyanobacterial blooms becomes readily available to other primary producers and stimulates summer production in the entire food web, from zooplankton and benthos to fish (e.g. Karlson et al., 2015; Motwani et al., 2018; Svedén et al., 2016). Diazotrophs thus relieve ecosystem production from nitrogen limitation and enable communities to make use of the phosphorus pool left in the water column at the end of the spring bloom (Nausch et al., 2008; Raateoja et al., 2011; Rahm et al., 2000; Schneider et al., 2017; Wasmund, 1997).

Nitrogen fixation is therefore an important link in the vicious circle of Baltic Sea eutrophication (Vahtera et al., 2007). When the oxygen demand for organic matter mineralization exceeds the limited oxygen supply to deep-water layers and sediments, oxidation of organic matter below the halocline gives rise to hypoxia/anoxia with corresponding redox alterations of the nutrient cycles (cf. Sections 2.5-6). In the vicious circle, the inorganic nitrogen removal due to denitrification in the hypoxic zone and the release of phosphates from iron-humic complexes in the anoxic zone result in a Redfield excess of phosphorus. Cyanobacteria channel the phosphorus excess into biotic cycling via nitrogen fixation, thus increasing primary production, sedimentation and decomposition of organic matter, which, in turn, leads to further expansion of hypoxic and anoxic zones with increased denitrification and DIP release (Savchuk, 2018 and references therein; Vahtera et al., 2007). In the Baltic Proper, cyanobacteria blooms followed the increasing phosphorus loads with a lag of 20 years (Kaiser et al., 2020). The large-scale manifestation of the vicious circle has been empirically supported by the significant correlations between satellite-detected cyanobacteria surface accumulations, water temperature, and integral phosphorus pool (Kahru et al., 2020; Savchuk, 2018). In recent years, this self-sustaining positive feedback is further reinforced by increased oxygen consumption in saline inflowing waters, making inflows less efficient in aerating the water below the halocline (Meier et al., 2018b).

**2.3.4. Expected future changes**

Given present day nutrient loads, Baltic Sea biogeochemical models show that nutrient turnover and productivity will increase in warmer climates (Meier et al., 2012a, 2012b; Meier et al., 2012; Meier et al., 2018a, 2014). Still, load reductions will affect Baltic Sea nutrient concentrations and productivity more than climate change, as seen by a large ensemble of six coupled physical-biogeochemical models and 58 transient simulations (Meier et al., 2018a). Expected changes differ between the more eutrophic southern areas and the northern basins. In the Baltic Proper, climate change will increase primary production because warming and reduced bottom-water oxygen levels will intensify nutrient turnover. Higher pelagic regeneration will benefit phytoplankton, whereas benthic production depending on export from the euphotic zone, will decline (Ehrnsten et al., 2020). In the more oligotrophic Bothnian Sea and Bothnian Bay primary production might decrease because higher DOC inputs reduce transparency and favor heterotrophic bacteria (Andersson et al., 2015).

Stratification and future bottom water oxygen conditions will play a major role for future productivity and nutrient concentrations in the Central Baltic Sea. With present nutrient loads bottom water oxygen is expected to decline because stratification increases (Meier et al., 2018a). Warmer inflows take up less oxygen from the atmosphere and carry less dissolved oxygen into bottom waters (Meier et al., 2011; Skogen et al., 2014), where hypoxia in turn intensifies phosphorus cycling (Meier et al., 2011). In high-warming scenarios, also global mean sea level rise will start to contribute to increased salt-water inflows and stratification (Meier et al., 2017). Stratification changes are uncertain in sub-basins that receive salt water intrusions from the Baltic Proper, in particular the Gulf of Finland (Meier et al., 2019), where lateral intrusions drive stratification (Vankevich et al., 2016), depending on halocline position in the Baltic Proper (Meier et al., 2019). Both the Gulf of Finland and the Bothnian Sea might become less stratified in the future, with higher bottom water oxygen concentrations and reduced phosphorus turnover (Meier et al., 2018a).

Future nutrient loads, warming, and changes in stratification and oxygen conditions will also shift the balance between nitrogen removal via denitrification and nitrogen fixation, which determines Baltic Sea nitrogen levels (Skogen et al., 2014). Future warmer climates will have longer periods suitable for cyanobacteria growth (Hense et al., 2013; Neumann et al., 2012). Simulations suggest that denitrification is able to counteract nitrogen fixation and external inputs except in load increase scenarios (Skogen et al., 2014), where the fraction of nitrogen inputs removed by denitrification starts to decline (Meier et al., 2012b).

The Baltic Sea, especially its entrance area, will also be affected by changes in nutrient concentrations in the inflowing North Sea water. These are highly uncertain and mostly determined by water exchange with the North Atlantic (Meier et al., 2019; Skogen et al., 2014). Assuming that changes at the North Sea – Atlantic boundary would halve Skagerrak nitrogen and phosphorus concentrations, primary production would drop by 40% in the Kattegat and 10% in the Gotland Sea (Meier et al., 2019).

In the more oligotrophic northern Baltic Sea, climate change effects will be modulated by increasing DOM inputs (Andersson et al., 2015). DOM from the northern part of the drainage basin contains high amounts of CDOM, which is conservatively mixed over a large salinity range (Harvey et al., 2015). Mesocosm experiments (Paczkowska et al., 2020) and field studies in estuaries (Andersson et al., 2018) show that the deteriorating light climate at high DOC inputs can counteract nutrient effects on primary production. In field studies this effect is found in estuaries with high CDOM combined with low nutrient input, leading to a decline in primary production and an increase in bacterial production (Andersson et al., 2018). Suppressed phytoplankton growth and food-webs shifts to microbial loop dominance are seen when DOC concentrations increase by 25%-30% on top of background levels (Andersson et al., 2013; Lefébure et al., 2013), probably modulated by nutrient competition between phytoplankton and bacteria (Meunier et al., 2017).

**2.4 Changes in organic matter remineralization and oxygen availability**

**2.4.1 Oxygen supply**

Dissolved oxygen concentration in the water column is controlled by the supply of oxygen by vertical and lateral transports and by oxygen consumption in the water column and sediment (e.g. Savchuk 2018). In the eutrophied Baltic Sea, water below the permanent pycnocline is oxygen depleted because both large but sporadic barotropic inflows of oxygenated saline water from the North Sea, so-called Major Baltic Inflows (MBIs) (e.g. Mohrholz et al., 2015) and smaller inflows preferably ventilating the halocline but also sometimes deeper layers (e.g. Feistel et al., 2003; Meier et al., 2004; Neumann et al., 2017), do not always compensate for the oxygen consumption due to organic matter remineralization after the spring and summer blooms. Three narrow straits and shallow sills in the western Baltic Sea constitute natural obstacles, constraining the free water exchange with the world ocean (Matthäus et al., 2008). As a result, the deep-water layer of the central basins is prone to hypoxic or even euxinic conditions.

Although MBIs explain only about 20% of the total salt input (Mohrholz, 2018), they are the only mechanism that are able to ventilate the deeper parts of the Baltic Sea. In this respect the Baltic Sea is special compared to other coastal seas because it is characterized by largely varying topography with the deepest areas in the central eastern (Gotland Deep, ~250 m) and northwestern (Landsort Deep, ~459m) Gotland Basin and long residence time. For the period 1887-2017, MBIs do not show a systematic trend, but a pronounced multi-decadal variability of about 25-20 years (Mohrholz, 2018). On average MBIs occur once per year. However, there are periods without any MBI, so-called stagnation periods, e.g. during 1983-1992. According to model simulations, such periods without MBIs and with decreasing salinity are part of the natural variability of the system and occur once per century on average (Schimanke and Meier, 2016).

In contrast to periods with stronger stratification, during periods with lower average salinity, deeper halocline and weaker vertical stratification, vertical fluxes of oxygen are larger and capable of ventilating the bottom water along the rim of the sub-basins with permanent halocline such as the Bornholm and Gotland basins (Väli et al., 2013). Hence, during stagnation periods without MBIs basin-wide hypoxic areas are smaller compared to periods with many MBIs although the very deep areas of the Baltic Sea suffer from oxygen depletion (Conley et al., 2002; 2009; Meier et al., 2017).

Another important process is entrainment of ambient water into the inflowing gravity-driven saltwater plumes (Kõuts and Omstedt, 1993; Meier et al., 2018; Neumann et al., 2017). Most of the oxygen arriving with MBIs in the bottom water at Gotland Deep is oxygen from the Baltic Sea interior and not from the Baltic Sea entrance area (Neumann et al., 2017). Due to a strong internal vertical re-circulation, the entrainment of inflowing water is considerable. From 20-year long records of observations Kõuts and Omstedt (1993) showed that the inflowing dense water is diluted by surface water and that the flow increased by a factor of four on its way between Kattegat into the Landsort Deep.

Further, no trend was found in the impact of wind-induced vertical turbulent mixing on the multi-decadal variations in salinity (Radtke et al., 2020).

In summary, the ventilation of the Baltic Sea deep water is of very intermittent character given by the frequency of MBIs and shows pronounced multi-decadal variations with improved bottom oxygen conditions during stagnation periods. Due to the large interannual to multi-decadal variability, systematic trends in deep water ventilation on centennial time scale are difficult to detect. However, long-term changes in oxygen consumption are pronounced (Gustafsson et al., 2012; Carstensen et al., 2014; Meier et al., 2018; 2019a; 2019b) and will be discussed in the following.

**2.4.2 Organic matter remineralization**

The predominant sink for oxygen and oxidants are bacterially mediated degradation processes of organic matter. These processes take place in the water column as well as in the marine sediments. Fluxes between water and sediment connect both environments and thus sedimentary oxidant sinks impact the water column and vice versa. Oxygen flux between atmosphere and ocean keep the oxygen concentration in the mixed surface layer close to equilibrium with atmospheric oxygen. Therefore, oxygen deficiency becomes important below a temporal or permanent pycnocline, which hampers an oxygen flux from the surface. In the Baltic Sea, both temporal and permanent regions with oxygen deficiencies exist.

Carstensen et al. (2014) reported about a 10-fold hypoxia increase within the last 115 years. Their analysis is based on oxygen profiles and estimated total apparent oxygen utilization. As primary cause for the increase, they detected increased nutrient loads from land, but also climate warming. Similar results were found using modeling (Gustafsson et al., 2012; Meier et al., 2019a; Meier et al., 2019b).

For the time period 2004-2014, Schneider and Otto (2019) did not find an interannual variability of oxygen consumption in the eastern Gotland Basin. Furthermore, the mineralization rate was relatively constant at 2 mol carbon m-2 year-1. The detected mineralization rate is independent of the redox conditions in the water column. The estimates were made on basis of observed total inorganic carbon dynamics during so-called stagnation periods.

The oxygen consumption rates after Major Baltic Inflows (MBI) have increased since the 1970s (Meier et al., 2018). This trend was detected from observations as well as from model simulations. The explanation, derived from the model study, is the increased abundance of particulate organic matter (POM), especially zooplankton, which is mixed into the inflowing water. The higher POM concentration in the inflowing bottom water accelerates oxygen consumption due to respiration and mineralization.

An additional explanation was provided in a model simulation study by Neumann et al. (2017). The focus on MBIs only is not sufficient to quantify the oxygen supply to the deep Baltic Sea waters. For the MBIs 2003 and 2014, it could be shown that the 2003 event was accompanied by several smaller events altogether exceeding the stronger 2014 MBI 1.5-fold. A difference in oxygen consumption rates immediately after the MBIs in 2003 and 2014 could not be shown.

An example for hypoxia dynamics in the Gulf of Finland (GoF) was given in an observational study by Stoicescu et al. (2019). The GoF shows a strong seasonality due to a temporal pycnocline. The oxygen consumption during an established pycnocline was estimated to 0.31-0.82 mg l-1 month-1 for 2016 and 2017, respectively.

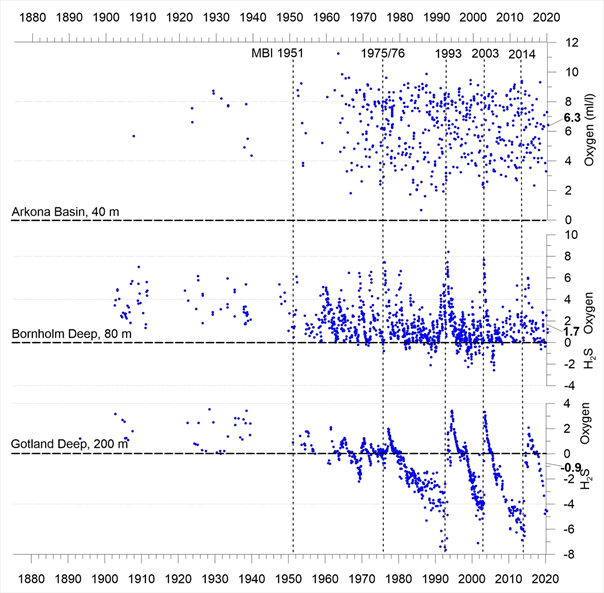
**2.4.3 Changes in oxygen concentration**

Figure 1 shows the exemplary development of dissolved oxygen and hydrogen sulphide concentrations in the near bottom layer of three key stations. The Arkona Basin is the westernmost one, located close to the sills and narrow straits. It shows highly dynamic oxic conditions in the deep water, which are rarely hypoxic (below 2 ml/l) and influenced even by weak and short inflow pulses via the sills, which occur in high frequency. The long-term mean of 6.3 ml/l is well above the hypoxic limit with no sign of reduction over time.

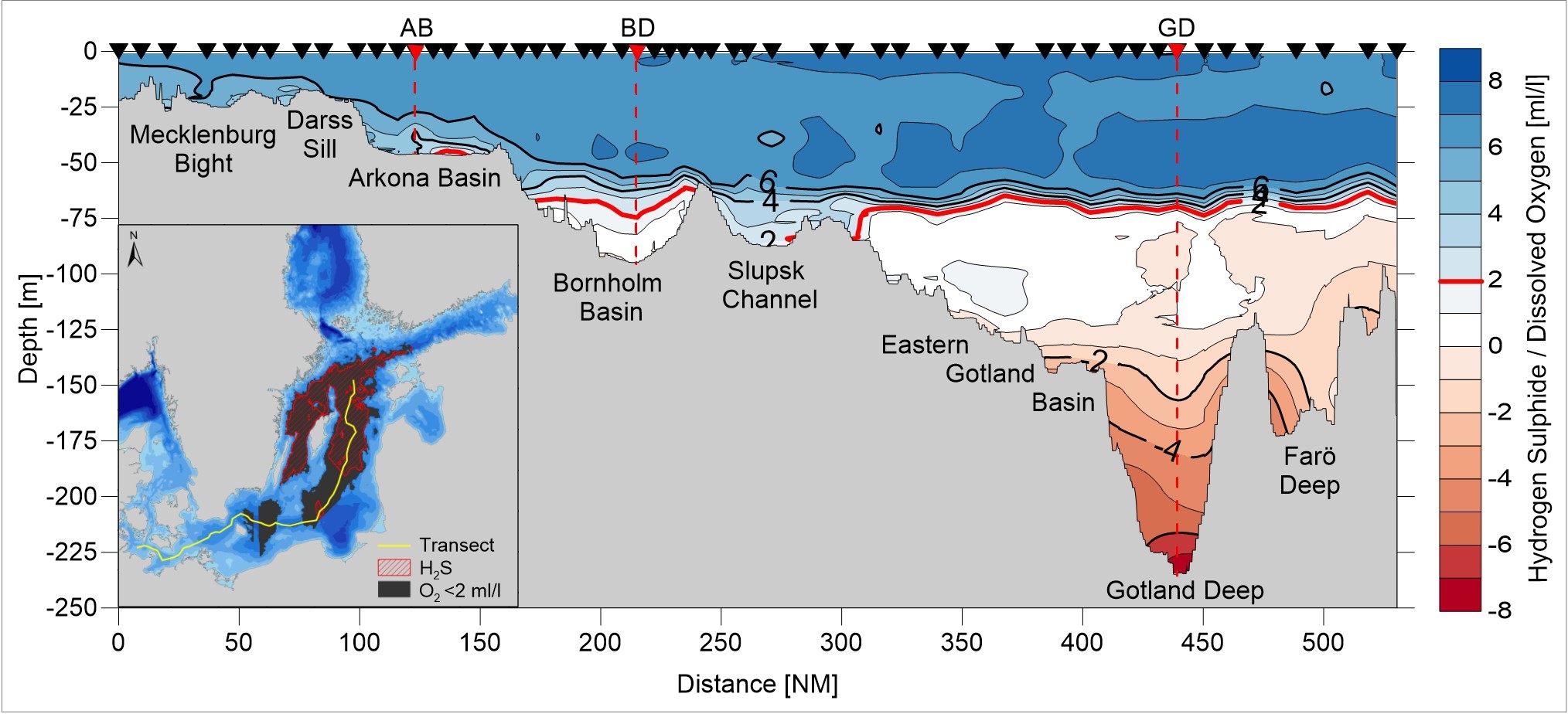
In the eastward connected Bornholm Basin of 90 m water depth the situation is less dynamic with larger distance to the world ocean. Only more intense inflow events, occurring with multiannual intervals, ventilate bottom waters of this basin, leading to alternating oxic and hypoxic deep-water conditions. The long-term mean of 1.7 ml/l tends to hypoxic bottom water conditions with alternating oxic phases. From 1980 to 2010 hydrogen sulphide was often observed, but this has not occurred during the last decade.

In the central sub-basins, conditions are more stagnant and deep-water renewals/ventilations are rare. As example, the conditions in the Gotland Deep (Eastern Gotland Basin) show persistency of hydrogen sulphide during the last decades with a mean of -0.9 ml/l over time (Fig. 5). A lateral view of recent oxic, hypoxic to euxinic conditions in the water column is displayed along a cross section from western to the central Baltic Sea (Fig. 6). After the latest period of intense inflow activity during 2014 to 2017 and several ventilations of the central deep water the environmental status switched back to stagnant conditions of large hypoxic to euxinic water volumes below 70 m water depth. Hansson et al. (2019) calculated a hypoxic area of 82.000 km2 (32 % of the Baltic Proper) and a hypoxic volume of 3100 km3 (22 % of the Baltic Proper water volume) for late summer 2019. Such levels were already reached sporadically in the 1970s but it has become more persistent during the recent two decades. Furthermore, hydrogen sulphide concentrations are higher and more persistent in the last two decades compared to earlier times. Under recent oceanographic and climatic conditions with high freshwater discharge and strong water column stratification, hypoxia nearly attains the maximum spatial extent possible for this ecosystem.

Worsening of the oxygen deficit by increasing concentrations of hydrogen sulphide due to amplified oxygen consumption rates over the last decades was shown by model simulations comparing the last 150 years in more detail (Meier et al. 2018).



*Figure 5: Long-term variations of dissolved oxygen concentrations and hydrogen sulphide (converted to negative oxygen equivalents) in the near bottom layer at the key stations Arkona Basin (BY2, TF0113), Bornholm Deep (BY5, TF0213) and Gotland Deep (BY15, TF0271) (source: IOW-DB, ICES).*



*Figure 6: Transect from the Great Belt to Baltic Proper, showing the recent state of dissolved oxygen and hydrogen sulphide (converted to negative oxygen equivalents) concentrations along the pathway of saltwater inflows from the western Baltic Sea to the deep basins (July 2020, unpublished data of cruise EMB242, IOW). Locations of CTD measurements and water sampling are marked by black triangles along the track. Red triangles mark the positions of the key stations Arkona Basin (AB), Bornholm Deep (BD) and Gotland Deep (GD). The map (left lower corner) shows the position of the transect (yellow line) and the hypoxic and euxinic area derived from this cruise data (dark grey and red hatch).*

**2.5 Burial and turnover of C, N, P in the Baltic Sea sediments**

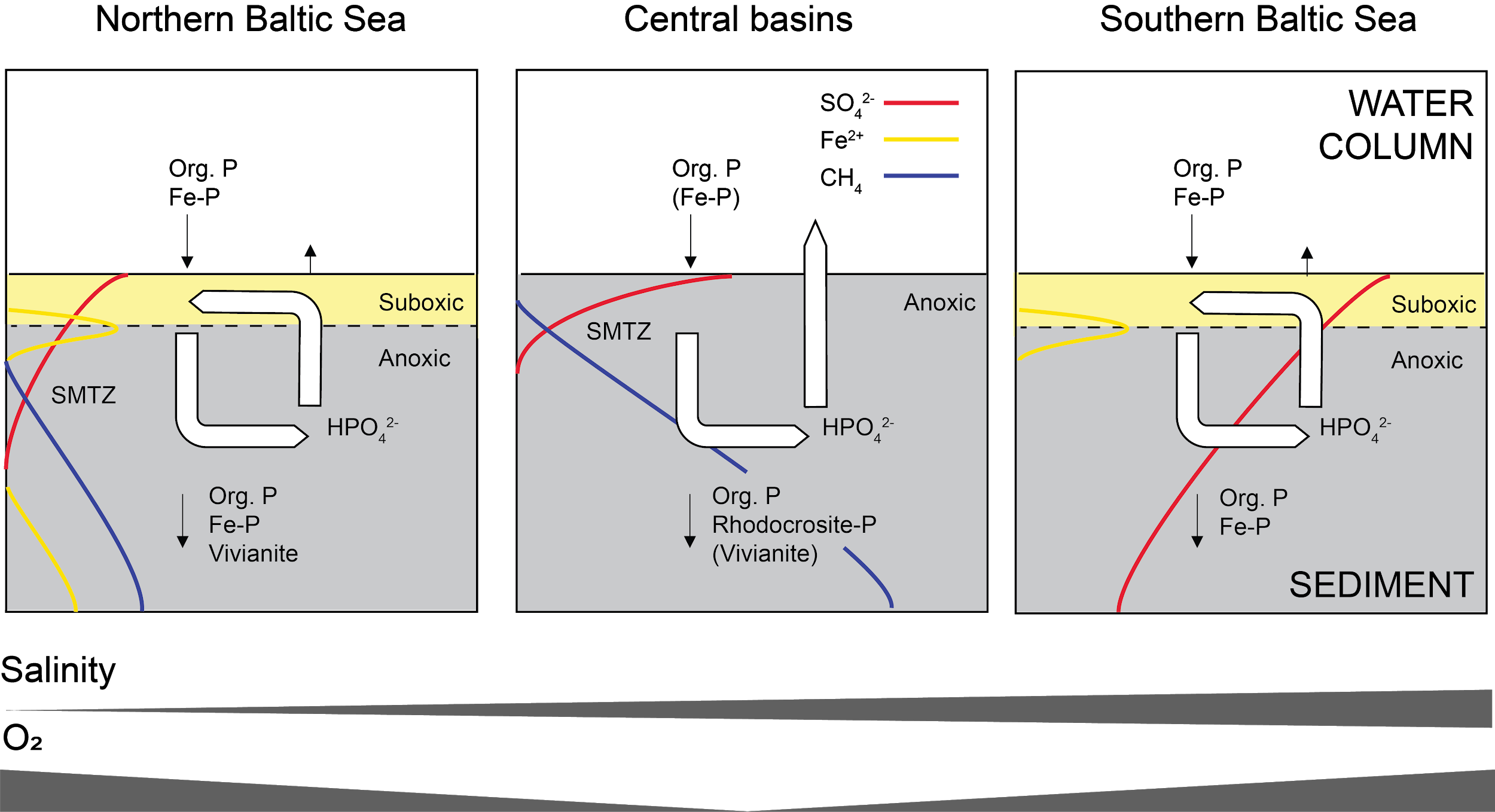
**2.5.1 Spatial variability in deposition of Corg at the sediment surface**

Aphotic seafloor sediments constitute an essential compartment within the marine biogeochemical cycles of C, N and P (Burdige, 2006, Aller, 2014). Settling of reactive organic matter at the base of the water column allows sediments to host a multitude of microbially-mediated and abiotic reactions involving organic matter and the products of its degradation (Berner, 1980, Schulz & Zabel, 2006). The rate of deposition of Corg at the seafloor is a key variable determining rates of sedimentary carbon remineralization, as well as the rate of sedimentary carbon burial. The Baltic Sea today is characterized by a strong N‒S gradient from oligotrophic through mesotrophic to eutrophic conditions (Section 2.4), which exerts a first order control on potential Corg deposition due to vertical settling of autochthonous organic matter (Section 2.5, Tamelander et al., 2017). Deposition of this material is modulated by transformations in the water column, including aggregation and degradation processes, and impacted by transport through currents and seafloor morphology (Leipe et al., 2011). A key process affecting net Corg deposition in the Baltic Sea is wind-wave driven resuspension and lateral transport of fine-grained sediments away from shallower areas (Leipe et al., 2000; Almroth-Rosell et al., 2011). This leads to the accumulation of fine-grained sediments in the central deep basins, with Corg contents of 12‒16 wt%, (Leipe et al., 2011). Terrestrial organic matter (Corg-T) delivered by rivers mainly enters the Baltic Sea in dissolved form (Mattsson et al., 2005; Gustafsson et al., 2014), but undergoes flocculation and settling along the salinity gradient, leading to accumulation in sediments (Asmala et al., 2014; Jilbert et al., 2018). Estimates based on lignin biomarker analyses suggest that 10‒30% of sedimentary Corg in the Baltic Sea is Corg-T, with clear inter-basin differences (Miltner and Emeis, 2001). Erosion of earlier-deposited marine sediments due to glacio-isostatic uplift is important in northern regions (Virtasalo and Kotilainen, 2008) and may constitute an additional source of Corg to modern accumulation areas.

Deposition rates of Corg in the Baltic Sea have been estimated from both sediment traps (e.g., Lehtonen and Andersin, 1998; Gustafsson et al., 2013, Heiskanen and Tallberg, 1999; Struck et al., 2004) and surface sediment data (e.g. Leipe et al., 2011; Winogradow and Pempkowiak, 2014; Nilsson et al., 2019). Of these approaches, the latter may be considered preferable due to the integration of deposition over a longer period (e.g. 2‒10 yr in a sediment slice of 0‒2 cm). An important consideration, as outlined by Nilsson et al. (2019), is that estimates for Corg accumulation based on surface sediment Corg contents and local mass accumulation rates represent deposition rates, rather than ultimate burial rates in sediments. Remineralization of Corg persists throughout the uppermost decimeters of the sediment column, modulating burial. Nilsson et al. (2019) therefore adopted a closed-sum budget approach, based on remineralization rates from DIC effluxes and true burial rates estimated from deeper sediment intervals, to calculate a net rate of Corg deposition of 22.8 ± 7.76 Tg C/yr for the entire Baltic Sea. Deposition is higher in deep accumulation areas than shallower erosion and transport areas, and varies per basin, with highest values observed in the Baltic Proper (Nilsson et al., 2019).

**2.5.2 Cycling of Corg in upper sediment column: controls on primary redox reactions**

Oxygen has the highest energy yield of any electron acceptor (EA) used in Corg remineralization, hence spatial and temporal variability in bottom water oxygen supply is a key factor determining the vertical zonation of diagenetic reactions in Baltic Sea sediments (e.g., Lehtoranta et al., 2009). In regions of oxic bottom waters, diffusive penetration of oxygen into muddy sediments is typically in the order 1‒5 mm (Hietanen and Kuparinen, 2008; Bonaglia et al., 2013). Below this depth, sediments are characterized by a light-colored *suboxic* zone in which reduction of nitrate (i.e. *denitrification*), and Mn and Fe oxides mostly dominate (Fig. 7). When these EAs are exhausted, sulfate reduction becomes the dominant remineralization pathway and the sediment acquires a darker color due to the associated formation of iron sulfides. In regions where Corg deposition is sufficiently high, sulfate too may be completely exhausted within the uppermost decimeters of the sediment column, leaving methanogenesis as the only remaining remineralization pathway. In persistently anoxic areas of the deep basins, the entire oxic and suboxic zone is usually absent from the sediment column (Fig 7) and remineralization is dominated by sulfate reduction and methanogenesis. Transitional states between the end-member examples in Fig. 2.6.1 exist around the margins of the deep basins, and may be observed transiently following major Baltic Inflows and the associated re-oxygenation of deep waters (Dellwig et al., 2018; Hermans et al., 2019). The dissolved reduced products of redox reactions in the sediment column, including methane, ammonium and divalent metal cations, are able to diffuse vertically and can consume EAs through secondary redox reactions, further influencing the diagenetic zonation.



*Figure 7: Schematic of vertical zonation of sediments in the Baltic Sea from a biogeochemical perspective (partially redrawn from Lehtoranta et al., 2009). Colored lines indicate pore water concentrations of key species (zero to left). In areas with oxic bottom waters, remineralization of OM in the suboxic zone proceeds by denitrification and metal oxide reduction. Remineralization proceeds by sulfate reduction and methanogenesis in the anoxic zone, above and below the sulfate-methane transition (SMTZ), respectively. Processes impacting on regeneration and burial of P are shown with annotated arrows. Remineralization produces orthophosphate (HPO42-),which may diffuse to the bottom waters, become trapped in surface-sediment Fe oxides, or incorporated into authigenic minerals, depending on the depositional environment (see text). Note that exceptions to these schematic representations can occur (e.g. a shallow SMTZ was observed in the Gdansk Basin, Southern Baltic, due to high rates of Corg deposition and sulfate reduction, Brodecka et al., (2013)*).

In addition to determining the zonation of primary redox reactions, oxygen availability also directly and indirectly influences the overall rates of microbial Corg remineralization. Organic matter degradation through sulfate reduction and methanogenesis have slow reaction kinetics (Moodley et al., 2005) and are typically assumed to proceed at half the rate of the higher-energy yield reactions in diagenetic models of Baltic Sea sediments (Reed et al., 2011; Radtke et al., 2019). Hence, respiration of organic matter proceeds more slowly in the sediments of the deep anoxic basins. Oxygen also strongly controls the distribution of benthic fauna in the Baltic Sea (Carman & Cederwall, 2001; Laine et al., 2003; Gogina et al., 2016). Within oxic areas, benthic fauna may directly ingest and respire Corg (Ehrnsten et al., 2019) as well as modify rates of microbial processes through bioturbation and bioirrigation (e.g., Kristensen et al., 2001). As a rule, the presence of fauna enhances Corg remineralization, for example through enhancing the supply of EAs in the upper sediments (Arndt et al., 2013). However, benthic community composition has an important role in determining the exact mechanisms. In a comparative study, Kristensen et al. (2011) showed that the invasive polychaete *Marenzellaria viridis* enhanced net sediment respiration by stimulation of microbial sulfate reduction in its I- and J-shaped >20 cm deep burrows. In contrast, the native *Nereis diversicolor* enhanced aerobic remineralization through well-flushed U-shaped burrows in the shallow sediments.

The North-South salinity gradient of the Baltic Sea exerts an important secondary control on the availability of two key EAs, namely sulfate and Fe oxides. The low salinity of the Bothnian Bay and Bothnian Sea leads to bottom water sulfate concentrations < 5 mmol/L, limiting the potential reservoir for microbial sulfate reduction. Under present-day conditions of relatively high Corg deposition, sulfate penetration in the sediment column of the Bothinan Sea is < 10 cm (Slomp et al., 2013, Egger et al., 2015a). Consequently, the sulfate-methane transition (SMTZ) and underlying methanogenic zone are encountered at a relatively shallow depth in the sediment column (Fig. 7), increasing the likelihood that a significant fraction of accumulated Corg will degrade via methanogenesis. Similar porewater profiles have been observed in other areas of the northern Baltic Sea (Sawicka and Bruchert, 2017, Jilbert et al., 2018, Myllykangas et al., 2020a) indicating a key role for methanogenesis in low-salinity, eutrophied regions. In contrast to sulfate, the availability of sedimentary Fe oxides is generally greater in the low salinity northern regions, as a result of high supply rates of Fe from peatland-rich catchments (Sarkkola et al., 2013), limited loss to flocculation due to the weak coastal salinity gradient (Jilbert et al., 2018), and less sulfidization of Fe in surface sediments.

**2.5.3 Quantifying remineralization and burial rates of Corg**

Using DIC fluxes in benthic chamber *in situ* experiments, Nilsson et al. (2019) estimated net sedimentary Corg remineralization of 21.8 ± 7.76 Tg C/yr for the entire Baltic Sea. This value is on average equivalent to 96% of the estimated depositional flux, implying that the vast majority of Corg deposited at the sediment surface is recycled to the water column as DIC. It is also considerably higher than the rate of 1.04 Tg C/yr estimated in an earlier budget (Kuliński and Pempkowiak, 2011), and approximately twice the value simulated in a modeling study by Gustafsson et al. (2017).

Attempts to quantify relative rates of primary redox reactions within the sediment column at a given location have typically been made by diagenetic modeling. Reed et al. (2011) showed that in a muddy sediment location in the Arkona Basin, oxygen accounted for >50% of total Corg remineralization throughout the year, even under hypoxic (<63 µmol/L O2) bottom water conditions during late summer. Reduction of nitrate, Fe oxides and sulfate each accounted for 10‒20%, depending on season, while methanogenesis was insignificant in shallow-sediment diagenesis at this southerly location. Conversely, a similar model estimated that up to 40% of total Corg remineralization at a muddy deep site in the Bothnian Sea proceeds via methanogenesis, due to the shallow penetration of porewater sulfate at this location (Rooze et al., 2016). In the deep anoxic basins of the Baltic Proper, remineralization in muddy sediments is dominated by sulfate reduction and methanogenesis. However, as shown by Reed et al. (2016), temporal fluctuations in shelf-to-basin transport of Fe oxides (cf. Lenz et al., 2015) or reoxygenation events associated with MBIs (Matthaus and Franck, 1992) may lead to a transient role for Fe oxides in Corg remineralization in the deep basins. Until recently, estimates of process rates in permeable sandy sediments have been lacking. However, new results from the south-western Baltic Sea region show that both oxygen uptake and sulfate reduction rates are similar in sandy sediments compared to nearby muds (Lipka et al., 2018). Similarly, Bartl et al. (2019) detected comparable rates of denitrification between permeable and non-permeable sediments offshore from the Vistula estuary. Extrapolation of location-specific process rate estimates to the basin-scale remains fraught with difficulty, but advances in high-resolution sediment typology (e.g. Tauber 2014) and coupled water column‒sediment biogeochemical modeling (Radke et al. 2019) offer high potential for improvement in this area.

Nilsson et al. (2019) estimate a modern Corg burial rate of 0.98 + 0.31 Tg C/yr for the entire Baltic Sea, based on Corg contents below the zone of upper-sediment diagenesis and estimated mass accumulation rates from 30 locations. The value corresponds well with the 0.91 Tg C/yr simulated in the modeling study of Gustafsson et al. (2017). Both approaches effectively consider burial to occur only in deep basin accumulation areas, for example as defined by Håkanson and Jansson (1983) and Carman and Cederwall (2001). However, high 137Cs-based sediment mass accumulation rates have also been observed in many coastal locations traditionally considered to be erosion and transport areas (Mattila et al., 2006). Using these data and sediment Corg contents, Leipe et al. (2011) showed that significant rates of Corg burial may occur in shallow settings. Therefore, it is likely that the true rate of carbon burial in the Baltic Sea as a whole is greater than the estimate of Nilsson et al. (2019). Future studies should try to identify locations (“depocenters”) with high sediment accumulation rates within the Baltic erosion and transport bottom areas to get an improved comprehensive estimate of the integrated Corg burial rate.

**2.5.4 Specific aspects of sedimentary nitrogen and phosphorus cycling**

Organic nitrogen and phosphorus are deposited, remineralized and buried in sediments together with Corg, hence several aspects of the sedimentary cycling of these elements follow what has already been described for carbon. However, N and P cycling is also impacted by multiple further reactions in sediments, which influence their ultimate fate. Sedimentary nitrogen cycling is most dynamic in areas of oxic bottom waters overlying organic rich sediments (Carstensen et al., 2014). Under these conditions, ammonium released during remineralization may be nitrified, facilitating nitrate reduction processes in the upper sediment column (principally denitrification, leading to loss of fixed N as N2 gas; and dissimilatory nitrate reduction to ammonium *DNRA,* which retains fixed N). Denitrification dominates in Baltic Sea sediments (Hietanen and Kuparinen, 2008; Jäntti et al., 2011; van Helmond et al. 2020) although DNRA may be more active under certain conditions (Jäntti and Hietanen, 2012; Bonaglia et al., 2017). Anaerobic ammonium oxidation (*anammox*), leading to loss of fixed N as N2 gas, has been found to be insignificant in Baltic Proper sediments (Hylén et al., unpublished results). Microbial denitrification rates have been shown experimentally to be accelerated by the presence of bioturbating meiofauna, especially of the phylum *Nemotoda* (Bonaglia et al., 2014). In the deep anoxic basins, ammonium released during remineralization diffuses out of the sediments and accumulates in the sub-halocline water mass, from where it may be oxidized upon entrainment into the halocline (Dalsgaard et al., 2013) or during inflow events (e.g., Myllykangas et al., 2017).

Similarly to ammonium, orthophosphate released during remineralization is most likely to participate in complex further cycling in sediments underlying oxic bottom waters. The association of P with sedimentary Fe oxides in the Baltic Sea is well established (e.g. Lehtoranta et al., 2009; Mort et al. 2010). Phosphorus is adsorbed or co-precipitated with oxide minerals when these are present in surface sediments, limiting the P efflux to bottom waters (Fig. 7). Conversely, when these oxides are reduced, the P efflux increases. Seasonal and multiannual variations in the size of the hypoxic area in the central Baltic therefore induce periods of net release or retention of P from sediments around the margins of the deep basins (Conley et al. 2002; Reed et al., 2011). Shelf-to-basin shuttling of oxide-bound P also generates a maximum in P efflux as this material crosses the hypoxic transition zone at the halocline (Almroth-Rosell et al., 2015). High P effluxes in this zone could be related to transient release from sulfur-oxidizing bacteria such as *Beggiatoa*, which form large mats at the sediment surface and have been shown to accumulate P in the form of polyphosphates in other marine systems (Noffke et al., 2016). In the deep anoxic basins, orthophosphate diffuses freely across the sediment-water interface in the absence of surface sediment Fe oxides (Mort et al., 2010; Jilbert et al., 2011; Viktorsson et al., 2012; Viktorsson et al., 2013). Sediment Corg/Porg ratios show that the relative rate of P regeneration from organic matter under anoxic conditions is elevated with respect to C (Jilbert et al., 2011), as confirmed by creating sub-Redfield C/P ratios in the efflux at the sediment-water interface (Viktorsson et al., 2012; Viktorsson et al., 2013). This phenomenon is related to the low P demand of the sediment microbial community (Steenbergh et al., 2011).

Despite efficient remineralization of P in the sediment column, the efflux of orthophosphate across the sediment water interface in the Baltic Sea is modulated by the formation of authigenic P-bearing minerals, which enhance solid-phase P burial rates. Three principal groups of minerals have been identified in Baltic Sea sediments: Ca-phosphates such as carbonate fluorapatite (CFA), Mn-Ca carbonates such as rhodocrosite (Jilbert and Slomp, 2013); and Fe (II) phosphates such as vivianite (Egger et al., 2015b; Dijkstra et al., 2016). Of these, CFA formation is relatively unimportant in the Baltic in comparison with open ocean margin sediments (Mort et al., 2010), while the others are characteristic of the Baltic Sea and may constitute important burial phases of P in certain locations. Both rhodocrosite-bound P and Fe (II) phosphates have been shown to accumulate in deep basin sediments due to shelf-to basin shuttling of precursor Fe and Mn oxide minerals (Jilbert and Slomp, 2013; Reed et al., 2016; Dijkstra et al., 2016). Fe (II) phosphates are also observed below the depth of sulfate penetration in northern regions such as the Bothnian Sea (Egger et al., 2015b), where reduction of deep-buried Fe oxides in the absence of hydrogen sulfide leads to high porewater Fe concentrations and supersaturation with respect to vivianite (Fig. 7). A modeling study by Lenstra et al. (2018) showed that fluctuations in inputs of Fe oxides, as well as salinity, regulate the importance of vivianite as a P burial sink in Bothnian Sea sediments. The study showed that vivianite may account for over 50% of total P burial in low-salinity, high Fe environments such as estuaries.

**2.6 Changes in the marine CO2 system and Ocean Acidification**

**2.6.1 The role of the inorganic carbon cycle in Baltic Sea ecosystem research**

Carbon is the main component of organic matter and thus, primary production and mineralization inevitably are connected to fixation or liberation of carbon. Other than for nutrients like nitrate or phosphate, the bioavailable pool for carbon in seawater as well as brackish water (e.g. the Baltic Sea), due to its inorganic carbon system, is large and usually not limiting. Carbon removal from the upper layer of the sea is thus a direct measure of net production and a quantitative measure of eutrophication (=> 2.3). As the bulk of oxygen production during photosynthesis and oxygen consumption during mineralization of organic matter is a consequence of reduction and oxidation of carbon, carbon transformation processes are the main driver of oxygen depletion (=> 2.4). As the main component of the acid-base system of seawater, the inorganic carbon system is the main buffering system with respect to an increase of atmospheric CO2 penetrating into the marine system, leading to a decrease in pH, a process referred to as ocean acidification (OA). The imbalance of the fugacities of CO2 in the atmosphere and at the sea surface determines the flux of CO2 at the air-sea interface. Investigations of the carbon cycle thus allow insights into some of the processes of main interest for today’s chemical oceanography and ecosystem research in the Baltic Sea, including the function as source/sink for atmospheric carbon, ocean acidification, and the coupling of ecosystem production and oxygen consumption

**2.6.2 Main components of the inorganic carbon system**

While the acid-base system controlling seawater pH is regulated by a number of substances which can act as a proton donor or acceptor (Dickson et al., 2007), the dominating species are bound to the inorganic carbonate system. The inorganic carbon system is comprised of the different forms of carbonic acid and its dissociation products in seawater, including CO2 solvated in water (including a very small fraction of undissociated carbonic acid) referred to as CO2\*, and the two deprotonated forms bicarbonate (HCO3−) and carbonate (CO32−). These are connected by two equilibrium constants (e.g. Millero et al., 2010). The conventional expression of seawater equilibrium reactions in terms of concentrations – instead of activities – results in a dependency of the equilibrium constants on salinity, in addition to temperature and pressure, because the equilibrium constants implicitly include the salinity-dependent activity coefficients. The concentration of CO2\* is related to the fugacity of CO2 (fCO2) by the solubility constant, and the direction of CO2 flux is determined the difference of the fCO2 in surface seawater and in the surface-near air. Lastly, the concentration of CO32− is related to the solubility product of calcium carbonate, either in the form of calcite or aragonite.

*Main variables used to describe the inorganic carbon system*

Analytically, the inorganic CO2 system is determined by four measurable parameters (Dickson et al., 2007); the total concentration of dissolved inorganic carbon (CT), total alkalinity (AT), the partial pressure of carbon dioxide (*p*CO2) and the negative decadic logarithm of protons (pH), where

CT = [CO2\*] + [HCO3−] + [CO32−]

represents the sum of all inorganic carbon forms in seawater;

AT = [HCO3−] + 2 [CO32−] + [B(OH)4−] + [OH−] + [HPO42−] +2 [PO43−] + [SiO(OH)3−] + [NH3] + [HS−] − [H+]F− [HF] − [HSO4−] − [H3PO4] + [minorbases − minoracids]

is defined as the excess of proton acceptors over donors, which is dominated by the carbonate and bicarbonate species, and can be seen as a measure for the buffer capacity of the water;

pCO2 , the partial pressure of CO2, is very closely related to the fugacity (e.g. Pfeill, 2013), and determines the flux direction of CO2 at the sea-air interface;

and pH is the negative decadic logarithm of the H+ activity, and thus the variable most closely related to the seawater “acidity”.

Due to thermodynamic equilibria, it is usually (open ocean) sufficient to determine two of the four variables (and pressure, temperature and salinity) to fully describe the inorganic CO2 system. Similarly, in the open ocean, OA is predictable (e.g. Doney et al., 2009), where only the rise in atmospheric concentrations and change in surface temperature have been considered. Neither of this holds true for the Baltic Sea due to peculiarities in the acid-base and carbon system.

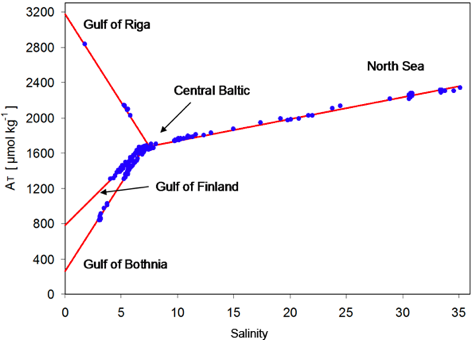
**2.6.3 Status of carbon system measurements in the Baltic Sea**

To date, there exists no harmonized long-term effort to monitor the inorganic carbon system in the Baltic Sea. Yet, apart from campaign and research project based data, some long-term data series exist. AT has been measured continuously as part of the Swedish monitoring program and temporarily by other countries, allowing robust trend analysis since the mid 1990s (Müller et al., 2016). pH has been measured mostly as a side parameter to the biological monitoring, mostly based on glass electrodes with some restrictions on a system with variable salinity. Yet, some long-term trends could be depicted from these data (Carstensen and Duarte, 2019). Recent technological and chemical breakthrough will allow the use of traceable precise spectrophotometric pH measurements in the Baltic (Müller et al., 2018ab; Müller and Rehder 2018). pCO2 has been measured continuously aboard commercial ships between Helsinki and Lübeck since 2003 (Schneider and Müller 2018), and have been used to extensively to describe and quantify productivity patterns in the Baltic Sea (REFs), and to constrain biogeochemical models for the Baltic Sea (REFS). CT measurements, though the most direct variable with respect to the transport of carbon, have so far only been continuously monitored on one station in the Central Baltic Sea since 2003, and used to quantify carbon mineralization in the deep basin during stagnation periods (Schneider and Otto, 2019).

**2.6.4 Peculiarities of the inorganic carbon system in the Baltic Sea**

**2.6.4.1 Alkalinity-salinity relations and main different end members**

The alkalinity of the Baltic sea is governed by a complex interplay of different waters entering the Baltic Sea, unusual contributions to alkalinity complicating carbon system calculations, and long-term trends. The surface alkalinity can be best described by considering the Central Baltic Sea as mixing bowl with a salinity around 6-7 and receiving water with higher salinity and alkalinity from the North Sea, freshwater surplus with lower alkalinity from the North and East (i.e. Gulf of Bothnia and Finnland), and freshwater sources with higher salinity from the southern part of the drainage basins through the, Gulf of Riga, or runoff from the Odra or Vistula Rivers (Fig. 8) (Beldowski et 2010; Kuliński et al., 2014&2017, Hjalmarsson et al., 2008, Stokowski et al., 2020). The robustness of AT-S relations in the Baltic is also reflecting a lack of calcifying primary producers in the Baltic Sea east of the Kattegat (Tyrell et al., 2009), mostly suppressing a change of AT during primary production and oxic mineralization. The range of AT in the Baltic from < 700 mmol/kg to > 3000 mmol/kg leads to a large range of pH under equilibrium with the atmosphere (Omstedt et al., 2010, Kulinski et al., 2017, Gustafsson and Gustafsson 2020).



*Figure 8: Different AT vs. S regimes observed in the Baltic Sea (modified after Beldowski et al., 2010).*

**2.6.4.2 Impacts of organic alkalinity and boron anomalies on carbon system calculations**

The impact of organic matter, which is far more abundant in Baltic Sea waters than in the open ocean, has been shown to have a non-negligible impact on the internal AT distribution through changing the contribution of different components to AT, despite not directly changing AT itself (Kulinski et al., 2014, Ulfsbo et al., 2015,). This leads to considerable differences (up to 50 mmol/kg; Hammer et al., 2017) between measured AT and AT derived from CT and either pCO2 or pH, which can lead to significant errors when using AT in carbon system calculations, in particular in the classical combination with measured CT. Kuliński et al., (2018) extended earlier data on total boron (TB), derived a Baltic specific TB-S relationship and inferred small errors when calculating pH or pCO2 from AT and CT, contributing to the uncertainty in the carbon system determination.

**2.6.4.3 Trends in alkalinity over the last decades and potential implications on acidification**

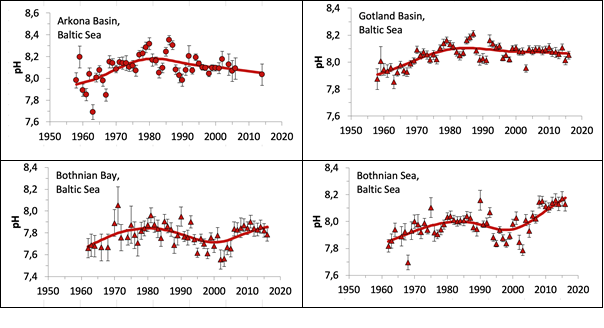
The Baltic Sea receives alkalinity from various sources, including the exchange with the North Sea, riverine alkalinity inputs, and internal alkalinity generation due to organic matter cycling, including anaerobic diagenetic processes in the sediments (e.g. Gustafsson et al., 2014 & 2019ab). Indications of increasing riverine alkalinity inputs (Hjalmarsson et al., 2008), and an increase of AT in the central Gotland Sea over the last century (Schneider et al., 2015) were confirmed by a statistical analysis of all available surface alkalinity data (< 20m water depth) until 2015 (Müller et al., 2016). The data suggest an ubiquitous increase in the AT/S-relations in the Baltic Sea between 1995 and 2015, with an increase of 3.4 mmol/kg yr in the central Gotland Basin, and up to 7.4 mmol/kg yr in the Bothnian Bay at a salinity of 3, corresponding to an increase of 70 mmol/ kg (~5%) or 140 mmol/ kg (~20%) in the two basins over two decades, respectively.

The reasons for this increase in alkalinity are not completely clarified. Müller et al. 2016 suggested a major contribution of weathering-induced external (riverine) input. Sun et al., 2017 support this interpretation by reporting 10-20% increased weathering rates in the pristine northern drainage basin based on a 40 year record of riverine water chemistry data. Gustafsson et al. (2019a, 2019b) report on an overall increase in flow-normalized AT loads to the Baltic Sea from Swedish rivers by approximately 21% over the period 1985–2012 (Gustafsson et al., 2019a, 2019b). Gustafsson et al. 2014, based on model-derived mass balance considerations, suggested that about one third of the alkalinity sources to the Baltic Sea should originate from internal sources, of which only a small fraction could be accounted for in the model (mainly denitrification), and suggested sulphate reduction with subsequent pyrite formation and silicate weathering as main underestimated sources of alkalinity. Yet, Gustafsson et al. (2019a) estimate that a maximum of 18% of the unaccounted alkalinity can be generated by pyrite formation, and suggest that both internal and external sources contribute to the unknown alkalinity contribution. The implication of trends in alkalinity on coastal acidification will be discussed below.

For the Northern Basins, the alkalinity of riverine endmembers derived from (a) chemical characterization of riverine waters and (b) extrapolation of AT-S relationships in the basins have been shown to be in excellent agreement (Gustafsson et al., 2014 & 2019a). A peculiar process leading to a considerable loss of alkalinity has been recently reported for the Odra estuary (Stokowski et al., 2020). Highly alkaline riverine waters during the productive period reach extreme carbonate oversaturation as a consequence to high biological productivity in the Szczecin Lagoon, which triggers inorganic carbonate precipitation. The authors calculated that the process led to a reduction of the alkalinity exported to the Baltic Sea of > 800 mmol/kg (~30%) during the spring productive phase in the lagoon. This recent work highlights the importance of the Baltic Sea with its carbon system peculiarities as a natural lab for coastal processes.

**2.6.5 pH-trends in the Baltic Sea on decadal timescales**

Ocean acidification (OA), i.e. the decrease of pH in seawater as a consequence of rising atmospheric CO2 levels, also referred to as “the other CO2 problem” is one of the major concerns related to ocean chemistry and its interplay with ecosystem functioning. In the open ocean, OA is predictable (e.g. Doney et al., 2009), where only the rise in atmospheric concentrations and change in surface temperature have to be considered. Trends in pH (and other carbon system parameters) in coastal areas are way more dynamic and less predictable, due to possible changes in the catchment area, including precipitation, weathering, liming, nutrient and organic matter supply (eutrophication), and timing and magnitude of production/respiration patterns occurring on similar time scales (e.g. Carstensen and Duarte, 2019). This is particularly true for stratified water bodies like the Baltic Sea, where production and respiration can be vertically separated, leading to different and potentially contrasting trends in surface and deep waters. Though pH measurements have been performed as part of the Swedish, Finnish and Danish monitoring programs since decades, measurement methodology and frequency would impede detecting the “climatologic” acidification signal caused by increasing atmospheric CO2 levels (Almén et al., 2017, Carstensen and Duarte, 2019), and new methods allowing high precision pH measurements with the required high frequency have only recently become available (Müller et al., 2018, Müller and Rehder, 2018). Yet, clear pH trends on decadal scales have been reported for some of the Danish coastal systems (Carstensen et al., 2018), and for the surface waters of most of the major Baltic Sea basins. The weakly buffered (low alkalinity) Northern Basins are amongst the coastal regions with highest seasonal and interannual pH variability (Carstensen and Duarte 2019; see Fig9). A common feature in the surface waters of all basins is an increase in pH until the early to mid 80th as a consequence of eutrophication, which in the central and western basins is followed by a decrease in pH, an expected trend due to the combined effects of oligotrophication and rising atmospheric CO2 levels. In the Bothnian Bay and Bothnian Sea, however, further increase in pH since the late 90th coincidences with the reported strong increase in alkalinity (Müller et al., 2016; see above). In a recent sensitivity study, Gustafsson and Gustafsson (2020) used the BALTSEM model to address the isolated effect of different drivers on pH in the Baltic Sea basins, including atmospheric CO2 and temperature increase, effects of eutro-and oligotrophication, as well changes in runoff and weathering (i.e. alkalinity input). They conclude that on the long run, the increase in atmospheric CO2 is likely to dominate surface pH trends. An important observation is that although summertime and annual mean surface water pH are enhanced in a eutrophied Baltic Sea with increasing hypoxia, the wintertime pH – and with this the annual pH minimum – is lower than under lower nutrient loads and better oxygen conditions, as e.g. under the BSAP scenario. It has to be emphasized that in a scenario of climate warming and atm. CO2 increase under effective restoration (i.e. oligotrophication) measures, various pH-reducing drivers work in the same direction, with natural weathering potentially counteracting. Despite large seasonal amplitude in pH and huge differences in the mean annual pH between the different basins due to the large alkalinity gradients, an observational strategy to allow determination of pH trends, including the reaction to ecosystem management actions, is an important task for the future.



*Figure 9: Trends in (recalculated) pHT for four major Baltic Sea Basins, adopted from Carstensen and Duarte, 2019, Suppl. Information.*

**2.7 Role of specific microorganisms in Baltic Sea biogeochemistry**

Prokaryotes (Archaea, Bacteria) have a pivotal role for the majority of biogeochemical processes in all ecosystems and similar principles as in other aquatic systems are also relevant for the Baltic Sea. However, there are some Baltic Sea-specific features which deviate from many other coastal and marginal seas and that have to be considered for understanding the link between microbial diversity and the biogeochemistry of the Baltic Sea. The most important ones are (1) the reduced circulation in the deeper basins and the resulting low-oxic to sulfidic conditions, (2) the salinity gradient from full marine to nearly limnic conditions and a large brackish area in the Baltic proper, and (3) constant inflow-borne supply of freshwater and terrestrial bacteria together with terrestrial organic matter.

**2.7.1 New insights into Baltic Sea microbiomes**

The application of molecular techniques to study natural microbial communities has resulted in major new insights into diversity, biogeography and functionality of Baltic Sea microbial communities within the last decade. High-throughput sequencing, using the 16S rRNA gene, provided the first comprehensive picture of bacterioplankton diversity along the entire salinity gradient (Herlemann et al. 2011). This approach also revealed, among others, strong seasonal changes in bacterial composition in surface waters (Andersson et al. 2009) and pronounced differences between free-living and particle-associated microbial assemblages (Rieck et al 2016). Single-cell genomics of sediment microbes was pioneered in the Baltic Sea and has given insight into the metabolic potential and function of uncultured Archaea (Lloyd et al. 2013) and Bacteria (Marshall et al., 2017), e.g., in protein degradation in the seafloor, but was also limited by the incompleteness of the single amplified genomes (SAGs).

Metagenomics illuminates the functional potential of complex microbial communities and has been applied in the Baltic Sea, for example, to reconstruct metabolic pathways and functional adaptations along an oxic-anoxic transition (Thureborn et al. 2013) and along the salinity gradient (Dupont et al. 2014), and to assess the response to environmental perturbations (Markussen et al. 2018). Metagenomic analyses were also applied to examine the functional potential of specific bacterial groups, revealing their ecological niches in the Baltic Sea (Hugerth et al. 2015). A Baltic Sea Reference Metagenome (BARM) is available for the taxonomic and functional annotation of genes (Alneberg et al. 2018). Nearly complete genomes from metagenomes (MAGS) enabled the physiological analysis of newly discovered prokaryotic groups such as Lokiarchaeota from Baltic Sea sediments (Cáceres et al. 2020), and allowed to predict ecological niches for dominant planktonic prokaryotes (Alneberg et al. 2020). Metatranscriptomics is a powerful approach to assess the complete set of expressed functions of communities; thereby indicating locations for specific transformations or microbial responses to external factors. It has been applied, for example, to examine the response to small-scale mixing in the oxic-anoxic transition zone (Beier et al. 2019), and to examine the physiology and performance of cyanobacterial isolates (Teikari et al. 2018).

**2.7.2 Microbially-mediated transformations in oxygen-deficient waters**

The strong stratification in the deep basins of the Baltic Sea, due to a permanent halocline, results in oxygen depletion and euxinic conditions in the bottom waters, only interrupted by major Baltic inflow (MBI) events which cause ventilation and extensive deepwater renewal (last ones in 1993, 2003, 2014) (Mohrholz et al. 2015). The oxic-anoxic transitions zones (pelagic redoxclines) are sites of important transformations within the C-, N-, S-cycles and have been well- studied with regard to the prokaryotic key players. Particularly the two deepest basins, the Landsort and the Gotland Deeps, served as model systems for the study of microbial communities in oxygen-deficient water columns. Overall, there is high similarity in microbial composition to other marine euxinic systems, but also to oceanic oxygen minimum zones (Wright et al. 2012, Jürgens and Taylor 2018).

It seems that relatively few taxa control the major inorganic biogeochemical transformations at these pelagic redoxclines. Epsilonproteobacteria of the genus *Sulfurimonas* dominate at the oxic-anoxic interface and in the upper sulfidic zone, are major contributors to chemoautotrophic production (Jost et al. 2008) and use nitrate to oxidize reduced sulfur compounds (Grote et al. 2012, Bruckner et al. 2013). This process of chemoautotrophic denitrification in the central basins can be considered as a major N loss process for the Baltic Sea, being equivalent to sedimentary denitrification (Dalsgaard et al. 2013). Depending on the actual sulfide concentration, an important side product of this denitrification process is nitrous oxide (N2O) (Dalsgaard et al. 2013). There is a close coupling of nitrification and denitrification around the oxic-anoxic interface. Interestingly, the process of ammonia oxidation is entirely dominated by Archaea (Thaumarchaeota related to the genus *Nitrosopumilus*) (Labrenz et al. 2010), probably partly due to their adaptation to cope with frequent exposure to sulfidic conditions (Berg et al. 2015). Ammonia oxidizing bacteria are insignificant here but seem to play a more important role in coastal waters with high nutrient loading (Happel et al. 2018). Nitrite oxidation is conducted by the globally distributed marine nitrite oxidizer phylum Nitrospinae (Beier et al. 2019). Other important chemoautotrophic sulfur oxidizers in the redoxcline are Gammaproteobacteria of the SUP05 clade (Glaubitz et al. 2013), a group also known from oceanic oxygen minimum zones (Wright et al. 2012).

Anammox does not seem to be an important process in the redoxcline (Hietanen et al. 2012), which is due to the fact that, unlike in the Black Sea, an extended anoxic and sulfide-free zone, the typical habitat for anammox bacteria, is missing in the Baltic Sea. Instead, the proximity of the nitrate and sulfide profiles favour chemoautotrophic denitrification. An interesting exception constitutes the situation after MBIs and the re-establishment of anoxia in bottom waters. As observed after the 2003 inflow event (Hannig et al. 2007), total (reduced and oxidized) manganese concentrations in a compressed anoxic zone were considerably increased and the enhanced downward flux of oxidized, particulate Mn was probably used for bacterial H2S oxidation, thus creating an anoxic sulfide-free zone, suitable for anammox (Hannig et al. 2007). After the MBI in 2003, the occurrence of anammox bacteria (*Candidatus* Scalindua) and significant anammox rates created such a “Black Sea-like” situation for the Gotland basin (Hannig et al. 2007). Bacteria of the genus *Sulfurimonas* probably also play an important role for manganese oxide-driven sulfide oxidation, as shown for the Black Sea (Henkel et al. 2019). Inflow events also result in the temporal displacement of water masses with anoxic microbial communities (and the processes triggered by them) to shallower water depths (Bergen et al. 2018).

In contrast to the inorganic transformations, the decomposition of organic material in the deeper anoxic basins, where also DOM composition differs from the oxic layers (Seidel et al. 2017) has been much less studied. Sulfate-reducing bacteria (mainly Deltaproteobacteria) become a dominating group here (Herlemann et al. 2011), and sulfate reduction is probably a major organic matter decomposition process. However, neither for this nor for other anaerobic metabolic pathways (e.g. different fermentations), field data from the central Baltic Sea exist.

**2.7.3 Pelagic microbial communities within the salinity gradient**

Meanwhile we have a fairly good picture of the salinity-related Baltic Sea bacterial biogeography (Herlemann et al. 2011, Dupont et al. 2014). At a broad phylogenetic level, the relative abundance of Gamma- and Alphaproteobacteria increases with salinity, whereas an opposite trend is exhibited by Actinobacteria and Betaproteobacteria. Further, Verrucomicrobia, mainly represented by one taxon affiliated with the Spartobacteriaceae, dominate large areas of mesohaline waters in the central Baltic Sea (Herlemann et al 2011). The reconstruction of the genome of this taxon by metagenomics revealed many genes potentially involved in processing of polysaccharides, which are produced by phytoplankton, especially cyanobacteria (Herlemann et al. 2013). Shifts in bacterial composition with declining salinity also occur on a finer phylogenetic level, e.g., within the globally dominating marine clade SAR11 (Herlemann et al. 2014). A challenging question is whether the strong shift in bacterial composition along the salinity gradient also involves functional changes, e.g., in the decomposition efficiency of different compounds. Some evidence for this was derived from mesocosm experiments in which bacterial communities from the Northern Bothnian Sea seemed to be better adapted to utilize river-born terrestrial DOC (tDOC) (Herlemann et al. 2017), and where DOM degradation differed between different communities (Logue et al. 2016). This is consistent with substantial remineralization rates of tDOC and CO2 supersaturation measured for this area (Fransner et al. 2019). Overall, freshwater bacteria can successfully migrate into the brackish Baltic Sea, where they might gain a selective advantage when riverine DOC is the main carbon source (Kisand et al. 2005, Riemann et al. 2008).

**2.7.4 Role of benthic microbial communities**

Like any sediment microbial community, also Baltic Sea communities exhibit a strong vertical stratification driven by organic carbon availability, redox zonation (Edlund et al. 2008), and environmental filtering (Starnawski et al. 2017; Marshall et al. 2019), where deep sediment communities assemble close to the sediment surface (Petro et al. 2019), at the bottom of the bioturbation zone (Chen et al. 2017). In the Western Baltic Sea, sulfate-reducing microorganisms (SRM) peak in abundance and richness just below the bioturbation zone, and their numbers strongly decline after sulfate is depleted in the sulfate-methane transition zone (SMTZ) (Marshall et al. 2019). When oxygen is absent from the bottom waters, the SRM peak is shifted up to the sediment surface, and also the overall community at the sediment surface differs between sites with permanently oxic (bioturbated), permanently anoxic (not bioturbated), and seasonally hypoxic bottom water. Microbial diversity is highest at oxic and lowest at anoxic sites (Broman et al. 2017, Sinkko et al. 2019), and oxygen availability thus largely controls, via bioturbation, benthic microbial community structure (Deng et al. 2020). The depth of the SMTZ on the other hand is controlled by the availability of organic matter (and thus sedimentation rate) and the concentrations of sulfate and other electron acceptors (and thus salinity and bottom water conditions). This implies that SRM should peak closer to the surface and decline faster with depth in the oligohaline, hypoxic areas of the Eastern Baltic Sea, for which, however, only few data are available (Sinkko et al. 2011, Reyes et al. 2016). Methanogenesis and anaerobic methane oxidation (AMO) are highest in the SMTZ, where methanogenic *Methanosarcina* may form syntrophic associations with acetate oxidisers, e.g., *Geobacter* (Rotaru et al. 2018) and ANME-1 Archaea may conduct both methanogenesis and AMO (Beulig et al. 2019); in other sites, ANME-1 and/or ANME-2 Archaea have been implicated in AMO (Treude et al. 2005, Shubenkova et al. 2010, Myllykangas et al., 2020). For a recent review on biogeochemical processes and microbial life in Baltic Sea sediments, including the deep biosphere, see Jørgensen et al. (2020).

Similar to the water column, salinity exerts an overarching control on benthic bacterial communities, with typical marine and typical freshwater taxa towards the extremes, and a broad overlap at mesohaline conditions (Klier et al. 2018). In coastal sediments, seasonal changes in community composition follow the major inflow of freshwater in spring and phytoplankton sedimentation after the spring bloom (Vetterli et al. 2015, Sinkko et al. 2013).

The important role of chemolithotrophs and metal-cycling microbes seen in the water column also applies to the sediment communities but with distinct taxa: nitrification (at oxic sites) appears dominated by ammonia-oxidizing bacteria and nitrite oxidizers of the genera *Nitrospina*, *Nitrospira*, and *Nitrobacter* (Reyes et al. 2017) and is closely coupled to sulfide-dependant denitrification and DNRA, e.g., by *Beggiatoa* and *Thiothrix* (Reyes et al. 2017, Klier et al. 2018). The role of epsilonproteobacterial *Sulfurimonas* and *Sulfurovum* remains unclear (Broman et al. 2017). In the absence of internal nitrate production, e.g. in permanently or periodically hypoxic basins, nitrate reduction may be fuelled by settling pelagic diatoms after phytoplankton blooms, who link pelagic and benthic N cycling by transporting intracellular nitrate to the oxygen-deficient seafloor (Kamp et al. 2018).

A coupling of N and Fe cycles is implicated by the detection of Fe(II)-oxidizing nitrate reducers (Laufer et al. 2016b), tentatively identified as e.g., *Thiobacillus, Hoeflea, Dechloromonas* in Danish fjord sediments (Laufer et al. 2016a, Otte et al. 2018). Also microaerophilic Fe/Mn oxidizers (*Mariprofundus, Gallionella*) and phototrophic Fe-oxidizers (e.g., *Rhodobacter, Chlorobium*) were found at several sites (Reyes et al. 2016, Otte et al. 2018), and metal-oxidizers were enriched from iron-manganese concretions in the Gulf of Finland (Yli-Hemminki et al 2014). Fe- and Mn-reduction appears to be coupled to organic carbon content (Laufer et al. 2016a), and members of *Arcobacter, Colwellia* and Oceanospirillaceae were implied in Mn reduction at Mn-oxide-rich marine sites of the Baltic Sea (Vandieken et al. 2012). However, identification of metal-cycling key players in Baltic Sea sediment remains often tentative, when known metal reducers (*Geobacter, Shewanella*) are rare, while versatile sulfate (and potentially metal-) reducers (Desulfobulbaceae, Desulfuromonadaceae, and Pelobacteraceae) appear associated with zones of metal reduction (Reyes et al. 2016, Otte et al. 2018).

The permanently or seasonally hypoxic sediments of the Baltic Sea are prone to release of free sulfide. Euxinia can be counteracted by benthic microbial sulfide oxidation as long as a minimum of oxygen (or nitrate) in the bottom water is available. This is seen in the hypoxic transition zone of the Eastern Gotland Basin, which is covered by dense mats of *Beggiatoa* (Noffke et al. 2016), which have been estimated to consume up to 70% of the sulfide flux towards the sediment/water interface (Yücel et al. 2017). *Beggiatoa* are also common at many other sites of the Baltic Sea (Reyes et al. 2017, Klier et al. 2018). Alternatively, filamentous cable bacteria (originally discovered in the Baltic Sea; Pfeffer et al. 2012) can link the oxidation of sulfide in deeper sediment horizons to the reduction of oxygen or nitrate by conducting electrons over centimeter distances. Cable bacteria belong to the deltaproteobacterial family Desulfobulbaceae, with the marine/brackish genus *Ca*. Electrothrix and the freshwater/oligohaline *Ca*. Eletronema (Trojan et al., 2016). They are widespread across all salinities in Baltic Sea sediments (Klier et al. 2018; Otte et al. 2018, Marzocci et al. 2019), show highest densities at seasonally hypoxic sites with high sulfate reduction rates (Hermans et al., 2019), and have been hypothesized to interact with Fe-cycling microbes (Otte et al. 2018). The metabolic activity of cable bacteria does not only remove sulfide but generates a suboxic zone with a pH minimum, which promotes the dissolution of FeS and the formation of Fe and Mn oxides (Risgaard-Petersen et al. 2012). These can act as "firewall" against euxinia long after the bottom waters have become hypoxic (Sejtaj et al. 2015). Hermans et al. (2019) proposed this cable bacterial firewall to explain why bottom waters in the highly eutrophic Gulf of Finland rarely contain sulfide in summer. The niche partitioning between *Beggiatoa* and cable bacteria, and the extent of their respective euxinia protection, are currently unresolved.

**2.7.5 Anticipated future development of microbial communities and activities**

Due to the fact that more comprehensive data on microbial communities have been gathered only in recent years and do not yet cover adequately the whole Baltic Sea, it is currently not possible to anticipate how these communities will respond to predicted future environmental changes of the Baltic Sea. From experimental and field studies it is clear that higher SST and increased river runoff will fuel the activity of heterotrophic microorganisms, result in higher carbon remineralisation and shift pelagic food webs towards the microbial components (Wikner and Andersson 2012). Bacterial communities respond quickly to changes in environmental factors, with the appearance of well-adapted taxa, probably due to the presence of a large seed bank both in the sediment and the water column. This has been experimentally investigated, for example, for shifts in salinity (Shen et al. 2018) and different environmental stressors (Markussen et al. 2018). For benthic communities, an increase in areas with hypoxic or anoxic bottom waters may not only lead to declining microbial diversities at the sediment surface but also to faster depletion of sulfate, an upwards migration of the SMTZ, and higher abundances and activities of methanogens.

**2.8 Interactions between biogeochemical processes and chemical contaminants**

Organic contaminants and toxic metals emitted through human activities have become ubiquitous and unwelcome entities in the environment. Their transport and fate are closely associated with biogeochemical cycles, mainly due to the tendency of many substances to sorb to organic matter (OM) (Nizzetto et al., 2010). For dissociating organic contaminants and metals, environmental factors such as pH and redox conditions influence speciation and sorption to organic matrices and mineral surfaces (Jones and Tiller, 1999; Pohl and Hennings., 1999). The influence of anthropogenic contaminants on biogeochemical processes is less studied, and many knowledge gaps remain.

**2.8.1 Catchment characteristics and biogeochemistry impact contaminant transport to the Baltic Sea**

*Organic contaminants*. Rivers collect contaminants emitted from point sources (e.g. WWTPs) or deposited on land and subsequently transported via runoff. Hydrological conditions are thereby important for contaminant river transport and retention. Contaminated sediment and soil particles are typically mobilized and eroded during high discharge events (Rügner et al., 2019; Schwientek et al., 2013). Contaminants are also released from the snow-pack during spring floods (Josefsson et al., 2016; Meyer et al., 2011) or mobilized from soil when snow-melt displace shallow groundwater (Filipovic et al., 2015). Concentrations of organic contaminants in boreal catchment rivers, and thus transport to the Baltic Sea, are influenced by vertical gradients of OM and contaminants in soil, and season and land-type dependent pathways for water at different depths in the soil (e.g., discharge from deep soil levels during snow-cover and overland flow in areas with frozen top layer during snowmelt). Another important factor that affects this transport pathway is the quality of suspended OM, which influences the sorption capacity for various compounds and helps explain varying retention of atmospherically deposited contaminants observed in forests and mire-landscapes (Bergknut 2010, 2011; Josefsson 2011, 2016).

Stable flow conditions reduce catchment runoff and therefore transport to rivers, and promote sedimentation of contaminants in rivers and lakes: a process that reduces water concentrations. During low flow conditions however, concentrations of contaminants emitted from human activities may increase if emissions occur at a constant rate (Urbaniak et al., 2019). Export of contaminants via rivers is counteracted by degradation processes. Field and flume studies of pharmaceuticals in rivers show that biodegradation at the sediment-water interface efficiently eliminates substances due to the relatively long residence time in sediment pore water and diverse microbial community, and is influenced by e.g., sediment composition, redox conditions and DOC concentrations in the water (Lewandowski et al., 2011; Schaper et al., 2018; Posselt et al., 2020).

Shifts in biological productivity in lakes and marine surface water impact the air-water exchange of many organic contaminants with a mechanism similar to the “biological pump” sequestering CO2 from the atmosphere (Dachs et al., 2002; Taylor et al 1991). Hydrophobic and stable compounds sorb to OM and are transported downwards, thereby potentially depleting contaminant concentrations in the surface water and enhancing diffusive air-water exchange, as has been observed in the field (Berrojalbiz et al., 2011; Galbán-Malagón et al., 2012; Josefsson et al., 2011).

*Metals*. Trace metals in freshwater are carried in dissolved, colloidal, or particulate form, mainly in association with OM, Fe-Mn (oxyhydr)oxides and clay particles. Metal speciation is more important than total concentration with respect to effects on organisms and export. Metals bound to silicate minerals are generally immobile and non-bioavailable (Tuzen 2003), while their ionic species have a high mobility and bioavailability (Sunda and Lewis 1978; Luoma 1983). Bioavailable trace metals in rivers can however rapidly precipitate in estuaries after mixing with fairly small amounts of the alkaline brackish seawater as observed in an estuary in Western Finland, thereby retaining the metals in coastal sediments (Nystrand et al., 2016). Metal speciation and distribution in the water column are affected by complex interactions like adsorption-, precipitation-, desorption- or dissolution processes, among them redox conditions and formation of metal sulfides play a key role during stagnant conditions (Pohl and Hennings., 1999). In sediments, metal cycling is driven by microbial oxidation of OM releasing complexed/sorbed metals or dissolution of Fe-Mn (oxyhydr)oxides upon changing redox conditions. Part of the metal-OM complexes are efficiently recycled in the surface sediments during diagenesis while a considerable fraction is permanently buried as refractory metal-OM complexes or is incorporated into insoluble sulfides, and excluded from biological processes, making sediments a sink for metals (Jokinen et al., 2020). Several processes such as water flows changing redox and oxygen conditions, resuspension of sediment particles, and release from dumped chemical warfare agents (linked to the presence of anaerobic sulfide reducing bacteria) contribute to returning of metals to the available marine trace metal pool (Bełdowski et al., 2016; Cybulska et al., 2020). Metals are also entering Baltic Sea bottom waters via submarine groundwater discharge (SGD) through porous sediments (Szymczycha et al., 2016; Virtasalo et al., 2019). The impact of SGD on the Baltic Sea ecosystem is still not well understood; the importance of this entry route depends on the physicochemical properties of groundwater, sediments and bottom water (Schlüter et al., 2004; Jakobsson et al., 2016; Szymczycha et al., 2016; Krall et al., 2017; Virtasalo et al., 2019).

**2.8.2 Impact of contaminants on biogeochemical processes**

Current research points towards that contaminants together with other stressors such as shifts in nutrient, DOM and oxygen concentrations contribute to changes in biogeochemical processes. Much knowledge is however lacking, partly because the chemical mixture in the Baltic Sea is not well characterized (Sobek et al., 2016; Wang et al., 2020), as illustrated in a modelling study performed on the Kattegatt and the North Sea (Everaert et al., 2015). Exposure to specific PCBs and pesticides measured in the seawater did not limit marine phytoplankton growth, but when compensating for the presence of unknown contaminants in the model, a growth limitation of 10% was estimated for Kattegatt (Everaert et al., 2015), which would be a significant impact on the marine carbon cycle.

*Observed effects on microbial communities in the water column.*

Exposure to contaminants can change the microbial community composition (van der Meer, 2006; Echeveste et al., 2016) which in turn may affect the functionality of the community (Allison and Martiny, 2008; Shade et al., 2012). In one of the very few studies in the Baltic Sea, effects on pelagic bacterial communities of exposure to frequently occurring organic contaminants were less pronounced than the effects observed due to increased terrestrial DOM concentrations. Still, exposure to contaminants contributed to an overall reduction of bacterial activity and diversity, particularly at elevated terrestrial DOM (Rodriguez et al., 2018). In the Mediterranean (Cerro-Galvez et al., 2019), nutrient conditions were found to be drivers of both microbial growth and extracellular enzymatic activities, but similar to the findings in the Baltic Sea, organic contaminants contributed to change of both endpoints. Further, a model bacterium culture relevant for the Baltic Sea (*Rheinheimera* sp. BAL341) exposed to contaminant mixtures had a significant decrease (9-18%) in abundance and production (when in the exponential growth phase) (Karlsson et al., 2019). Genes that responded to contaminant exposure were involved in several distinct cell functions (Karlsson et al., 2019), suggesting that contaminants may influence microbial community composition and function.

*Effects on biogeochemical processes in sediment.*

Field observations from the eastern Gulf of Finland demonstrate effects of metals and oil contamination on sediment microbial activity (Polyak et al., 2017). Experimental research on sediment from the Baltic Sea showed that in sediments exposed to Cd at environmentally relevant concentrations and at varying oxygen conditions including hypoxia, Cd affected microbial denitrification (Broman et al., 2019). In contrast, no effects on nitrification rates or the bacterial ammonia oxidizer gene were found in Baltic Sea sediment contaminated by PAHs and PCBs, while significant effects on the microbenthic community structure were observed (Iburg et al. Submitted).

*Indirect effects due to reduced bioturbation.*

Organic contaminants, such as PCBs and DDT, in sediment can impact sediment reworking and burrowing of benthic invertebrates (Landrum et al., 2004; Mulsow et al., 2002). Contaminants may however indirectly also affect sediment biogeochemical processing, as was recently observed in a river system where PAH contamination led to reduced bioturbator activity of the worm *Tubifex tubifex*, which in turn negatively impacted aerobic respiration and denitrification rates in the sediment (Mermillod-Blondin et al., 2013).

**3. Knowledge gaps and future research needs**

**Ad. Chapter 2.1:**

The future status of the Baltic Sea will be greatly influenced by both socioeconomic and climate drivers. There are considerable uncertainties to the importance of both these drivers and their interactions. These uncertainties arise in particular from how global drivers will play out at the regional level. The current downscaling of such drivers, in particular for socioeconomics, are associated with great uncertainties. There is a need to develop socioeconomic scenarios that align with current policy trends at both regional and global scales and to explore how this meets the global sustainability targets and the regional environmental targets of the Baltic Sea Basin.

The historical nutrient loads to the Baltic Sea prior to the 1970’s are poorly quantified and uncertain. This limits the understanding of the previous biogeochemical and ecological interactions in the Baltic Sea. There is a need to better quantify the amounts and geographical distributions of nutrient loads to provide a reliable baseline the modelling of the ecological status of the Baltic Sea,.

**Ad. Chapter 2.2:**

The biogeochemical processes governing the elemental cycling of C, N and P are generally well known, whereas the environmental and biological factors stimulating certain biogeochemical pathways over others in the complex mosaic of interacting and competing processes are poorly understood. Quantification of the regulatory effects that salinity, temperature, light, oxygen, concentrations of nutrients and trace elements, and biological composition have on process rates are important for calibrating coupled hydrodynamic-biogeochemical models for assessing the role of the coastal filter in the future.

Furthermore, the pool of compounds constituting dissolved organic matter (DOM) is still poorly characterized with measurement techniques available today. Since DOM is an important constituent of the coastal filter, as a key characteristic of coastal ecosystems itself and as a regulatory factor for other processes, it is important to understand how the quantity and quality of DOM changes across the land-sea interface, and if DOM inputs and processing in the coastal zone will change in the future.

**Ad. Chapter 2.3:**

The major biogeochemical transformations of inorganic nitrogen and phosphorus in the Baltic Sea are fairly well understood, in contrast to the dynamics of the organic nutrient pools. Organic compounds make up 81% of the pelagic nitrogen and 30% of the pelagic phosphorus pool (Savchuk, 2018). Organic nitrogen and phosphorus are produced as well as taken up by both phytoplankton and bacteria. The net effect of temperature and salinity changes on organic nutrient dynamics is poorly understood, and will be further complicated by the expected future increases in terrestrial organic matter inputs, in particular in the Northern Baltic Sea.

**Ad. Chapter 2.4:**

Due to the lack of oxygen concentration data, the estimated sizes of hypoxic area for the open sea and coastal zones have considerable errors. The sizes of hypoxic and euxinic areas of available map products calculated from measured oxygen and hydrogen sulfide concentration profiles differ by up to 20% (Meier et al., 2019c).

Better knowledge on sources and sinks of the oxygen budget is needed. In particular, oxygen consumption and water-sediment fluxes of nutrients are not well known.

The dynamics of small and large saltwater inflows is still insufficiently known. In particular, our knowledge about mixing processes and their role for the ventilation of the deep water needs to be improved.

**Ad. Chapter 2.5:**

An important general knowledge gap in the study of sediment biogeochemical processes arises from limited spatial and temporal coverage of sediment data. Remineralization and burial rates may be highly heterogeneous, introducing problems for interpolation from a small number of study locations to the scale of the Baltic Sea or its sub-basins. Improvements in sediment typology (e.g. Tauber 2014) and coupled water column‒sediment biogeochemical modeling (Radke et al. 2019) show the way forward towards more accurate depiction of spatio-temporal variability, but these tools are not yet viable in all locations due to gaps in data and expertise. A particular problem concerns estimates of process rates, and especially carbon and nutrient burial, in shallow shelf areas of the Baltic Sea. These areas have traditionally been considered to be dominated by sediment erosion and hence omitted from estimates of basin-wide remineralization and burial of OM. An increasing number of studies has demonstrated the importance of coastal and shallow areas as potential traps of carbon and nutrients (Section 2.3), implying that a subset of these environments should be considered as accumulation zones and are therefore important for carbon and nutrient cycling. Improved knowledge of the distribution of sediment accumulation in bathymetrically complex shallow areas, as well as the distribution of water column redox conditions (e.g. Virtanen et al., 2019), will aid future study of large-scale process rates. Other areas requiring further study include the role of benthic fauna in carbon and nutrient cycling, in particular turnover rates of elements within the faunal biomass, as well as the long-term effects of bioturbation and bioirrigation on microbial process rates in a system with changing benthic communities. Also, the role of terrestrial organic matter in diagenetic processes in the Baltic Sea is understudied considering the importance of this material to the total sediment carbon pool and its variable rate of input from the surrounding land masses (Section 2.2).

**Ad. Chapter 2.6**

Several important knowledge gaps addressing the carbon system and cycling in the Baltic Sea call for future research actions. The reasons for the observed trends in the alkalinity/salinity relations for the different basins need to be identified, which requires an integrated approach addressing changes in runoff from land (=> 2.1), sedimentary diagenetic processes (2.6), and the water carbon chemistry (=> 2.7). The current inability to close the mass balance for AT, as one of the key biogeochemical parameters and main variable describing the buffer capacity with respect to changes in the acid-base system (i.e. acidification), needs to be overcome. This research need is actually linked to the even larger topic of a consistent carbon inventory for the Baltic Sea. The role of coastal systems in the land-ocean-atmosphere continuum (LOAC) as a transformation system and possible sink for carbon entering the marine system from land is currently one of the hot topics of marine biogeochemistry. Yet, an integrated assessment and mass balance for carbon in the Baltic Sea has not been attempted since the work by Kuliński and Pempkowiak (2011). Without this knowledge, important questions such as the vulnerability to acidification or the current and future role of the Baltic as a source/sink for atmospheric carbon cannot be answered.

Dedicated research is needed to unravel the variations in C/N/P stoichiometry during parts of the spring and the summer bloom, which have been revealed in particular by carbon observations, and have forced several amendments to biogeochemical models. Carbon observations are a unique tool to validate and constrain biogeochemical models. Recent technological advances with respect to the measurement of carbon system parameters even on autonomous platforms, and the recognition that long-term observations of carbon parameters are needed to improve the biogeochemical assessment of the Baltic Sea, calls for a model-guided development of a cost-effective, fit-for purpose long-term observation strategy for carbon system parameters in the Baltic Sea, which is an important scientific and management challenge on its own.

**Ad. Chapter 2.7:**

We still lack a complete overview of prokaryotic taxonomic distribution and functionality in the different subsystems of the Baltic Sea. For example, it is not known whether seasonal coastal hypoxia triggers the development of communities comparable to the oxygen-deficient deep basins. For benthic systems, the data base is even weaker, and it is not clear how the different types of sediment biogeochemistry across the Baltic Sea, outlined in 2.6 (Fig. 2.6.1), are reflected in different prokaryotic communities. More important, it is currently not understood whether the documented shifts in phylogenetic composition of microbial communities along the main environmental gradients of the Baltic Sea have significant implications for some biogeochemical processes, or whether a high functional redundancy buffers these community shifts.

Further, the key players for a couple of important biochemical transformations have not been identified yet, such as for methane production and consumption, and metal cycling (Fe and Mn oxidizers and reducers). For some processes (e.g., sulfur oxidation) where pelagic (*Sulfurimonas*, SUP05) and benthic (*Beggiatoa*, cable bacteria) key players are known, niche differentiation and the specific ecological impacts are not sufficiently clear to incorporate the performance of these specific taxa into biogeochemical models. This knowledge gap is due to the lack of representative isolates that can be studied in lab experiments as well as the lack of in situ methods that can quantify the contribution of specific taxa to a particular biogeochemical process.

**Ad. Chapter 2.8:**

Several knowledge gaps regarding the mechanism and importance of interactions between contaminants and biogeochemical processes remain. One challenge is to predict how conditions in the catchment influence river transport, where more research is needed to understand factors that govern degradation processes in rivers and how differences in sorption of contaminants to different types of particles influence export to the Baltic Sea. Another knowledge gap is how the consequences of climate change, such as shifts of salinity, pH, redox, and oxygen conditions, will affect the speciation and bioavailability of metals. Research regarding the quantitative impact of exposure to environmental contaminant mixtures on biogeochemical processes in the Baltic Sea is still in its early stages. One key question is whether exposure to environmental contaminant mixtures makes environmental systems, such as microbial communities, more vulnerable to other environmental stressors, such as climate change.

**4. Conclusions and key messages**

**Ad. Chapter 2.1**

* Nitrogen and phosphorus loads to the Baltic Sea Basin are driven by both climate change and socio-economic factors. Modelling shows that changes in societal factors generally outweigh effects of changes in the climate.
* Projections of climate change in the Baltic Sea Basin by 2050 shows increases for temperature of about 2.5 °C and with a 10% increase in precipitation. However, the opposing effects of increased rainfall and increased evapotranspiration results in overall little or no change in stream runoff.
* A range of socio-economic scenarios have been developed and explored related to the nutrient loadings of the Baltic Sea. Only very few scenarios that focus on sustainability with less agricultural land use and targeted technologies for reducing emissions from land use and waste water can comply with the BSAP.
* Measures to reduce nutrient emissions have succeeded to reduce inputs significantly during the past decades.

**Ad. Chapter 2.2:**

* Denitrification is the most important removal process for nitrogen with the highest rates observed in lagoons and estuaries receiving high inputs of nitrate and organic matter from land
* Phosphorus is removed through permanent burial, but the burial forms have different stabilities that depend on salinity, availability of iron and oxygen.
* The composition of dissolved organic matter changes drastically during passage of the coastal zone through processes of heterotrophic consumption, photochemical degradation, flocculation and burial.
* Coastal ecosystems around the Baltic Sea are highly diverse in their hydromorphology and physical-chemical conditions as well as the inputs of nutrients and organic matter from land, resulting in a broad span for the coastal filter efficiency, today and in the future.

**Ad. Chapter 2.3:**

* The dynamics of nitrogen and phosphorus in the Baltic Sea are driven by nutrient inputs, modified by biogeochemical processes. At present, denitrification and burial remove about 87% of the annual nitrogen inputs, whereas only 69% of the annual phosphorus load is lost by burial in bottom sediments. About 1% of the annual nitrogen and 4% of the phosphorus load accumulate in water column and sediments; the remainder is exported to the North Sea.
* In addition to a long-term, eutrophication driven increase, pelagic nitrogen and phosphorus pools fluctuate with changing redox conditions. Expanding hypoxia causes the nitrogen pool to decline, whereas the phosphorus concentrations increase.
* Changes in insulation, wind-speed, temperature and stratification, have prolonged the phytoplankton growth season with an earlier spring bloom and a delayed autumn bloom.

**Ad. Chapter 2.4:**

* Saltwater inflows ventilating the deep water with oxygen-rich water show a considerable multi-decadal variability including stagnation periods but no statistically significant trend.
* Oxygen depletion has amplified during the past decades and oxygen consumption in the water column has increased relatively more than oxygen consumption in the sediment.
* In 2019, the size of hypoxic area is with >80,000 km2 one of the three largest on record (Hansson et al., 2019).

**Ad. Chapter 2.5:**

* Carbon, nitrogen and phosphorus are deposited in sediments principally as organic matter. In Baltic Sea sediments, autochthonous OM is the dominant source, with terrestrial OM contributing up to 30%.
* Physical redistribution processes lead to accumulation of fine-grained, OM-rich sediments in deeper areas. Shallow areas are typically characterized by erosion and transport, although bathymetry may favor local accumulation in these areas too.
* Remineralization in the upper sediments returns >90% of deposited organic carbon to the water column as DIC. The remaining material is buried. Nitrogen and P are also released from OM during remineralization, but their ultimate fate depends on redox conditions at the sediment-water interface.
* Both the N-S salinity gradient of the Baltic Sea, and the redox gradient between shallow and deep areas, influence relative and absolute rates of sediment biogeochemical processes.

**Ad. Chapter 2.6:**

* The Baltic Sea, with its gradient from open ocean to freshwater salinity, (at least) two very distinct freshwater endmembers with respect to alkalinity, and measurable contributions from unusual acid-base systems, provide a unique system to study the coastal carbon system.
* Clear trends in alkalinity have been observed over the last 25 years, with strong implication on acidification.
* The range of pH in the Baltic Sea is large, both on temporal (high productivity in connection to a partial separation of production and mineralization) and spatial scale, and past and future trends are determined by a complex interplay of different drivers, some of which can or even are managed
* The non-stoichimetric uptake of C/N/P has been shown for the central and recently also for the northern basins, and enhanced efforts to monitor the carbon system are essential for the understanding of primary production.

**Ad. Chapter 2.7:**

* Microbial community composition and key taxa show pronounced shifts along the environmental gradients of the Baltic Sea with hitherto unknown consequences for overall ecosystem functions
* Biogeochemical processes in the redoxcline of the deep oxygen-deficient basins are carried out by relatively few key taxa which control transformations within the C-, N- and S-cycles with ecosystem-wide impact
* Salinity, bottom water oxygen conditions, and bioturbation are the key determinants for the composition, diversity, and function (e.g., sulfate reduction and sulfide oxidation) of the microbial communities at the sediment surface.
* Deeper sediment communities, including those driving important processes like sulfate reduction and methanogenesis, assemble close to the sediment surface (just below the bioturbation zone) by environmental filtering of the founding surface communities.

**Ad. Chapter 2.8:**

* Organic carbon cycling influences contaminant export from land to sea due to the tendency of many contaminants to sorb to organic matter
* Biogeochemical processes that change ambient conditions, e.g. redox conditions, both in the water column and in sediments, has a large impact on speciation of metals which in turn governs metal cycling and bioavailability
* Exposure to contaminants at environmental concentrations can contribute to shifts in e.g., microbial community composition and enzyme activity.

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