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Fate of legacy ammonium in the coastal Baltic Sea

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Abstract. Eutrophication has enriched coastal sediments globally with organic matter (OM), fuelling internal nutrient loading once benthic hypoxia occurs. Internal ammonium (NH_4^+) loading may be particularly prominent in the coastal Baltic Sea due to the existence of a substantial NH_4^+ pool in its sediments, accumulated via OM burial and mineralization over long-term eutrophication. However, despite its potential to exacerbate eutrophication, internal NH_4^+ loading has so far received little attention in the coastal Baltic Sea. It remains poorly understood for how long the benthic legacy OM may affect both the benthic NH_4^+ pool and the internal NH_4^+ loading via sediment-water NH_4^+ effluxes, especially under varying oxygen availabilities. To reconstruct past and predict future NH_4^+ effluxes in response to different OM loading and bottom water oxygen conditions, we developed a reactive transport model for muddy, organic-rich sediments of the coastal Baltic Sea. Our model results suggest that the legacy OM in the coastal sediments is the key driver sustaining benthic NH_4^+ pools and effluxes both today and well into the future. As today's OM loading is constantly adding new OM to the already existing legacy loading, the benthic NH_4^+ pool will continuously grow and result in persistently elevated NH_4^+ effluxes in the future, even under oxic conditions. If external measures strongly reduce OM loading to eventually pre-industrial levels, the NH_4^+ pool still continues to grow for at least 80 years due to the continued mineralization of legacy OM, which keeps NH_4^+ effluxes elevated for at least 180 years before eventually returning to pre-industrial levels in the year 2300. These model results highlight the persistence

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of eutrophication legacy effects and their importance for ecosystem management of the coastal Baltic Sea. The knowledge obtained is beneficial also for other anthropogenically impacted coastal seas with similar geomorphology as the Baltic Sea.

1 Introduction

Excessive land-derived nutrient loading has led to eutrophication of the coastal oceans globally, thereby strongly enriching these systems with organic matter (OM). The combination of high production rate and fast sedimentation of OM in coastal systems can quickly deplete oxygen (O_2) in the benthic habitat, leading to ecosystem degradation and loss of biodiversity (Smith, 2003; Breitburg et al., 2018), as well as the rapid accumulation of OM in the sediments, creating a eutrophication-induced OM legacy pool (Gustafsson et al., 2012). Over time, aerobic and anaerobic mineralization of this OM generates nutrient pools in the sediment, including ammonium (NH_4^+) and phosphate (PO_4^{3-}; Aller, 2014). The retention (PO_4^{3-}) and consumption (NH_4^+) of these nutrients in the sediment is usually high under oxic bottom water conditions; yet, when benthic hypoxia (O_2 concentration < 63 µM or 2 mg L^{-1}, Middelburg and Levin, 2009) develops, both nutrients are preferentially released into the water column (Koop et al., 1990; Cowan and Boynton, 1996; Norkko et al., 2015), leading to internal nutrient loading with high potential to reinforce eutrophication (Pitkänen et al., 2001; Conley et al., 2007). Research on internal nutrient loading has often focused on PO_4^{3-} due to its crucial role in the repeated occurrences of massive cyanobacteria blooms and metal recycling (Conley et al., 2002; Vahtera et al., 2007; Viktorsson et al., 2013; Matisoff et al., 2016), whereas internal NH_4^+ loading has so far received little attention. This is a critical oversight, considering that excess bioavailable nitrogen (N) is a key driver of coastal eutrophication (Ryther and Dunstan, 1971).

Internal NH_4^+ loading may be particularly prominent in the long-term eutrophied coastal Baltic Sea. Compared to coastal regions worldwide, the coastal Baltic Sea displays the highest benthic NH_4^+ effluxes of all studied sites and a highly pronounced relationship between NH_4^+ efflux and bottom water O_2 condition (Fig. 1A). NH_4^+ effluxes increase significantly with decreasing O_2 concentrations across all investigated sites in the coastal Baltic Sea, despite their dispersed geographic locations (Denmark, Sweden, and Finland) and only moderate number of replicates in the cited studies (n= 6−14; Fig. 1B).

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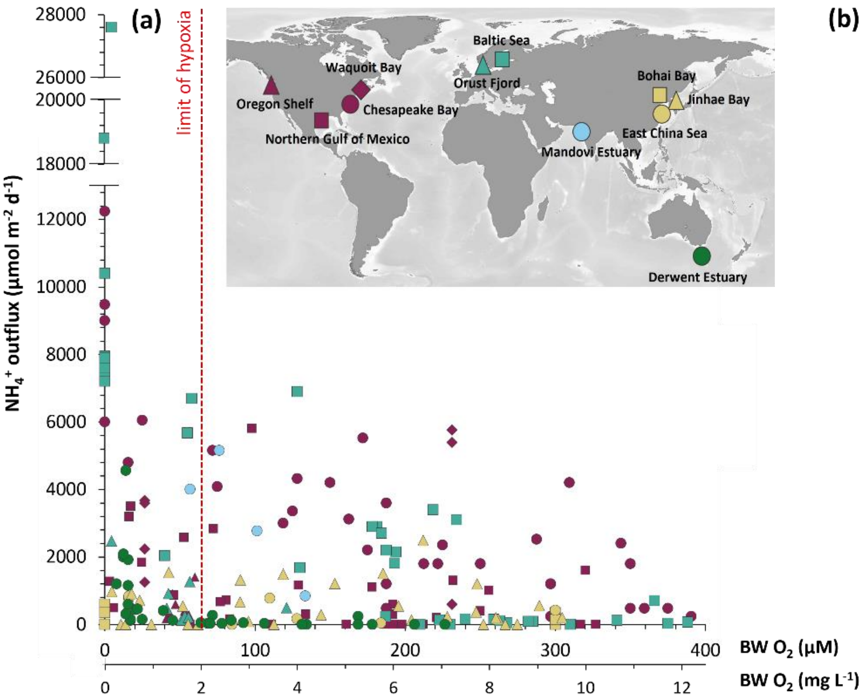

(b)

Area (n)	p
Oregon Shelf (7)	0.239
Northern Gulf of Mexico	
Louisiana Shelf (25)	0.102
Corpus Christie Bay (6)	0.110
Chesapeake Bay (33)	<0.001*
Waquoit Bay (8)	0.804
Orust Fjord (8)	0.156
Baltic Sea	
Himmerfjärden (14)	0.002*
Kanholmsfjärden (7)	0.001*
Skive Fjord (6)	0.041*
Tvärminne Archipelago (9)	<0.001*
Mandovi Estuary (4)	0.123
Bohai Bay (8)	0.528
Coastal East China Sea (15)	0.315
Jinhae Estuary (31)	0.736
Derwent Estuary (30)	0.010*

* = statistical significance

Figure 1. (a): Benthic ammonium (NH_4^+) efflux at different bottom water oxygen (BW O_2) concentrations from coastal sites around the world, and **(b):** statistical significance (p≤ 0.05) of each site's relationship between NH_4^+ and BW O_2 tested with linear regression analysis (n= number of measurements, *= statistical significance; 0 µM O_2 was changed to 0.1 µM O_2 to conform with the analysis). Sites include the Oregon Shelf (Fuchsman et al., 2015), northern Gulf of Mexico (McCarthy et al., 2008; Nunnally et al., 2014; Roberts and Doty, 2015), Chesapeake Bay (Cowan and Boynton, 1996; Testa and Kemp, 2012), Waquoit Bay (Foster and Fulweiler, 2019), Orust Fjord (Norkko et al., 2019), Baltic Sea (Conley et al., 2007; Bonaglia et al., 2014; Ekeroth et al., 2016; Gammal et al. 2017), Mandovi Estuary (Pratihary et al., 2009), Bohai Bay (Mu et al., 2017), coastal East China Sea (Song et al., 2021), Jinhae Bay (Huang and An, 2022), and Derwent Estuary (Banks et al., 2012). All data originate from core- or chamber-incubations and were included in this overview if they supplied NH_4^+ flux measurements under varying O_2 concentrations; we do not claim an exhaustive literature review, such as can be found by Boynton et al. (2017). NH_4^+ effluxes are given as average of replicate measurements per investigation without standard-deviation or -error; NH_4^+ uptake into the sediment is shown as zero outflux. The red dashed line gives the limit of hypoxia (O_2 concentration < 63 µM, resp. < 2 mg L^{-1}, Middelburg and Levin, 2009). Map is drawn with Ocean Data View (Schlitzer 2023).

This clear pattern might be explained by Baltic Sea-specific environmental factors that reduce the influence of natural variability in measurements, such as low species diversity, negligible tidal influence, and a long water residence time, as well as by the presence of a significant benthic legacy pool of NH_4^+. Since the acceleration of Baltic Sea eutrophication in the 1950s (Zillen and Conley, 2010; Gustafsson et al., 2012), OM and eventually NH_4^+ have accumulated in the porewaters of muddy accumulation bottoms, visible today as characteristic NH_4^+ profiles of strongly increasing concentrations with depth that are

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consistent across seasons (Fig. 2). In a first-order estimate, using coastal boundaries as defined by the Water Framework Directive (European Communities, 2003) including Kattegat, coastal sediment classifications according to Al-Hamdani and Reker (2007), and example 0–20 cm porewater NH_4^+ profiles from a representative sandy (site VE15, Vistula Estuary, Bay of Gdansk, Thoms et al., 2018) and muddy (site Storfjärden, Tvärminne Archipelago, Gulf of Finland, D. Hellemann Fig. 2) sediment, we suggest that > 100 kt NH_4^+ could be stored in the top 20 cm of Baltic Sea coastal sediments. This is not only a larger pool of bioavailable N than the pelagic pool of dissolved inorganic nitrogen (DIN) in the coastal Baltic Sea, estimated at ~ 61 kt N using a 21 % share of DIN in total nitrogen (TN; C. Lønborg, pers. comm.) and the pelagic TN pool estimate for the coastal Baltic Sea from Lønborg and Markager (2021), but, furthermore, it is a minimum estimate of the entire NH_4^+ sediment pool considering that concentrations continue to increase with depth below 20 cm.

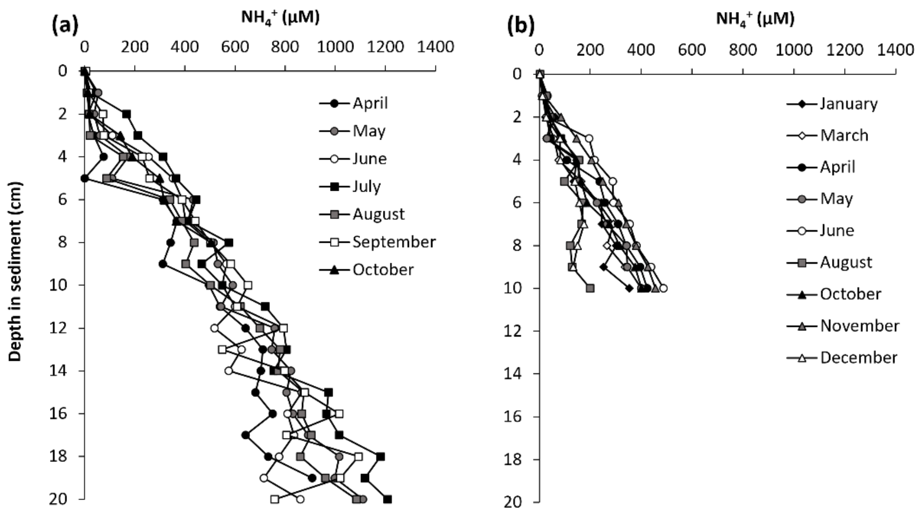

Figure 2. Example porewater profiles of ammonium (NH_4^+) in muddy sediments of the coastal Baltic Sea, taken from **(a)** site Storfjärden (33 m deep, 0–20 cm sediment depth, April–October 2016) and **(b)** site Långholmsbranten (21 m deep, 0–10 cm sediment depth, December 2022–November 2023), north-western Gulf of Finland; data: D. Hellemann. The pattern of NH_4^+ concentrations increasing with sediment depth and not showing significant variations between seasons is in line with profiles from other muddy sites of the coastal Baltic Sea, such as the Himmerfjärden estuary, western Baltic Proper (Bonaglia et al., 2014), coastal Luebeck and Mecklenburg Bight, and Tromper Wiek, southwestern Arkona Basin (Gogina et al., 2018).

The impact of this NH_4^+ pool on water column biogeochemistry is largely regulated by O_2 availability, although factors such as OM availability and benthic infauna also affect the magnitude of sediment-water fluxes (Boynton et al., 2017). Commonly, under normoxic conditions, most of the NH_4^+ that diffuses upwards from porewater is oxidized to nitrate (NO_3^-) via nitrification in the oxic surface sediment and subsequently is reduced to bio-unavailable di-nitrogen gas (N_2) via denitrification in the underlying anoxic sediment (Fig. 3A). This also happens in the numerous 3-dimensional oxic-anoxic interfaces created by

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benthic infauna (Andersen and Kristensen, 1991). If microphytobenthos is present on the sediment surface, it additionally takes up nutrients from the underlying sediments (Sundbäck and Miles, 2000). Together, these factors commonly buffer against large NH_4^+ effluxes from the sediment, leaving only residual fluxes originating from faunal activity (Andersen and Kristensen, 1991; Karlson et al., 2007). When benthic hypoxia or anoxia develops, the oxic sediment surface is lost, benthic infauna and microphytobenthos are extirpated, and the benthic N turnover processes change from N removal to N recycling (Kemp et al., 1990; Jensen et al., 1990; Childs et al., 2002; Karlson et al., 2007; Jäntti and Hietanen, 2012). The extent of these effects depends on frequency and duration of O_2 deficiency (Conley et al., 2007; Jäntti and Hietanen, 2012; Villnäs et al., 2012), but they all fuel NH_4^+ release into the water column compared to normoxic conditions, enriching the pelagic pool of bioavailable N (Fig. 3B). While exceptions to this pattern exist (Foster and Fulweiler, 2019; Testa et al., 2025), the available field data from the coastal Baltic Sea show a significant trend following this paradigm, resulting in internal loading of bioavailable N under hypoxia (Fig. 1). In the open Baltic Sea, such internal N load would be efficiently removed via denitrification at the redoxcline of the permanently hypoxic deep waters (Hietanen et al., 2012; Dalsgaard et al., 2013). However, in the coastal Baltic Sea, pelagic denitrification is not a common feature as density- and wind-driven water mixing usually prevent large-scale permanent hypoxia (Leppäranta and Myrberg, 2009; Conley et al., 2011) and thus the establishment of a pelagic denitrifier community (Falk et al., 2007; Kim et al., 2011). Consequently, in the coastal Baltic Sea, the internal N load from benthic NH_4^+ efflux remains in the pelagic system and thus available for coastal primary production, potentially enhancing coastal eutrophication.

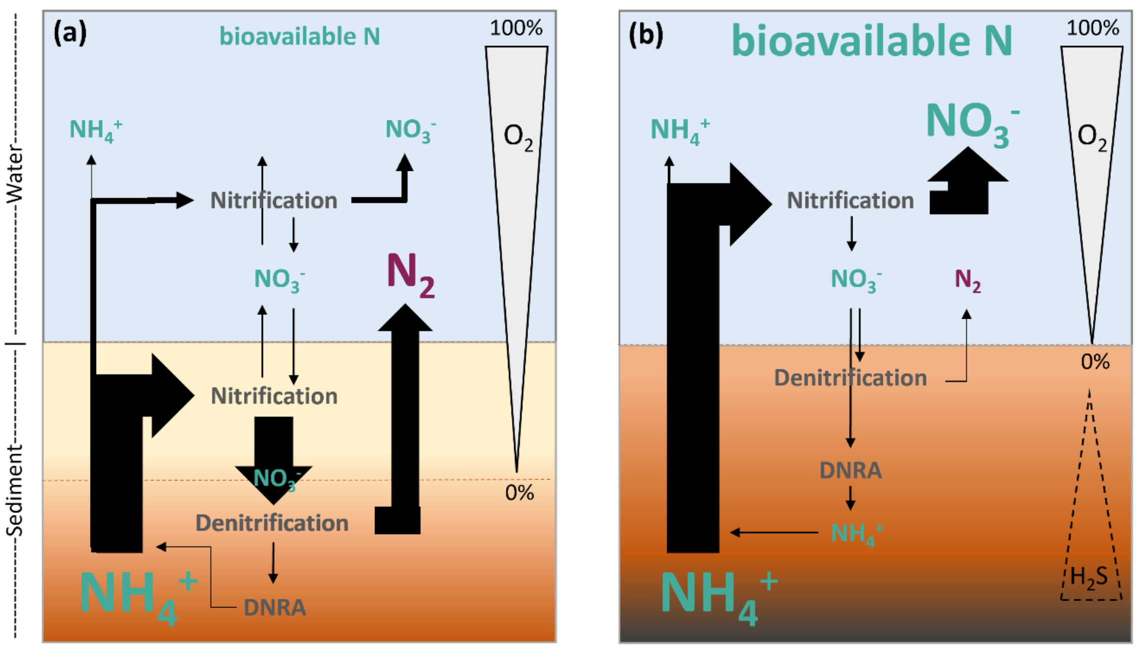

Figure 3. Conceptualized ammonium (NH_4^+) turnover in coastal sediments under **(a)** normoxic and **(b)** hypoxic conditions, based on Kemp et al. (1990), Jensen et al. (1990), Conley et al. (2007), Jäntti and Hietanen (2012) and others. Bioavailable nitrogen (N)-compounds shown in green, non-bioavailable N-compounds shown in violet. Arrow thickness symbolizes relative flux magnitude; font size symbolizes relative

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compound abundance. Anaerobic ammonium oxidation is not shown as often negligible in organic-rich coastal sediments (Dalsgaard et al., 2005). **(a) normoxic conditions**: NH_4^+ flux across the sediment-water interface is small, as most of the upwards diffusing NH_4^+ is oxidized to nitrate (NO_3^-; nitrification) when passing the oxic layer. Due to typically strong coupling of nitrification and denitrification in coastal sediments, most of the resulting NO_3^- is subsequently reduced to non-bioavailable di-nitrogen gas (N_2; denitrification). Hence, the addition of bioavailable N compounds to the water column is relatively small. **(b) hypoxic conditions:** NH_4^+ flux across the sediment-water interface is large, as the oxic sediment layer is lost, disabling nitrification in the sediment; consequently, upwards diffusing NH_4^+ is not transformed to NO_3^- and thus can neither be removed in denitrification. Once in the water column, NH_4^+ accumulates under hypoxic conditions or is oxidized to NO_3^- under oxic conditions, either way increasing the pelagic pool of bioavailable N. Only a small part of water column NO_3^- is assumed to diffuse back into the sediments, as coastal water turbulence is stronger than diffusion. In the absence of hydrogen sulfide (H_2S), this NO_3^- can be reduced to N_2, while in the presence of H_2S dissimilatory nitrate reduction to ammonium (DNRA) dominates, further enriching the NH_4^+ pool.

The timeline of internal NH_4^+ loading affecting the Baltic Sea coastal ecosystem is uncertain, as the long-term fate and persistence of the benthic legacy NH_4^+ pool is largely unknown. For the TN pool in Baltic Sea sediments, Lønborg and Markager (2021) suggest a legacy effect of several centuries. This gap in knowledge limits the capacity of eutrophication mitigation measures, such as the Baltic Sea Action Plan (BSAP), an agreement between the Baltic states on specific actions to bring the Baltic Sea back to a good environmental status (HELCOM 2007; 2021). Coastal hypoxia is likely to increase in the future due to climate change driven water warming (Caballero-Alfonso et al., 2015; Breitburg et al., 2018), increasing the potential for internal NH_4^+ loading. Furthermore, present-day OM loading to coastal Baltic Sea sediments remains at ~10 mmol OM m^{-2} d^{-1} (estimated from the physical-biogeochemical BALTSEM model, Gustafsson et al., 2012, Fig. 4A), which is lower than the peak loading of ~14 mmol OM m^{-2} d^{-1} in the 1990s, but still several-fold higher than before the onset of eutrophication. This means that new OM is continuously added to sediments still containing the OM legacy from past eutrophication, which is likely to extend the lifetime of the existing NH_4^+ pool and its potential to impact the coastal ecosystem.

Interestingly, while data from the Chesapeake Bay and the Derwent Estuary also show a significant relationship between benthic NH_4^+ efflux and O_2 deficiency (Fig. 1B), the NH_4^+ porewater profiles of the Chesapeake Bay do not indicate the existence of a NH_4^+ legacy pool (Kemp et al., 1990; Boynton et al. 2017; no porewater data available for the Derwent Estuary). Despite a eutrophication history similar to the Baltic Sea, the OM deposition environment of the Chesapeake Bay does not seem to support the build-up of benthic legacy pools, owing to higher energy of physical processes, including tidal flushing, and shorter water residence time due to a more open geomorphology. Hence, eutrophication-induced legacy effects similar to the ones from the coastal Baltic Sea might be expected more commonly in eutrophic systems of a semi- to fully enclosed geomorphology and long water residence time, such as the Black Sea (BSC, 2019), Sea of Marmara (Demirel et al., 2023) or even inland waters such as Lake Erie (Watson et al., 2016). Owing to its unique data-richness, the coastal Baltic Sea can serve as a key example for these systems and their potential future evolution (Reusch et al., 2018).

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2 Reactive transport modelling to investigate benthic NH_4^+ turnover and eutrophication legacy effects

To shed light on the long-term fate and persistence of the legacy NH_4^+ pool in the coastal Baltic Sea, we investigate future scenarios of sediment-water NH_4^+ effluxes in response to OM loading and bottom water O_2 availability, using a 1-dimensional reactive transport model (RTM). The RTM is also used to investigate seasonal variability, which allows comparison with existing flux measurements (Fig. 1).

2.1 The model set-up

The 1-dimensional RTM is developed based on the R package "ReacTran" and comprises a multi-component diagenetic setting to examine benthic NH_4^+ efflux in response to two different OM loading scenarios and three different bottom water O_2 conditions (Table 1). The RTM assumes a 1-meter-long sediment column, as the top meter of sediment is usually highly reactive and determines the mass exchange between water and sediment. Into this sediment column, the redox ladder of marine OM degradation is fully integrated, so that OM is sequentially oxidized with O_2, NO_3^-, iron and manganese, and sulfate (SO_4^{2-}), followed by methanogenesis when SO_4^{2-} concentrations are low. These six primary reactions of OM oxidation are complemented by eight secondary reactions (Appendix A, Table A1), largely following Reed et al. (2011) with some modifications. Reaction rates, and reaction and transport parameters are given in Appendix A, Tables A2−A4. Nitrification and denitrification, the two main processes controlling benthic N cycling, are also included, whereas anaerobic ammonium oxidation is not considered due to its commonly negligible occurrence in organic-rich coastal sediments (Dalsgaard et al. 2005). Boundary conditions at the top and bottom of the sediment model domain are defined for both solid and liquid phases. For solid phases, an OM deposition flux (µmol m^{-2} d^{-1}) derived from the simulation of the BALTSEM model (Gustafsson et al., 2012) is prescribed at the sediment–water interface and as the model runs, the OM accumulates over time in the sediment column. OM is removed from the model domain bottom by deep burial. For liquid phases (porewater), the upper boundary condition is set to be determined by the concentration gradient between sediment surface and bottom water via diffusion, while the lower boundary is open so that the RTM determines how much ions could be "leaked out" of the model domain via diffusion and sedimentation. The ion concentrations of the bottom water are assumed to be constant, i.e. most are zero except for SO_4^{2-} (5 mmol L^{-1}), NO_3^- (0.01 mmol L^{-1}) and O_2 (0, 0.063, and 0.2 mmol L^{-1}, depending on the scenario run). Together, this set-up allows to investigate how the sediment system responds to changes in OM loading, which in turn regulates the sedimentary NH_4^+ pool and efflux.

The RTM runs on a 1 cm depth resolution and a default 1-year time step. It is designed for a muddy, OM-rich accumulation bottom, which is with ~41 % the predominant bottom type of the coastal Baltic Sea (calculated using Al-Hamdani and Reker (2007) and coastal boundaries following the Water Framework Directive including Kattegat (European Communities, 2003)). To verify the model, we used the environmental conditions and geochemical porewater profiles of the coastal site "Storfjärden", north-western Gulf of Finland (Jilbert et al. 2018, Appendix B, Fig. B1, doi.org/10.5281/zenodo.17082148).

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2.2 Scenario modelling

The OM loading generated from the BALTSEM model (Gustafsson et al., 2012) covers a period from 1850 to 2400. Initially, the model is run into a steady state using pre-industrial levels of OM flux to the sediment surface as a baseline prior to eutrophication. Then, an OM overloading dataset is used as a forcing from 1850 to 2020, followed by two future scenarios from 2020 to 2400: a "Present scenario", that keeps the OM loading rate as a constant of the value of the year 2020, and a "Pre-industrial scenario", that reduces OM loading in accordance with the BSAP target until 2100, and continues to further reduce OM loading beyond 2100 until pre-industrial levels are reached (Table 1). By adopting this approach, we are able to compare the NH_4^+ pool and fluxes in 2100, when BSAP targets of "good environmental status" are considered to be achieved, with the later situation when true pre-industrial conditions are re-established. Each scenario is run for three classes of bottom water O_2 conditions (normoxia, hypoxia, and anoxia; Table 1), representing best and worst case O_2 conditions. To account for O_2 transport by bioturbating infauna during normoxia, O_2 is forced to penetrate 2 cm deep into the sediment and subsequently decreases exponentially to 0 µM at 5 cm depth. Initial tests showed no significant difference in NH_4^+ fluxes between a forced O_2 penetration depth of 1 or 2 cm.

Table 1. Key input variables and scenario assumptions used in the reactive transport model; with OM = organic matter, C = carbon, O_2 = oxygen, and BSAP = Baltic Sea Action Plan.

OM loading scenarios	**"Present scenario"** OM loading is kept constant on the value of year 2020 (381 µmol C m^{-2} d^{-1})			**"Pre-industrial scenario"** OM loading is reduced following BSAP targets until 2100, and continues until reaching pre-industrial levels		
Bottom water O_2 conditions	Normoxia (200 µM)	Hypoxia (63 µM)	Anoxia (0 µM)	Normoxia (200 µM)	Hypoxia (63 µM)	Anoxia (0 µM)
Salinity	7					
Sulfate (mM)	5					
OM loading data	BALTSEM model (Gustafsson et al. 2012)					

Due to ongoing climate change, bottom water temperatures will likely rise over the modelled future period and affect reaction rates and resulting NH_4^+ effluxes (Boynton et al. 2017), as heterotrophic metabolic processes are expected to increase at twice the rate of autotrophic production (Harris et al. 2006). To provide a first-order assessment of temperature effects on NH_4^+ effluxes, we carried out a sensitivity test by increasing all used rate constants in the model by 20 % (Gudasz et al., 2015). The resulting effect on the modelled NH_4^+ effluxes was negligible, hence we do not discuss further the effects of climate change on our simulations. However, since our model is a generalization that does not account for variable temperature effects, for

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example along water depth gradients, our modelled NH_4^+ fluxes may be considered to represent a lower limit of potential values if bottom waters continue to warm in the future.

In a second set of simulations, the RTM was set to a monthly resolution to assess potential seasonal fluctuations, such as likely included in the field data of Fig. 1, using the example period 2010–2020 and the same set of parameters as given in Table 1. Only the bottom water O_2 conditions deviated from Table 1 and instead were (i) normoxia of 200 µM all year round, (ii) anoxia in summer (May to August) and normoxia in the remaining year, and (iii) anoxia all year round. The model code for all scenarios and simulations can be found at doi.org/10.5281/zenodo.17082148.

3 Coastal NH_4^+ effluxes under future scenarios of OM loading and O_2 availability

The BALTSEM-derived OM loading in both model scenarios can be divided into four phases (Fig. 4, 5). Phase I gives the pre-industrial OM level representing natural loading. Phase II is characterized by a sharp increase in OM loading from the onset of eutrophication in the 1950s to the early 2000s, when the loading reaches its peak of ~14 mmol OM m^{-2} d^{-1}, tripling the OM loading of Phase I. Following this peak, Phase III covers the decrease in OM loading to its present-day value of ~10 mmol m^{-2} d^{-1} (Present scenario), or respectively, to the projected pre-industrial baseline values (Pre-industrial scenario). Phase IV represents the far-future return to pre-industrial OM loading and thus can only be seen in the Pre-industrial scenario.

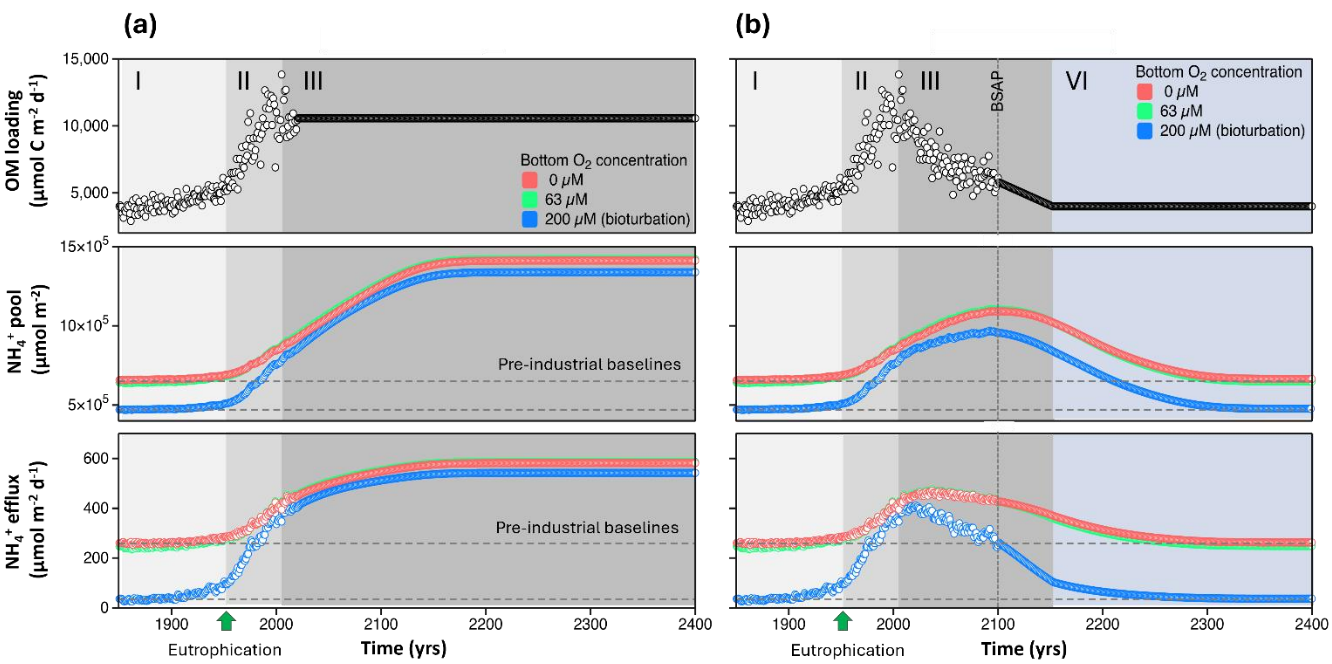

Figure 4. Sediment organic matter (OM) loading, sediment ammonium (NH_4^+) pool, and sediment-water NH_4^+ efflux in the northern coastal Baltic Sea under anoxic (red), hypoxic (green) and normoxic (blue) bottom water oxygen (O_2) conditions, modelled for the OM loading

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scenarios **(a)** "Present scenario" (constant OM loading) and **(b)** "Pre-industrial scenario" (reduced OM loading down to pre-industrial levels). Modelling period 1850 to 2400. The NH_4^+ pool is the annual amount of porewater NH_4^+ integrated over a 1-m sediment model domain. Grey shadings indicate the different phases of OM loading with (I) pre-industrial, (II) strongly increasing, (III) after-peak, and (VI) return to pre-industrial. Horizontal dashed lines give the pre-industrial baselines of NH_4^+ pool and efflux at normoxic and deteriorated O_2 conditions. The vertical dashed line gives the end point of OM load reductions if following the target of the Baltic Sea Action Plan (BSAP, see Table 1). The green arrow indicates the onset of widespread eutrophication around the 1950s.

During Phase I, the model reaches a steady state of a constant NH_4^+ pool in the sediment and low NH_4^+ efflux to the water column. In phase II, the NH_4^+ pool and efflux respond closely to the fast increase in OM loading. In phase III, the OM loading decreases to varying degrees, while both the pool and the efflux of NH_4^+ continue to increase, indicating a legacy effect of eutrophication. In the Present scenario, the NH_4^+ pool and efflux continue to rise for over a century until reaching a stable high plateau shortly after 2150. In the Pre-industrial scenario, a maximum NH_4^+ pool is observed at around 2100, while the NH_4^+ efflux reaches its peak quickly in the first half of the 21st century. Such legacy effects are caused in the model by NH_4^+ being continuously regenerated from the in sediment accumulated OM over subsequent years. The external OM supply is larger than the sediment-internal mineralization of deposited OM, which leads to a long-term gradual accumulation of OM and consequently growth of the NH_4^+ pool during phase II and III, as the sediment column fills up with degrading OM (Fig. 4). When OM loading and NH_4^+ efflux are plotted against each other, the legacy effect is clearly observed in the Pre-industrial scenario as a negative correlation during phase III, meaning that the NH_4^+ efflux continues to increase despite the decline in OM loading (Fig. 5B III). Historical field investigations in the coastal Baltic Sea suggest that in the early stages of eutrophication in the mid-late 20th century, benthic NH_4^+ dynamics were strongly driven by seasonal changes in OM loading and their resulting O_2 conditions (e.g. Nedwell et al., 1987; Jensen et al., 1990). This contrasts with the present-day situation of consistently elevated NH_4^+ concentrations in porewaters (Fig. 2) and is consistent with the concept of a legacy NH_4^+ pool in the sediments that has developed over time to dominate the flux at the sediment-water interface.

Eventually, both the NH_4^+ pool and efflux return to pre-industrial levels in the Pre-industrial scenario (Fig. 4B, 5B IV), for which the reduction in OM loading is key and thus, no return to pre-industrial levels happens in the Present scenario (Fig. 4A, 5A III). From the start of nutrient reductions in the early 2000s, it takes about 150 years for the OM pool, and more than 300 years for the NH_4^+ pool, to reach pre-industrial levels in the Pre-industrial scenario. Interestingly, by 2100, when BSAP-derived OM load reductions to achieve good environmental status are reached (HELCOM, 2007; 2021; BALTSEM), the benthic NH_4^+ pool in our model is still at its maximum, sustaining high NH_4^+ effluxes similar to the 1990s (Fig. 4B). This implies that it might take longer than previously thought for the BSAP to achieve its environmental targets.

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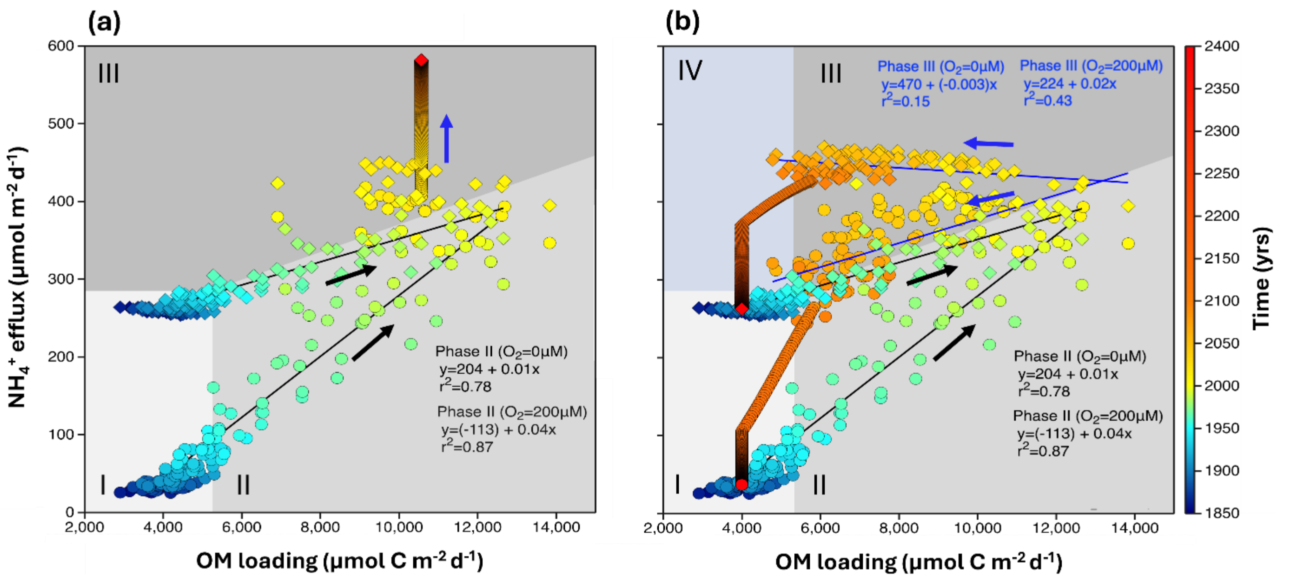

Figure 5. Sediment ammonium (NH_4^+) efflux plotted against organic matter (OM) loading under two conditions of bottom water oxygen (diamonds = 0 µM; circles = 200 µM O_2), shown for the (**A**) Present scenario and (**B**) Pre-industrial scenario of OM loading. Grey-shading indicates the different phases of OM loading with (I) pre-industrial, (II) strongly increasing, (III) after-peak, and (IV) return to pre-industrial, corresponding to Figure 3. Arrows and color-coding indicate the temporal evolution. A negative correlation between OM and NH_4^+, indicative of legacy effects, is only observed in (B). Note that in (B) data points of the late years (phase IV) seem to enter phase I, which is only a graphical glitch as they return to pre-industrial conditions.

As the NH_4^+ effluxes are driven by the NH_4^+ pool (Fig. 4), they are clearly elevated compared to pre-industrial levels for an indefinite time in the Present scenario (Fig. 4A III), and for the next at least 180 years in the Pre-industrial scenario (Fig. 4B IV). The only exception is the NH_4^+ efflux under normoxic bottom water conditions in the Pre-industrial scenario, which appears to respond more closely to changes in OM loading rather than the NH_4^+ pool (Fig. 4B). This observation indicates a more efficient consumption of the NH_4^+ pool within the sediment under oxic conditions (nitrification with subsequent denitrification), thereby reducing its influence on sediment-water NH_4^+ effluxes (Fig. 3). This is supported by the oxic NH_4^+ efflux returning to pre-industrial levels already at around 2200, despite of the NH_4^+ pool still being above baseline conditions.

Interestingly, a similar pattern cannot be seen in the Present scenario (Fig. 4A), suggesting that OM loading could overrule the role of O_2 in regulating sediment-water NH_4^+ effluxes, as outlined in the following. Under low OM availability, such as in the pre-industrial period before widespread eutrophication (Fig. 4 I) or in the Pre-industrial scenario after drastic reductions in OM loading (Fig. 4B IV), our modelling results show NH_4^+ effluxes clearly following bottom water O_2 conditions with negligible efflux under normoxia and high efflux under anoxia and hypoxia as outlined in Fig. 3. However, with increasing OM availability, such as during phase II and III in Fig. 4A, modelled NH_4^+ effluxes under all O_2 conditions including normoxia strongly increase regardless of O_2 conditions. This suggests that nitrification in oxic surface sediments, the predominant

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process buffering against large NH_4^+ effluxes, can lose its function in environments of high OM availability, simply due to being overwhelmed by excessive NH_4^+ from OM mineralization (Prosser 2007). These results suggest that under low OM availability O_2 remains the key parameter regulating benthic NH_4^+ efflux as commonly acknowledged (Fig. 3), whereas under high OM availability NH_4^+ fluxes are increasingly subject to OM loading history and subsequent legacy effects of NH_4^+ regeneration. In strongly eutrophied systems, this mechanism could result in high NH_4^+ effluxes even under oxic bottom water conditions, potentially explaining some of the high oxic NH_4^+ effluxes observed in published field data (Fig. 1).

Despite being derived from OM-rich settings, the field data from the coastal Baltic Sea, however, do not reflect the model observation of almost O_2-independent NH_4^+ effluxes, but rather display a strong differentiation between normoxic and anoxic NH_4^+ effluxes (Fig. 1). This offset between model and field observation can be explained with the resolution of the RTM, which runs on an annual basis and thus does not capture seasonal fluctuations and extreme events that can drastically increase the efflux but rather merges them to an annual average. Thus, to take a closer look at the impact of seasonality on NH_4^+ effluxes, we also ran the model on a monthly resolution for one decade to assess the response of NH_4^+ efflux to three months of seasonal anoxia. High OM loading in summer can quickly lead to anoxic conditions in bottom waters and enhance NH_4^+ efflux by an order of magnitude in comparison to seasons with low OM loading and oxic bottom waters. Our model results show that the NH_4^+ efflux quickly responded to OM loading and resulting anoxia, leading to high values in summer (Appendix C, Fig. C1). The range of values captured a large part of coastal Baltic Sea field data, except for the extremely high NH_4^+ effluxes during anoxia (Fig. 1). While these values originate from 3 different sites (Kanholmsfjärden, Sweden; Tvärminne Archipelago, Finland; Skive Fjord, Denmark) and thus show representation across the coastal Baltic Sea area, they are nevertheless specific seasonal events driven by an individual combination of OM loading and loading history, anoxia duration, and temperature, which explains why they cannot be fully captured in our RTM that was calibrated using one representative site (Storfjärden). Without these extreme values adding to the annual average, the modelled anoxic NH_4^+ efflux is lower than it can periodically be in nature, explaining the low differentiation between modelled anoxic and oxic NH_4^+ effluxes. This affects only the anoxic, not the oxic fluxes (Fig 1., Fig. C1).

4 Sensitivity of the benthic NH_4^+ legacy pool to changed boundary conditions

To test the sensitivity of our modelled NH_4^+ pool and efflux to different marine environmental conditions, we applied a range of salinities (representative of different SO_4^{2-} concentrations in bottom water), OM loadings, and sedimentation rates to our RTM, both under normoxic and anoxic bottom water conditions, using the northern coastal Baltic Sea as a baseline (salinity of 6 with a SO_4^{2-}concentration of 5 mmol L^{-1}, Pre-industrial OM loading scenario, and a sedimentation rate of 6 mm yr^{-1}). The OM loadings were generated by the BALTSEM simulations. The sedimentation rate affected how much and for how long these OM loadings would remain in each sediment depth layer.

Under normoxic conditions and compared to the baseline, an increase in salinity would increase the size of both NH_4^+ pool and efflux (Fig. 6A I). The key reason for this observation is the higher overall rate of OM mineralization regenerating NH_4^+

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due to a higher share of energetically more favourable SO_4^{2-} reduction compared to methanogenesis. Hence, systems with higher salinity than the Baltic Sea may experience more pronounced legacy effects in terms of direct production and resulting efflux of NH_4^+. A 50 % decrease in OM loading would reduce the magnitude of the NH_4^+ pool and efflux similarly by 50 %, whereas the duration until the efflux returns to pre-industrial baseline levels would stay unchanged (Fig. 5A II), reflecting the coherent process of NH_4^+ turnover in the sediment (kinetics of nitrification and denitrification). Further, at high OM loading (200 %), the NH_4^+ efflux becomes less sensitive to the OM loading rate (Fig. 6B II). Increase or decrease in sedimentation rate would accelerate or delay, respectively, the build-up of the NH_4^+ pool and the subsequent legacy period (Fig. 6A III). Under the highest sedimentation rate, the legacy effect disappears already after around 150 years, which however could be due to the set-up of the RTM; the model domain is set to 1 m with simulations running from 1850 to 2400, hence, the OM legacy pool would be moved out of the domain 100–250 years into the future depending on the sedimentation rate. This assumption rests on our general understanding that the top one meter is the most reactive sediment layer that directly determines sediment-water fluxes. Under anoxic bottom water conditions, similar patterns as the ones described can be observed but with slightly stronger effects on the build-up and efflux of the NH_4^+ pool (Appendix D, Fig. D1).

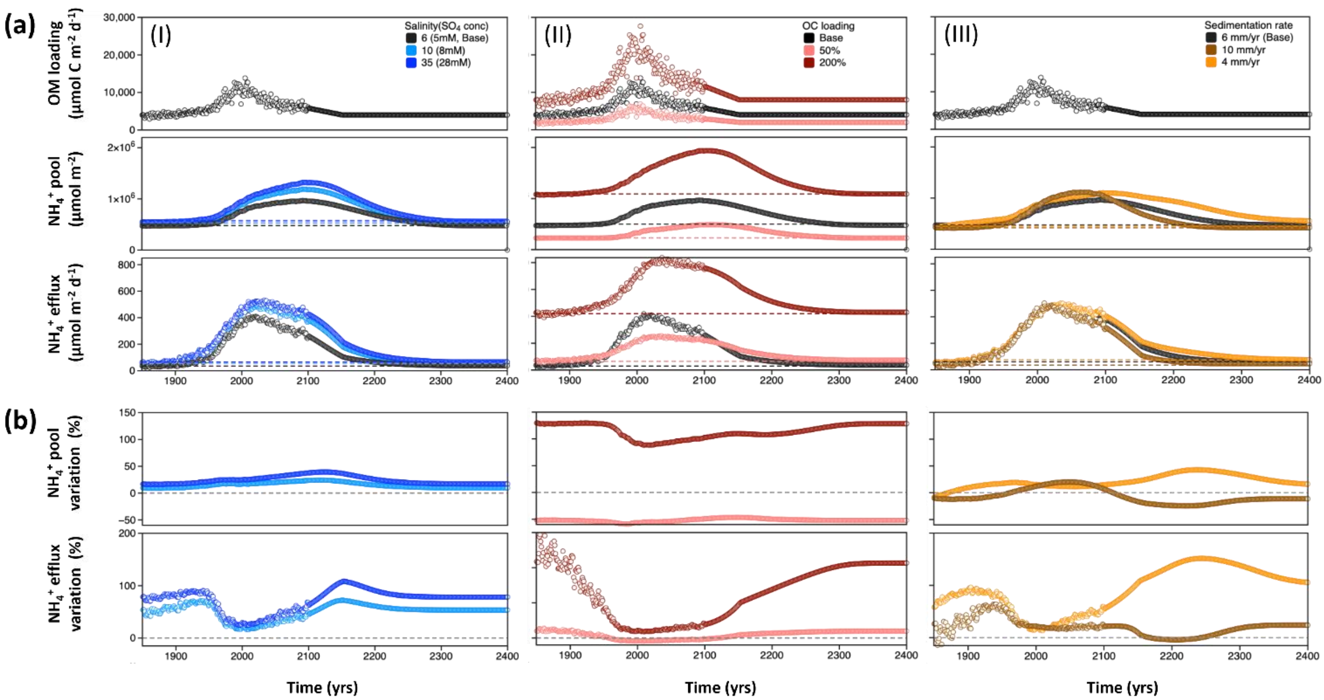

Figure 6. Effect of changed salinity (I), organic matter (OM) loading (II), and sedimentation rate (III) compared to baseline conditions representative of the coastal northern Baltic Sea (i.e. salinity of 6, Pre-industrial OM loading scenario, sedimentation rate of 6 mm yr^{-1}; black line) on **(a)** sediment ammonium (NH_4^+) pool and sediment-water NH_4^+ efflux, and **(b)** its percent change relative to baseline condition. Modelled for normoxic bottom water conditions (O_2 = 200 µM). Dashed lines give the pre-industrial baselines of NH_4^+ pool and efflux.

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While these results point to a general direction for nutrient cycling in eutrophied coastal systems of different environmental conditions, they need to be interpreted while considering the geomorphological context for each system. For instance, a higher salinity is often found in open coastal systems; while more SO_4^{2-} contributes to the mineralization of OM and potential generation of an NH_4^+ pool in the sediment, those systems are commonly highly dynamic with high energy depositional settings. Dispersion or remineralization of OM prior to accumulation in the sediment is expected to reduce the build-up of a legacy pool of OM and NH_4^+ in the sediment, explaining, for example, the absence of such observations in the eutrophied Chesapeake Bay (Kemp et al., 1990; Boynton et al. 2017), a system of much higher tidal range and thus more dynamic environment than the Baltic Sea. Hence, understanding a habitats geomorphological setting and environmental conditions are highly important when aiming to realistically model nutrient flux predications of coastal sediments.

5 Implications for restoring eutrophied coastal ecosystems

Our results highlight the persistence and thus importance of legacy effects from past eutrophication for present and future nutrient cycling and corresponding ecosystem health in the coastal Baltic Sea. We demonstrate that the eutrophication-derived OM pool in the sediments of the coastal Baltic Sea is the key factor that continuously maintains the sedimentary NH_4^+ pool of today and well into the future, which in turn acts as an internal supply of NH_4^+ to the overlaying water column, particularly under anoxic conditions. The model results of the Present scenario show that, if no further nutrient reductions are undertaken and OM loading remains at today's level, mineralization of both legacy OM and continuously new deposited OM could maintain a large pool of NH_4^+, whose magnitude remains constant for an indefinite period of time, resulting in accordingly high sediment-water NH_4^+ effluxes and intensified internal N loading to the coastal ecosystem. This could happen even under oxic bottom water conditions, as high OM loading almost overruled O_2 as the key factor governing sediment-water NH_4^+ effluxes in our RTM. If strong, external nutrient load reductions are applied and, consequently, OM loading drastically decreases, such as in the Pre-industrial scenario, legacy OM will still maintain the coastal benthic NH_4^+ pool on a timescale of centuries, resulting in elevated sediment-water effluxes of NH_4^+ and thus internal N loading to the coastal ecosystem for at least 180 years from the present day. The same holds true for OM load reductions following the BSAP, whose reduction target is assumed to be reached in the year 2100, when the benthic NH_4^+ pool is, however, still at its maximum due to the legacy effect (Fig. 4B). These results are similar to Lønborg and Markager (2021), who estimated a period of about 400 years until a 20 % reduction of external nutrient inputs could overcome the TN legacy in Baltic Sea sediments and lead to good ecological status of the open sea water body. The demonstrated persistency of eutrophication legacies emphasizes the need for early mitigation measures at the first signs of eutrophication (Ehrnsten et al., 2025), as well as policies and implementation that commit to long time scales for restoration.

Considering that our RTM is 1-dimensional and not directly coupled to the water column, we cannot estimate any direct feedback between the sediment and water column. In a natural system, the additional N from internal loading would be taken

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up in primary production and eventually settle back to the sediment to become new OM, which adds complexity to internal nutrient loops in both scenarios and is not included in the current model. It is likely, that the coastal current of the Baltic Sea will keep OM and nutrients for a certain time in the coastal zone, before both eventually end up in the open Baltic Sea (Radtke et al., 2012), which increases the chance that the internal N load is ultimately removed from the aquatic system via denitrification at the permanent redoxcline. How long it takes to remove the internal N load from the coastal zone and thus from the potential to favour coastal primary production, is, however, unclear. Hence, our results provide a baseline for assessing the size and persistency of the legacy pool, while these estimates are still associated with uncertainties.

We acknowledge that the impact of internal N loading on the coastal ecosystem is localized, as despite a predicted rise in hypoxia it is highly unlikely that the entire coastal zone of the Baltic Sea will eventually become O_2 deficient. Localized hypoxic events can lead to localized increases in N availability and subsequent primary production, yet this increase is likely masked by the overall coastal eutrophication. However, for the management efforts to mitigate coastal eutrophication and restore a healthy ecosystem, every addition of biologically available nutrients that is not accounted for in budgets should be noteworthy, even if only localized. In the big picture, internal loading adds more nutrients to a system than it should have and even if these nutrients remain only for a limited time, they can cause ecological harm to the ecosystem and impede eutrophication mitigation measures. The latter is particularly true when considering the persistence of the NH_4^+ pool and thus the long timeframe it could "sabotage" mitigation efforts.

6 Conclusions

Our model results clearly show the persistency of NH_4^+ accumulations in Baltic Sea coastal sediments as a legacy of past and ongoing eutrophication, and their potential impact on the coastal ecosystem in the form of elevated sediment-water NH_4^+ effluxes well into the future, if no strict external nutrient load reductions are enforced. We therefore call for sustained control of N loading to the coastal Baltic Sea in future decades and centuries, especially in the context of climate change, impacting biogeochemical processes rates (Meier et al., 2022). Further, as internal N loading will continue even under the strictest OM load reduction scenario until the legacy pool is depleted (Fig. 6A II), particularly under O_2 deficient conditions (Fig. 5B), it would be advisable to account for it and its associated timeframes when setting nutrient load target values for the coastal Baltic Sea. Our results are beneficial also for other coastal areas of similar geomorphology as the Baltic Sea that face similar challenges in nutrient reduction and ecological management.

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Appendix A

Table A1. Chemical reactions in the RTM; organic carbon (OC) is of the form $(CH_2O)_a(NH_4^+)_b(H_3PO_4)_c$, where a = 106, b = 16, and c = 1.

Primary OC reactions:	
$OC + aO_2 \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + aH_2O$	(R1)
$OC + \frac{4a}{5}NO_3^- + \frac{4a}{5}H^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + \frac{2a}{5}N_2 + \frac{7a}{5}H_2O$	(R2)
$OC + 2aMnO_2 + 4aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + 2aMn^{2+} + 3aH_2O$	(R3)
$OC + 4aFe(OH)_3^{\alpha} + 12aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + 4aFe^{2+} + 13aH_2O$	(R4)
$OC + \frac{a}{2}SO_4^{2-} + aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + \frac{a}{2}H_2S + aH_2O$	(R5)
$OC \rightarrow \frac{a}{2}CO_2 + bNH_4^+ + cH_3PO_4 + \frac{a}{2}CH_4$	(R6)
Secondary reactions:	
$2O_2 + NH_4^+ + 2HCO_3^- \rightarrow NO_3^- + 2CO_2 + 3H_2O$	(R7)
$O_2 + 2Mn^{2+} + 4HCO_3^- \rightarrow 2MnO_2 + 4CO_2 + 2H_2O$	(R8)
$2O_2 + FeS \rightarrow SO_4^{2-} + Fe^{2+}$	(R9)
$2O_2 + H_2S + 2HCO_3^- \rightarrow SO_4^{2-} + 2CO_2 + 2H_2O$	(R10)
$O_2 + CH_4 \rightarrow CO_2 + H_2O$	(R11)
$2Fe(OH)_3 + H_2S + 4CO_2 \rightarrow 2Fe^{2+} + S_0 + 4HCO_3^- + 2H_2O$	(R12)
$Fe^{2+} + H_2S \rightarrow FeS + 2H^+$	(R13)
$SO_4^{2-} + CH_4 \rightarrow 2HCO_3^- + HS^-$	(R14)

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Table A2. Reaction and transport parameters used in the RTM; a) Boudreau 1997, b) Reed et al., 2011. Limiting concentrations double as inhibition coefficients in the Monod scheme (Boudreau 1996).

Parameter	Value or expression	Units
Porosity at depth x[a]	$\phi(x) = \phi_\infty + (\phi_0 - \phi_\infty)exp\,(-\max\,(0, x - x_0)/x_{att})$	cm³ cm⁻³
Porosity at surface sediment[b]	$\phi_0 = 0.95$	$cm^3\ cm^{-3}$
Porosity at 1 meter[b]	$\phi_\infty = 0.6$	$cm^3\ cm^{-3}$
Depth to start exponential decrease	$x_0 = 2$	cm
Attenuation coefficient in exponential decrease	$x_{att} = 1$	
Solid volume fraction[b]	$\phi_S(x) = 1 - \phi$	$cm^3\ cm^{-3}$

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Tortuosity[a]	$\theta^2(\phi) = 1 - 2\ ln\phi$	—
Sediment density	$\rho_S = 2.65$	g cm^{-3}
Sediment accumulation rate[b]	$F_{sed} = 0.6$	cm yr^{-1}
Biodiffusion	$D_b(x) = D_{b0} exp(-\max(0, x - x_0)/x_{att}))]$	cm^2 yr^{-1}
Biodiffusion coefficient	$D_{b0} = 1500$	cm^2 yr^{-1}
Limiting concentration of O_2[b]	$k_{O_2} = 20$	µmol L^{-1}
Limiting concentration of NO_3^-[b]	$k_{NO_3^-} = 4$	µmol L^{-1}
Limiting concentration of MnO_2[b]	$k_{MnO_2} = 4$	µmol g^{-1}
Limiting concentration of $Fe(OH)_3$[b]	$k_{Fe(OH)_3} = 65$	µmol g^{-1}
Limiting concentration of SO_4^{2-}[b]	$k_{SO_4^{2-}} = 1.6$	mmol L^{-1}
R1-6 decay constant (reactive *OC)*	$k_{OC1} = 1.62$	yr^{-1}
R1-6 decay constant (less reactive *OC)*	$k_{OC2} = 0.0086$	yr^{-1}
R7 reaction constant	$k_{R7} = 100000$	$mmol^{-1}$ L yr^{-1}
R8 reaction constant	$k_{R8} = 20$	$mmol^{-1}$ L yr^{-1}
R9 reaction constant	$k_{R9} = 300$	$mmol^{-1}$ L yr^{-1}
R10 reaction constant	$k_{R10} = 1$	$mmol^{-1}$ L yr^{-1}
R11 reaction constant	$k_{R11} = 160$	$mmol^{-1}$ L yr^{-1}
R12 reaction constant	$k_{R12} = 8$	$mmol^{-1}$ L yr^{-1}
R13 reaction constant	$k_{R13} = 100$	$mmol^{-1}$ L yr^{-1}
R14 reaction constant	$k_{R14} = 10$	$mmol^{-1}$ L yr^{-1}

Table A3. Primary chemical reaction rates; i stands for the reactive or less reactive OC pools.

$$R_{O_2} = k_i OC\left(\frac{[O_2]}{k_{O_2} + [O_2]}\right)$$

$$R_{NO_3^-} = k_i OC\left(\frac{k_{O_2}}{k_{O_2} + [O_2]}\right)\left(\frac{[NO_3^-]}{k_{NO_3^-} + [NO_3^-]}\right)$$

$$R_{MnO_2} = k_i OC\left(\frac{k_{O_2}}{k_{O_2} + [O_2]}\right)\left(\frac{k_{NO_3^-}}{k_{NO_3^-} + [NO_3^-]}\right)\left(\frac{[MnO_2]}{k_{MnO_2} + [MnO_2]}\right)$$

$$R_{Fe(OH)_3} = k_i OC\left(\frac{k_{O_2}}{k_{O_2} + [O_2]}\right)\left(\frac{k_{NO_3^-}}{k_{NO_3^-} + [NO_3^-]}\right)\left(\frac{k_{MnO_2}}{k_{MnO_2} + [MnO_2]}\right)\left(\frac{[Fe(OH)_3]}{k_{Fe(OH)_3} + [Fe(OH)_3]}\right)$$

$$R_{SO_4^-} = k_i OC\left(\frac{k_{O_2}}{k_{O_2} + [O_2]}\right)\left(\frac{k_{NO_3^-}}{k_{NO_3^-} + [NO_3^-]}\right)\left(\frac{k_{MnO_2}}{k_{MnO_2} + [MnO_2]}\right)\left(\frac{k_{Fe(OH)_3}}{k_{Fe(OH)_3} + [Fe(OH)_3]}\right)\left(\frac{[SO_4^{2-}]}{k_{SO_4^{2-}} + [SO_4^{2-}]}\right)$$

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$$R_{CH_4}^{i} = k_i OC\left(\frac{k_{O_2}}{k_{O_2} + [O_2]}\right)\left(\frac{k_{NO_3^-}}{k_{NO_3^-} + [NO_3^-]}\right)\left(\frac{k_{MnO_2}}{k_{MnO_2} + [MnO_2]}\right)\left(\frac{k_{Fe(OH)_3}}{k_{Fe(OH)_3} + [Fe(OH)_3]}\right)\left(\frac{k_{SO_4^{2-}}}{k_{SO_4^{2-}} + [SO_4^{2-}]}\right)$$

Table A4. Secondary chemical reaction rates.

$r_{R7} = k_{R7}[O_2][NH_4^+]$
$r_{R8} = k_{R8}[O_2][Mn^{2+}]$
$r_{R9} = k_{R9}[O_2][Fe^{2+}]$
$r_{R10} = k_{R10}[O_2][H_2S]$
$r_{R11} = k_{R11}[O_2][CH_4]$
$r_{R12} = k_{R12}[Fe(OH)_3\ ][H_2S]$
$r_{R13} = k_{R13}[Fe^{2+}][H_2S]$
$r_{R14} = k_{R14}[SO_4^{2-}][CH_4]$

Appendix B

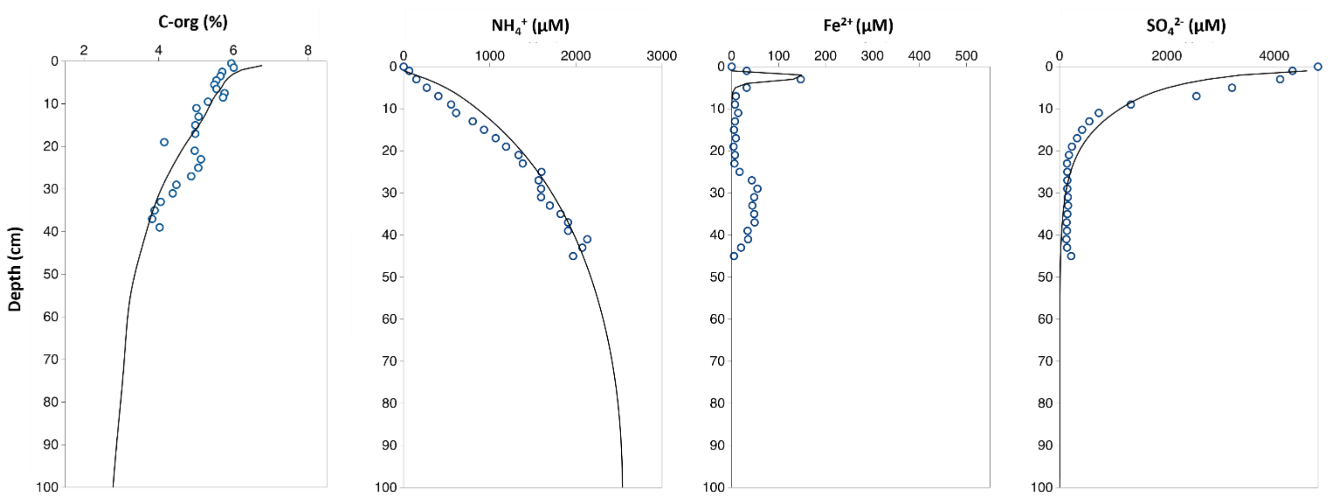

Figure B1. RTM model validation. Sediment and porewater profiles of major model parameters, measured in 2016 at the coastal site "Storfjärden", a 30 m deep site at the south-western coast of Finland (open circles; Jilbert et al. 2018), and modelled results of those parameters (solid line); with C-org = organic carbon content, NH_4^+ = ammonium, Fe^{2+} = iron(II), and SO_4^{2-} = sulfate.

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Appendix C

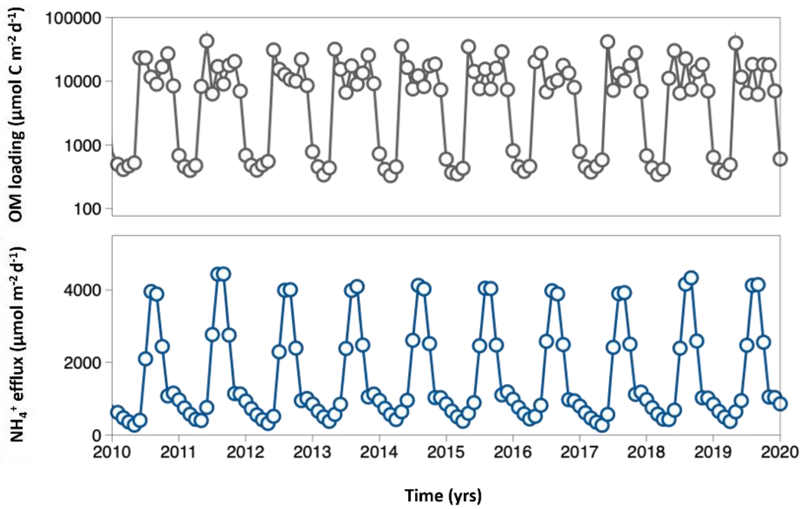

Figure C1. Monthly organic carbon (OC) loading and sediment-water ammonium (NH_4^+) efflux between 2010–2020 calculated by the RTM running on a monthly resolution. The bottom water condition was set to be anoxic (O_2 = 0 µM) from May to August and oxic (O_2 = 200 µM) in the remaining months.

Appendix D

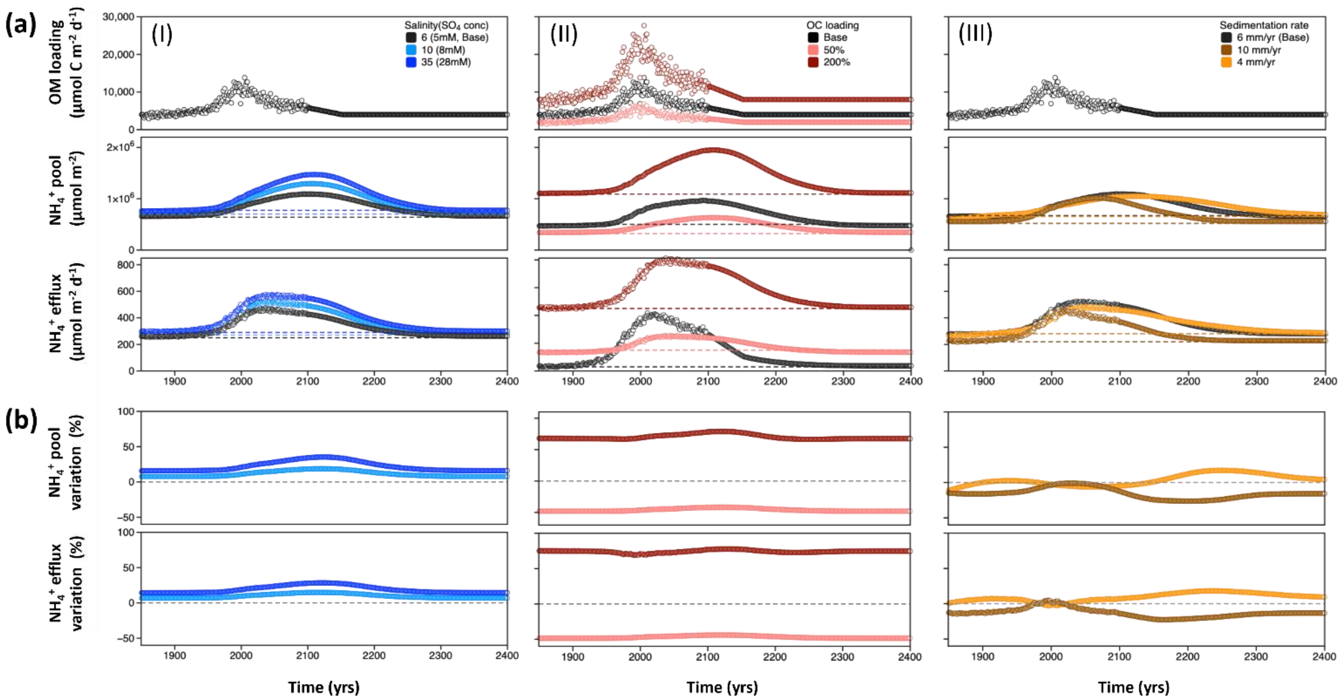

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Figure D1. Effect of changed salinity (I), organic matter (OM) loading (II), and sedimentation rate (III) compared to baseline conditions representative of the coastal northern Baltic Sea (salinity of 6, Pre-industrial OM loading scenario, sedimentation rate of 6 mm yr^{-1}; black line) on (**A**) sediment ammonium (NH_4^+) pool and sediment-water NH_4^+ efflux, and (**B**) its percent change relative to baseline condition. Modelled for anoxic bottom water conditions (O_2 = 0 µM). Dashed lines give the pre-industrial baselines of NH_4^+ pool and efflux.

Code and data availability

The R model code and the model output data are available at https://doi.org/10.5281/zenodo.17082148.

Author contribution

DH: conceptualization, investigation, visualization, writing - original draft (lead), review and editing (lead); XS: conceptualization, investigation, methodology (lead), visualization, writing – review and editing (co-lead); TJ: investigation, funding acquisition, validation, writing – review and editing; EE: validation, writing – review and editing; LH: validation, writing – review and editing; BG: validation, writing – review and editing; CH: validation, writing – review and editing; AN: funding acquisition, validation, writing – review and editing

Competing interests

The authors declare that they have no conflict of interest.

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