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 deposition~~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 and it~~. It~~ remains poorly understood for how long the sediment ~~benthic legacy OM may affect both the benthic~~ NH_4^+ pool could sustain ~~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, OM~~organic~~-rich sediments of the coastal Baltic Sea. Our model results suggest a strong legacy effect in the sediment~~arythe~~ NH_4^+ pool, driven by mineralization of OM that accumulated during past and ongoing eutrophication, as expected. The influence of the sediment NH_4^+ pool on the sediment-water NH_4^+ effluxes in our model is, however, smaller than the direct influence of the external OM loading. The sediment NH_4^+ pool keeps the sediment-water NH_4^+ effluxes 5-10% elevated compared to pre-industrial baseline values for at least 140 years after OM loading has reached constant baseline levels, albeit at a small efflux magnitude. While this NH_4^+ efflux magnitude can increase under increasing OM loading and O_2-deficiency, the legacy effect is generally mild and NH_4^+ effluxes react well to external OM load reductions. If strict OM load reductions following the direction of the Baltic Sea Action

Plan are implemented, sediment-water NH_4^+ effluxes can naturally return to a level similar to the 1950s, the start of wide-spread eutrophication, within 110 years, given that sediments stay normoxic and bioturbated during the reduction period. Without external OM load reductions, NH_4^+ effluxes stay indefinitely elevated and fuel continuous internal NH_4^+ loading. Our findings highlight the complexity of eutrophication legacy effects in regard to NH_4^+ and their varying impact on the ecosystem. The knowledge obtained is beneficial also for other anthropogenically impacted coastal seas with similar geomorphology as the Baltic Sea.~~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 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 not only to ecosystem degradation ~~and loss of biodiversity (Smith, 2003; (~~Breitburg et al., 2018) but also ~~, as well as~~to ~~the~~ 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 dissolved inorganic phosphorous ~~phosphate~~ (DIP~~PO_4^{3-}~~; Aller, 2014). The retention (DIP~~PO_4^{3-}~~) and turnover~~consumption~~ (NH_4^+) of these nutrients within 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 DIP ~~PO_4^{3-}~~ due to its crucial role in the repeated occurrences of massive cyanobacteria blooms and close coupling to metal ~~re~~cycling (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 omission 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 (defined here as the coastal areas of the Baltic Sea extending one nautical mile seaward from territorial water baselines, following the Water Framework Directive; European Communities, 2003). Compared to coastal regions worldwide, the coastal Baltic Sea displays the highest

sediment-water ~~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).

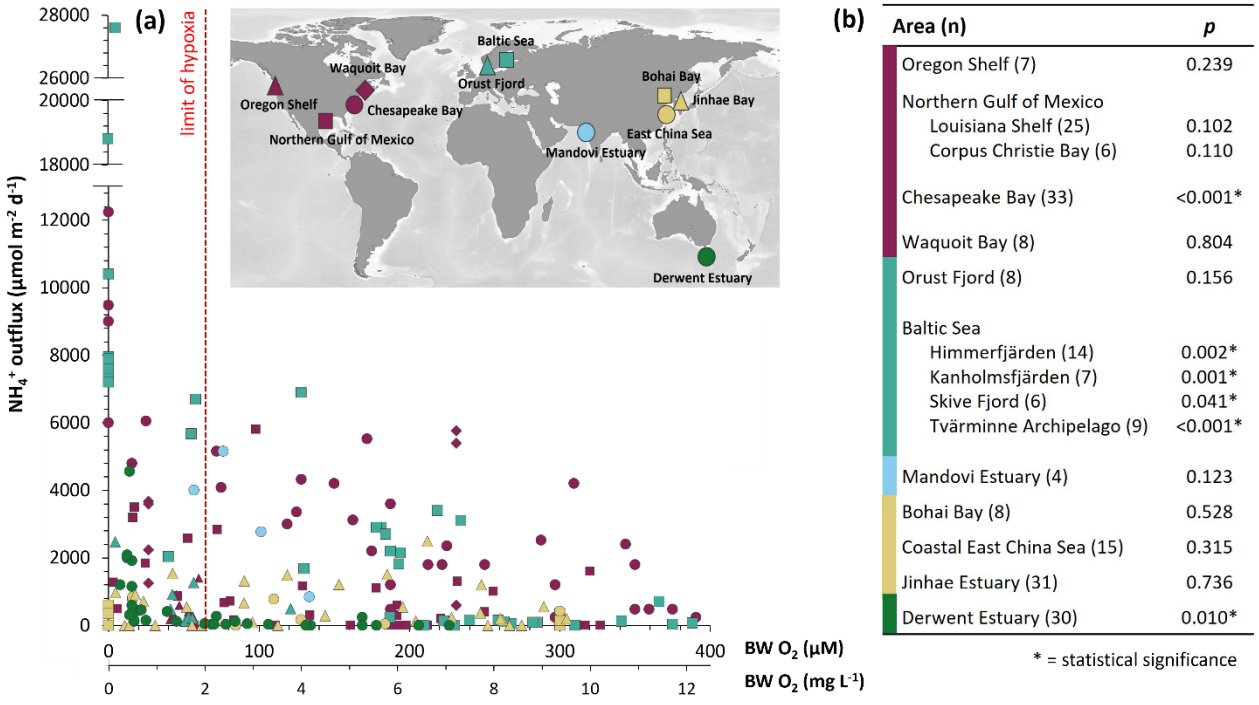


Figure 1. (a): Sediment-water ~~Benthic~~ ammonium (NH_4^+) efflux at different bottom water oxygen (BW O_2) concentrations from coastal sites around the world, and **(b):** statistical significance ($p \leq 0.05$) of each site's relationship between NH_4^+ efflux and BW O_2 tested with linear regression 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 consistent across seasons (Fig. 2). In a first-order estimate, using coastal boundaries as defined above ~~as defined by the Water Framework Directive~~ (European Communities, 2003) ~~including Kattegat~~, coastal sediment classifications from~~according to~~ Al-Hamdani and Reker (2007), and example ~~0–20 cm~~ porewater ~~NH_4^+~~ profiles of NH_4^+ from a representative sandy (site VE15, ~~Vistula Estuary,~~ southern Bay of Gdansk, Thoms et al., 2018) and muddy (site Storfjärden, ~~Tvärminne Archipelago,~~ northern 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~~ benthic pool of NH_4^+~~bioavailable N~~ is not only larger 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 ~~a~~the coastal pelagic TN pool estimate ~~for the coastal Baltic Sea~~ from Lønborg and Markager (2021), but, furthermore, it is only a minimum estimate of the entire NH_4^+ ~~sediment~~ pool in the coastal sediments considering that concentrations continue to increase with depth below 20 cm.

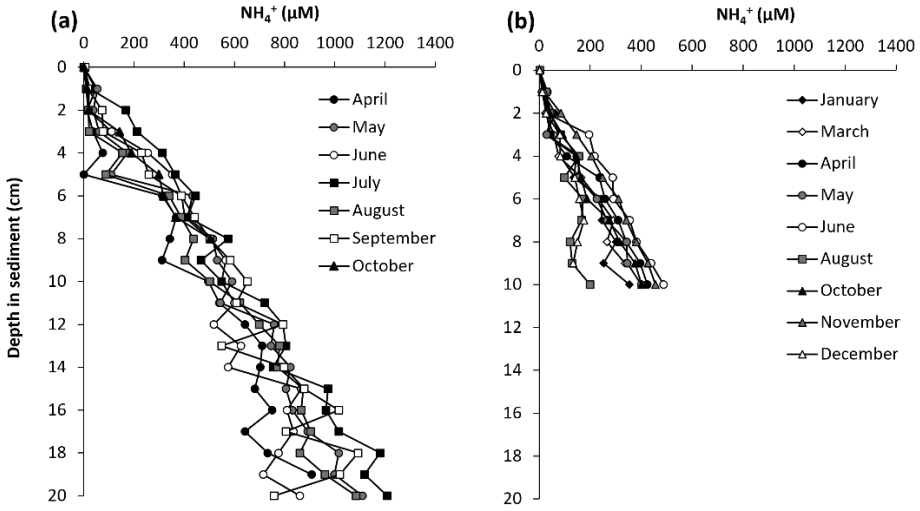


Figure 2. Example porewater profiles of ammonium (NH_4^+) in muddy sediments of the coastal Baltic Sea, taken from **(a)** site Storfjärden (33 m deep, ~~0–20 cm sediment depth,~~ April–October 2016) and **(b)** site Långholmsbranten (21 m deep, ~~0–10 cm sediment depth,~~ December 2022–November 2023), 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), and the coastal Luebeck ~~and Mecklenburg~~ Bight~~,~~ and Tromper Wiek, southwestern Arkona Basin (Gogina et al., 2018).~~-~~

The impact of this sediment 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, a substantial portion ~~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). The same ~~is~~ also happens in the numerous 3-dimensional oxic-anoxic interfaces created by benthic infauna (Andersen and Kristensen, 1991). Further, m~~-~~icrophytobenthos, if present on the sediment surface, can intercept and assimilate nutrients effluxing from the underlying sediments ~~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, for example, 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 sediment-water~~benthic~~ NH_4^+ efflux~~es~~ remains in the pelagic system and is thus available for coastal primary production, potentially enhancing coastal eutrophication.

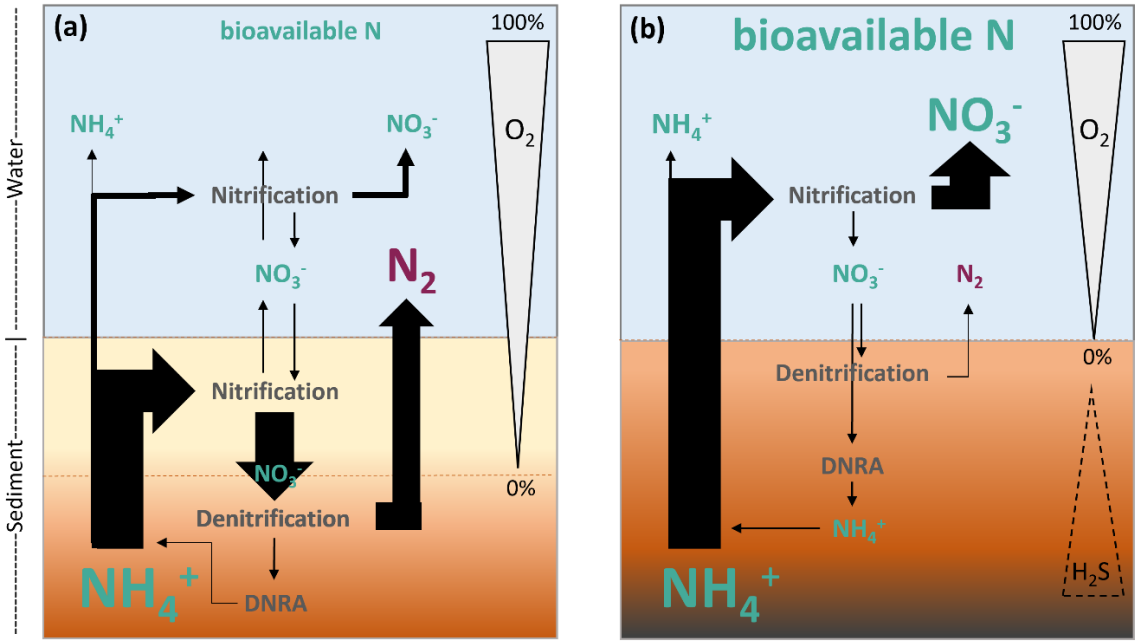


Figure 3. Conceptualized ammonium (NH_4^+) turnover in coastal sediments under **(a)** normoxic and **(b)** hypoxic conditions, based on Kemp et al. (1990), Jensen et al. (1990), Conley et al. (2007), Jäntti and Hietanen (2012) and others. Bioavailable nitrogen (N)-compounds shown in green, non-bioavailable N-compounds shown in violet. Arrow thickness symbolizes relative flux magnitude; font size symbolizes relative compound abundance. Anaerobic ammonium oxidation is not shown as it is often negligible in organic-rich coastal sediments (Dalsgaard et al., 2005). **(a) N~~n~~ormoxic 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) H~~h~~ypoxic conditions:** NH_4^+ flux across the sediment-water interface is large, as the oxic sediment layer is lost, disabling nitrification in the sediment and thus the transformation of upwards diffusing NH_4^+ to NO_3^- and its ~~the latter one's~~ subsequent removal in denitrification. ~~; 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. Some of this~~Only a small part of~~ water column NO_3^- can be ~~is assumed~~expected to diffuse back into the sediment~~s~~ where it can be removed in denitrification or transformed to NH_4^+ in dissimilatory nitrate reduction to ammonium (DNRA); however, being subject to a dynamic coastal system, the additional N might also be quickly exported to other water layers and, for instance, taken up in primary production. If hydrogen sulfide (H_2S) builds up in the surface sediment, DNRA commonly dominates over denitrification (Jäntti et al., 2021), further enriching the NH_4^+ pool.~~, 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 sediment ~~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 C~~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 C~~OM~~ m^{-2} d^{-1} in the 1990s, but still several-fold higher than before the onset of eutrophication. As a result, ~~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 sediment 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 sediment-water ~~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; Akçay et al., 2025) 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).

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~~ sediments of 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 M~~The m~~odel set-up

The 1-dimensional RTM is developed based on the R package "ReacTran" (Soetaert and Meysman, 2012) and comprises a multi-component diagenetic setting to examine sediment-water~~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 oxides, and sulfate (SO_4^{2-}), followed by methanogenesis when porewater 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. All reaction and transport parameters and ~~R~~reaction rates ~~, and reaction and transport parameters~~ are given in Appendix A (~~,~~Tables A2 and A3).~~=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 C 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 or is~~. OM is~~ removed from the ~~model~~ domain bottom by deep burial. For this, the model uses three OM pools: a reactive pool with a molar carbon/nitrogen (C/N) ratio of 6.6 representing marine-derived fresh OM (62% of deposited OM), a less reactive pool with a molar C/N ratio of 13 representing terrestrial-derived OM (15% of deposited OM), and a non-reactive pool that gets buried directly without degradation (23% of deposited OM), according to model calibration data from Jilbert et al. (2021; see also Appendix A). ~~molar (C/N)deposited molar C/Ndeposited deposited~~ For liquid phases ~~(porewater)~~, the upper boundary condition is set to be a constant bottom water concentration~~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 burial~~sedimentation~~. The ion concentrations of the bottom water are assumed to be constant and zero, ~~, i.e. most are zero~~ except for SO_4^{2-} (5 mmol L^{-1}), NO_3^- (0.007~~1~~ mmol L^{-1}) and O_2 (0, 0.063, and 0.2 mmol L^{-1}~~, depending on the scenario run~~, equivalent to anoxia, hypoxia, and normoxia; Table 1). Together, th~~is~~ set-up allows to investigate how the sediment system responds to changes in OM loading, which in turn regulate~~s~~ the sediment~~sedimentary~~ NH_4^+ pool and the sediment-water NH_4^+ efflux.

The RTM runs on a ~~runs on a~~ 1 ~~-~~cm depth resolution ~~and a default 1 year time step. It is designed for~~ and uses sediment porosities representative of ~~-~~muddy, OM-rich accumulation bottoms, which, ~~is~~ with ~41 %, is the predominant bottom type of the coastal Baltic Sea (calculated using Al-Hamdani and Reker (2007) and coastal boundaries ~~f~~according to ~~ollowing the Water Framework Directive including Kattegat (~~European Communities (~~,~~2003))~~)~~. The RTM output is ~~Model output is~~ given in 1-year time steps.

To calibrate~~verify~~ and verify the model, we used the geochemical porewater profiles and environmental conditions, such as sedimentation rate, nutrient concentrations, and salinity, ~~and geochemical porewater profiles~~ of the coastal site "Storfjärden"

in the ;—north-western Gulf of Finland (Jilbert et al. 20212018, Appendix AB, Fig. AB1-A3, Table 1 doi.org/10.5281/zenodo.17082148). , doi.org/10.5281/zenodo.22120110).

2.2 Scenario modelling

The OM loading used in the RTM is generated from the BALTSEM model (Gustafsson et al., 2012) covers a period from 1850 to 2400. Initially, the sediment modelRTM is run into a steady state using pre-industrial levels of OM flux to the sediment surface as a baseline prior to eutrophication (period 1800 to 1850). Then, hindcast an OM sedimentation rates overloading dataset areis used as a forcing for the period from 1850 to 2020, followed by two future scenarios from 2020 to 2400: In thea "Present scenario", that keeps the OM loading is held rate as a constant at the of the value of the year 2020 level, and in thea "Pre-industrial scenario", that reduces OM loading is reduced in accordance with the BSAP target until 2100, and subsequently continues to decline continues to further reduce OM loading beyond 2100 until pre-industrial levels are reached again around 2160 (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 (Murray et al., 2019), with the later situation when true pre-industrial conditions are re-established. Each scenario is run for three classes of bottom water O_2 availabilitiesconditions (normoxia, hypoxia, and anoxia; Table 1), representing best and worst case O_2 conditions. To account for the effects of bioturbating infauna on OM burial and O_2 transport during normoxia, we use the empirical parameter D_b (Boudreau 1997; Appendix A, Table A2) and furthermore force O_2 transport by bioturbating infauna during normoxia, O_2 to penetrate 2 cm deep into the sediment and subsequently to decrease exponentially to 0 µM at 5 cm depth. 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. The model code for all scenarios and simulations can be found at–, doi.org/10.5281/zenodo.22120110.

Table 1. Key input variables and scenario assumptions used in the RTM. reactive transport modelSalinity, sulfate, and sedimentation rate are adopted from the coastal site "Storfjärden", north-western Gulf of Finland. ; with OM = organic matter, C = carbon, O_2 = oxygen, and BSAP = Baltic Sea Action Plan.

OM loading scenarios	**"Present scenario"** OM loading is held is kept constant aton the level of year the value of year 2020 level (10338381 µmol C m^{-2} d^{-1})	**"Pre-industrial scenario"** OM loading is reduced according to following BSAP targets until 2100, and subsequently furtherthen further reduced until reaching to and continues until reaching pre-industrial levels (3719 µmol C m^{-2} d^{-1})

Bottom water O_2 conditions	Normoxia (200 µM)	Hypoxia (63 µM)	Anoxia (0 µM)	Normoxia (200 µM)	Hypoxia (63 µM)	Anoxia —(0 µM)
Salinity	6~~7~~					
Sulfate (mM)	5					
Sedimentation rate (mm yr^{-1})	5					
OM loading data	BALTSEM model (Gustafsson et al. 2012)					

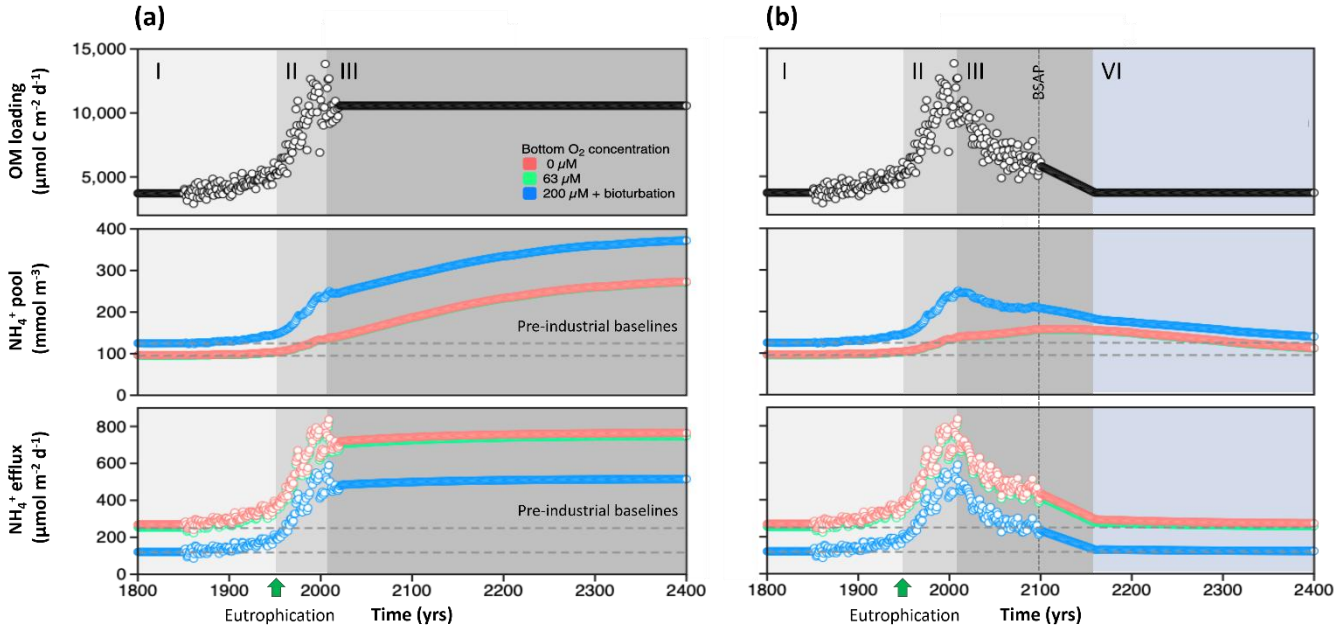
Due to ongoing climate change, bottom water temperatures will likely rise over the modelled future period and affect reaction rates and resulting NH_4^+ effluxes (Boynton et al. 2017), as heterotrophic metabolic processes are expected to increase at twice the rate of autotrophic production (Harris et 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 further 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 example along water depth gradients or seasons, 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-eutrophication~~industrial~~ OM level (including pre-industrial baseline levels) 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 C~~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 level~~value~~s (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.



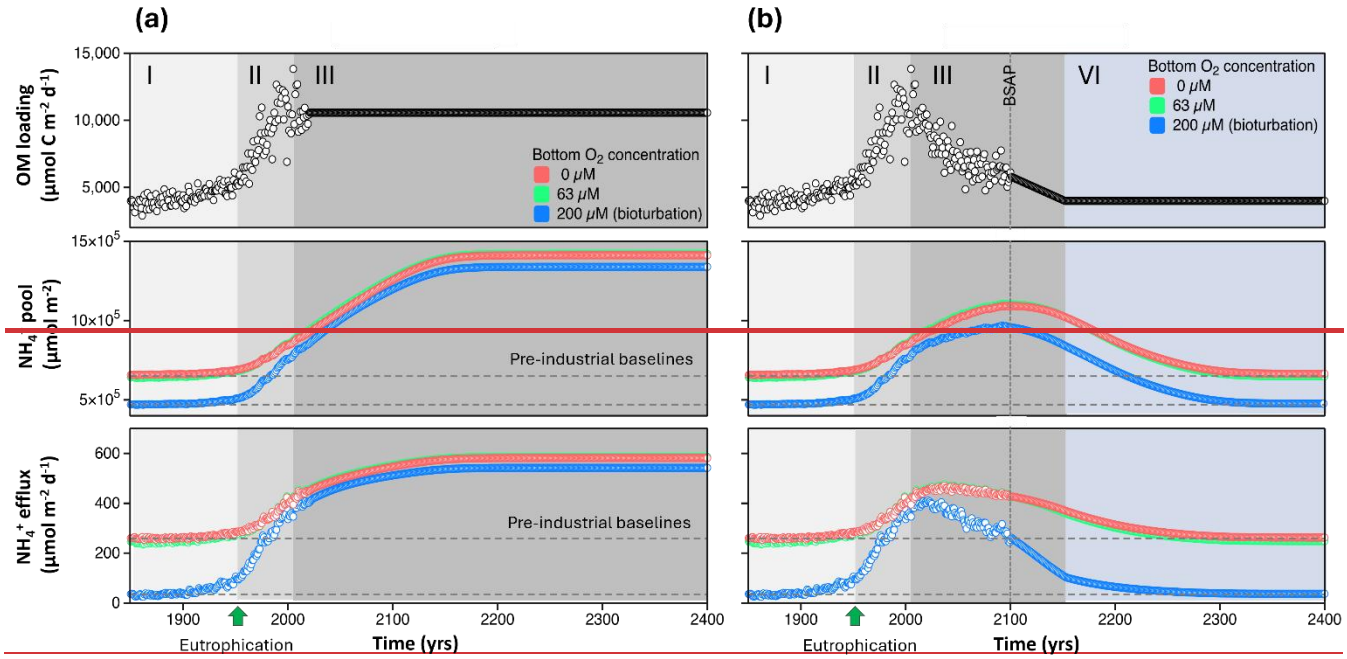


Figure 4. Sediment organic matter (OM) loading (µmol C m^{-2} d^{-1}), sediment ammonium (NH_4^+) pool (mmol m^{-3} bulk sediments), and sediment-water NH_4^+ efflux (µmol m^{-2} d^{-1}) in the northern coastal Baltic Sea under anoxic (red), hypoxic (green) and normoxic (blue) bottom water oxygen (O_2) conditions, modelled for the OM loading scenarios **(a)** "Present scenario" (constant OM loading) and **(b)** "Pre-industrial scenario" (reduced OM loading down to pre-industrial levels). Normoxic O_2 conditions include bioturbation, which is absent under O_2-deficient conditions as the benthic infauna dies. Modelling period 1805 to 2400. The NH_4^+ pool is the ~~annual~~ amount of porewater NH_4^+ integrated over a 1-meter sediment model domain. Grey shadings indicate the different phases of OM loading with (I) pre-eutrophication~~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 deficient 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. Please note, that the hypoxic NH_4^+ pool differentiates very little from the anoxic NH_4^+ pool and therefore is only marginally visible.

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~~ to the fast increase in OM loading. In phase III of the Present scenario (Fig. 4a), the OM loading becomes ~~decreases~~ constant in 2020, while the NH_4^+ pool continues to increase steadily for over two centuries before approximating a slight plateau around 2250; in contrast, NH_4^+ effluxes reach an actual plateau already around 2100 and thereafter stay high indefinitely under all O_2 conditions. ~~to varying degrees, while both the pool and~~

~~the efflux of NH_4^+ continue to increase, indicating a legacy effect of eutrophication.~~ In phase III of the Pre-industrial scenario (Fig. 4b), the NH_4^+ pool under normoxic conditions decreases in accordance with the drastic decrease in OM loading, albeit at a comparatively much slower rate: when OM loading reaches pre-industrial baseline values around 2160, the normoxic NH_4^+ pool is still as large as in the 1970s. In contrast, the NH_4^+ pool under O_2-deficient conditions continues to increase for about 100 years despite the reductions in OM loading, before eventually starting to slowly decrease. Under neither O_2 condition does the NH_4^+ pool of the Pre-industrial scenario reach pre-industrial baseline levels by year 2400, the end of our modelling period (Fig. 4b phase IV). Despite the difference in pool development, NH_4^+ effluxes in phase III of the Pre-industrial scenario follow the reduction of OM loading closely under all O_2 conditions (Fig. 4b). Nevertheless, NH_4^+ effluxes do not reach pre-industrial baseline levels by the end of phase III, but show remaining efflux-"tails" as can be seen when zooming into the model-output (Appendix B, Fig. B1). True pre-industrial baseline levels of NH_4^+ efflux are reached in phase IV first after 2300 (normoxic conditions) or even around 2400 (anoxic conditions; Appendix B, Fig. B1); the flux magnitude of these "tails" under all O_2 conditions is however small and comparable to the pre-eutrophication period (Appendix B, Fig. B1). In both scenarios, the NH_4^+ pool is smaller and increases at a lower rate under O_2-deficient than under normoxic conditions, whereas the NH_4^+ effluxes are larger under O_2-deficient than under normoxic conditions (Fig. 4). The latter one is in line with field data from the coastal Baltic Sea (Fig. 1) and the common understanding of more efficient internal NH_4^+ turnover under normoxic conditions (Fig. 3).

Depending on the O_2 condition, the NH_4^+ effluxes of the Pre-industrial scenario remain elevated compared to pre-industrial baseline levels for 280 to 380 years after the start of drastic OM load reductions (Appendix B, Fig. B1). However, already about 110 years following the reductions, i.e. in 2130, normoxic NH_4^+ effluxes have shrunk to a level comparable to the 1950s, the starting point of wide-spread eutrophication; under O_2-deficient conditions, this level will be reached about 140 years following the reductions, i.e. in 2160 (Fig. 4b). This modelled timeframe of decline in NH_4^+ efflux is 30–60 years longer than the estimate of when BSAP-driven good environmental status is assumed to be reached (year 2100, Fig. 4; Murray et al. 2019), which could suggest that it might take longer than previously thought for the BSAP to achieve its environmental targets.

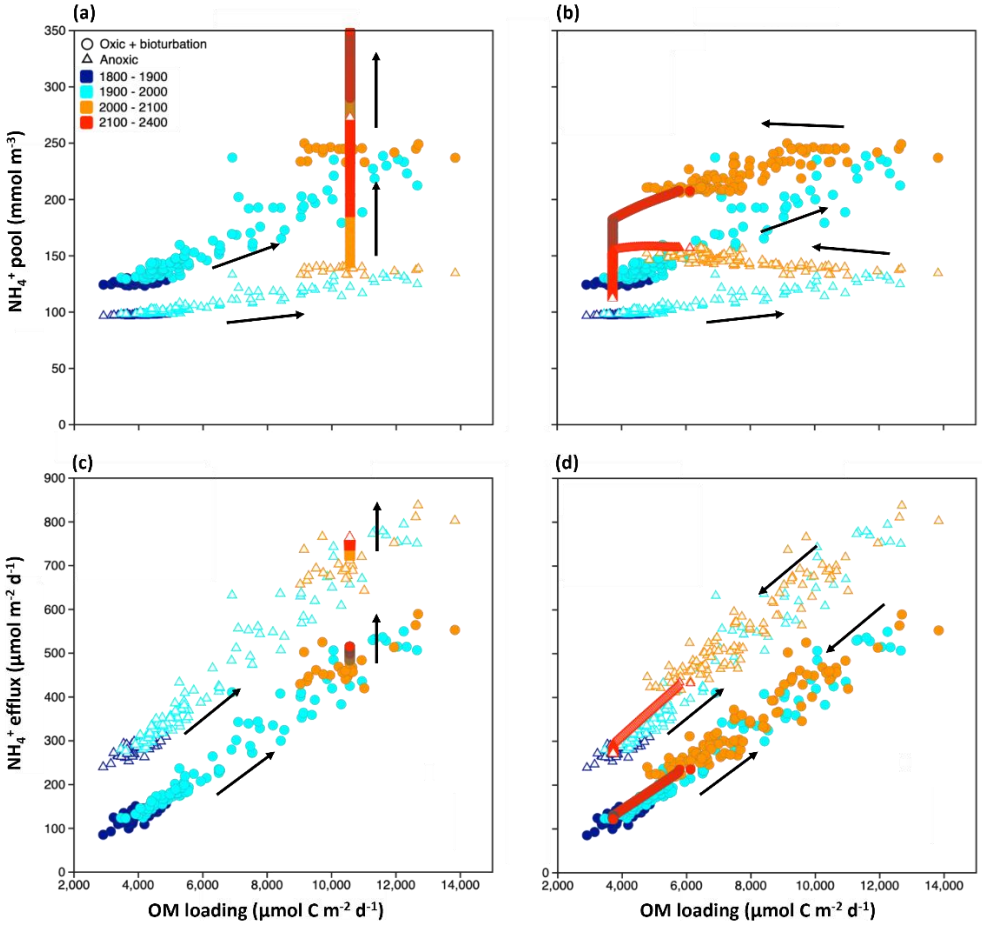


Figure 5. Sediment ammonium (NH_4^+) pool **(a, b)** and efflux **(c, d)** plotted against organic matter (OM) loading under two conditions of bottom water oxygen (O_2) and its consequential absence/presence of benthic fauna (triangles = 0 µM O_2; circles = 200 µM O_2 and bioturbation), shown for the Present scenario **(a, c)** and the Pre-industrial scenario **(b, d)**. Arrows and color-coding indicate the temporal evolution, with dark blue = period 1800-1900, light blue = period 1900-2000, orange = period 2000-2100, and red = period 2100-2400.

~~.~~ NH_4^+ effluxes in both scenarios seem to respond more rapidly to the external OM loading than to the internal NH_4^+ pool (Fig. 4, Fig. 5b, d). This is interesting, as the NH_4^+ pool shows a strong legacy effect and thus creates high internal NH_4^+ availability, even long after strong external OM loading has ceased, such as in the Pre-industrial scenario (Fig. 5b). This legacy effect is caused by NH_4^+ being continuously regenerated from the OM that accumulate in the sediment over subsequent years of strong eutrophication (Fig. 4, phase II), and results in (i) the increase of the NH_4^+ pool in the Present scenario, despite constant OM loading (Fig. 4a, phase III), as well as (ii) the slow decline (normoxic conditions) and initial increase (O_2-deficient conditions) of the NH_4^+ pool in the Pre-industrial scenario, despite rapid OM load reductions (Fig. 4b, phase III-IV). The different evolution of the normoxic and O_2-deficiant NH_4^+ pools can be attributed to the more efficient mineralization and

internal NH_4^+ turnover under normoxic than O_2-deficiant conditions (Fig. 3). The influence of the NH_4^+ pool on the NH_4^+ effluxes eventually becomes visible in the Pre-industrial scenario, where the NH_4^+ effluxes remain 5−10% above pre-industrial baseline values after OM loading has returned to constant baseline levels (Appendix B, Fig. B1, Fig. 4b), creating NH_4^+ efflux-"tails". While these tails are small in magnitude, they last for 140 to 240 years, depending on the O_2 condition (Appendix B, Fig. B1), clearly indicating the influence of the NH_4^+ pool and its strong legacy effect. In consequence, also the NH_4^+ effluxes are subject to a eutrophication legacy effect, which is highly persistent, but only small in effect size. Hence, the majority of the NH_4^+ effluxes remain controlled by the external OM loading, which suggests that NH_4^+ effluxes and their subsequent impact on the coastal environment can be mitigated rather promptly by reducing the external OM loading, such as in our Pre-industrial scenario.

While our modelled NH_4^+ effluxes in both scenarios show distinctly different responses between normoxic and O_2-deficient conditions, confirming the common assumption of O_2 as a key control of sediment-water NH_4^+ effluxes (Fig. 3), the magnitude of our modelled O_2-deficient fluxes is below the range of field-observations from the coastal Baltic Sea (Fig. 1). This underestimation of O_2-deficient~~anoxic~~ NH_4^+ effluxes can be explained by the resolution of our RTM, which runs on an annual basis and thus does not capture seasonal fluctuations and anoxic extreme events that can drastically increase the efflux. Such events are generally challenging to incorporate in a model, as they originate from individual combinations of OM loading and loading history, anoxia duration, temperature, and more. Without the addition of extreme values to the RTM's annual average, the modelled O_2-deficient NH_4^+ effluxes are lower than they can periodically be in nature, and our model consequently~~Therefore, our model~~ provides an estimate of O_2-deficient NH_4 effluxes at the~~a~~ lower range of their~~its~~ annual value. This underrepresentation of seasonal extreme values in our model affects only the O_2-deficient, but not the normoxic fluxes, which fit well to the range observed in the coastal Baltic Sea (Fig. 1). ~~Therefore, our model provides an estimate of NH_4 efflux at a lower range of its annual value.~~

~~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 that accumulated in the sediment 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.

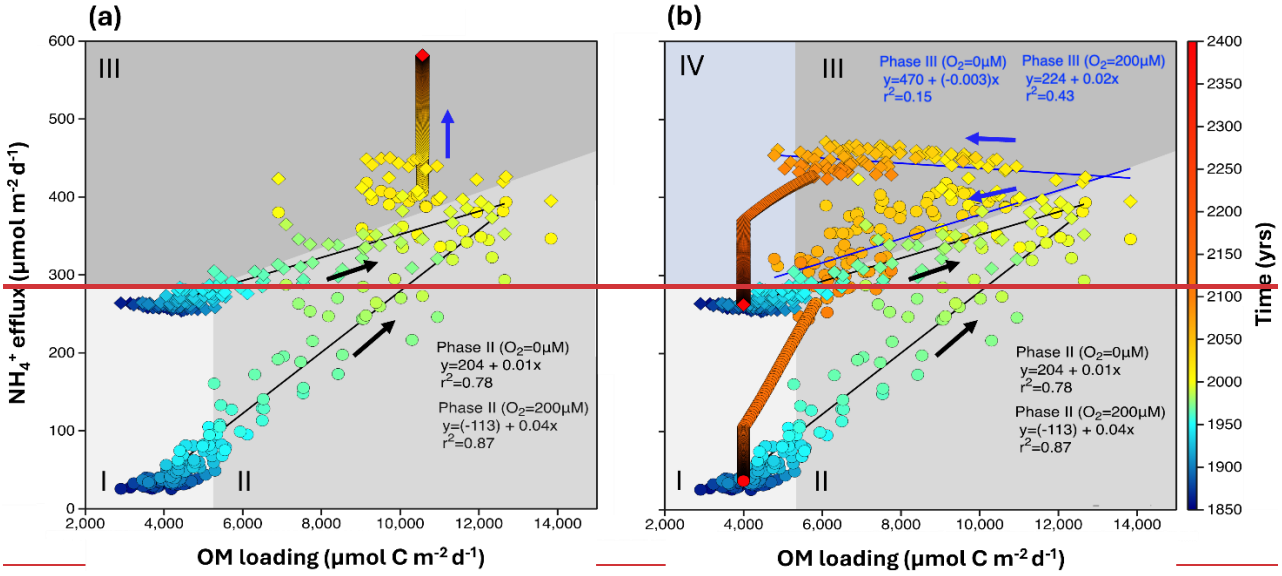


Figure 5. Sediment ammonium (NH_4^+) efflux plotted against organic matter (OM) loading under two conditions of bottom water oxygen (diamonds = 0 µM; circles = 200 µM O_2), shown for the (**A**) Present scenario and (**B**) Pre-industrial scenario of OM loading. Grey shading indicates the different phases 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 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).

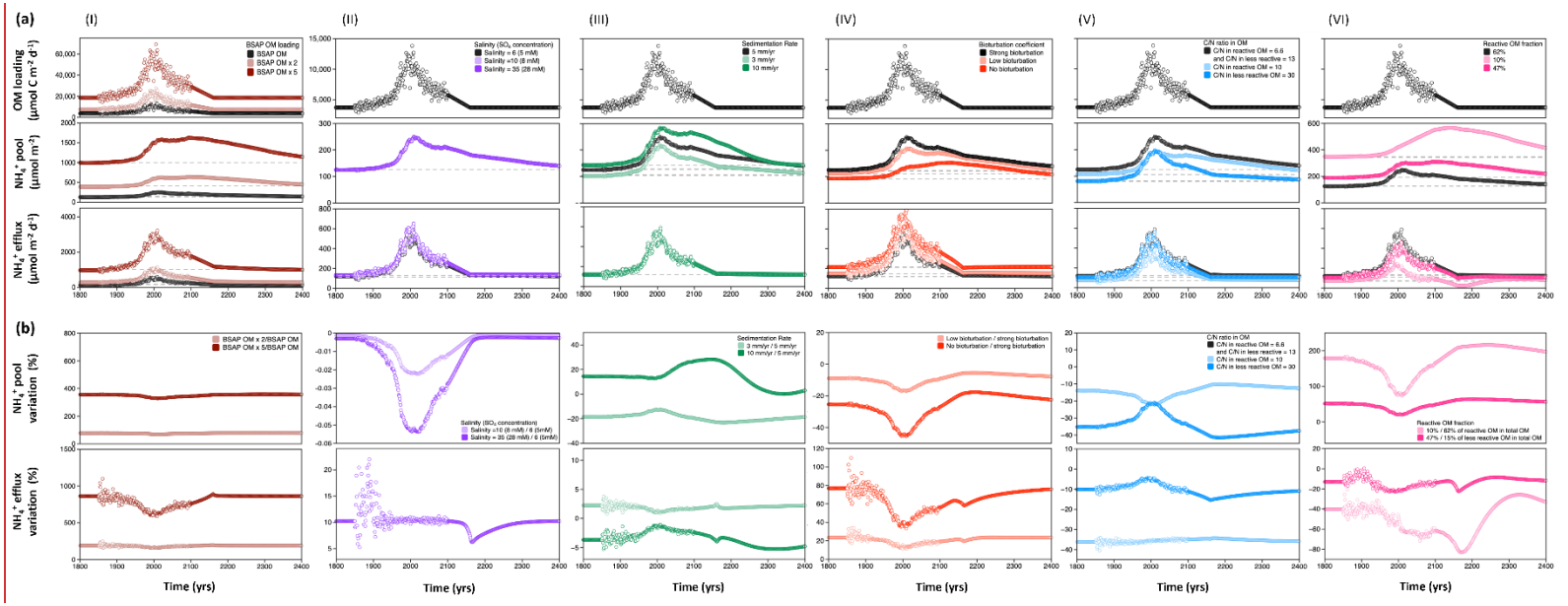
4 Sensitivity of ~~the~~ benthic NH_4^+ turnover~~legacy pool~~ to changed boundary conditions

Our model results ~~model~~ represent ~~is calibrated~~ one site in the coastal northern Baltic Sea, which is considered representative for a muddy, OM-rich accumulation bottom in low-saline, eutrophic conditions. To test the sensitivity of our modelled NH_4^+ pool and efflux to different ~~marine~~ environmental conditions, we applied a range of OM loadings, salinities (representative of different SO_4^{2-} concentrations in bottom water), ~~OM loadings, and~~ sedimentation rates, bioturbation coefficients, ~~and~~ C/N ratios of OM, and different shares of reactive material in OM to our RTM, both under normoxic (Fig. 6) and anoxic (Appendix B~~D~~, Fig. B2~~D1~~) bottom water conditions. ~~As~~ As a baseline for comparison, we used the conditions of our model setting under the Pre-industrial OM loading scenario, i.e. ~~, using the northern coastal Baltic Sea as a baseline (~~a 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}, strong bioturbation under normoxic conditions, and a C/N ratio of 6.6 in the reactive fraction and 13~~0~~ in the less reactive fraction. ~~).~~ 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 OM loading would strongly increase the NH_4^+ pool and efflux and both their legacy effects (Fig. 6 I). While the increase in legacy effect in the NH_4^+ pool is very visible, its increase in the NH_4^+ efflux is more subtle in larger flux magnitudes of the efflux-"tails" (Fig. 6 I-a), proportionally to the level of OM loading. The highest OM loading (5-times the BSAP OM loading) shows the strongest effect on the NH_4^+ effluxes (Fig. 6 I-b) ~~is ef~~. An increase in salinity does not change the size of the NH_4^+ pool and only marginally increases the NH_4^+ efflux (Fig. 6 IIa), with the largest change observed when salinity changes from 6.6 to 10 (Fig. 6 IIb). This result is caused by the comparatively low availability of OM that limits the reduction of SO_4^{2-}; hence, a higher salinity can only increase the NH_4^+ pool and efflux, if also the OM availability is higher, such as can be expected under the Present scenario. A change in sedimentation rate affects the size of the NH_4^+ pool but not the NH_4^+ efflux (Fig. 6 III). The presence or absence of bioturbation has a strong effect on both NH_4^+ pool and efflux (Fig. 6 IV). In the presence of bioturbation, the NH_4^+ pool increases, likely because bioturbation promotes the degradation of OM (Andersen and Kristensen, 1991) and thus the release of NH_4^+ to the sediment porewater. However, at the same time, the NH_4^+ efflux decreases, as bioturbation generally increases the availability of O_2 in surface sediments, thereby facilitating the sediment internal NH_4^+ turnover (Fig. 3). If bioturbation is absent, the opposite happens. This result highlights the importance of bioturbating infauna for sediment O_2 and N-turnover. An increase in C/N ratio towards less bioavailable and more refractory OM will decrease both the NH_4^+ pool and efflux (Fig. 6 V), as the OM becomes less rich in N and thus less NH_4^+ is produced in mineralization. Different shares of reactive OM also play a role in regulating the NH_4^+ pool and efflux by impacting the C/N ratios used in the model (Fig. 6 VI). In this test, the share of refractory OM is kept at 23%, and the share of reactive OM is 47% and 10%. Fig. 6 VI shows that lower shares of reactive OM lead to a larger NH_4^+ pool and a lower NH_4^+efflux, which is caused by higher shares of less reactive OM pool that degrade slowly but last longer in the sediments. ~~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).~~ 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- and efflux (Appendix B~~D~~, Fig. B2~~D1~~).

While these results point to a general direction for NH_4^+ cycling in eutrophied coastal systems of different environmental conditions, they need to be interpreted while considering the geomorphological context for each system and the understanding that parameters occur in clusters not in isolation. For instance, high salinity might often occur together with a low sedimentation rate, given that many coastal systems of high salinity are connected to the open ocean and thus experience high water energy from waves and tidal force, which enhances dispersion of OM and ultimately affects the sedimentation rate. In our sensitivity analysis, we do not consider such parameter combinations. Hence, understanding a habitat's geomorphological setting and environmental conditions are highly important when aiming to realistically model NH_4^+ flux predications of coastal sediments.


(a)
(I)
(II)
(III)
(IV)
(V)
(VI)
OM loading (µmol C m⁻² d⁻¹)
NH4+ pool (µmol m⁻²)
NH4+ efflux (µmol m⁻² d⁻¹)
BSAP OM loading
BSAP OM
BSAP OM x 2
BSAP OM x 5
Salinity (SO4 concentration)
Salinity = 6 (5 mM)
Salinity =10 (8 mM)
Salinity = 35 (28 mM)
Sedimentation Rate
5 mm/yr
3 mm/yr
10 mm/yr
Bioturbation coefficient
Strong bioturbation
Low bioturbation
No bioturbation
C/N ratio in OM
C/N in reactive OM = 6.6 and C/N in less reactive = 13
C/N in reactive OM = 10
C/N in less reactive OM = 30
Reactive OM fraction
62%
10%
47%
(b)
NH4+ pool variation (%)
NH4+ efflux variation (%)
BSAP OM x 2/BSAP OM
BSAP OM x 5/BSAP OM
Salinity (SO4 concentration)
Salinity =16 (8 mM) / 6 (5mM)
Salinity = 35 (28 mM) / 6 (5mM)
Sedimentation Rate
3 mm/yr / 5 mm/yr
10 mm/yr / 5 mm/yr
Low bioturbation / strong bioturbation
No bioturbation / strong bioturbation
C/N ratio in OM
C/N in reactive OM = 6.6 and C/N in less reactive = 13
C/N in reactive OM = 10
C/N in less reactive OM = 30
Reactive OM fraction
10% / 62% of reactive OM in total OM
47% / 15% of less reactive OM in total OM
Time (yrs)

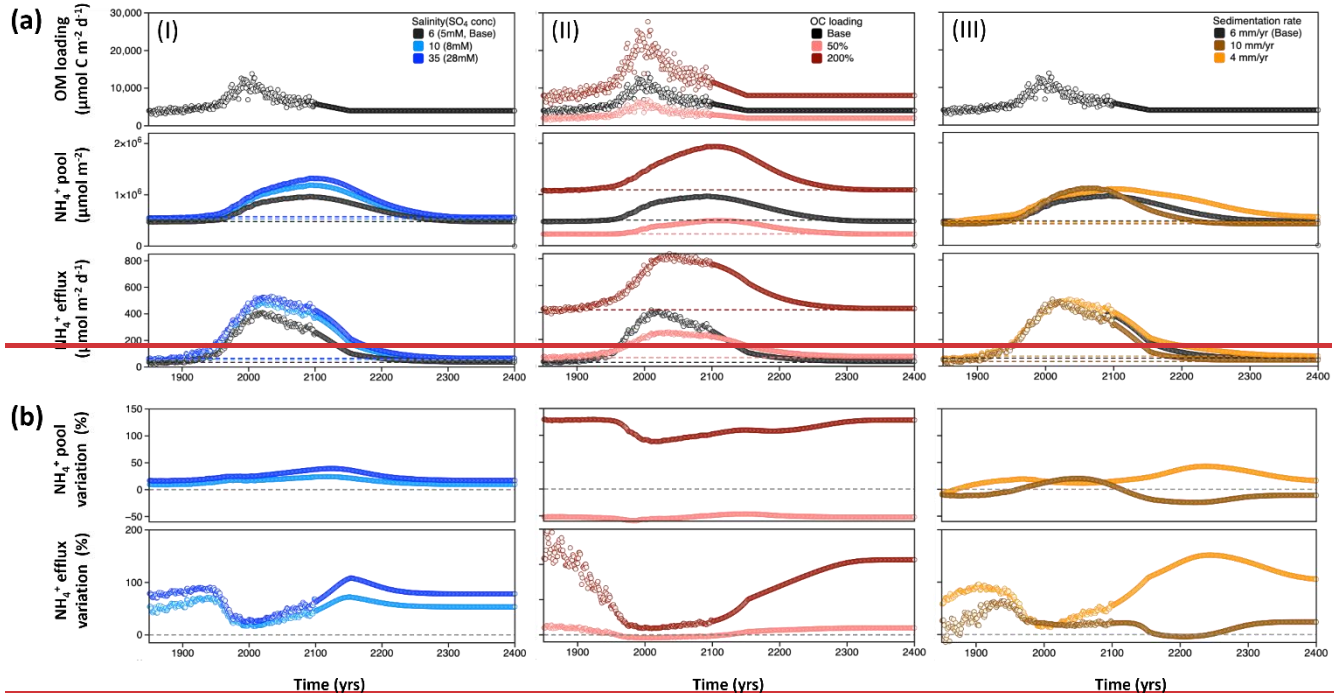


Figure 6. Effect of changed organic matter (OM) loading (I), salinity (II), sedimentation rate (III), bioturbation coefficient (IV), carbon to nitrogen (C/N) ratio in OM (V)~~salinity (I), organic matter (OM) loading (II), and sedimentation rate (III)~~, and different shares of reactive material in OM (VI) compared to baseline conditions representative of the coastal northern Baltic Sea (~~i.e. salinity of 6,~~ Pre-industrial OM loading scenario, salinity of 6, sedimentation rate of 6 mm yr^{-1}, strong bioturbation, and OM with a C/N ratio of 6.6 (reactive fraction) and 13 (less reactive fraction);~~,~~ 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 O_2). Dashed lines give the pre-industrial baselines of NH_4^+ pool and efflux. Note the different y-axis scales in (a) of (I) and (VI) vs. (II−V), as well as in (b) between all parameters (I−VI).

~~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 habitat's 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 modelling results highlight~~highlight~~ the presence ~~persistence and thus importance~~ of legacy effects from past eutrophication in present-day ~~for present~~ and future NH_4^+~~nutrient~~ cycling ~~and corresponding ecosystem health~~ in coastal sediments of the ~~the coastal~~ Baltic Sea. While the legacy effects are highly persistent in both the sediment NH_4^+ pool and the sediment-water NH_4^+ effluxes, the legacy's effect size is much smaller in the NH_4^+ effluxes than in the NH_4^+ pool. This means, that the NH_4^+ pool affects the NH_4^+ effluxes for a long time, but potentially plays a minor role in its ecological impact. However, the impact of the NH_4^+ efflux legacy on the coastal ecosystem is ultimately determined by the external OM loading, which not only strongly drives the effluxes (Fig. 4b) but also ~~ultimately~~determines the magnitude and thus ecological significance of the legacy effluxes (Fig. 6 I). The strong connection between external OM loading and NH_4^+ effluxes gives a clear direction for the restoration of eutrophied coastal ecosystems. Our model results show that if strict OM load reductions are implemented, sediment-water NH_4^+ effluxes will decrease promptly and nearly proportionally to the load reductions. About 110 years after the start of OM load reductions, NH_4^+ effluxes could reach the same level as at the start of eutrophication in the 1950s. This is a relatively short time-frame for natural ecosystem recovery of a heavily eutrophied system, such as the coastal Baltic Sea. The time-frame can be met when bottom waters are normoxic and sediments are strongly bioturbated, meaning that the benthic system stays as healthy as possible during the duration of OM load reductions. If bottom waters become O_2-deficient and the bioturbating infauna dies, the duration to reach 1950s levels of NH_4^+ effluxes will be extended by approximately 30 years, during which also the magnitude of NH_4^+ effluxes will be higher than under normoxic conditions (Fig. 4b), thereby sustaining the strain on the ecosystem. Hence, the key factors affecting internal N loading via sediment-water NH_4^+ effluxes in the coastal Baltic Sea are the external OM loading and the bottom water O_2 conditions. Consequently, internal N loading and its impact on the coastal environment cannot be mitigated if OM loading remains on its present-day level (Fig. 4a).

The limited direct impact of the sediment NH_4^+ pool on the sediment-water NH_4^+ effluxes in our model is unexpected, but important knowledge for ecosystem management, as it suggests that ecosystem recovery in regard to N overloading is achievable within realistic time-frames. A stronger influence of the NH_4^+ pool on the NH_4^+ effluxes than what we see in our results would increase both the magnitude of the NH_4^+ effluxes and their resistance to external management measures, potentially extending their impact on the coastal ecosystem by many years. It is worth to keep in mind that, despite its limited direct impact, our modelled sediment NH_4^+ pool nevertheless maintains the long-lasting legacy effect of the NH_4^+ effluxes, whose magnitude and thus ecological impact can increase under O_2-deficient and OM enriched conditions (Appendix B, Fig. B2). The estimated duration of legacy effect in the NH_4^+ effluxes is similar to Lønborg and Markager (2021), who suggested a TN legacy in Baltic Sea sediments of several centuries.

Considering that our model 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, a large part of the N released to the water column via sediment-water NH_4^+ efflux would be taken up in primary production and eventually settle back to the sediment to become new OM, which adds complexity to the internal nutrient loops in both modelling 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 end up in the open Baltic Sea (Radtke et al., 2012), which increases the chance that the coastal N load is ultimately removed from the aquatic system via denitrification at the permanent redoxcline. How long it takes to remove the internal N loading from the coastal zone and thus from the potential to favour coastal primary production, remains, however, unclear. Furthermore, the impact of internal N loading on the coastal ecosystem may be 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 or anoxic events can lead to increases in N availability and subsequent primary production, yet, such increases are likely masked by the overall coastal eutrophication. Nevertheless, for management efforts to mitigate coastal eutrophication and restore a healthy ecosystem, every addition of biologically available nutrients that is not accounted for in mass balance budgets should be noteworthy. In the big picture, internal loading adds nutrients to a system and even if these nutrients are transient, they can cause ecological harm and impede eutrophication mitigation measures.

Following OM load reductions, NH_4^+ effluxes in our model decrease slightly faster and with a smaller flux magnitude under normoxic than under O_2-deficient conditions (Fig. 4b), which might raise the question whether artificial aeration methods could be a justifiable option to aid the ecosystem recovery. However, the effects of artificial aeration on benthic N-cycling in the coastal Baltic Sea are not straight forward and sediment-water NH_4^+ effluxes both did (site *Lännerstasundet*, Lehtoranta et al., 2022) and did not (site *By Fjord*, De Brabandere et al., 2015) significantly decrease under aeration, or even increased (site *Sandöfjärden*, Lehtoranta et al., 2022) due to aeration induced sediment warming and a return of anoxia despite aeration. Similar conflicting results were found also outside the coastal Baltic Sea (e.g. Harris et al., 2015, Testa et al., 2025), including increased air-water methane exchange under conditions of aeration (Lapham et al., 2022), which suggests that engineered aeration approaches often have unexpected and unintentional impacts on both nutrient cycling and greenhouse gas emissions.

Given these variable responses, we do not consider artificial aeration to be a clear-cut solution to shorten the duration of NH_4^+ effluxes in the coastal Baltic Sea.

The results suggest We demonstrate that the eutrophication-derived OM pool in the sediments of the coastal Baltic Sea is likely 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 deposited, new 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 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 may beis 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 isnot accounted for in mass balance 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 are transient 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 where and thus the a long timeframe it could "sabotage" mitigation efforts.~~

6 Conclusions

Muddy sediments in the coastal Baltic Sea commonly contain large NH_4^+ accumulations from past and ongoing eutrophication, which are favourably released to the water column under O_2-deficient conditions, resulting in internal N loading with the potential to facilitate coastal primary production. To better understand the fate and persistency of this NH_4^+ legacy and its impact on the coastal ecosystem, we modelled the past to future development of the sediment NH_4^+ pool and sediment-water NH_4^+ effluxes in the coastal Baltic Sea under two contrasting OM loading scenarios and three O_2 conditions. Our model results ~~clearly~~ clearly show a strong eutrophication legacy effect in the development of the sediment NH_4^+ pool, which affects the sediment-water NH_4^+ effluxes for a long time, but with only a mild effect size. T~~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~~ he key driver of the sediment-water NH_4^+ effluxes is the external OM loading, which continuously adds to the OM- and NH_4^+ accumulations in coastal surface sediments. This result strongly indicates, that present-day elevated sediment-water NH_4^+ effluxes can be effectively reduced if strict OM load reductions are realized, such as proposed by the BSAP. Oxic bottom water and strong bioturbation aid the reduction of NH_4^+ effluxes, as both aspects increase the sediment internal turnover of NH_4^+. However, these factors are secondary behind the reduction in OM loading: following our model results, internal N loading to the coastal Baltic ecosystem cannot be mitigated if OM loading continues as at present, which has been suggested repeatedly in other studies (Pitkänen et al. 2001, Saraiva et al. 2019). ~~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 ~~o~~ for other coastal areas of similar geomorphology as the coastal Baltic Sea that face similar challenges in nutrient reduction and ecological management.

Appendix A

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

Primary OM~~C~~ reactions:	
$OMC + aO_2 \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + aH_2O$	(R1)
$OMC + \frac{4a}{5}NO_3^- + \frac{4a}{5}H^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + \frac{2a}{5}N_2 + \frac{7a}{5}H_2O$	(R2)
$OMC + 2aMnO_2 + 4aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + 2aMn^{2+} + 3aH_2O$	(R3)
$OMC + 4aFe(OH)_3^{\alpha} + 12aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + 4aFe^{2+} + 13aH_2O$	(R4)
$OMC + \frac{a}{2}SO_4^{2-} + aH^+ \rightarrow aCO_2 + bNH_4^+ + cH_3PO_4 + \frac{a}{2}H_2S + aH_2O$	(R5)
$OMC \rightarrow \frac{a}{2}CO_2 + bNH_4^+ + cH_3PO_4 + \frac{a}{2}CH_4$	(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)

Partial differential equations.

~~Equations A1, A2.~~ We established a 1-dimensional RTM to simulate the variations of OM and porewater ions given in Table A1. The RTM was achieved by the ReacTran package in R language (Soetaert and Meysman, 2012) via the following partial differential equations:

$$\phi \cdot \frac{\partial C}{\partial t} = \frac{\partial(\phi \cdot (D_s + D_b) \cdot \frac{\partial C}{\partial x})}{\partial x} - \frac{\partial(\phi \cdot v \cdot C)}{\partial x} + \phi \cdot \sum R_1 \quad \text{(Eq. A1)}$$

$$(1-\phi) \cdot \frac{\partial S}{\partial t} = \frac{\partial((1-\phi) \cdot D_b \cdot \frac{\partial S}{\partial x})}{\partial x} - \frac{\partial((1-\phi) \cdot w \cdot S)}{\partial x} + (1-\phi) \cdot \sum R_2 \quad \text{(Eq. A2)}$$

where C denotes dissolved ion concentration in porewater (mmol L^{-1}), S represents OM contents, x is depth (cm below sea floor), and ∑R1 and ∑R2 define the net reaction rates in porewater and dry sediments, respectively. More parameters and definitions are given in Table A2.

Table A2. Reaction and transport parameters used in the RTM with; a) Boudreau 1997, b) Jilbert et al., 2021, and c) Reed et al., 2011. ~~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]	$\phi(x) = \phi_\infty + (\phi_0 - \phi_\infty)exp(-\max(0, x - x_0)/x_{att})$	$cm^3\ cm^{-3}$
Porosity at surface sediment[b]	$\phi_0 = 0.945$	$cm^3\ cm^{-3}$
Porosity at 1 meter[b]	$\phi_\infty = 0.86$	$cm^3\ cm^{-3}$
Depth to start exponential decrease	$x_0 = 2$	cm
Attenuation coefficient in exp. ~~onential~~ decrease	$x_{att} = 1$	cm
Solid volume fraction[b]	$\phi_S(x) = 1 - \phi$	$cm^3\ cm^{-3}$
Tortuosity[a]	$\theta^2(\phi) = 1 - 2\ ln\phi$	—
Sediment density[b]	$\rho_S = 2.4865$	$g\ cm^{-3}$
Sediment accumulation rate[b]	$v_0 F_{sed} = 0.56$	$cm\ yr^{-1}$
Burial velocity of porewater	$v = v_0 \frac{\Phi_{inf}}{\Phi}$	$cm\ yr^{-1}$
Burial velocity of OM	$w = v_0 \frac{1 - \Phi_{inf}}{1 - \Phi}$	$cm\ yr^{-1}$
Bioturbation~~diffusion~~	$D_b(x) = D_{b0} exp(-\max(0, x - x_0)/x_{att}))]$	$cm^2\ yr^{-1}$
Bioturbation~~diffusion~~ coefficient	$D_{b0} = 1500$	$cm^2\ yr^{-1}$
Limiting concentration of O_2[c~~b~~]	$k_{O_2} = 20$	$\mu mol\ L^{-1}$
Limiting concentration of NO_3^-[c~~b~~]	$k_{NO_3^-} = 24$	$\mu mol\ L^{-1}$
Limiting concentration of MnO_2[c~~b~~]	$k_{MnO_2} = 4$	$\mu mol\ g^{-1}$
Limiting concentration of $Fe(OH)_3$[c~~b~~]	$k_{Fe(OH)_3} = 65$	$\mu mol\ g^{-1}$
Limiting concentration of SO_4^{2-}[c~~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	$k_{R7} = 1500000$	$mmol^{-1}\ L\ yr^{-1}$

R8 reaction constant	$k_{R8} = 20$	mmol^{-1} L yr^{-1}
R9 reaction constant	$k_{R9} = 14300$	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	$k_{R13} = 1000$	mmol^{-1} L yr^{-1}
R14 reaction constant	$k_{R14} = 10$	mmol^{-1} L yr^{-1}

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Table A3. Rates of reactions given in Table A1~~Primary chemical reaction rates~~; i stands for the reactive or less reactive OM~~C~~ pools.

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

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

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

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

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

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

$$r_{R7} = k_{R7}[O_2][NH_4^+]$$

$$r_{R8} = k_{R8}[O_2][Mn^{2+}]$$

$$r_{R9} = k_{R9}[O_2][Fe^{2+}]$$

$$r_{R10} = k_{R10}[O_2][H_2S]$$

$$r_{R11} = k_{R11}[O_2][CH_4]$$

$$r_{R12} = k_{R12}[Fe(OH)_3][H_2S]$$

$$r_{R13} = k_{R13}[Fe^{2+}][H_2S]$$

$$r_{R14} = k_{R14}[SO_4^{2-}][CH_4]$$

~~**Table A4.** Secondary chemical reaction rates.~~

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

~~A1.~~ RTM~~Model~~ calibration and validation.~~

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The RTM is calibrated using the sediment property and geochemical porewater data of the coastal site Storfjärden, north-western Gulf of Finland, Baltic Sea (Jilbert et al., 2021). The included reactions and parameters are given in Tables A1-A3. The following Figures A1-A3 show the model outputs fitted on the measured sediment profiles.

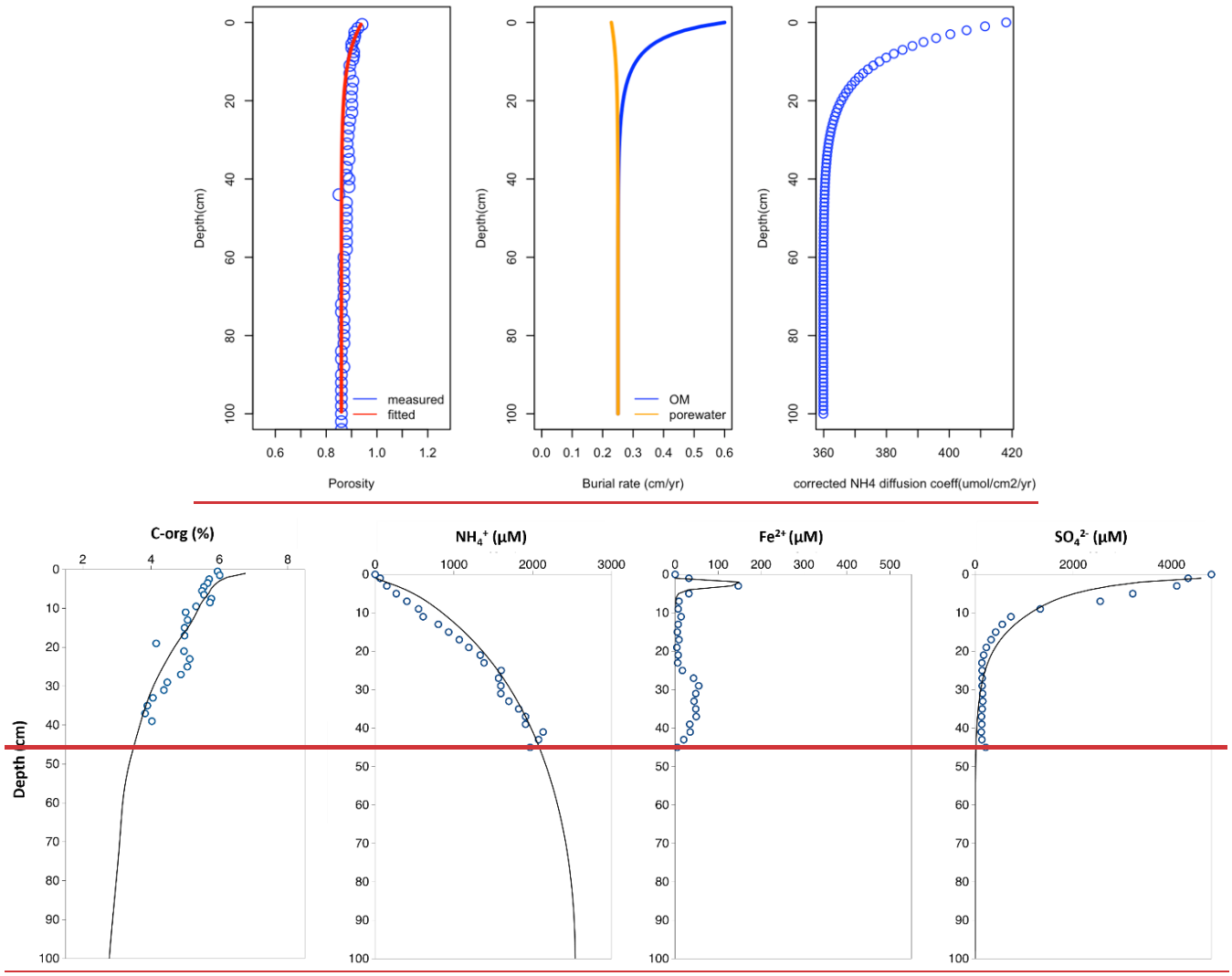


Figure A~~B~~1. The fit of porosity variation with depth, the calculation of burial rates of OM and porewater, and the effective NH_4^+ diffusion coefficient corrected by tortuosity.

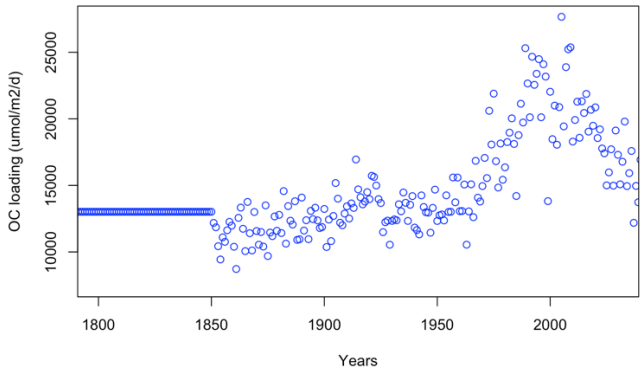


Figure A2. The prescribed OM loading onto surface sediments (3-4 times BALTSEM output) in order to fit the measured OM profile of the coastal site "Storfjärden" by Jilbert et al (2021), as shown in Figure A3.

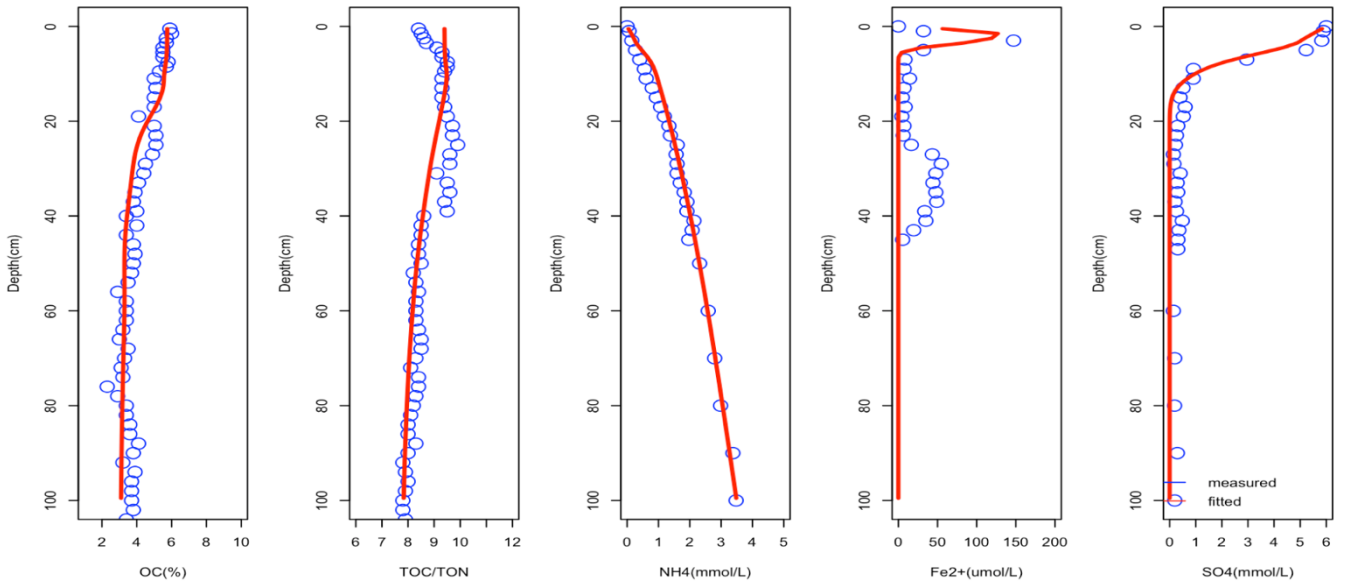
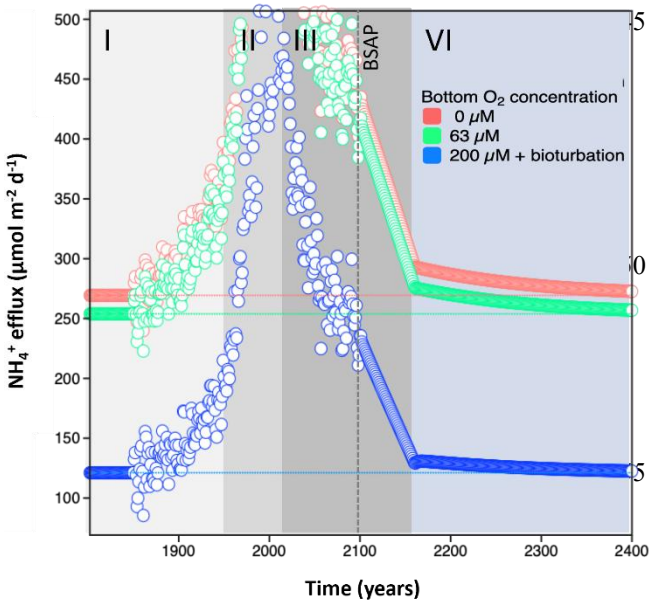


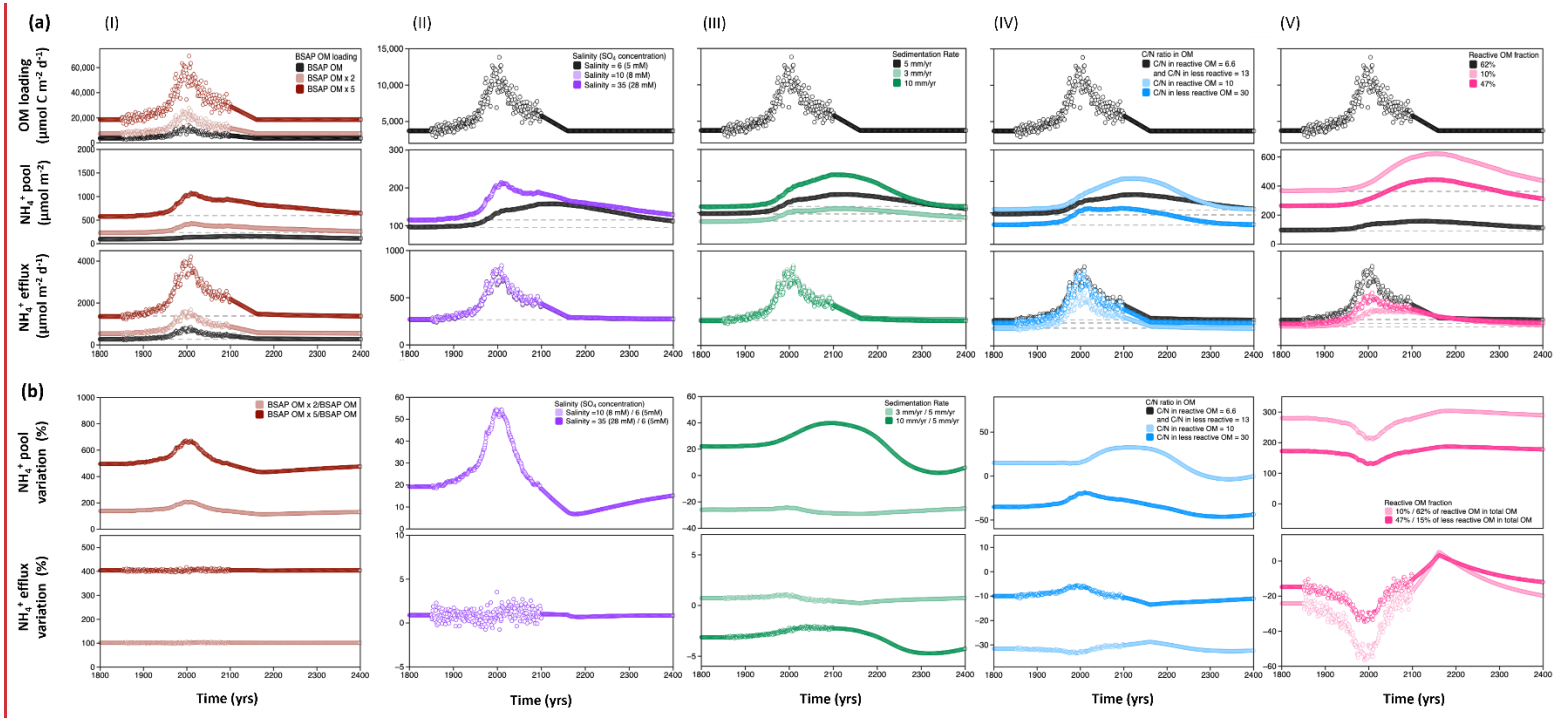
Figure A3. Sediment and porewater profiles of major model parameters, measured at the coastal site "Storfjärden", north-western Gulf of Finland (open blue circles; Jilbert et al., 2021), and modelled results of those parameters (solid red line); with OC = organic carbon content, TOC/TON = total organic carbon to nitrogen ratio, NH_4^+ = ammonium, Fe^{2+} = iron(II), and SO_4^{2-} = sulfate.

~~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.~~



Appendix B

Figure B1. Zoom into the normoxic, hypoxic, and anoxic NH_4^+ effluxes along time of the Pre-industrial scenario, ~~showing~~showing ~~an~~ efflux-"tails" starting from circa~~round~~ 2160 when OM loading reaches pre-industrial baseline values. These NH_4^+ efflux-"tails" are indicative of a eutrophication legacy effect. Due to the effect, true baseline values of NH_4^+ efflux are first reached after 2300, depending on the O_2 conditions. ~~The magnitude of the efflux "tails" is, however, likely ecologically negligible.~~ Grey shadings indicate the different phases of OM loading with (I) pre-eutrophication, (II) strongly increasing, (III) after-peak, and (VI) return to pre-industrial, in line with Fig. 4. The vertical dashed line gives the end point of OM load reductions if following the target of the Baltic Sea Action Plan (BSAP, Table 1).


(a)
(I)
(II)
(III)
(IV)
(V)
OM loading (µmol C m-2 d-1)
NH4+ pool (µmol m-2)
NH4+ efflux (µmol m-2 d-1)
BSAP OM loading
BSAP OM
BSAP OM x 2
BSAP OM x 5
Salinity (SO4 concentration)
Salinity = 6 (5 mM)
Salinity =10 (8 mM)
Salinity = 35 (28 mM)
Sedimentation Rate
5 mm/yr
3 mm/yr
10 mm/yr
C/N ratio in OM
C/N in reactive OM = 6.6 and C/N in less reactive = 13
C/N in reactive OM = 10
C/N in less reactive OM = 30
Reactive OM fraction
62%
10%
47%
(b)
NH4+ pool variation (%)
NH4+ efflux variation (%)
BSAP OM x 2/BSAP OM
BSAP OM x 5/BSAP OM
Salinity (SO4 concentration)
Salinity =10 (8 mM) / 6 (5mM)
Salinity = 35 (28 mM) / 6 (5mM)
Sedimentation Rate
3 mm/yr / 5 mm/yr
10 mm/yr / 5 mm/yr
C/N ratio in OM
C/N in reactive OM = 6.6 and C/N in less reactive = 13
C/N in reactive OM = 10
C/N in less reactive OM = 30
Reactive OM fraction
10% / 62% of reactive OM in total OM
47% / 15% of less reactive OM in total OM
Time (yrs)

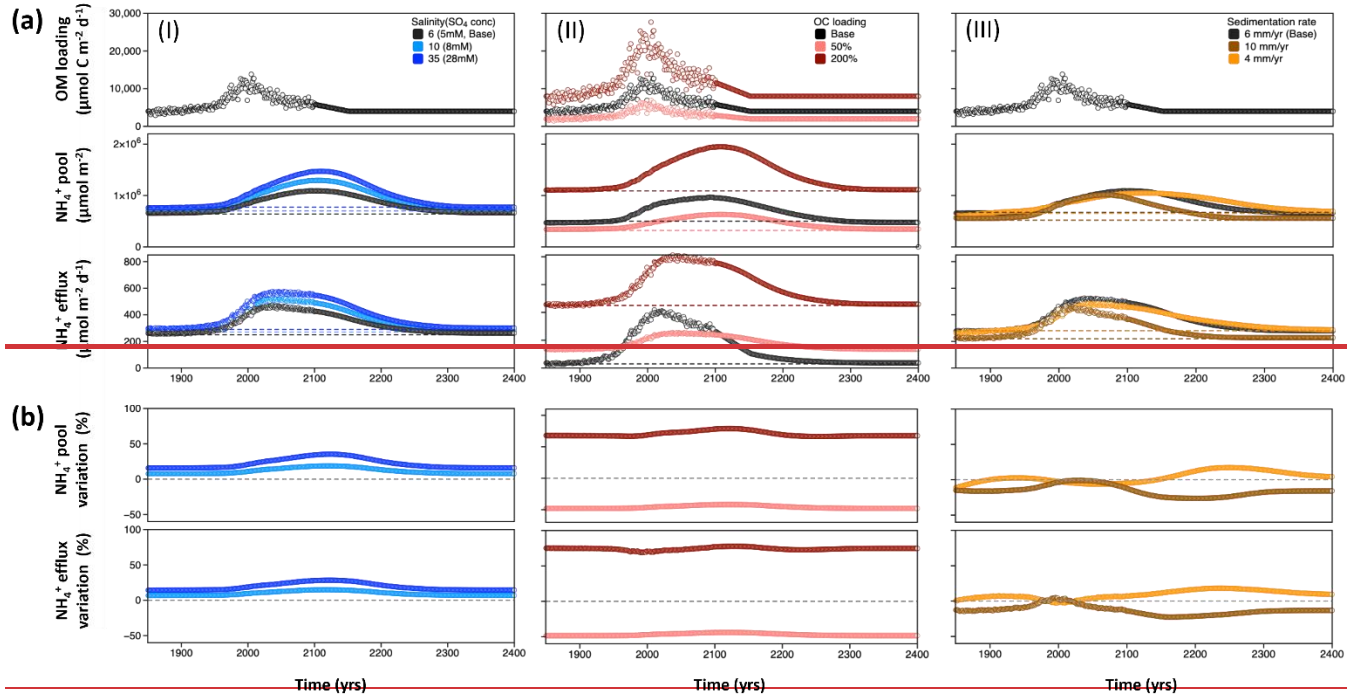


Figure BCD21. Effect of changed ~~salinity (I),~~ organic matter (OM) loading (I~~I~~), salinity (II), ~~and~~ sedimentation rate (III), carbon to nitrogen (C/N) ratio in OM (IV), and different shares of reactive material in OM (V) compared to baseline conditions representative of the coastal northern Baltic Sea (salinity of 6, Pre-industrial OM loading scenario, sedimentation rate of 6 mm yr^{-1}, and OM with a C/N ratio of 6.6 (reactive fraction) and 13 (less reactive fraction); ~~:~~ black line) on (a~~A~~) sediment ammonium (NH_4^+) pool and ~~sediment water NH_4^+~~ efflux, and (b~~B~~) its percent change relative to baseline condition. Modelled for anoxic bottom water conditions (~~O_2 =~~ 0 µM O_2). Dashed lines give the pre-industrial baselines of NH_4^+ pool and efflux. Please note the different y-axis scales in (a) between (I) and (II–IV), as well as in (b) between all parameters (I–IV).

Code and data availability

The R model code and the model output data are available at ~~https://doi.org/10.5281/zenodo.17082148.~~ doi.org/10.5281/zenodo.22120110.

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; BG: validation, writing – review and editing; LH: validation, writing – review and editing; ~~BG: validation, writing – review and editing;~~ CH: validation, writing – review and editing; BMK: methodology, 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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