

First of all, we'd like to thank the anonymous reviewers for their detailed and constructive comments which will greatly help us in preparing a revised manuscript. Here we provide our replies (in bold text) to their comments (in plain text).

Reviewer #1

The authors apply a Bayesian mixing model to determine the relative contribution of various nitrate sources to 4 regions of the Baltic Sea. The assessment is based on dual nitrate isotopes and relies on the fact that different nitrate sources carry distinct isotopic signatures. Based on model estimates, the authors conclude that the southern regions of the Baltic Sea are mainly influenced by river inputs, whereas the influence of nitrate from pristine soils is highest in northern basins. The contribution of atmospheric deposition is always low and negligible in the southern part of the study area.

The manuscript and dataset is interesting, and the use of the SIAR mixing model is a promising approach. Nevertheless, the manuscript in my opinion will still need some revision. The main part of the study, the use of the mixing model (albeit a valuable way of addressing the different input sources) is where I see the most serious issues:

(1) In general, I doubt the source assessment for $\delta^{18}\text{O}$ in nitrate from N_2 fixation. Naturally, this fixed nitrogen will primarily not contain any O. The authors try to solve this by using a fractionation factor computed by Buchwald and Casciotti (p. 5874, lines 15-19), but this is problematic. Buchwald and Casciotti actually state that the $\delta^{18}\text{O}$ value of freshly produced nitrate should be

$$\delta^{18}\text{O}_{\text{NO}_3} = \frac{2}{3}\delta^{18}\text{O}_{\text{source}} + \frac{1}{3}(\delta^{18}\text{O}_{\text{H}_2\text{O}})$$

It is true that they, based on their measurements of $\delta^{18}\text{O}$ in nitrite and the fractionation factor $\delta^{18}\text{O}$ during nitrite oxidation (12.8‰, get to a range of $\delta^{18}\text{O}$ for freshly produced NO_3^- that is 0.7 to 8.3‰ below that of seawater. However, they also note that this is *considerably* lower than measured values of NO_3^- in the open ocean, and reckon that the largest uncertainty stems from the isotope signature of nitrite. This isotope signature is not known, mentioned, or measured in this study. How plausible is it then to use this theoretical value as a source signature? I am not aware of any study that actually found such low nitrate isotope signatures in any system that is influenced by nitrification. A good example are actually pristine (or agricultural, for that matter) soils/catchments. The authors themselves assume that pristine soils have an isotope signature of 2‰ – but this actually is a nitrification signal. Hence, I don't think it is valid to use the theoretically assumed value of (seawater minus X) as a source signature for freshly produced nitrate.

Is it possible to estimate the contribution of N_2 fixation based on nitrogen isotope data alone - especially as N_2 fixation was excluded in areas with possible input of atmospheric deposition, the other source of depleted N? That would make this specific part of the model much more trustworthy, even if uncertainties rise. While I do not specifically question the relevance of N_2 fixation, I do doubt the general applicability of the model if these source signatures are used.

Authors: The reviewers concerns regarding the estimated $\delta^{18}\text{O}$ of nitrate deriving from N_2 fixation are legitimate and it is true that our assumed values of -14.3 to -6.7‰ are lower than all known measured values in the ocean. Therefore we changed our assumptions which were based on a theoretically assumed value (seawater minus X) as a source signature for freshly produced nitrate from N_2 fixation.

Our new assumptions are based on both measured and theoretically assumed values from the literature. Measurements from Bourbonnais et al., 2009 ($\delta^{18}\text{O}$ values of 2.0‰) and estimated $\delta^{18}\text{O}$ of NO_3^- deriving from N_2 fixation from Sigman et al. (2009) ($\delta^{18}\text{O}$ values of -0.2‰) and Bourbonnais et al. (2012) ($\delta^{18}\text{O}$ values of -3.8‰) were taken. The estimated values for nitrate from N_2 fixation are now between -3.8 and 2.0‰ (mean -0.7 ± 2.9 ‰). The text now reads "The $\delta^{18}\text{O}$ values were estimated to be between -3.8‰ and 2.0‰, based on measurements in the subtropical northeast

Atlantic where N_2 fixation was the main source of N (Bourbonnais et al., 2009) ($\delta^{18}O-NO_3^- = 2\text{‰}$) and the estimated $\delta^{18}O$ of NO_3^- deriving from N_2 fixation by Sigman et al. (2009) ($\delta^{18}O-NO_3^- = -0.2\text{‰}$) and Bourbonnais et al. (2012) ($\delta^{18}O-NO_3^- = -3.8\text{‰}$).” Even though we changed the estimated values for nitrate from N_2 fixation as a potential nitrate source the percentage contribution of the four sources are similar and the trend that the southern regions of the Baltic Sea are mainly influenced by river inputs, whereas the influence of nitrate from pristine soils is highest in northern basins remained.

Values for nitrate from N_2 fixation in table 1 were also changed.

Concerning the isotope signature of nitrite it is true that we did not measure it, but in the “Material and methods” section it is written that “ NO_2^- was not removed since its concentrations were always less than 2% (referring to the procedure described in Casciotti et al., 2007).” We therefore assume that there is no impact from nitrite on the nitrate stable isotope signal of oxygen according to (Casciotti et al., 2007) and later publications from her.

(2) In line with this, I can imagine that it is nearly impossible to separate influences by atmospheric deposition and nitrogen fixation. I believe the authors were aware of this problem, as they excluded N_2 fixation from the northern parts of the study area – but how much would it have altered the results if all 4 potential sources had been considered?

Authors: It is true if we include N_2 fixation in the Gulf of Bothnia and the rivers our results for those parts would look different since the isotopic values of nitrate from N_2 fixation and from pristine soils are similar. Especially in the Gulf of Bothnia, N_2 fixation as a potential nitrate source would contribute up to $63.7 \pm 4.3\%$ if we consider it as a potential nitrate source.

However, we know that in the Gulf of Bothnia no N_2 fixers occur (Dahlgren et al., 2010, Mar. Ecol. Prog. Ser., Vol. 409: 77–94), since - compared to the rest of the Baltic Sea - this area is not nitrogen limited but phosphorus limited (Graneli et al., 1990) hence N_2 fixation does not take place (see page 5876). Therefore we excluded N_2 fixation in the northern part of the Baltic Sea and the rivers. The contribution of atmospheric deposition stays the same in all areas, it does not matter if we include or exclude N_2 fixation.

(3) the source assessment is an integral part of the model and the study. In Table 1, source signatures are listed, but clear references are missing (this study? Previous studies?) This might all be in the text, but it is well-hidden. This is especially important as the referencing scheme for ^{18}O changed when a new value was assigned to IAEA- N3 – older data can have an offset of 3‰

Authors: The reviewer is correct. The references were only in the text and are now added to Table 1. Additionally, we have now compared our taken source values with more literature data in the Material and method section “2.3 NO_3^- sources”. See also Reviewer 2 point 3.

(4) The importance of atmospheric deposition should be discussed. Mayer et al (2002), cited in this study, find only a slight elevation of ^{18}O values even at 40% contribution of atmospheric deposition. Is this due to recycling? That may also apply to the Northern rivers, where atmospheric deposition seems to be the most important N source- but after assimilation into plant biomass, and upon remineralization/nitrification, the ^{18}O signal will of course be lost. These effects should be mentioned

Authors: The reviewer is correct. We write on page 5882 in line 17 to 19: “Additionally, the NO_3^- from atmospheric deposition is presumably intensively cycled through the organic N pool in spring and summer such that after several mineralization cycles its origin is difficult to recognize isotopically (Mayer et al., 2002).” However, our samples were taken in winter and therefore assimilation into biomass and remineralization/nitrification should hardly affect the isotopic signal of atmospheric deposition. It just seems that the influence from rain events is too small to leave a detectable isotopic imprint in the water column (see p. 5882 line 3 to17).

Specific comments:

- I find it quite awkward that the authors chose to merge the results and discussions section. I find this very unpleasant to read, as it is not always easy to disentangle their own work and previous

studies or assumptions based on those. If this is in accordance with the BGD style, I would highly recommend splitting this up into separate sections.

Authors: The results and discussion sections are now separated.

Introduction, p. 5870, line 25: N-fluxes have doubled” – please specify. Which fluxes?

Authors: riverine fluxes. The text now reads: “Consequently, riverine nitrogen (N) fluxes have doubled in recent years, which has strongly impacted the marine N cycle and ecosystem health, both at regional and global scales.”

p. 5872, lines 8-15: might be a stylistic issue, but to me, the Baltic Sea is anything but a typical temperate ocean.

Authors: The sentence with the temperate ocean was deleted and the text now reads: “In the Baltic Sea the NO_3^- pool present in the surface waters in spring originates from the previous growth season and is consumed during the onset of the phytoplankton spring bloom, in February/March. Stratification in summer hinders circulation down to the halocline, thus atmospheric deposition and N_2 fixation are the major N sources, whereas in coastal areas riverine discharge dominates (Radtke et al., 2012; Voss et al., 2011).

- I suppose it has been ensured that sampling (February) was before the onset of the spring bloom (Feb/March, p. 5872, line 11). Still, this should be mentioned in the Material and methods section.

Authors: Yes the samples were taken before the onset of the phytoplankton spring bloom. Chlorophyll a samples were taken at the same stations. It is now mentioned in the material and method section right at the beginning.

p. 5873, line 10: What was the sampling interval, i.e., how long were samples left in the samplers before filtration?

Authors: Rain was collected always at least after 24 h. The text now reads: “In Warnemünde, precipitation was collected on an event basis, and retrieved daily to limit microbial degradation, using a sampler consisting of a plastic funnel (diameter: 24 cm) connected to a 1-L polyethylene bottle.”

Model results: I noticed that mean values for individual source contributions are not the mean of minimum and maximum values. This is probably due to the distribution of model results?

Authors: May be the reviewer missed that we calculated a mean value and not a median which would be a value lying at the midpoint of a frequency distribution of observed values, such that there is an equal probability of falling above or below it.

- p. 5878, lines 12: “concentrations in winter were almost identical” – This is impossible to see in figure 2. The color coding encompasses values from 0-250 M. This figure could be split, and low-concentration ranges could be expanded in the color scale.

Authors: If this is ok with the editor we include a table of our raw data in the supplements and concentrations can be checked here.

- p.5878/5879, lines 27-4: This does not sound like a novelty, but like a mere reporting issue to me.

Authors: Here we wanted to point out a difference between the only two studies using SIAR for source apportionment and ours. The studies from Xue and Yang, used SIAR for nitrate source identification based upon literature values, while we used source values which were obtained in the study area.

-p. 5879, line 27: Referring to 3.9‰ here is not quite correct, as far as I know, Knapp et al refer to DON, not nitrate. This number is not needed, it is quite clear that N from N_2 fixation is relatively light.

Authors: 3.9‰ was deleted.

- p. 5880, line 3: where do these $-9.8 \pm 3.8\%$ come from? I can't follow that calculation.

Authors: This part was changed since the estimated $\delta^{18}\text{O}$ value of nitrate deriving from N_2 fixation was changed. See also point 1 in general comments from reviewer 1.

-p. 5888, lines 21-23: I would be interested to know if this result can be attributed to the (questionable) ^{18}O source assignment for N_2 -fixation.

Authors: See answer of point 1. Even though we changed the estimated values for N_2 fixation as a potential nitrate source the percentage contribution of the four sources are similar and the trend that the southern regions of the Baltic Sea are mainly influenced by river inputs, whereas the influence of nitrate from pristine soils is highest in northern basins remained. Also the N_2 fixation in the three areas where we expect N_2 fixation to occur are almost identical. See Table 3 for comparison of the revised values and the old values.

- p. 5882, lines 2-19: Can you please include some numbers on pool sizes etc. to put these calculations into perspective? same later in this section, when referring to the Kalix river.

Authors: In this section pool sizes were included for the Baltic Sea, Baltic Proper and the Kalix River.

-The last section, 3.3, the comparison of water column versus sediment isotope values, is entirely isolated from the rest of the manuscript. Can you define a specific objective to incorporate these results in your study? Else, they might as well be omitted. Also, I don't quite get what you're trying to say in line 10/11. I can imagine that during a spring bloom, there is lots of uptake, and afterwards, sinking particles, and complete consumption, too, but this is a strange way to put it.

Authors: With the comparison of surface water and sediment $\delta^{15}\text{N}$ values we can demonstrate how consistent the nitrogen input to the sediments is over years. Coastal areas preserve the isotope signature of riverine sources while open Baltic Sea sediments indeed mirror the nitrogen input dominated by N_2 fixation. Moreover the data demonstrate that no change over time in the input of the sources has occurred. Our surface water samples were sampled from 2008 to 2011, the surface sediment samples collected 1993 to 2003, and deposited in the period of app. 10 years before collection. This is now included in section "4.3 Comparison of isotope patterns in the water column and sediments".

Does a comparison of $7.9 \pm 1.8\%$ to $7.3 \pm 2.1\%$ really suggest significant differences? Did you test this?

Authors: The reviewer is correct; the values are not significantly different. In the beginning of the paragraph we write: "The $\delta^{15}\text{N}$ values from surface water correlated significantly with those from surface sediments, as reported in Voss et al. (2005) ($p < 0.001$)." We deleted the section where we tried to argue that when nitrate is only partially consumed, which seems to be the case in the near coastal area, the $\delta^{15}\text{N}$ of surface sediment incorporate this incomplete NO_3^- utilization as a lower isotopic value than the NO_3^- in the overlying water and therefore the $\delta^{15}\text{N}$ values were slightly higher in the surface water compared to the sediments.

Conclusions – Highlighting the importance of atmospheric deposition, right at the end of a study that basically showed that this source is not relevant, seems a bit strange. To catchments, maybe, but to the Baltic?

Authors: Good suggestion. We have weakened our statement that increasing atmospheric deposition will affect the Baltic Sea and reformulated the sentences. The text now reads: "Because they are particularly sensitive to human pressure and global climate change, marginal seas, including the Baltic Sea, will no doubt be affected by the increases in temperature and precipitation predicted for the near future (BACC, 2008). Indeed, increasing atmospheric

depositions of NO_3^- in the world's oceans have already been reported, by Duce et al. (2008) and Kim et al. (2011) and, may impact northern catchments of the Baltic Sea to a larger extent. Additionally, in coastal waters under increasing eutrophication pressure the efficiency of NO_3^- removal was shown to be reduced (Lunau et al., 2013; Mulholland et al., 2008), and this additional NO_3^- may alter the biogeochemical cycle. Therefore, the identification of NO_3^- sources, especially as anticipated in response to global climate change, is important for future environmental management strategies for the Baltic Sea and other marine environments. We suggest that with an adaption of the potential sources the approach used in this study can easily be applied in other environments where NO_3^- is a major N contributor.”

technical corrections:

p. 5871, line 8: change influences to influence

Authors: Done

p. 5883, line 25 – change remain to remains

Authors: Done