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 d18O in nitrate from N2 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 18O value of freshly produced nitrate should be

^18 O_NO3=2/3^18 O_source+1/3(^18 O_H2O-")

It is true that they, based on their measurements of d18ONO2 and the fractionation factor 18" during nitrite oxidation (12.8‰, get to a range of d18O for freshly produced NO3 that is 0.7 to 8.3‰ below that of seawater. However, they also note that this is *considerably* lower than measured values of NO3 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 N2 fixation based on nitrogen isotope data alone especially as N2 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 N2 fixation, I do doubt the general applicability of the model if these source signatures are used.

Authors: The reviewers concerns regarding the estimated δ^{18} O of nitrate deriving from N₂ 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 N₂ fixation.

Our new assumptions are based on both measured and theoretically assumed values from the literature. Measurements from Bourbonnais et al., 2009 (δ^{18} O values of 2.0‰) and estimated δ^{18} O of NO₃⁻ deriving from N₂ fixation from Sigman et al. (2009) (δ^{18} O values of -0.2‰) and Bourbonnais et al. (2012) (δ^{18} O values of -3.8‰) were taken. The estimated values for nitrate from N₂ fixation are now between -3.8 and 2.0‰ (mean -0.7±2.9‰). The text now reads "The δ^{18} O values were estimated to be between -3.8‰ and 2.0‰, based on measurements in the subtropical northeast

Atlantic where N₂ fixation was the main source of N (Bourbonnais et al., 2009) (δ^{18} O-NO₃⁻ = 2‰) and the estimated δ^{18} O of NO₃⁻ deriving from N₂ fixation by Sigman et al. (2009) (δ^{18} O-NO₃⁻ = -0.2‰) and Bourbonnais et al. (2012) (δ^{18} O-NO₃⁻ = -3.8‰)." Even though we changed the estimated values for nitrate from N2 fixation as a potential nitrate source the percentage contribution of the four sources are almost identical 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 N2 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±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 180 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 180 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 remineralizaton/nitrification, the 180 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 NO₃" 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 N2 fixation is relatively light.

Authors: 3.9‰ was deleted.

- p. 5880, line 3: where do these -9.8 +/- 3.8‰ come from? I can't follow that calculation. Authors: This part was changed since the estimated δ^{18} O value of nitrate deriving from N2 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) 180 source assignment for N2-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 N2 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 $\delta^{15}N$ 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 N₂ 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 +/- 1.8‰ to 7.3 +/- 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 δ^{15} 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 δ^{15} N of surface sediment incorporate this incomplete NO₃⁻ utilization as a lower isotopic value than the NO₃⁻ in the overlying water and therefore the δ^{15} 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 NO₃⁻ 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 NO₃⁻ removal was shown to be reduced (Lunau et al., 2013; Mulholland et al., 2008), and this additional NO₃⁻ may alter the biogeochemical cycle. Therefore, the identification of NO₃⁻ 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 NO₃⁻ 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**

Reviewer #2

In general, the manuscript is well written and contains interesting information regarding the nitrate source apportionments estimated by SIAR, a Bayesian isotopic mixing model in the Baltic Sea. However, the paper needs some clarification and additional support for some of its interpretations.

1. Page 5871 lines 26-27: When low water temperatures reduce microbial activity, the fraction nation can be minimized and neglected, is that right? I do not think so; at least I think nitrification is still going on. We conducted incubation experiments using coastal water at a temperature around 10 C and we found continued nitrification in the time sequence. Can you provide a detailed explanation?

Authors: Nitrification continues at low temperatures you reported. In our study the water temperature was even lower with 2 to 4 °C. On page 5878 we provide further information why nitrification should not be a minor process impacting our values. Here the text now reads: "In this study, we assumed that the effects of fractionation by microbial processes were negligible because all our samples were collected in winter, at a mean temperature of 3.1 ± 1.3 °C (data not shown), when microbial activity is low (Pfenning and McMahon, 1997), as confirmed in a study of nitrification in the Baltic Sea by Jäntti et al. (2011). They showed that in the Gulf of Finland although nitrification potentials may be high during cold months, in situ nitrification is undetectable whereas the rate increases progressively towards the summer.

2. Page 5872: considering your field sampling, it included 2008, 2009, 2010 and 2011. You sampled the surface water in four years for the Baltic sea and compare the performance of source contributions of atmospheric deposition, N2 fixation, pristine soils and agricultural runoff. My question is that did you consider about the uncertainty regarding source variations in these four years? Are there any sudden events that may alter the N pool?

Authors: Even though samples were taken from 2008 to 2011 we expect source variation to be negligible since, for example river dual nitrate isotope values were rather stable over the years of sampling in a period from 2000-2002 in the 12 Baltic rivers (See Voss et al. 2006; Biogeosciences, 3, 663–676). Therefore we assume that the annual mean river nitrate has not changed significantly. The same should hold true for nitrogen fixation since those values were estimated from the process as such. Finally atmospheric deposition nitrate had been collected by Rollf et al. 2008 (Biogeosciences, 5, 1657–1667) in 2001 to 2002 and the δ^{15} N-NO₃⁻ values in winter are very similar

to our values collected almost ten years later. Moreover we are not aware of any major changes to any of the sources or climate impacts.

Additionally, the comparison of surface water $\delta^{15}N$ values with sediment surface $\delta^{15}N$ values showed that no change over time in the input of the sources has occurred. Since the values were almost identical we assume that the nitrate winter stock in the Baltic Sea seems not to be influenced by any sudden events that may have altered the N pool. (See also answer to point 8)

3. Page 5874 lines 20-21: How do you consider the data from Deutsch et al. (2006) as representative agricultural runoff? Moreover, the data was collected in 2003, the land use types, fertilizer application etc. may also change. Please provide a detailed explanation.

Authors: We expect the data from Deutsch et al (2006) to be representative for agricultural runoff since both δ^{15} N-NO₃⁻ and δ^{18} O-NO₃⁻ of 9.9±1.5‰ and 4.6±1.0‰ are in range to literature values from areas where agriculture activity is high. Johannsen et al. (2008) found in the rivers Rhine, Elbe, Weser and Ems δ^{15} N-NO₃⁻ values between 8.2 and 11.2‰ and δ^{18} O-NO₃⁻ values from 0.4 to 0.9‰ and similar isotopic ratios for nitrate where also found by Wankel et al. (2006). Even though we add the data from Johannsen et al. (2008) and Wankel et al. (2006) or add the δ^{15} N-NO₃⁻ and δ^{18} O-NO₃⁻ values from sewage and manure of 8.9±1.4‰ and 7.7±6.8‰ from Koszelnik and Gruca-Rokosz (2013) we get a slightly higher source values for NO₃⁻ from agricultural runoff but results in SIAR are not significantly different and the trend that the southern Baltic is mainly influenced by agricultural runoff stays the same. We have now compared our taken values from Deutsch et al. (2006) with literature data in the Material and method section 2.3 NO₃⁻ sources.

4. Page 5876: SIAR mixing model: Please provide the mean and standard deviation of the potential sources.

Authors: This has been already done in Table 1. Here, mean and standard deviation of the 4 sources are given.

5. Since your isotopic values of sources are not from literature, the specific source composition should be representative for your study. It is better you provided more detailed and more references to support your assumption for isotopic composition of the potential nitrate sources. Authors: We did that in the revised manuscript. Please see authors answer to point 3. In the Material and method section "2.3 NO₃" sources" a more detailed comparison to literature data is given.

6. Page 5880, lines 1-5: the three rivers you mentioned were all influenced by agricultural activities? You mentioned ERGOM model, but without any description, it is so strange! Authors: Yes all three rivers are influenced by agricultural activities. A description concerning the model ERGOM was included. The text now reads: "However, Neuman (2000) estimated that 13% of the N input of the Oder River is transported to the central Baltic Sea, while Radtke et al. (2012) could show, using a source attribution technique in the three-dimensional ecosystem model ERGOM (Ecological ReGional Ocean Model), that at least a part of the dissolved inorganic nitrogen (DIN) load from the Vistula River, the main NO₃⁻ contributor to the Baltic Sea (Wulff et al., 2009), enters the Baltic Proper. This 3D model comprises a circulation model, a thermodynamic ice model, and a biogeochemical model and utilizes the Modular Ocean Model, MOM3.1 (Radtke et al., 2012)."

7. Page 5883 NO3- from pristine soils: this section is confusing. Lines 10-12: I did not see a low d15N and high d18O values.

Authors: To present the data better we now show the raw data in the supplements. The value is the one from the northern rivers.

For clarification the section was rearranged as followed: "In general, in rivers such as the Kalix River, whose catchments include pristine vegetation, δ^{15} N-NO₃ values are low while those of δ^{18} O- NO_3 are high (Voss et al., 2006). This finding was confirmed in the present study, in which $\delta^{15}N$ -NO₃ and δ^{18} O-NO₃ values of 1.6‰ and 10.6‰, respectively, were determined. In the Kalix River, the NO₃⁻ contribution from the runoff of pristine soils as determined by SIAR is 33.8–92.8% (mean 75.3±7.9%). In pristine soils the isotopic NO₃ signal is mainly derived from nitrification, which is in agreement with previous studies of small catchments, where much of the NO₃⁻ was shown to be of microbial origin (Campbell et al., 2002; Kendall et al., 2007; Mayer et al., 2002). Similar δ^{15} N-NO₃⁻ values were reported for areas where pristine soils were also sampled. For example, δ^{15} N-NO₃ and δ^{18} O-NO₃ values of 1.9 and 2.8‰ were determined for Biscuit Bay (Burns et al., 2009) and 2.9 and 2.8‰ for the San River (Koszelnik and Gruca-Rokosz, 2013), respectively. The higher δ^{18} O-NO₃⁻ values of the Kalix River can, as discussed above, be attributed to atmospheric deposition. For the Gulf of Bothnia, where the catchment is dominated by pristine areas like forests (50%) and shrubs (20%), NO₃⁻ from pristine soils contributes 91.7-100% (99.0±0.9%). However, for the Baltic Proper the NO₃ contribution from pristine soils is negligible, because the NO₃ derived from nitrification is very low in concentrations and remains in the Gulf because of the cyclonic circulation in the Bothnian Sea and Bothnian Bay (Humborg et al., 2003) and the high residence time of the water (7.4 years) which results in a rather slow exchange with the rest of the Baltic Sea (Myrberg and Andrejev, 2006)."

8. Page 5884, lines 15-17: again, the sediments data were from 2005, and you compare it to the present ones for coastal water. This means you assume the difference in these years were not significant, right?

Authors: The data from the sediment samples published in 2005 give not the information on a certain time point, rather the $\delta^{15}N$ pattern in surface sediments mirrors the deposition over the last few years to decades since the first 1 to 2 cm of the cores were sampled. The timescale depends on the sedimentation rates which have been found to vary between 0.2 and 4.3 mm yr⁻¹ (see Voss et al. 2005). If we now compare the $\delta^{15}N$ sediment values to our $\delta^{15}N$ values from the water column we do not find any differences in the Baltic Sea. The values are identical, which indicates that the nitrate winter stock in the central Baltic Sea seems not influenced by any event that may have altered the N pool.

We included following text in the section 4.3: "Additionally, we could show how consistent the nitrogen input to the sediments is over the years. Even though, our surface water samples were sampled from 2008 to 2011, the surface sediment samples from 1993 to 2003 and deposited in the period of approximately 10 years before collection, the comparison of the δ^{15} N values showed that there is no significant difference. Coastal areas preserve the isotope signature of riverine sources while the open Baltic Sea sediments indeed mirror the nitrogen input dominated by N₂ fixation. Moreover the data demonstrate that no change over time in the input of NO₃⁻ sources has occurred."

9. Considering the source apportionment for the Baltic Sea, you specify source by source, why not sampling part by part (western Baltic Sea, Baltic Proper etc.), which may be easier to understand. Authors: During the writing process we also considered to structure the discussion not by source but rather by area but decided to structure according to the sources. Otherwise details about the basins, their specific potential differences in source apportionment would have to be discussed. This makes the discussion difficult to read for readers outside the Baltic Sea community. Moreover the data base may be too small to find proper source descriptions for all sub-areas.