

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 #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 fractionation 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  $3.1 \pm 1.3$  °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, N<sub>2</sub> 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 Rolf et al. 2008 (Biogeosciences, 5, 1657–1667) in 2001 to 2002 and the  $\delta^{15}\text{N-NO}_3^-$  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  $\delta^{15}\text{N}$  values with sediment surface  $\delta^{15}\text{N}$  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  $\delta^{15}\text{N-NO}_3^-$  and  $\delta^{18}\text{O-NO}_3^-$  of  $9.9 \pm 1.5\text{‰}$  and  $4.6 \pm 1.0\text{‰}$  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  $\delta^{15}\text{N-NO}_3^-$  values between 8.2 and 11.2‰ and  $\delta^{18}\text{O-NO}_3^-$  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  $\delta^{15}\text{N-NO}_3^-$  and  $\delta^{18}\text{O-NO}_3^-$  values from sewage and manure of  $8.9\pm 1.4\text{‰}$  and  $7.7\pm 6.8\text{‰}$  from Koszelnik and Gruca-Rokosz (2013) we get a slightly higher source values for  $\text{NO}_3^-$  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  $\text{NO}_3^-$  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  $\text{NO}_3^-$  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  $\text{NO}_3^-$  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  $\text{NO}_3^-$  from pristine soils: this section is confusing. Lines 10-12: I did not see a low  $\delta^{15}\text{N}$  and high  $\delta^{18}\text{O}$  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,  $\delta^{15}\text{N-NO}_3^-$  values are low while those of  $\delta^{18}\text{O-NO}_3^-$  are high (Voss et al., 2006). This finding was confirmed in the present study, in which  $\delta^{15}\text{N-NO}_3^-$  and  $\delta^{18}\text{O-NO}_3^-$  values of  $1.6\text{‰}$  and  $10.6\text{‰}$ , respectively, were determined. In the Kalix River, the  $\text{NO}_3^-$  contribution from the runoff of pristine soils as determined by SIAR is 33.8–92.8% (mean  $75.3\pm 7.9\%$ ). In pristine soils the isotopic  $\text{NO}_3^-$  signal is mainly derived from nitrification, which is in agreement with previous studies of small catchments, where much of the  $\text{NO}_3^-$  was shown to be of microbial origin (Campbell et al., 2002; Kendall et al., 2007; Mayer et al., 2002). Similar  $\delta^{15}\text{N-NO}_3^-$  values were reported for areas where pristine soils were also sampled. For example,  $\delta^{15}\text{N-NO}_3^-$  and  $\delta^{18}\text{O-NO}_3^-$  values of  $1.9$  and  $2.8\text{‰}$  were determined for Biscuit Bay (Burns et al., 2009) and  $2.9$  and  $2.8\text{‰}$  for the San River (Koszelnik and Gruca-Rokosz, 2013), respectively. The higher  $\delta^{18}\text{O-NO}_3^-$  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%),  $\text{NO}_3^-$  from pristine soils contributes 91.7–100% ( $99.0\pm 0.9\%$ ). However, for the Baltic Proper the  $\text{NO}_3^-$  contribution from pristine soils is negligible, because the  $\text{NO}_3^-$  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  $\delta^{15}\text{N}$  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<sup>-1</sup> (see Voss et al. 2005). If we now compare the  $\delta^{15}\text{N}$  sediment values to our  $\delta^{15}\text{N}$  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  $\delta^{15}\text{N}$  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 N<sub>2</sub> fixation. Moreover the data demonstrate that no change over time in the input of NO<sub>3</sub><sup>-</sup> 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.