

Response to comments by Referee #2

We are grateful the referee for thoughtful comments and helpful suggestions. We have accounted all of them and made the necessary changes.

1. Specific Comments Title: It would be useful to use dissolved inorganic nitrogen (DIN) instead of inorganic fixed nitrogen, since the measured species are not fixed. These species simply presented in the water-soluble fraction.

DIN had been typically used for dissolved inorganic forms of nitrogen in seawater until questions on di-nitrogen (N_2) was raised. Di-nitrogen is also dissolved inorganic nitrogen. Besides, its concentration is much higher, as compared to nitrate, nitrite, or ammonium. In order to recognize between di-nitrogen and nitrogen compounds, the later are often called "fixed nitrogen" (Examples are numerous in publications on nitrogen cycling). Thus, we use IFN (inorganic fixed nitrogen) to identify inorganic compounds of nitrogen (nitrate, nitrite, ammonium), but di-nitrogen.

2. Abstract As stated, DIN deposition is reported for rainwater samples. Therefore, it would be suitable to apply wet deposition instead of atmospheric deposition throughout the whole document (for example, see line 1: wet deposition and line 16: atmospheric deposition)

We explain at page 11164 that "The sampler was open for collection of wet atmospheric precipitations and closed on other time in Sevastopol. The sampler collected dry and wet atmospheric deposition in Katsiveli". We quantify specifically and point further at lines 20-23 that the input of dry deposition in on average 14%.

3. Lines 13-15: Please specify riverine input as $t\ N\ yr^{-1}$. On the other hand, Ludwig et al. (2009) have assessed the riverine input of $1.12\ t\ N\ yr^{-1}$ for the Black Sea. Regarding reported riverine input, the DIN from wet deposition is only 28 % of the riverine input. Ludwig et al. (2009): Progress in Oceanography 80 (2009) 199–217.

We have double-checked our text to text to find that the riverine input is always specifies as $t\ N\ yr^{-1}$.

First of all, we have to change line 14 at page 11160 to "which is **on average** 39%". We have used several other published estimates for the riverine input of IFN to the Black sea to reveal (page 11172 lines 16-17) that "the atmospheric input of IFN can account for 32 – 48% (average 39%) and to conclude (page 11172 line 18) that "this is definitely an important contribution to the total budget of nitrogen". We agree and we will add another reference for Ludwig et al. (2009): Progress in Oceanography 80 (2009) 199–217. This will result in the range of 28 – 48% for the atmospheric input, but it will not change our conclusion on the importance of this input for the total budget of nitrogen.

4. Keywords: It seems that the keywords are missing.

It looks like keywords are not required for publications in Biogeosciences. If keywords are required, we suggest "inorganic fixed nitrogen, atmospheric input, the Black Sea".

5. Introduction The words 'yet' and 'but' are applied quite often. Please use another word from time to time (see page 3,lines: 4, 5, 6, 15).

We agree and we will follow this suggestion preparing the final version.

6. It seems that the usage of the 'recent' is awkward. For example, (page 3, line 11-13) 'it has been recently found that... (Donaghay et al., 1991; Duce et al., 2008). The cited studies are not recent; on the other hand, the former one is published 24 years ago. The same mistake can be

throughout whole text. Please correct these mistakes.

We agree and we will reword the text. Page 11161 line 11 "it has been found though that dry and wet ...", page 11161 line 18 remove "until recently", page 11162 line 3 remove "recently".

7. Page 4, Line 18: Kubilay et al. (2013) should be Kubilay et al. (1995). Please check cited studies.

Yes, it's our misprint. It's corrected.

8. Page 5, Lines 4-6: Medinets and Medinets (2012) have discussed the temporal variability of the atmospheric deposition of DIN between 2004 and 2010. For example, they found the highest wet DIN deposition in 2009. Please consider aforementioned publication and rephrase lines from 4 to 6.

We agree. It's corrected. "However, neither published data on spatial variations in the atmospheric input of IFN, nor information on effects of this deposition is available".

9. Data and Methods Page 6: Please use official pages while addressing sampling sites Katsiveli and Sevastopol. Remove Wikipedia links.

We can suggest official pages for Sevastopol (<https://sevastopol.gov.ru/>) and Katsiveli (<http://yalta.rk.gov.ru/rus/about.htm>), but they are only in Russian and less useful. Data presented at Wikipedia pages are good enough in this case, as we demonstrate most general information of population and location of these settlements and we do not use this information for any purposes that could mislead.

10. Page 6, third paragraph: A total of 228 and 217 rainwater samples are obtained from Sevastopol and Katsiveli, respectively, between 2003 and 2008. Please give information about sampling coverage so that the reader may assess the rigorousness of the sampling campaign.

This information is presented in Fig. 1 and Fig. 2. It has been also discussed in section 2.1 "sampling and observational data".

11. Page 7, lines 5-16: The manuscript gives information about observed levels of nitrogen species in rainwater samples. However, there is no information how the authors calculate nitrate, nitrate and ammonium in rainwater samples. As it is well known, concentrations in rain must be calculated considering volume weighted means (VWM). Without applying VWM, concentrations will be misleading. Moreover, there is information about calculation of wet deposition (see Herut et al., 1999, 2002). Please give details about these issues.

We have recalculated VWM for data presented in Fig. 2 and Fig. 3. The difference is less than 12% for Sevastopol and below 15% for Katsiveli. This difference is less than the reported operational accuracy and does not change the major conclusions of this work. But we agree and update Fig. 2 and Fig. 3. (Look, please, for updated Fig. 2 and Fig.3) We also make it clear that VWM are calculated and used throughout the text.

12. Results and Discussion 3.1. IFN speciation, average concentrations and temporal variations As the title implies (see comment above), it seems that manuscript use arithmetic mean values instead of VWM. If VWM are applied, clarify it? If not, then use VWM to present your data.

We have also used corrected volume weighted values in section 3.1 and 3.2. Where it comes to data presented and discussed in section 3.3 and 3.4, equation 2 has been used to calculate IFN inputs, rather than concentrations. Thus, the rate of precipitation has been accounted.

13. Page 10, lines 24-28: It is suggested that the observed values of DIN might be affected by long range transport (LRT) and local sources (LS). Indeed, values might be affected by LRT and LS. However, there is no attempt to clarify or support aforementioned suggestion. Thus, it would be useful to categorize air masses back trajectories and assess the influence of air flow on the DIN composition. In addition to fuel combustion, are there any possible sources for DIN, such as residential heating, wood burning, and vehicle emissions?

We have estimated the extent of the influence of local source (section 3.2). “We have found that the effect of local sources associated with large cities for typical conditions of Sevastopol is limited to coastal zone within 25 km distance. Despite the fact that local sources have no significant direct effect on off-shore areas of the sea, monitoring of IFN deposition remains important to correctly evaluate the budget of nitrogen in coastal waters near industrial sites. It is specifically true for winter, when these sources are most significant. Thus, the input of IFN to the major off-shore part of the Black Sea can be correctly estimated applying the multiple regression (Eq. 2). This is specifically true and important for open off-shore areas, where direct observations for rain events and sampling are hardly possible.

Where it comes to air masses back trajectories, they have been analyzed by Kubilay et al. (1995) in detail and we have used those data.

14. Page 12, lines 21-23: The emission from ship should be considered since the urban site is near to port.

It is very possible, but we have not discussed the input of various individual local sources and their contribution to the total local input. It's beyond the scope of this work.