Response to comments by Referee #1

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

1. Page 11160, Line 23-24: "This input of IFN can potentially double the level of primary production." Can be reworded as " The input of IFN to Black Sea has potential to enhance level of primary production".

   This sentence has been changed to "The input of IFN to the Black Sea has potential to enhance two-fold the level of primary production".

2. Page 11161, Line 6-8: "Yet, this data is exceptionally important for understanding, modeling, and fighting negative effects of anthropogenic eutrophication and other effects in the Black Sea ecosystem." Can be reworded as "Such data are very important for understanding, modelling and assessing negative impacts of anthropogenic eutrophication as well as other effects in the Black Sea ecosystem."

   Accepted.

3. Page 11161, Line 17: "Yet, when it comes to …" can be replaces by "However, in case of …"

   Accepted.

4. Page 11161, Line 19-24: Too long statement. Can be reworded as "Black Sea being surrounded by industrialized areas (or countries), it is obvious as well as highlighted by few studies ((Kubilay et al., 1995; Chaykina et al., 2006; Medinets and Medinets, 2012), the significant influx of IFN via atmospheric wet deposition. However, till date, neither basin-wide magnitude, nor spatial and temporal variations, or aftereffects of this input to the Black Sea has been ever evaluated."

   This long sentence has been reworded. "Though scarce published data (Kubilay et al., 1995; Chaykina et al., 2006; Medinets and Medinets, 2012) demonstrate an input of inorganic fixed nitrogen (IFN) with atmospheric precipitations, neither basin-wide magnitude, nor aftereffects of this input to the Black Sea have been ever evaluated".

5. Page 11162, Line 8-14: Again, it's too long statement. Can be divided into two lines.

   It's edited. "A number of studies has been completed over the last two decades to show that this source can account for 60% of the total continental supply of IFN to the oligotrophic Mediterranean (Guerzoni et al., 1999), but it may also reach 8% for the highly eutrophic Baltic Sea (Langer et al., 2009). It can level riverine inputs and support a large short term increase in phytoplankton growth under certain meteorological conditions (Spokes and Jickells, 2005)".

6. Page 11162, Line 18: Kubilay et al. (2013) is missing in the reference list.

   It's our misprint. It's corrected to Kubilay et al. (1995).

7. Page 11163, Line 4-6: Repeated here from Page 11161, Line 22-24.

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

8. Page 11163, Line 9-11: 19 % of air parcels are derived from local region. How they can contribute to long-range transport?

   It's corrected. "Atmospheric transport in the Black Sea region is dominated by trajectories from

Eastern Europe (38 %), Russia (33 %), local region (19 %), and North Africa (10 %) (Kubilay et al., 1995)".

9. Page 11163, Line 11-14: What I understand here that 90 % of precipitation events have back-trajectories associated with long-range transport (which are crossing anthropogenic IFN emission regions). However, in the next line, the local processes are highlighted. I think, both play significant role in bringing pollutants (in this case IFN) and their delivery to the surface ocean.

We agree with this comment and have reworded this statement. "This means that long-range transport is important for seasonal variations in the content of IFN in the atmosphere and its input to the Black Sea, but local processes of scavenging below clouds on time of precipitation are important for regional spatial and short-term temporal variations".

10. Page 11163, Line 17-22: Objectives can be reworded as: (i) to present multi-annual observational dataset of IFN (: : :..) via wet atmospheric deposition...(ii): : :(iii): : : and (iv) to evaluate impact of IFN deposition to the Black Sea on various time scales.

We agree with this suggestion and reworded this part of the text. "This paper is aimed (i) to present multi-annual observational dataset of IFN (ammonium, nitrate and nitrite) with wet atmospheric deposition in an urban (Sevastopol) and rural (Katsiveli) sites of the Crimean coast of the Black Sea; (ii) to analyze interannual, seasonal, and mesoscale variations in IFN deposition; (iii) to parameterize this deposition; and (iv) to evaluate impact of IFN deposition to the Black Sea on various time scales".

11. Data and Methods: It will be useful to mention rainfall pattern and sample collection procedure at the sampling site. What is the collection period of each samples? Collected sample represent a single or multiple event in a day?

The rainfall pattern is discussed referencing Ivanov and Belokopytov (2013) in section 3.3. Yet, we add information on sample collection. "Wet atmospheric precipitations of every single rain event have been collected. The collection procedure has conformed to requirements of EMEP (EMEP Manual, 2001)."

12. Page 11165, Line 5-18: This should go to Result and discussions.

We consider observational data to keep in section "Data and methods", while results derived from observational data in section "Results and discussions".

13. Page 11165, Line 19-21: Briefly discuss meteorological parameter and their seasonality as they will be used in regression analyses.

We discuss meteorological information important for this paper in section "Results and discussion" following the referenced publication by Ivanov and Belokopytov (2013). There are many published papers and several books to present and discuss meteorological data for the Black Sea in detail. We use that information and apply it for the purposes of our work. Data on precipitation has been found important and it is discussed in section "Results and discussion". Data on wind is less important for in depth analysis and it is mentioned briefly. But it is well presented in the referenced publications, if needed.

14. Page 11168, Line 14-19: Can be reworded.

"Our data are also in good agreement with those reported for the North Sea (Jickells, 1995), the eastern Mediterranean (Cretan) Sea (Guerzoni et al., 1999), and the northern Levantine basin (Kocak et al., 2010)."

15. Page 11169, Line 10-11: Why there is a seasonal variation in the fuel consumption? Is this a local phenomenon near sampling site (Sevastopol)? Or is it attributed to any socio-economic activity?

    Fuel consumption is increased on winter time mostly for heating purposes. It's maximum is on February due to local intra-annual variations in temperature.

16. Page 11170, Line 4: Are concentration same for both sites?

    It says: "The average concentrations and the ratio of nitrate to ammonium are very the same for Sevastopol and Katsiveli in summer. The average summer concentration of IFN is equal to $1.3 \pm 0.29 mgN\ L^{-1}$ and the ratio of nitrate to ammonium is equal to 1.40." We have found no statistically significant difference between two sites on summer time.

17. Page 11171, Line 19-23: How the concentrations are calculated on spatial scale over Black Sea? Does the contribution from other source region (especially near coast) are considered while extrapolating the IFN concentration?

    We used the derived multiple regression equation (section 2.2) and meteorological data (section 2.3) to calculate both concentrations and atmospheric deposition of IFN over the Black Sea. In 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."

Technical comments: There are several grammatical errors and typo. Few are mentioned below.

1. Page 11161, Line 2: "… marine environments has … . Should be "… marine environment have …"

   "The importance … has been revealed and demonstrated …" It's all right, unless we miss the point of this comment.

2. Page 11163, Line 8: "… below clouds on …" should be "… below clouds during … ".

   We agree. "… below clouds during atmospheric precipitation …"

3. Page 11165, Line 6: "… April 2005, accordingly …" Should be "… April 2005, respectively …"

   We agree. It's "… April 2005, respectively."

4. Page 11167, Line 19: chlorophyll …

   Yes, it is "chlorophyll …"

5. Page 11168, Line 8: Remove region.

   Yes, we agree.

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.