Answer to the reviewers

Throughout the paper we have made smaller corrections to the text to improve readability and sharpening the descriptions and conclusions. Based to large extend on the many valuable comments from the reviewers we have added additional references, and extended some sections with additional explanation. Finally we have again based on suggestions from the reviewers added an additional figure with dry and wet deposition calculations from the EMEP model. In the following 7 pages we have described the various changes made to the text on basis of the suggestions from the two reviewers. It is our belief that the review paper has improved considerably on basis of these modifications.

Reviewer 1:

We are very pleased that reviewer 1 finds our review useful and recommends publication of our manuscript after consideration of addressed comments and suggestions for additional references. We believe to have taken all comments carefully into consideration in the revised manuscript.

Page 9352 ln 8 – we have added the suggested reference to Phillips et al (2004) with a comment that dry deposition velocities for ammonia may be close to 40 mm/s.

Page 9352 Ln 13 – we have added the suggested reference to Duyzer et al. (1994) with a comment that dry deposition velocities for NH_4^+ containing aerosols are in the range 1 to 10 mm/s.

Page 9352 Ln 17 – we have added a reference to personal communication with Johannes van Jaarsveld with a comment that this is found in model calculations with the Dutch OPS model.

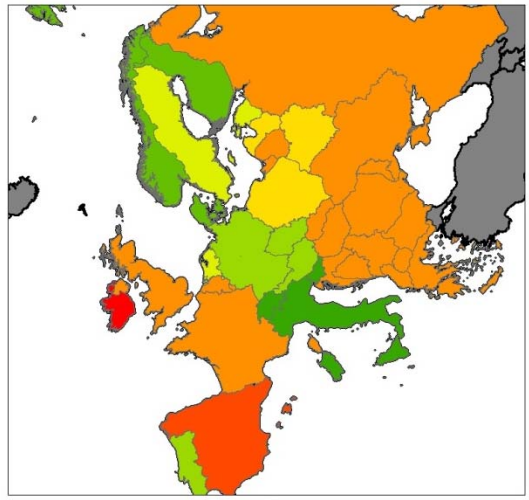
Page 9353 Ln 7 – we have added the suggested reference to Luke et al. (2010) with a comment that studies in urban areas of Houston Texas have shown that PAN and HNO_3 may be of similar magnitude and may be up to 1.5 ppbv in the urban area.

Page 9353 Ln 19 – the reviewer suggested to add the disregarded NOy compounds in a side bar to the figure or caption. We have extended the figure caption to explicitly mention the disregarded compounds. We decided not to extend the sketch itself in order to keep simplicity.

Page 9355 Ln 20 – The reviewer asks for reference concerning emissions from manufacturing processes – and we have accordingly added a reference to Reis et al. (2009) and a link to the EDGAR inventories.

Page 9355 Ln 23 – The reviewer comments on the fact that we group landfills together with non-anthropogenic sources like natural fires. We have therefore moved landfills to the Bullet above that include humans, pets and sewage systems, which is in line with the suggestion from the reviewer.

Page 9357 Ln 24 – The reviewer asks for more elaboration on the differences between emissions in Northern and Southern Europe. We have accordingly added a sentence telling that in Southern Europe animals are in the field most of the year and referred to Figure 2 in Skjøth et al. (2011) – this figure is shown here:



Page 9359 – the reviewer asks for a table showing research programs/projects – these were already listed in the text but we have formed a table as requested.

Page 9360 ln 4 – the reviewer asks for identification of national experts – we have extended the sentence so in reads: "…national inventories that originates from national experts appointed by the single countries". These experts may be academic or governmental depending on the country.

Page 9361 ln 22 – the reviewer asks for specific height of releases imposed by regulators. However, this depends on the specific case – the owners of a planned plant or factory with high release will be requested to document that their emissions will not affect health or nature. In order to do so they will usually have model calculations performed to determine the release height necessary for compliance with guidelines. The result will depend on orographic and climatic conditions as well as temperature and exit velocity of the release. We have decided not to change the text, and hope the above explanation is sufficient.

Page 9362 Sec 2.2.2 – the reviewer finds that the text in this paragraph has an unacceptable paucity of references. We have added references to road traffic research.

Page 9362 Ln 24 – the reviewer request references concerning impact of out phasing of nuclear power putting more pressure on coal based power plants. We have added references to Reis et al. (2009) and link to EMEP emission inventories http://www.ceip.at/ stating that projections in NOx emissions are more uncertain due to this out phasing. Currently emissions are still following a downward trend due to regulation.

Page 9364 ln 11 – the reviewer asks for a quantification of the decrease in sulphuric acid – we have added reference to Estonian study (Treier et al., 2008) that shows decrease in measured sulphate contents in precipitation of up to a factor of four at monitoring sites.

Page 9364 ln 13 – the reviewer asks for time period for our reference – we have added that this refers to UK in the 1960ties.

Page 9365 ln 4 – the reviewer asks for more elaboration on the reversible nature – we have extended the text with more detailed description concerning conditions where the pollutants are released to gas phase again stating that: Changes in temperature and/or humidity will lead to changes in the partitioning between gas phase NH_3/HNO_3/HCl and aerosol phase NH_4NO_3/NH_4Cl, with increasing humidity and decreasing temperature moving the partitioning towards the aerosol phase compounds

Page 9365 Ln 15 – the reviewers wants us to be explicit concerning relative abundance of HNO_3 and HCl. We have added a reference to de Leeuw et al. (2003) that from both measurements and model calculations indicated HNO3 concentrations in the range of 300 to 600 pptv – this has been added just after the sentence referring HCl concentrations around 250 pptv also in marine boundary layer.

Page 9365 Ln 19 – the reviewers finds that the statement about growing SO_2 emissions is in contradiction to previous statements about decline in SO_2 emissions. The statement refers to overall trends in NOx and SOx emissions whereas the statement in section 2 on trends in NH_3 influenced by decrease in SO_2 emissions refers to conditions in Europe. We have added a sentence stating that "global NOx and Sox emissions are still increasing whereas European emissions are decreasing" in section 3.

Page 9366 Ln 19 – the reviewers asks for information about the trends in European ozone levels. We have added a reference to Tørseth et al. (2012): An analysis of long-term O_3 trends at EMEP monitoring sites showed a mixed pattern. A substantial year-to-year variability in O_3 concentrations is caused by varying meteorological conditions and this variability made it hard to separate trends related to emission change from other effects. For the Nordic countries the data indicated a reduction over the years in occurrence of very low concentrations. The most pronounced change in the frequency distribution was seen for the UK sites and for the sites in the Netherlands, showing a reduction in the higher values. Smaller changes are seen in Germany, while in Switzerland and Austria, no change is seen in the frequency distribution of O_3.

9367 – the reviewers finds that the text has a unacceptable paucity in references. Concerning the conversion between NO to NO2 in reaction with O3, we have added reference to Palmgren et al. (1996) and the text book of Seinfeld & Pandis (2006). For the importance of the OH radical we have added reference to the text book of Finlayson-Pitts and Pitts (1986). For the CO chemistry we again refer to Seinfeld and Pandis (2006).

Page 9369 Ln 3 – the reviewer asks us to specify the height of the tower in London from which NO3 – we have added the information that the tower has a height of 160m.

Page 9369 Ln 27 – the reviewer asks for references to nighttime/daytime chemistry of HONO. We have added references to Finlayson-Pitts and Pitts (1986) for the daytime reactions, and we have added additional reference to Harrison and Collins (1998) concerning the heterogeneous formation of HONO from the reaction of NO2 on aerosol surfaces. For the morning dissociation of HONO we have added reference to Schiller et al. (2001).

Page 9370 Ln 3 – the reviewer question whether only few studies regarding HONO chemistry in urban areas have been carried out. We have added reference to the recent review on HONO chemistry by Indarto (2012) who states that the chemistry of HONO is still poorly understood. We have not found many studies in this regard and have thus not changed the statement in the paper.

Page 9371 Ln 13 – in compliance with the suggestion from the reviewer we have added reference to Walker et al. (2012)

Page 9371 Ln 17 – in compliance with the suggestion from the reviewer we have added reference to Lin et al. (2010)

Page 9372 – the reviewer finds that this text section on this page has paucity in references. We have in compliance with the reviewers suggestions added references to Gaffney and co-workers. Furthermore we have added references to Finlayson-Pitts and Pitts (1986) regarding the basic chemistry of PAN. Again in compliance with the reviewer's suggestions we have added reference to Tanner et al. (1988) on the reactions of aldehydes leading to formation of PAN.

Page 9372 Ln 26 to 27 – in compliance with the suggestions from the reviewer we have added references to Lamb et al. (2000) for the NADP and to Totsuka et al. (2005) for the EANET.

Page 9373 Ln 1 – in compliance with the suggestions from the reviewer we have added a reference to Meyers et al. (1998) regarding inferential models.

Page 9373 Ln 4 – following the comment of the reviewer we have rephrased line 4 so that it reads "which are underpinned by often sparse databases of campaign based process studies or long-term monitoring in both cases with limited geographical coverage"

Page 9373 Ln 11 – again in compliance with the reviewers suggestions we have added a reference to the inter-comparison study by Milford et al. (2009) on NH3 fluxes deploying various techniques.

Page 9373 Ln 22 – in compliance with the reviewer we have added the word "presently" regarding that comprehensively detailed coverage can only be obtained through modeling

Page 9373 Ln 25 – the reviewer stresses that the contribution to deposition depends not only on climate but also on local and regional meteorology and emissions. In the text we talk about "pollution climate" and not "climate" alone. Pollution climate refers to pollutant levels at the specific site that will of course depend on local and regional emissions as well as meteorological conditions leading to transport and dispersion. However, the deposition process itself depends of course also on meteorology and we have therefore added this to the sentence that now reads: "Their relative contributions to N deposition depend on the pollution climate and meteorological conditions"

Page 9374 Ln 3 – We assumed the reviewer had Walker et al. (2012) in mind (and not (2010)) and this reference has therefore been added in the text

Page 9374 Ln 9 – We assume that the reviewer had Wu et al. (2009) in mind (and not Yu et al.) and this reference has therefore been added in the text together with the suggested reference to Pleim et al. (2012)

Page 9376 – In compliance with the reviewer suggestions we have added references to Cellier & Brunet (1992), and to Spokes & Jickells (2005). Concerning Sorensen et al. (2003), this reference is used in the section on bi-directional fluxes.

Page 9379 – in compliance with the reviewers suggestions we have added a sketch of the resistance method

Page 9379 Ln 7 – again in compliance with the suggestion from the reviewer we have added an expression for the Henry's law coefficient for ammonia

Page 9385 Ln 25 – in compliance with the suggestion from the reviewer we have added a reference to the EMEP network concerning wet deposition measurements.

Figure 1 – the reviewer requests a more detailed sketch to truly illustrate the complexity of the system. The reviewer furthermore asks for percentages through the various pathways, but this is not easy as this depends on the specific environment. It would through detailed modeling be possible to come up with figures for different situations, but we find this is beyond the scope of the current publication. We have added reservoir species to the explanation in the Figure caption.

Figure 2 – the reviewer does not find that this figure adds value to the presentation

The reviewer asks for avoiding double use of wordings – as an example in the first sentence of the abstract "different and differences" which we do not find is a problem and therefore have left unchanged. Another example given by the reviewer is on Page 9357 Ln 21 "affect" – we have here modified the sentence by substituting the first "affect" the word "alter".

Reviewer 2:

Unfortunately reviewer 2 has not used the page and line numbers in the discussion paper but refers instead to page and line numbers in own printout of the paper. This makes it considerably harder to locate especially the specific corrections.

We have in compliance with the suggestion from the reviewers modified the title of the paper so that it covers better the content. The new title reads: "Governing processes for reactive nitrogen compounds in the European atmosphere"

Page 9354 Ln 22 – the reviewer states that despite we are listing emissions of major reactive nitrogen compound as consisting of NH_3, NO_x and organic N, we only treat the first two in the subsequent sections. We do state, however, that emissions of organic N such as amines are poorly quantified. We have added a short sub-section 2.4 regarding emissions of organic N including amines.

The reviewer points our rightfully that we do not follow the listed separation of NH_3 emission sources in our structuring of the sub-sections: 2.1.1-2.1.4. We agree with the reviewer and have therefore restructured

the sub-sections accordingly, and have now 9 sub-sections for each of the listed emission source groups + the two sub-sections on temporal variation and long-term trends.

The reviewer points out rightfully that we have another structure on the section 2.2 on NOx emissions compared with the structure of section 2.1 on NH3 emissions, and that this difference is not justified. We agree with the reviewer and have therefore restructured section 2.2, creating a number of new short sub-sections for the various emission sectors for NOx emissions.

Reviewer 2 asks why NO from agriculture plays a major role on global scale but in negligible in Europe! – *In a global perspective, NOx emissions from soil contribute > 40 % (Davidson and Kingerlee, 1997) (IGAC Newsletter Dec 2000), and > 10 % for some European countries (Butterbach-Bahl et al., 2004; Skiba et al., 1997; Stohl et al., 1996).* The soil contribution to a countries NO budget is much smaller for industrialised countries with high fossil fuel combustion rates than in countries with small fossil fuel combustion rates. This is especially the case for countries in warm dry climate zones, as these conditions favour microbial NO production (Skiba et al, 1997).

In compliance with the suggestion of Reviewer 2 we have moved the section on discussion of dry deposition of PAN to the sub-sequent section.

Reviewer 2 asks for references regarding problems in quantifying precipitation input and finds the discussion of relative importance of dry and wet deposition to be rather short, and asks for budgets – We have according added a plot showing EMEP model calculations of dry and wet deposition (Figure 11).

Reviewer 2 asks for identification of knowledge gaps in reactive nitrogen research –The last section has been renamed to “Conclusions and Perspectives”, and we have accordingly added some sentences in the text.

Reviewer 2 noted a typo in reference /Butterbach-Bah/Butterbach-Bahl/, that has now been corrected

In compliance with the suggestion of Reviewer 2 we have changed /breakdown of fertilizer/breakdown of mineral fertilizer/

The reviewer 2 noted that Simpson et al. (2006) mas missing and this has now been added to the reference list

Reviewer 2 asks for explanation of RH – we have now added “Relative Humidity” first time the abbreviation RH is used.

We have rephrased “Organic $NO3^-$ ” to “Organic nitrate”

We have removed (2011) from “NitrogenEurope (2011)”

We have added explanation on DMS “dimethyl sulphide”

The reference to Simpson et al. (2007) has been corrected

We have added the word “former” in front of NitroEurope project to indicate that this is no longer on-going like e.g. the EMEP

We have in accordance with Reviewer 2 harmonized to seeder-feeder (previously also “seeder – feeder” was used).

We have modified Cair to C_{air} in order to follow the notation in the formula

The Reviewer 2 finds our figure with the farm scale nitrogen deposition confusing as he would imagine an effect of trees on the deposition. The main effect is an impact on the turbulence but this is not yet well described. The results concerning impact of trees on local deposition is not showing a very clear picture as these affect local turbulence and the deposition process in a rather complex way. We have not modified the text due to this unclear picture of the impact.

The figure caption has been corrected /bas/base/