Replies to Referees 1, 2 and Editor comments

We provide here replies to all referee and editor comments. Some new references are suggested, and included at the end of this common reply. One Table and one Figure needed minor correction also, see reply to Editor.

1 Reply to Referee #1, Tony Dore

1.1 General comments

Deposition of reactive nitrogen to natural ecosystems is currently a high profile environmental issue. Levels of input from atmospheric nitrogen deposition result in the exceedance of critical loads across much of Europe as well as other parts of the world, causing a major global concern. Both scientists and policy makers are interested in future projections of emissions of pollutants (i.e. NOx and NH3) and in assessing how effective technical measures to control emissions will be in reducing the threat to the natural environment. More recently the potential impact of future climate change on air pollution has been the focus of interest. This article is therefore clearly of significance as it deals with both the impacts of climate change and emissions changes over the European continent in the framework of a multi-model study. The paper is carefully written and well referenced. The supplementary material, figures and tables are relevant to the study. The use of an ensemble of models in the study serves both to provide greater confidence in future predictions of nitrogen deposition as well as to highlight areas (where modelled results diverge) where further research is needed. I am pleased to recommend this paper for publication. A number of points are raised below where some additional clarification could usefully be provided.

Reply: We thank the referee for these comments

1.2 Specific comments

(1) Page 6670: The future projection of emissions to the year 2050 inevitably involves considerable uncertainty as the influence of new technology and changes in global commerce and energy consumption are not well known over the next four decades. Can the authors explain what assumptions have been made in the emissions projec- tions (in particular the large emissions reductions for SO2, NOx and VOCs) and give some indication of uncertainty?

Reply: The future emissions are of course very uncertain. The values we use are an updated version of the so-called ECLIPSE version 4 data, produced by IIASA for the EU ECLIPSE project as well as for the ECLAIRE and PEGASOS projects, and the TF HTAP study. These projections assume business-as-usual economic development and implementation of all currently agreed emission control legislation. Unfortunately these emissions are not yet fully documented, but the report by Amman et al (2012) describes the baseline scenarios, and the paper by Amann et al (2013) gives a good overview of the range of possible emission scenarios. Some details are also given on the IIASA web-site. Referee #2 also asked about RCP scenarios. In order to

explain these emissions in more detail, we will first denote the IIASA emissions as ECLIPSE v.4^e (Adding this label to Table 2 also), then add two pieces of text to Sect. 2.1:

It should be noted that these ECLIPSE v.4^e 2050 emissions are not the same as the so-called RCP emissions which were developed recently for the IPCC process (van Vuuren et al., 2011), because of very different assumptions concerning energy pathways and legislation. The ECLIPSE projections assume business-as-usual economic development and implementation of all currently agreed emission control legislation (cf Amann et al., 2012, 2013). They also make much more use of detailed national data, and are believed more appropriate than RCP for air quality modelling. However, the large (67%) NOx emission reductions seen in Table 2 are broadly consistent with all RCP changes for EU27 presented in Winiwarter et al. (2011). Emissions of NHx are predicted to remain almost constant in Table 2, whereas RCP estimates suggest either a significant increase (ca. 25% for RCP8.5), or a decrease (ca. 25% for RCP2.6 and RCP4.5). There are of course considerable uncertainties in all these projections, arising from assumptions concerning technical measures, growth and policies (Amman et al., 2013).

(2) Page 6672, line 1: Both models show rather similar large-scale changes in precipitation to the 2050s though. A comment on the distribution of precipitation changes (i.e. increases in southern Europe, decreases in northern Europe would be useful in the text here).

Reply: We have added the following text to the end of the paragraph:

..., with rather large increases (ca. 10%) in north-eastern Europe, and decreases of around 10% around the Mediterranean.

(3) Tables 2: BIC (Boundary and Initial Conditions) should be defined

Reply: Done (Table 1)

(4) Tables 3: stns: (number of stations) should be defined. Can some general comments be made on the overall correlation statistics? Why are models more successful in their correlation with measurements of NO2 than SO2 (treatment of elevated point sources) and of NH3 than HNO3 (complexity of oxidised nitrogen chemistry)? Are there thought to be any systematic errors in measurements (i.e. are the precipitation chemistry collections made using wet-only collectors or are they made with bulk collectors which may be subject to dry deposition?)

Reply: We deliberately kept discussion of Table 3 short since it wasn't the main focus of the paper, but we have now added some extra text in response to this comment:

The reasons for better performance of some compounds compared to others are complex, and not always understood. However, in general we expect better performance for 'simple' precursors from mainly ground-level sources (e.g. NO_2) than from high-level point sources (SO_2), or for compounds with complex chemical pathways and strong deposition-induced gradients, notably HNO_3. HNO_3 measurements are also affected by partitioning issues with ammonium nitrate and NH_3 reactions.

Regarding wet-deposition, we can note that the EMEP network is a mixture of bulk and wet-only collectors, with each country choosing the most appropriate method for its condtions (see `http://ebas.nilu.no`*). For daily sampling, there is not thought to be be a large difference in the results in many areas, but with bulk collectors some dry-deposited material will be incorrectly assessed as wet-deposition. The quality of measurement also differs; results for sulphate tend to be somewhat better than nitrate, worse for ammonium measurements (EMEP/CCC, 2014). Given these uncertainties (and the use of climate-model based meteorology), the level of discrepancies seen in Table 4 can be regarded as satisfactory.*

(5) Measurement data has been considered covering an 11 year period, taking average annual mean values to compare with modelled values. Have studies been undertaken on how the models respond to emissions changes and whether the modelled changes in concentration and deposition agree with observed changes? Reference to such studies would be useful here, particularly as the models are subsequently used in a predictive mode.

Reply: Yes, this is a good point, although the answer quickly gets complicated. We have added a new subsection 2.4.5 (Comparison with trends) to the paper though to address this issue:

Most model-measurement comparisons address the issue of how well model results match observations in current conditions. It is much harder to show that the models can capture changes in pollution with time accurately, although it can be noted that if the models work well across all of Europe, this in itself suggests they do capture the effects of changing pollution conditions in differing meteorological conditions. Some trend studies are available though, which we briefly summarise here.

For EMEP, such studies include Jonson et al (2006) for ozone and NO_2, Fagerli and Aas (2008) for Nr compounds in air and precipitation, and Colette et al. (2011) for NO_2, O_3 and PM_{10}. Schulz et al. (2013) presented comparisons for 1990, 2000-2011, for S-compounds as well as Nr. For DEHM, previous analysis of multi-year model runs show that the model in general reproduce the observed trends in concentrations and depositions of N and S components caused by emission changes (Geels et al., 2005; Geels et al., 2012b). For MATCH, Hansen et al, 2013 compared a MATCH simulation over 1980-2011, forced by EMEP emissions and ERA-Interim meteorology, to observed trends in annual mean wet deposition of NOy and NHx over different regions of Sweden.

Summarising these studies, it is generally found that the models capture the broad features of trends for the S and Nr compounds over large areas, although capturing results for specific sites is more difficult. It should be noted however that comparisons of observed and modelled trends rely on consistency in the measurement network (sites, techniques and quality), and on accurate estimates of emission trends. Problems associated with these factors have been discussed in for example Fagerli & Aas (2008) and Colette et al. (2011).

(6) Page 6682 exceedance of critical loads. The EMEP model has been developed as a European policy model and therefore calculates ecosystem specific deposition. How do the other models deal with dry deposition to different vegetation types? The use of a simple ecosystem-

independent critical level suggests that these models do not generate vegetation/ecosystem specific dry deposition values.

Reply: Yes, there is a well-developed system to use EMEP model results for estimates of CL exceedances, and we made use of that here. As noted in Sect. 2.4.3, MATCH discriminates between four different surface-types: water, low vegetation, high vegetation (i.e. forest), barren land (including urban areas). The SILAM setup used for this study used a grid-average roughness-length to estimate aerosol deposition, and for gases discriminated just land-sea, wet-dry, frozen-not frozen. It would thus have been impossible to compare the detailed CL exceedances in a coherent way across the models.

We have added a few words on the SILAM methodology to Sect. 2.4.4 to make these land-cover assumptions more explicit, but have left the text in Sect. 4 as-is, since it explains that we make use of the LRTAP procedures with the EMEP model.

(7) Conclusions, page 6686, line 1: The conclusion that for future scenarios ammonia concentrations and dry deposition of reduced nitrogen will increase due to the reduction in the availability of sulphuric acid and nitric acid to react with ammonia is quite important. This illustrates clearly the non-linearity of atmospheric chemical reactions in controlling patterns of deposition. It therefore follows that in the future nitrogen deposition will be more correlated to local emissions (of ammonia) and that long range transport (of particulates) will become relatively less important. This has implications for the effectiveness of national (as opposed to international) policy to reduce emissions and bring down the exceedance of critical loads for nitrogen deposition. I think it is worth mentioning this here.

Reply: This issue is rather complicated. As noted by our paper and the referee, the increased NH_3 deposition implies more local control. On the other hand, we note on line 25 of this page that consideration of more complex bi-directional exchange might increase the transport distance of NHx. Engardt and Langner (2013) also estimated longer lifetimes of S and NOy compounds in the future, thus increasing the international transport of some particles.

In order to avoid a very long discussion of issues that clearly need more research, we have re-arranged the last paragraph on p6686, now also including:

With regard to emissions control strategies, the increased NH_3 deposition noted above (and in e.g. Engardt & Langner 2013) implies that local control measures might become more effective. On the other hand, Engardt and Langner (2013) also estimated longer lifetimes of S and NOy compounds in the future, thus increasing the international transport of some particles.

2 Referee #2

The impact of climate change and anthropogenic emission scenarios upon future air quality is an important topic for scientists and policy makers. The issue of how reactive nitrogen deposition may change in the future is a subject containing many uncertain- ties and variables that clearly requires further analysis. This article describes how an ensemble of four chemistry transport

models was used to investigate future nitrogen deposition as a result of decreased NOy emissions, increased NH3 emissions, and climate change. The article is well-written and well-organised and presents interesting results while highlighting the areas that require further study. I would like to recommend this article for publication in ACP.

Reply: We thank the referee for these comments.

2.1 Specific Comments

In Section 2.1, it is stated that the current legislation (CLE) scenario was used for future emissions, but the representative concentration pathway (RCP) 6.0 was used for ship emissions. For further clarity, please explain how the CLE scenario compares to the RCP scenarios.

Reply: The RCP scenarios were aimed mainly at climate modellers with a focus on CO_2 emissions and radiative forcing targets. It proved very difficult to relate these to air quality precursor emissions, especially at the national scale. As explained in Amann et al. (2012, 2013), the IIASA GAINS model was therefore used to make more detailed estimates, taking into account national legislation. The GAINS current legislation estimate (CLE) was that adopted for EU ECLIPSE and PEGASOS projects and the UN-ECE Task Force on Hemispheric Air Pollution. (As even this CLE estimate changes with time, we have added the notation ECLIPSE v.4^e to the text, to distinguish from newer estimates.)

We have added text to explain these emissions and the relation to RCP – see the answer to Tony Dore's query (1) above.

The choice of RCP6 for shipping was made in discussions with IIASA on the most appropriate emission pathway (in terms of CO_2 emission) to match their CLE scenario. This choice was also harmonised across the projects mentioned above. We will add some brief text to note this.

On page 6670, line 15, the authors state that the future increase in NH3 emissions have the potential to offset many of the beneficial effects of European NOy emission reductions. Please clarify that this is expected to be a partial offset

Reply: Yes, we will make this change

2.2 technical comments

Reply: We will correct the typos and hyphenation issues as suggested by the referee. Thanks for careful reading!

3 Comments from Editor, Eiko Nemitz

We reply here to a few comments made by the Editor.

a) It may be worth showing the differences in emissions that were not harmonised between models (p.6669, Lines 23-25Lines 137-140)?

Reply: Langner et al 2012b give a Table and plot of these emissions, so we simply added some text:

(Differences in isoprene emissions were indeed substantial, ranging from ca. 1600–8000 Gg/yr as annual average for the models used here, see Langner et al. for details and more discussion).

b) The effect of the change in NH3/NH4+ on transport distance could be quantified and discussed in more detail. For example, the export from Europe and the country-to-country transport is likely to go up. What are the relative importance of decreases in SOx/NOx emission and temperature increase in changing the gas/aerosol partitioning.

Reply: We have addressed this to some extent in the reply to Tony Dore above. In addition, Engardt and Langner (2013) discussed changes in atmospheric lifetime of sulphur and nitrogen components as a result of emissions and climate change, but all such studies respond to many changes (temperature, precipitation, boundary layer properties, etc.) and we cannot extract specific response to e.g. temperature. In general, sulphur and oxidised nitrogen is expected to have longer atmospheric lifetimes in the future thereby increasing the international transport. Reduced nitrogen, on the other hand, becomes more short-lived, thereby increasing the local deposition and making national actions more relevant for securing the environment in the respective country. However, even such statements need qualification, as noted in the last paragraph on p6686 and the reply to Referee 1, point (7).

c) The results should be compared more with the previous studies mentioned in the introduction.

Reply: We have added text at the end of section 3.3:

The magnitude and distribution of changes in Nr deposition over Europe is sensitive to which climate projection that is used. Engardt and Langner (2013) compared three different climate projections (including the one used here) using the MATCH model and found changes due to climate change until 2050 less than 1 kg(N)/ha for both NOy and NHx. These changes are

comparable to the ensemble mean changes presented here. Hedegaard et al. (2013) reported a general reduction in the Nr deposition over Europe above 0.2 kg(N)/ha due to climate change in the period 1990 to 2090 using the hemispheric DEHM model. This could be compared to the case with changing BCs and changing climate in this study which gives an increase in central/southern Europe for NOy and a more widespread increase for NHx. These differences in results are however small enough to be explained by differences in the climate projection used. Engardt and Langner (2013) also reported changes in Nr deposition due to emission changes until 2050 using the RCP4.5 scenario. The reductions in deposition are comparable to those reported here for NOy but for NHx the distribution of the changes are different primarily due to differences in the emission data.

d) Figure 10. The sum of NH_3+NH_4^+ appears to be the same in the E50-M560-BC3 run as in the other runs. The change in partitioning in favour of NH_3 converts a slowly depositing compound into a fast depositing compound and should reduce the NHx burden?

Reply: This comment is of course correct, but spatial variability masks this in Fig. 10. A proper discussion and evaluation of this would be rather complex (and involve full 3-D budgets that we unfortunately do not have). However, we have added the following text to Sect. 3.4:

According to Fig. 10, the sum (NHx) of NH_3+NH_4^+ is approximately constant from the year 2000s to the 2050s scenario. However, Fig. 10 shows averages over a large area. In fact, as seen in Fig 11, the deposition of NHx (NH_3+NH_4^+) decreases in most parts of western Europe (especially France), and increases in many parts of central and eastern Europe (see also Figs 2b for emissions). The EU28+ area includes areas in both regimes.

New references

Amann et al., TSAP Report #1, Future emissions of air pollutants in Europe . http://webarchive.iiasa.ac.at/Admin/PUB/Documents/XO-12-011.pdf, 2012

EMEP/CCC 2014. Results from EMEP laboratory intercomparison. Results published on web: http://www.nilu.no/projects/ccc/intercomparison/index.html

Hansen, K., et al., Trender i kvvenedfall ver Sverige 1955-2011. IVL Rapport B 2119, 86 pp (In Swedish, abstract also in English). http://www.ivl.se/download/18.372c2b801403903d2757ade/1383582083173/E 2013.

Jonson, J. et al., Can we explain the trends in European ozone levels? Atmos. Chem. Physics, 2006, 6, 51-66

Schulz et al., Modelling and evaluation of trends in the EMEP network, EMEP Report 1/2013, Norwegian Meteorological Institute, 85-104, 2013.

Winiwarter, et al., Future scenarios of nitrogen in Europe, in European Nitrogen Assessment (Sutton et al., eds.) Chapter 24 , 551-569, 2011

3.1 Corrected Table, Figure

In double-checking the data behind Fig.10 we found that the averaging area was from a larger domain than EU28+. This also applies to the numbers in Table 5. There is no major change to either in terms of differences between models or components, but the values are somewhat higher since we now include more of the highly polluted EU28 area. The revised manuscript will include these new data and Figure. We give them here:

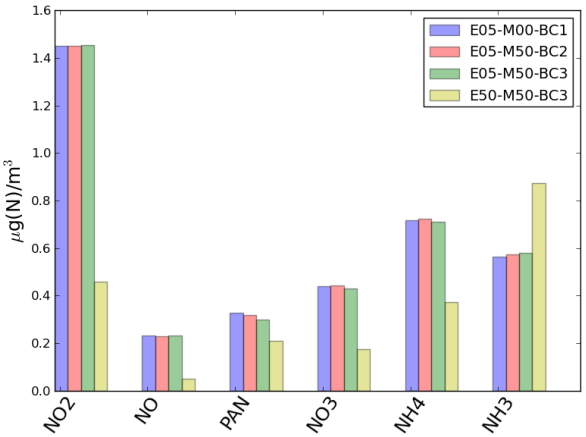

Fig. 10, revised: Calculated concentrations of major Nr species from the EMEP MSC-W model for four scenarios.

Table 5. Base-case depositions of Nr ($\mathrm{kg\,(N)\,ha^{-1}}$) components for the four CTMs, along with the 3CTM-ensemble mean and spread. Values are average depositions over the EU28^{+} ~~the inner~~ domain. ~~(see Fig. 1)~~

	DEHM	EMEP	MATCH	SILAM	3CTM-Ensemble mean	3CTM-Ensemble spread (%)
TDEP-Nr	11.9	8.5	9.7	9.3	9.2	13
TDEP-NO_y	4.9	3.7	4.6	5.1	4.5	32
TDEP-NH_x	7.0	4.8	5.1	4.2	4.7	21
WDEP-Nr	5.8	5.5	6.4	6.5	6.1	16
WDEP-NO_y	2.5	2.5	3.0	3.6	3.0	39
WDEP-NH_x	3.3	3.0	3.4	2.8	3.1	18
DDEP-Nr	6.1	3.0	3.3	2.8	3.1	17
DDEP-NO_y	2.4	1.3	1.6	1.5	1.5	24
DDEP-NH_x	3.7	1.8	1.7	1.3	1.6	29

Notes: the 3CTM ensemble consists of the three European-scale CTMs driven by RCA3. Spread is defined as $(\max - \min)/\mathrm{mean}$ of these 3 models.