Referee #1

We thank the two reviewers for their helpful comments and suggestions which significantly improved our paper.

This is a solid paper that accumulates the evidence for an important role of nitrate aerosol in present and future conditions. I particularly appreciate the careful job that the authors did trying to understand similarities and differences with other studies. The emphasis on the key role of agricultural (NH3) emissions in future is justified, but perhaps some more emphasis should be placed on the strong limitations resulting from the use of the RCP emissions dataset. For almost all pollutant emissions, except for SO2, they are to some extent assuming Kuznets assumptions, and it is very questionable what pollution levels in 2100 could be. Why not focus on 2050, a somewhat more foreseeable future?

In this paper we indeed focus on the RCP scenarios which have been the reference for the CMIP5 and ACCMIP simulations. Our choice for this first paper was to put the nitrate forcing in the context of these recent exercises on which the community concentrated over the past years. The RCP limitations for air quality are indeed clear and we mention them. For this paper we have simulated 2030, 2050 and 2100. The key figures of the article illustrate these three time horizons, again to put our results in the context of the CMIP5 and ACCMIP simulations. Several figures (now in the supplement) illustrate 2030 and 2100. 2030 (or 2050) are indeed more seeable futures. 2100 is also interesting despite the uncertainty on emissions in a context of a changing climate which will be more pronounced at the end of the century. We are now working on new simulations with new scenarios prepared by the IIASA institute in the framework of the ECLAIRE EU project. These scenarios are more 'realistic' in terms of air quality legislation than the RCPs and focus on 2030 and 2050. Results from this ongoing work will be presented in forthcoming publications.

On the more technical level, the authors have formulated a simplified thermodynamic aerosol formation scheme. While it has been demonstrated in the past (e.g. Metzger et al), that such parameterizations are relatively accurately mimicking more accurate schemes, such evaluation is missing in the present manuscript. I suggest that the authors present such evaluation in the supplement, especially because the whole paper is building on the accuracy of this scheme. Some simple plots scanning concentration/T/RH space would be indicative of the model's performance.

Based on this interesting suggestion, we have performed box model simulations and compared the results to the ISORROPIA model results. For these simulations we vary the key parameters such as T, RH, total sulfate, total ammonia, total nitrate and compare the sensitivity of the two models. The comparison is quite encouraging and provides good confidence in the INCA model performance despite the simplified treatment. These results are mentioned in section 2.2 and are presented in the supplementary material under figure S1.

The amount of figures is quite excessive (26). On the hand there will be always somebody who will appreciate particular plots- but it may preclude others to start reading the paper. I would encourage the authors to place less essential plots in the supplementary material, and work on some good summary plots and tables. For instance I have seen a number of scatter plots, but it difficult to grasp if the overall picture is consistent among these figures.

Yes we agree. We have significantly reduced the number of plots in the main paper from 27 to 16. These former figures are now in the supplementary material. In addition, a table has also been *introduced (Table 2) summarizing the correlations between model results and measurements. The correlation plots are now in the supplement as well.*

With regard to the measurements evaluation, especially the ammonium-nitrate-sulfate system, the authors should be more explicit on the limitation of the measurements that they have used. For instance it is known that filter-pack measurements at temperatures above 20 C, are prone to large sample losses, and at most present a lower limit to the model. Can this explain the overestimates of nitrate/nh4 reported in the paper? Likewise an evaluation sub-regions- (perhaps in a table, regions from Chin et al??) could give more specific information on issues and model performance. We don't need more figures- but something that could summarize North-South differences in Europe/USA, for instance.

The reviewers are correct in pointing to the measurement limitations. We have added the following text in the supplementary material where the model-measurement plots have been moved:

To obtain a large spatial coverage with respect to evaluation of nitrate and ammonium model values we have included measurements from different methods. These use different combinations of filterpacks and denuders. Little harmonization of the methods has been achieved globally and there is virtually no choice to pick just one method. Depending on filter type, sampling set-up, temperature and changing ambient conditions during a sampling interval considerable sampling artefacts may occur. At higher temperatures, evaporation of ammonium nitrate from filters has been shown to lead to losses of up to 50% in summer conditions (Vecchi et al., 2009; Sickles and Shadwick, 2002; Allegrini et al., 1994; Yu et al., 2005, Hering and Cass, 1999; Chow et al., 2005, Ashbaugh and Elfred, 2004, Schaap et al., 2004, Schaap et al., 2002). Such sampling artefacts may explain in total a positive model bias, while negative model bias clearly points to model errors. We do not find a clear north-south gradient in bias, neither in Europe nor in Northen America, higher temperatures in the south are not associated with higher positive model bias. However, in central Europe there are several sites, where the positive bias of the model is high in summer and almost absent in winter. In depth inspection of nitrate bias on a map and per measurement site may be accessed via the AeroCom web interface (http://aerocom.met.no/cgi-bin/aerocom/surfobs_annualrs.pl?PROJECT=INCA &Run0=LOI_DH10n&Parameter0=SCONC_NO3).

We also added a remark in section 3.1 and in the conclusion:

A positive bias in simulated nitrate aerosol is suspected to be partly linked to negative sampling artefacts in measurements, because evaporation of ammonium nitrate has been frequently reported to create occasionally losses of up to 50%, in particular in warm weather. Further work is needed to better characterize the individual nitrate measurement error, to see where modeled nitrate is consistent with measurements.

The following citations have been added in the supplement:

Allegrini, I., A. Febo, C. Perrino and P. Masia. "Measurement of Atmospheric Nitric-Acid in Gas-Phase and Nitrate in Particulate Matter by Means of Annular Denuders." International Journal of Environmental Analytical Chemistry 54, no. 3 (1994): 183-201.

Ashbaugh, L. L. and R. A. Eldred. "Loss of Particle Nitrate from Teflon Sampling Filters: Effects on Measured Gravimetric Mass in California and in the Improve Network." Journal of the Air & Waste Management Association 54, no. 1 (2004): 93-104.

Chow, J. C., J. G. Watson, D. H. Lowenthal and K. L. Magliano. "Loss of Pm2.5 Nitrate from Filter Samples in Central California." Journal of the Air & Waste Management Association 55, no. 8 (2005): 1158-1168.

Hering, S. and G. Cass. "The Magnitude of Bias in the Measurement of Pm2.5 Arising from Volatilization of Particulate Nitrate from Teflon Filters." Journal of the Air & Waste Management Association 49, no. 6 (1999): 725-733.

Schaap, M., K. Muller and H. M. ten Brink. "Constructing the European Aerosol Nitrate Concentration Field from Quality Analysed Data." Atmospheric Environment 36, no. 8 (2002): 1323-1335.

Schaap, M., G. Spindler, M. Schulz, K. Acker, W. Maenhaut, A. Berner, W. Wieprecht, N. Streit, K. Muller, E. Bruggemann, X. Chi, J. P. Putaud, R. Hitzenberger, H. Puxbaum, U. Baltensperger and H. ten Brink. "Artefacts in the Sampling of Nitrate Studied in the "Intercomp" Campaigns of Eurotrac-Aerosol." Atmospheric Environment 38, no. 38 (2004): 6487-6496.

Sickles, J. E. and D. S. Shadwick. "Biases in Clean Air Status and Trends Network Filter Pack Results Associated with Sampling Protocol." Atmospheric Environment 36, no. 29 (2002): 4687-4698.

Vecchi, R., G. Valli, P. Fermo, A. D'Alessandro, A. Piazzalunga and V. Bernardoni. "Organic and Inorganic Sampling Artefacts Assessment." Atmospheric Environment 43, no. 10 (2009): 1713-1720.

Yu, X. Y., L. Taehyoung, B. Ayres, S. M. Kreidenweis, J. L. Collett and W. Maim. "Particulate Nitrate Measurement Using Nylon Filters." Journal of the Air & Waste Management Association 55, no. 8 (2005): 1100-1110.

The authors mention a potential issue with heterogeneous reactions of N2O5, which is much lower than in other studies, but even compared to the same model (Bauer et al. 2007). I appreciate the honesty, but it is something that has to be clarified, since it could influence the model performance especially in winter.

We do not really see it as an issue but more a difference we found comparing our budgets and HNO_3 formation terms with those presented by Xu and Penner (2012). In our case the formation from heterogeneous reaction of N_2O_5 on sulfate aerosols is 4 TgN to be compared to 18 TgN in Xu and Penner. Bauer et al. (2004) used the LMDz-INCA model but in a very different version since the NMHC chemistry and the role played by gas phase NO_3 or PANs for instance was not accounted for in this early version. With their version this term was 19 TgN. Interestingly, writing this reply, we realized that Xu and Penner also used a simplified nitrogen chemistry with fixed oxidants and without considering the role played organics. In addition, the reaction probability γ for the reaction of N_2O_5 hydrolysis has also been significantly updated from this very early version of the model. Therefore we have the feeling this discrepancy with Xu and Penner we mentioned is not an issue and the formation of HNO_3 has been higher in previous work based on different and simplified treatments used for gas phase chemistry and reaction probability. We also realized that it was unclear which aerosols where actually considered for this hydrolysis of N_2O_5 in Xu and Penner. In our case, only sulfate aerosols are concerned. Moreover, as mentioned by Evans and Jacob (2005) who used an even smaller γ coefficient, there is still a large uncertainty on this reaction probably and on the role of this reaction. A few sentences have been added in order to mention briefly these elements in the paper.

Finally, I appreciate the attempts of the authors to compare to other studies. Nevertheless it remains difficult to do this comparison more systematically due to a lack of standard evaluation sets and methods. To my opinion AEROCOM could play a more a substantial role in defining such benchmark sets- that would allow easy comparison of model results (beyond the joint exercises that take place from time to time), and monitor progress. I would appreciate some paragraph on this as well.

Yes. We agree on the important role that AeroCom could play in this type of model intercomparison for nitrates and the role of AeroCom to reduce uncertainties in the nitrate particle budget is mentioned in our conclusion. The internet link to the model intercomparison protocol currently underway within AeroCom is now provided. The results from these simulations will be presented in the framework of the AeroCom conferences and in forthcoming papers.

Despite these criticisms, I recommend the paper to be published in ACP, as a welcome addition to the not very extensive literature on nitrate aerosol, after taking into account the remarks above, and the detail comments below.

Detailed comments:
p. 6864 l. 8 same magnitude as ; represent=>compares to

The text has been clarified.

p. 6864 l. 18 All nitrates, or mainly NH4NO3?

Yes ammonium nitrates. This is now specified.

p. 6864 l. 20 RCP scenarios only?

Yes, only RCPs have been used in this paper. Other scenarios are currently being introduced in the model.

p. 6867 l. 7 The thermodynamic scheme is used in some regional/global models. Was it evaluated against more comprehensive scheme? How good was it?

We have now added such an evaluation of the thermodynamic model results against the ISORROPIA results by varying most of the input parameters. This evaluation is now summarized in the supplementary material by Figure S1. A reference to this evaluation is also provided in the model description section.

p. 6868l. 5-20 What is the thickness of the surface layer? How can the mixing of the model be characterized. These are essential parameters to understand NH3.

In this 19 level version the first level is located at 70 m. In several papers, the transport and mixing of tracers in the LMDz GCM have been investigated and evaluated against obervations for both inert tracers (e.g., Hourdin and Issartel, 2000; Hauglustaine et al., 2004; Rivier et al., 2005) and in the framework of inverse modelling (e.g., Bousquet et al., 2005; Pison et al., 2009; Bousquet et al., 2010). These studies concluded that the model at this vertical resolution is well suited for the transport of tracers and chemical species. However it was also pointed out that the simulated inter-hemispheric exchange time is 1.1 years for fossil CO2 (Hauglustaine et al., 2004), in the lowest part of the range (1.1–2.1 years) provided by TRANSCOM1 model inter-comparison (Law et al., 1996; Rayner et al., 1995). This means that LMDZ has one of the fastest inter-hemispheric mixing among the models of TRANSCOM 1 and efficient mixing and transport. This limitation has been added in the model description.

p. 6870 l. 11 I am not sure, but to me equation 3 just looks like a rewrite of equation 2? Or is the point that the factor Beta is independent evaluated?

The text has been clarified to keep only the two key equations.

p. 6871 Given the equations in R1 to R9: what is the number of N_2O production?

We added a short paragraph on N_2O production and comparison with Dentener and Crutzen (1994) numbers when we discuss the actual Table 4. We find a production of 0.6 TgN/yr based on the budget presented in our actual table 4 providing the budget for NH_3. This number is similar to the previous estimate.

p. 6872/6873 could probably move to supplement together with a more thorough evaluation.

The parameterization used for the equilibrium constant based on Mozurkewich (1993) has been moved to the Supplementary Material with the box model evaluation against ISORROPIA as suggested by the reviewer.

p. 6874 9- and further Only on dust and seasalt? Did the authors consider uptake on sulfate? And on Nh_4NO_3?

The reaction on sulfates and NH_4NO_3 formation is only considered for the formation of accumulation mode nitrate particles. In order to form coarse nitrates particles we consider the reaction on dust and sea-salt. In this model only two very distinct classes are considered as described in 2.1 and references therein.

p. 6877 I assume one year was used as spin-up?

Yes. The year 2005 is used as a spin-up. This has now been specified in the paper.

p. 6878 I found the color scheme giving too little information in the regions below 1 ug/m3

Sorry. We have tried to keep a color scale suitable for most of the surface plots for the various gaseous and particulate species. We found this one as the best compromise among all others we have tried. In the original plots we have, the green between 0.2-0.5 and the green for 0.5-1 are more different than on the pdf version. We will provide the original postscript files for the final version.

p. 6878 It is important to tell how the data were used. E.g. can summer measurements on filters be used? Is there a threshold of # days to calculate a monthly average. Were there quality issues, and how were they used?

A monthly average is constructed from at least 2 days of measurements, but the filter measurements used cover almost at all stations more than 90% of the days in a given month. Temporal coverage is more a problem at some AERONET sites, when cloudy conditions prevail. Note that we have used matching daily data, which means that in a given month at a given site the same number of days form model and observation are used for the monthly averages. With respect to data quality: see added text in response to general remark above.

p. 6879 l. 29 and further. The authors seem to suggest that it is physics of global models that are causing the issue. What about resolution? What about measurements that may be biased? This aspect needs to be more expanded also for abstract and conclusions.

It is indeed quite a challenge to represent nitrates in a global model with coarse resolution, simplified chemistry and also simplified mixing and transport. The emission dataset is also crucial. We agree that the uncertainty on the measurements is not to be neglected but clearly regional models succeed better to represent the distribution of particles and nitrates in particular than global models. The uncertainty on the measurement is however now mentioned in the paper in reponse to the comment above. The uncertainty associated with the resolution, the simplified aerosol scheme, and physics has been added in this specific paragraph as well.

p. 6882 l. 27 I am confused about the sulfate remark, since previously the authors only talk about reactions with dust and seasalt.

The standard version of the LMDz-INCA model as described by Hauglustaine et al. (2004) and Folberth et al. (2006) includes the formation of HNO3 from the reaction of N2O5 on aerosols. We have emphasized the reference to these papers in Section 2.1.

p. 6883 l 4 ensured=>caused, or most nitrate is lost by wet deposition

Yes. Text corrected.

p. 6885 cloudiness formed?

No. We changed the text to specify that only the direct radiative forcings of aerosols are considered and hence the cloudiness is not affected by the presence of aerosols in this version of the model.

p. 6888 Future evolution of what?

Text modified : Future evolution of nitrate aerosols

p. 6893 It is good that the authors perform sensitivity studies, but we should realize that the RCP scenarios are not internally consistent. Possibly more useful is to increase/decrease within a scenario the emissions of specific components.

These sensitivity scenarios are only intended to illustrate the most important emissions explaining the differences seen in the two extreme RCP scenarios and the relative importance of NO_x, NH_3 *and* SO_2 *emissions. We found these tests useful to, at least, better understand our results. Of course we agree that other scenarios could be used and tested in forthcoming studies.*

p. I found the ‘future’ section relatively lengthy- and as I explained before, very dependent on relatively similar scenarios. Perhaps this is a section that could be reduced, while retaining the main points: the increasing importance of nitrate under future conditions.

This section has been significantly reduced and a lot of figures and related text moved to the Supplementary Material.

Referee #3

We thank the two reviewers for their helpful comments and suggestions which significantly improved our paper.

In this study, the authors document the addition of a nitrate aerosol scheme in the LMDz-INCA model, and use that model to highlight the dramatic change in aerosol composition that would happen over the 21st century if RCP are followed. The paper is well written and presents interesting results, notably that ammonium burden remains fairly constant in the future as ammonium nitrate formation replaces ammonium sulphate formation; that ammonia is the limiting factor explaining differences between different RCPs; and that the change in aerosol composition has important consequences on direct radiative forcing (Figures 24, 26, and 27). The authors also do a good job at placing their study within the existing scientific literature, although they could be bolder in highlighting the strengths of their study.

For those reasons, I recommend publication of the manuscript, with minor changes to address the comments below.

1 Main comments

- The authors state several time that their results replicate those by previously published papers. Replication is important, but the authors should evaluate previous results more critically. For example, some of the previous studies did not include coarse-mode nitrate, or did not model ammonium explicity. Do those limitations matter? The scheme implemented by the authors is fairly complete – at least from a global modelling point of view – and I believe the results in this paper can be seen with more confidence than some of the previous, more simple, analyses.

Thank you for this encouraging comment. The recent nitrate models by Xu and Penner (2012), Bellouin et al. (2011), and Pringle et al. (2010) were actually already fairly detailed and in the case of Xu and Penner (2012) quite detailed in their nitrate chemistry. We do not pretend to have a more complex model. However, Xu and Penner used a very simplified nitrogen chemistry excluding NMHC chemistry and with imposed monthly mean OH and O_3 *fields and didn't investigate the RCP future evolution of nitrates and the associated future forcing. Bellouin et al. look into this but didn't focus too much on the chemistry evolution and did not include the coarse particle formation. This can be quite important since* HNO_3 *is used to form coarse nitrate particles instead of fine particles and hence this reduces the nitrate forcing for present-day conditions. This is an interesting aspect of our work in addition of applying the model for future evolution and looking into the relative contribution of nitrates to the forcing. This added value of our study has been added at several places in the manuscript and in particular in the abstract, conclusion, and when the present-day forcing of nitrate is mentioned. The fact that Xu and Penner used a simple chemistry is also mentioned now since we overlooked this limitation in their work.*

- One defining characteristics of nitrate aerosols is their ability to easily dissociate back into the gas phase. This means that nitrate aerosols have a very pronounced diurnal cycle, spending most of daytime hours in the gas phase (Dall'Osto et al., 2009). This is crucial for their direct radiative effects, which is exerted by the aerosol phase only. Previous studies have overlooked that aspect, so it would be very interesting to add a short discussion of the diurnal cycle in LMDz and demonstrate that it follows the observed behaviour.

This is an interesting suggestion. Even if it is quite out of the scope of this paper to evaluate a global model on such a fine temporal scale, we have output the surface concentrations with a 30 min time step. Interestingly, a diurnal cycle with minimum concentrations of NO_3 *during daytime is obtained at several stations and coincides with peak* HNO_3 *concentrations. Even if we don't pretend to perform a detailed comparison with Dall'Osto et al. (2009), the model results at a similar location and time of year than the performed measurements show very similar features, variability and concentration* *range. A few words on this are now given in Section 3.1 and a figure provided in the supplementary material (Fig. S2).*

- There is a large number of figures in the paper, some of which being only discussed briefly in the text. I would suggest merging some of the figures together, especially Figures 1, 2, and 6; Figures 4 and 5; Figures 11 and 12; and possibly Figures 22 and 23.

Yes we agree. We have now significantly reduced the number of figures in the main paper from 27 to 16. The figures which were not key to the discussion are now in the supplementary material together with new figures requested by the reviewers. A table has been introduced (Table 2) which summarizes the model-measurement comparisons and the correlation plots sent to the supplement.

- Furthermore, the same kind of information (change in surface concentrations) is presented in different ways across Figures 16 to 18. The reason for that choice is unclear: Why doesn't Figure 17 show RCP4.5? Why does Figure 18 show timeseries rather than distributions? With regard to the subject matter, a Figure similar to Figure 21, but showing in addition regional changes, would be very helpful. By showing the evolution of all the species involved in nitrate formation (NH_3, HNO_3, SO_4) it would provide an efficient way of identifying the causes for the change in nitrate concentrations.

Most of these figures showing future changes in atmospheric composition are now in the supplement. The change in surface sulfate concentration for scenario RCP4.5 has been added to the former Figure 17 (now Fig. S9). We also added a new figure (Fig. S10) showing the map of changes in nitrate surface concentration in 2030 and 2100 for both RCP8.5 and RCP4.5. All these figures showing the regional changes help to have a better insight into the future changes of nitrate particles under the RCP scenarios.

2 Other comments

Page 6866, line 1: Are the radiative forcings reported in this sentence for the direct effect only, or is it a mixture of direct-only and direct+indirect studies?

The text has been clarified. Since we focus only on direct forcings in this paper, those are for the direct forcing only.

Page 6868, line 8: Out of curiosity, what does the z in LMDz stand for?

'z' stands for 'zoom'. But this capability of the model to zoom over a specific area is not used in the present version.

Page 6869, lines 6–7: The distinction between anthropogenic and natural aerosols made here is a bit arbitrary, since sulphate aerosols can be both, for example. I suggest rephrasing that statement.

Absolutely. The text has been clarified.

Page 6872, line 1: It is worth stating here that the fact that sulphate formation takes precedence over that of nitrate is justified by the low vapour pressure of sulphuric acid.

Text modified accordingly.

Page 6874, line 13: In the introduction, limitations to the first-order removal method have been mentioned, with Feng and Penner (2007) as a reference. In section 2.2, it would be useful to shortly summarise what those limitations are.

We have modified to text to mention two major limitations of the formulation: the relative humidity and aerosol chemical composition dependences. The method used to partly overcome these limitations are then described.

Page 6878, lines 4 and 23: Does "agreement" refer to patterns, magnitudes, or both?

It is more the patterns which agree since the magnitude can vary significantly among the various studies mentioned. The text has been clarified.

Page 6878, line 19: What do the authors mean by "anthropogenic nitrates" in that context?

Fine mode nitrates. The text has been corrected.

Page 6879, line 24: Another difficulty when comparing with measurements of nitrate aerosol concentrations is that they are difficult to measure, because nitrate aerosols dissociate easily. See for example Schaap et al. (2002). It is likely that those difficulties translate into underestimated concentrations in the observations.

The reviewers are correct in pointing to the measurement limitations. We have added the following text in the supplementary material where the model-measurement plots have been moved:

To obtain a large spatial coverage with respect to evaluation of nitrate and ammonium model values we have included measurements from different methods. These use different combinations of filterpacks and denuders. Little harmonization of the methods has been achieved globally and there is virtually no choice to pick just one method. Depending on filter type, sampling set-up, temperature and changing ambient conditions during a sampling interval considerable sampling artefacts may occur. At higher temperatures, evaporation of ammonium nitrate from filters has been shown to lead to losses of up to 50% in summer conditions (Vecchi et al., 2009; Sickles and Shadwick, 2002; Allegrini et al., 1994; Yu et al., 2005, Hering and Cass, 1999; Chow et al., 2005, Ashbaugh and Elfred, 2004, Schaap et al., 2004, Schaap et al., 2002). Such sampling artefacts may explain in total a positive model bias, while negative model bias clearly points to model errors. We do not find a clear north-south gradient in bias, neither in Europe nor in Northen America, higher temperatures in the south are not associated with higher positive model bias. However, in central Europe there are several sites, where the positive bias of the model is high in summer and almost absent in winter. In depth inspection of nitrate bias on a map and per measurement site may be accessed via the AeroCom web interface (http://aerocom.met.no/cgi-bin/aerocom/surfobs_annualrs.pl?PROJECT=INCA &Run0=LOI_DH10n&Parameter0=SCONC_NO3).

We have also added a remark in section 3.1 and in the conclusions:

A positive bias in simulated nitrate aerosol is suspected to be partly linked to negative sampling artefacts in measurements, because evaporation of ammonium nitrate has been frequently reported to create occasionally losses of up to 50%, in particular in warm weather. Further work is needed to better characterize the individual nitrate measurement error, to see where modeled nitrate is consistent with measurements.

The following citations have been added:

Allegrini, I., A. Febo, C. Perrino and P. Masia. "Measurement of Atmospheric Nitric-Acid in Gas-Phase and Nitrate in Particulate Matter by Means of Annular Denuders." International Journal of Environmental Analytical Chemistry 54, no. 3 (1994): 183-201.

Ashbaugh, L. L. and R. A. Eldred. "Loss of Particle Nitrate from Teflon Sampling Filters: Effects on Measured Gravimetric Mass in California and in the Improve Network." Journal of the Air & Waste Management Association 54, no. 1 (2004): 93-104.

Chow, J. C., J. G. Watson, D. H. Lowenthal and K. L. Magliano. "Loss of Pm2.5 Nitrate from Filter Samples in Central California." Journal of the Air & Waste Management Association 55, no. 8 (2005): 1158-1168.

Hering, S. and G. Cass. "The Magnitude of Bias in the Measurement of Pm2.5 Arising from Volatilization of Particulate Nitrate from Teflon Filters." Journal of the Air & Waste Management Association 49, no. 6 (1999): 725-733.

Schaap, M., K. Muller and H. M. ten Brink. "Constructing the European Aerosol Nitrate Concentration Field from Quality Analysed Data." Atmospheric Environment 36, no. 8 (2002): 1323-1335.

Schaap, M., G. Spindler, M. Schulz, K. Acker, W. Maenhaut, A. Berner, W. Wieprecht, N. Streit, K. Muller, E. Bruggemann, X. Chi, J. P. Putaud, R. Hitzenberger, H. Puxbaum, U. Baltensperger and H. ten Brink. "Artefacts in the Sampling of Nitrate Studied in the "Intercomp" Campaigns of Eurotrac-Aerosol." Atmospheric Environment 38, no. 38 (2004): 6487-6496.

Sickles, J. E. and D. S. Shadwick. "Biases in Clean Air Status and Trends Network Filter Pack Results Associated with Sampling Protocol." Atmospheric Environment 36, no. 29 (2002): 4687-4698.

Vecchi, R., G. Valli, P. Fermo, A. D'Alessandro, A. Piazzalunga and V. Bernardoni. "Organic and Inorganic Sampling Artefacts Assessment." Atmospheric Environment 43, no. 10 (2009): 1713-1720.

Yu, X. Y., L. Taehyoung, B. Ayres, S. M. Kreidenweis, J. L. Collett and W. Maim. "Particulate Nitrate Measurement Using Nylon Filters." Journal of the Air & Waste Management Association 55, no. 8 (2005): 1100-1110.

Section 3.1, and Figures 2,4,5,6: It is worth clarifying early in the text and figure captions the units used for concentrations and burdens, i.e. whether they are given as [S] or [N], or as $[SO_4]$, $[NO_3]$, etc.

We have now specified at the beginning of Section 3.1 that all concentrations and burdens are expressed in mass of the species (e.g. $\mu g/m^3$ *for* $\mu g\text{-}NO_3/m^3$*) unless otherwise stated (e.g.,* $\mu g\text{-}N/m^{-3}$ *or* $\mu g\text{-}S/m^3$*).*

Page 6881, line 12: I'm not sure I understand the statement "nitrate on pure sulfates" here. By redoing the calculation, it seems that it means "fine mode nitrates".

Yes. The text has been modified accordingly.

Page 6886, line 13: A global-, annual-averaged total AOD of 0.059 is a surprisingly small number. Is that really correct? Looking at Table 5, it seems it is in fact not the total AOD, but excludes mineral dust and sea-salt aerosols.

The value of 0.059 is indeed for accumulation mode. The total AOD is 0.1355. The text has been corrected.

Figure 13a: looking at Figures 5, I would have expected a more equal contribution of fine and coarse mode nitrate to total column around 50N, but it is difficult to judge by eye.

We double checked the figure and the results are correct. Please note the coarse mode shown on Figure 13a is the sum of dust particles (Fig. 5b) and sea-salt particles (Fig 5c).

Figure 13b: Has the y-axis really been multiplied by 100? The magnitudes and the lines 18–20 on page 6887 suggest that is not the case.

The figure is correct but the text didn't account for the x100 factor! The text has been corrected.

Page 6891, lines 17–25: The discussion of Figure 20 is confusing, since it mixed hemispheric averages (which decrease) with regional changes that are of varying signs. It would be clearer to say that although Figure 20 seems to show an increase in nitrate column in most regions of the north hemisphere, that increase is in fact more than compensated by strong decreases over Europe and China, where present-day nitrate concentrations are large. Having said that, I am surprised by how localised the European and Chinese changes are: Why don't they extend downwind?

This figure has been moved to the supplementary material (Fig. S10) and we have added another figure showing maps of the change in nitrate surface concentration (Fig. S11). The text has been clarified according to the reviewer's comment. This change in surface nitrates shows are localized the decreases are over Europe and Asia due to a strong reduction in HNO_3 *concentrations and hence nitrate formation. Since nitrates are short-lived, these changes remain localized to these regions and are not transported aloft or only to a small extend.*

Page 6895, lines 5–8: I would rephrase this statement slightly as "because deposition of nitric acid, which dissociates readily in water, causes a significant drop in pH, but deposition of NHx increases the water alkalinity."

The statement has been rephrased accordingly.

Page 6901, line 16: At this stage, I would think that understanding the sources of uncertainty is a more likely outcome of an intercomparison project than reducing those uncertainties.

Yes, the sentence has been modified accordingly.

3 Technical comments

The level of English language is very good, but there are recurring mistakes when plurals are used as adjectives, and a few other grammatical mistakes. To list but the first instances:

- Page 6864, line 2: fine nitrate particle formation

- Page 6865, line 2: nitrate exerts; line 9: nitrogen oxide emissions; line 10: soil emissions.

- Page 6870, line 14: organic carbon

I encourage the authors to pay particular attention to these matters when proof-reading the paper.

Thank you. We paid attention to those and other additional mistakes.