

Response to Referees Comments

AR: Authors' response

Anonymous Referee #1

This paper presents an evaluation of modeled trends in wet deposition of sulfur and nitrogen compared to observations from the EMEP network for the periods 1990-2000 and 2000-2010. The paper provides a very detailed analysis of the trends, including examining factors contributing to model performance. Overall, the paper is well written, but in some sections becomes a bit of a recitation of statistics with little analysis. Section 3.6 is probably one of the more important sections, yet it is one of the shortest. Understanding why the observed trends are (or aren't) reproduced by the models is important. Page numbers or continuous line numbering would have been helpful.

AR: We thank the referee for their constructive comments. In the revised manuscript we have shortened the analysis by removing references to the performance metrics of individual models except where we want to highlight the performance of a particular model, for example a model that gives a large bias. We have also expanded Section 3.6 (Trend attribution analysis, now Section 3.5) by including an analysis of the spatial distributions of the factors influencing the trends. This new analysis suggests that the influence of changing meteorology on the wet deposition trends is mostly due to changing precipitation patterns during the two periods and that the "Residual" component is also driven by changes in precipitation. This gives strength to our suggestion that changes in precipitation partially offset the decreasing trends due to emission reductions during the first period but not the second, at the measurement sites.

Specific comments:

Page 3: What is the difference between the Collette et al. (2016) work and the Torseth et al. (2012) analysis?

AR: The main difference between these two studies is the time periods they cover. Torseth et al. covers the period 1980–2009 whereas Collette et al. covers the period 1990–2012. In fact the study by Collette et al. was designed as an extension to that of Torseth et al. with updated methodologies and site selection, which were agreed on during meetings of the Task Force on Measurements and Modelling and a dedicated workshop. Collette et al. also contains additional information, such as the modelled air quality trends. In the revised manuscript we have reduced the size of this section and now only include a summary of the trends estimated from observations without listing the trends from each study.

Page 3, line 5: Consider a comma after "periods"

AR: A comma has been added

Page 3, line 6: Consider a comma before "but"

AR: A comma has been added

Page 3 - 4: There are several multi-model studies that are cited. It is impractical to provide the list of models and citations in this paper. It would be helpful to know, though, if the models used in the present study were included in those studies as well.

AR: The results of the multi-model studies have been summarised to remove the detail and highlight the variability of model performance for wet deposition estimates. Previous results of models used in this study are discussed in the Discussion section.

Page 6, line 10: If the other models were run with a lat-long grid, why wasn't CMAQ?

AR: For the CMAQ model the horizontal grid coordinate system is the same as the other models (i.e. latitude-longitude), however the CMAQ model uses Lambert conformal Conic map projection in its native state with 25 km resolution. To be comparable with the other models, the CMAQ output was interpolated to the common domain used by the other models. Also note that the common domain consists of a regular latitude-longitude grid with increments of 0.25° and 0.4° in the latitude and longitude, respectively, which is about 25 km × 25 km at European latitudes. This means that both grids are comparable.

Page 7: Did any of the models include the bidirectional flux of NH₃? This is noted in Table S2, but not discussed in the text. What is the impact on the model results of not considering this?

AR: Only one model, LOTO-EUROS, included bidirectional fluxes of NH₃. This model includes compensation points for stomata and leaf, soil and water surfaces (although the compensation point for soil surfaces is currently set to zero). Wichink Kruit et al. (2012) showed that the inclusion of compensation points in the LOTOS-EUROS model decreased annual NH₃ dry deposition, especially in ammonia source areas, leading to an increase in the atmospheric lifetime of NH₃ and an increase in WNH_x over most of the continent. However, the relative increases in WNH_x were very small over land areas and were much smaller than the inter-model differences found in our study. EMEP has a simplified approach with no dry deposition of NH₃ to growing crops, which also increases NH₃ concentrations slightly. If compensation points were included in the other models then this would be expected to increase the estimates of WNH_x slightly although it would not be enough to correct the negative biases found for some of the models. We have added these comments to section 4.2 in the revised manuscript.

Ref.: Wichink Kruit, R. J., et al. "Modeling the distribution of ammonia across Europe including bi-directional surface-atmosphere exchange." Biogeosciences 9.12 (2012): 5261-5277.

Page 7, line 7: Organic species were included in the modeled estimates of wet deposition. Are they included in the measurements? What about NO, NO₂ and N₂O₅?

AR: Organic species are not included in the measurements, although they should be considering that they are estimated to contribute to around 25% of N wet deposition in Europe (Cornell, 2011). Taking CHIMERE as an example, although the model includes organic species in the wet deposition estimates the actual contribution is zero for these simulations. It is expected that the contribution from organic species (if they include them) in the wet deposition output of the other models is also zero or negligible. By contrast, CHIMERE estimates that organic species contribute up to 13% of the grid cell dry deposition of oxidised nitrogen, which is clearly not negligible although we have not evaluated dry deposition in this study. NO and NO₂ are

relatively insoluble in water compared to other gases such as NH₃, HNO₃ and SO₂ and so are not expected to make a large contribution to the measured N wet deposition. N₂O₅, however is highly soluble but atmospheric concentrations are generally quite low and so its concentration to wet deposition is also expected to be small.

Ref.: Cornell, Sarah. "Atmospheric nitrogen deposition: revisiting the question of the invisible organic fraction." Procedia Environmental Sciences 6 (2011): 96-103.

Page 7, line 13: Doesn't the CMAQ model provide information to distinguish sea-salt sulfate?

AR: CMAQ provides sulfate in three modes (Aiken, accumulation and coarse) without distinguishing their source. In this version of the model, the coarse sulfate is from sea salt emissions. In this revised analysis we removed the coarse sulfate from the CMAQ estimates of total sulphate concentrations, as now described at the end of Section 2.1.

Page 7, line 20: consider rewording "network data of"

AR: The sentence has been rewritten as "For the evaluation of modelled atmospheric concentration estimates, the EMEP network data of mean annual concentrations of total nitrate, ammonium and sulfate (non-sea-salt component) were used." [Page 6, lines 16-18]

Page 8, line 13: Note that these criteria were developed for atmospheric concentrations and not deposition values.

AR: This is a very valid point and the following disclaimer has been added to the manuscript "It should be noted, however, that these criteria were developed for evaluating the atmospheric concentrations estimated by air quality models using specially designed model evaluation field experiments. They may not, therefore, be an appropriate tool for evaluating operational wet deposition estimates using monitoring data and can only be used as an indicator of model acceptability." [Page 7, lines 13-16]

Page 8, line 17-18: Clarify what the observed and modelled trends are for on line 17 and what trends on line 18 are more difficult to evaluate compared to annual wet deposition.

AR: Line 17 refers to the observed and modelled wet deposition trends and line 18 refers to the evaluation of the modelled wet deposition trends. However, the statistical evaluation of the wet deposition trends has been removed in the revised manuscript since it was considered confusing and did not contribute much additional information to the analyses.

Page 8, lines 20-23: suggest splitting the sentence at "then" on line 20.

AR: The sentence has been modified and split into two sentences

Page 8, line 21: "were" should be "was" as it refers to magnitude

AR: OK, the change has been made

Page 9, line 2: How were the tau values determined?

AR: This is described in the subsequent lines: “..approximated as the difference in wet deposition over the eleven year period for simulations where the other factors are kept constant, divided by ten (to obtain the mean annual trend). For example, the change in wet deposition over the period 1990–2000 due to changes in emissions can be calculated from two simulations with emissions for 1990 and 2000, both with meteorology and boundary conditions for 2000.”

Page 9, line 24: Does “European” start a new paragraph?

AR: Yes it does. The preceding paragraph is on the NO_x emission trends and the new paragraph is on the NH₃ emission trends

Page 9, lines 25-30: Specific information is given from Sutton et al. (2003) about why NH₃ emissions decreased but the same level of detail is not provided for other species.

AR: This decrease was specifically highlighted since it was the result of political change and not a result of the implementation of control measures to reduce emissions mentioned in the introduction

Page 10, line 15: Consider listing the meteorological models

AR: We agree. The model names have been included in the revised manuscript

Page 10, line 19: Consider specifying “meteorological models” rather than just models.

AR: We agree. We have made the recommended change

Page 10, line 25: It seems odd that one WRF run (used for CMAQ) would have such very different precipitation compared to the other WRF runs. What was different about the WRF runs? It might be helpful to have a table in the supplemental that provides details on the meteorological models.

AR: The main difference between the two WRF simulations is that the simulation used as the common meteorological driver used nudging whereas the WRF simulation used for the CMAQ simulations was free-running (only forced at the domain boundaries). This difference along with the different grid spacing could lead to the discrepancy in precipitation. The use or not of nudging has been added to Table S1. The specifics of the met model runs are presented in Table 4 of Colette et al. (2017a) and we considered that it was not efficient reproducing this information in the manuscript.

Page 15, section 3.5: This section seems to repeat information that was presented earlier.

AR: This section presents a statistical evaluation of the modelled trends at the measurement sites by comparing the modelled and observed trends (and their significance) at each site. This is different to the results presented in the preceding section which compares the distributions of the modelled and observed trends (i.e. not a direct comparison of the modelled and observed trends at each site) in order to show the bias of modelled trends, on average. However, section 3.5 was considered confusing and did not contribute much additional information to the analyses and has been removed in the revised manuscript.

Page 21, lines 30-33: Do these studies use different versions of the EMEP model? Please indicate what versions were used.

AR: Yes they did use different versions. This has been clarified in the manuscript

Page 22, line 13: Why is the trend for observed WNO_x for 1990-200 in Figure 16 so different than the emissions trend? Is this realistic?

AR: Yes, we believe that it is realistic. This difference is due to low significance of the observed WNO_x trends for this period. Increasing trends were observed at 15 of the 34 sites but only 3 of these were significant (see Fig. 4). Decreasing trends were observed at 19 sites but only 6 were significant. The fact that increasing trends were observed at 44% of the sites and decreasing trends at 56% of the sites leads to a median trend close to zero. If only the significant trends are used (26% of sites), the median trend is -3.3% per year, which is larger than the emission trend. However, the sites with the most significant trends are most likely to be located in the regions with the largest emission reductions and, therefore, are not representative of the model domain.

Page 24, line 18-19: what would be the effect on mass conservation of doing a bias correction?

AR: The bias correction will invalidate any assumptions of mass conservation since the correction only applies to the simulated wet deposition, leaving other components (e.g. atmospheric concentrations) unchanged. The bias correction is proposed as a post-processing step to provide more accurate estimates of future wet deposition. If mass conservation is required for these estimates, however, the bias correction should not be applied.

Figures 4 and 9: The legend text is too small.

AR: We have increased the legend text size of Figure 4 in the revised manuscript. Figure 9 is not in the revised manuscript

Figure 12: Are these period (i.e. seasonal) totals values?

AR: Yes they are. This has been clarified in the caption

Table S2: - Consider adding a table with specifics of the met model runs - Are the vertical layers for the CTM or the met model? - For Chimere, CMAQ, and MINNI, give an approximate value for the 1st model layer. - CMAQ description is incomplete and incorrect. No citation is given for the dry deposition of gases. CMAQ does include a bidirectional NH₃ model (but maybe it wasn't used). Wesely (1989) is not the correct reference for the stomatal resistance. This is calculated in the Pleim-Xu land surface model and is described in papers by Pleim and Xu.

AR: The specifics of the met model runs are presented in Table 4 of Colette et al. (2017a) and we considered that it was not efficient reproducing this information in the manuscript. However, we now refer to this table in the text so that the reader can easily access this information if they require. The vertical layers shown are for the CTM. This has been clarified in Table S1 of the revised Supplementary Material and we have also included an estimate of the depth of the first layer used in Chimere, CMAQ and MINNI. The information regarding the parameterisation of CMAQ has also been corrected.