acp-2022-82: A new assessment of global and regional budgets, fluxes and lifetimes of atmospheric reactive N and S gases and aerosols

Ge et al.

Response to Reviewer #2

We thank the reviewer for their time spent reading our manuscript. Below we include all the reviewer comments and provide in blue text our point-by-point responses. All line numbers mentioned in our responses refer to the clean revised manuscript (not the track-changed version).

General comments:

The manuscript provides a comprehensive analysis of the concentrations, wet and dry deposition, fluxes, and lifetimes of reactive N and S gases and aerosols globally, and for 10 world regions. The goal of the manuscript is to conduct a more up-to-date examination of atmospheric processes affecting the fate of reactive N and S, particularly of the reduced N species NH_3 and NH_4^+, for developing mitigation strategies according to different regional conditions. Overall, this study is an important contribution to the field as it highlights the substantial regional variation in Nr and Sr budgets and the need for modelling to simulate the chemical and meteorological linkages underpinning atmospheric responses to precursor emissions. The manuscript is well written and structured clearly and fits well the scope of this journal. I recommend that this study can be accepted for publication after the specific comments below are addressed.

Response: We thank the reviewer for their supportive comments for our work and for their recommendation of publication after attention to some minor revisions.

Major comments:

1. Section 3.1.2, lines 225-228: What was the rationale for choosing the fine SIA concentration of 0.5 μg m^{-3} as the threshold?

Response: The intention of applying such a threshold is to help readers concentrate on regions with relatively larger values of fine SIA concentration where the knowledge of chemical domain is important for formulating anthropogenic emission reduction policies to mitigate the concentrations of NH_4^+, SO_4^{2-} and fine NO_3^-. As no such threshold value is reported in other studies, we have made our own arbitrary choice based on visualisation of the global SIA distribution. We also trialled other threshold values

(e.g., 0.1 μg m^{-3}, 0.7 μg m^{-3}, 1.0 μg m^{-3}, etc.) and chose 0.5 μg m^{-3} as an appropriate option to present Fig. 3. Perhaps our choice can serve as a reference for future studies.

2. Section 3.3.1, lines 460-464: To increase study reproducibility and transparency, the authors may want to consider providing detailed calculation equations of atmospheric burdens and lifetimes (perhaps adding a section in the supplementary).

Response: We thank the reviewer for making this useful suggestion. We have added a new section in the Supplement (Sect. S1.2) that describes the detail of the calculation procedures for atmospheric burdens and lifetimes used in this manuscript. Reviewer #1 also requested more information on this point so see also our responses to this reviewer for more information.

3. Section 3.3.1, lines 517-520: In general, gaseous NH_3 deposits quicker than aerosol-phase NH_4^+. Could you provide more information on the difference between NH_3 deposition and NH_4^+ deposition in the model as deposition is crucial for lifetime calculations?

Response: Both NH_3 and NH_4^+ have dry and wet deposition. The dry deposition of gaseous NH_3 to the surface is modelled using the dry deposition velocity parameter. In the model, the deposition velocity is calculated at the centre of the lowest grid (~25 m). Different landcover types (e.g., crops, forests, grassland, seminatural, etc.) have different deposition velocities. For grids containing multiple landcover types, the grid-average deposition rate is calculated based on fractional coverage of different land-cover types within each surface grid.

The NH_4^+ dry deposition follows the basic formulation for fine-mode aerosol components in EMEP model. Its deposition velocity is dependent on landcover categories and the atmosphere stability (Obukhov length) of turbulent flows in the lower boundary layer. For example, NH_4^+ in forests with different Leaf Area Index has different deposition velocity. For stable and unstable stratification conditions, its surface deposition velocity is also different. Detailed formulation is described in Simpson et al. (2012) as cited in the main paper.

Parameterisation of the wet deposition processes in the EMEP model includes both in-cloud and sub-cloud scavenging of gases and particles. As both NH_3 and NH_4^+ are soluble species, their in-cloud scavenging, following the same equation, is correlated with their own atmospheric mixing ratio and in-cloud scavenging ratio. The parameters of precipitation rate, characteristic scavenging depth, and water density are the same for the two species. For below-cloud scavenging, gaseous NH_3 and particle-phase NH_4^+ follows two different equations. The former is controlled by its mixing ratio and sub-cloud scavenging ratio, while the latter is controlled by its mixing ratio, the raindrop fall speed, and the size-

dependent collection efficiency of aerosols by the raindrops. Again, the detailed formulation is described in the Simpson et al. (2012) citation in the paper.

4. Section 3.3.3, lines 618-619: Is there any explanation for why does South America have such large proportion of RDN in the form of NH_3?

Response: This may be related to South America's distinctive NH_3 emission characteristic. Chen et al. (2013), Whitburn et al., (2014), van Marle et al. (2017) and Evangeliou et al. (2021) all showed that natural NH_3 emissions from forest, savanna, and agriculture fires are more significant in this region compared to other world regions. NH_3 emitted from biomass burnings is mainly concentrated on remote or rural areas, and away from anthropogenic emissions of NO_x and SO_x. As a result, most NH_3 stays in the gaseous phase and quickly deposits back to ground without forming stable NH_4^+ aerosol (i.e., $(NH_4)_2SO_4$; NH_4NO_3 is easily converted back to NH_3 under high temperature conditions), which explains the large proportion of NH_3 in total RDN burden over South America. Our discussion of regional RDN deposition in Sect. 3.2.1 also shows that RDN deposition in South America is dominated by NH_3 (86%; Fig. 5), which is consistent with our explanation here.

Technical corrections:

Line 758: DOI information is missing in this citation. Please add the "doi" number before "2013b".
Response: Requested change made.

Line 809: Please add the "doi" number before "2005".
Response: Requested change made.

Line 896: Please add the "doi" number before "2015".
Response: Requested change made.

Line 966: Please add the "doi" number before "2012".
Response: Requested change made.

References

Chen, Y., Morton, D. C., Jin, Y., Gollatz, G. J., Kasibhatla, P. S., Van Der Werf, G. R., Defries, R. S., and Randerson, J. T.: Long-term trends and interannual variability of forest, savanna and agricultural fires in South America, Carbon Manag., 4, 617–638, https://doi.org/10.4155/cmt.13.61, 2013.

Evangeliou, N., Balkanski, Y., Eckhardt, S., Cozic, A., Van Damme, M., Coheur, P. F., Clarisse, L., Shephard, M. W., Cady-Pereira, K. E., and Hauglustaine, D.: 10-year satellite-constrained fluxes of ammonia improve performance of chemistry transport models, Atmos. Chem. Phys., 21, 4431-4451, 10.5194/acp-21-4431-2021, 2021.

van Marle, M. J. E., Field, R. D., van der Werf, G. R., Estrada de Wagt, I. A., Houghton, R. A., Rizzo, L. V., Artaxo, P., and Tsigaridis, K.: Fire and deforestation dynamics in Amazonia (1973–2014), Global Biogeochem. Cy., 31, 24–38, https://doi.org/10.1002/2016GB005445, 2017.

Whitburn, S., Van Damme, M., Kaiser, J. W., Van Der Werf, G. R., Turquety, S., Hurtmans, D., Clarisse, L., Clerbaux, C., and Coheur, P. F.: Ammonia emissions in tropical biomass burning regions: Comparison between satellite-derived emissions and bottom-up fire inventories, Atmos. Environ., 121, 42–54, https://doi.org/10.1016/j.atmosenv.2015.03.015, 2014.