

Atmos. Chem. Phys. Discuss., author comment AC1 https://doi.org/10.5194/acp-2021-342-AC1, 2021 © Author(s) 2021. This work is distributed under the Creative Commons Attribution 4.0 License.

Reply on RC1

Jan Eiof Jonson et al.

Author comment on "Modelling changes in secondary inorganic aerosol formation and nitrogen deposition in Europe from 2005 to 2030" by Jan Eiof Jonson et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-342-AC1, 2021

Comments to remarks from reviewer 1

General comments:

Several papers have investigated the interplay between SOx/NOx and NH3 emission changes. Nenes et al. (Atmos. Chem. Phys., 20, 3249–3258, 2020 and Atmos. Chem. Phys., 21, 6023–6033, 2021) provide a very interesting modeling framework to evaluate when particulate matter and dry deposition of inorganic reactive nitrogen are sensitive to ammonia and nitrate availability using aerosol pH and liquid water content as drivers. I think the present manuscript would benefit from a relevant discussion and comparison to these findings.

We have included a discussion of the implications of the results from the Nenes et al. papers in section 5 (Discussion and and conclusions):

As discussed in Nenes et al. (2020.021), gas/aerosol partitioning of total reduced and oxidised nitrogen are affected by aerosol pH level and water content, so that low (high) pH is favourable for NH_4^+ (NO_3^-) formation. The increase in the aerosol fraction in total reduced and oxidised nitrogen would lead to decreases in their dry deposition, and subsequently their residence times and transport distances. This effect has not been accounted for in the EMEP model, thus some limited local effects might have been missed in our model simulations. For instance, based on the Nenes et al. (2021) results, there may be additional NO₃ formation in areas with low acidity, such as coastal or dusty regions. Potentially this may reduce the deposition of total nitrate near these local sources, somewhat enhancing the accumulation of particles. Furthermore, as future emissions of SOx and NOx are expected to decrease, the pH of the particles is likely to increase, potentially favouring NO3-formation, and thus decreasing dry deposition and increasing the transport distances of oxidised and total nitrogen in some regions. On the other hand, our results show that overall, the fraction of reduced nitrogen in the total nitrogen has been increasing, and this increase is expected to continue until 2030. Assuming that the deposition rates for total nitrogen are mostly driven by those of reduced nitrogen (following Nenes et al. 2021)), the local effects of NO₃⁻ formation bursts would probably not play a major role across the regions in different present and future chemical regimes. Therefore we believe that overall, the main conclusions presented in

our paper remain valid.

Minor corrections are listed below:

Line 51: sulphate

sulphate replaced by SO₄²⁻

Line 73: differs

Corrected

Line 100-102: could you provide an equation for this?

The calculation is based on an extensive set of input data and equations. A detailed description of the calculation of critical loads is described in Chapter 5 in the mapping manual, see new text.

New text:

The CL exceedances presented here were calculated using the current CL database, which is described in Hettelingh et al. (2017) and stored by the current Coordination Centre for Effects (CCE) at the German Federal Environmental Agency. The calculation is based on an extensive set of input data and equations. A detailed description is included in the Mapping Manual of the ICP Modelling and Mapping.(CLRTAP (2017), Chapter 5). This dataset is also used, among other things, to support European assessments and negotiations on emission reductions (Hettelinghet al, 2001, Reis et al. 2012, EEA, 2014).

Line 134: as ammonium is either...

Changed as suggested.

Line 145: is deposited than is emitted

Changed as suggested.

Line 212: is small

Changed as suggested.

Figure 1a: emission units should be Gg.y⁻¹

Changed as suggested.

Figure 3 caption units should be mg(N).m⁻².y⁻¹

Changed as suggested.

Table 2 – please clarify what meteorological data are used for each simulation year.

We have added:

All model runs have been performed with 2017 meteorological conditions as described in Section 3.

In figure 5 the CL exceedances are given in eq ha⁻¹ y^{-1} while in Figure 3 for the deposition fluxes the surface unit is m^2 – could you make them uniform?

Deposition fluxes and CL are addressing different user groups. The practice in the air pollution community has been to use $mg(N)m^{-2}$ y $^{-1}$, whereas the effects community prefers eq ha $^{-1}$ y $^{-1}$. This practice is also followed in the annual EMEP reporting (https://emep.int/mscw/mscw_publications.html) to the Convention on "Long-range Transboundary Air Pollution"