Review of 'Role of ammonia ...' by Sebnem Aksoyoglu et al.

This paper is a nice overview of the changing role of ammonia in the European atmosphere. I think it is generally well written, and my only major criticism is that a number of important uncertainties which might affect the conclusions are not discussed or addressed.

Major comments

The major areas of uncertainty that I missed include:

1. Bi-directionality. It is well established that ammonia can be emitted as well as deposited as a result of the equilibria between atmospheric and surface concentrations. It displays a so-called compensation point (Nemitz et al., 2001; Flechard et al., 1999, 2013), and this can affect the deposition close to source areas and long-range transport in general (Bash et al., 2013; Wichink Kruit et al., 2012).

2. This study seems to ignore the impacts of co-deposition, in which the acidity of the surface (affected by both SO_2 and NH_3 emissions, and their trends) changes. The impacts of this process on trends have been explored in for example Wichink Kruit et al. (2017).

3. A similar issue with trends, also not mentioned, is changing pH of rainwater (Banzhaf et al., 2012).

4. Meteorological variability. The current study mainly uses meteorology from just two years, 1990 and 2010, but Wichink Kruit et al. (2017) showed that meteorology can also account for a significant contribution to NH_3 trends.

5. Ship-plumes. It is well known that models tend to mis-represent HNO_3 production from NO emitted from ships into clean marine environments (von Glasow et al., 2003; Vinken et al., 2011, 2014). This could potentially have been handled with the CAMx model's plume-in-grid approach, but this doesn't seem to have been used. However, some of the comments made about HNO_3 (e.g. L187 onwards) may be impacted by this issue.

6. In the introduction, I missed some discussion of trend studies on land-based emissions and concentration/deposition trends which have already been done, e.g. Fowler et al. (2007); Fagerli and Aas (2008) or Wichink Kruit et al. (2017). How does the current study add to these? (Page 3 gives a lot of information given on the impacts of shipping, but not much about land.)

Smaller comments

1. L36. The Maas & Grennfelt reference is not peer-reviewed. There are plenty of peer-reviewed publications on this subject.

2. L37. The authors only mention ammonium sulfate here, but bi-sulfate is an important component of European aerosol too.

3. L42. The Dentener ref is 14 years old now; find something more

4. L53. Are you sure it is ammonium sulfate, and not bisulfate?
5. L54. The Colette et al 2016 reference seems to be some grey literature, with no address and no url. What is this? And surely there are some peer-reviewed papers that be cited to support this statement?
6. L65-17. The SECA's came into effect at the start of 2015
7. L80-84. It is unclear where the cited 1-14% PM2.5 applies. This number sounds very different to those cited for Karl et al., and so this paragraph is a little confusing. Are the Karl et al results similar to, or very different from those cited from Viana et al.?
8. L100-102 How is the coarse-mode aerosol (e.g. for nitrate) handled in this model system?
9. L103. Be explicit with a reference to the Zhang scheme (not just the cited CAMx user's guide). And whether co-deposition is included or not?
10. L134 on. Brief details on the measurement networks underlying EDT work should be given.
11. L140 I won't repeat Ref #1's comments, but agree with them.
12. L145. As noted above, many processes not discussed in this manuscript might also contribute to model-measurement bias. Another issues is scale, which is very briefly mentioned on L156, but which can be a very important factor for NH_3 (Theobald et al., 2016; Denby et al., 2020).
13. Notation. Better to use pNH4, pNO3, pSO4 than PNH4 etc, to avoid mixing chemical and atmospheric nomenclature.
14. L242 states that the amount of precipitation is crucial, but no figures are given on this here; please expand.
15. Many of the figure legends and colors need to be re-done. For example, in Fig2c reds are used for positive values and blues for negative, which is great, but in Figs. 2d and 2e the color-scale shows white for levels both above and below certain thresholds! Later Figures also show such strange behaviors. I suggest using the same color-scale for all subplots, and do not have the same color for different values.
16. Fig. S2 - which measurements? Be explicit.

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