

Interactive comment on “Spatial, temporal and vertical distribution of ammonia concentrations over Europe – comparing a static and dynamic approach with WRF-Chem” by M. Werner et al.

Anonymous Referee #1

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This paper deals with an important topic, the spatial, temporal and vertical distribution of NH_3 in Europe, and the impacts of dynamic approaches. Although I agree with the comment by Dr. A. Dore that this subject is important, I think this paper does little to advance our knowledge of the subject. The authors claim that the aim is to improve the basic understanding of ammonia in the atmosphere, but essentially all they show is that WRF-Chem performs poorly for hourly NH_3 when used with static emissions, and still performs poorly when used with dynamic emissions. No real attempt to solve the problem with this model is presented, and no attempt is made to show if the problem is general for other models and locations.

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Detailed Comments

1. The use of dynamic emissions compared to static emissions has already been shown by Skjoth et al (2004, 2011) and Werner, et al. 2015. The main new thing in this paper is that the dynamic emissions still result in poor reproduction of the diurnal cycle. This would have been worth exploring, but essentially no exploration is done. The authors did no sensitivity tests of their own, and seem unaware of the much more detailed work done on this subject by other workers.
2. Hourly measurements are presented for just one station (Harwell, UK), and only NH_3 concentrations are presented. As discussed below, hourly data are available for other stations in Europe, and for at least some of the other key compounds which one would normally look at when trying to further understanding of NH_3 in the atmosphere (e.g. HNO_3 , sulfate).
3. Indeed, the authors seem unaware of many of the studies done in Europe to highlight problems in the understanding of NH_3 diurnal cycles, or that other models do not show such poor performance for hourly data. Work with the LOTOS-EUROS model in particular has extensively looked into model comparisons against hourly data in Europe, and these studies did a much better job of analyzing the reasons for any discrepancies and of testing alternative model. Some examples:
 - Aan de Brugh, JMJ et al. Modelling the partitioning of ammonium nitrate in the convective boundary layer Atmos. Chem. Physics, 2012, 12, 3005-3023 - investigate hourly data and partitioning of $\text{NH}_3\text{-NH}_4^+$ at Cabauw, including a number of model tests with ISORROPIA to explain the observed diurnal variations. This paper has a much more thorough analysis of both the diurnal cycle and vertical profiles (and sensitivity analysis) than the submitted manuscript.

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- Schaap, M. et al., Illustrating the benefit of using hourly monitoring data on secondary inorganic aerosol and its precursors for model evaluation. *Atmos. Chem. Phys.*, 11, 11041-11053, doi:10.5194/acp-11-11041-2011, 2011
- one of the first papers to show comparisons of modeled versus observed diurnal cycles of the key components, and with an extensive discussion of equilibrium issues. The model used showed better results than those of WRF-Chem in the submitted manuscript.
- Wichink Kruit, R. et al., Improving the understanding of the secondary inorganic aerosol distribution over the Netherlands, TNO report TNO-060-UT-2012-00334, 2012 (available online),
- again, more examples of more successful evaluation and testing of models against hourly data at Cabauw.
The Cabauw site has seen a large number of measurements over the years, including vertical profiles, that would be very relevant to this investigation. For example,
- Kulmala, M. et al., General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales, *Atmos. Chem. Phys.*, 11, 13061-13143, 2011,
- shows vertical profiles of sulfate, nitrate and ammonium at Cabauw
- Aas, W et al., Lessons learnt from the first EMEP intensive measurement periods, *Atmos. Chem. Physics*, 2012, 2, 8073.8094,
- show hourly NH_3 , HNO_3 , NH_4^+ and NO_3^- for the EMEP sites Harwell, Ispra, and Cabauw, and data for Auchencorth Moss are said to be available.
This paper also showed that different sites had rather different diurnal cycles (e.g. some had peak NH_3 in daytime, others at night), which implies that a model cannot be evaluated by comparison with one site alone. This

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Aas paper spends more time discussing the reasons and caveats of diurnal cycles than the submitted manuscript.

In fact, both the LOTOS-EUROS and EMEP models seem to capture the diurnal pattern of NH_3 quite well in many cases, although sometimes with significant bias. Why do these models perform better than WRF-Chem with either static or dynamic NH_3 emissions? If other pollutants and sites had been considered we might have learned how well or badly WRF-Chem performs in general before trying to draw conclusions for just one pollutant.

4. The paper actually claims that hourly NH_3 measurements are only available at one site in Europe, Harwell. This is clearly not true, as the above studies testify.
5. The paper stresses some points which are obvious and well known from even decades old studies, in particular that the concentrations of a pollutant released at ground level are inversely correlated with PBL height. This is basic air-pollution meteorology.
6. The authors try to make the point that dissociation of NH_4NO_3 is not a strong source of NH_3 . This would have been interesting to quantify, but instead the authors simply cite that fact that there is a phase-shift between the NH_3 and NH_4^+ concentrations
7. The paper is also careless in many places, for example the lack of proper labeling on Figures and use of citations that aren't appropriate, e.g. the Sutton et al. paper given as a reference for ECLAIRE doesn't mention ECLAIRE.
8. The authors claim that they have analyzed the vertical distribution of NH_x , but they haven't. They have simply illustrated this, without comparison to measurements or even earlier studies that have done this before in a more thorough way.

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9. The authors concentrate on WRF's bias with respect to temperature, but what about wind-speed, or even friction velocity? The paper does cite other studies (e.g. Jimenez and Dudhia, 2013) but since these were done by other groups in different areas and likely with other WRF settings, those studies are not necessarily relevant for the European area or Harwell.
10. The authors compare a BASE case with a DYNAMIC case, but nowhere do we see the annual time-series compared with each other.
11. Page 22937, Line 15. Are not hourly measurement of ammonia rare because of their expense and complexity?
12. Page 22938, Line 21. It is claimed that the work of Werner et al., 2015 shows significant improvements when dynamic approaches are used, which is a slight exaggeration. Werner only examined few locations, and found worse results for some statistics in some seasons.
13. Page 22939, Line 5. What is FP7?
14. Page 22940, last paragraph. This is very hand-waving. The model performs 'well', there were 'biases', the 'biases are significant', and I have no idea what any of that means. Quantify.
15. Page 22941, Line 6. Are the Schaap 2005 profiles commonly used? Which models use them?

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