Review of: Global and regional impacts of HONO on the chemical composition of clouds and Aerosols, Y. F. Elshorbany, P. J. Crutzen, B. Steil, A. Pozzer, H. Tost, and J. Lelieveld
acpd-13-23599

Summary

This paper examines the role that HONO plays in the affecting the global distribution and composition of aerosol. HONO is often neglected or poorly simulated in chemistry and aerosol models, but here the authors show that realistic HONO concentrations affect simulated concentrations gas phase oxidants, which impacts the production of sulphate, nitrate and ammonium aerosol. In particular realistic HONO concentrations can increase wintertime sulphate concentrations, which are typically low biased in chemical transport models. The parameterisation used is empirically based and very simple thus it would be easy to incorporate into all chemistry and aerosol models.

The paper is interesting, scientifically sound, well written and the content is within the scope of ACP. I would recommend publication after the following revisions.

Main Comments

1) The parameterisation of HONO concentration is extremely simple; a constant NOx:HONO ratio is assumed. Thus rather that treating the complex HONO chemistry, the HONO concentration is assumed to be [NOx]*0.02. This parameterisation has been published previously in ACP (Elshorbany et al, 2012). I would like a brief description of the parameterisation to appear in the abstract of this paper and a more detailed description under a clear subheading in the Model Description section, even if this requires a little repetition from the Elshorbany et al, 2012 paper.

2) The sensitivity study simulations are not named in a way that describes their setup. Please either re-name more clearly or include a table that summarised the setup used in each simulation. Understanding these simulations is key to understanding the paper and a little more clear explanation would help.

3) The hope for the parameterisation is that it would improve wintertime sulphate concentrations but the comparison to observations shows that generally with the additional parameterisation the model performs more poorly wrt winter sulphate in Europe and Asia, although there is some increase in performance in North America. This process is therefore not the main source of model / observation bias in these regions and this should be outlined clearly. Also, the Zhang et al (2007) AMS data used by Pringle et al (2011) is freely available as a supplement to the Zhang paper so it would be worth repeating the analysis rather than expressing what you would "expect" might happen. Does the model skill at simulating the seasonal cycle increase? I would expect that the simulation of the seasonality to improve even if the absolute model bias is not reduced by this new parameterisation. Please discuss.

4) I am not fully convinced by the argument about particle growth rate (page 23613 and Conclusion). One would expect an increase in H2SO4 to result is faster aging of the particles from hydrophobic to hydrophilic modes, as is shown in the paper. But if the H2SO4 grew a

fraction of the particles in the Aitken hydrophilic mode so they were transferred to the accumulation hydrophilic mode other factors would also change; their ability to form cloud droplets, the rate of sedimentation and deposition. In other words the relative contribution of the different modes is controlled by a range of microphysical factors. I think it is difficult to draw the conclusions that you have drawn on this point from the data you have available. Please consider revising this paragraph. Also I think the paper does not benefit from this discussion as it distracts from the main point of the paper.

Minor Comments

1) Figures 1 and 2, there are so many gradients it's hard to read the values from the colour bar (I can't tell the difference between all the greens, or all the reds).
2) Figure 5; colour scale is hard to read. Caption does not fully describe the plots. Which simulations are being compared?
3) Figure 9; Percentage change is potentially misleading as it is being dominated by the large change in Asia, but I think this is because the baseline is low in these locations. Why else would there be these two large "spots" of change? Consider re-scaling or explaining in text.

Refs

Elshorbany, Y. F., Steil, B., Brühl, C., and Lelieveld, J.: Impact of HONO on global atmospheric chemistry calculated with an empirical parameterization in the EMAC model, Atmos. Chem. Phys., 12, 9977–10000, doi:10.5194/acp-12-9977-2012, 2012.

Description and evaluation of GMXe: a new aerosol submodel for global simulations (v1) K. J. Pringle, H. Tost, S. Message, B. Steil, D. Giannadaki, A. Nenes, C. Fountoukis, P. Stier, E. Vignati, and J. Lelieveld, Geosci. Model Dev., 3, 391-412, 2010

Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Allan, J. D., Coe, H., Ulbrich, I., Alfarra, M. R., Takami, A., Middlebrook, A. M., Sun, Y. L., Dzepina, K., Dunlea, E., Docherty, K., De-Carlo, P. F., Salcedo, D., Onasch, T., Jayne, J. T., Miyoshi, T., Shimono, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R. J., Rautiainen, J., Sun, J. Y., Zhang, Y. M., and Worsnop, D. R.: Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes, Geophys. Res. Lett., 34, L13801+, doi:10.1029/2007GL029979, 2007.