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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Size-resolved aerosol pH over Europe during summer" *by* Maria Zakoura et al.

Anonymous Referee #2

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In this work, Zakoura et al. used a chemical transport model to investigate aerosol acidity over Europe and its variations with particle size, altitude, and time. In addition, they emphasize on the central role of non-volatile mineral cations, notably calcium, on pH calculations and nitrate concentrations. This study is of definite interest to the ACP audience by contributing to one of the least understood atmospheric aerosol properties, the aerosol acidity. The manuscript is very well written, the methodology is scientifically sound, and the presentation is clear. However, I have one major comment regarding the omission of mineral dust emissions from the Sahara Desert. Overall, I recommend this study for publication. Below are a few comments to be considered prior to publication.

Major comment:

1. The authors mention that they have not included the dust emissions from the Sahara Desert. However, it is well known that dust particles from the Sahara can travel towards Europe and influence the air quality over Southern Europe but in some cases as far as the Central Europe. Given the importance of NVCs to aerosol pH calculations and aerosol nitrate formation, as vividly presented in this work, the authors should include the Saharan mineral dust and NVC emissions in their analysis. Mineral dust emissions can be calculated online from the WRF model that has been used here for meteorological inputs. Alternatively, there are available reliable emission inventories to be used offline such as the AEROCOM emission inventory (Dentener, 2006).

Specific comments:

1. Page 2, lines 52-61: A reference to the AeroCom phase III study for aerosol nitrate can also fit in the discussion here (Bian et al., 2017).

2. Page 2 line 64: The value of 9 for aerosol pH seems unrealistically high and certainly not in line with the results of your study.

3. Page 4 line 112: Please change "was" with "is"

4. Page 4 Eq. 1: The [W] needs to be inside the log.

5. Page 4 Eq. 1: Is this the water from ISORROPIA II only or do you also consider the water associated with the organics? If so, please discuss briefly how you calculate the aerosol water associated with the organics and state their hygroscopicity if needed.

6. Page 4 Section 2: Based on the model description, I assume that you don't take into account the impact of organic acids on aerosol pH. Can you briefly discuss the implications of such a simplification?

7. Page 5 line 144: Can you add a reference for these fractions?

8. Page 5 line 154: Do you assume stable or metastable aerosols for the present study?

9. Page 5 line 152-156: Can you explain more here? How the insoluble CaSO4 is removed from the particles? Do you have soluble and insoluble size sections in your model and you calculate the aerosol pH only for the soluble sections or do you have only one well mixed particle for each size section?

10. Page 6 Section 4: I found the map projection used in Fig. 1 and the rest of the manuscript quite confusing, making hard to follow the results. Can you use a different map projection (e.g., Mercator)?

11. Page 6 1st paragraph: Can you state the domain average (or the continental average) aerosol pH for each of the particle sizes?

12. Page 6 1st paragraph: Can you comment on why the tropical Atlantic Ocean in figure 1a looks very acidic with pH values lower than 2? Also, Northern Scotland looks more acidic than the rest of the Great Britain.

13. Page 6 line 167: Why lower NH3 results in higher pH? Do you mean lower sulphate?

14. Page 6 lines 181-184: Very interesting. You can also specifically comment on the Mediterranean Sea where the change of pH between sizes is large. Furthermore, over the Mediterranean, submicron aerosol nitrate is very low and super-micron nitrate very high, corroborating your hypothesis.

15. Page 7 line 225: Do you mean all of them (and not both)?

16. Page 8 lines 231-234: This is very interesting. Can you expand the discussion here? Do these diurnal profiles of aerosol pH correlate with any of the diurnal profiles of the variables stated here (e.g., RH, T, PBL)?

17. Page 8, lines 244-245: This is also very useful result. Can the authors comment if this acidification of aerosols can affect their CCN activity and/or the pH of the formed cloud droplets?

18. Page 8 Section 4.4: Why there is a nitrate-free zone between North and South Europe in Figure S4? I would expect that NOx and NH3 emissions are everywhere in central Europe. Furthermore, it looks like you have more nitrates over the oceans than over land.

19. Page 8 Line 260: Do you mean the mass transfer to the aerosol?

20. Page 10 lines 296-304: Why the effect over oceans is so large in Fig. 7? What is the composition of sea salt emissions? Have you changed their composition here as well?

21. Page 10 lines 296-304: The impact of NVCs on aerosol pH and nitrate is quite impressive, given that you only use urban dust emissions. This emphasizes the need to include Saharan emissions as well.

22. Page 10 line 303: Do you mean by up to 0.5 units?

23. Page 10 line 308: This is not the case here. Over the northern coast of continental Europe and Southern England, the impact of NVCs on nitrate concentrations is significant despite the fact that the impact on pH is negligible. Why submicron aerosol nitrate has such a strong increase (almost twofold) in the presence of NVCs?

24. Page 10 Section 4.5.1: Similar to NVCs, The impact of calcium on the pH all over the oceans is very strong. Does your sea salt contain any Ca? Can you comment why pH increases almost uniformly even over the remote oceanic locations of your domain?

References:

Bian, H. S., Chin, M., Hauglustaine, D. A., Schulz, M., Myhre, G., Bauer, S. E., Lund, M. T., Karydis, V. A., Kucsera, T. L., Pan, X. H., Pozzer, A., Skeie, R. B., Steenrod, S. D., Sudo, K., Tsigaridis, K., Tsimpidi, A. P., and Tsyro, S. G.: Investigation of global particulate nitrate from the AeroCom phase III experiment, Atmospheric Chemistry and Physics, 17, 12911-12940, 10.5194/acp-17-12911-2017, 2017.

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J., Generoso, S., Ginoux, P., Gong, S., Hoelzemann, J. J., Ito, A., Marelli, L., Penner, J. E., Putaud, J. P., Textor, C., Schulz, M., van der Werf, G. R., and Wilson, J.: Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed data-sets for AeroCom, Atmos. Chem. Phys., 6, 4321-4344, 2006.

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