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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Acid gases and aerosol measurements in the UK (1999–2015): regional distributions and trends" *by* Y. Sim Tang et al.

Anonymous Referee #2

Received and published: 20 August 2018

Review of Tang et al This is a nice paper that presents the analysis of UK monitoring sites and documents changes in the atmospheric composition over the past fifteen years. This results in a comprehensive view of how the atmosphere has changed during a period of large emission reductions. The authors then discuss the changes in emissions with the observed changes in atmospheric composition. Overall, the manuscript is well written but could benefit from some additional editing in areas outlined below.

General comments

1. Currently the manuscript is primarily focused on documenting trends and events and with a smaller focus on the changes in atmospheric composition and pollutant fate due to changes in emissions. This manuscript would benefit from a bit more focus. I

suggest focusing more on the trends and how they relate to emission changes and less on specific events captured in the data.

2. The discussion of the trends of NH3 and HNO3 are sometimes a bit difficult to follow as the change in aerosol composition and loading over the time frame of the measurements impacts the gas phase concentrations. Consider discussing these trends as total nitrate (gaseous HNO3 + aerosol NO3) and NHx (gaseous NH3 + aerosol NH4).

3. There are lots of small sections in this manuscript, some consisting of single sentences. Consider combining them into more general sections. Specifically, 2.3.1-2 and 2.5-6.

4. Many sentences leading paragraphs are structured as "For {atmospheric constituent}, . . .". This is a bit formulaic and the authors may want to revise these sentences.

Specific comments

1. Abstract: I find the final two sentences of the abstract to be the most compelling. There is a lot of detail, primarily on page 1, that would be better suited for the results section. Consider summarizing the text on the spatial and temporal trends and better connecting them to the changes in atmospheric HNO3 and NH3.

2. Abstract Page 2 lines 5-6: ". . . indications that the atmospheric lifetime of HNO3 and NH3 has increased . . .". This does not seem correct to me. The lifetime of these gases has not increased but rather the phase/composition of these species have. There are now more gaseous and less aerosol bound NO3 and NH3 due to changes in SO2. This likely decreases the atmospheric lifetime of total nitrate and reduced nitrogen compounds as NH3 and HNO3 typically dry deposit faster than aerosol NO3 and NH4.

3. Page 4 lines 17-29: This paragraph contains similar information as the previous paragraph. Consider combining it with the previous paragraph

4. Page 6 lines 21-22: This is an awkward introductory sentence for this paragraph.

Consider revising or adding an introductory paragraph that introduces the importance of the denuder base coating.

5. Sections 2.5: Are data that failed the quality checks removed from the analysis?

6. Section 2.5 iv) What is the criteria to determine anomalies and outlies?

7. Section 2.6 Line 22: Does the empirical factor used for HNO3 bias correction exhibit any dependence on season, temperature or solar radiation? If the bias is due to oxidants, then I would expect a dependence in the bias on seasonal and environmental parameters.

8. Page 9 lines 28-29: The mean difference between the measurements are given here but what is the scatter between the measurements and the median difference. A correlation coefficient would provide some information about the scatter and a median difference would indicate how normal the distribution is and if the bias is being driven by high values in one of the measurement techniques.

9. Page 9 line 32: Difference in the instrumentation flow rates and/or inlets could result in the instruments measuring different sized aerosols and my influence the differences in SO4.

10. Page 16 lines 1-2: The peaks in NHx and SO4 in the spring may just be coincidental. The spring time could also be a time in which the aqueous formation pathway of SO4 is at its maximum or the SO2 emissions from heating or transportation may be larger. In the US, the SO4 concentrations typically peak in the summer while the NH3 concentrations peak in the spring.

11. Page 16 Line 5: "Na+ and Cl-" have highest concentrations during winter . . .' Is salt used for the treatment of road surfaces in the Winter in the UK?

12. Page 20 line 18: Significant has a specific statistical meaning. I think "larger" would be a more appropriate.

13. Page 21 line 5: ". . . SO2 towards it being dominated by NH3, . . ." This appears to be a bit binary. There are lots of constituents in the air, many of which were note measured here. More context is needed.

14. Page 22 line 12: "expected to increase residence times of NH3 and HNO3 in the atmosphere" If we are in an NH3 limited environment, I can see how this would increase HN4NO3 and how that could increase the atmospheric lifetime of HNO3 as it is partitioned to NO3 aerosols. However, I do not see how this increases the NH3 lifetime. NH3 will preferentially partition with SO4, which is more thermodynamically stable than NH4NO3, this should decrease the lifetime of NH3 if anything as the NH4NO3 will evaporate where the (NH4)2SO4 aerosol would not.

15. Figure 8: I am happy to see a measure of scatter on these plots as the SD. However, a 5% and 95% CI would be more informative as it would give the reader an idea about the distribution of the data.

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