

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

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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 NH_x (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.

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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 outliers?
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 may influence the differences in SO₄.
10. Page 16 lines 1-2: The peaks in NH_x 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.

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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 not 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 NH₄NO₃ 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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