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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Drivers for spatial, temporal and long-term trends in atmospheric ammonia and ammonium in the UK" *by* Yuk S. Tang et al.

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RESPONSE TO REVIEWER

We have carefully considered Referee #2's comments.

Reviewer Comment: "The trend analysis in this study is superficial and does not meet a criteria for publishing in a high-impacted journals such as ACP."

Author response: The objective of the statistical trend analysis presented in our research paper was to identify trends in the long-term datasets (univariate monotonic, see e.g. Hirsch et al., 1991), estimate the rate of change and to address

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the question of whether trends in NH3 and NH4+ concentrations (if any) are consistent with the changes in estimated UK annual NH3 emissions (data downloaded from: http://naei.beis.gov.uk/data/data-selector-results?q=101505)?" The dataset is sufficiently long-term (i.e. gaseous NH3: 17 years and particulate NH4+: 16 years) and collected by consistent methods, to allow for effective statistical trend analyses to be carried out.

To identify and quantify monotonic trends in the paper, trend assessment was carried out using (i) linear regression (LR), (ii) Mann-Kendall (MK) test (Hirsch et al., 1981; Gilbert, 1987) on annually averaged and monthly mean data, and (iii) Seasonal Mann-Kendall (SMK) test (Hirsch et al., 1982) on monthly data only. We think that this is not a superficial trend analysis - rather we applied the relevant methodologies. We referred to overviews of some of the more widely used techniques in time series modelling and analysis are widely available (see e.g., Chatfield, 2016; Hamilton, 1994; Meals et al., 2011). Online resources (e.g. https://cran.r-project.org/web/views/TimeSeries.html) also provide information on the range of statistical tests to identify and quantify trends in environmental data. It is noted that the non-parametric Mann-Kendall (MK) statistical approach is also commonly employed to detect monotonic trends in series of environmental data in many papers and scientific reports (e.g. Colette et al., 2012., Gurreiro et al., 2014, Li et al., 2016, Meals et al., 2011; Serrano et al., 1999; Torseth et al., 2012., Yao et al., 2016) and hydrological data (e.g. Hirsch et al., 1981, 1982). Trend analysis using the Mann-Kendall approach are also described in publications by ACP (e.g., Gurreiro et al., 2014, Li et al., 2016, Torseth et al., 2012., Yao et al., 2016). The advantages of the MK approach over linear regression for trend assessments are in that (i) it does not require normally distributed data, (ii) it is not affected by outliers, and (iii) it removes the effect of temporal auto-correlation in the data. The Seasonal Kendall test deployed also is highly robust and relatively powerful, recommended for water quality trend monitoring (Meals et al., 2011) and most recently applied in air pollution trend assessments in Europe (Colette et al., 2016; Torseth et al., 2012).

The causes of observed trends were subsequently interpreted in terms of three main drivers: 1) Meteorological: influence of temperature/rainfall 2) Changes in emissions from 3 dominant source sectors (cattle, pigs & poultry, sheep) 3) Changes in chemical climate, e.g. effects of large decrease in SO2 emissions and concentrations on co-deposition relationship of NH3 with SO2, and shift in form of particulate NH4+ from (NH4)2SO4 to NH4NO3.

It is noted that the MK and linear trend approach have been used in EMEP and in UK Air quality monitoring network reports respectively, therefore it was important to look at both and the differences.

Reviewer comment: "The authors are strongly encouraged to conduct a literature review for which trend analysis tools are the most suitable for this work."

Author Response: A literature review for trend analysis tools as suggested by the reviewer is considered outwith the scope of this research paper. We have added a sentences discussing the previous use of trend analysis methods by TFMM/EMEP and in UK Air quality monitoring network reports - primary users of these datasets. As noted in the text of the manuscript both analysis methods lead to similar results.

Reviewer Comment: "Incorrect adopting trend analysis tools also leads that several discussion such as "Trends in NH3 concentrations vs trends in NH3 emissions", "Influence of climate" and "Influence of local emission sources" is full of augments and lack of solid scientific values. The reviewer believes a substantial revision to be required to make the current version publishable"

Author response: Given our opinion that we have used appropriate methods, and a lack of detailed critique by Reviewer #2, we are unable to directly respond to this comment. However we do not think a substantial revision of our manuscript is required.

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