

Supplement of Atmos. Chem. Phys., 18, 705–733, 2018  
<https://doi.org/10.5194/acp-18-705-2018-supplement>  
© Author(s) 2018. This work is distributed under  
the Creative Commons Attribution 3.0 License.



*Supplement of*

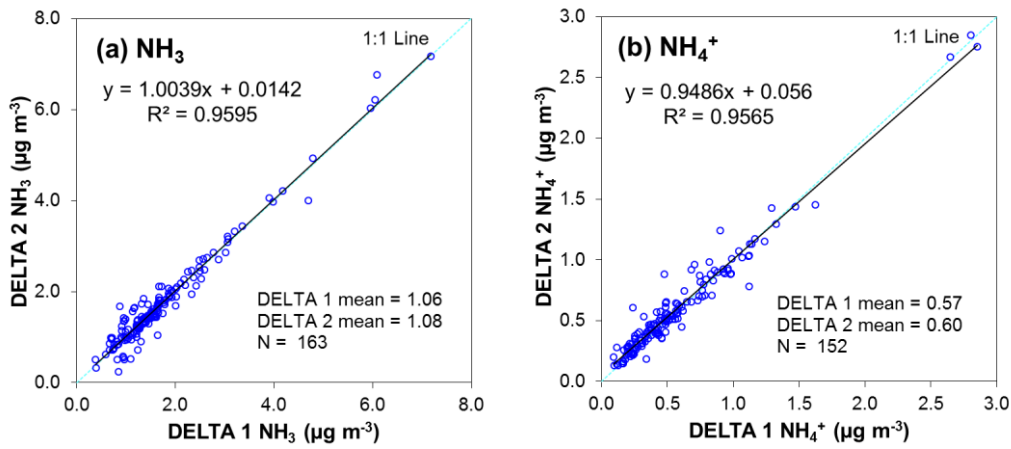
## **Drivers for spatial, temporal and long-term trends in atmospheric ammonia and ammonium in the UK**

**Yuk S. Tang et al.**

*Correspondence to:* Yuk S. Tang (yst@ceh.ac.uk)

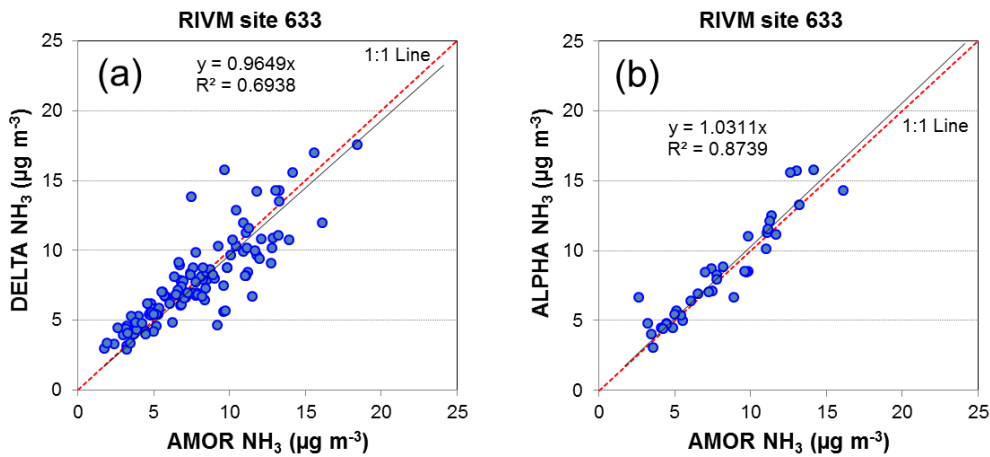
The copyright of individual parts of the supplement might differ from the CC BY 3.0 License.

# Supplementary material

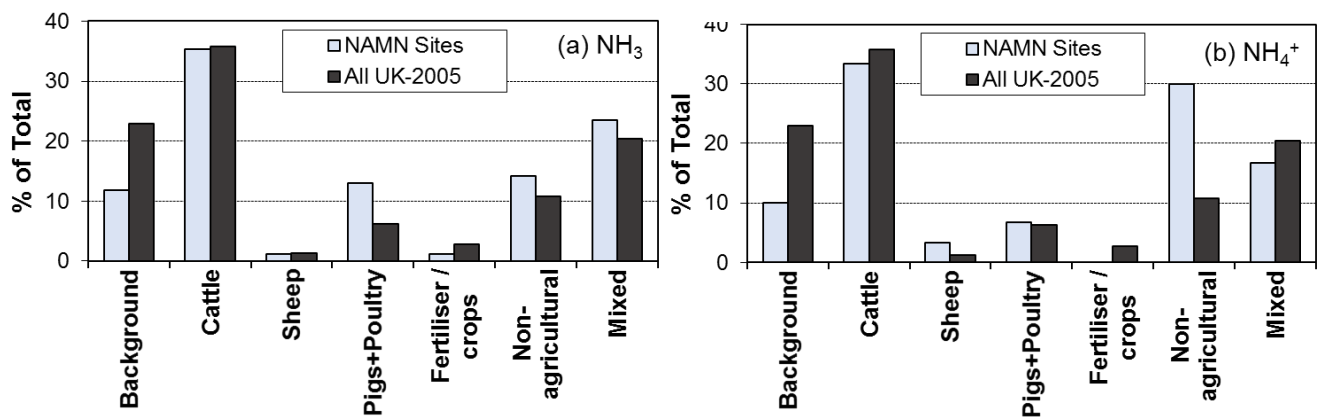


- 5 Supp. Figure S1: Comparisons of parallel measurement of monthly (a)  $\text{NH}_3$  and (b) particulate  $\text{NH}_4^+$  concentrations from duplicate DELTA sampling at the UK National Ammonia Monitoring Network (NAMN) site Bush OTC (UKA00128) for the period 1999 to 2014.

10

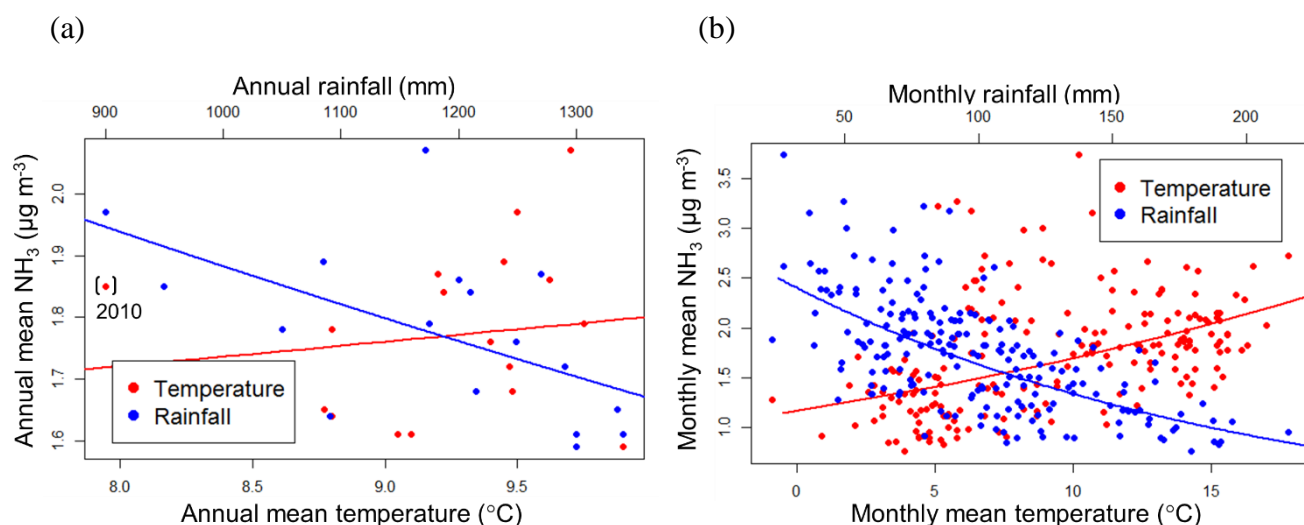


- 15 Supp. Figure S2: Comparison of the monthly  $\text{NH}_3$  concentrations determined by: a) DELTA (mean of replicate results), and b) ALPHA methods with the AMOR results derived from the average of hourly AMOR data at the Zegfeld Dutch National Air Quality Monitoring Network site (ID 633) for the corresponding DELTA and ALPHA sampling periods (unpublished data).

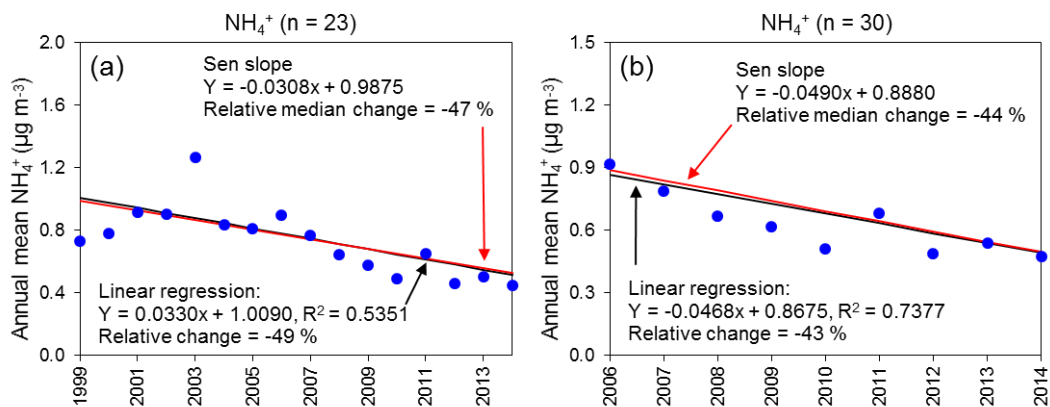


Supp. Figure S3: Frequency distribution of sites in the UK National Ammonia Monitoring Network NAMN measuring (a)  $\text{NH}_3$  (85 sites) and (b)  $\text{NH}_4^+$  (30 sites), according to each of seven dominant  $\text{NH}_3$  source sectors Error! Reference source not found. (based on the network structure for 2005) compared with the dominant source classification for the whole land area of the UK.

5

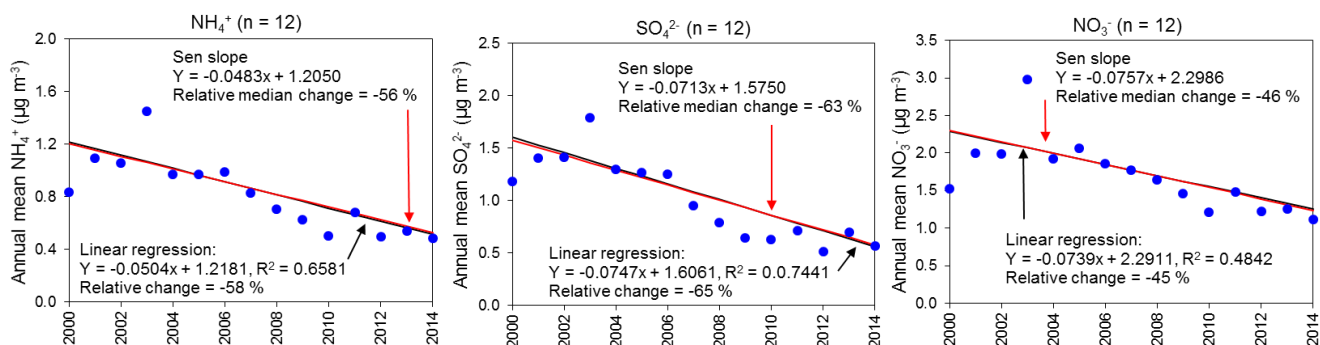


- 10 Supp. Figure S4: (a) Relationships between UK mean annual measured  $\text{NH}_3$  concentrations from 59 sites in the National Ammonia Monitoring Network (NAMN) and mean annual temperature and rainfall (data downloaded from <http://www.metoffice.gov.uk/>) for the period 1998 to 2014.  $\text{NH}_3$  was negatively correlated with rainfall (blue line:  $\text{Log}(\text{NH}_3) = -0.0003 \cdot \text{Log}(\text{rain}) + 0.9656$ ,  $R^2 = 0.32$ ,  $n = 17$ ,  $p = 0.02$ ). For the relationship between  $\text{NH}_3$  and temperature, although most of the data shows an increase in  $\text{NH}_3$  with temperature, the correlation was not significant (red line:  $\text{Log}(\text{NH}_3) = 0.0227 \cdot \text{Log}(\text{temp}) + 0.3618$ ,  $R^2 = 0.02$ ,  $n = 17$ ,  $p = 0.59$ ).
- 15 2010 (data point marked on graph) was an unusual year with considerably lower annual mean temperature (7.9 °C) than normal (mean = 9.2 °C for period 1998 – 2014). This was due to exceptionally cold winter temperatures occurring in Jan, Feb, Nov and Dec 2010, with Dec 2010 being the coldest for over 100 years. While the mean temperature for 2010 was lower than usual, the mean annual  $\text{NH}_3$  concentration for 2010 was in fact similar to other years, since the lowest  $\text{NH}_3$  concentrations occurred in the winter months. (b) Relationships between UK mean monthly measured  $\text{NH}_3$  concentrations from the NAMN and mean monthly temperature and rainfall from the same selection of sites for the period 1998 to 2014.  $\text{NH}_3$  was negatively correlated with rainfall (blue line:  $\text{Log}(\text{NH}_3) = -0.0057 \cdot \text{Log}(\text{rain}) + 1.0579$ ,  $R^2 = 0.45$ ,  $n = 204$ ,  $p < 0.01$ .) and positively correlated with temperature (red line:  $\text{Log}(\text{NH}_3) = 0.0370 \cdot \text{Log}(\text{temp}) + 0.1580$ ,  $R^2 = 0.24$ ,  $n = 204$ ,  $p < 0.01$ ).
- 20

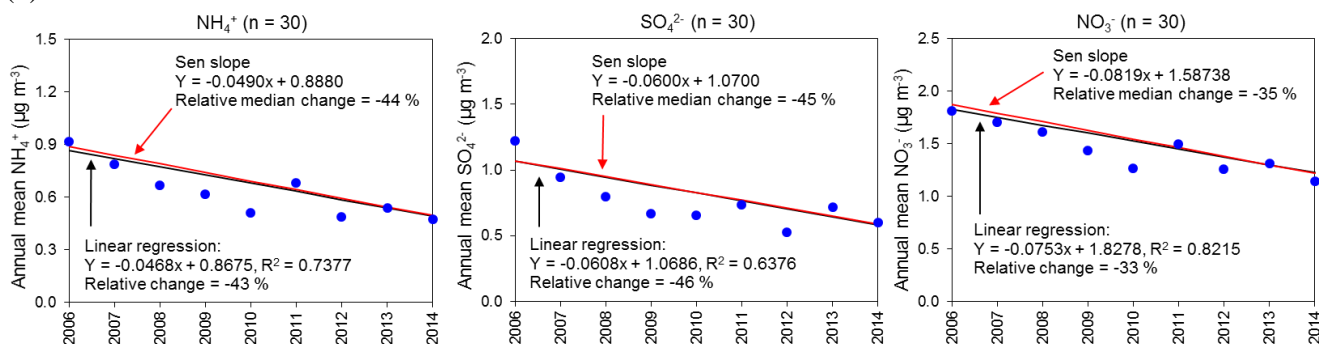


5 **Supp. Figure S5: Time series trend analysis by Mann-Kendall Sen's slope vs squares linear regression on annually averaged particulate  $\text{NH}_4^+$  and gaseous  $\text{NH}_3$  concentrations from the UK National Ammonia Monitoring Network (NAMN) for a)  $\text{NH}_4^+$  (1999-2014, n=23), b)  $\text{NH}_4^+$  (2006-2014, n= 30), c)  $\text{NH}_3$  (1999-2014, n=23, same sites as a), and d)  $\text{NH}_3$  (2006-2014, n=30, same sites as b). Individual data points are annually averaged concentrations.**

10 (a)



(b)



15 **Supp. Figure S6: Time series trend analysis by Mann-Kendall Sen's slope vs linear regression on annually averaged particulate  $\text{NH}_4^+$  from the UK National Ammonia Monitoring Network (NAMN) and particulate  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  from the UK Acid Gas and Aerosol Network (AGANet for time periods a) 2000 – 2014 (12 sites) and b) 2006 – 2014 (30 sites). Individual data points are annually averaged concentrations.**

20