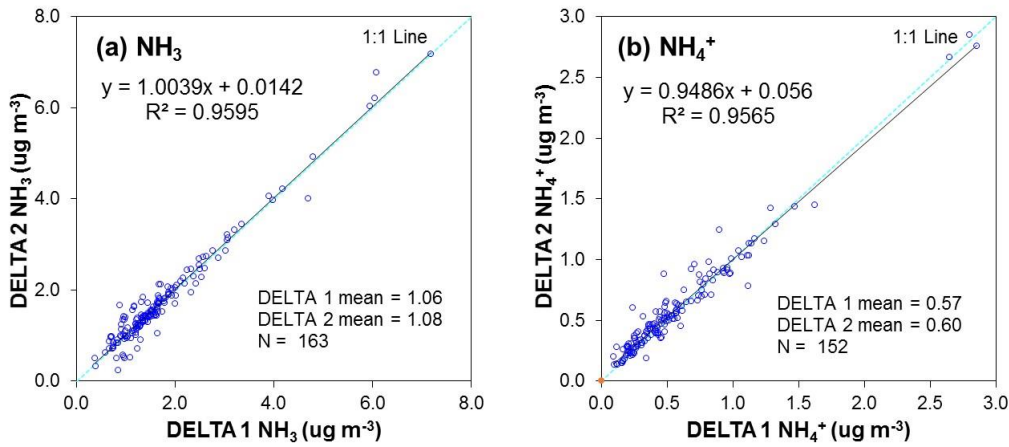
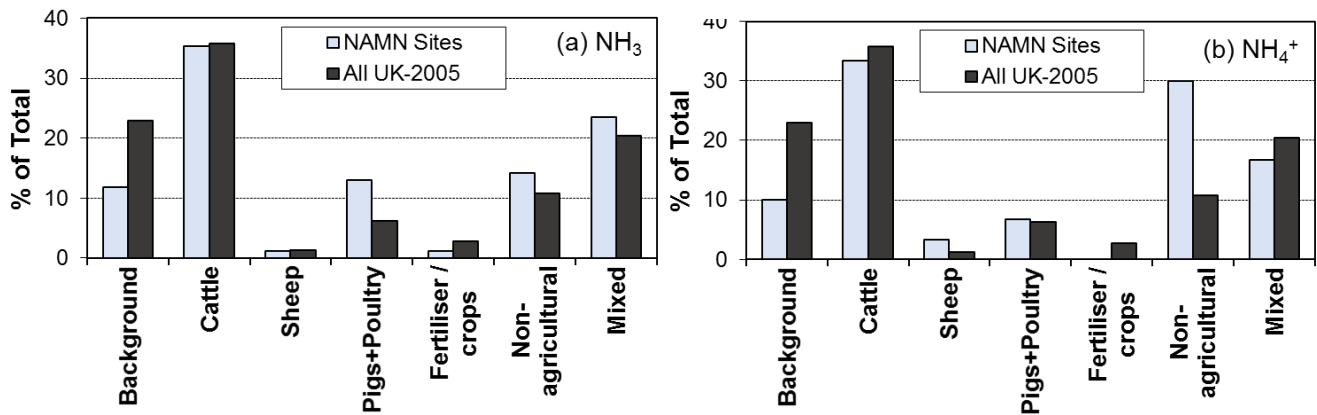
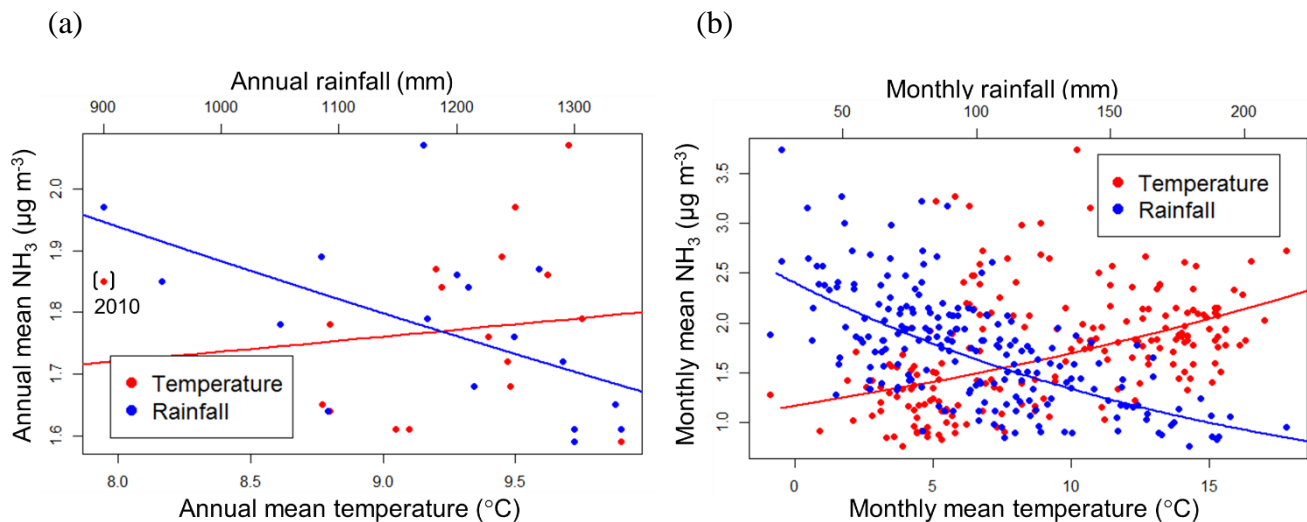
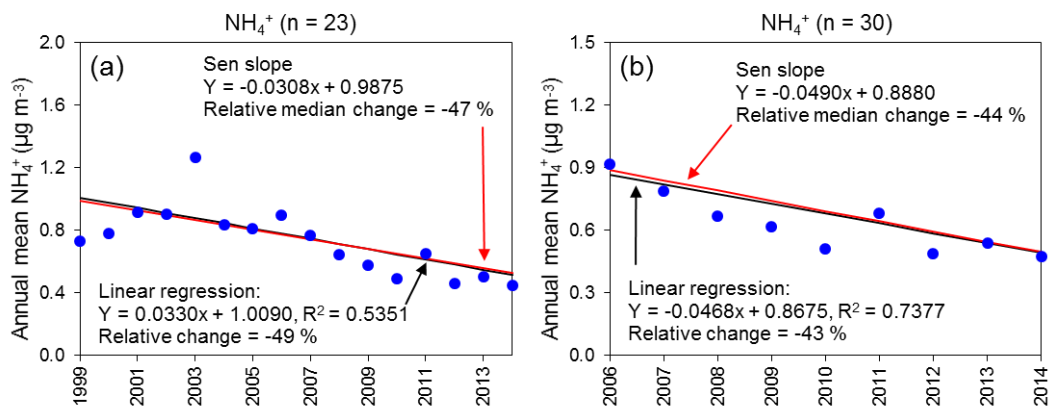
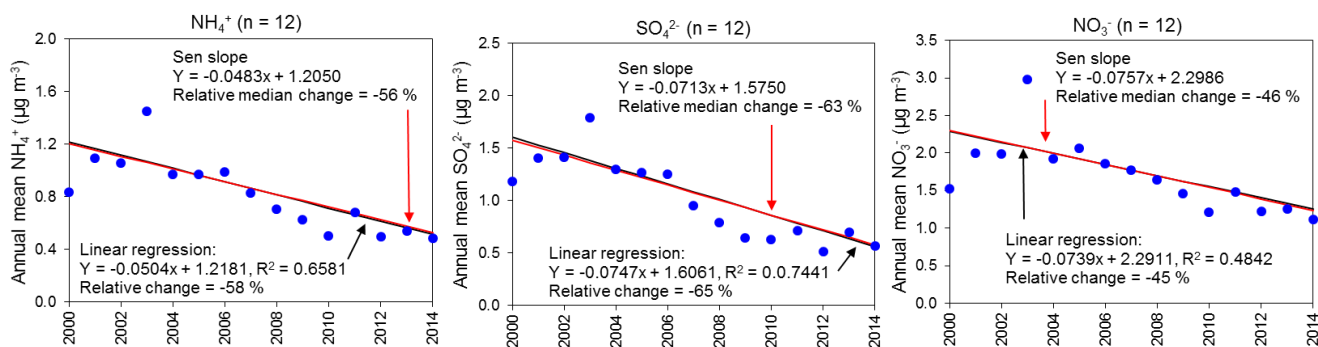
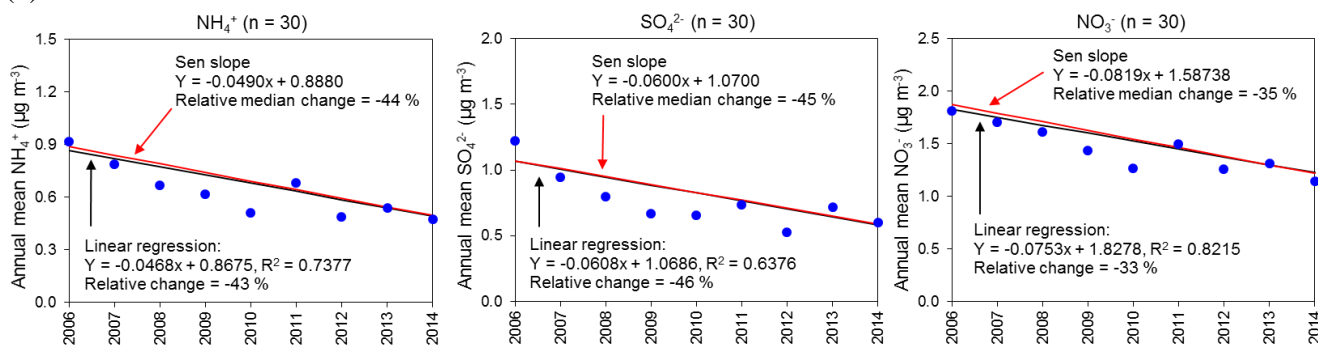
Supplementary material

Supp. Figure S1: Comparisons of parallel measurement of monthly (a) NH_3 and (b) particulate 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.

Supp. Figure S2: Frequency distribution of sites in the UK National Ammonia Monitoring Network NAMN measuring (a) NH_3 (85 sites) and (b) NH_4^+ (30 sites), according to the dominant NH_3 source sectors identified in 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.

Supp. Figure S3: (a) Relationships between UK mean annual measured 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. NH_3 was negatively correlated with rainfall (blue line: $Log(NH_3) = -0.0003*Log(rain) + 0.9656$, $R^2 = 0.32$, $n = 17$, $p = 0.02$). For the relationship between NH_3 and temperature, although most of the data shows an increase in NH_3 with temperature, the correlation was not significant (red line: $Log(NH_3) = 0.0227*Log(temp) + 0.3618$, $R^2 = 0.02$, $n = 17$, $p = 0.59$). 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 NH_3 concentration for 2010 was in fact similar to other years, since the lowest NH_3 concentrations occurred in the winter months. **(b)** Relationships between UK mean monthly measured NH_3 concentrations from the NAMN and mean monthly temperature and rainfall from the same selection of sites for the period 1998 to 2014. NH_3 was negatively correlated with rainfall (blue line: $Log(NH_3) = -0.0057*Log(rain) + 1.0579$, $R_2 = 0.45$, $n = 204$, $p < 0.01$).) and positively correlated with temperature (red line: $Log(NH_3) = 0.0370*Log(rain) + 0.1580$, $R2 = 0.24$, $n = 204$, $p < 0.01$).

Supp. Figure S4: Time series trend analysis by Mann-Kendall Sen's slope *vs* squares linear regression on annually averaged particulate NH₄⁺ and gaseous NH₃ concentrations from the UK National Ammonia Monitoring Network (NAMN) for a) NH₄⁺ (1999-2014, n=23), b) NH₄⁺ (2006-2014, n= 30), c) NH₃ (1999-2014, n=23, same sites as a), and d) NH₃ (2006-2014, n=30, same sites as b). Individual data points are annually averaged concentrations.

(a)

(b)

Supp. Figure S5: Time series trend analysis by Mann-Kendall Sen's slope *vs* linear regression on annually averaged particulate NH₄⁺ from the UK National Ammonia Monitoring Network (NAMN) and particulate NO₃⁻ and SO₄²⁻ 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.