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*Supplement of*

# Acid gases and aerosol measurements in the UK (1999–2015): regional distributions and trends

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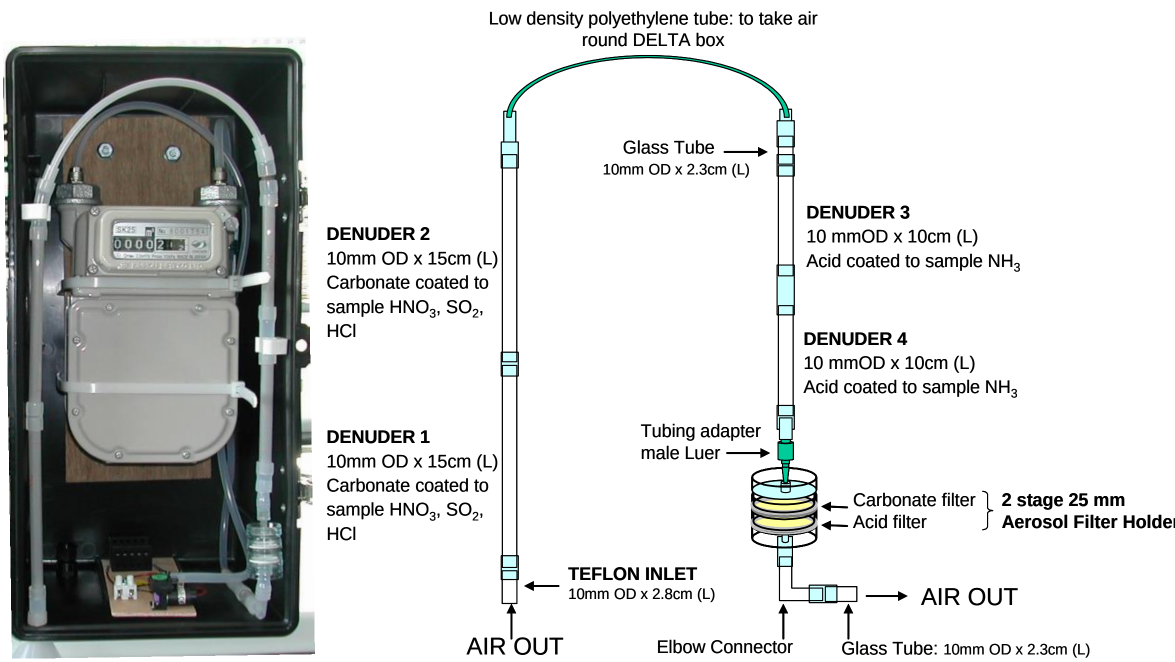
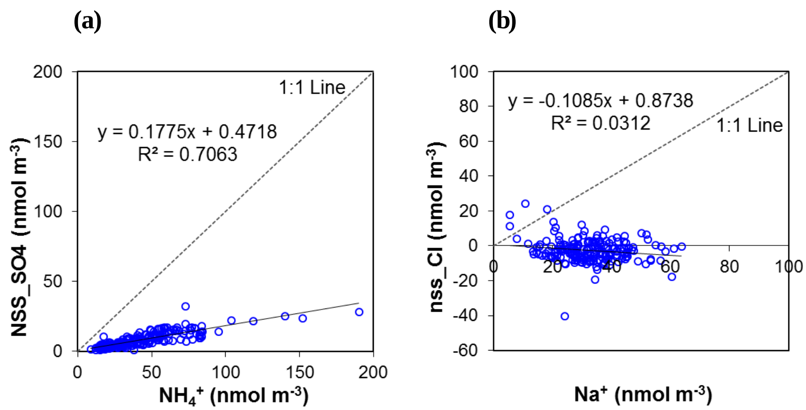
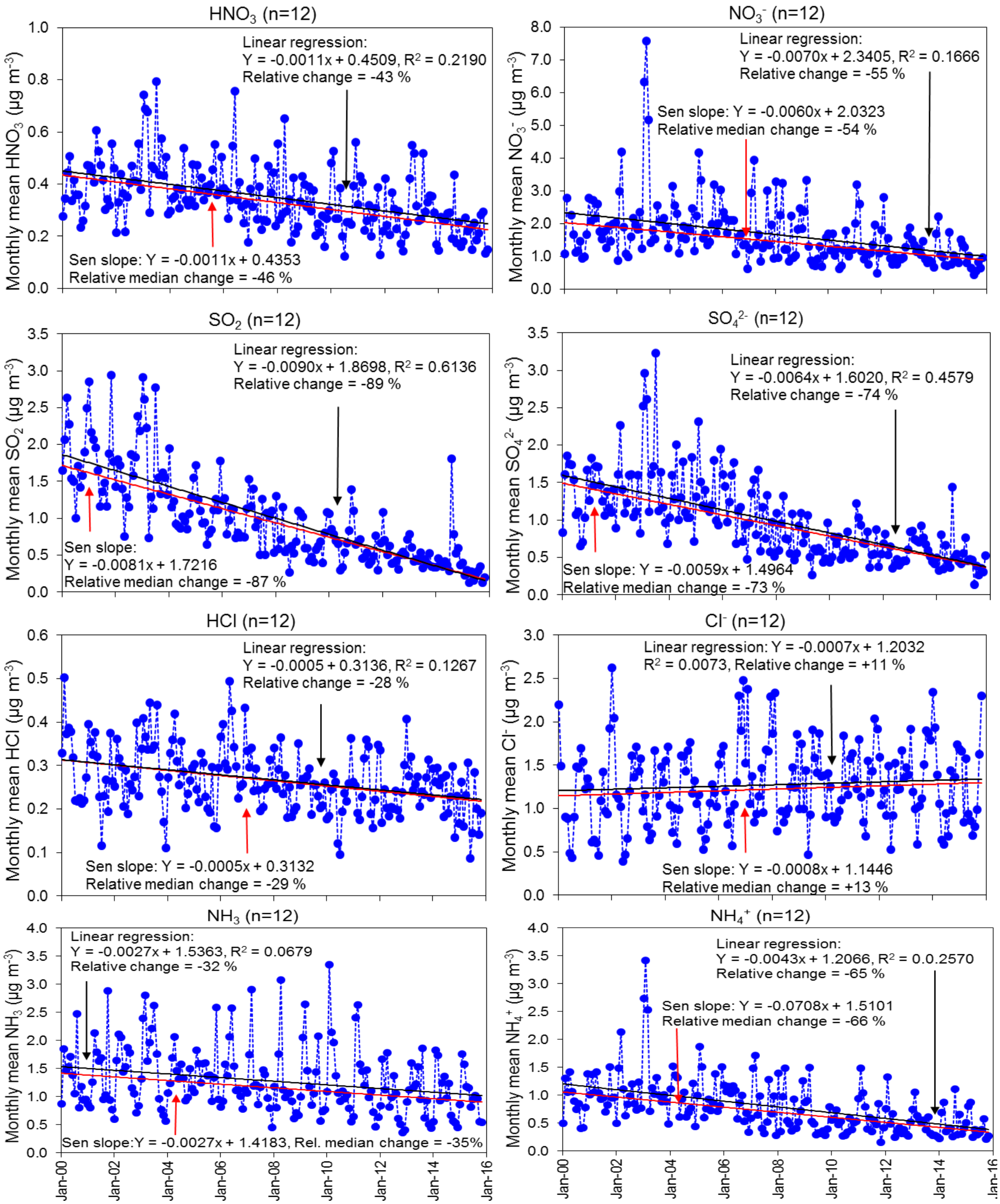


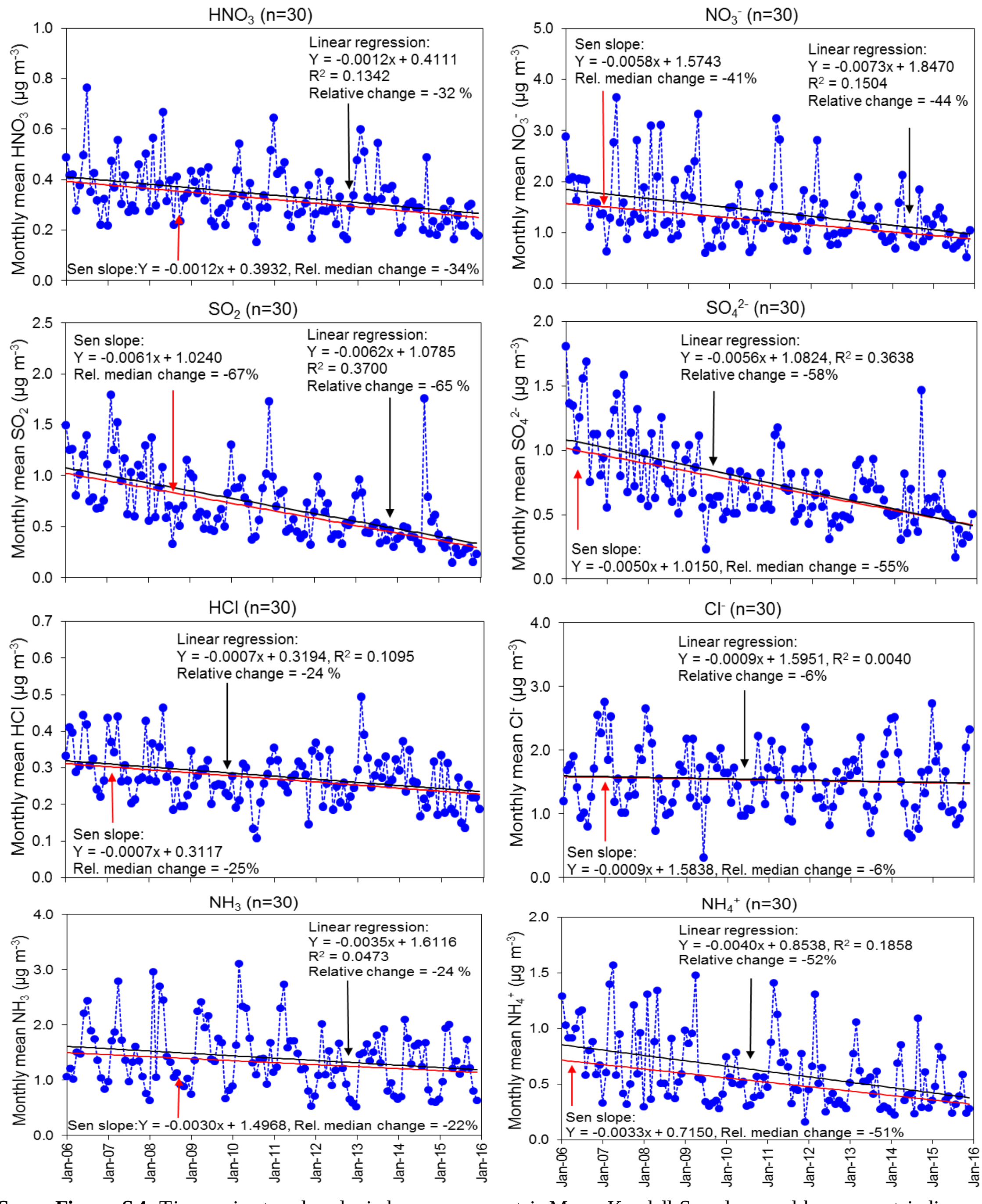

**Supp. Figure S1:** Left: DEnuder for Long-Term Atmospheric sampling (DELTA) as applied for the monthly measurements of reactive gases and particulate matter composition in the UK Acid Gases and Aerosol Network (AGANet), with sampling train *in situ*. Right: sampling train consisting of 2 x 15 cm long $K_2CO_3$ + glycerol coated denuders (determination of $HNO_3$, $SO_2$, HCl), 2 x 10 cm long acid coated denuders (determination of $NH_3$), carbonate coated filter (determination of $NO_3^-$, $SO_4^{2-}$, $Cl^-$, $Na^+$, $Ca^{2+}$, $Mg^{2+}$) and acid coated filter (determination of evolved $NH_4^+$).



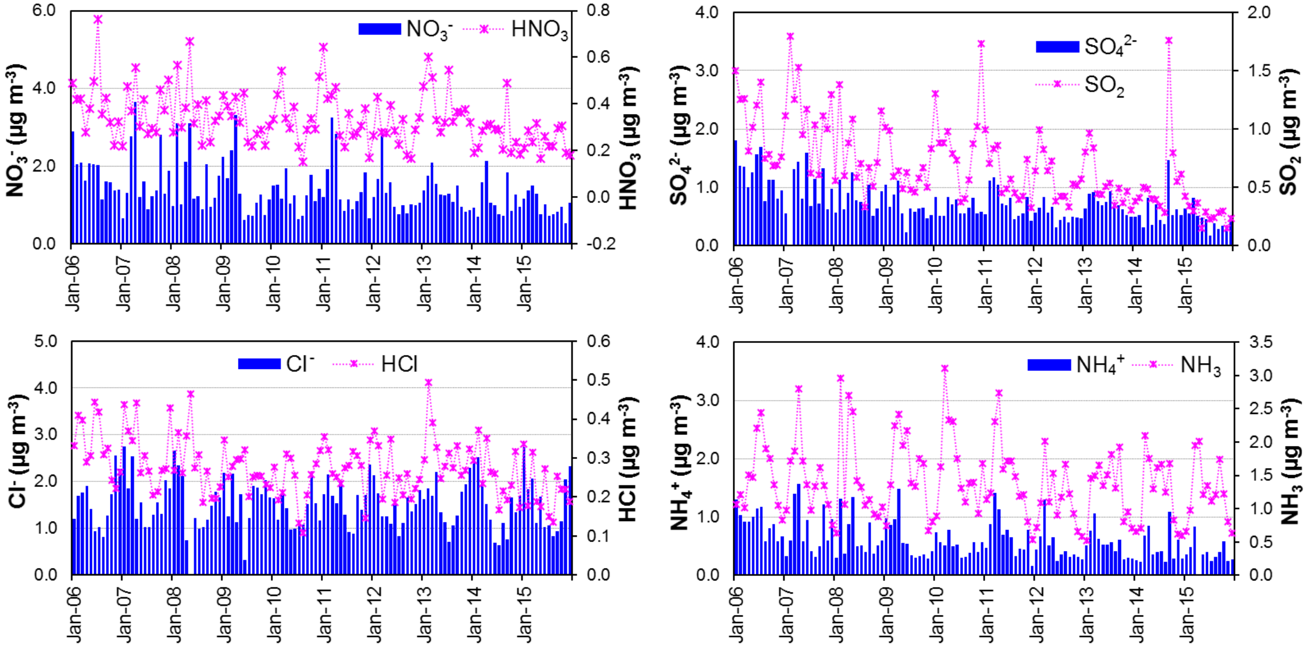

**Supp. Figure S2:** Scatter plots between concentrations of (a) non-sea salt sulphate (nss_SO4) vs $NH_4^+$, and (b) non-sea salt chloride (nss_Cl) *vs* $Na^+$ from mean monthly measurements (1999-2015) for the 12 sites in the UK Acid Gas and Aerosol Monitoring Network (AGANet) that were operational over the whole period. $NH_3$ and $NH_4^+$ data are from the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) made at the same time.



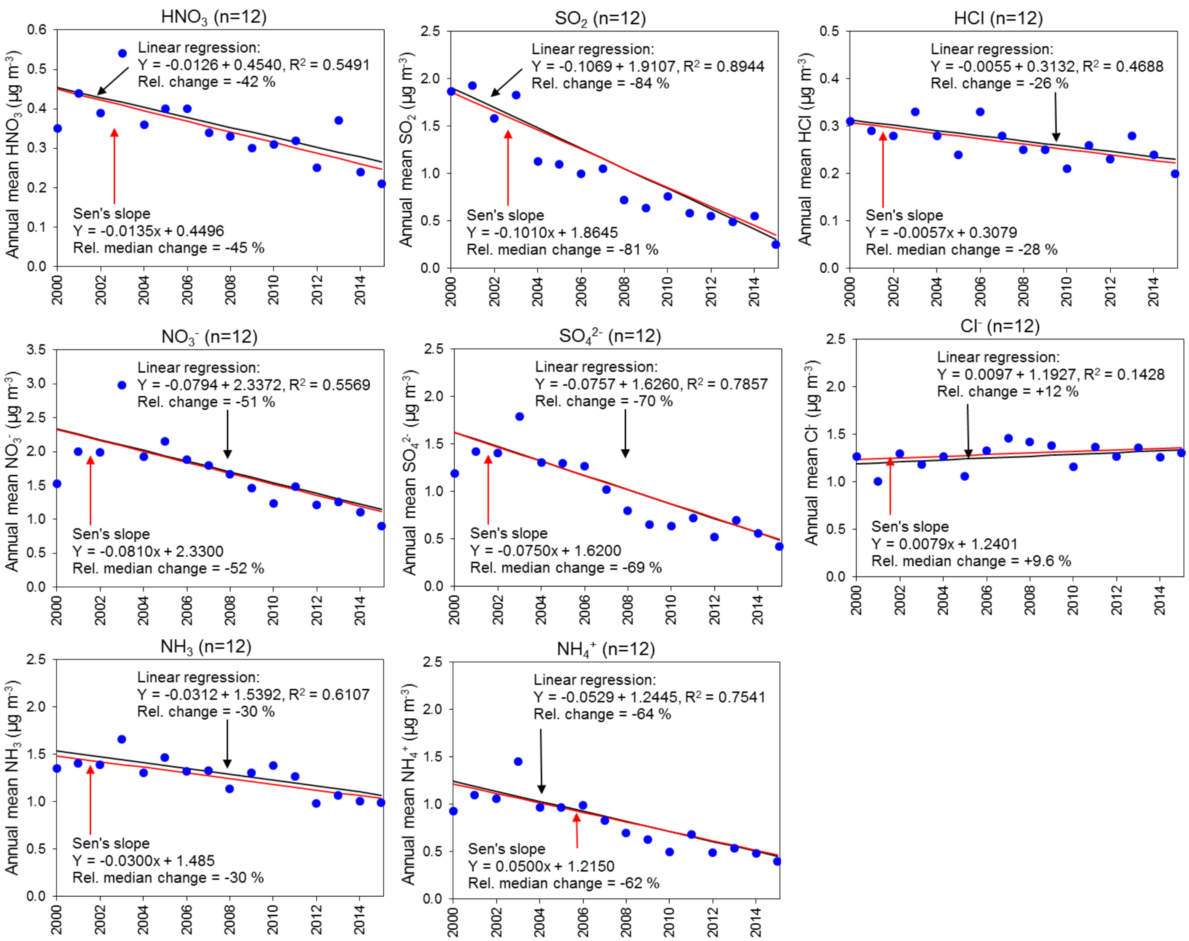

**Supp. Figure S3**: Time series trend analysis by non-parametric Mann-Kendall Sen slope and by parametric linear regression on monthly mean gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 12 sites that were operational over the period 2000 to 2015. $NH_3$ and $NH_4^+$ concentrations data measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. Individual data points are monthly mean concentrations across 12 sites.



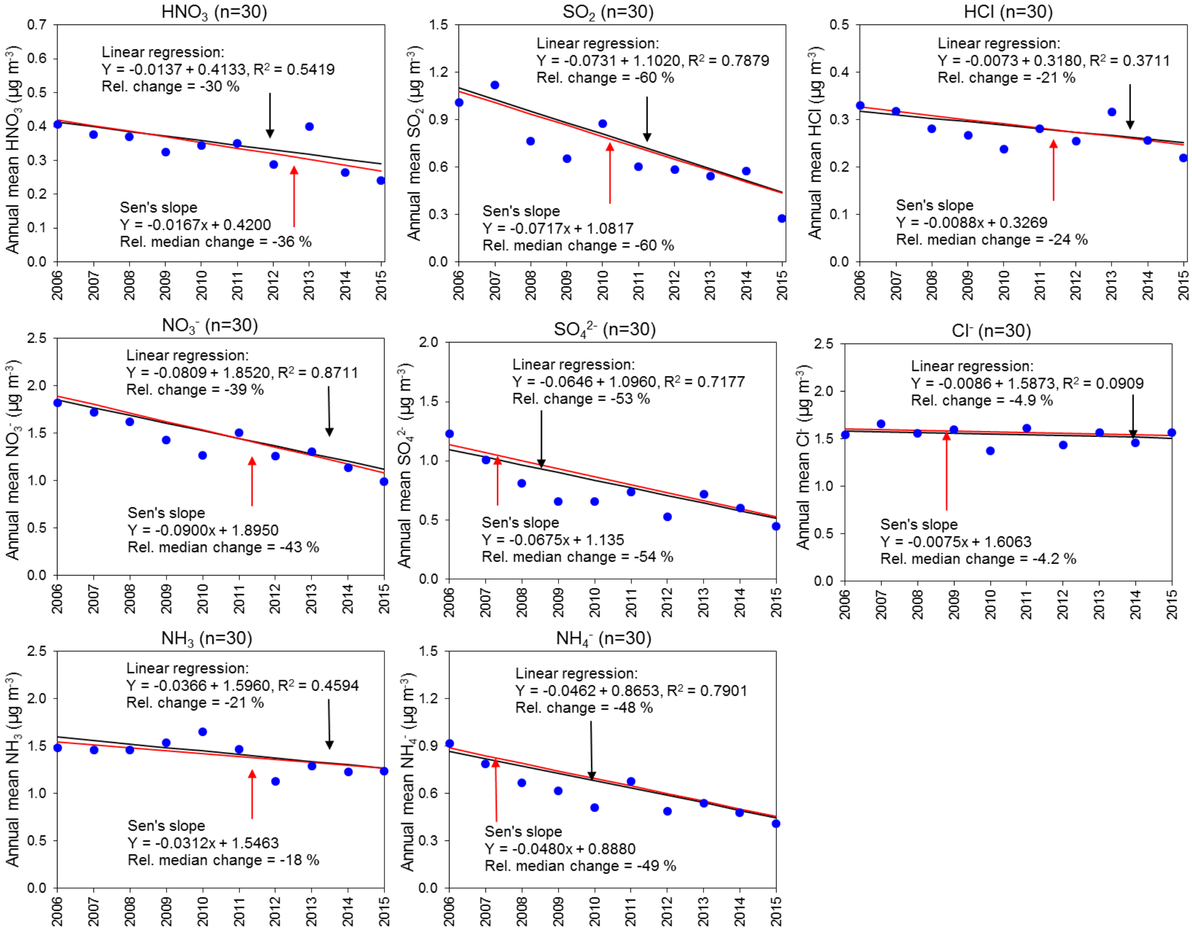

**Supp. Figure S4:** Time series trend analysis by non-parametric Mann-Kendall Sen slope and by parametric linear regression on monthly mean gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 30 sites that were operational over the period 2006 to 2015. $NH_3$ and $NH_4^+$ concentrations data measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. Individual data points are monthly mean concentrations across 30 sites.



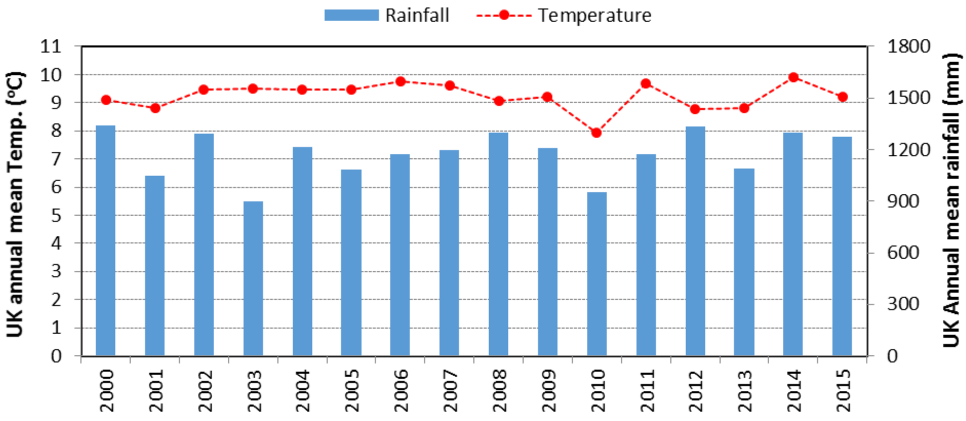

**Supp. Figure S5:** Monthly mean concentrations in gaseous $HNO_3$, $SO_2$, HCl and aerosol $NO_3^-$, $SO_4^{2-}$, $Cl^-$ from the UK Acid Gases and Aerosol Monitoring Network (AGANet) over the period 2006 - 2015. Monthly mean concentrations of $NH_3$ and $NH_4^+$ that were measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also shown for comparison. Each data point in the graphs represents the mean of monthly measurements of 30 sites operational in the network over the period 2006 to 2015.




**Supp. Figure S6:** Time-series trend analysis by non-parametric Mann-Kendall Sen slope and by parametric linear regression on annually averaged gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) of 12 sites that were operational over the period 2000 to 2015. $NH_3$ and $NH_4^+$ concentrations measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison.




**Supp. Figure S7:** Time series trend analysis by non-parametric Mann-Kendall Sen slope and by parametric linear regression on annually averaged gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) of 30 sites that were operational over the period 2006 to 2015. $NH_3$ and $NH_4^+$ concentrations data measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison.




**Supp. Figure S8**: UK annual mean temperature and rainfall (data source: https://www.metoffice.gov.uk/climate/uk/summaries)

**Supp. Table S1:** Major ions measured in DELTA extracts and typical limits of detection (LOD).

| Analytes (denuder aqueous extracts) | Harwell Laboratory (Sep99 – Jun09) | | CEH Lancaster (from Jul09 ) | |
|---|---|---|---|---|
| | Analytical Method | Typical LOD ($\mu g\ m^{-3}$) | Analytical Method | Typical LOD ($\mu g\ m^{-3}$) |
| $NO_3^-$ | IC | 0.05 ($HNO_3$) | IC | 0.05 ($HNO_3$) |
| $NO_2^-$ | Colorimetry | 0.05 (HONO) | IC | 0.05 (HONO) |
| $SO_4^{2-}$ | IC | 0.05 ($SO_2$) | IC | 0.05 ($SO_2$) |
| $Cl^-$ | IC | 0.05 (HCl) | IC | 0.05 (HCl) |

**Supp. Table S2:** Major ions measured in aerosol filter extracts and typical limits of detection (LOD).

| Analytes (aerosol filter aqueous extracts) | Harwell Laboratory (Sep99 – Jun09) | | CEH Lancaster (from Jul09 ) | |
|---|---|---|---|---|
| | Method | Typical LOD ($\mu g\ m^{-3}$) | Method | Typical LOD ($\mu g\ m^{-3}$) |
| $NO_3^-$ | IC | 0.05 ($NO_3^-$) | IC | 0.06 ($NO_3^-$) |
| $NO_2^-$ | Colorimetry | 0.05 ($NO_2^-$) | IC | 0.05 ($NO_2^-$) |
| $SO_4^{2-}$ | IC | 0.06 ($SO_4^{2-}$) | IC | 0.06 ($SO_4^{2-}$) |
| $Cl^-$ | IC | 0.08 ($Cl^-$) | IC | 0.16 ($Cl^-$) |
| $Ca^{2+}$ | IC (Sep99-Jun08) | 0.05 | ICP-OES | 0.09 |
| $Mg^{2+}$ | IC (Sep99-Jun08) | 0.05 | ICP-OES | 0.05 |
| $Na^+$ | IC (Sep99-Jun08) | 0.1 | ICP-OES | 0.16 |
| $Ca^{2+}$ | ICP-AES (Jul08-Jun09) | 0.05 | | |
| $Mg^{2+}$ | ICP-AES (Jul08-Jun09) | 0.05 | | |
| $Na^+$ | ICP-AES (Jul08-Jun09) | 0.1 | | |

**Supp. Table S3:** Calculated lengths of chemically impregnated denuders (borosilicate glass tubes, 10 mm o.d, 0.65 mm i.d) to capture 95 % of gas of interest at a flow rate of 0.4 LPM under laminar flow.

| Reactive gas | $HNO_3$ | $SO_2$ | HCl | $NH_3$ |
|---|---|---|---|---|
| Diffusion coefficient @ 10ºC | $1.15 \times 10^{-5}\ m^2\ s^{-1}$ (Massmana 1998) | $1.22 \times 10^{-5}\ m^2\ s^{-1}$ (Durham & Stockburger 1986) | $5.25 \times 10^{-5}\ m^2\ s^{-1}$ (Mumallah 1986) | $2.01 \times 10^{-5}\ m^2\ s^{-1}$ (Hargreaves & Atkins, 1986) |
| L (cm): for 95 % capture efficiency at flow rate of 0.4 LPM | 14 | 13 | 3 | 8 |

**Note:** 10 cm and 15 cm long denuders are used in the UK National Ammonia Monitoring Network (NAMN) and UK Acid Gases and Aerosol network (AGANet) to sample $NH_3$ and acid gases ($HNO_3$, $SO_2$, HCl), respectively.

**Supp. Table S4:** Summary of Mann-Kendall (MK) and Linear Regression (LR) time series trend analysis on annually averaged gas and aerosol concentrations from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 12 sites that were operational over the period 2000 to 2015. $NH_3$ and $NH_4^+$ concentrations measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. For the MK tests, the 95% confidence interval (CI) for the median trend and relative change are also estimated.

| 2000 - 2015 (12 sites: annual data) | Mann-Kendall (MK) | | Linear Regression (LR) | | |
|---|---|---|---|---|---|
| | [a]Median annual trend & [95% CI] (µg $y^{-1}$) | [b]Relative median change 2000-2015 & [95% CI] (%) | [c]Annual Trend (µg $NH_3$ $y^{-1}$) | [d]Relative change 2000-2015 [%] | $R^2$ |
| $HNO_3$ | -0.0135 [-0.0067, -0.0180] | -45** [-26, -55] | -0.0126 | -42** | 0.549 |
| $SO_2$ | -0.1010 [-0.0729, -0.1250] | -81*** [-72, -91] | -0.1069 | -84*** | 0.894 |
| HCl | -0.0057 [-0.0020, -0.0100] | -28*** [-11, -42] | -0.0055 | -26** | 0.469 |
| $NH_3$ | -0.0300 [-0.0125, -0.0433] | -30** [-13, -39] | -0.0312 | -30*** | 0.611 |
| $NO_3^-$ | -0.0810 [-0.0520, -0.1125] | -52** [-37, -63] | -0.0794 | -51*** | 0.557 |
| $SO_4^{2-}$ | -0.0750 [-0.0450, -0.-0988] | -69** [-52, -82] | -0.0757 | -70*** | 0.786 |
| $Cl^-$ | 0.0079 [-0088, 0.0236] | $9.6^{ns}$ [-9.5, 33] | 0.0097 | $+12^{ns}$ | 0.143 |
| $NH_4^+$ | -0.0500 [-0.0375, -0.0675] | -62** [-51, -74] | -0.0529 | -64*** | 0.754 |

Significance level: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, $^{ns}$ non-significant ($p > 0.05$)

[a]Median annual trend = fitted Sen's slope of Mann-Kendall linear trend (unit = µg $y^{-1}$)

[b]Relative median change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the Sen's slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

[c]Annual trend = fitted slope of linear regression (unit = µg $NH_3$ $y^{-1}$)

[d]Relative change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

**Supp. Table S5:** Summary of Mann-Kendall (MK) and Linear Regression (LR) time series trend analysis on annually averaged gas and aerosol concentrations from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 30 sites that were operational over the period 2006 to 2015. $NH_3$ and $NH_4^+$ concentrations data measured at the same time from the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. For the MK tests, the 95% confidence interval (CI) for the median trend and relative change are also estimated.

| 2006 - 2015 (30 sites: annual data) | Mann-Kendall (MK) | | Linear Regression (LR) | | |
|---|---|---|---|---|---|
| | [a]Median annual trend & [95% CI] (µg $NH_3$ $y^{-1}$) | [b]Relative median change 2000-2015 & [95% CI] (%) | [c]Annual Trend (µg $NH_3$ $y^{-1}$) | [d]Relative change 2000-2015 [%] | $R^2$ |
| $HNO_3$ | -0.0167 [-0.0075, -0.0200] | -36* [-18, -41] | -0.0137 | -30* | 0.542 |
| $SO_2$ | -0.0717 [-0.0300, -0.0108] | -60*** [-33, -73] | -0.0731 | -60*** | 0.788 |
| HCl | -0.0088 [0.0000, -0.0200] | -24* [0.0, -47] | -0.0073 | -21$^{ns}$ | 0.371 |
| $NH_3$ | -0.0312 [0.0033, -0.0625] | -18$^{ns}$ [+2.0, -31] | -0.0366 | -21* | 0.459 |
| $NO_3^-$ | -0.0900 [-0.0580, -0.1300] | -43*** [-30, -56] | -0.0809 | -39*** | 0.871 |
| $SO_4^{2-}$ | -0.0675 [-0.0233, -0.1167] | -54** [-25, -78] | -0.0646 | -53*** | 0.718 |
| $Cl^-$ | -0.0075 [+0.0167, -0.0300] | -4.2$^{ns}$ [+12, -16] | -0.0086 | -4.9$^{ns}$ | 0.091 |
| $NH_4^+$ | -0.0480 [0.0267, -0.0700] | -49** [-33, -64] | -0.0462 | -48*** | 0.790 |

Significance level: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, $^{ns}$ non-significant ($p > 0.05$)

[a]Median annual trend = fitted Sen's slope of Mann-Kendall linear trend (unit = µg $y^{-1}$)

[b]Relative median change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the Sen's slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

[c]Annual trend = fitted slope of linear regression (unit = µg $NH_3$ $y^{-1}$)

[d]Relative change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

**Supp. Table S6:** Comparison of % change in estimated UK $NO_x$, $SO_2$ and $NH_3$ emissions reported by the National Atmospheric Emission Inventory (NAEI) (data from http://naei.defra.gov.uk/) with % change between 2000-2015 (12 sites with complete time series) and between 2006-2015 (30 sites with complete time series) in annually averaged $HNO_3$ / $NO_3^-$ and $SO_2$ / $SO_4^{2-}$ concentrations from the UK Acid Gas and Aerosol Monitoring Network (AGANet), and annually averaged $NH_3$ / $NH_4^+$ concentrations from the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018).

| **Components** | **2000 – 2015 (12 sites)** | | | **2006 – 2015 (30 sites)** | | |
|---|---|---|---|---|---|---|
| | **UK emissions** % change | **MK Sen Slope** % relative median change[a] | **LR** % relative change[b] | **UK emissions** % change | **MK Sen slope** % relative median change[a] | **LR** % relative change[b] |
| **Gas $HNO_3$** | -58 ($NO_x$) | -45** | -42** | -41 ($NO_x$) | -36* | -43* |
| **Particulate $NO_3^-$** | | -52*** | -51*** | | -30** | -39*** |
| **Gas $SO_2$** | -80 ($SO_2$) | -81*** | -84*** | -64 ($SO_2$) | -60*** | -60*** |
| **Particulate $SO_4^{2-}$** | | -69*** | -70*** | | -54** | -53*** |
| **Gas $NH_3$** | -10 ($NH_3$) | -30** | -30*** | -3.5 ($NH_3$) | -18[ns] | -21* |
| **Particulate $NH_4^+$** | | -62*** | -64*** | | -49** | -48*** |

Significance level: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, [ns] non-significant ($p > 0.05$)

**Supp. Table S7:** Summary of Mann-Kendall (MK) and Linear Regression (LR) time series trend analysis on monthly mean gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 12 sites that were operational over the period 2000 to 2015. $NH_3$ and $NH_4^+$ concentrations measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. For the MK tests, the 95% confidence interval (CI) for the median trend and relative change are also estimated.

| 2000 - 2015 (12 sites: monthly data) | Mann-Kendall (MK) | | Linear Regression (LR) | | |
|---|---|---|---|---|---|
| | [a]Median annual trend & [95% CI] (µg $NH_3$ $y^{-1}$) | [b]Relative median change 2000-2015 & [95% CI] (%) | [c]Annual Trend (µg $NH_3$ $y^{-1}$) | [d]Relative change 2000-2015 [%] | $R^2$ |
| $HNO_3$ | -0.0132 [-0.0096, -0.0156] | -46*** [-36, -52] | -0.0128 | -43*** | 0.219 . |
| $SO_2$ | -0.0972 [-0.0864, -0.1080] | -87*** [-82, -90] | -0.1078 | -89*** | 0.614 |
| HCl | -0.0060 [-0.0036, -0.0084] | -29*** [-19, -40] | -0.0058 | -28*** | 0.127 |
| $NH_3$ | -0.0324 [-0.0168, -0.0468] | -35*** [-20, -46] | -0.0325 | -32*** | 0.068 |
| $NO_3^-$ | -0.0720 [-0.0528, -0.0900] | -54*** [-44, -63] | -0.0839 | -55*** | 0.167 |
| $SO_4^{2-}$ | -0.0708 [-0.0600, -0.0804] | -73*** [-66, -77] | -0.0770 | -74*** | 0.458 |
| $Cl^-$ | 0.0096 [-0.0048, 0.0252] | +13[ns] [-5.7, +36] | +0.0085 | +11[ns] | 0.007 |
| $NH_4^+$ | -0.0456 [-0.0360, -0.0552] | -66*** [-57, -74] | -0.0516 | -65*** | 0.257 |

Significance level: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, [ns] non-significant ($p > 0.05$)

[a]Median annual trend = fitted Sen's slope of Mann-Kendall linear trend (unit = µg $y^{-1}$)

[b]Relative median change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the Sen's slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

[c]Annual trend = fitted slope of linear regression (unit = µg $NH_3$ $y^{-1}$)

[d]Relative change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

**Supp. Table S8:** Summary of Mann-Kendall (MK) and Linear Regression (LR) time series trend analysis on monthly mean gas and aerosol concentration data from the UK Acid Gases and Aerosol Monitoring Network (AGANet) for the 30 sites that were operational over the period 2006 to 2015. $NH_3$ and $NH_4^+$ concentrations measured at the same time in the UK National Ammonia Monitoring Network (NAMN, Tang et al., 2018) are also included for comparison. For the MK tests, the 95% confidence interval (CI) for the median trend and relative change are also estimated.

| 2006 - 2015 (30 sites: monthly data) | Mann-Kendall (MK) | | Linear Regression (LR) | | |
|---|---|---|---|---|---|
| | [a]Median annual trend & [95% CI] (µg $NH_3$ $y^{-1}$) | [b]Relative median change 2006-2015 & [95% CI] (%) | [c]Annual Trend (µg $NH_3$ $y^{-1}$) | [d]Relative change 2006-2015 [%] | $R^2$ |
| $HNO_3$ | -0.0144 [-0.0072, -0.0204] | -34*** [-18, -44] | -0.0146 | -32*** | 0.134 |
| $SO_2$ | -0.0732 [-0.0564, -0.0924] | -67*** [-59, -74] | -0.0749 | -65*** | 0.370 |
| HCl | -0.0084 [-0.0036, -0.0120] | -25** [-11, -33] | -0.0084 | -24*** | 0.109 |
| $NH_3$ | -0.0360 [-0.0036, -0.0696] | -22* [-26, -39] | -0.0425 | -24* | 0.047 |
| $NO_3^-$ | -0.0696 [-0.0408, -0.0101] | -41*** [-26, -54] | -0.0875 | -44*** | 0.150 |
| $SO_4^{2-}$ | -0.0600 [-0.0432, -0.0768] | -55*** [-44, -64] | -0.0674 | -58*** | 0.364 |
| $Cl^-$ | -0.0108 [-0.0228, -0.0432] | $-6^{ns}$ [-14, +23] | -0.0111 | $-6^{ns}$ | 0.004 |
| $NH_4^+$ | -0.0396 [-0.0024, -0.0588] | -51*** [-35, -64] | -0.0480 | -52*** | 0.186 |

Significance level: * $p < 0.05$, ** $p < 0.01$, *** $p < 0.001$, $^{ns}$ non-significant ($p > 0.05$)

[a]Median annual trend = fitted Sen's slope of Mann-Kendall linear trend (unit = µg $y^{-1}$)

[b]Relative median change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the Sen's slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

[c]Annual trend = fitted slope of linear regression (unit = µg $NH_3$ $y^{-1}$)

[d]Relative change calculated based on the estimated annual concentration at the start ($y_0$) and at the end ($y_i$) of time series computed from the slope and intercept (=100*[($yi$-$y_0$) /$y_0$])

**Denuder theory:**

The length of denuder required to obtain near complete capture of a reactive gas is a function of the diffusion rate of the reactive gas and the air sampling rate. For cylindrical tubes, with laminar flow and where the tube wall is a perfect sink for the gas of interest, Gormley & Kennedy (1949) and Ferm (1979) showed that the collection efficiency of a simple denuder for a reactive gas may be calculated using Equation 1.

$$\eta = 1 - \frac{\beta_1}{\beta_0} = 1 - 0.819 \cdot e^{-14.6272\delta} + 0.0976 \cdot e^{-89.22\delta_r} + 0.01896 \cdot e^{-212\delta_r} \quad (1)$$

where

$\eta$ is the collection efficiency of the denuder;
$\beta_1$ is the mass concentration of gas at the denuder outlet
$\beta_0$ is the mass concentration of gas at the denuder inlet

$\delta$ is described by Equation 2:

$$\delta = \frac{\pi D L}{4\phi} \quad (2)$$

where

$D$ is the molecular diffusion coefficient of reactive gas, in $cm^2/s$
$L$ is effective length of the denuder, in cm
$\phi$ is the air flow rate through the denuder, in $cm^3/s$.

For collection efficiencies ≥ 95%, contributions from terms 2 and 3 in Equation 1 are insignificant (< 0.3 %) and only the first term is significant. Equation 1 may then be simplified to Equations 3 and 4:

$$\eta = 1 - \frac{\beta_1}{\beta_0} = 1 - 0.819 \cdot e^{-14.6272\left(\frac{\pi DL}{4\phi}\right)} \quad (3)$$

$$\frac{\beta 1}{\beta o} = 0.819\, e^{-14.6272\ \left(\frac{\pi DL}{4\phi}\right)} \quad , \frac{\beta 1}{\beta o} = 0.05 \text{ for 95 \% capture efficiency} \quad (4)$$

Laminar flow is achieved a short distance from the inlet. The minimum length of tube at inlet not coated with sorbent, $L_{min}$ to fully develop laminar flow is given by Equation 5.

$$L_{min} = 0.05 . \mathrm{Re} . d \quad (5)$$

where

Re is the Reynolds number
$d$ internal diameter of tube

An inlet length of 2.8 cm (uncoated Teflon tube: 10 mm o.d, 6.5 mm i.d) is used in the AGANet sampling train to develop laminar flow. Reynolds number must be <2000 for laminar flow. The Reynolds number for this is calculated to be 86 (at flow rate = 0.4 LPM and internal tube diameter = 6.5 mm).

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