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Trends in bulk deposition of acidity in the UK, 1988–2007, assessed using additive models

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ABSTRACT

Previous analyses of deposition trends in the UK have used traditional linear methods, but recognised that neither emissions nor acid deposition had followed linear trends. Here we employ a non-linear technique, an additive model, to determine trends in both concentrations and bulk deposition loads (precipitation-weighted) at 12 Acid Deposition Monitoring Network (ADMN) sites most closely co-located with sites in the UK Acid Waters Monitoring Network (AWMN). Bulk deposition data were collected weekly or two-weekly over the 20 year period from 1988 to 2007 and all samples were analysed according to common protocols. Periods of significant increase or decrease over the period of monitoring were identified using the first derivative of the fitted trend, computed using finite differences. Results of the trend analysis show that:

1. 11 out of 12 bulk deposition sites show significant increasing trends in pH.
2. Concentrations of non-marine sulphate (xSO_4^{2-}) show significant decreasing trends at ten out of 12 sites and appear to be the main driver of changes in bulk deposition pH since the late 1990s, though earlier trends in bulk deposition pH are so far unexplained.
3. Trends in concentrations of nitrogen (N) species are mixed with many sites showing no significant trend. For NO_3^- three sites show a general decline while four sites show a short period of significant increase in the early 1990s which in three cases reverses to a significant declining trend in the late 1990s. For NH_4^+ five sites show some periods of significant decline in concentration but these are only prolonged at three sites.
4. Precipitation-corrected deposition loads show very similar trends to concentrations for xSO_4^{2-} and N species, but for N species there are more sites with significant trends.
5. For total Cl^- concentrations, seven sites show declining trends but only four of these remain significant with precipitation weighting. Non-marine chloride (xCl^-) concentrations decline significantly at nine sites, reducing to six sites with precipitation weighting. Hence reductions in xCl^- are acting alongside xSO_4^{2-} as drivers of declining acidity in bulk deposition.
6. Small declining trends in precipitation measured in the bulk deposition collectors may reflect changes in sampling methodology as sampling frequency changed from weekly to 2-weekly during the ADMN monitoring period.

These non-linear trends explain the lack of significant trends using linear methods on shorter data series in previous analyses and may help to explain non-linear patterns in chemical recovery in surface waters.

1. Introduction

Acid deposition has been recognised as a driver of ecological change in sensitive aquatic ecosystems in the UK for over 25 years (e.g. Battarbee, 1990). During the 1980s the UK government Department of the Environment set up a series of research and monitoring programmes including the Acid Deposition Monitoring Network (ADMN; Lawrence et al., 2008) to monitor deposition loads to ecosystems and the Acid Waters Monitoring Network

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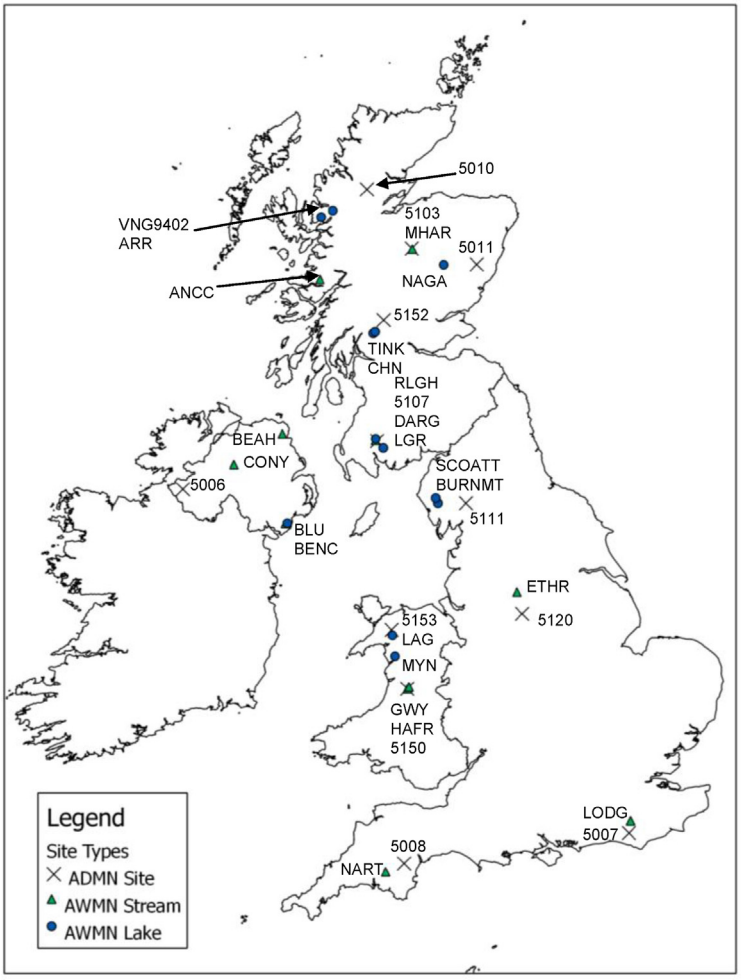

Acid Deposition Monitoring Network	
Site Code	**Site Name**
5006	Lough Navar
5007	Barcombe Mills
5008	Yarner Wood
5010	Strathvaich Dam
5011	Glen Dye
5103	Allt a' Mharcaidh
5107	Loch Dee
5111	Bannisdale
5120	Wardlow Hay Cop
5150	Pumlumon
5152	Balquhidder
5153	Llyn Llydaw
Acid Waters Monitoring Network	
Site Code	**Site Name**
ANCC	Allt na Coire nan Con
ARR	Loch Coire nan Arr
BEAH	Beaghs Burn
BENC	Bencrom River
BLU	Blue Lough
BURNMT	Burnmoor Tarn
CHN	Loch Chon
CONY	Coneyglen Burn
DARG	Dargall Lane
ETHR	River Etherow
GWY	Afon Gwy
HAFR	Afon Hafren
LAG	Llyn Llagi
LGR	Loch Grannoch
LODG	Old Lodge Stream
MHAR	Allt a'Mharcaidh
MYN	Llyn cwm Mynach
NAGA	Lochnagar
NART	Narrator Brook
RLGH	Round Loch of Glenhead
SCOATT	Scoat Tarn
TINK	Loch Tinker
VNG9402	Loch Coire Fionnaraich

Fig. 1. Location of AWMN sites and ADMN sites assessed for bulk deposition trends.

(AWMN: Shilland et al., this issue) to monitor chemical and biological responses of aquatic ecosystems to changes in acid deposition loads. While the initial remit of the ADMN and AWMN was primarily to monitor the effectiveness of measures to reduce emissions of sulphur, the main agent of acidification in the 1980s, both networks have been invaluable in demonstrating the additional impacts of nitrogen (N) deposition, which has increased in importance over the last decade as sulphur deposition has declined dramatically from its peak in the 1970s (RoTAP, 2012). Furthermore, it has been recognised in recent years that as well as contributing to acidification (Curtis et al., 2005), the enhanced nitrate leaching caused by atmospheric N deposition is also changing nutrient balances in upland rivers and lakes, where N limitation of primary production is found to be much more widespread than previously recognised (Maberly et al., 2002; Curtis et al., 2014). Here we utilise selected sites from the ADMN to assess the trends in the concentrations and deposition loads of anthropogenic sulphur, nitrogen and chloride compounds in bulk deposition, collected weekly or 2-weekly, at sites in the AWMN over the twenty year period from 1988 to 2007. Given the non-linearities reported in previous trend analyses of shorter deposition data series (Fowler et al., 2007; Matejko et al., 2009), we employ non-linear methods to model trends in the data without any aggregation to annual time scales.

1.1. Context

The most recent assessment of deposition trends in the UK was presented in the Review of Transboundary Air Pollution (RoTAP, 2012), which summarised trends in acid deposition up to 2006 based on 35 sites in the ADMN. Sulphur deposition in the UK declined by 80% from 1986 to 2006, while total N deposition did not change significantly through this period. Despite a decline in emissions of oxidised N species (NO_x), ammonia emissions remained level and most of the reductions in NO_x deposition related to pollution that was exported overseas, with much lower reductions in deposition within the UK of only 15%. Concentrations of acidity, non-marine SO_4^{2-} (hereafter xSO_4^{2-}) and non-marine chloride (xCl^-) decreased by 85%, 75% and 95% respectively, with xSO_4^{2-} accounting for 75% of the trend in deposited acidity. Currently levels of reduced N deposition are similar to those for oxidised N (RoTAP, 2012).

For all these pollutants, total deposition at the national scale is dominated by the wet deposition component, but there is great spatial variation with distance from sources (RoTAP, 2012). The relative importance of wet deposition inputs for xSO_4^{2-}, NO_x (as NO_3^-) and reduced N (as NH_4^+) for the 5 km grid square containing AWMN site sampling locations (Fig. 1) in the UK is shown in Table 1. Data are annual mean concentration-based estimated deposition (CBED) loads averaged over the three year period 2004–06 and derived partly from ADMN data across the UK. Wet deposition of xSO_4^{2-} makes up between 70 and 95% of total non-marine inputs, with the lowest proportion at Old Lodge in south-east England and the highest proportion in Scotland. A much greater range is found for the proportion of wet deposited NO_3^-, from 36% at Old Lodge and 53–66% in Wales to a maximum of 85% in the Trossachs of Scotland. For wet deposited NH_4^+ the proportion varies from 51% of