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Trends and variability in weather and atmospheric deposition at UK Environmental Change Network sites (1993 – 2012).

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Abstract

We characterised temporal trends and variability in key indicators of climate and atmospheric deposition chemistry at the twelve terrestrial UK Environmental Change Network (ECN) sites over the first two decades of ECN monitoring (1993 – 2012) using various statistical approaches. Mean air temperatures for the monitoring period were approximately 0.7 °C higher than those modelled for 1961-1990, but there was little evidence for significant change in air temperature over either the full monthly records or within individual seasons. Several upland ECN sites, however, warmed significantly over the first decade before cooling in the second. Summers at most sites became progressively wetter, and extremes in daily rainfall increased in magnitude. Average wind speeds in winter and spring declined at the majority of sites. Directional trends in summer precipitation could be linked to an atypically prolonged negative deviation in the summer North Atlantic Oscillation (NAO) Index. Several aspects of air quality improved markedly. Concentrations and fluxes of sulphate in precipitation declined significantly and substantially across the network, particularly during the earlier years and at the most polluted sites in the south and east. Precipitation concentrations of nitrate and ammonium, and atmospheric concentrations of nitrogen dioxide also decreased at most sites. There was less evidence for reductions in the loads of wet deposited nitrogen species, while trends in atmospheric ammonia concentration varied in direction and strength between sites. Reductions in acid deposition are likely to account for widespread gradual increases in the pH of soil water at ECN sites, representing partial recovery from acidification. Overall, therefore, ECN sites have experienced marked changes in atmospheric chemistry and weather regimes over the last two decades that might be expected to have exerted detectable effects on ecosystem structure and function. While the downward trend in acid deposition is unlikely to be reversed, it is too early to conclude whether the trend towards wetter summers simply represents a phase in a multi-decadal cycle, or is indicative of a more directional shift in climate. Conversely, the first two decades of ECN now provide a relatively stable long term baseline with respect to air temperature, against which effects of anticipated future warming on these ecosystems should be able to be assessed robustly.

1. Introduction

The environmental, biogeochemical and ecological character of most of the non-urban UK land surface is subject to a range of local-scale pressures resulting from its use for agriculture, water supply, forestry, recreation, etc. However, the "natural" environment is also under the dynamic influence of regional-scale pressures from climate and air pollution. The magnitude and temporal dynamics of these more pervasive drivers of change need to be taken into account in any assessment of the causes of long term changes in ecosystem structure and function, regardless of the spatial scale of interest. The UK Environmental Change Network (ECN) was established in the early 1990s to provide UK-wide evidence supporting the detection, quantification and attribution of the impacts of environmental change on the ecological state of a wide range of UK habitats, including 12 terrestrial sites (the terrestrial ECN). While gradual long-term shifts in land use are sometimes inevitable (see Dick et al. in review, this issue), management regimes at ECN terrestrial sites have been kept as constant as possible since the onset of monitoring, thereby maximising the sensitivity of environmental and ecological indicators to regional-scale influences, and particularly changes in climate and air pollution (Sier et al., this issue).

ECN monitoring has been conducted over a period when changes in regional-scale environmental drivers might have been expected to be substantial. Global carbon emissions increased at a rate of 1.0% yr^{-1} in the 1990s and 3.1 % yr^{-1} since 2000, while atmospheric concentrations of carbon dioxide, the principal greenhouse gas (GHG), measured at the Mauna Loa Observatory, increased from 357 to 394 ppm between 1993 and 2012 (NOAA-ESRL data). This has been accompanied by a considerable and progressive increase in the heat content of the world's oceans (Abraham et al., 2013) and global surface temperatures (Hansen et al., 2010). Sea levels have risen by a global average of approximately 6 cm (Blunden and Arndt, 2013), while summer minimum Arctic Sea Ice area extent has contracted by between 9.4 to 13.6 % $decade^{-1}$ over the past three decades (Swart et al., 2015).

Global anthropogenically-driven warming is predicted to affect the climate of the UK in the long term through increased air temperatures, changes in the amounts and seasonal distribution of precipitation, and increases in the frequency of extreme climatic events including floods and droughts (Jenkins et al., 2009). These trends will be mediated at more local scales by relatively short-term variation in ocean temperatures and the position, and pressure gradients, of the earth's teleconnected regional atmospheric circulation systems. Inter-annual variability in UK weather is particularly well summarised by the North Atlantic Oscillation Index (NAOI) (Hurrell and Van Loon, 1997) – the standardised difference in sea level atmospheric pressure between fixed points in the Azores and Iceland. The NAO tends to vary at an approximately decadal frequency and has, in turn, been argued to be sensitive to the extra-terrestrial influence of subtle variation in solar activity (Brown and John, 1979; Lockwood et al., 2010; Scaife et al., 2013).

Separately, major reforms to energy policy in recent decades in northern hemisphere industrialised countries, influenced by statutory controls on the emission of acidifying, eutrophying and other toxic pollutants from industrial, agricultural and domestic sectors, have been implemented nationally and internationally (Schöpp et al., 2003), while the UK economy has shifted from a largely manufacturing- to service-based economy. This has resulted in large reductions in emissions of sulphur and heavy metals to the atmosphere across Europe and North America, and smaller reductions in the emissions of reactive nitrogen species (Fowler et al., 2007). Recent reductions in the deposition of sulphur and acidity across the UK have been linked to marked chemical improvements in soil and surface water chemistry (RoTAP, 2012). In the meantime, however, rapid economic development in parts of the developing world has contributed to broadly opposite trends (Lu et al., 2011), with major implications for human health and environmental sustainability (Zhang et al., 2012).

Finally, air pollution, particularly in the form of sulphate (SO_4^{2-}) aerosol, can itself have a marked effect on climate, both directly, by reflecting short-wave radiation, and by providing condensation nuclei for cloud formation, in a process known as “solar dimming” (Stanhill and Cohen, 2001). Gedney et al. (2014) reported that river flows in some of the most polluted regions of northern Europe were up to 25% higher than normal when aerosol levels peaked around 1980, and attributed this to reduced evaporative loss. The authors proposed that hydrological trends might be reversed with more recent “global brightening”. Changes in climate can also influence fluxes and concentrations of atmospheric pollutants to the land surface. Ambient pollution levels are heavily dependent on prevailing air mass trajectories (Fleming et al., 2012), while rainfall events can increase both fluxes and concentrations of pollutants, particularly in upland environments through the feeder-seeder effect (Inglis et al., 1995).

The ambient environment of semi-natural systems across the UK, including those represented by ECN, may therefore be expected to have undergone significant shifts in both climate- and pollution-related ecological stressors over the past two decades. Quantification and characterisation of these changes are essential prerequisites for the appropriate attribution of single and interactive effects on soils, surface waters, species and ecosystems. The first broad assessment of trends in physical and biogeochemical drivers of environmental change at ECN sites was conducted by Morecroft et al. (2009), who also reported on trends in ecological indicators of change. Statistical analyses were largely confined to tests of linear change in variables summarised at an annual scale. The study period (1993-2008) was characterised by marked increases in air temperature across most seasons (amounting to circa 1 °C decade^{-1}), in addition to large reductions in concentrations of SO_4^{2-} and acidity in precipitation. However, Swart et al. (2015), in their assessment of recent trends in Arctic sea ice, emphasise that climate change is unlikely to be uniform, and characterisation of trends using linear methods alone is thus vulnerable to the specific period chosen for analysis. A succession of relatively cool years since 2008 (e.g.(Cattiaux et al., 2010)) and evidence for a recent stabilisation of atmospheric pollutant deposition rates (Curtis and Simpson, 2014), render a simple repeat of previously applied linear analysis of restricted value, while acquisition of a further five years data has increased options to characterise trends using non-linear approaches.

In the following assessment, therefore, we apply both linear and non-linear statistical analyses of key meteorological and air pollutant variables at the 12 terrestrial ECN sites covering the period 1993-2012 with the aim of: 1) quantifying net change over the full period, 2) characterising temporal variation in the rate and statistical significance of change, and 3) testing for evidence of changing frequency of extreme meteorological events. We also refer to modelled meteorological data to provide longer term context for the changes recorded over the past two decades at each site.

2. Methods

2.1 Sites

For most of the last two decades the terrestrial ECN comprised 12 sites spanning much of the UK. Information regarding location, biogeographical characteristics and dates of initiation of monitoring is provided by Sier et al (in review, this issue), but sites range from lowland agricultural systems, including Rothamsted and Drayton, lowland forested sites - Wytham and Alice Holt, upland low intensity agricultural - Sourhope, Glensaugh and Moor House, to more extreme montane environments - Snowdon and Cairngorm. Twenty year mean meteorological and air chemistry measurements are provided in Table 1 to illustrate the gradients of climate and deposition covered by the sites. The mean annual temperature of ECN terrestrial sites is inversely related to both altitude and latitude (Monteith et al., 2015) and sites in the lowlands of the south and east are, inevitably, substantially warmer than those in more elevated locations. Sites in the south and east also receive the least precipitation, while Snowdon, in North Wales is by far the wettest of all monitoring locations. The long-term average chemical composition of precipitation varies substantially across sites reflecting their relative proximity to major industrial, domestic and agricultural sources.

2.2 Meteorology

Automatic Weather Stations (AWSs) were established in the vicinity of the ECN Targeted Sampling Site at all ECN sites at the onset of monitoring and operated according to ECN protocols set out by Sykes and Lane (1996). Dry bulb temperature within a non-aspirated screen, wind speed (anemometer) and solar radiation (Kipp solarimeter) were recorded at a 5 or 10 second frequency and logged as mean hourly values. Precipitation was recorded continuously using a tipping bucket rain gauge and summed at an hourly frequency. During the early years of monitoring, several sites also operated co-located manually maintained weather stations that provided measurements at either daily or weekly frequency.

Meteorological data (Rennie et al., 2015b) (air temperature, precipitation, wind speed and solar radiation) from January 1st 1993 to 31st December 2012 were summarised at two levels to meet requirements of different statistical analyses. First, daily averages (or sums with respect to precipitation) of hourly measurements, required for assessment of changes in daily extremes, were computed for all variables. Entries for days on which fewer than 24 hours of data were available were returned as “missing”. Daily data were then summarised as monthly means (or sums with respect to precipitation), with values for months with any missing days of data returned as “missing”.

All ECN AWS instruments were subject to regular (normally annual or bi-annual) calibration checks. However, as an additional quality control step we examined correlations between the monthly means or sums, and comparable monthly estimates derived for the corresponding 1 km^2 from the NERC Centre for Ecology and Hydrology’s CHESS model (Robinson et al., 2015) (temperature, solar radiation and wind speed), or GEAR model (Tanguy et al., 2014) (precipitation). Correlations for all variables were found to be strong and no monthly mean data were rejected on this basis. Recording of precipitation data during autumn to spring seasons at Cairngorm, the highest altitude ECN site, was limited due to frequent freezing up and blocking of the automatic rain gauge so no data were analysed for this site for these seasons. The final monthly datasets, covering the period 1993 to 2012, varied with respect to the number of months of where data were incomplete (temperature = 17%; precipitation = 13%). Where possible, missing monthly precipitation data were replaced by co-located manual measurements. Otherwise, gaps in monthly estimates were filled using modelled estimates based on site-specific linear regression equations for relationships between available monthly ECN measurements and CHESS or GEAR modelled equivalents. Consequently, complete matrices of 20 years of monthly estimates for all variables were compiled for all sites.

2.3 Atmospheric chemistry

Weekly samples of precipitation for chemical analysis were collected using Warren Spring Laboratory standard precipitation collectors situated in the vicinity of the meteorological station of all ECN sites. Precipitation volumes were estimated by weight, while pH and concentrations of major ions were assessed by analytical laboratories linked to the individual sites according to protocols provided by Sykes and Lane (1996). Samples reported to have been affected by bird strikes or other interference were removed from the initial dataset. Types of laboratory analytical accreditation varied across the network but further quality control measures were applied to the data collated in the ECN database (Rennie et al., 2015a), including the removal of clear outliers in linear relationships between: total anions and total cations when expressed in equivalent concentrations; sodium and chloride (since seasalt is assumed to be the dominant source of both ions); and measured conductivity vs theoretical conductivity derived from the sum of ionic strength of individual ions. Any samples with concentrations of phosphorus greater than 1 mg L^{-1} were assumed to be contaminated and also removed. Monthly volume-weighted mean concentrations of individual ions were determined by dividing the total estimated monthly flux of the remaining samples by the sum of their volumes.

Monthly nitrogen dioxide (NO_2) concentrations were measured using diffusion tubes. Initially, NO_2 diffusion tubes were assembled and analysed locally according to the specification provided by Sykes and Lane (1996), but these were replaced at some sites with commercially available tubes manufactured and analysed by Gradko Ltd. in more recent years. A comparison of both methods deployed in parallel at two ECN sites, Wytham and Moor House, in 2007 and 2008 respectively, revealed no consistent difference in estimates (Rose pers. comm). Monthly ammonia (NH_3) concentrations have also been measured at terrestrial ECN sites as a contribution to the National Ammonia Monitoring Network (Tang et al. 2015), although mostly only from around 1998 or later. Ammonia measurements were made using either CEH Delta (DEnuder for Long Term Atmospheric sampling) samplers (Sutton et al., 2001b), or, where a local power source was not available, CEH Alpha (Adapted Low-cost Passive High Absorption) samplers (Sutton et al., 2001a) .

2.4 Data analysis

Visual inspection of both the meteorological and atmospheric chemistry time series demonstrated substantial within year variation and clear signs of seasonality. Prior to the characterisation of long-term trends, therefore, within-year variation in each variable of interest at each site was quantified using a generalised additive model (GAM; (Hastie and Tibshirani, 1990)) with a gamma error distribution and log link. Julian date was treated as a continuous explanatory predictor and cyclic basis functions (Wood, 2006) were used within the GAM to capture the seasonality and ensure that the continuum across the year (i.e. December through to January) was maintained. Residuals from the model, representing zero mean de-trended estimates, were extracted and used in all subsequent analyses.

Having accounted for seasonality, linear trends were fitted to monthly meteorological and chemistry data using generalised linear models (GLMs,(McCullagh and Nelder, 1989)). This modelling framework was used to account for the often skewed distribution of the response variable by applying a Gamma error distribution with a log link function. Autocorrelation was accounted for within the model by the inclusion of a lag-1 autoregressive term. Each variable was fitted with a single explanatory variable representing a running day-of-year value across the whole time period (1993 – 2012). Model coefficients were extracted representing the change in the indicator variable per day, and the *p* value, i.e. the probability that the slope of a linear trend did not differ from zero. Similar models, without the requirement of an autocorrelation term, were fitted to annual mean data taken from monthly observations within specific seasons: winter (December - February); spring (March - May); summer (June - August); and autumn (September - November).

To investigate evidence for non-linear trends over the period, GAMs were used to fit a smoothly varying function to the de-trended time series. In common with the linear models, a running day-of-year metric was used as the sole explanatory variable together with a gamma error distribution and log link function. Autocorrelation in the monthly time series data was similarly accounted for by the inclusion of an autoregressive lag-1 component, AR(1), in the model. Following a similar approach to Monteith et al. (2014) and Large et al. (2013), the gradient of the fitted smooth trend was evaluated at monthly intervals along the whole time series and the associated standard error was obtained to assess whether the gradient was significantly different from zero. This allowed the assignation of all points along the time series to one of three categories – stationary, increasing or decreasing, thus allowing the nature and direction of the trend in each variable to be determined across the whole time series.

In addition, we tested for evidence of changes in extreme values in the daily meteorological data. The threshold against which extreme values were defined was determined using the approach of Northrop and Jonathan (2011) in which quantile regression (Koenker and Bassett, 1978) was applied to fit a linear trend to the upper (99%) or lower (1%) boundaries of the data. We then examined evidence for any changes in the distribution of data points for each variable of interest falling beyond these thresholds. To test for evidence of a significant trend in extreme values, a bootstrap (Efron and Tibshirani, 1994) procedure was used to create 1000 pseudo time series by resampling the original daily series with replacement. Quantile trends were then fitted to each of these artificial datasets to provide a distribution of the regression trend coefficient under the null hypothesis. *P* values, representing the probability of a non-significant trend in the quantile regression, were obtained by comparing the observed trend to the distribution of the 1000 bootstrapped samples. Threshold exceedances beyond this fitted trend were analysed in terms of the number of occurrences within each year, the average exceedance value and the 95% quantile of the exceedance values within each year. Simple linear regression models were fitted to each of these metrics to assess whether there had been any change over the course of the time series. All analyses were performed in the R statistical environment using the mgcv (Wood, 2006) and MASS (Venables and Ripley, 2002) libraries.

Finally we tested for evidence of trends in three potentially ecologically important climatic indices derived at an annual level using a simple linear model. These included the number of frost days (the number of days in the year that minimum air temperature fell below 0 °C), length of growing season (starting, each year, when the temperature on five consecutive days exceeded 5 °C, and ending after five consecutive days of temperatures below 5 °C), and the total solar radiation flux over the growing season (integrated hourly solar radiation flux) – as an indicator of potential net primary productivity. Annual meteorological data were included in the analysis only for those years when more than 300 daily mean measurements were available.

3. Results

3.1 Trends in weather

3.1.1 Linear analysis of meteorological variables

Linear trend statistics for monthly mean air temperature, wind speed, solar radiation and total monthly precipitation for all months and individual seasons respectively are presented in Tables 2a and 2b. The only variable to show widespread linear change in the full monthly time series was mean monthly wind speed. This declined at all sites but one (Porton) for which full data runs were available. Negative trends in mean monthly wind speed at four sites in the south of England were significant at $p<0.001$, while p values for linear trends at several other sites fell marginally above 0.05.

Network-wide patterns of change were more marked with respect to the seasonal data (Table 2b). The most spatially consistent trend was an often highly significant ($p<0.05$) increase in summer precipitation - North Wyke and Hillsborough were the only sites where trends were not significant at this threshold despite positive slopes. The rate of change in summer precipitation (indicated by the trend slope) was strongly correlated with annual precipitation ($r^2 = 0.80$), and the largest increases in summer precipitation were therefore seen at the wettest sites, namely Snowdon (8.2 mm yr^{-1}) and Moor House (5.9 mm yr^{-1}). Unsurprisingly, given the pattern in rainfall, summer solar radiation trends were exclusively negative, but were significant at three sites, all in the south of England, only, i.e. Alice Holt, Porton and Wytham.

Trends in solar radiation in spring were almost all positive, although statistically significant at five sites only - four in the south of England (Alice Holt, Drayton, North Wyke and Rothamsted) in addition to Snowdon. Spring temperatures also showed exclusively positive slopes, but significant change was confined again mostly to southern sites (Alice Holt, Rothamsted, Wytham) in addition to Sourhope.

Trends in wind speed during winter and spring were consistently negative and provided the main explanation for trends in wind speed at an annual scale. Statistically significant negative trends in spring wind speed occurred at four sites in the south of England including Alice Holt, Drayton, Rothamsted and Wytham, in addition to Snowdon, while Drayton, Rothamsted and Wytham also showed statistically significant declines in winter wind speed.

Two sites, one lowland (Hillsborough) and one upland (Sourhope), experienced a significant increase in the length of the growing season (Table 2a) while trends in this metric were positive but not statistically significant at seven of the other nine sites analysed. These, in addition to Porton, were the only sites to show negative slopes in the number of frost days, although no sites showed significant trends in this parameter. No site showed a significant trend in the total solar radiation flux over the growing season.

3.1.2 Non-linear analyses of meteorological variables

The GAM fitting approach applied to the four meteorological variables (full monthly datasets only) provided relatively little indication of non-linear temporal trends. In most cases, variables that showed significant linear trends tended to increase or decrease relatively monotonically throughout the full monitoring period, and the method rarely identified specific sub-periods of significant change within the 20-year records. An exception, however, was a tendency amongst some upland sites, and Hillsborough (Northern Ireland), for gradual increases in mean monthly air temperatures over the first half of the record followed by comparable declines in the second decade. With respect to four of these sites a hump-shaped curve provided a significantly better fit than a horizontal line, i.e. no trend (Figure 1). Fitted splines for temperature for the remaining lowland sites were essentially horizontal and not significant. Otherwise, significant fits for wind speed at Drayton, North Wyke, Rothamsted and Wytham were effectively linear – matching the negative linear trends reported earlier. At the other sites, neither increases nor decreases in wind speed were found to be statistically significant.

3.1.3 Analysis of daily extremes in meteorological variables

Statistically significant trends in daily extremes in meteorological variables were confined largely to precipitation and wind speed. In common with the linear trend directions, these exhibited increases and decreases in extreme values respectively (Supplementary Information: Table 1). Both southern lowland and northern upland locations showed significant increases in the 99th percentile of precipitation, indicating an increase in the amount of precipitation on extremely wet days. The number of unusually wet days, after accounting for the upward trend in extremes (i.e. days when precipitation amounts exceeded the 99th percentile), did not increase over time at any site, although average daily precipitation on extremely wet days increased significantly at Wytham, where there was also a significant increase in the precipitation levels in the most extreme of the extreme events. There were widespread reductions in both 99th percentile and 1st percentile wind speeds at the majority of sites, while the mean of the most extreme windy days also declined at Snowdon, Alice Holt, Rothamsted and Wytham. Other significant trends identified were more site specific and difficult to interpret with respect to wider site characteristics. We observed a significant increase in the 1st percentile of daily air temperatures at three sites, Sourhope, Snowdon and Porton, implying that the coldest days at these sites had become less severe over time. The 99th percentile value for daily air temperatures increased significantly at Snowdon and Rothamsted, but declined significantly at Cairngorm, Glensaugh and Hillsborough.

3.2 Trends in atmospheric pollutants

3.2.1 Linear analysis of atmospheric pollutant variables

The most acidic bulk precipitation over the two decades occurred at Alice Holt, Glensaugh and Rothamsted, where both non-marine SO_4^{2-} and NO_3^- concentrations were relatively high. The least acidic bulk precipitation occurred at Hillsborough, where NO_3^- concentrations were relatively low and NH_4^+ concentrations relatively high. Mean long-term bulk precipitation pH at each site could be explained effectively by a linear model comprising two explanatory variables - the equivalent sum of mean non-marine sulphate (SO_4^{2-}) and nitrate (NO_3^-) concentrations (negative effect), and mean ammonium (NH_4^+) concentration (positive effect); adjusted $r^2 = 0.73$).

Linear modelling indicated that monthly volume-weighted average concentrations of most ions fell significantly at most sites over the two decades (Table 4). The largest reductions were for SO_4^{2-} and non-marine SO_4^{2-} which were invariably highly significant and showed similar slopes, thus implying a dominance of reductions in pollutant SO_4^{2-} over any changes in seasalt SO_4^{2-} deposition. Non-marine SO_4^{2-} trend slopes were most marked at sites in the south and east of the UK where rates of sulphur emissions from power stations and other industrial plant are likely to have been largest historically and hence to have fallen the most.

Rates of reductions in NO_3^- concentration were strongly correlated with those for non-marine SO_4^{2-} (r^2= 0.70), indicating that these signals were dominated by common sources. Rates of change in the former were approximately half of those in the latter and were not statistically significant for the more remote upland sites Snowdon and Glensaugh. Downward slopes in NH_4^+ concentration were significant at all sites other than Snowdon, Sourhope and North Wyke. Unsurprisingly, since the deposition of reduced N tends to be dominated by agricultural sources, trend slopes for NH_4^+ were only weakly correlated with those for non-marine SO_4^{2-} ($r^2 = 0.34$), and better correlated with those for NO_3^- ($r^2 = 0.64$) suggesting that a proportion of the NO_3^- signal was also of agricultural origin.

Indeed, trend slopes in NO_3^- concentration could be explained effectively by a linear model comprising trend slopes for non-marine SO_4^{2-} and NH_4^+ (adjusted $r^2 = 0.80$; both variables significant at $p<0.05$).

Chloride concentration declined significantly at all sites other than Hillsborough and Moor House, although the direction of trend in the latter was still negative. Trend slopes in chloride were closely correlated with trends in sodium, suggesting a common and dominant sea salt source, but correlations with slopes in other base cations were weak.

Nitrogen dioxide concentrations declined significantly at seven sites, five of which also showed significant reductions in NO_3^- concentration in bulk deposition. The largest reductions in both cases were seen at the agricultural stations Rothamsted and Hillsborough. In contrast, gaseous concentrations of NH_3 declined significantly at Drayton only, and increased (significantly) slightly at Moor House and more markedly at Hillsborough.

While both concentrations and fluxes of non-marine SO_4^{2-} declined across the network, reductions in nitrogen species in bulk deposition were confined largely to concentrations. The agricultural stations, Hillsborough (NO_3^- and NH_4^+) and Drayton (NH_4^+) were the only sites to record significant declines in wet nitrogen fluxes.

3.2.2 Non-linear analyses of atmospheric pollutants

Fitted GAM splines for the monthly volume-weighted average concentrations revealed differences in temporal patterns between ions. In common with the results of the linear analysis, the clearest cross-network signals of long term change were seen in total SO_4^{2-} and non-marine SO_4^{2-} concentrations. At most sites, change at all points along the splines was statistically significant, indicating a progressive reduction in anthropogenic SO_4^{2-} deposition throughout the 20 year monitoring record. Downward trends in both variables at most sites were curvilinear with the most rapid reductions occurring in the early years of monitoring, since when the rate of decline in concentration has slowed but remained significant (Figure 2). Trends in these determinands for Snowdon, records for which only began in 2008, were essentially flat.

Patterns of change in NO_3^- concentration differed from those for SO_4^{2-} at most sites. Indeed, change in NO_3^- at some of the sites that are most distant from major fossil fuel burning sources, i.e. Glensaugh, Snowdon and North Wyke, as well as Wytham, was not statistically significant at any point in the record. Most other sites showed relatively linear long term declines. Trends in NH_4^+ concentration were also not significant throughout the records of North Wyke and Sourhope. However, patterns of change mimicked those for SO_4^{2-} at the three agriculturally influenced sites, Drayton, Hillsborough and Rothamsted, with the largest reductions at all three sites again occurring before 2000, since when the rate of change has slowed but remained significant. Ammonium concentrations at all other sites declined significantly, but more gradually and more linearly.

The fitted GAMs indicated progressive and generally linear reductions in both chloride and sodium concentrations at most sites throughout the monitoring period.

4. Discussion

Our analyses show that prevailing environmental conditions changed substantially across the network over the first two decades of ECN monitoring. In general, changes in air chemistry variables were largely in line with expectations, particularly given the international adoption in 1994 of the "Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution on Further Reduction of Sulphur Emissions", in which the UK agreed to reduce emissions of sulphur dioxide by at least 80% by 2010 relative to 1980 levels. Also according to expectations, inter-annual variation in weather was heavily dominated by regional-scale synoptic variation, and provided relatively little evidence of the warming that is apparent over longer timescales than currently represented by the network. Nevertheless, a recent progressive increase in summer precipitation, most notably at wetter upland sites, was particularly striking and had not been anticipated at the time the network was initiated.

4.1 Changes in weather

With the exception of wind speed, which declined significantly across much of the network, there was little evidence for long-term directional change in weather variables when represented as full monthly time series. The non-linear trend analysis revealed a tendency at some northern/upland sites for statistically significant warming over the first decade of monitoring followed by a similar level of cooling in the second, but this behaviour was not detectable at the more southerly/lowland sites. This is broadly consistent with the observation of Morecroft et al. (2009) that upland ECN sites experienced more rapid warming over the first fifteen years of monitoring than lowland sites, and hints at greater sensitivity to regional temperature variability with respect to the sites at higher elevations. Holden and Rose (2011) previously reported a faster rate of warming in recent decades during winter at the upland ECN site Moor House relative to a lowland meteorological station at nearby Durham, and linked this to the suggestion that winter warming might be expected to be more rapid in colder environments as a consequence of more marked variations in a surface albedo influenced by occasional snow and/or ice cover (Pepin and Lundquist, 2008).

Spring temperatures were found to have increased significantly at four sites, three of which are in south-east England. This could have significant ecological implications, e.g. with respect to influencing the phenology of biota (Thackeray et al., 2010), but, at an annual scale, these changes appear to have been balanced by generally negative, although statistically insignificant, trends in winter and summer temperatures. Three mid-latitude sites provided an indication of small increases in the length of the growing season, but no site showed significant trends in total solar radiation flux over the growing season, perhaps as a consequence of the run of wetter and thus cloudier summers.

The absence of evidence for sustained increases in air temperatures over the full ECN record contrasts with the strong positive trends at both annual and seasonal levels identified in the 15 year dataset (Morecroft et al., 2009). We did not perform the composite analysis of trends in upland and lowland site groupings applied in the earlier study, so results are not directly comparable. However a recent sequence of relatively cool years has clearly influenced overall trends. This serves to emphasise the sensitivity of environmental trend assessments to the precise period under investigation.

While there is a common perception that the global temperature rise has "paused" in recent years, the global instrumental record demonstrates that the rate of temperature increase has at most slowed (Mann, 2014), possibly reflecting recent phases in competing multi-decadal oscillations in Atlantic and Pacific surface temperatures (Steinman et al., 2015). However, Karl et al. (2015) asserts that warming over the current century to date is at least as rapid as that during the second half of the previous one. In this context it is important to note that the mean annual temperature of the Central England Temperature Series, (annual variation in which is strongly correlated with mean air temperatures across the ECN), was approximately 0.7 °C higher, over the 1993-2012 ECN monitoring period, than during the commonly used 1961-1990 baseline period. Similar differences are apparent when comparing mean measured 1993-2012 air temperatures for individual sites with their mean CHESS-modelled 1961-1990 estimates. The smoothed spline fitting approach applied to a longer run of air temperatures (1970-2012) modelled for ECN sites using CHESS, indicate that temporal variation in air temperatures over the last 20 years is entirely consistent with a longer-term progressive upward trend at all sites (Figure 3). Comparison of temperature records over the two time scales therefore hints at the likelihood of a forthcoming upward step-change in air temperatures at ECN sites to compensate for recent stationarity. A possible portent is that the year 2014, without the period covered by this analysis, was reported by the UK Met Office to be the warmest year in the UK instrumental record and the warmest year for the Central England Temperature Series that stretches back to 1659 (http://www.metoffice.gov.uk/news/releases/archive/2015/Record-UK-temps-2014); 2014 was also the warmest year globally (Trenberth and Fasullo, 2013).

Variation in seasonal temperatures at ECN sites over the first two decades of monitoring was very closely linked with fluctuations in the NAO. During the winter months, the positive phase of the NAO is associated with vigorous westerly dominated systems bringing relatively warm wet and windy conditions, and the negative phase with blocking highs over the northern Atlantic, and considerably colder, drier, and calmer weather (Hurrell and Van Loon, 1997). Conversely, during summer, a positive NAOI describes a synoptic situation favouring anticyclonic warm and dry weather, while periods with a negative NAOI are characterised by the movement of North Atlantic storm tracks over the UK, with consequent increased precipitation and cooler temperatures (Folland et al., 2009). Winter (December-February) and summer (June-August) mean daily temperatures at ECN sites for the 20-year period were positively correlated with respective winter and summer NAO indices (NAO data source: National Oceanic and Atmospheric Administration (NOAA) Climate Prediction Centre) (Figure 4). Winter temperatures tended to show the stronger Pearson correlation coefficients, ranging from 0.72 for North Wyke to 0.37 for Cairngorm, with particularly tight relationships evident for the southerly lowland ECN sites. Slight negative (although invariably statistically insignificant), winter temperature trend slope coefficients across the network are broadly consistent, therefore, with a weak negative slope in the winter NAOI over the monitoring period.

Correlations between summer temperatures and the summer NAOI were slightly weaker (ranging from 0.64 at Glensaugh to 0.17 at Snowdon) than winter relationships. However, in contrast to the winter index, the summer NAOI showed a strong negative trend over much of the monitoring period. Furthermore, a series of five consecutive summer NAOI values below -0.5 from 2008 onwards is without precedent in the 60 year NAO record analysed, and there is no evidence for a similar prolonged period of negative summer indices in the series when extended back to 1821 by Jones et al. (1997). As a postscript to this analysis, the NAOI for the summers of 2013 and 2015 was also strongly negative although it was positive in 2014. Despite a generally weaker correlation between the summer NAOI and summer temperatures at ECN sites, the more directional nature of the summer NAOI may, therefore, have exerted a negative influence on air temperature trends over the full monitoring period.

Statistically significant linear increases in monthly precipitation across all seasons were identified at three sites only – all in north eastern locations (i.e. Sourhope, Glensaugh and Cairngorm), and slope coefficients were invariably slight. In contrast, summer precipitation increased dramatically across the network and was highly statistically significant at all sites other than North Wyke and Hillsborough, with the greatest absolute increases occurring at the wettest sites.

Again, Figure 4 demonstrates negative relationships between summer precipitation and the summer NAOI with Pearson Correlation coefficients that range from very strong (-0.74 for Snowdon), to weak (-0.27 for Hillsborough) but are generally high (mean = -0.59). Strong correlations between summer precipitation and summer NAOI have been identified previously for periods preceding the shift in the latter to consistently negative values (Folland et al., 2009). The trend in the summer precipitation signal over the ECN monitoring period is therefore clearly linked to the recent, apparently anomalous, uni-directional behaviour of the summer NAOI (Figure 5).

This sequence of wet summers has been observed throughout much of northern Europe and has led to speculation that it may be linked to global-scale climate change. Screen (2013) used a climate simulation approach to demonstrate that the position of the jet stream, that determines the direction of air mass trajectories over northern Europe, can be linked directly to Arctic sea ice extent, and thus explain the recent run of wet conditions. Others argue, however, that recent patterns are governed primarily by the recent positive phase in the Atlantic Decadal Oscillation (Sutton and Dong, 2012), an indicator of North Atlantic Ocean temperatures that shows natural variability at a frequency of 65-70 years (Schlesinger and Ramankutty, 1994) and is itself linked to, and generally precedes, the NAOI with a series of lag times. Currently, therefore, the extent to which recent trends in precipitation may be linked to global climate change as opposed to natural variability cycles remains unclear.

Daily precipitation extremes also increased in intensity at several sites in both upland and lowland locations. Thus the recent increase in summer rainfall is likely to have been influenced by the magnitude of events and not just their frequency. This conforms with reports of an intensification of heavy rainfall events across much of the northern hemisphere which been attributed to anthropogenic increases in GHGs (Min et al., 2011). However, as in our study, most analyses of changes in precipitation extremes to date are based on changes in daily precipitation amounts. High spatial resolution modelling suggests that increases in convective precipitation events, associated with rising temperatures, will have even greater influence on very short term (e.g. hourly), as opposed to daily, extremes (Kendon et al., 2014). Such changes are likely to have major implications, not only with respect to the increased frequency of flash flooding (Ruiz-Villanueva et al., 2012), but also on soil erosion (Martínez-Casasnovas et al., 2002). Other impacts on ecosystems are very difficult to predict, but the co-location of weather stations and ecological measurements at ECN sites makes ECN well placed to begin to assess their importance. It has not been possible in this generic review of trends to include assessment of data at hourly resolution, but further work on more fine scale analyses of ECN meteorological data is in progress.

The most commonly significant linear trends detected at an annual scale were reductions in wind speed. Trends were most apparent during winter and spring, but the analysis of extremes also demonstrates a general tendency for reductions in the wind speeds during both the most windy and the calmest days on record. All sites showed strong positive correlations between mean winter wind speed and the winter NAOI, with the weakest relationship ($r = 0.54$) observed for Alice Holt (Figure 4). Summer wind speeds showed weaker, and generally negative, correlations with the summer NAOI. In addition to likely links with decadal scale variation specific to UK climate, land surface wind speeds have also been reported to have been declining globally over several decades, contributing to reductions in pan evaporation (Roderick et al., 2007), and leading to concerns over long-term consequences for wind power generation (Pryor et al., 2006). Various causes of this long-term decline have been proposed for different parts of the globe, but Vautard et al. (2010) suggest links with both changes to atmospheric circulation patterns and increased surface roughness associated with increased urbanization and forest cover.

We found relatively little evidence for significant linear trends in solar radiation at ECN sites despite changes in air quality that might be expected to have been accompanied by reductions in sulphate aerosol, and thus a reversal of “global dimming”. However, average levels increased significantly at four relatively low rainfall sites in the south of England and Snowdon during spring, when trends at other sites were also consistently positive (with the exception of Hillsborough). In contrast, trends in solar radiation in summer were consistently negative, but were not as marked as the positive trends in precipitation in this season, with only three sites showing statistically significant trends over the full monitoring period.

4.2 Changes in atmospheric chemistry

The concentration of most major ions in bulk precipitation declined significantly at most ECN sites over the 20 year period. The strong and highly significant downward trends in non-marine SO_4^{2-} concentration across the network were expected, given the controls imposed on major sulphur emission sources, i.e. coal and oil burning power stations and other industrial plant in recent decades. UK emissions of sulphur to the atmosphere are estimated to have declined by 93% between 1970 and 2008 (RoTAP, 2012). Large reductions in non-marine SO_4^{2-} concentration across the UK have been reported previously for ECN sites (Morecroft et al., 2009) and trends measured across the UK Acidifying and Eutrophying Atmospheric Pollutants network (UKEAP) have been well documented elsewhere (e.g. RoTAP (2012); Curtis and Simpson (2014)). The regional variation, and the magnitude of change, in non-marine SO_4^{2-} concentration we observed was highly consistent with previous analysis of UKEAP data by Fowler et al. (2005), with a gradient of declining deposition rates from high (south-east England) to low (north west Scotland). Rates of change in non-marine SO_4^{2-} concentration were most marked for the south easterly sites Rothamsted, Alice Holt and Drayton, but large trends were also observed at North Wyke (south-west England) and Hillsborough (Northern Ireland). Surprisingly, the trend at Wytham, also in the south-east, was relatively muted, suggesting local sources may have had an important influence on local air quality here. Wet and dry sulphur deposition have been the primary contributors to the acid load in the UK since the onset of major industrial activity, although relative rates of change in the two vary, with dry deposition (not measured at ECN sites) falling more rapidly close to sources and wet deposition dominating in more remote regions. The curvilinear fits (Figure 2) demonstrated that while reductions in non-marine SO_4^{2-} were sustained throughout the full monitoring period at all sites, change occurred at the greatest rate in the earlier years.

In recent years emission controls are generally reported to have resulted in much stronger effects on deposited sulphur compared to the other major acidifying agent, nitrogen, to a point where nitrogen deposition is beginning to dominate the residual acid load. Nitrogen deposition in the UK is derived from three major sectors: power plants and other industrial fossil fuel combustion sources, motorised transport and agriculture. The former two emit N primarily in oxidised forms (NO_x-N) which are either deposited to the surface dry or as NO_3^- in precipitation after reaction within clouds. UK emissions of NO_x-N are thought to have declined by over 50% between 1970 and 2008, with much of the reduction occurring after 1990 (RoTAP, 2012). The strong correlations we found between rates of change in NO_3^- and non-marine SO_4^{2-} are indicative of the common primary source of this pollutant, although patterns of change were more linear for the former than the latter, with more consistent and more gradual change occurring across the two decades. Concentrations of NO_2 also showed net declines at the majority of sites, although the patterns of change were much more variable between sites, possibly reflecting a greater contribution from local sources which are likely to have varied much more than regional-scale sources.

Rates of change in NO_3^- were also independently correlated, although to a lesser extent, with rates of change in NH_4^+. Agricultural livestock is considered to be the primary source of emissions of ammonia to the atmosphere, which is then either deposited in gaseous form or is transformed to NH_4^+ and deposited in precipitation. The largest reductions in NH_4^+ were seen at agricultural research sites where concentrations have historically been the highest in the network (Hillsborough, Drayton and Rothamsted). When ammonia is sufficiently abundant in the atmosphere to have neutralised any sulphuric acid, it can react with gaseous nitric acid to form ammonium nitrate aerosol (Metzger et al., 2002). This may therefore explain the link between trends in the two N species in wet deposition. In contrast to previous studies that have shown a dominance of S deposition over N deposition, we found that rates of change in NO_3^- and NH_4^+ concentrations combined (when expressed in terms of equivalent acidity) roughly equated to, or exceeded, rates of change in non-marine SO_4^{2-} at the agricultural sites on the network, while at the other sites the latter usually dominated.

Strong correlations between rates of change in chloride and sodium were indicative of a dominant influence on the concentrations of these ions from sea salt. Reductions over the 20 years are therefore consistent with significant declines in winter and spring wind speeds, since sea salt aerosol dispersion over land is highly dependent on wind strength and a reduction in the winter NAOI (Evans et al., 2001). However, it should also be noted that rates of change in chloride concentration at the majority of sites were disproportionally higher than rates of change in sodium (equivalent ratio of Na:Cl in sea salt = 0.86), indicating reduction in supply from another source of chloride. In an assessment of ECN, UKEAP and Upland Waters Monitoring Network data, Evans et al. (2011) demonstrated that the UK had experienced a significant reduction in hydrochloric acid (HCl) deposition in recent years. A product of the combustion of coal with a high chlorine content, it would appear that this decline in HCl has made a significant contribution to the overall reduction in the acid load.

In common with the recent assessment of the UKEAP data by Curtis and Simpson (2014), linear analysis indicated significant reductions in hydrogen ion concentration at most sites, thus illustrating the impact of the reduction in S and N species on the acid load. Alice Holt, Drayton, Moor House, Snowdon and Wytham all showed clear curvilinear declines that mimicked reductions in non-marine SO_4^{2-}. Clear increases in soil water pH (particularly upland soils with lower buffering potential) have been observed at several ECN sites over the full monitoring period (Supplementary Information; Figure 1) which would appear to be directly linked to reductions in acid deposition over the period. Only the least acidic and most manipulated soils at the agricultural sites Rothamsted, Drayton, Hillsborough and Wytham showed no clear indication of a pH increase in either shallow or deep soil samplers. Soil pH exerts a tight control on a variety of biogeochemical processes and has been found to strongly influence the structure of soil microbial assemblages (Griffiths et al., 2011). Given the ubiquity of soil pH changes and the paucity of evidence for marked climatic trends over the last 20 years it would therefore seem likely that changes to S and N deposition will have dominated regional scale changes in the ecological characteristics of much of the UK semi-natural landscape.

5. **Conclusion**

The primary purpose of this study, and the continuing meteorological and atmospheric chemical monitoring at terrestrial ECN sites, is to inform assessments of biogeochemical and biological change across the network. The first twenty years of monitoring has revealed significant directional changes in the ambient environment. The most notable of these have been substantial reductions in acid deposition, which is leading to progressive increases in the pH of acid-sensitive soil types, and marked increases in the amount of precipitation during summer and the amount of precipitation on very wet days. There has also been a tendency for wind speeds during winter and spring to decrease. The main changes identified in weather are likely to have been dominated by regional scale variability rather than global climate change *per se*. However, it remains to be seen whether the apparently unprecedented recent series of exceptionally wet summers and associated negative deviation in the North Atlantic Oscillation simply represents the upward limb of a multi-decadal scale cycle as opposed to a more directional shift in climate. While the environments of each ECN site are in some respects unique, similarities in patterns of change between sites are sufficiently strong for us to assume that similar trends are occurring nationally. However, data from other more-issue focussed networks are clearly vital to provide the necessary spatial context for these observations.

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Table 1. Summary of mean weather and air quality parameters for ECN sites for the period of up to the end of 2012. Mean nitrogen dioxide (NO_2) concentrations provided for the period 1995 – 2012 at most sites (Cairngorm record covers 2000 – 2012). Mean ammonia (NH_3) concentrations provided for the period 1998 – 2012 at most sites (Alice Holt record covers 2000 – 2012); Snowdon record covers 2009 – 2012). NA = insufficient data available to determine long-term mean.

Metric	Alice Holt	Cairngorm	Drayton	Glensaugh	Hillsborough	Moor House	North Wyke	Porton	Rothamsted	Snowdon	Sourhope	Wytham
First full year of ECN monitoring	1993	1999	1993	1993	1993	1993	1993	1994	1993	1995	1993	1993
Meteorological variables												
annual precipitation (mm)	854	NA	657	1121	1172	2106	1058	859	708	3623	1002	740
mean monthly temperature (°C)	10.7	5.0	10.2	7.4	9.3	5.9	10.0	10.0	10.1	7.4	7.3	9.9
mean monthly wind speed ($m\ s^{-1}$)	1.1	7.7	1.5	3.0	2.2	4.2	2.8	3.0	2.4	3.9	5.1	2.3
mean monthly solar radiation ($W\ m^{-2}$)	191	117	194	163	152	164	192	200	203	161	168	204
Bulk deposition variables												
pH	4.7	NA	5.1	4.6	5.2	5.0	5.0	NA	4.7	5.0	4.8	5.0
SO_4^{2-} ($\mu eq\ L^{-1}$)	42	NA	43	43	43	26	33	NA	57	25	34	34
non-marine SO_4^{2-} ($\mu eq\ L^{-1}$)	33	NA	36	30	32	19	17	NA	50	16	24	28
NO_3^- ($\mu eq\ L^{-1}$)	36	NA	34	35	24	17	24	NA	43	12	26	26
NH_4^+ ($\mu eq\ L^{-1}$)	32	NA	44	34	47	21	27	NA	51	14	24	34
Cl^- ($\mu eq\ L^{-1}$)	72	NA	60	108	124	57	151	NA	76	85	91	52
Na^+ ($\mu eq\ L^{-1}$)	67	NA	59	105	90	55	130	NA	63	72	89	44
Mg^{2+} ($\mu eq\ L^{-1}$)	16	NA	15	24	25	12	29	NA	10	17	20	10
Ca^{2+} ($\mu eq\ L^{-1}$)	33	NA	33	16	36	11	27	NA	23	7	15	23
K^+ ($\mu eq\ L^{-1}$)	4	NA	5	6	5	2	18	NA	6	2	5	2
Gaseous concentration variables												
NO_2 (ppb)	4.5	1.0	5.6	0.6	3.0	2.4	3.2	4.9	13.7	1.7	0.7	6.9
NH_3 (ppb)	0.6	0.1	2.0	0.3	4.1	0.4	1.7	2.3	1.5	0.3	0.3	1.1

Table 2a.

Linear trends with time (change per year) in monthly meteorological variables (all seasons included) and annual climate indices recorded at UK ECN sites over the period 1993-2012. Trends significant at p<0.05 highlighted by grey shading. The criterion for analysis of trends in climate indices was at least 15 years where a minimum of 300 days of meteorological data were available.

NA = data not available.

	Temperature (deg C yr^{-1})		**Precipitation (mm yr^{-1})**		**Solar radiation (W m^{-2} yr^{-1})**		**Wind speed (m s^{-1} yr^{-1})**		**No. frost days (yr^{-1})**		**Length of growing season (days yr^{-1})**		**Cumulative solar radiation flux**	
Site	*Slope*	*p-value*	*Slope*	*p-value*	*Slope*	*p-value*	*Slope*	*p-value*	*Slope*	*p-value*	*Slope*	*p-value*	*Slope*	*p-value*
Alice Holt	0.012	0.497	0.584	0.250	0.110	0.735	-0.005	0.057	0.302	0.308	0.602	0.750	-0.609	0.306
Cairngorms	-0.012	0.538	NA	NA	0.950	0.017	-0.055	0.059	1.208	0.392	-2.369	0.289	0.389	0.761
Drayton	-0.007	0.685	0.329	0.351	0.073	0.723	-0.026	<0.001	0.876	0.208	1.050	0.468	-0.057	0.916
Glensaugh	0.026	0.310	1.680	0.024	0.073	0.808	-0.045	0.006	0.436	0.327	1.482	0.245	-0.337	0.607
Hillsborough	-0.001	0.956	0.584	0.537	-0.694	0.430	-0.008	0.067	-0.451	0.318	2.603	0.044	-0.607	0.559
Moorhouse	0.002	0.911	0.183	0.872	-0.219	0.528	-0.020	0.102	1.061	0.115	1.088	0.439	-3.025	0.048
North Wyke	-0.012	0.492	-0.256	0.699	0.183	0.415	-0.018	<0.001	0.438	0.278	1.112	0.459	0.632	0.304
Porton	0.001	0.969	-0.183	0.711	-0.438	0.134	0.009	0.164	-0.079	0.834	1.713	0.253	-0.276	0.787
Rothamsted	0.016	0.390	0.146	0.717	1.278	0.001	-0.020	<0.001	0.115	0.708	-0.418	0.676	0.915	0.051
Snowdon	0.042	0.040	1.424	0.470	0.402	0.256	-0.013	0.154	NA	NA	NA	NA	NA	NA
Sourhope	0.027	0.141	0.950	0.091	0.475	0.128	NA	NA	-0.658	0.277	2.242	0.019	0.533	0.213
Wytham	0.009	0.640	0.329	0.399	-0.365	0.208	-0.014	<0.001	0.109	0.722	0.344	0.765	0.189	0.660

Table 2b. Linear trends with time (change per year) in monthly meteorological variables summarised for individual seasons recorded at UK ECN sites over the period 1993-2012. Trends significant at p<0.05 highlighted by grey shading.

	Temperature (deg C)		Precipitation (mm)		Solar radiation (W m^{-2})		Wind speed (m s^{-1})	
	Slope coeff.	*p-value*	*Slope coeff.*	*p-value*	*Slope coeff.*	*p-value*	*Slope coeff.*	*p-value*
SPRING								
Alice Holt	0.050	0.049	0.505	0.719	1.248	0.041	-0.007	0.045
Cairngorms	0.017	0.555	NA	NA	2.587	0.172	-0.106	0.166
Drayton	0.030	0.229	-0.039	0.958	1.209	0.012	-0.030	0.001
Glensaugh	0.030	0.328	-0.062	0.972	2.200	0.087	NA	NA
Hillsborough	0.034	0.107	-0.240	0.842	-1.995	0.723	-0.005	0.569
Moorhouse	0.035	0.216	-2.953	0.036	0.715	0.335	-0.033	0.235
North Wyke	0.011	0.644	-0.745	0.455	1.584	0.027	-0.017	0.056
Porton	0.043	0.107	-0.073	0.932	0.785	0.209	-0.003	0.848
Rothamsted	0.056	0.021	-0.121	0.914	2.428	0.029	-0.025	0.029
Snowdon	0.059	0.099	-4.893	0.157	2.154	0.001	-0.032	0.026
Sourhope	0.071	0.041	0.082	0.929	2.337	0.064	NA	NA
Wytham	0.054	0.006	0.111	0.880	0.755	0.152	-0.024	0.009
SUMMER								
Alice Holt	-0.009	0.768	2.461	0.006	-1.500	0.038	0.006	0.237
Cairngorms	-0.032	0.435	6.024	<0.001	0.347	0.843	-0.039	0.481
Drayton	-0.042	0.113	2.424	0.005	-1.183	0.115	-0.022	0.011
Glensaugh	-0.043	0.294	5.598	0.000	-1.024	0.515	NA	NA
Hillsborough	0.004	0.908	2.413	0.579	-4.378	0.551	0.001	0.739
Moorhouse	-0.002	0.971	5.864	0.001	-1.483	0.075	-0.016	0.262
North Wyke	-0.020	0.496	2.467	0.152	-1.195	0.187	0.001	0.898
Porton	-0.041	0.172	2.394	0.027	-2.502	0.016	0.033	0.004
Rothamsted	-0.013	0.691	1.889	0.036	0.368	0.783	-0.001	0.893
Snowdon	0.010	0.781	8.246	0.049	-1.315	0.283	-0.002	0.845
Sourhope	0.000	0.989	3.579	0.006	-0.917	0.365	NA	NA
Wytham	-0.034	0.378	2.894	0.001	-2.158	0.041	0.002	0.628
AUTUMN								
Alice Holt	0.037	0.128	-0.282	0.734	0.376	0.412	0.000	0.934
Cairngorms	-0.007	0.833	NA	NA	0.984	0.277	0.073	0.028
Drayton	0.016	0.534	-0.599	0.337	0.268	0.571	-0.017	0.039
Glensaugh	-0.014	0.535	0.718	0.677	0.456	0.331	NA	NA
Hillsborough	0.000	1.000	1.233	0.457	-1.286	0.650	-0.004	0.525
Moorhouse	0.013	0.755	2.360	0.189	-0.571	0.334	0.019	0.418
North Wyke	0.014	0.575	-0.494	0.597	0.467	0.235	-0.015	0.146
Porton	0.034	0.169	-1.782	0.017	0.293	0.628	0.024	0.004
Rothamsted	0.044	0.126	-0.444	0.535	1.416	0.042	-0.011	0.193
Snowdon	0.079	0.014	4.747	0.386	0.304	0.583	0.020	0.357
Sourhope	0.032	0.236	0.752	0.357	0.686	0.088	NA	NA
Wytham	0.035	0.213	-1.049	0.116	-0.008	0.988	-0.001	0.943
WINTER								
Alice Holt	-0.020	0.668	0.786	0.524	-0.123	0.697	-0.014	0.084
Cairngorms	-0.037	0.495	NA	NA	-0.355	0.610	-0.159	0.038
Drayton	-0.037	0.416	0.160	0.820	-0.506	0.139	-0.038	<0.001
Glensaugh	-0.039	0.361	2.366	0.257	-1.388	0.071	NA	NA
Hillsborough	-0.043	0.341	0.031	0.986	-2.300	0.213	-0.027	0.056
Moorhouse	-0.035	0.536	-2.915	0.399	0.053	0.909	-0.053	0.065
North Wyke	-0.036	0.506	1.482	0.657	-0.561	0.056	-0.029	0.123
Porton	-0.025	0.646	-0.195	0.859	-0.861	0.172	-0.005	0.811
Rothamsted	-0.019	0.697	0.158	0.874	0.145	0.725	-0.038	0.003
Snowdon	0.025	0.678	1.385	0.768	-0.047	0.921	-0.032	0.255
Sourhope	-0.004	0.921	0.410	0.694	-0.935	0.040	NA	NA
Wytham	-0.012	0.809	0.386	0.732	-0.619	0.270	-0.033	0.002

Table 3. Linear trends with time in monthly volume weighted average ionic concentrations (μeq L^{-1} yr^{-1}) and fluxes (μeq m^{-2} yr^{-1}), and trends in mean monthly NO_2 concentrations and mean monthly NH_3 concentrations recorded at UK ECN sites over the period 1993-2012 for which at least 15 year runs of data are available. Insufficient time series available for Cairngorm and Porton. Values presented are linear slope coefficients. Those shaded grey significant at $p<0.05$; those in bold significant at $p<0.01$).

	Ca^{2+}	**Cl^-**	**H^+**	**K^+**	**Mg^{2+}**	**Na^+**	**NH_4^+**	**NO_3^-**	**SO_4^{2-}**	**xSO_4^{2-}**	**NO_2**	**NH_3**
Site	**Annual rate of change in volume weighted concentration (μeq L^{-1} yr^{-1})**										**ppb yr^{-1}**	**μg m^{-3} yr^{-1}**
Alice Holt	**-1.44**	-1.52	**-0.94**	**-0.17**	-0.28	-1.01	**-0.89**	**-0.83**	**-2.23**	**-2.07**	**-0.077**	0.004
Drayton	**-0.79**	-1.11	**-0.64**	**-0.18**	-0.17	-0.53	**-3.17**	**-1.64**	**-2.34**	**-2.25**	**-0.109**	**-0.093**
Glensaugh	-0.16	**-3.09**	-0.19	**-0.50**	**-0.52**	-1.89	**-0.80**	-0.29	**-1.96**	**-1.73**	**-0.012**	-0.002
Hillsborough	-0.85	-0.03	**-0.35**	**-0.35**	-0.64	-0.49	**-3.65**	**-1.04**	**-2.30**	**-2.21**	-0.023	**0.115**
Moor House	**-0.49**	-0.77	**-0.69**	**-0.08**	**-0.25**	**-1.16**	**-0.55**	**-0.31**	**-1.18**	**-1.02**	**-0.054**	**0.020**
North Wyke	**-2.15**	-3.57	-0.02	**-1.63**	-0.79	-1.99	-0.88	-0.76	**-2.36**	**-2.08**	**-0.069**	0.002
Rothamsted	**-1.27**	**-3.63**	**-1.47**	**-0.49**	-0.12	**-2.55**	**-2.05**	**-1.48**	**-4.01**	**-3.65**	**-0.441**	0.112
Snowdon	<0.04	**-2.35**	**-0.62**	<0.03	-0.37	-1.75	-0.20	-0.14	**-0.75**	**-0.53**	0.022	-0.006
Sourhope	**-0.52**	**-2.70**	**-0.57**	**-0.41**	-0.545	-1.41	-0.31	**-0.58**	**-1.77**	**-1.58**	**-0.016**	0.041
Wytham	**-1.08**	-1.13	**-0.81**	-0.03	**-0.32**	**-1.14**	**-0.76**	-0.40	**-1.27**	**-1.13**	-0.062	0.004
	Annual rate of change in bulk precipitation flux (μeq m^{-2} yr^{-1})											
Alice Holt	**-75.7**	-85.3	-60.8	-8.9	-14.7	-53.6	-19.3	-0.7	**-140.9**	**-133.6**		
Drayton	-4.0	3.4	**-25.2**	-7.3	9.1	43.4	**-113.5**	-24.5	**-84.6**	**-84.3**		
Glensaugh	0.2	-424.8	55.0	**-58.3**	-55.1	-189.8	5.5	66.8	**-177.5**	**-154.4**		
Hillsborough	**-89.5**	-263.4	13.9	**-33.2**	**-125.9**	-228.9	**-469.5**	**-113.8**	**-267.8**	**-224.9**		
Moor House	**-67.2**	-73.8	**-112.9**	-14.5	-34.5	-158.5	-29.1	-12.9	**-147.8**	**-122.7**		
North Wyke	**-0.2**	-0.3	0.0	**-0.2**	-0.1	-0.2	-0.1	-0.1	**-0.2**	**-0.2**		
Rothamsted	**-60.4**	**-203.0**	**-100.7**	**-28.7**	0.9	**-135.9**	-46.8	-30.8	**-215.4**	**-197.1**		
Snowdon	-33.9	**-1576.2**	**-230.3**	2.9	-263.5	**-1217.8**	-99.7	-85.2	**-332.7**	**-185.3**		
Sourhope	**-25.4**	-229.2	-58.2	**-40.2**	-31.5	-85.3	-9.7	-38.4	**-170.9**	**-159.8**		
Wytham	**-52.2**	-63.3	**-62.1**	2.0	-18.7	-62.6	-2.3	10.3	-46.0	-38.2		

Figure 1. Statistically significant curvilinear generalised additive model (GAM) fits for ECN meteorological variables.

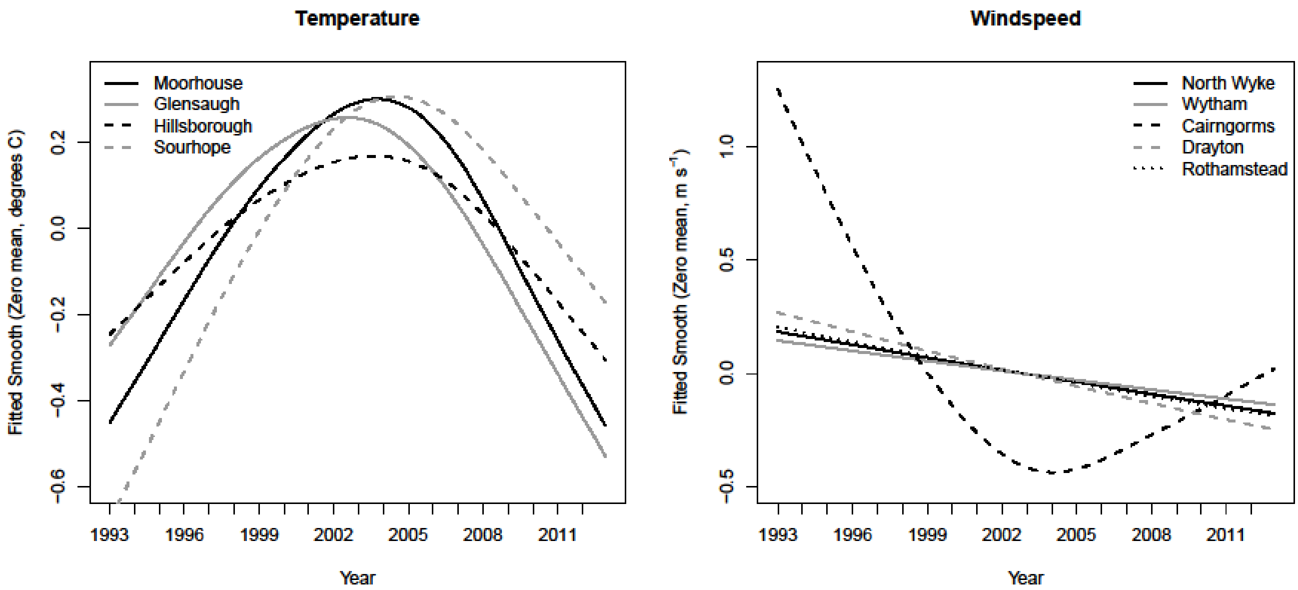

Figure 2. Statistically significant curvilinear generalised additive model (GAM) fits for ECN atmospheric chemistry variables: including volume weighted concentrations of nitrate-N (NO_3), sulphate (SO_4) , non-marine sulphate (xSO_4), hydrogen ion and ammonium in bulk collectors, and nitrogen dioxide concentrations measured by diffusion tube. Non-significant fits not included.

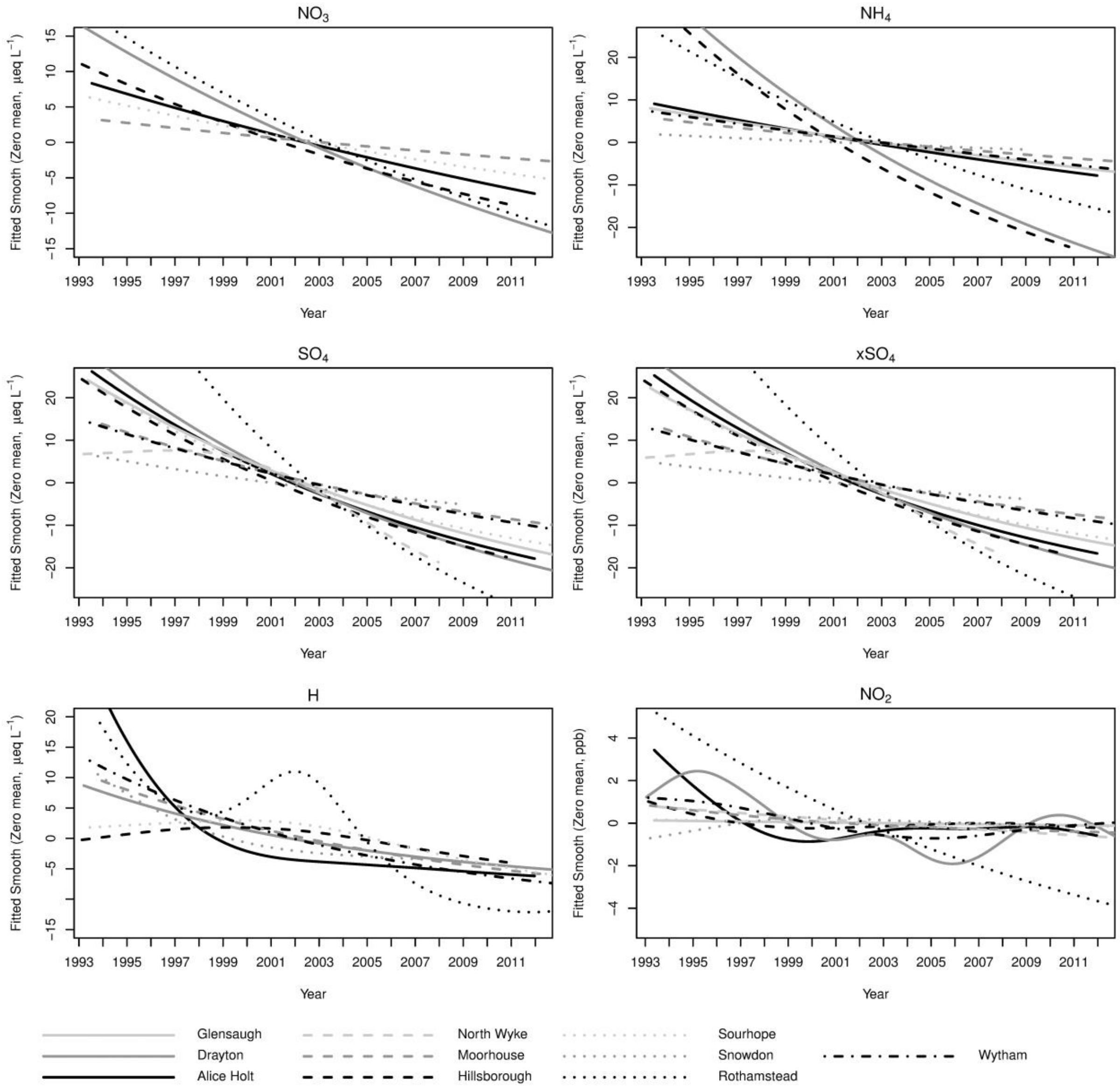

Figure 3. Curvilinear Generalised Additive Model (GAM) fits for modelled (CHESS) monthly air temperature data for three ECN sites, (a) Rothamsted, (b) Moor House and (c) Glensaugh (1970-2012). Data represent variation (in °C) relative to a mean of zero °C. In each case the trend is significant throughout these records.

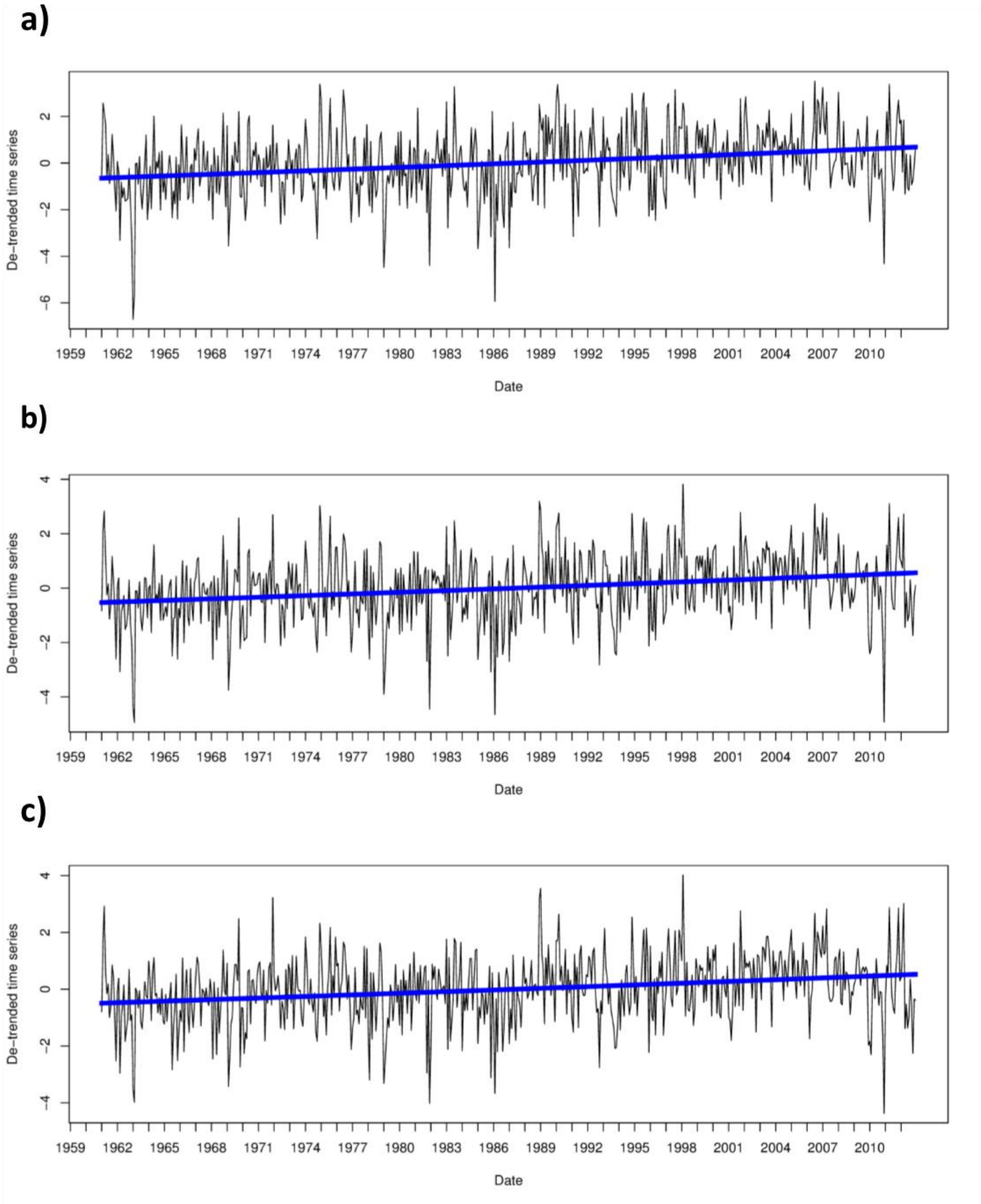

Figure 4 Pearson correlation coefficients (r) for relationships between annual winter and summer North Atlantic Oscillation Indices and respective seasonal precipitation, air temperature, wind speed and solar radiation variables for the twelve ECN sites. NAO Data source: NOAA Climate Prediction Service.

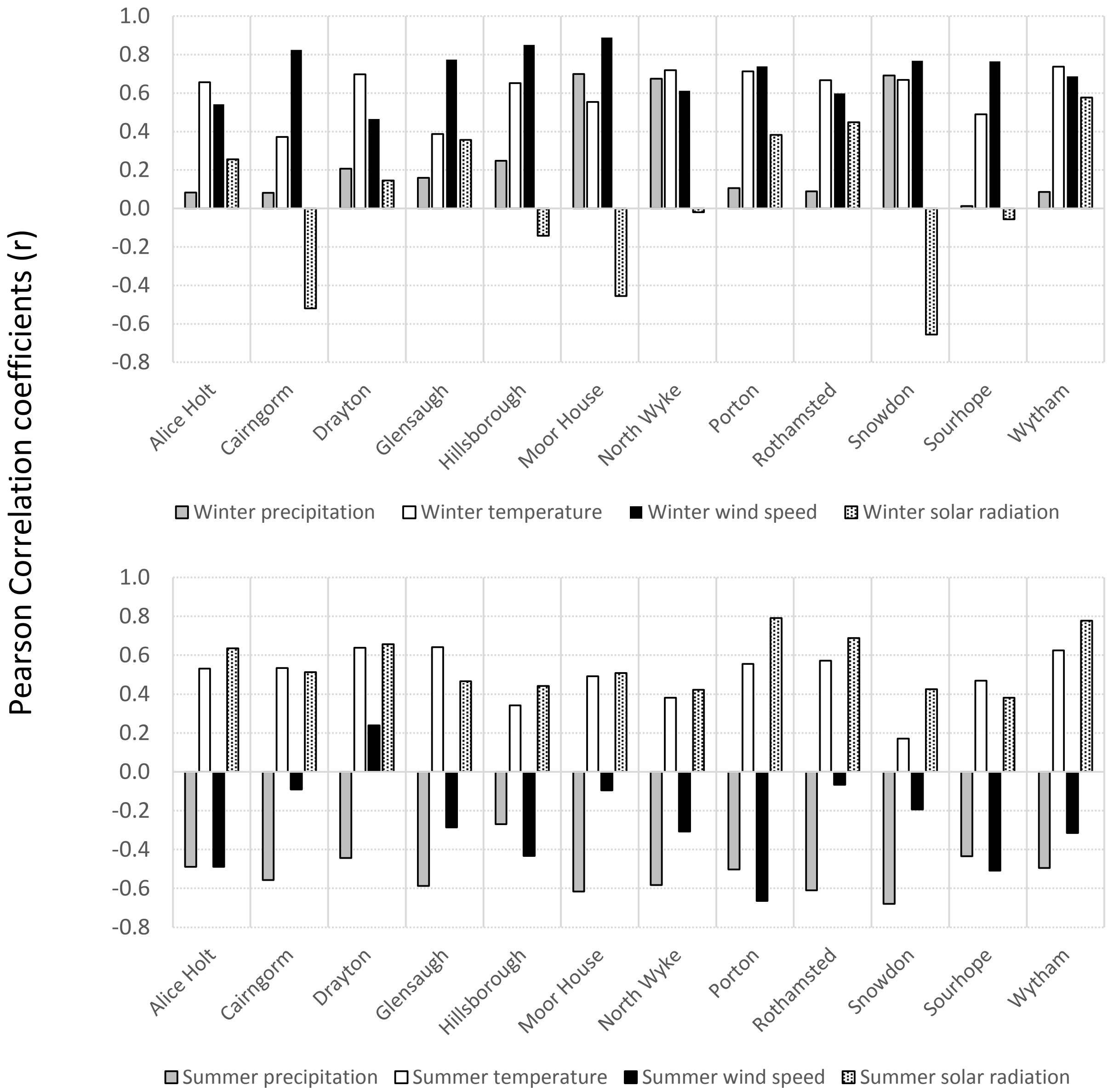

Figure 5 Trend in mean winter (December to February) and summer (June to August) North Atlantic Oscillation indices from 1950 to 2012. NAO Data source: NOAA Climate Prediction Service. ECN monitoring period indicated by shaded box.