

Article (refereed) - postprint

Monteith, Don; Henrys, Peter; Banin, Lindsay; Smith, Ron; Morecroft, Mike; Scott, Tony; Andrews, Chris; Beaumont, Deborah; Benham, Sue; Bowmaker, Victoria; Corbett, Stuart; Dick, Jan; Dodd, Bev; Dodd, Nicki; McKenna, Colm; McMillan, Simon; Pallett, Denise; Pereira, M. Gloria; Poskitt, Jan; Rennie, Sue; Rose, Rob; Schafer, Stefanie; Sherrin, Lorna; Tang, Sim; Turner, Alex; Watson, Helen. 2016. **Trends and variability in weather and atmospheric deposition at UK Environmental Change Network sites (1993–2012)** [in special issue: Assessing ecosystem resilience through long term ecosystem research: observations from the first twenty years of the UK Environmental Change Network]

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

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29 **Keywords:**

30 **ECN, trends, climate change, weather, extreme events, air pollution, acid deposition.**

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32 **Published online in Ecological Indicators – 29-2-2016**

33 <http://dx.doi.org/10.1016/j.ecolind.2016.01.061>

34

36 **Abstract**

37 We characterised temporal trends and variability in key indicators of climate and atmospheric
38 deposition chemistry at the twelve terrestrial UK Environmental Change Network (ECN) sites over the
39 first two decades of ECN monitoring (1993 – 2012) using various statistical approaches. Mean air
40 temperatures for the monitoring period were approximately 0.7 °C higher than those modelled for
41 1961-1990, but there was little evidence for significant change in air temperature over either the full
42 monthly records or within individual seasons. Several upland ECN sites, however, warmed significantly
43 over the first decade before cooling in the second. Summers at most sites became progressively
44 wetter, and extremes in daily rainfall increased in magnitude. Average wind speeds in winter and
45 spring declined at the majority of sites. Directional trends in summer precipitation could be linked to
46 an atypically prolonged negative deviation in the summer North Atlantic Oscillation (NAO) Index.
47 Several aspects of air quality improved markedly. Concentrations and fluxes of sulphate in
48 precipitation declined significantly and substantially across the network, particularly during the earlier
49 years and at the most polluted sites in the south and east. Precipitation concentrations of nitrate and
50 ammonium, and atmospheric concentrations of nitrogen dioxide also decreased at most sites. There
51 was less evidence for reductions in the loads of wet deposited nitrogen species, while trends in
52 atmospheric ammonia concentration varied in direction and strength between sites. Reductions in
53 acid deposition are likely to account for widespread gradual increases in the pH of soil water at ECN
54 sites, representing partial recovery from acidification. Overall, therefore, ECN sites have experienced
55 marked changes in atmospheric chemistry and weather regimes over the last two decades that might
56 be expected to have exerted detectable effects on ecosystem structure and function. While the
57 downward trend in acid deposition is unlikely to be reversed, it is too early to conclude whether the
58 trend towards wetter summers simply represents a phase in a multi-decadal cycle, or is indicative of
59 a more directional shift in climate. Conversely, the first two decades of ECN now provide a relatively
60 stable long term baseline with respect to air temperature, against which effects of anticipated future
61 warming on these ecosystems should be able to be assessed robustly.

62

63

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

79 ECN monitoring has been conducted over a period when changes in regional-scale environmental
80 drivers might have been expected to be substantial. Global carbon emissions increased at a rate of
81 $1.0\% \text{ yr}^{-1}$ in the 1990s and $3.1\% \text{ yr}^{-1}$ since 2000, while atmospheric concentrations of carbon dioxide,
82 the principal greenhouse gas (GHG), measured at the Mauna Loa Observatory, increased from 357 to
83 394 ppm between 1993 and 2012 (NOAA-ESRL data). This has been accompanied by a considerable
84 and progressive increase in the heat content of the world’s oceans (Abraham et al., 2013) and global
85 surface temperatures (Hansen et al., 2010). Sea levels have risen by a global average of approximately
86 6 cm (Blunden and Arndt, 2013), while summer minimum Arctic Sea Ice area extent has contracted by
87 between 9.4 to $13.6\% \text{ decade}^{-1}$ over the past three decades (Swart et al., 2015).

88 Global anthropogenically-driven warming is predicted to affect the climate of the UK in the long term
89 through increased air temperatures, changes in the amounts and seasonal distribution of
90 precipitation, and increases in the frequency of extreme climatic events including floods and droughts
91 (Jenkins et al., 2009). These trends will be mediated at more local scales by relatively short-term
92 variation in ocean temperatures and the position, and pressure gradients, of the earth’s teleconnected
93 regional atmospheric circulation systems. Inter-annual variability in UK weather is particularly well
94 summarised by the North Atlantic Oscillation Index (NAOI) (Hurrell and Van Loon, 1997) – the
95 standardised difference in sea level atmospheric pressure between fixed points in the Azores and
96 Iceland. The NAO tends to vary at an approximately decadal frequency and has, in turn, been argued
97 to be sensitive to the extra-terrestrial influence of subtle variation in solar activity (Brown and John,
98 1979; Lockwood et al., 2010; Scaife et al., 2013).

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

110 Finally, air pollution, particularly in the form of sulphate (SO₄²⁻) aerosol, can itself have a marked effect
111 on climate, both directly, by reflecting short-wave radiation, and by providing condensation nuclei for
112 cloud formation, in a process known as “solar dimming” (Stanhill and Cohen, 2001). Gedney et al.
113 (2014) reported that river flows in some of the most polluted regions of northern Europe were up to
114 25% higher than normal when aerosol levels peaked around 1980, and attributed this to reduced
115 evaporative loss. The authors proposed that hydrological trends might be reversed with more recent
116 “global brightening”. Changes in climate can also influence fluxes and concentrations of atmospheric
117 pollutants to the land surface. Ambient pollution levels are heavily dependent on prevailing air mass
118 trajectories (Fleming et al., 2012), while rainfall events can increase both fluxes and concentrations of
119 pollutants, particularly in upland environments through the feeder-seeder effect (Inglis et al., 1995).

120 The ambient environment of semi-natural systems across the UK, including those represented by ECN,
121 may therefore be expected to have undergone significant shifts in both climate- and pollution-related
122 ecological stressors over the past two decades. Quantification and characterisation of these changes
123 are essential prerequisites for the appropriate attribution of single and interactive effects on soils,
124 surface waters, species and ecosystems. The first broad assessment of trends in physical and
125 biogeochemical drivers of environmental change at ECN sites was conducted by Morecroft et al.
126 (2009), who also reported on trends in ecological indicators of change. Statistical analyses were largely
127 confined to tests of linear change in variables summarised at an annual scale. The study period (1993-
128 2008) was characterised by marked increases in air temperature across most seasons (amounting to
129 circa 1 °C decade⁻¹), in addition to large reductions in concentrations of SO₄²⁻ and acidity in
130 precipitation. However, Swart et al. (2015), in their assessment of recent trends in Arctic sea ice,
131 emphasise that climate change is unlikely to be uniform, and characterisation of trends using linear
132 methods alone is thus vulnerable to the specific period chosen for analysis. A succession of relatively
133 cool years since 2008 (e.g. (Cattiaux et al., 2010)) and evidence for a recent stabilisation of atmospheric
134 pollutant deposition rates (Curtis and Simpson, 2014), render a simple repeat of previously applied
135 linear analysis of restricted value, while acquisition of a further five years data has increased options
136 to characterise trends using non-linear approaches.

137 In the following assessment, therefore, we apply both linear and non-linear statistical analyses of key
138 meteorological and air pollutant variables at the 12 terrestrial ECN sites covering the period 1993-
139 2012 with the aim of: 1) quantifying net change over the full period, 2) characterising temporal
140 variation in the rate and statistical significance of change, and 3) testing for evidence of changing
141 frequency of extreme meteorological events. We also refer to modelled meteorological data to
142 provide longer term context for the changes recorded over the past two decades at each site.

143

144

145 **2. Methods**

146 2.1 Sites

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

160 2.2 Meteorology

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

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

176 All ECN AWS instruments were subject to regular (normally annual or bi-annual) calibration checks.
177 However, as an additional quality control step we examined correlations between the monthly means
178 or sums, and comparable monthly estimates derived for the corresponding 1 km² from the NERC
179 Centre for Ecology and Hydrology’s CHESS model (Robinson et al., 2015) (temperature, solar radiation
180 and wind speed), or GEAR model (Tanguy et al., 2014) (precipitation). Correlations for all variables
181 were found to be strong and no monthly mean data were rejected on this basis. Recording of
182 precipitation data during autumn to spring seasons at Cairngorm, the highest altitude ECN site, was
183 limited due to frequent freezing up and blocking of the automatic rain gauge so no data were analysed
184 for this site for these seasons. The final monthly datasets, covering the period 1993 to 2012, varied
185 with respect to the number of months of where data were incomplete (temperature = 17%;
186 precipitation = 13%). Where possible, missing monthly precipitation data were replaced by co-located
187 manual measurements. Otherwise, gaps in monthly estimates were filled using modelled estimates
188 based on site-specific linear regression equations for relationships between available monthly ECN

189 measurements and CHESS or GEAR modelled equivalents. Consequently, complete matrices of 20
190 years of monthly estimates for all variables were compiled for all sites.

191 2.3 Atmospheric chemistry

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

206 Monthly nitrogen dioxide (NO₂) concentrations were measured using diffusion tubes. Initially, NO₂
207 diffusion tubes were assembled and analysed locally according to the specification provided by Sykes
208 and Lane (1996), but these were replaced at some sites with commercially available tubes
209 manufactured and analysed by Gradko Ltd. in more recent years. A comparison of both methods
210 deployed in parallel at two ECN sites, Wytham and Moor House, in 2007 and 2008 respectively,
211 revealed no consistent difference in estimates (Rose pers. comm). Monthly ammonia (NH₃)
212 concentrations have also been measured at terrestrial ECN sites as a contribution to the National
213 Ammonia Monitoring Network (Tang et al. 2015), although mostly only from around 1998 or later.
214 Ammonia measurements were made using either CEH Delta (DENuder for Long Term Atmospheric
215 sampling) samplers (Sutton et al., 2001b), or, where a local power source was not available, CEH Alpha
216 (Adapted Low-cost Passive High Absorption) samplers (Sutton et al., 2001a) .

217 2.4 Data analysis

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

227 Having accounted for seasonality, linear trends were fitted to monthly meteorological and chemistry
228 data using generalised linear models (GLMs,(McCullagh and Nelder, 1989)). This modelling framework
229 was used to account for the often skewed distribution of the response variable by applying a Gamma
230 error distribution with a log link function. Autocorrelation was accounted for within the model by the
231 inclusion of a lag-1 autoregressive term. Each variable was fitted with a single explanatory variable
232 representing a running day-of-year value across the whole time period (1993 – 2012). Model

233 coefficients were extracted representing the change in the indicator variable per day, and the p value,
234 i.e. the probability that the slope of a linear trend did not differ from zero. Similar models, without the
235 requirement of an autocorrelation term, were fitted to annual mean data taken from monthly
236 observations within specific seasons: winter (December - February); spring (March - May); summer
237 (June - August); and autumn (September - November).

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

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

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

273

274 **3. Results**

275 3.1 Trends in weather

276 *3.1.1 Linear analysis of meteorological variables*

277 Linear trend statistics for monthly mean air temperature, wind speed, solar radiation and total
278 monthly precipitation for all months and individual seasons respectively are presented in Tables 2a
279 and 2b. The only variable to show widespread linear change in the full monthly time series was mean
280 monthly wind speed. This declined at all sites but one (Porton) for which full data runs were available.
281 Negative trends in mean monthly wind speed at four sites in the south of England were significant at
282 $p < 0.001$, while p values for linear trends at several other sites fell marginally above 0.05.

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

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

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

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

307 *3.1.2 Non-linear analyses of meteorological variables*

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

320

321 *3.1.3 Analysis of daily extremes in meteorological variables*

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

339

340 3.2 Trends in atmospheric pollutants

341 *3.2.1 Linear analysis of atmospheric pollutant variables*

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

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

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

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

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

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

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

379

380 *3.2.2 Non-linear analyses of atmospheric pollutants*

381 Fitted GAM splines for the monthly volume-weighted average concentrations revealed differences in
382 temporal patterns between ions. In common with the results of the linear analysis, the clearest cross-
383 network signals of long term change were seen in total SO_4^{2-} and non-marine SO_4^{2-} concentrations. At
384 most sites, change at all points along the splines was statistically significant, indicating a progressive
385 reduction in anthropogenic SO_4^{2-} deposition throughout the 20 year monitoring record. Downward
386 trends in both variables at most sites were curvilinear with the most rapid reductions occurring in the
387 early years of monitoring, since when the rate of decline in concentration has slowed but remained
388 significant (Figure 2). Trends in these determinands for Snowdon, records for which only began in
389 2008, were essentially flat.

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

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

401

402 **4. Discussion**

403

404 Our analyses show that prevailing environmental conditions changed substantially across the network
405 over the first two decades of ECN monitoring. In general, changes in air chemistry variables were

406 largely in line with expectations, particularly given the international adoption in 1994 of the “Protocol
407 to the 1979 Convention on Long-Range Transboundary Air Pollution on Further Reduction of Sulphur
408 Emissions”, in which the UK agreed to reduce emissions of sulphur dioxide by at least 80% by 2010
409 relative to 1980 levels. Also according to expectations, inter-annual variation in weather was heavily
410 dominated by regional-scale synoptic variation, and provided relatively little evidence of the warming
411 that is apparent over longer timescales than currently represented by the network. Nevertheless, a
412 recent progressive increase in summer precipitation, most notably at wetter upland sites, was
413 particularly striking and had not been anticipated at the time the network was initiated.

414 4.1 Changes in weather

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

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

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

441 While there is a common perception that the global temperature rise has “paused” in recent years,
442 the global instrumental record demonstrates that the rate of temperature increase has at most slowed
443 (Mann, 2014), possibly reflecting recent phases in competing multi-decadal oscillations in Atlantic and
444 Pacific surface temperatures (Steinman et al., 2015). However, Karl et al. (2015) asserts that warming
445 over the current century to date is at least as rapid as that during the second half of the previous one.
446 In this context it is important to note that the mean annual temperature of the Central England
447 Temperature Series, (annual variation in which is strongly correlated with mean air temperatures
448 across the ECN), was approximately 0.7 °C higher, over the 1993-2012 ECN monitoring period, than
449 during the commonly used 1961-1990 baseline period. Similar differences are apparent when
450 comparing mean measured 1993-2012 air temperatures for individual sites with their mean CHES-

451 modelled 1961-1990 estimates. The smoothed spline fitting approach applied to a longer run of air
452 temperatures (1970-2012) modelled for ECN sites using CHESSE, indicate that temporal variation in air
453 temperatures over the last 20 years is entirely consistent with a longer-term progressive upward trend
454 at all sites (Figure 3). Comparison of temperature records over the two time scales therefore hints at
455 the likelihood of a forthcoming upward step-change in air temperatures at ECN sites to compensate
456 for recent stationarity. A possible portent is that the year 2014, without the period covered by this
457 analysis, was reported by the UK Met Office to be the warmest year in the UK instrumental record and
458 the warmest year for the Central England Temperature Series that stretches back to 1659
459 (<http://www.metoffice.gov.uk/news/releases/archive/2015/Record-UK-temps-2014>); 2014 was also
460 the warmest year globally (Trenberth and Fasullo, 2013).

461 Variation in seasonal temperatures at ECN sites over the first two decades of monitoring was very
462 closely linked with fluctuations in the NAO. During the winter months, the positive phase of the NAO
463 is associated with vigorous westerly dominated systems bringing relatively warm wet and windy
464 conditions, and the negative phase with blocking highs over the northern Atlantic, and considerably
465 colder, drier, and calmer weather (Hurrell and Van Loon, 1997). Conversely, during summer, a positive
466 NAOI describes a synoptic situation favouring anticyclonic warm and dry weather, while periods with
467 a negative NAOI are characterised by the movement of North Atlantic storm tracks over the UK, with
468 consequent increased precipitation and cooler temperatures (Folland et al., 2009). Winter
469 (December-February) and summer (June-August) mean daily temperatures at ECN sites for the 20-
470 year period were positively correlated with respective winter and summer NAO indices (NAO data
471 source: National Oceanic and Atmospheric Administration (NOAA) Climate Prediction Centre) (Figure
472 4). Winter temperatures tended to show the stronger Pearson correlation coefficients, ranging from
473 0.72 for North Wyke to 0.37 for Cairngorm, with particularly tight relationships evident for the
474 southerly lowland ECN sites. Slight negative (although invariably statistically insignificant), winter
475 temperature trend slope coefficients across the network are broadly consistent, therefore, with a
476 weak negative slope in the winter NAOI over the monitoring period.

477
478 Correlations between summer temperatures and the summer NAOI were slightly weaker (ranging
479 from 0.64 at Glensaugh to 0.17 at Snowdon) than winter relationships. However, in contrast to the
480 winter index, the summer NAOI showed a strong negative trend over much of the monitoring period.
481 Furthermore, a series of five consecutive summer NAOI values below -0.5 from 2008 onwards is
482 without precedent in the 60 year NAO record analysed, and there is no evidence for a similar
483 prolonged period of negative summer indices in the series when extended back to 1821 by Jones et
484 al. (1997). As a postscript to this analysis, the NAOI for the summers of 2013 and 2015 was also
485 strongly negative although it was positive in 2014. Despite a generally weaker correlation between
486 the summer NAOI and summer temperatures at ECN sites, the more directional nature of the summer
487 NAOI may, therefore, have exerted a negative influence on air temperature trends over the full
488 monitoring period.

489
490 Statistically significant linear increases in monthly precipitation across all seasons were identified at
491 three sites only – all in north eastern locations (i.e. Sourhope, Glensaugh and Cairngorm), and slope
492 coefficients were invariably slight. In contrast, summer precipitation increased dramatically across the
493 network and was highly statistically significant at all sites other than North Wyke and Hillsborough,
494 with the greatest absolute increases occurring at the wettest sites.

495 Again, Figure 4 demonstrates negative relationships between summer precipitation and the summer
496 NAOI with Pearson Correlation coefficients that range from very strong (-0.74 for Snowdon), to weak
497 (-0.27 for Hillsborough) but are generally high (mean = -0.59). Strong correlations between summer
498 precipitation and summer NAOI have been identified previously for periods preceding the shift in the
499 latter to consistently negative values (Folland et al., 2009). The trend in the summer precipitation

500 signal over the ECN monitoring period is therefore clearly linked to the recent, apparently anomalous,
501 uni-directional behaviour of the summer NAOI (Figure 5).

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

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

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

540 We found relatively little evidence for significant linear trends in solar radiation at ECN sites despite
541 changes in air quality that might be expected to have been accompanied by reductions in sulphate
542 aerosol, and thus a reversal of "global dimming". However, average levels increased significantly at
543 four relatively low rainfall sites in the south of England and Snowdon during spring, when trends at
544 other sites were also consistently positive (with the exception of Hillsborough). In contrast, trends in
545 solar radiation in summer were consistently negative, but were not as marked as the positive trends

546 in precipitation in this season, with only three sites showing statistically significant trends over the full
547 monitoring period.

548 *4.2 Changes in atmospheric chemistry*

549 The concentration of most major ions in bulk precipitation declined significantly at most ECN sites over
550 the 20 year period. The strong and highly significant downward trends in non-marine SO_4^{2-}
551 concentration across the network were expected, given the controls imposed on major sulphur
552 emission sources, i.e. coal and oil burning power stations and other industrial plant in recent decades.
553 UK emissions of sulphur to the atmosphere are estimated to have declined by 93% between 1970 and
554 2008 (RoTAP, 2012). Large reductions in non-marine SO_4^{2-} concentration across the UK have been
555 reported previously for ECN sites (Morecroft et al., 2009) and trends measured across the UK
556 Acidifying and Eutrophying Atmospheric Pollutants network (UKEAP) have been well documented
557 elsewhere (e.g. RoTAP (2012); Curtis and Simpson (2014)). The regional variation, and the magnitude
558 of change, in non-marine SO_4^{2-} concentration we observed was highly consistent with previous
559 analysis of UKEAP data by Fowler et al. (2005), with a gradient of declining deposition rates from high
560 (south-east England) to low (north west Scotland). Rates of change in non-marine SO_4^{2-} concentration
561 were most marked for the south easterly sites Rothamsted, Alice Holt and Drayton, but large trends
562 were also observed at North Wyke (south-west England) and Hillsborough (Northern Ireland).
563 Surprisingly, the trend at Wytham, also in the south-east, was relatively muted, suggesting local
564 sources may have had an important influence on local air quality here. Wet and dry sulphur deposition
565 have been the primary contributors to the acid load in the UK since the onset of major industrial
566 activity, although relative rates of change in the two vary, with dry deposition (not measured at ECN
567 sites) falling more rapidly close to sources and wet deposition dominating in more remote regions.
568 The curvilinear fits (Figure 2) demonstrated that while reductions in non-marine SO_4^{2-} were sustained
569 throughout the full monitoring period at all sites, change occurred at the greatest rate in the earlier
570 years.

571 In recent years emission controls are generally reported to have resulted in much stronger effects on
572 deposited sulphur compared to the other major acidifying agent, nitrogen, to a point where nitrogen
573 deposition is beginning to dominate the residual acid load. Nitrogen deposition in the UK is derived
574 from three major sectors: power plants and other industrial fossil fuel combustion sources, motorised
575 transport and agriculture. The former two emit N primarily in oxidised forms ($\text{NO}_x\text{-N}$) which are either
576 deposited to the surface dry or as NO_3^- in precipitation after reaction within clouds. UK emissions of
577 $\text{NO}_x\text{-N}$ are thought to have declined by over 50% between 1970 and 2008, with much of the reduction
578 occurring after 1990 (RoTAP, 2012). The strong correlations we found between rates of change in
579 NO_3^- and non-marine SO_4^{2-} are indicative of the common primary source of this pollutant, although
580 patterns of change were more linear for the former than the latter, with more consistent and more
581 gradual change occurring across the two decades. Concentrations of NO_2 also showed net declines at
582 the majority of sites, although the patterns of change were much more variable between sites,
583 possibly reflecting a greater contribution from local sources which are likely to have varied much more
584 than regional-scale sources.

585 Rates of change in NO_3^- were also independently correlated, although to a lesser extent, with rates of
586 change in NH_4^+ . Agricultural livestock is considered to be the primary source of emissions of ammonia
587 to the atmosphere, which is then either deposited in gaseous form or is transformed to NH_4^+ and
588 deposited in precipitation. The largest reductions in NH_4^+ were seen at agricultural research sites
589 where concentrations have historically been the highest in the network (Hillsborough, Drayton and
590 Rothamsted). When ammonia is sufficiently abundant in the atmosphere to have neutralised any
591 sulphuric acid, it can react with gaseous nitric acid to form ammonium nitrate aerosol (Metzger et al.,

592 2002). This may therefore explain the link between trends in the two N species in wet deposition. In
593 contrast to previous studies that have shown a dominance of S deposition over N deposition, we found
594 that rates of change in NO_3^- and NH_4^+ concentrations combined (when expressed in terms of
595 equivalent acidity) roughly equated to, or exceeded, rates of change in non-marine SO_4^{2-} at the
596 agricultural sites on the network, while at the other sites the latter usually dominated.

597 Strong correlations between rates of change in chloride and sodium were indicative of a dominant
598 influence on the concentrations of these ions from sea salt. Reductions over the 20 years are therefore
599 consistent with significant declines in winter and spring wind speeds, since sea salt aerosol dispersion
600 over land is highly dependent on wind strength and a reduction in the winter NAOI (Evans et al., 2001).
601 However, it should also be noted that rates of change in chloride concentration at the majority of sites
602 were disproportionately higher than rates of change in sodium (equivalent ratio of Na:Cl in sea salt =
603 0.86), indicating reduction in supply from another source of chloride. In an assessment of ECN, UKEAP
604 and Upland Waters Monitoring Network data, Evans et al. (2011) demonstrated that the UK had
605 experienced a significant reduction in hydrochloric acid (HCl) deposition in recent years. A product of
606 the combustion of coal with a high chlorine content, it would appear that this decline in HCl has made
607 a significant contribution to the overall reduction in the acid load.

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

622

623 5. Conclusion

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

637 similar trends are occurring nationally. However, data from other more-issue focussed networks are
638 clearly vital to provide the necessary spatial context for these observations.

639

640 **Acknowledgements**

641 The authors wish to thank the many people who have contributed over the years to the collection
642 and storage of ECN data both at sites and at the Central Coordination Unit. ECN monitoring and
643 research at the sites included in our study is funded by: the Agri-Food and Biosciences Institute; the
644 Biotechnology & Biological Sciences Research Council; the Defence Science & Technology
645 Laboratory; Natural Resources Wales; Department for Environment, Food and Rural Affairs (Defra);
646 the Forestry Commission; Natural England; the Natural Environment Research Council (NERC); the
647 Scottish Government; Scottish Natural Heritage and the Welsh Government. Essential scientific
648 support has been provided by the NERC Centre for Ecology & Hydrology, ADAS, Forest Research,
649 Rothamsted Research and The James Hutton Institute. We are also very grateful to three reviewers
650 for constructive comments that have helped improve the manuscript.

651

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813 **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 (NO₂)**
814 **concentrations provided for the period 1995 – 2012 at most sites (Cairngorm record covers 2000 – 2012). Mean ammonia (NH₃) concentrations provided**
815 **for the period 1998 – 2012 at most sites (Alice Holt record covers 2000 – 2012); Snowdon record covers 2009 – 2012). NA = insufficient data available to**
816 **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⁻ (µeq L ⁻¹)	42	NA	43	43	43	26	33	NA	57	25	34	34
non-marine SO ₄ ²⁻ (µeq L ⁻¹)	33	NA	36	30	32	19	17	NA	50	16	24	28
NO ₃ ⁻ (µeq L ⁻¹)	36	NA	34	35	24	17	24	NA	43	12	26	26
NH ₄ ⁺ (µeq L ⁻¹)	32	NA	44	34	47	21	27	NA	51	14	24	34
Cl ⁻ (µeq L ⁻¹)	72	NA	60	108	124	57	151	NA	76	85	91	52
Na ⁺ (µeq L ⁻¹)	67	NA	59	105	90	55	130	NA	63	72	89	44
Mg ²⁺ (µeq L ⁻¹)	16	NA	15	24	25	12	29	NA	10	17	20	10
Ca ²⁺ (µeq L ⁻¹)	33	NA	33	16	36	11	27	NA	23	7	15	23
K ⁺ (µeq 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

819 **Table 2a.**

820 **Linear trends with time (change per year) in monthly meteorological variables (all seasons included) and annual climate indices recorded at UK ECN sites**
 821 **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**
 822 **years where a minimum of 300 days of meteorological data were available.**

823 NA = data not available.

Site	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	
	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

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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

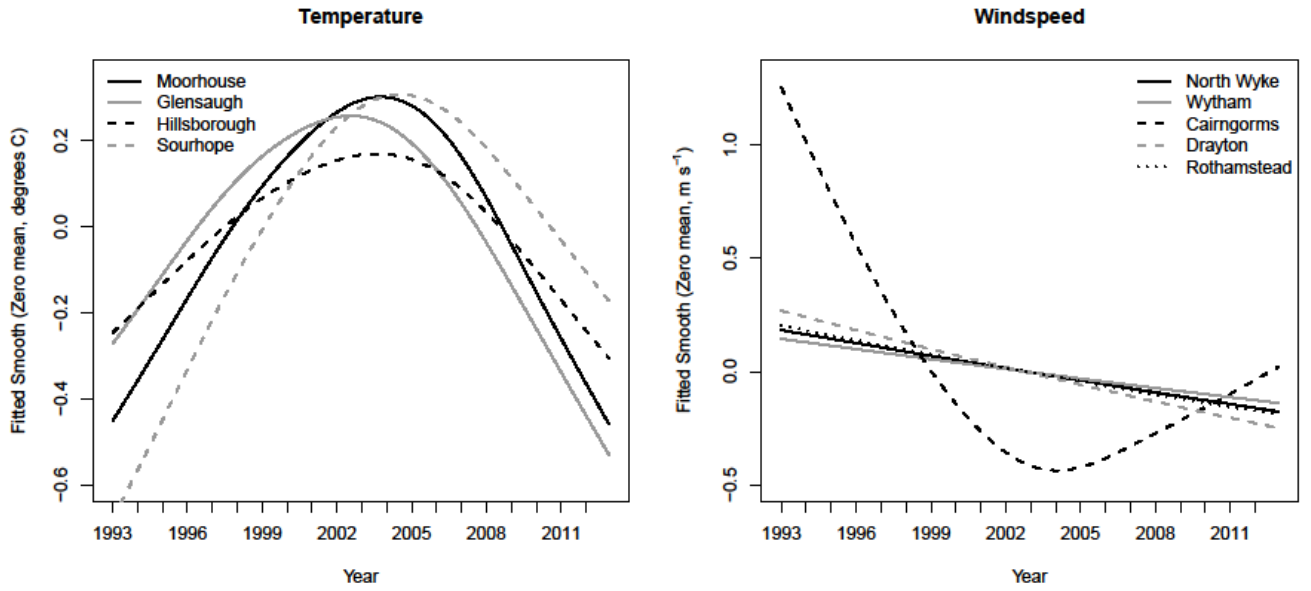
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Table 3. Linear trends with time in monthly volume weighted average ionic concentrations ($\mu\text{eq L}^{-1} \text{yr}^{-1}$) and fluxes ($\mu\text{eq m}^{-2} \text{yr}^{-1}$), and trends in mean monthly NO_2 concentrations and mean monthly NH_3 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 ($\mu\text{eq L}^{-1} \text{yr}^{-1}$)										ppb yr^{-1}	$\mu\text{g m}^{-3} \text{yr}^{-1}$
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 ($\mu\text{eq m}^{-2} \text{yr}^{-1}$)											
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		

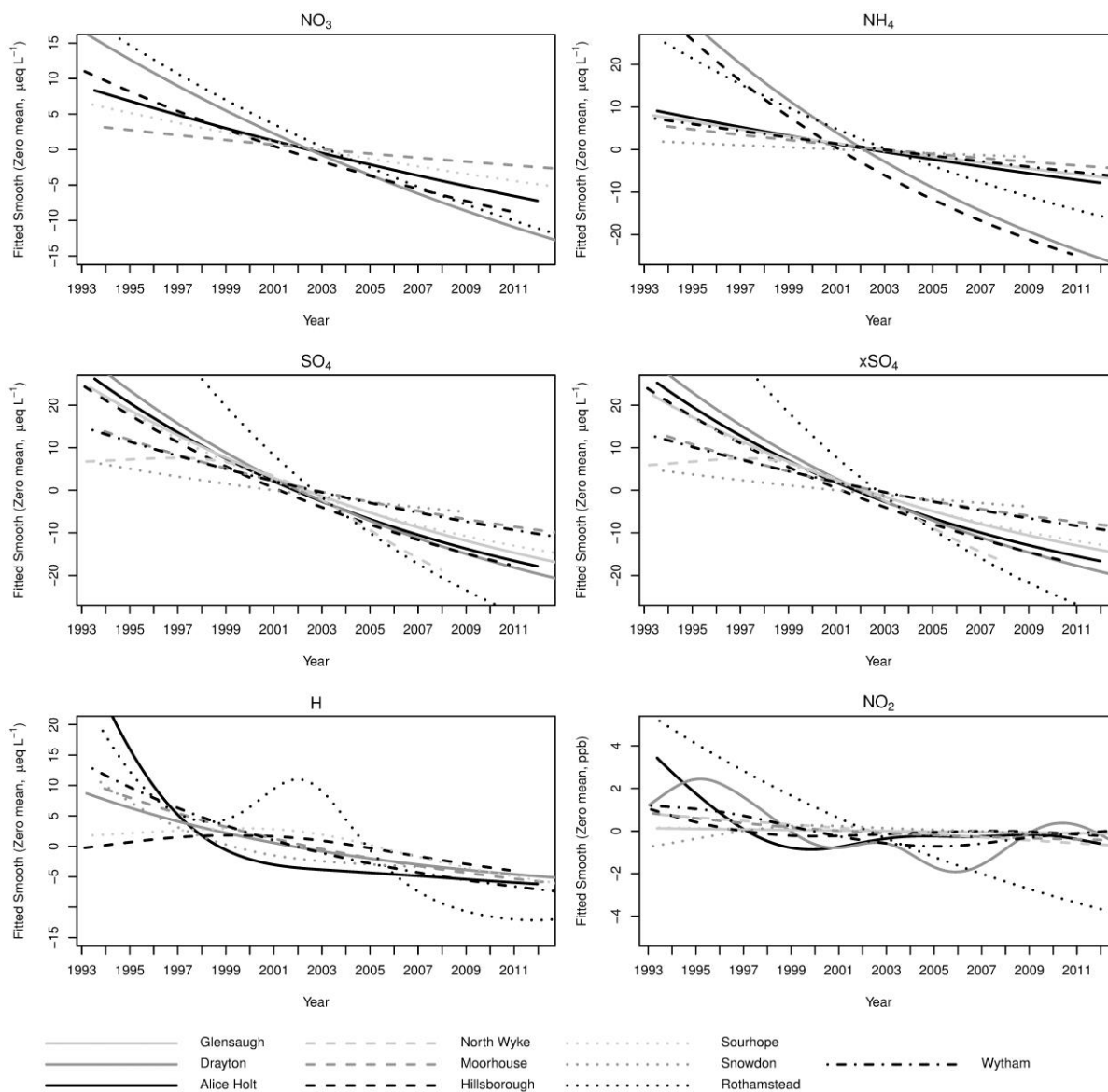
1 **Figure 1. Statistically significant curvilinear generalised additive model (GAM) fits for ECN**
2 **meteorological variables.**

3



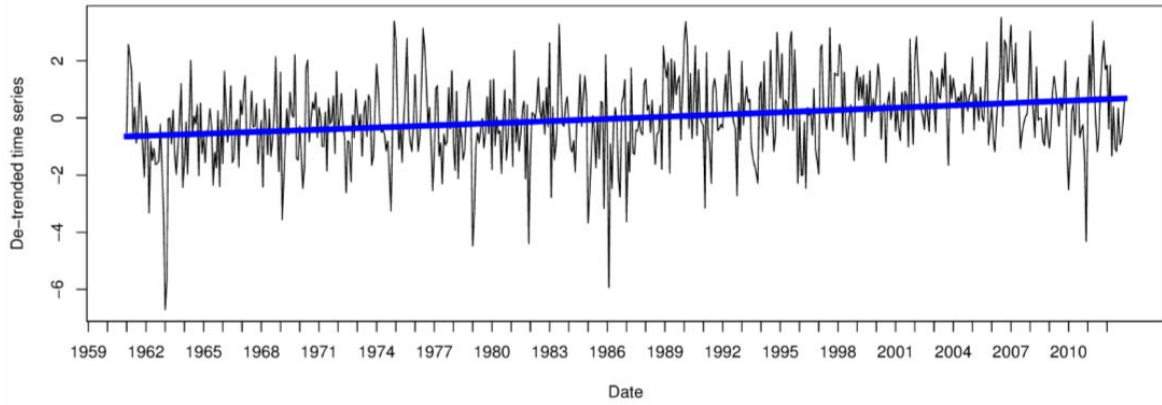
4

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

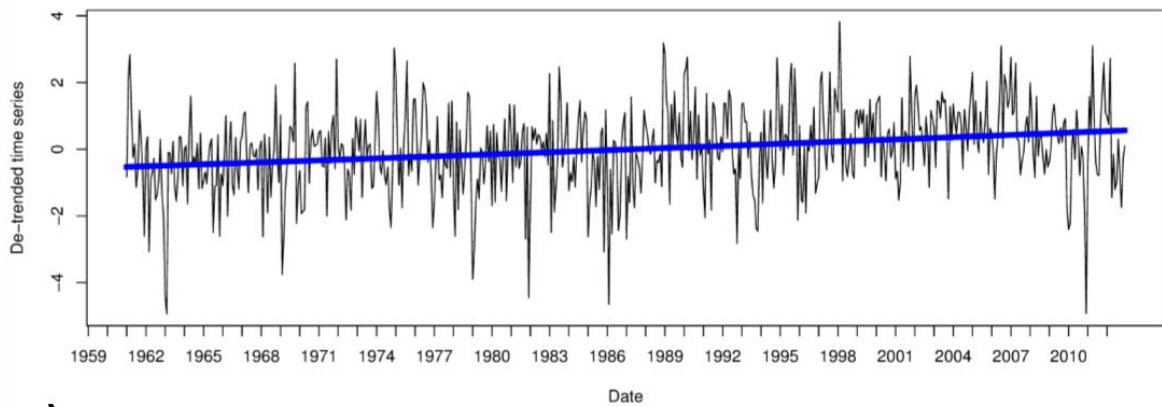


10 **Figure 3. Curvilinear Generalised Additive Model (GAM) fits for modelled (CHES) monthly air**
11 **temperature data for three ECN sites, (a) Rothamsted, (b) Moor House and (c) Glensaugh (1970-**
12 **2012). Data represent variation (in °C) relative to a mean of zero °C. In each case the trend is**
13 **significant throughout these records.**

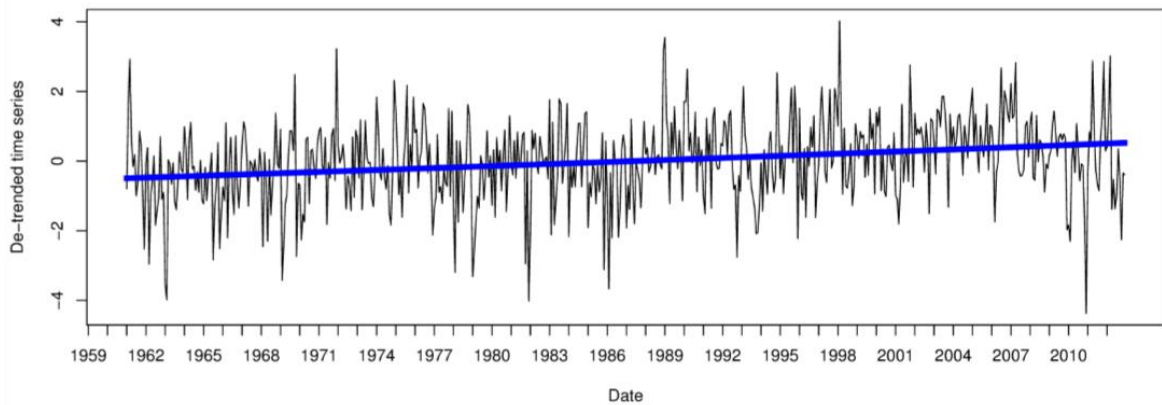
a)



b)



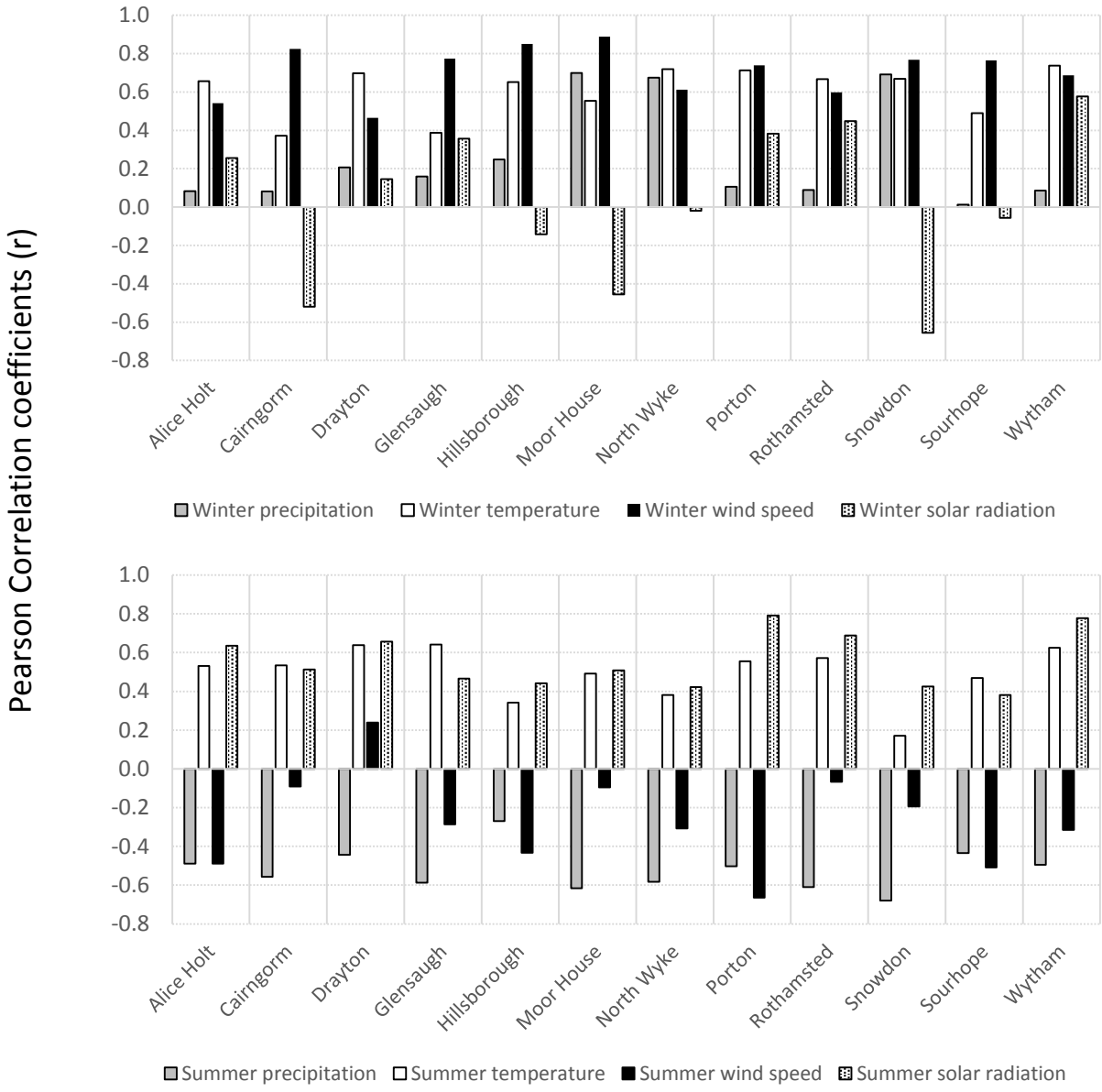
c)



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15

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

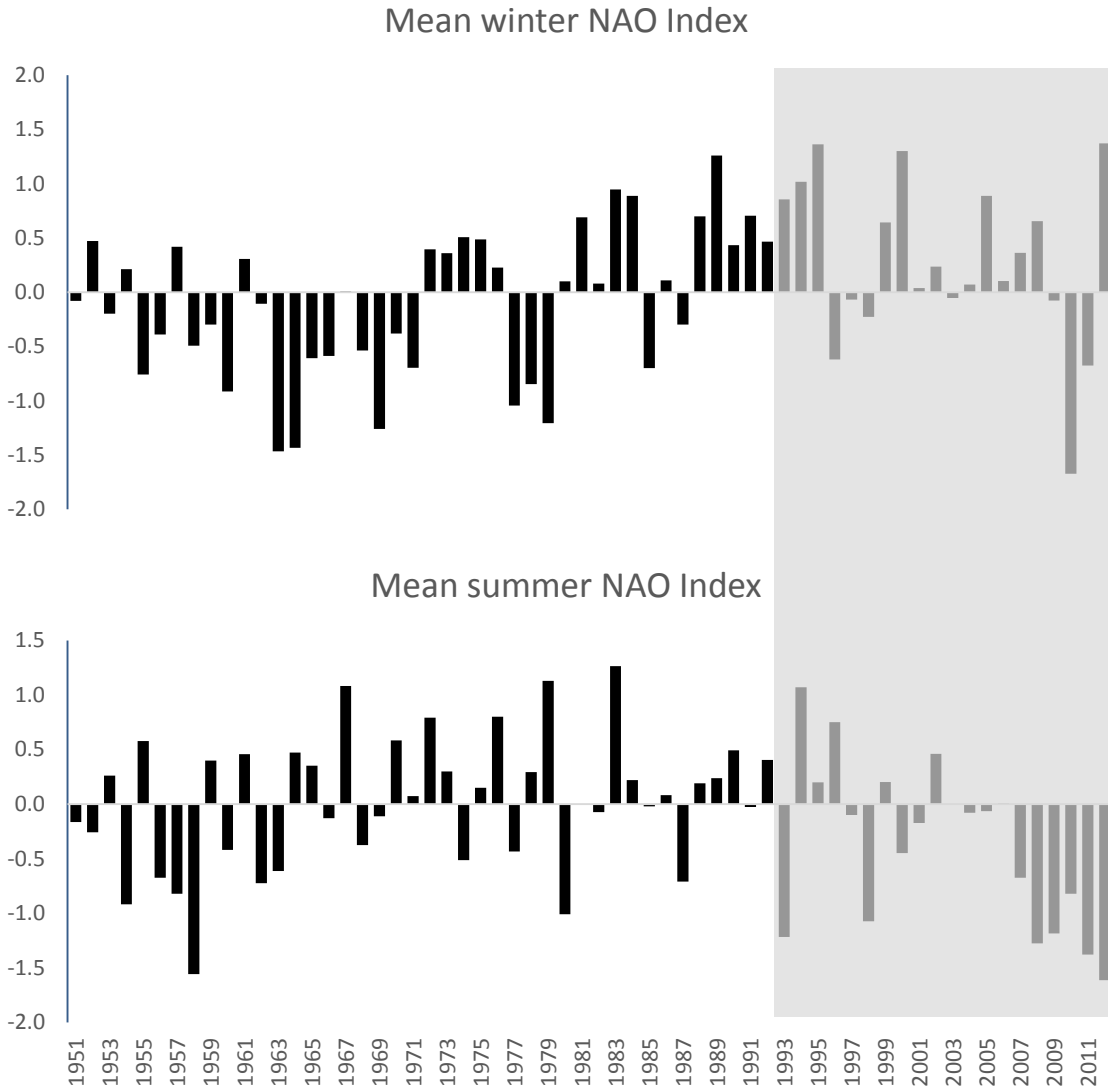


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22

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

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