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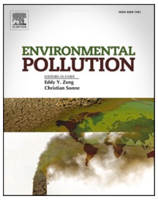

The evolving role of weather types on rainfall chemistry under large reductions in pollutant emissions☆

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ABSTRACT

Long-term change and shorter-term variability in the atmospheric deposition of pollutants and marine salts can have major effects on the biogeochemistry and ecology of soils and surface water ecosystems. In the 1980s, at the time of peak acid deposition in the UK, deposition loads were highly dependent on prevailing weather types, and it was postulated that future pollution recovery trajectories would be partly dependent on any climate change-driven shifts in weather systems. Following three decades of substantial acidic emission reductions, we used monitoring data collected between 1992 and 2015 from four UK Environmental Change Network (ECN) sites in contrasting parts of Great Britain to examine the trends in precipitation chemistry in relation to prevailing weather conditions. Weather systems were classified on the basis of Lamb weather type (LWT) groupings, while emissions inventories and clustering of air mass trajectories were used to interpret the observed patterns. Concentrations of ions showed clear differences between cyclonic-westerly-dominated periods and others, reflecting higher marine and lower anthropogenic contributions in Atlantic air masses. Westerlies were associated with higher rainfall, higher sea salt concentrations, and lower pollutant concentrations at all sites, while air mass paths exerted additional controls. Westerlies therefore have continued to favour higher sea salt fluxes, whereas emission reductions are increasingly leading to positive correlations between westerlies and pollutant fluxes. Our results also suggest a shift from the influence of anthropogenic emissions to natural emissions (e.g., sea salt) and climate forcing as they are transported under relatively cleaner conditions to the UK. Westerlies have been relatively frequent over the ECN monitoring period, but longer-term cyclicity in these weather types suggests that current contributions to precipitation may not be sustained over coming years.

1. Introduction

Atmospheric deposition is the process by which chemical species are transported from the atmosphere to terrestrial and aquatic surfaces (Pacyna, 2008). It contributes acidity and other toxic contaminants, provides nutrients to plants and is attributed with maintaining key biogeochemical cycles in more nutrient poor environments (Tipping et al., 2014). The deposition loads of major ions and nutrients can therefore induce both acidification and eutrophication. Deposited acidity in upland areas depleted soils of base cations, and resulted in the chronic acidification of soils and acid-sensitive freshwater ecosystems over centennial time scales (Battarbee and Charles, 1986), and is also linked to forest decline (Grennfelt et al., 2020). Eutrophication by atmospherically deposited reactive nitrogen can have detrimental effects on some taxa (e.g. heathland herb and shrub species) while benefitting nitrophilous competitors, leading to declines in biodiversity

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(Stevens et al., 2020). In addition atmospheric deposition determines the ionic strength of precipitation and catchment runoff in a way that can have a profound effect on terrestrial organic matter solubility and transport, and hence serves to regulate riverine carbon fluxes (Hruška et al., 2009; Lawrence and Roy, 2021).

The period of peak atmospheric deposition of sulphur and acidity in the UK (1970s–80s) coincided with growing awareness of a threat to the global climate system posed by the emission of greenhouse gasses. Among issues considered at the time was the potential for climate change to influence acid deposition patterns through its influences on synoptic weather conditions. Synoptic weather conditions in the British Isles are often summarized by simple atmospheric circulation types (i.e. Lamb weather types, LWTs) (Lamb, 1972). Specifically, westerlies deliver unsettled weather and variable wind directions as depressions cross the UK, with most rain in northern and western districts. Cyclonic weather brings wet unsettled conditions with variable wind directions over most of the country, while anticyclonic weather is mainly dry with light winds, and warm in the summer with occasional thunderstorms. In the 1980s, it was shown that LWTs provided a robust proxy for assessing the weather dependence of precipitation composition. For example, Davies et al. (1986) showed that the highest H^+ loadings in bulk precipitation and fraction of total rainfall was associated with cyclonic weather, while Farmer et al. (1988) showed the interplay of emission and climatological factors on deposition fluxes by studying trends of NO_3^- and SO_4^{2-} deposition. Davies et al. (1990) demonstrated the utility of LWTs as indicators of synoptic meteorology using data from Eskdalemuir, which led to a LWT classification approach based on the meteorological controls on rainfall ion content between 1981 and 1985 at three UK sites (Davies et al., 1991). They showed that LWTs were effective in discriminating between pollutant loads, but it was also demonstrated that the LWT-deposition relationship was highly site specific. Since these investigations, the implementation of clean air policies, both nationally and internationally, has led to major reductions in emissions of acidifying pollutants in recent decades. Sulphur dioxide (SO_2) emissions declined in the UK and EU by 71% and 72% respectively between 1986 and 2001 (Fowler et al., 2005), and the downward trend has continued more recently (see section 3.1). Concomitantly, emissions of reduced N and oxidized N emissions declined by 10% and 40% respectively, which agrees with reports in the US and China that reduced N emissions is becoming more important since it is unregulated (Li et al., 2016; Liu et al., 2013). Declining acid emissions has led to major reductions in the wet and dry deposition of sulphur species (Fowler et al., 2005) and hydrochloric acid (Evans et al., 2011), with smaller reductions in nitrogen species, and subsequently reductions in concentrations of sulphate (SO_4^{2-}) and chloride (Cl^-) in soil waters (Sawicka et al., 2017) and surface waters (Evans et al., 2011; Stoddard et al., 1999). More recently, clear regional scale reductions in nitrate concentration in surface waters are also becoming apparent (Austnes et al., 2018). Evidence is also beginning to emerge of widespread improvements in both acid-sensitive freshwater plants and animals (Monteith et al., 2005; Murphy et al., 2015) and acid-sensitive terrestrial vegetation (Rose et al., 2016).

Global emissions of greenhouse gasses have resulted in a rise in global ocean and land surface temperatures and evidence for impacts on atmospheric circulation patterns (IPCC, 2021, 2014), both of which are shown to influence UK precipitation (Blenkinsop et al., 2015). The UK's geographic location renders its weather highly sensitive to the dominant modes of circulation and fluctuations in the position of the jet stream, which could progressively migrate northward beyond its natural variability under unabated future warming (Osman et al., 2021). It is therefore reasonable to expect that climate change, through its influence on weather patterns, will exert influences on the concentrations and loads of residual atmospheric pollutants and marine salts deposited to the UK land surface.

Given the major reductions in atmospheric pollutants in the UK since the original LWT characterisation work, it is important to understand whether the relationships between deposition concentrations and fluxes and LWTs is changing as the effect of climatic forcing become more dominant. Such understanding could provide important insights into the evolving relationship between deposition chemistry and terrestrial biogeochemical dynamics as climate change progresses in a new low-sulphur environment.

In this study, we analysed amounts and chemical compositions of long-term weekly precipitation samples collected at four UK Environmental Change Network (ECN) sites located in contrasting parts of the British Isles, and characterised deposition chemistry measurements by LWT in the week preceding each sampling date. We sought to understand i.) the role of geographical location in the relationships between deposition chemistry and weather metrics (i.e. LWTs), ii.) whether these relationships have been maintained over a period of very large changes in atmospheric emissions and iii.) whether certain types of weather types have become more frequent over the ECN monitoring period. In addressing iii.) we also assessed longer-term trends in weather-type records in order to determine whether changes in weather patterns observed over the period covered by ECN are consistent with longer term shifts in climate and may thus be influencing atmospheric deposition patterns over the British Isles in the longer term.

2. Methods and data

2.1. UK environmental change network data

The UK Environmental Change Network (ECN) is the only long-term monitoring scheme providing co-located measurements of physical, chemical and biological variables for a range of UK ecosystems (see Rennie et al. (2020) for data and site descriptions). Its eleven sites span much of the UK and represent a variety of land use classes. It is a member of International Long-term Ecological Research (ILTER) network and is part of its European regional network (LTER-Europe). In this study, we analysed bulk deposition chemistry data collected weekly at ECN sites between 1993 and 2015 (Rennie et al., 2017b), together with hourly rainfall data collected by the Automatic Weather Station(AWS) at each site (Rennie et al., 2017a). Methodological protocols for both datasets follow Sykes and Lane (1996). Specifically, we assessed trends in the electrical conductivity (EC) and the concentration of major ions in precipitation samples. The major ions analysed included the strong acid anions, non-marine sulphate (xSO_4^{2-}), nitrate (NO_3^-) and chloride (Cl^-), and the cations ammonium (NH_4^+) and sodium (Na^+). Deposition of nitrogen species can exert both acidifying and eutrophying effects depending on how they are biogeochemically processed in soils. Na^+ and Cl^- ions in bulk precipitation across the UK are derived predominantly from seasalt, although the burning of coal with a high chlorine content has also contributed to atmospheric loads of Cl historically (Evans et al., 2011).

Sulphate concentrations were converted to xSO_4^{2-} by subtracting a hypothetical marine sulphate fraction from total sulphate, based on the assumptions that all chloride was derived from seasalt and that the chloride-sulphate ratio in seasalt composition was 9.62 (based on a molar ratio of 0.14^{-1} (Morris and Riley, 1966)) when all concentrations are expressed as $\mu eq/L$. Each weekly bulk precipitation measurement was matched with the total volume of precipitation and the dominant daily LWT from the day after the previous measurement to the day of the current measurement (spanning 6–8 days). We limited our assessment to four geographically spread ECN sites, Glensaugh (northeast Scotland), Moor House (northern England), Rothamsted (southeast England), and Snowdon (northwest Wales). These sites provide some of the most complete long-term precipitation chemistry records, while the geographical spread ensured a range in levels and amount of change in atmospheric deposition as a consequence of relative distance from major pollution emission sources. The sites also differed in their distance from the west coast, thus influencing contributions of seasalt deposition that is most pronounced in the west of Britain.

Recent trends and shorter-term variability in weather and atmospheric deposition variables at ECN sites over the period 1993–2012 have previously been reported by Monteith et al. (2016). They found directional trends in summer rainfall that could be linked to a prolonged negative excursion in the summer North Atlantic Oscillation (NAO) Index, and a significant reduction in wind speed, particularly during winter and spring. Sulphate, NO_3^- and NH_4^+ concentrations in bulk deposition declined significantly at most sites, and these changes could be linked to gradual increases in soil water pH, reflecting the partial recovery of soil chemistry from acidification.

To determine chemical fluxes, we converted weekly sampled chemical concentrations to annual fluxes using the following equation:

$$flux = 52.18 \times concentration \times V \div A \quad (1)$$

where V is the collected sample volume, and A is the area of the collector funnel.

2.2. Back trajectory analysis of air masses and source apportionment

Back trajectory analysis was applied to provide a clearer insight into the pathways air masses take before arriving at an observation location (e.g. Baker, 2010; Dorling et al., 1992; Dorling and Davies, 1995; McGregor and Bamzelis, 1995 in the UK). We used the single particle back trajectory model HYSPLIT (Draxler and Hess, 1997; Draxler and Rolph, 2015) via the R package OpenAir (Carslaw and Ropkins, 2012). HYSPLIT is one of the most widely used models for atmospheric trajectory and dispersion calculations (Stein et al., 2015). The arrival height and the trajectory run time were set to 10 m and 96 h, respectively, while the meteorological database used was the Global NOAA-NCEP/NCAR reanalysis data archives. Since we needed to visualise patterns over multiple years, we applied a clustering algorithm (also conducted via OpenAir) to group similar trajectories. The 'angle' distance measure is used to group back trajectory points with a similar angle of origin.

Key to the interpretation of deposition chemistry is the temporal evolution of emissions from large point sources and from different sectors. We therefore surveyed publicly available databases for large combustion plants (European Environmental Agency, 2019) and the NO_x and SO_2 emissions by sector from 1970 to 2019 from the UK National Air Emission Inventory (NAEI) database (https://naei.beis.gov.uk/data, see supplementary information (SI)) to study their temporal evolution and shifts in relative importance.

2.3. Lamb weather types

The Objective Jenkinson classification of LWTs has 27 classes, which includes 8 directional types (e.g. N, NE), non-directional types (cyclonic, anti-cyclonic), combined complex hybrid types (e.g. CN, CNE) and unclassifiable types (U) (Jenkinson and Collison, 1977). Daily records of the objective LWTs are available from 1871 to present (https://crudata.uea.ac.uk/cru/data/lwt/).

The 27 objective LWTs belong to several sub-groups that partially overlap. Principal Component Analysis (PCA) of annual frequencies of LWT on annual weather type frequencies from 1861 to 1980 suggested only six were needed to define and monitor changes in atmospheric circulation (Jones and Kelly, 1982). LWTs are widely used and have been shown to influence rainfall oxygen isotopes level (Tyler et al., 2016), heavy rainfall (Barnes et al., 2021) and air quality (Graham et al., 2020; Pope et al., 2014). In a recent study, five out of the 27 objective LWTs were found to account for >80% of flood events in the River Eden, Cumbria, UK since 1873 (Pattison and Lane, 2012). These 'event-generating' weather types are westerlies, cyclones, and their combinations (i.e. C, W, SW, CW, CSW), which are highly related to North Atlantic climate forcing metrics, e.g. NAO. In this study, we grouped rainfall chemistry samples into classes on the basis of the frequency of occurrence of these LWTs during the period of sample collection. Hereinafter, these five weather types (i.e. C, W, CSW, SW, CW) are grouped together under the term *"westerlies"* while all other weather types are grouped as "*other*". The weekly rainfall chemistry samples are classified by the number of days of westerlies in the previous week.

To quantify the temporal trends in concentrations associated with the main classes and compare changes across sites, we fitted a linear model for each chemical species against the number of westerlies per week and per year. The model took the form:

$$concentration = \beta_0 + \beta_1 lambcount + \beta_2 year + \beta_3 year \times lambcount + \varepsilon \quad (2)$$

where β_0 is the intercept, β_1, β_2 and β_3 are linear model coefficients, *year* is the calendar year, *lambcount* is the number of westerlies per week, and *year* × *lambcount* is the interaction term. We found that the variability (or spread) of concentration increases with the westerlies counts. Therefore, a weighted least-squares approach was taken to account for the heteroscedasticity, involving fitting a linear model but with weights prescribed at each data point. The weights were determined by first fitting an unweighted linear model and obtaining its fitted values and residuals, and then fitting another linear model between them and take the reciprocal of the squared fitted values of the second model.

Finally, to test whether any effect of weather type on chemical concentrations was consistent or deviated over time, we performed a likelihood ratio test (LRT) on each model to see whether the addition of an interaction term between two variables (i.e. number of westerlies and year) would significantly improve the accuracy of the model. The test statistic for the LRT follows a chi-squared distribution with degrees of freedom equal to the difference in dimensionality of the models. The equation for the test statistic is provided below:

$$\lambda_{LR} = -2(\ell(model\ with\ interaction) - \ell(model\ without\ interaction)) \quad (3)$$

where $\ell(.)$ is the log-likelihood function. If the LRT indicated it is beneficial to add the interaction term, this would provide evidence that the effects of westerlies on concentrations have changed over the ECN monitoring period. The LRT is performed using the R package *lmtest* (Zeileis and Hothorn, 2002).

2.4. Time series analysis of count data

We used two methods for time series analysis of count data (i.e. number of westerly days per year). In the first method, a changepoints algorithm provided by the R package *changepoint* (Killick and Eckley, 2014), was used to define segments with distinctively different means and variances. A Poisson distribution was assumed and the efficient Pruned Exact Linear Time (PELT) algorithm (Killick et al., 2012) was used to identified segments with significantly different mean and variance. In the second and separate method, we normalized the time series and fitted a generalized additive model (GAM). Segments of the time series during which the count was deemed to have been increasing or decreasing on the basis of first derivative being significantly different from zero were highlighted.

3. Results and discussion

3.1. Air mass trajectories based on lamb weather types and source apportionment

To better understand the pathways and source of air masses under different rainfall and weather types, we conducted 96 h back trajectory analysis for air masses arriving at the four ECN sites from 1993 to 2015. We grouped the trajectories by the LWTs then performed a 10-member clustering of the trajectories. The cluster analysis revealed distinctive patterns for the different LWTs (Fig. 1), and emphasised the classic anti-clockwise spirals for cyclones in the Northern hemisphere.

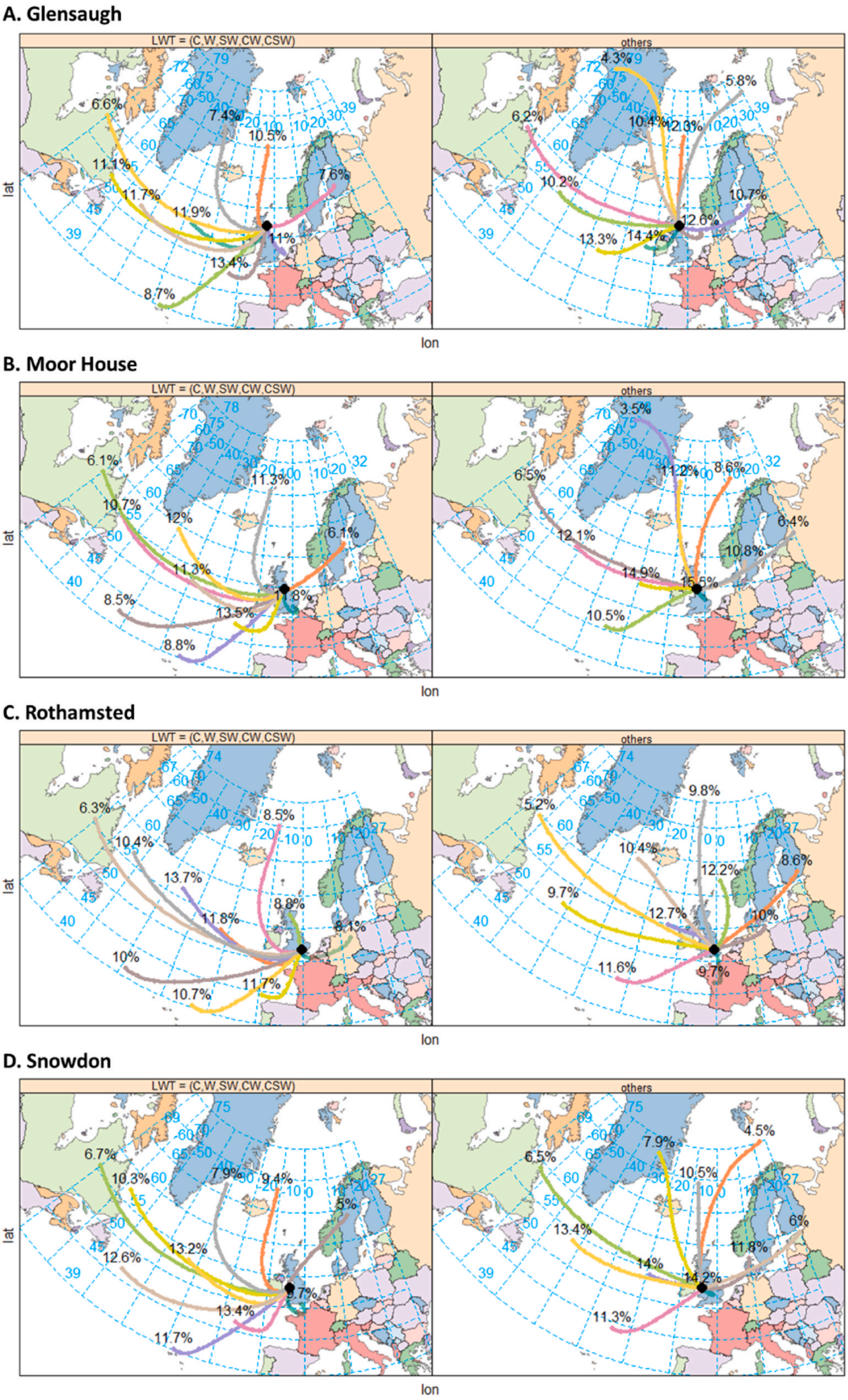

Fig. 1. Mean trajectories showing the 10-cluster solution to air mass back trajectories (previous 96h) at the four ECN sites (black dot) grouped by Lamb weather types (LWTs). Note that clustering is performed for each group independently. The percentages next to each mean trajectory denotes the percentage of trajectories associated with each cluster.

For all sites, roughly 17% of air mass originated between 45 and 60° N along or off the coast of N. America for westerlies, in contrast to only 6% for the other LWTs. Note that for both groups, at least 40% of air masses originated from the mid-Atlantic Ocean. When classifying the trajectories into those that travelled from the east or the west of a site, up to 20% only of westerly trajectories originated from the east, in contrast to up to 40% for the other LWTs. A perhaps surprising feature for the other LWTs was that their air masses were more likely to travel relatively straight paths north of the UK. The other LWTs also included a small but larger fraction of air masses originating from Scandinavia than the westerlies. Only a small percentage of trajectories travelled from continental Europe and they only tended to travel short distances.

Key to the interpretation of deposition chemistry is the temporal evolution of emissions from large point sources. Fig. 2 shows the significant reduction of NOx or SO2 emissions from large combustion plants within the UK, which were emitting more than 10,000 tonnes per year of NOx or SO2 in 2005, from 2004 to 2017 (European Environmental Agency, 2019). In general, reductions in the emission of SO2 have been greater than NOx. However, significant reductions in emissions of SO2 from Longannet (south Scoatland) and Aberthaw (south Wales) are not apparent until 2014 (see Fig. SI 2). Other than these sites, almost all of the remaining SO2 emissions originated from sources in central England. In terms of proximity of the residual sources to the ECN sites used in this study, Glensaugh is nearest to the Scottish plants; Rothamsted is closest to Didcot A; Snowdon is relatively close to Fiddlers Ferry, while Moor House lies almost equidistant between the Scottish Plants and Drax and Eggborough in south Yorkshire, England.

As the emissions from large combustion plants decrease, emissions from other sectors become more important. A survey of the NAEI database reveals that SO2 emissions reduced drastically from 3425 kt to 268 kt during the ECN monitoring period (1992–2015) and further reduced to 163 kt in 2019 due to a very significant decrease in emissions from public electricity and heat production, with residential combustion becoming the largest remaining source. Meanwhile, NOx as NO_2 emissions reduced from 2834 kt to 1015 kt largely due to reductions in coal and oil-based public electricity production. Public electricity production remains a major source of N emissions, but is now at a similar level to stationary manufacturing processes, road traffic and shipping. During the same period, ammonium emissions reduced only slightly from 304 kt to 272 kt, since emissions from fertilizer applications were largely unchanged. The above highlights the shift of pollutant emissions from large point sources to more localised sources such as major roads and intensive agriculture. During the ECN monitoring period, there was a widespread decline in atmospheric sulphur emissions across Europe (Aas et al., 2019). Between 2005 and 2019, emissions of SOx and NOx fell by 76% and 42% respectively, largely due to specific caps on energy, industry and transport sectors, while NH_3 emissions (90% agricultural) reduced by 8% only (European Environmental Agency, 2021).

3.2. Rainfall chemistry signatures based on lamb weather types

3.2.1. Concentrations of major ions

There is a strong relationship between the concentration of major ions and the number of days in the week represented by Lamb westerlies over time (Fig. 3). Differences in concentrations between the two LWT groups are apparent for most ions at most sites, although this is less apparent for Glensaugh. At the remaining sites, the effect is apparent through consistently increasing or decreasing concentrations with increasing number of westerlies within each year group. Median concentrations of pollutant ions mostly decreased with the number of westerlies. Nitrate and ammonium concentrations in the 0–2 westerly day class are clearly higher than more westerly dominated classes. These relationships are observed relatively consistently through time despite large reductions in some pollutant ion concentrations over time, particularly for xSO_4^{2-}.

For ions of predominantly seasalt origin, concentrations generally increase with the number of westerlies. Again, however, LWT effects at Glensaugh are least clear. Chloride and Na^+ ion concentrations decreased slowly over time at Snowdon and Rothamsted, while remaining relatively stable at Moor House and Glensaugh. Our analysis

Fig. 2. (a) NOx and (b) SO_2 emissions map from UK large combustion plant emissions with emissions greater than 10,000 tonnes per year in 2005. The size of the bubble is proportional to emissions amount. The red and purple bubble indicate the emissions in 2005 and 2015 respectively. The time series between 2004 and 2017 are provided in Fig. SI 2. ECN site names are enclosed by a box. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

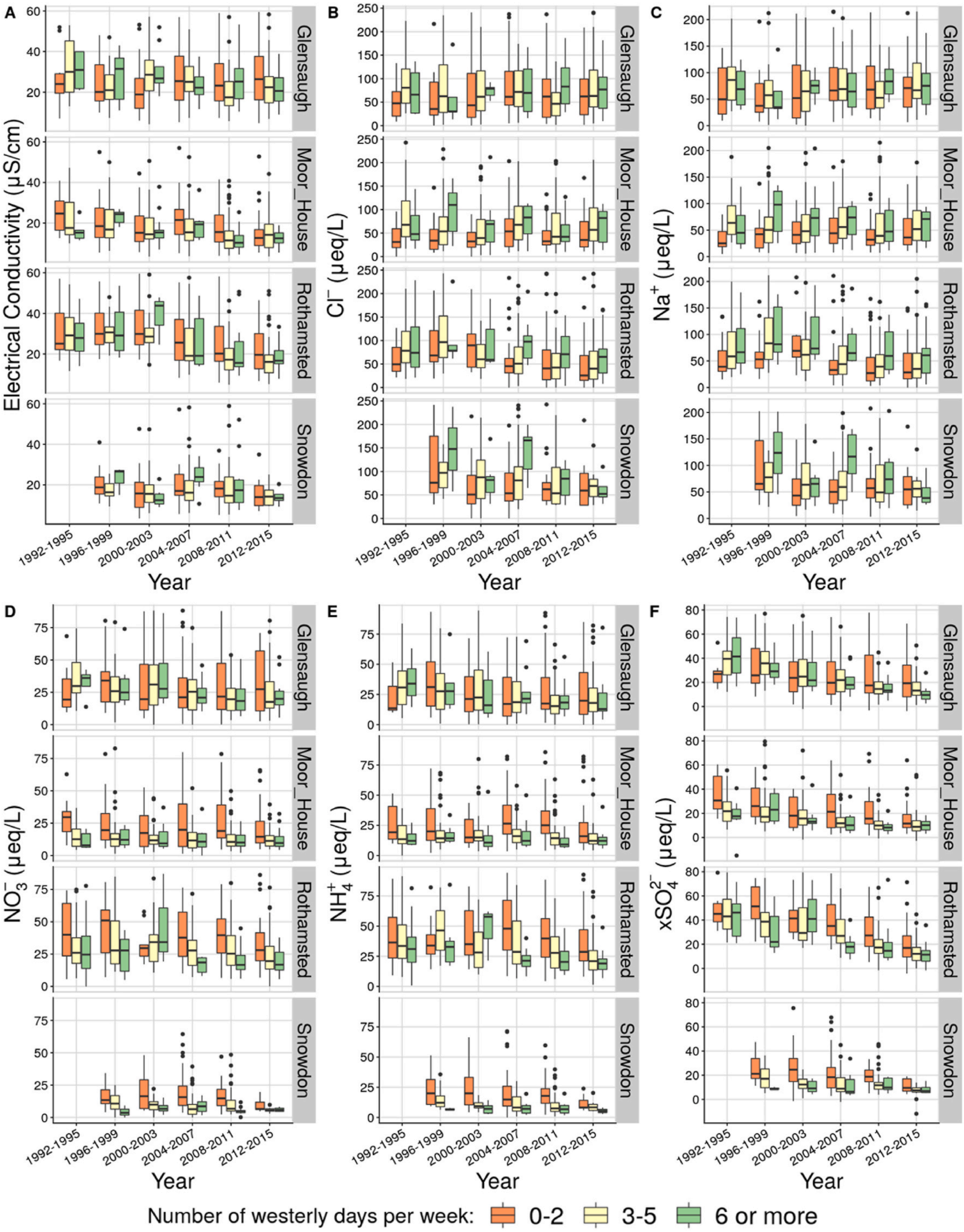

Fig. 3. The evolution of concentrations of major ions in precipitation at four ECN sites grouped by the number of days dominated by westerlies (LWT = C, W, SW, CW, or CSW) per week. Samples were generally collected at weekly intervals and each lettered panel represent one of the ions. The whiskers extends up to 1.5 times of the interquartile range beyond the upper and lower hinges.

provided little indication of influences of LWTs on EC, although long term reductions in EC were apparent at all sites other than Glensaugh. It would appear that the opposing influences of LWTs on pollutants and seasalts have largely cancelled out effects on total ionic concentrations. For all sites, up to 70% of westerlies air masses (Fig. 1) arrive in the UK in the southwest quadrant, in contrast to 20–40% for the other LWTs. Both Glensaugh and Rothamsted are located in the east of the country, with Rothamsted being an urban site. Glensaugh is a rural site but its southwest quadrant includes population centres such as Edinburgh and Glasgow, which may contribute to the more subtle LWT effects observed there.

The linear model fits for ionic concentration versus year and number of westerlies (Table SI 1 and Table SI 2, the latter including an additional interaction term) were used to quantify the trends observed in the boxplots. Chloride and Na^+ concentrations increased significantly with the number of westerlies at Moor House only, and decreased significantly over time at Rothamsted only ($p < 0.001$). Concentrations of most pollutant ions decreased significantly with both westerly counts ($p <$

0.001) and year ($p < 0.05$) at most sites other than Moor House where NO_3^- and NH_4^+ concentrations increased slightly (note the low intercepts), and Rothamsted, where the reduction in xSO_4^{2-} with respect to westerly counts was large but not statistically significant.

Table SI 2 shows the fit with the addition of an interaction term which is used to test for evidence that the relationship between ion concentrations and LWTs is changing significantly over time. The sign of the coefficients for the westerly count and year terms is less consistent than in the previous analysis (Table SI 1), but the effects are compensated by the interaction term. The coefficients for the intercept and year terms at Moor House and Rothamsted is significant ($p < 0.05$) for most species. Only a few of the site-ion combinations show a significant interaction term in the model (e.g. xSO_4^{2-} at Snowdon and Rothamsted). However, likelihood ratio tests show that all model fits (with the only exception of seasalt at Snowdon) improve significantly ($p < 0.001$) with the addition of the interaction term. The combination of these two observations indicate the relationship between westerly counts and concentration, in general, have not changed significantly

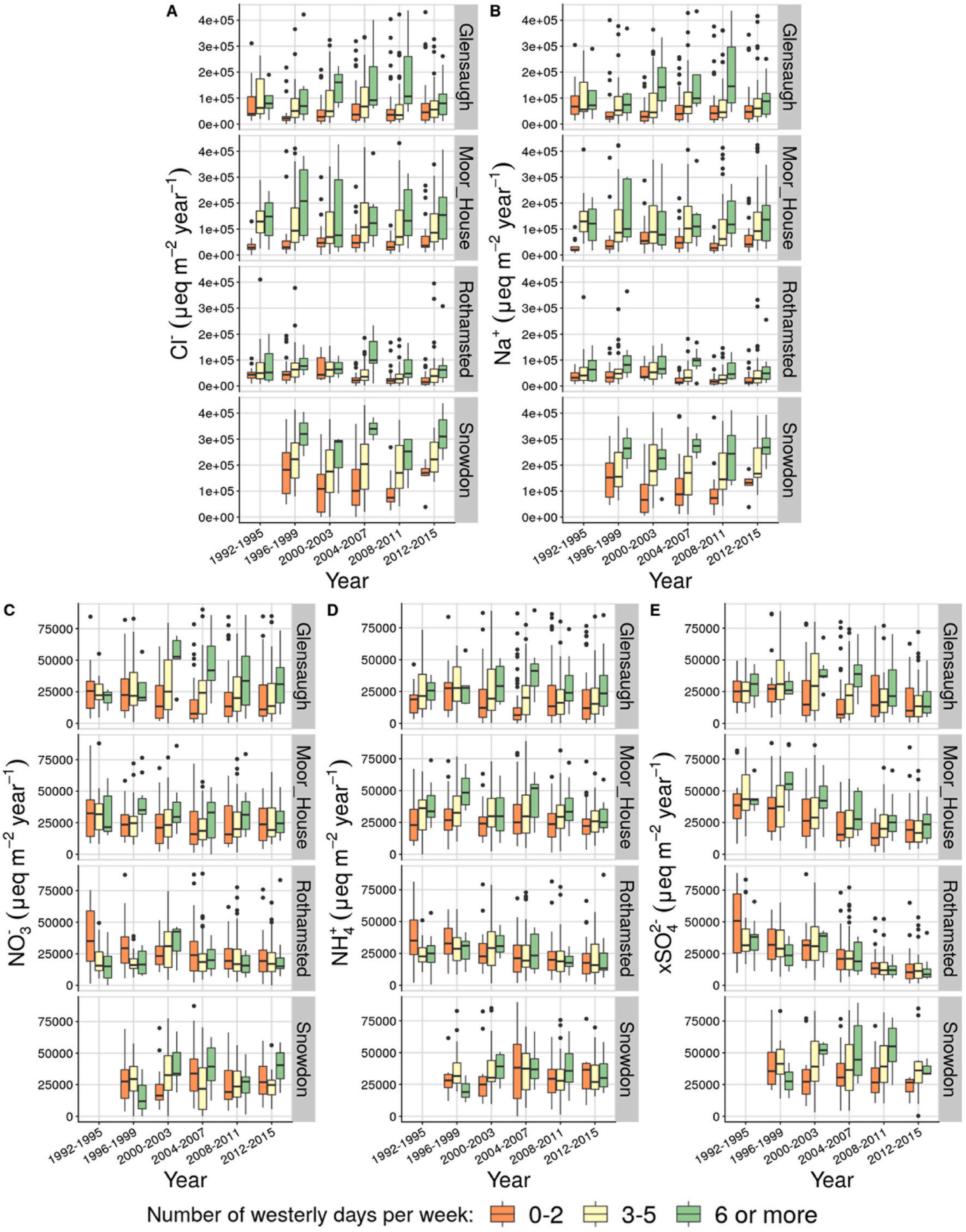

Fig. 4. The evolution of annual fluxes of major ions in precipitation at four ECN sites grouped by the number of days dominated by westerlies (LWT = C, W, SW, CW, or CSW) per week. Samples were generally collected at weekly intervals. The whiskers extends up to 1.5 times of the interquartile range beyond the upper and lower hinges.

over the ECN monitoring period studied here (1993–2015), while the addition of an interaction term between westerly counts and year terms have led to better modelled concentrations.

3.2.2. *Annual fluxes of major ions*

As a consequence of seasalt ion concentration and rainfall amount (see Fig. SI 1) both increasing with the number of days of westerlies, seasalt ion fluxes (Fig. 4) show very clear increases with the number of westerlies, both with respect to median values and the spread or interquartile range. Sea salt ion fluxes remained relatively constant for each group throughout the monitoring period at each site. The decline in pollutant ion concentrations with the number of westerlies might be expected to counteract the influence of increasing precipitation on the total major ion flux. However, overall, pollutant fluxes also tended to increase with the number of westerlies, demonstrating the dominant influence of the amount of rainfall. A notable exception was Rothamsted (especially prior to 2000), where (and when) pollution levels were high but fluxes were relatively low as a consequence of the relatively low rainfall in south-east England. Sharp reductions in xSO_4^{2-} fluxes were observed while the reductions in NO_3^- and NH_4^+ fluxes were less rapid. As air quality is projected to continue to improve in future, pollutant fluxes will become increasingly dominated by the amount of rainfall, and hence most associated with cyclonic and westerly days.

3.3. *Occurrences of different lamb weather types and their implications on changing climate*

Our analysis of weekly rainfall chemistry trends from 1993 to 2015 across a number of UK locations revealed significant differences in the chemistry of precipitation between the two LWT groupings. Weeks with no westerlies yielded the highest pollutant concentrations, and lower concentrations of ions derived largely from seasalt. Large reductions in pollutant ion concentrations over time, particularly with respect to xSO_4^{2-}, reflect changes in UK energy strategy and the implementation of national and international regulations on emissions over the monitoring period, such as the Industrial Emissions Directive, the Large Combustion Plant Directive and Euro standards for vehicles. Differences in concentrations between LWT groups remained apparent throughout the monitoring record, although there was a general tendency in recent years (i.e. post-2010) for xSO_4^{2-} concentration ranges in the 3–5 and >5 westerly classes to increasingly overlap. For the ions predominantly of seasalt origin, concentrations generally increased with the number of westerlies, consistent with the importance of westerly weather in the delivery of seasalt across the British mainland. Progressive reductions in Cl^- at Rothamsted are likely to be indicative of a reduction in the contribution from HCl, resulting from controls on the burning of coal with a high chlorine content (Evans et al., 2011).

As pollutant levels decline to levels not encountered for a century, and since weather patterns are vulnerable to climate change, it is reasonable to ask whether future climate change is likely to ameliorate or exacerbate residual pollutant concentrations and fluxes. In order to address this, we first considered whether the relative occurrence of westerly weather types had changed directionally over recent decades. Daily records of LWTs date back to 1871. Fig. 5a presents a time series of the number of cyclones and westerlies (including their LWT sub-types) per year between 1871 and 2020. This demonstrates that there has been a substantial increase in the prevalence of these weather types since the 1950s, but earlier data demonstrates that the longer term trend has not been monotonic. A changepoints algorithm was used to define segments with distinctively different means and variances. This identified six discrete periods over the last 150 years. The second (1874–1909) and fourth segments (circa 1954–1975) have a mean of 127 westerly

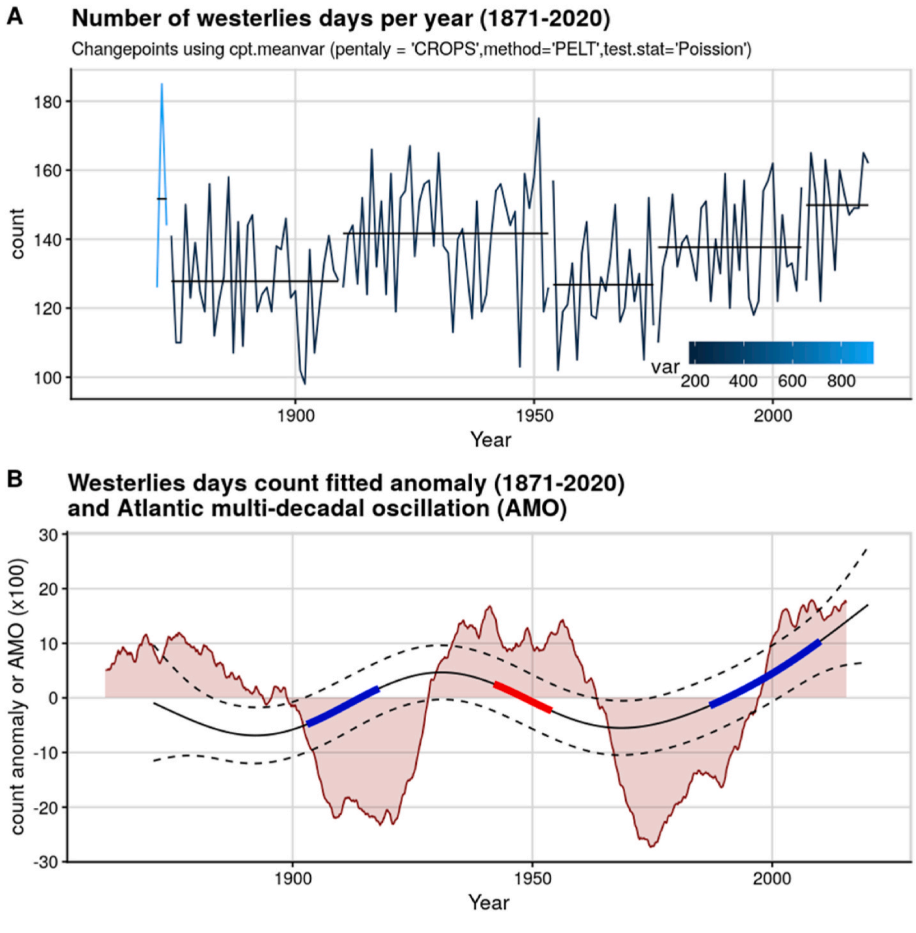

Fig. 5. A time series showing the number of westerlies (LWT = C, W, SW, CW, or CSW) per year from 1871 to 2020. (a) A changepoint analysis is applied to the time series for segment changes in mean and variance. The horizontal line designates the mean of each segment while the colour shows the variance of the segment. (b) The number of westerlies anomalies from 1871 to 2020 modelled by generalized additive models (GAMs). The solid line denotes the mean modelled deviation while the dashed lines shows the 95% prediction intervals based on its first derivatives. The red and blue bold lines shows periods of increase and decrease respectively based on first derivatives. The red shaded area is the Atlantic multi-decadal oscillation (AMO) during the same period. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

days per year, while the third and fifth segments (1910–1953; 1976–2006) have a mean of about 140 days. The most recent period (2007–2020) with a relatively high frequency of westerlies and cyclones (mean of 150 days) is therefore not particularly unusual in a longer term context.

Fig. 5b presents the fitted anomaly of westerly days count per year using a generalized additive model (GAM). Again, this plot highlights the multi-decadal nature of variation in the main weather groupings and the relatively persistent upward trend in the occurrence of westerlies and cyclones since the 1990s. Also overlaid on the same figure is the Atlantic Multi-decadal Oscillation (AMO) (Enfield et al., 2001; Kerr, 2000) of the same period. Although the two appear to have similar periods, they are not synchronous. Other drivers, possibly factors more local to the British Isles (e.g. NAO may be more relevant to the UK), may govern the patterns of westerly counts.

While there has been a recent tendency toward a progressively increasing frequency of westerlies, the degree to which the recent shift toward a greater prevalence of westerlies might be linked to global warming is unclear. Any reversal of the recent trend would be expected to result in lower concentration and fluxes of marine ions across the UK, and as these increasingly dominate the chemical signatures of deposition across the country this would also result in a reduction in precipitation electrical conductivity.

3.4. Limitations and alternative weather and rainfall type classification

The LWT classification approach is tailored to UK weather types and is not applicable to other geographical regions. A more flexible approach to define weather types based on k-means clustering of daily mean sea level pressure (Neal et al., 2016) has recently emerged which can potentially be used instead of LWTs for similar work.

We have also explored the use of convective rainfall fractions based on ERA5 reanalysis (Hersbach et al., 2018) as an alternative way to group rainfall samples. Overall, the concentration and flux signatures and trends based on convective rainfall fraction is less clear than those based on LWTs (see Fig. SI 3–4). This can be attributed to issues such as matching point chemistry measurements to a 31 km^2 grid, omission of orographic enhancements (Inglis et al., 1995) by ERA5, aggregating hourly convective rainfall at a grid to weekly total, and ignoring the wider weather patterns outside the model cell collocated with a ECN site.

3.5. Potential effects on soil chemistry

Changes in atmospheric chemistry exert a major bearing on the biogeochemical functioning of soils and waters in remote upland catchments particularly. Over the last two centuries, changes in the anthropogenic deposition of acids resulted first in the acidification of soils and then surface waters (Battarbee et al., 1985). A major reduction in acid emissions since the 1980s has seen a substantial reversal in soil and water acidity (Stoddard et al., 1999) and associated effects that include reductions in base cation concentrations (Weyhenmeyer et al., 2019) and large increases in the solubility of soil organic matter and concentrations of dissolved organic matter concentrations in waters (de Wit et al., 2021; Monteith et al., 2014). As acid deposition levels begin to return toward pre-industrial levels, ionic concentrations in more remote waters, particularly in lakes and streams in western Britain, are increasingly becoming dominated again by fluctuations in the deposition of seasalts. Soil organic matter dynamics, and the related browning of surface waters by dissolved organic matter, continue to be highly sensitive to changes in the ionic strength of soil water resulting from reductions in the ionic strength of deposition (Hruška et al., 2009; Lawrence and Roy, 2021; Monteith et al., 2007). Future variation in upland soil and water biogeochemistry will therefore become increasingly dependent on the effects of changing weather patterns on seasalt deposition (Lee et al., 2020), with periods of relatively low seasalt inputs resulting in higher soil organic matter solubility and hence higher concentrations of dissolved organic matter – an issue of potential concern to UK water companies dependent on upland drinking water sources (Ritson et al., 2014). Advancing our understanding of how changes in climate might be expected to moderate future atmospheric deposition patterns is therefore highly relevant to the monitoring and management ecosystems. While the present study focuses on analysing the trends in rainfall chemistry observations, further work will be necessary to assess the likely implications of future shifts in climate on deposition chemistry for soil chemistry and plant communities.

4. Conclusions

North Atlantic weather types such as cyclones and westerlies have previously been shown to exert significant controls over rainfall patterns, flooding events and pollutant loadings in the British Isles. Our analysis, based on long-term weekly monitoring data collected at sites in contrasting locations across the UK over the past 25 years demonstrates that weather types have continued to exert a significant influence on precipitation chemistry until close to the present day, despite the fact that atmospheric pollution loads have declined dramatically over this period. We observed strong links between the frequency of westerlies and both sea salt ion concentrations and (decreasing) pollutant ion concentrations, with their extent strongly controlled by air trajectory paths. Since cyclones and westerlies are associated with higher rainfall, they are associated with higher overall fluxes of sea salts and, in most cases, pollutant ions. As precipitation chemistry across the UK becomes increasingly dominated by marine, as opposed to pollutant, ions, fluctuations in total ion fluxes will become increasingly sensitive to variation and any long-term shift in dominant weather types. Our findings therefore demonstrate the continuing importance of weather in influencing precipitation chemistry and hence upland soil and water biochemistry to future changes in climate associated with the impact of long-term global warming. This work also highlights the benefits of long-term collocated ecological monitoring (e.g. ECN, LTER-Europe and ILTER networks) as well as adding contextual information by incorporating third-party datasets.

Data availability

The ECN data are available at the Environmental Information Data Centre (EIDC), a UK Natural Environmental Research Council (NERC) data centre hosted by the UK Centre for Ecology and Hydrology (Rennie et al., 2020, 2017b; 2017a):

An interactive R Shiny web app to reproduce elements of this paper is hosted on: https://cptecn-sandboxdemo.datalabs.ceh.ac.uk/(Source code: Tso, 2022). This is a demonstrator to illustrate improved use of notebook technology for reproducing elements of a scientific study more readily (Tso et al., 2022).

Emissions data are obtained from the National Atmospheric Emissions Inventory (NAEI) under the Open Government License (OGL). We acknowledge Defra, BEIS and https://naei.beis.gov.uk/data as the source of the information.

Credit author statement

Michael Tso: Conceptualization, Visualization, Software, Formal Analysis, Writing- Original draft preparation. Don Monteith: Supervision, Conceptualization, Methodology, Data curation, Resources. Tony Scott, Helen Watson, Bev Dodd, M. Glória Pereira, Susannah Rennie: Data curation, Resources. Michael Hollaway, Aaron Lowther: Methodology, Supervision. Gordon Blair, Rebecca Killick, Peter Henrys, John Watkins: Supervision, Funding acquisition. Everyone: Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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