

Air pollution and its effects on lichens, bryophytes, and lichen-feeding Lepidoptera: review and evidence from biological records

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Changing air quality has been one of the most important drivers of change for bryophytes and lichens in Britain and Ireland over the 20th Century, with acidic pollutants such as sulphur dioxide having large effects on the ranges and abundances of many species. At the same time, expert amateur and professional naturalists have put enormous efforts into recording the distributions of species within these groups. These efforts have provided much evidence for declines and recoveries within the bryophytes and lichens, with species distribution data being linked to airborne pollutants in many different ways. We provide a broad overview of some of the changes that have occurred in affected species, using biological records collected by national recording schemes to illustrate the various effects of air pollution; we also review the direct and indirect impacts of air pollutants on these groups. Environmental change affecting one group of species is also likely to cascade to other groups where trophic or other relationships exist between them. Using data from the volunteer-based National Moth Recording Scheme, we provide the first evidence for an indirect association between reductions in air pollution and increases in lichenivorous moths. © 2015 The Linnean Society of London, *Biological Journal of the Linnean Society*, 2015, **115**, 611–635.

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AIR POLLUTION: A SHORT REVIEW

Across the globe, both the extent and the impacts of air pollution are highly variable (Hill, 2010). For some pollutants in some areas, the outlook is not positive, and many currently detrimental trends are predicted to continue or worsen (UNEP, 2007, 2012). These impacts are also spread unevenly across species and ecosystems. Many pollutants, such as particulate matter, heavy metals, and ozone, have severe impacts

on human health, particularly in the poorest areas of the world (Hill, 2010; UNEP, 2012); others, including nitrogen and sulphur deposition, impact ecosystem functioning, and may interact with physical changes, such as the warming of the Earth's climate, to exacerbate these effects (Millennium Ecosystem Assessment, 2005). The present review focuses on the impacts of a subset of air pollutants that have had, or are having, large impacts on the populations of lichenized fungi, bryophytes, and associated Lepidoptera in Britain since the Industrial Revolution.

The air pollutants that are addressed in the present review are sulphur dioxide (SO₂), nitrogen oxides

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(NO_x) and ammonia (NH₃); the UK has committed to reducing its emissions of all these pollutants under the European Union (EU) National Emissions Ceiling Directive (Directive 2001/81/EC) and the 1999 'Gothenburg Protocol', which also currently includes several European non-EU member states, Canada and the USA (<http://www.unece.org/env/lrtap/>). In the UK, emissions of these pollutants have been declining (Fig. 1), although this is not the case for most developing countries (UNEP, 2012). The historical impacts

of pollutants such as SO₂ in the UK, and the ongoing recovery from these, may therefore be of use for the development of biological indicators of air pollution in the developing world (Nimis, Scheidegger & Wolseley, 2002), and for a more general understanding of the changes that naturalists and ecologists in those countries may be seeing in their natural environment, as well as being a fascinating and positive story of environmental improvement in the UK. To more thoroughly understand the past and future biological

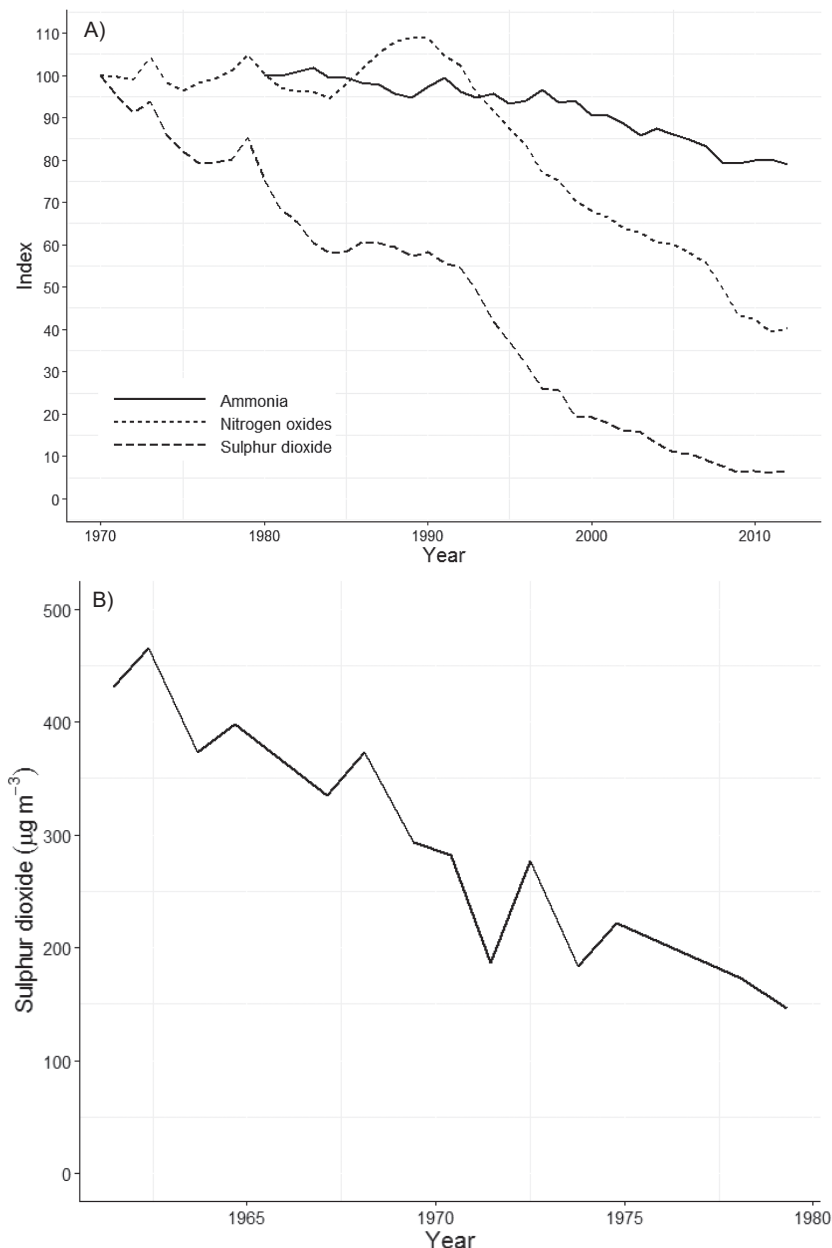


Figure 1. A, relative national trends in three air pollutants; data from Defra (2013). NO_x and SO₂ trends are relative to 1970; the NH₃ trend is relative to 1980. B, historic SO₂ trend in an urban centre (Westminster, London), redrawn from data presented in Rose & Hawksworth (1981).

impacts of the air pollutants under scrutiny, a brief review of their physical forms, atmospheric chemistry, and emission sources is provided.

SO₂

An acidic, corrosive gas that is a major air pollutant around the world; historically, it has been, along with particulate matter, the main component of air pollution as a result of the burning of fossil fuels (WHO, 2006). In the atmosphere, the gas SO₂ is oxidized, through the intermediate sulphite ion stage (SO₃⁻²), to produce liquid sulphuric acid (H₂SO₄) and solid sulphate (SO₄⁻²) aerosols, the latter often through reaction with ammonia gas (NH₃). It is these compounds that produce the acidifying effects of SO₂; these may be realized through washout in rain (wet deposition), or through direct, dry deposition onto surfaces (Hill, 2010). The aerial lifetime of the gas SO₂ is estimated to be only of the order of a day; however, sulphuric acid and sulphate have much longer residence times in the atmosphere, and it is these compounds that produce many of the impacts on ecosystems and species through acidic deposition (Hill, 2010). Sulphites can also be highly toxic to many cryptogamic (i.e. nonflowering) species (Lee & Studholme, 1992), although these ions may largely be generated on or within plants or lichens from deposited SO₂. The main sources of SO₂ in the UK in 2012 were from burning in energy production and transformation (66%), followed by combustion in manufacturing industries (14%; Defra, 2013; Salisbury *et al.*, 2014; <http://naei.defra.gov.uk/reports>); the massive reduction in UK emissions from 1970 (6.37 million tonnes) to 2012 (0.43 million tonnes; displayed in relative terms in Fig. 1A) has mainly been driven by a change in fuel use from coal to gas, and by the fitting of flue gas desulphurization units in the power industry (Defra, 2013).

NO_x

The chemical symbol NO_x represents the nitrogen oxides, a group of gases of which the most important here are nitric oxide (NO) and nitrogen dioxide (NO₂); NO is converted rapidly into NO₂ on exposure to air. Nitrogen oxides can also be converted into nitrate (NO₃⁻) and nitric acid (HNO₃) in the atmosphere, again forming aerosols that are significant components of acid deposition (RoTAP, 2012). Both nitrate and nitric acid can also contribute to the eutrophication of certain sensitive habitats (RoTAP, 2012). As with oxides of sulphur, these derived aerosols of NO_x have longer residence times in the atmosphere than their precursor gases (UNEP, 2007). The two major sources of nitrogen oxides in the UK in 2011 and 2012

were road transport (350 000 tonnes; approximately 34%) and energy production (315 000 tonnes; approximately 30%) (Defra, 2013). As will be noted from the above, many of the biological impacts of NO_x are shared with SO₂ (Hill, 2010), a point that should be borne in mind when using information on their prevalence in correlative ecological analyses. Despite a large decline in NO_x emissions in the UK overall (Fig. 1A) (Defra, 2013), roadside monitoring of the levels of NO₂ in London and other cities indicates that there has been little decline in the emissions in the urban environment over the recent past (UNEP, 2007; Stedman & Grice, 2009; GLA, 2010). For example, the World Health Organization's limit value for harmful effects on human health (40 µg m⁻³; WHO, 2006) is still regularly exceeded in these environments in the UK (RoTAP, 2012).

NH₃

Gaseous ammonia has a relatively short residence time in the atmosphere of the order of days (UNEP, 2007) but, as with SO₂ and NO_x, it undergoes reactions yielding a more stable ion (ammonium, NH₄⁺) that can form an aerosol. Reduced nitrogen can be deposited in wet or dry forms, although the dry deposition of reduced nitrogen is mostly in the gaseous form, with ammonium aerosol deposition making a smaller contribution. Emissions of ammonia are dominated by the agricultural sector in the UK, particularly the management of manure (RoTAP, 2012); however, the contribution of agriculture to total emissions of NH₃ has fallen slightly in recent years, from 89% in 2010 to 82% in 2012 (Fig. 1A) (RoTAP, 2012; Defra, 2013). This is reported to be a result of reductions in cattle numbers and more efficient fertilizer use (Defra, 2013). Because NH₃ is primarily emitted from diffuse sources, there is typically more uncertainty around the measurement of emissions than for SO₂ or NO_x; however, the available data indicate that UK emissions peaked in the early 1980s, and have decreased by approximately 20% subsequently (RoTAP, 2012; Defra, 2013). Point sources from pig and poultry farms can still have considerable impacts on biodiversity locally (Leith *et al.*, 2005) and, at the landscape scale, there is much concern that many ecosystems continue to be adversely affected across the UK (Leith *et al.*, 2005).

Other air pollutants of high importance globally include ground-level (tropospheric) ozone and fine particulate matter (Hill, 2010). Aerosols of all of the pollutants described above can contribute to particulate matter pollution of different sizes, which often present severe problems for human health (WHO, 2006). However, from a biodiversity impacts point of view, large particles of the air pollutants under

discussion in the present review are typically included as part of the phenomenon of dry deposition. Other pollutants not discussed here, such as metals or organic chemicals, can also contribute to particulate matter pollution, and these may also have their own impacts on specific taxa (e.g. Bargagli & Mikhailova, 2002). Ground-level ozone is another serious component of air pollution with considerable effects on human health (WHO, 2006); the impacts of this pollutant on crop yields are well understood (van Dingenen *et al.*, 2009) but wider biodiversity impacts have not been as well investigated as they have for sulphur and nitrogen compounds (Davison & Barnes, 1998; Ashmore, 2005), although some evidence for impacts on lichens and bryophytes has been reported (Bates, 2000; Bell & Treshow, 2002). Nitrogen oxides are important precursors of ground-level ozone (Hill, 2010), which may make it hard to separate NO_x impacts from ozone impacts in some study systems.

ENVIRONMENTAL POLLUTION, NATURAL HISTORY, AND PUBLIC ENGAGEMENT

Alongside the increasing monitoring and knowledge of pollutants and their impacts during the 20th Century, the early public face of ecology was coloured strongly by pollution-related issues (Carson, 1962; Mabey, 1974). The hugely influential advocacy of Carson's *Silent Spring* (1962) was followed in Britain by Mellanby's New Naturalist title *Pesticides and Pollution*, which was also aimed at raising the awareness of the wider reading public (Mellanby, 1967). Kenneth Mellanby was subsequently instrumental in the organization of a successful citizen science initiative, a survey of water pollution in Britain conducted by approximately 8000 school children (Mellanby, 1974). This project was part of a larger initiative organized by the Cambridge-based Advisory Centre for Education, the Nature Conservancy, and the *Sunday Times* newspaper (Mabey, 1974). As well as water pollution, the project featured several surveys of air pollution. This included a lichen-mapping exercise, again carried out by young people, resulting in an impressive national map of SO₂ pollution levels (Gilbert, 1974). The writings of Mellanby (1967) and Mabey (1974) recall a time when heavy pollution of several types was a commonplace in the British landscape: with descriptions of black to grey pollution-sullied sheep as 'ready-reckoners' of air pollution (Mellanby, 1967); of neat privet hedges becoming deciduous as a result of the depositions of soot on their leaves (Mabey, 1974); and of growers of chrysanthemums having to concern themselves with protecting blooms from spotting by air pollutants, using bags and polythene sheets (Bennett, 1968); all of

these remind us of the pervasive presence of heavy air pollution in parts of Britain at that time.

Forty years later, the accumulation of historic and contemporary species' distributional data from amateur and professional naturalists is yielding many new opportunities for the study of trends in the abundance and frequency of species in different groups, and allowing us to relate these to changes in the wider environment (Roy *et al.*, 2014; Pocock *et al.*, 2015). This legacy of 'citizen scientist'-led natural history in Britain and Ireland, and the wealth of resulting data, has been complemented by a long tradition of inference from maps (Preston, 2013), with, older, largely descriptive approaches to environmental change being supplemented over the past 20 years with new methods designed to model potential biases in biological records datasets (Prendergast *et al.*, 1993; Isaac *et al.*, 2014). The present review provides a broad overview of the evidence for air pollutant effects on lichens, bryophytes, and lichen-feeding Lepidoptera contained within biological records, presents original supporting analyses, and brings this information together with experimental findings on direct and indirect impacts for lichens and bryophytes. Although it is recognized that lichens and bryophytes are very different types of cryptogamic organisms, we treat them here together because they comprise 'pocket-sized autotrophs that occupy similar habitats' (Bates, 2002), following the lead of many other studies (During, 1992).

EFFECTS OF AIR POLLUTION ON LICHENS, BRYOPHYTES, AND LICHEN-FEEDING LEPIDOPTERA

LICHENS

Evidence from biological records over time

Lichens are perhaps best known amongst ecologists and naturalists for their value as indicators of various types of pollution (Nimis *et al.*, 2002). This indicator value will be species and pollutant dependent (Nash, 2008), and a very large amount of research effort amongst lichenologists has been dedicated to exploring these relationships (Bates, 2002). The first observation of air pollution impacts on lichens has been attributed on several occasions to Erasmus Darwin (Brightman, 1982; Hawksworth, 2002; Kricke & Loppi, 2002), and this historical starting point often features at the beginning of reviews of this topic. Erasmus Darwin wrote the following lines in his educational poem *The Botanic Garden*: 'No grassy mantle hides the sable hills,/No flowery chaplet crowns the trickling rills,/Nor tufted moss nor leathery lichen creeps/In russet tapestry o'er the crumbling steep's' (Darwin, 1791). These lines have been inter-

preted by Brightman (1982) as referring to the local impacts of the copper mines at Parys Mountain in Anglesey (North Wales), both on the strength of the scientist and writer Arthur Aikin's use of the lines above to describe the scenery on Parys during a visit there in the late 18th Century (Aikin, 1797), and on a reference to a footnote in *The Botanic Garden* stating that Darwin's lines did also in fact refer to the landscape of Parys Mountain (Brightman, 1982). Unfortunately, this footnote does not appear to exist in copies of *The Botanic Garden* that one of us (OLP) has examined, and, on reading the relevant section of the poem and the accompanying notes, it is clear that the lines quoted above actually describe a 'Traveller's Tale' concerning the purported results of a poisonous miasma emitted by the Upas tree (*Antiaris toxicaria*, Moraceae), a species found in Indonesia and throughout the Old World tropics. It is not disputed here that Aikin (1797) uses Darwin's lines on the Upas tree to describe the landscape at Parys Mountain (Brightman, 1982; Purvis, 2010); however, attributing the first observation of air pollution impacts on lichens to a fragment of poetry describing a generically desolate scene from a romantic standpoint is not very satisfactory. With interests spanning botany, chemistry, and mineralogy (Torrens, 2004), one might expect Aikin to have made more of an apparently original observation. Of course, given that the pollution on Parys was reportedly so bad that 'not a single shrub or hardly a blade of grass [was] . . . able to live in the sulphurous atmosphere' (Aikin, 1797), it is likely that Aikin did not feel that he had to spell out the parallel loss of the lower plants. The resulting quality of the observation, however, is of a rather different type to that of later observers, such as William Borrer (Laundon & Waterfield, 2007), who explicitly described the loss of lichens under 'smoky' urban conditions. The first clear observation of pollution impacts on lichens, therefore, shifts to Borrer, who, with Dawson Turner in his *Specimen of a Lichenographia Britannica* (Turner & Borrer, 1839), clearly comments on the prevalence of '*Lepraria viridis*' (almost certainly the alga *Desmococcus olivaceus*) where 'the air is so impure that scarcely any other Lichen can exist'. This observation was apparently first penned sometime around 1807 (Laundon & Waterfield, 2007).

Histories of the impacts of the Industrial Revolution on both bryophytes and lichens have, however, been reviewed or overviewed elsewhere on numerous occasions (Hawksworth, 1971, 1973, 2002; Gilbert, 1973; Seaward & Letrouit-Galinou, 1991; Adams & Preston, 1992; Bates, 2002) and we do not repeat them again here. The development of a conceptual framework under which to organize the observations of earlier naturalists, namely the idea of a clear and

quantifiable relationship between assemblages of lichens and zones of pollution, was first put forward by the Scandinavian Sernander (Sernander, 1912), and subsequently developed by numerous other continental lichenologists, before being taken up by Eustace Jones in Britain (Hawksworth, 1973). The work of Jones (1952) is perhaps most notable for the thorough attempt at finding the best 'indicator' metric of changes in lichen assemblages along pollution gradients (at this time, the emphasis was still on smoke pollution, the link with SO₂ not having been fully elucidated; Adams & Preston, 1992); different measures such as overall lichen cover, lichen overall frequency, and species richness were all investigated for different functional groups. The transects of Jones (1952) clearly demonstrated gradients of lichen richness and cover around SO₂ pollution sources in the British landscape for the first time (Fig. 2), an innovation that was soon to take off in lichenology (Hawksworth, 2002). However, rather unexpectedly given the long-standing emphasis on mapping lichens in relation to air quality, vascular plants actually appear to furnish the first example of national, hectad (10 × 10 km) species distribution maps being using to interpret potential air pollution impacts: Bowen (1965) used maps of juniper (*Juniperus communis*) to infer impacts of SO₂ on that species, a hypothesis that does not appear to have been followed up with experimental work. However, Hedgcock (1912) reported *J. communis* as the most resistant of eight conifer species in the proximity of the Washoe Anaconda copper smelter in Montana, USA, with the species showing 'little or essentially no injury from smelter fumes', an observation no doubt unavailable to Bowen (1965).

Numerous authors have subsequently used hectad maps of biological records in reviews to demonstrate the thesis that particular lichen species or genera have declined, or declined and then recovered, over the 20th Century (Seaward & Hitch, 1982; Brown, 1996; Seaward, 1998; Coppins, Hawksworth & Rose, 2001; Bates, 2002; Nash, 2008; Purvis, 2010). In all these cases, however, the commentaries have been based on largely subjective estimates of distribution limits under SO₂ pollution, or on the visual comparison of maps made at different time points. This approach can be more flexible than indicated by these reviews: depending on the temporal and spatial resolution of the biological records in question, such data are used in increasingly creative ways (Isaac *et al.*, 2014). For example, here, using the British Lichen Society's (BLS) Mapping Scheme and Lichen Database datasets, we make a comparison of hectad records for the periods 1900–1979 and 1980–2013, and relate this to an index of peak SO₂ concentrations. For a given species in this analysis, each hectad

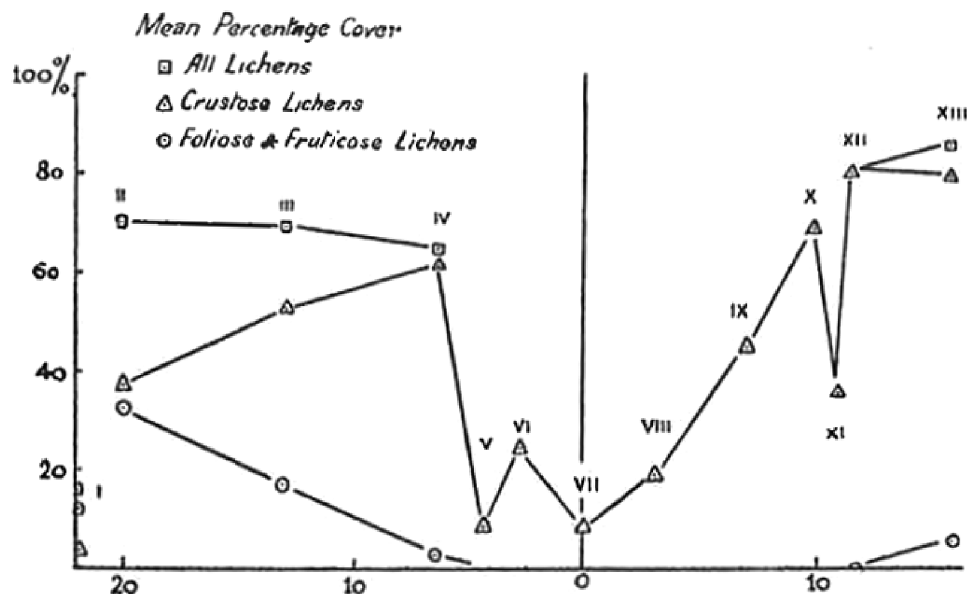


Figure 2. The results of a transect from Ludlow (Site I), on the left hand side of the graph, via Walsall (Site VII; 'no conifers thrive here, and even many deciduous trees are not prosperous' Jones, 1952), to Wootton Wawen (Site XIII) near Stratford-upon-Avon [about 13 miles (21 km.) from the nearest sources of smoke']. Adapted from Jones (1952).

was classified into one of three classes: gain (hectad gained between the two time periods); loss (hectad lost between time periods); and stable (hectad presence maintained between time periods); note that maintained hectad absences were excluded. The resulting categorizations were used in a multinomial logistic model (Agresti, 2002). Annual mean SO_2 concentrations for the year 1970 were calculated with the FRAME atmospheric chemical transport model (Dore *et al.*, 2007; Matejko *et al.*, 2009). This model uses emissions estimates of SO_2 , NO_x , and NH_3 from the UK National Atmospheric Emissions Inventory (<http://naei.defra.gov.uk/reports/>) to simulate dry and aqueous phase atmospheric chemical reactions and to subsequently calculate the dry and wet deposition of sulphur and nitrogen compounds using transport models. The resulting SO_2 concentrations (Fig. 6A) were used as a covariate (aggregated to 10×10 km) in species' multinomial logistic models. The modelled relationships give the probabilities of category membership as a function of the SO_2 concentration in 1970, which we use as an index of historic pollution load per hectad (Fig. 3). An SO_2 sensitive species that has subsequently recolonized large areas of previously polluted territory should show an increasing relationship for 'gain' versus SO_2 concentration because it is those areas that are most likely to have been recolonized after pollution-inflicted losses. Figure 3 demonstrates this for one genus (*Usnea*) and two species (*Hypogymnia physodes*, *Lecanora conizaeoides*) long known to exhibit differing sensi-

tivities to acidic pollution (Hawksworth & Rose, 1970). These species have been chosen not only to demonstrate the uses of a simple hectad mapping approach to distributional change, but also to highlight its limitations.

Usnea, a genus of epiphytic fruticose lichens, has often been used as an example of the large effects of SO_2 on the national distribution of sensitive species (Hawksworth, Rose & Coppins, 1973; Seaward, 1998). The map presented here (Fig. 3A) clearly demonstrates the partial infilling of the range between the two periods examined; the losses in the north and west of Britain may be more likely to be the result of uneven recording between periods. The predicted probability graph for *Usnea*, showing the probability that a hectad was lost, gained, or stable as a function of SO_2 concentration, demonstrates that gain was the most likely state above $26 \mu\text{g m}^{-3}$; hectads with this average level of pollution are most likely to have been without *Usnea* spp. in the period 1900–1979, and to have been recolonized subsequently. This value is close to the $35\text{--}40 \mu\text{g m}^{-3}$ SO_2 concentration given by British field scales as the tolerance level for this genus (Hawksworth & Rose, 1970). Comparison with the less SO_2 -sensitive foliose lichen *Hypogymnia physodes* (Fig. 3B), a species that is given an approximate tolerance of approximately $70 \mu\text{g m}^{-3}$ on the Hawksworth–Rose scale, shows a higher crossing point at which the probability of gain over the other two categories becomes most likely (approximately $60 \mu\text{g m}^{-3}$). Indeed, for the set of common, variously

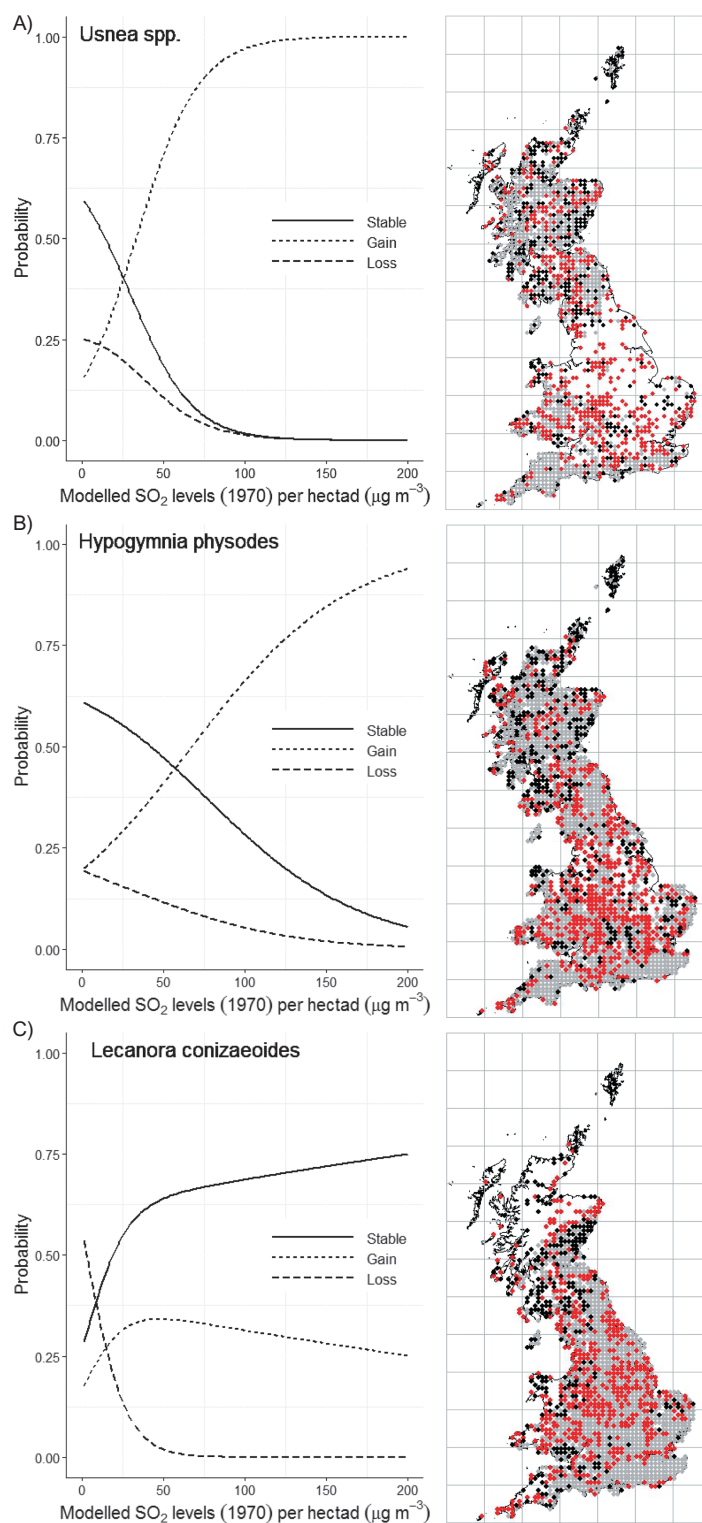


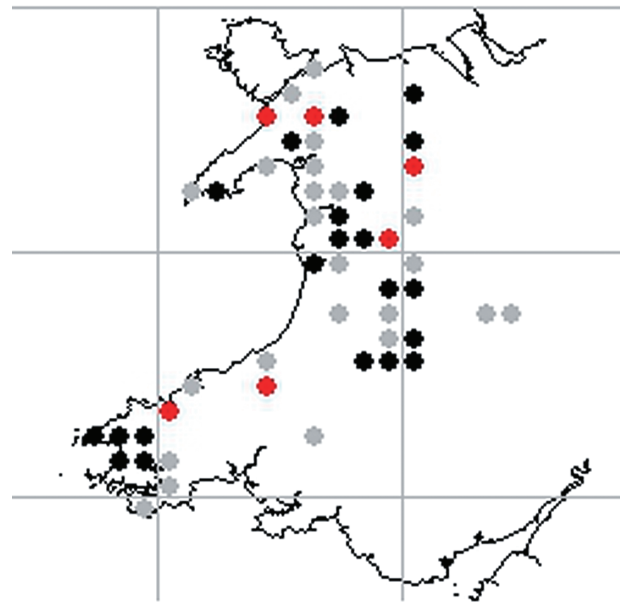
Figure 3. Predicted probability graphs and the hectad distributional data modelled for *Usnea* spp. (A), *Hypogymnia physodes* (B), and *Lecanora conizaeoides* (C). For the distribution maps: grey, stable; red, gain; black, loss between the time periods 1900–1979 and 1980–2013. The graphs were derived by solving the multinomial logistic regression equation for each species for unit increases (1–200 $\mu\text{g m}^{-3}$) in SO₂ concentration per hectad.

Table 1. Modelled SO₂ concentrations at which the gain of a species became more likely than either loss or stability, between the periods 1900–1979 and 1980–2013

Species	SO ₂ concentration (µg m ⁻³) where $P(\text{gain}) > P(\text{loss OR stable})$	First appears on Hawksworth & Rose (1970) scale (scale point and concentration)
<i>Hypogymnia physodes</i>	58	4: ~70 µg m ⁻³
<i>Parmelia sulcata</i>	56	4: ~70 µg m ⁻³
<i>Parmelia saxatilis</i>	42	4: ~70 µg m ⁻³
<i>Evernia prunastri</i>	37	5: ~60 µg m ⁻³
<i>Ramalina farinacea</i>	30	5: ~60 µg m ⁻³
<i>Lecanora chlarotera</i>	25	5: ~60 µg m ⁻³
<i>Usnea</i> spp.	26	7: ~40 µg m ⁻³

sensitive species listed in Table 1, the predicted SO₂ extinction/recolonization point derived from the comparison of the two broad hectad date classes puts the species into almost exactly the order given by the Hawksworth–Rose scale; the estimates given here are lower than the Hawksworth–Rose scale (Table 1), although other correlative work has also indicated that this scale generally overestimates a species' SO₂ tolerance (van Dobben & ter Braak, 1999). The model of hectad distribution change for *Lecanora conizaeoides* (Fig. 3C), a highly SO₂ tolerant species that spread under acidic pollution (Bates, 2002), suggests that this species has generally remained stable between the two periods analyzed, except in historically unpolluted areas, where loss is the most likely condition; however, this may be a result of under-recording in the Scottish Highlands (Fig. 3C). In this case, the model may be less accurate given that field lichenologists have typically reported severe declines of *Lecanora conizaeoides* in previously highly polluted areas (Gilbert, 1992; Purvis, 2010; Hauck *et al.*, 2011), and that analyses of biological records using different time periods have indicated recent declines at the hectad scale (Simkin, 2012).

The impacts of pollutants on the uplands should also be highlighted here: despite the separation of the uplands from the main historic sources of heavy industrial pollution in Britain (Fig. 6B), clean-air legislation in the 1950s and 1960s led to the introduction of taller chimneys in an attempt to reduce local pollution impacts. Unfortunately, these tall stacks efficiently exported pollutants as aerosols to areas, such as the uplands, that had not previously experienced the effects of SO₂ or NO_x deposition (Farmer, Bates & Bell, 1992). The resulting acidic wet deposition has been linked to the declines of lichens in upland *Lobarion* communities on several occasions (Day, 1985; Gilbert, 1986). Parts of Wales (e.g. the Cambrian Mountains) have been amongst the worst affected places in Britain (Gee & Stoner, 1989), and the BLS datasets referred to above indicate the loss of

**Figure 4.** Changes in the distribution of *Lobaria* spp. (*Lobaria amplissima*, *Lobaria pulmonaria*, *Lobaria scrobiculata*, and *Lobaria virens*) in Wales between the time periods 1900–1979 and 1980–2013 (BLS data); grey, stable; red, gain; black, loss.

Lobaria spp. from a number of locations across Wales (Fig. 4). Although the effects of acidification in the uplands have lessened in recent years (RoTAP, 2012), some field lichenologists consider that the long-range transport of acidifying pollutants is still affecting sensitive lichen communities (Sanderson, 2014). The BLS are currently running a *Lobarion* survey, where biological recorders are encouraged to visit previously known locations of key species, and it will be of great interest to see whether additional recoveries, or new locations, are reported in the coming years.

Future work on the use of biological records for this type of analysis would ideally capture information on the recorder effort that went into producing the records analyzed (Hill, 2012; Isaac *et al.*, 2014), and

would also benefit from analysis at finer temporal and spatial scales where records allow. However, a historic focus on summarizing data at the hectad-broad date-class level (i.e. a lack of separation between the act of biological recording and the use of data for hectad mapping) has meant that much lichen data may not be suitable for the types of more detailed trend analyses that have recently been applied to bryophytes (see below). Additional covariates can be incorporated into logistic models, and partial responses to particular covariates calculated (Hames *et al.*, 2002); this could potentially allow for changes in SO₂ to be modelled alongside changes in other pollutants. Stevens *et al.* (2012) used a similar approach to analyze terricolous lichen hectad occurrences for all records within a single date class, incorporating indicators of SO₂ and total nitrogen-deposition into a single model. Although Stevens *et al.* (2012) reported the existence of numerous species showing negative relationships with nitrogen-deposition at this scale, the complexities of the changing pollution environment experienced by lichens over the past half-century, exerting its influence at different scales and with largely unknown interactive effects, may mean that, in some cases, finer scale investigations will be more successful at characterizing reliable pollutant–species relationships. For example, recent work on NH₃ and NO_x has used detailed studies across smaller scale pollution gradients to infer such indicator relationships for epiphytic species (Wolseley *et al.*, 2006; Davies *et al.*, 2007).

Direct and indirect effects of air pollutants on lichens

Much of the research into the direct impacts of air pollutants on lichens has investigated effects on photosynthesis and respiration, as well as the methods of detoxification in apparently tolerant species; much of this work has focused on SO₂ (Nash, 2008). One study investigating multiple species and SO₂ concentrations has indicated that the oxidation of toxic bisulphite to sulphate, which is subsequently leached, can account for as much as 70% of the sulphur uptake of lichens (Gries *et al.*, 1997). Nash (2008) suggests that understanding how lichens deal with the remaining sulphur is the key factor determining species differential sensitivities. Gries *et al.* (1997) documented differences in H₂S emissions between species, which were interpreted as indicating differential bisulphite detoxification abilities because H₂S is the product of the reduction of sulphite by enzymes; however, this mechanism is only considered to be a minor part of SO₂ resistance (Nash, 2008). There is also evidence suggesting that peroxidase activity is an important part of SO₂ resistance in lichens because SO₂ uptake results in the production of free radicals (Modenesi,

1993); correspondingly, several studies have now demonstrated an association between superoxide dismutase (a free oxygen radical scavenging enzyme) activity and SO₂ tolerance or sensitivity in several species (Silberstein *et al.*, 1996a, b; Deltoro *et al.*, 1999; Kong *et al.*, 1999). Regardless of the particular mechanisms underlying lichen sensitivities to SO₂, it is worth noting that the ordering of particular lichen species sensitivities, as deduced from field studies calibrated using SO₂ gauges (Hawksworth & Rose, 1970), has been largely supported by laboratory fumigations, validating the use of lichens as bioindicators of SO₂ (Nash, 1988; van Dobben & ter Braak, 1999).

The indirect effects of pollutants on lichens are also well studied. Considerable effort has been spent on investigating the relationships between bark pH and epiphytic lichen presence and abundance. Early work demonstrated an association between the amount of pH buffering provided by a host species' bark, SO₂ levels and lichen community composition (Skye, 1968); in recent years, as acidification has become less of a problem, research interest has shifted to determining the indirect effects of nitrogenous pollution on lichen communities (Wolseley *et al.*, 2006; Davies *et al.*, 2007). One general theme arising from this work is the widespread shift from acidophyte (or 'nitrogen-sensitive') dominated communities to nitrophyte (or 'nitrogen-tolerant') dominated ones, particularly near to point sources of ammonia pollution, which are generally animal husbandry-related (Wolseley *et al.*, 2006). As discussed earlier, nitrogen pollution exists in various forms, and effort has also been spent on attempting to determine the relative importance of NH₃, NH₄⁺ and NO_x to the observed changes in lichen communities. Although conclusions have varied between studies, no doubt as a result of the number of additional factors that can also control lichen communities, and because of differences in how pollutant concentrations were quantified, the current evidence suggests that NH₃-driven increases in bark pH can be a significant local driver of community change (Wolseley *et al.*, 2006) but SO₂ declines may be a stronger national factor determining recent general increases in nitrophytes (van Dobben & ter Braak, 1998, 1999). Ammonium (NH₄⁺) deposition has been associated with acidification where it has been quantified from individual tree stemflows (Mitchell *et al.*, 2005), although this relationship has often not been clear in studies using broader, modelled estimates of atmospheric pollutants, with high collinearity between predictors (van Herk, Mathijssen-Spiekman & de Zwart, 2003). As with most correlative studies of complex, multivariable systems, where issues of scale, accuracy of measurements, collinearity between variables, and nonlinear relationships may all be present,

careful modelling and interpretation of results is always required (Smart *et al.*, 2012).

BRYOPHYTES

Evidence from biological records over time

The recent publication of a new bryophyte atlas of Britain and Ireland (Blockeel *et al.*, 2014) has allowed a much clearer insight into the changes that have taken place over the past half century. The first national bryophyte atlas (Hill, Preston & Smith, 1991) already revealed the beginnings of a recovery in the populations of some epiphytes (Proctor, 1991; Adams & Preston, 1992), now, the considerable increase in the number of records collected, and better geographical coverage (Preston, 2014; Preston & Rorke, 2014), has allowed a comprehensive and robust analysis of change in the new atlas (Hill & Preston, 2014). Hill & Preston (2014) applied a new model to the new atlas dataset, Frescalo ('FREquency SCAling LOcal'), formulated to deal with geographical variation in recorder effort (Hill, 2012), and also analyzed the relative change in the proportion of species-hectad occurrences out of all such occurrences for individual species between 1960–1989 and 1990–2013 using chi-squared goodness-of-fit tests. This latter test is essentially a simple measure of a species relative change between periods: if a species has neither increased nor decreased its distribution, it can be assumed that the number of hectad-level occurrences accounted for by a species will be a fixed proportion of all such records made, even if the absolute number of species-hectad occurrences increases or decreases (i.e. even if there is an overall shift in recording effort; Ball *et al.*, 2011; Hill & Preston, 2014; Isaac *et al.*, 2014). These analyses showed that epiphytes as an ecological group had, by a considerable margin, undergone the most dramatic change out of any of the groups analyzed. Overall, 28 species of epiphyte were found to have significantly increased between the two time periods (Table 2) (Hill & Preston, 2014). Figure 5 provides a coincidence map of richness for these species, strikingly demonstrating the result of the late 20th Century recovery in epiphyte diversity. Studies focusing on reviewing or demonstrating the recovery of epiphytes for more restricted localities have also increased in frequency [Epping Forest and nationally: Adams & Preston (1992); Sheffield area: Lake & Egan (2007); London: Duckett & Pressel (2009); Lancashire: Lowell (2009)]; all of these discuss increases and recoveries in their areas. A number of bryophyte Floras have also provided commentary on the effects of air pollution on a variety of taxa (Adams, 1974; Gardiner, 1981; Jones, 1991; Bates, 2005; Sanford & Fisk, 2010; Boon & Outen, 2011). For example, in Suffolk, '[s]pecies like

Table 2. Epiphytic bryophytes that significantly increased between the periods 1960–1989 and 1990–2013 (Hill & Preston, 2014)

L: <i>Cololejeunea minutissima</i>	M: <i>Orthotrichum pulchellum</i>
L: <i>Colura calyptrifolia</i>	M: <i>Orthotrichum pumilum</i>
L: <i>Frullania dilatata</i>	M: <i>Orthotrichum stramineum</i>
L: <i>Metzgeria fruticulosa</i> s.l.	M: <i>Orthotrichum striatum</i>
L: <i>Metzgeria furcata</i>	M: <i>Orthotrichum tenellum</i>
L: <i>Microlejeunea ulicina</i>	M: <i>Platygyrium repens</i>
L: <i>Radula complanata</i>	M: <i>Pylaisia polyantha</i>
M: <i>Cryphaea heteromalla</i>	M: <i>Syntrichia latifolia</i>
M: <i>Daltonia splachnoides</i>	M: <i>Syntrichia papillosa</i>
M: <i>Dicranum tauricum</i>	M: <i>Syntrichia virescens</i>
M: <i>Orthotrichum affine</i>	M: <i>Ulotia calvescens</i>
M: <i>Orthotrichum diaphanum</i>	M: <i>Ulotia crispa</i> s.l.
M: <i>Orthotrichum lyellii</i>	M: <i>Ulotia phyllantha</i>
M: <i>Orthotrichum obtusifolium</i>	M: <i>Zygodon conoideus</i>

L, liverwort; M, moss.

Cryphaea heteromalla, *Orthotrichum lyellii*, *Ulotia phyllantha* and *Frullania dilatata*, which fifteen years ago were exciting finds, are now almost commonplace' (Sanford & Fisk, 2010); whereas, in the Sheffield area, '[o]ne very good indicator of the improvement in conditions for epiphytes is the moss *Orthotrichum affine* which was seen in the South Yorkshire region only on rare occasions in the late 1990's and then mostly in the damp areas of the river valleys. Some six or seven years later there are now records for this species in over 120 tetrads [2 × 2 km squares] and it is recorded, seemingly, almost every time a new tetrad is visited, given that a suitable habitat is available' (Lake & Egan, 2007). Perhaps most encouragingly, the highly sensitive moss *Antitrichia curtipendula* (Adams & Preston, 1992) is also beginning to show signs of a recovery (Preston & Blockeel, 2014).

Finally, we review two examples where interesting trends have been displayed at the hectad level over the past 55 years; these are species for which air pollution drivers of change are considered to be highly likely. In these examples, the hectad occurrence information has been separated into high SO₂ and low SO₂ regions; Frescalo time trends were then generated separately for these two regions (Hill & Preston, 2014). The high SO₂ zone was defined as that where the mean atmospheric concentration of SO₂ exceeded 3.3 p.p.b. (~8.8 µg m⁻³ according to current Defra equivalence guidelines) in 1986 (Fowler *et al.*, 2007), and is mapped in Figure 6B. The first example is for two common and widespread epiphytes: *O. affine* and

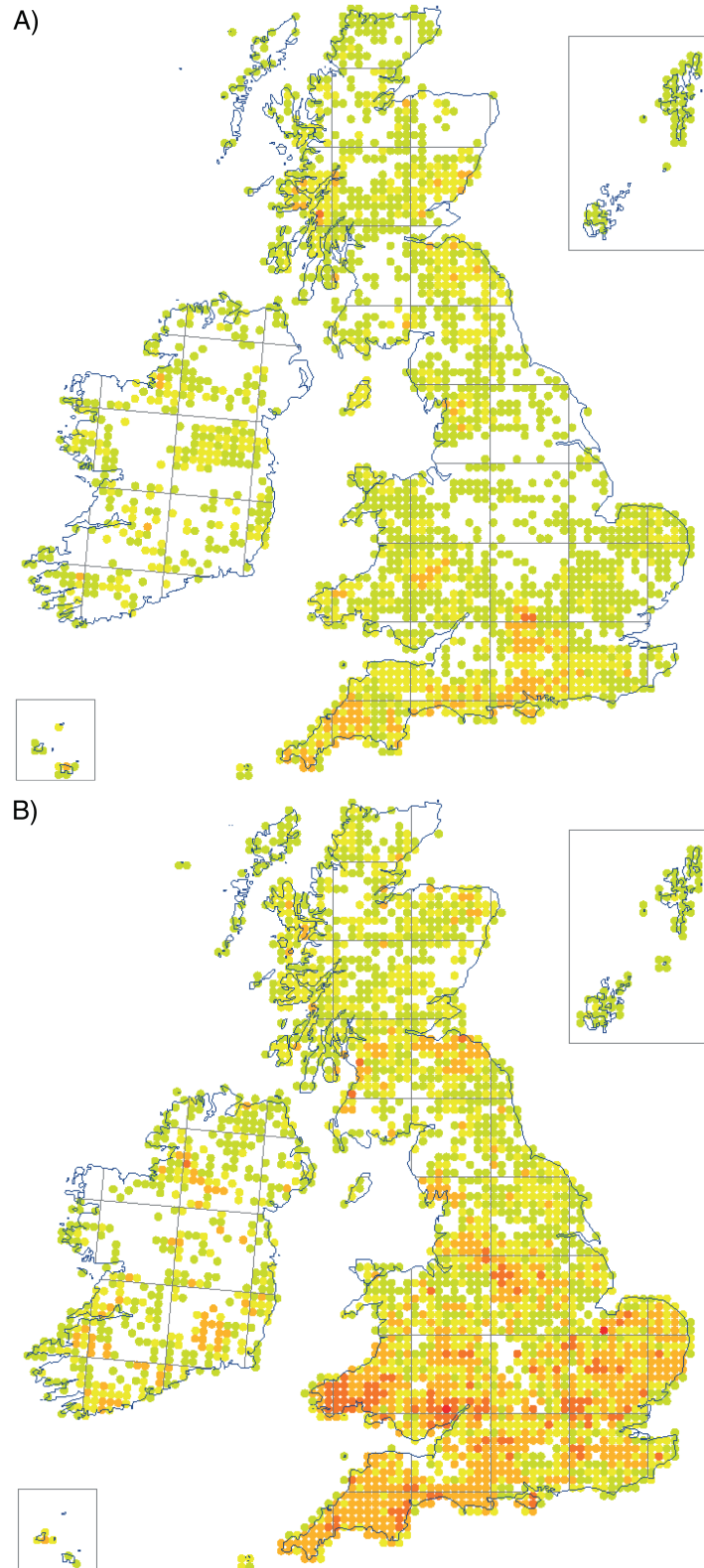


Figure 5. Coincidence map for 28 epiphytic bryophytes for 1960–1980 (A) and 1990–2010 (B). These are species that were found to have significantly increased in abundance, after adjustments for estimated local recorder effort, between 1960–2013 (Hill & Preston, 2014). Dots of increasing density indicate species per hectad counts of 1–5, 6–10, 11–15, 16–20, 21–25, and > 25.

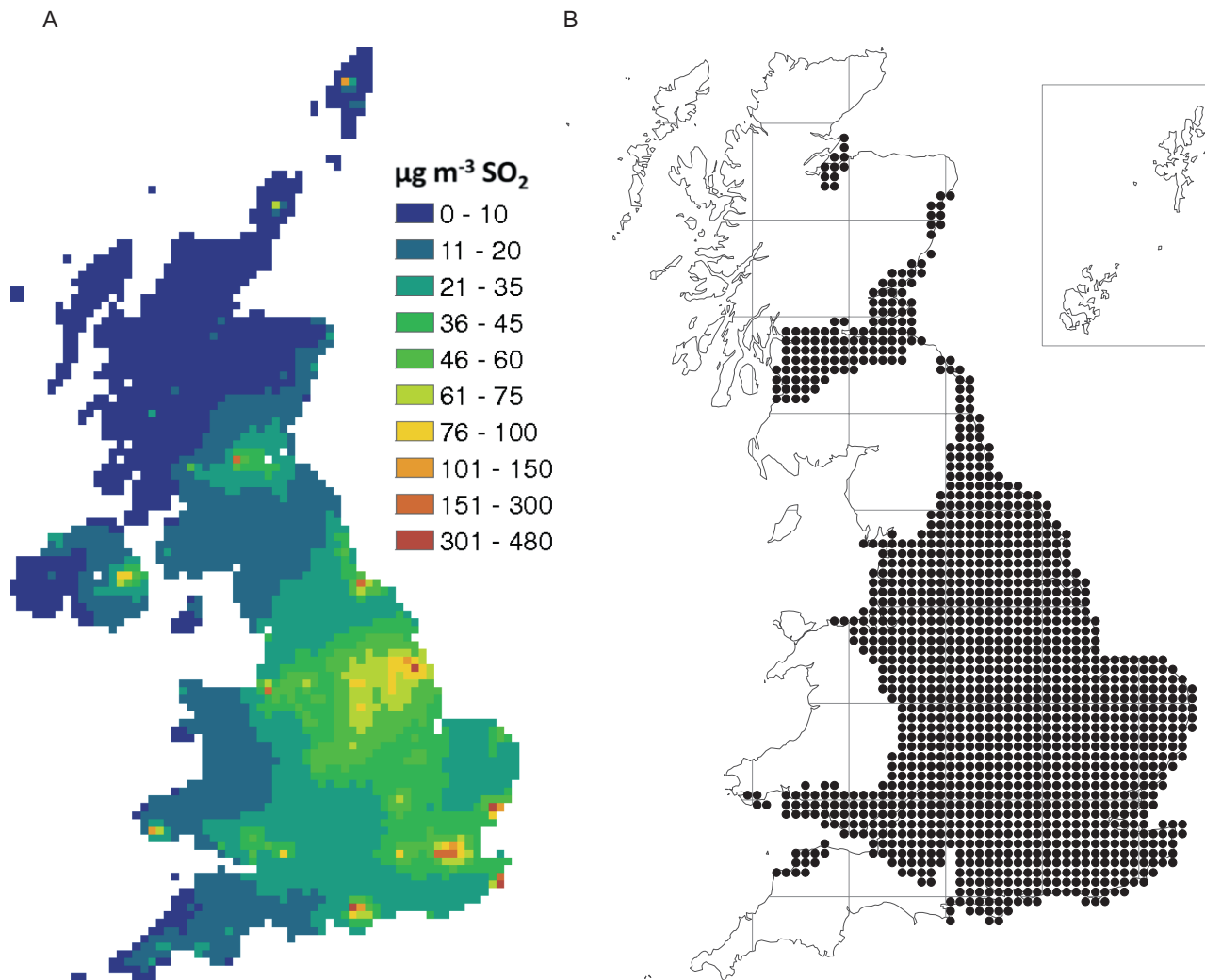


Figure 6. A, modelled SO₂ pollution levels for Britain in 1970 (Dore *et al.*, 2007; Matejko *et al.*, 2009). B, high (black) and low (white) SO₂ pollution zones for Great Britain for the bryophyte and Lepidoptera analyses of change.

its congener *Orthotrichum diaphanum*. These two epiphytes have overlapping niches, with *O. affine* generally restricted to trees and shrubs but with rare occurrences on walls and stones, whereas *O. diaphanum* is equally at home on either organic or inorganic surfaces (Hill *et al.*, 1991). Both of these species have been affected by acidic air pollution, although their distinct ecologies have led to different patterns of change. As early as 1968, Oliver Gilbert demonstrated a difference in the distributions of these species in the Newcastle area, showing that, although the epiphytic *O. affine* was restricted to the cleanest environments, *O. diaphanum* was able to survive on asbestos roofs some distance along the pollution gradient studied (Gilbert, 1968, 1970). This has led some authors to describe *O. diaphanum* as pollution tolerant (Stevenson, 1999); however, it would be more accurate to state that the species

avoids the effects of pollution, with its broader niche allowing it to penetrate inside sulphur polluted areas by growing on basic substrata such as asbestos (Gilbert, 1970). The most recent evidence from the recorder-effort adjusted national trends produced by Hill & Preston (2014) suggests that *O. diaphanum* may have recovered more quickly under decreasing sulphur pollution than *O. affine* (GB high sulphur zone) (Fig. 7). Although *O. diaphanum* was more common under polluted conditions as a result of its relatively catholic niche (Gilbert, 1970), the fact that it has also undergone a considerable recovery (albeit a weaker one than *O. affine*) (Fig. 7), clearly indicates that it too suffered as an epiphyte (Gilbert, 1970). *Orthotrichum diaphanum* initially appears to have increased more rapidly than *O. affine*, this may be at least partly a result of its existing populations on basic, inorganic substrata providing it with a head-

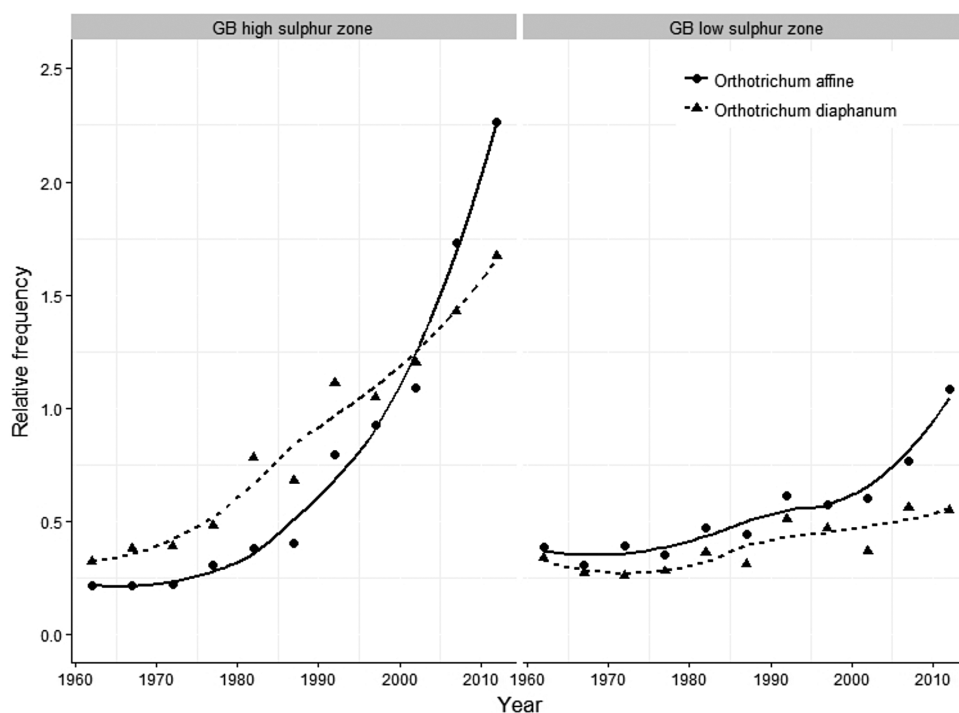


Figure 7. Frescalo time-trends for *Orthotrichum affine* and *Orthotrichum diaphanum* in Great Britain, calculated separately for high and low sulphur zones (see text); smoothers are local regression ('loess') lines. Frequencies are relative to the average local occupancy of a set of 'benchmark species' (Hill, 2012); frequencies above 1 indicate that a taxon has become commoner than the average benchmark species.

start from increased propagule pressure over other expanding epiphytes, as well as its broader niche (Fig. 7). Before the national analysis undertaken for the new bryophyte atlas, few studies appear to have commented on the increases in *O. diaphanum* in comparison to other sensitive taxa (but see Adams & Preston, 1992 and Boon & Outen, 2011). This may be mainly result from its continued existence in calcicolous inorganic refugia during the worst period of acidic pollution. However, it is now clear that *O. affine* has overtaken *O. diaphanum* by some margin, and that it is also increasing in the low sulphur, mainly upland, zone; whether this is a result of other environmental drivers, such as climate change, or merely a return to pre-pollution era biogeographical zones [Hill *et al.* (2007) classify *O. affine* as a member of the Boreo-temperate element compared to the Southern-temperate *diaphanum*] remains to be seen.

The second example of recent change in the British and Irish bryophyte flora concerns acidophilic species. Rose & Wallace (1974) included examples of these species in their review of the changing bryophyte flora of Britain, highlighting *Dicranum montanum*, *Dicranum tauricum* (syn. *Dicranum strictum*), *Dicranoweisia cirrata*, and *Orthodontium lineare* as acidophile species that had increased their ranges

(Rose & Wallace, 1974). Although the connection between SO_2 and substrate acidification was not explicitly made by Rose & Wallace (1974), it was suggested that these species were tolerant of 'moderate air pollution and possibly . . . selectively favoured by this in competition with other species' (Rose & Wallace, 1974). Jones (1991) provided a thorough review of the increasing acidophilic element in the Oxfordshire flora in the 20th Century, and also drew on much additional evidence from across Britain. The new bryophyte atlas (Blockeel *et al.*, 2014) now suggests that the late 1980s and early 1990s were a turning point for many of these species, with the available evidence demonstrating a subsequent decline in many acidophilic species (Hill & Preston, 2014). The Frescalo-generated (Hill, 2012) 5-yearly time trends shown in Figure 8 demonstrate this phenomenon for four species. The rise and fall (*Aulacomnium androgynum*, *D. montanum*, *Lophocolea heterophylla*), or drop off from high levels (*Pohlia nutans*), are clearer, or larger in magnitude, in the high SO_2 region compared to the low region, as would be expected for a set of species that are considered to have been favoured by acidic air pollution in the mid 20th Century. Strikingly, a very clear distinction between continued stability (low SO_2), and increase followed by decline (high SO_2), was also

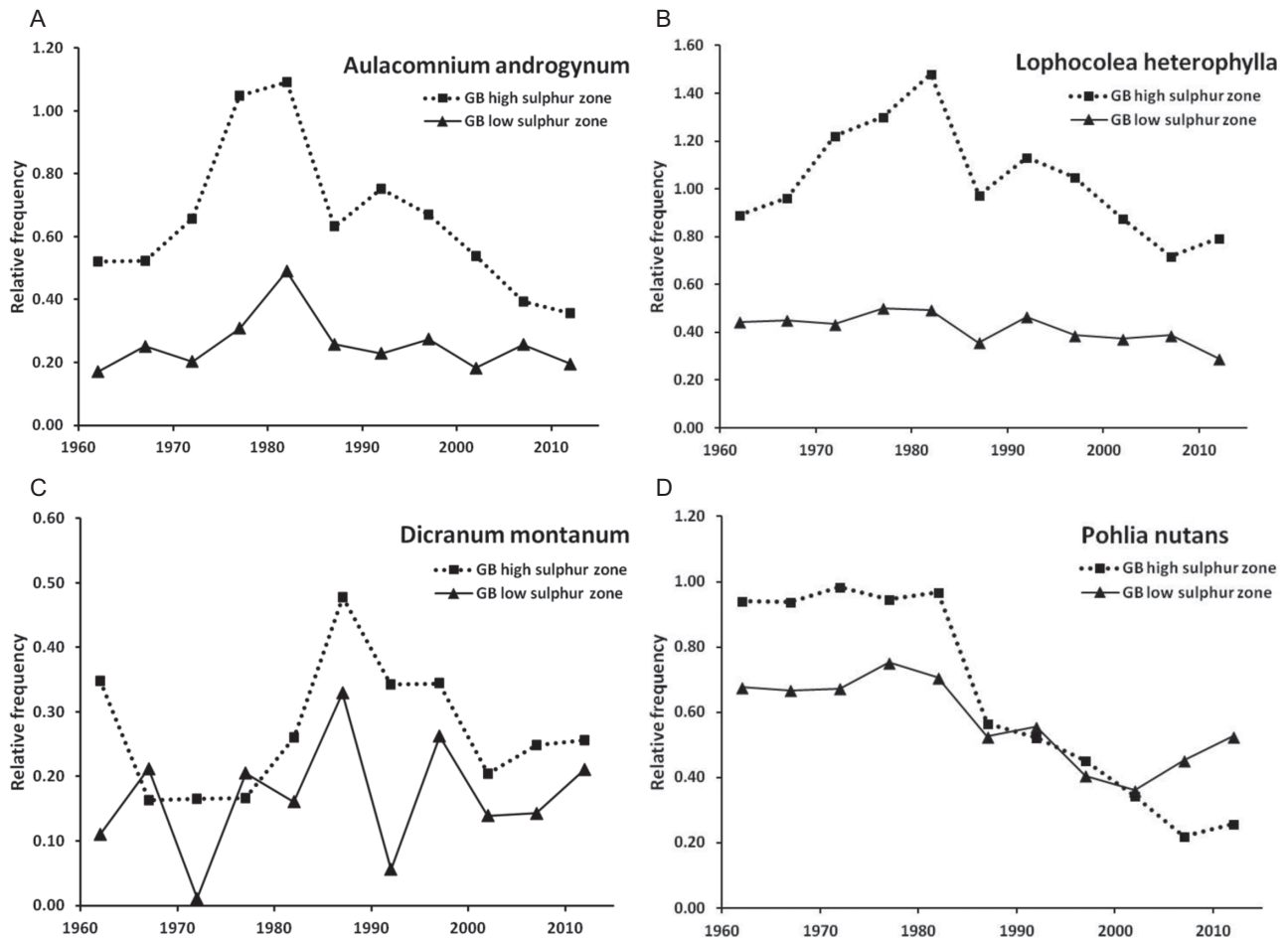


Figure 8. Frescalo time-trends for four acidophyte species in Great Britain, calculated separately for high and low sulphur zones (see text); note the different scales. A, *Aulacomnium androgynum*; B, *Lophocolea heterophylla*; C, *Dicranum montanum*; D, *Pohlia nutans*. Frequencies are relative to the average local occupancy of a set of 'benchmark species' (Hill, 2012); frequencies above 1 indicate that a taxon has become commoner than the average benchmark species.

found for an ecological grouping of bryophytes characteristic of lowland acid woodlands by Hill & Preston (2014). Unfortunately, experimental evidence for the colonization of trees and shrubs by acidophytic bryophytes under increasing SO_2 does not appear to exist for bryophytes as it does for lichens (Bates, McNee & McLeod, 1996), although the common patterns found across species of similar ecology, coupled with much anecdotal evidence for increases in acidophiles up to the early 1990s (Jones, 1991), as well as evidence for acidophytic lichen increases and declines under experimental SO_2 fumigation (Bates *et al.*, 1996) and increasing pH respectively (Hauck *et al.*, 2011) lend strong support to the national picture of increase followed by decline deduced from the analysis of relative hectad occurrences over time (Fig. 8; Hill & Preston, 2014).

As for lichens, we should not ignore the specific effects of pollutants in the uplands. For example,

significant effects of reduced nitrogen deposition on the growth and cover of *Racomitrium lanuginosum* were demonstrated by Pearce, Woodin & Van Der Wal (2003). The historic effects of sulphur and nitrogen pollution on ombrotrophic *Sphagnum* spp. on the southern Pennines are also well known (Lee & Studholme, 1992) and, encouragingly, recent surveys of experimental transplants and transects established during the 1980s are now revealing improvements in these habitats (Caporn *et al.*, 2006). The acidification and ongoing recovery of upland streams (Murphy *et al.*, 2014) is also likely to have had effects on aquatic bryophytes: the early developmental stages of species such as *Chiloscyphus polyanthus* and *Platyhypnidium riparioides*, which are characteristic of more neutral waters, are not able to tolerate acidity (Vanderpoorten & Goffinet, 2009), whereas species characteristic of acid waters, such as *Scapania undulata*, might be expected to have benefitted.

Indeed, it has been suggested that the abundance of *Nardia compressa* in the streams of mid-west Wales may be an indication of acidification (J. W. Bates, pers. comm.). Focused monitoring has provided some evidence for recoveries in upland acidic streams, generally relating to increased occurrences of the more acid-sensitive mosses *Hygrohypnum ochraceum* and *Fontinalis* spp. (Monteith & Shilland, 2007). Despite all of the above evidence for the upland impacts of acidic pollutants, none of the species noted above showed significant trends in the national hectad-scale analyses of Hill & Preston (2014). This is perhaps not surprising, given the abundance of these species across the British uplands.

Direct and indirect effects of air pollutants on bryophytes

Bryophytes are mostly poikilohydric, relying on their environment at any moment for moisture, and having no mechanism for the regulation of water loss (Proctor, 2011). Existing thus in an equilibrium with their water environment makes them particularly susceptible to atmospheric pollution, which can arrive in the form of dry, wet or occult (i.e. in cloud or fog aerosols) deposition (Bates, 2002). However, as Nash (2008) has pointed out in the context of lichen sensitivities to pollution, this is 'a generalisation that requires cautious interpretation and limited extrapolation'. Clearly all bryophytes are not equally sensitive to all pollutants, and it is the combination of physiological sensitivity to the direct effects of a pollutant, the actual exposure to a pollutant, as well as indirect effects, such as changes to a plant's interactions with other plants or animals, or pollution-driven changes to a substrate, that ultimately determine the risk of weakening or death. Many naturalists, and probably all bryologists, will be familiar with the particular susceptibility of epiphytic bryophytes to air pollution, although we may ask whether epiphytes are uniquely susceptible as a group, or whether the emphasis on this group has been a result of other factors, such as a bias in research interests because of the discrete nature of epiphyte habitats, or to more general aspects of their ecological life strategies or physiology; for example, the fact that many epiphytes have been categorized as 'shuttle' species (During, 1992) with relatively short life-cycles and dynamic populations; this point is returned to below.

Although there are many challenges to overcome in experimentally assessing the impacts of different types of atmospheric pollution on bryophyte species, much interesting work has been carried out using field or laboratory fumigations, or immersion techniques with aqueous solutions of pollutants in their ionic forms (Bates, 2002). A focus of several experiments has been the effects of SO₂ on growth

and photosynthesis (Ferguson, Lee & Bell, 1978). Investigations into the biochemical processes underpinning these effects have suggested that the acid-driven phaeophytinization (i.e. the replacement of Mg²⁺ with 2H⁺) of chlorophyll could be the main mechanism behind SO₂ toxicity, although other studies have suggested that this may be misleading because of the unrealistically high levels of SO₂ used in these early experiments (Bates, 2002). A particularly fascinating experiment, using the bog-pool moss *Sphagnum cuspidatum*, revealed evidence for inter-population variation for tolerance to SO₂ effects on photosynthesis and dark respiration (Baxter, Emes & Lee, 1991). This was revealed through the comparison of plants from a polluted area (the South Pennines) with those from a relatively unpolluted area (North Wales): the plants from the polluted mire were more tolerant to SO₂ than those from the cleaner area. This difference was found to be mediated by metal ions oxidizing phytotoxic bisulphite ions; the tolerance conferring metal ions were themselves present in the South Pennine mires as a result of industrial pollution. The increased SO₂ tolerance could be induced in the population from North Wales through the experimental addition of metal cations (Baxter *et al.*, 1991); an additional genetic contribution to the observed differences between populations cannot be ruled out but was not revealed by isozyme analyses (Lee & Studholme, 1992). Experimental evidence for differential SO₂ tolerance between species also exists: Winner & Bewley (1983) revealed differences in SO₂ sensitivity between the pleurocarps *Pleurozium schreberi*, *Hylocomium splendens*, and *Ptilium crista-castrensis*, despite finding no difference in SO₂ uptake between species. Fertility may also be affected: for example, fruiting in *P. schreberi* has been shown to be reduced around a copper smelter in Finland (Huttunen, 2003), and effects on the fertility of several species has been previously inferred from historic observations (Rose & Wallace, 1974). The physiological basis of SO₂ tolerance in two terricolous mosses has recently been investigated in greater detail by Bharali & Bates (2006). It was shown that the process of detoxification has both metabolically passive and active components. The passive oxidation of phytotoxic bisulphite is enabled by the uptake of Fe³⁺, as shown by Baxter *et al.* (1991) for *S. cuspidatum*, whereas evidence was also found for active processes such as metabolically-driven extracellular oxidation and internal metabolic detoxification.

The direct effects of the dry deposition of nitrogen oxides on bryophytes have been studied very little (Lee *et al.*, 1998), despite their being among the most important pollutants of urban environments. In what appears to be the only study utilizing experimental

fumigations of NO and NO₂, Morgan, Lee & Ashenden (1992) found effects of NO_x on nitrate assimilation but no effects on photosynthesis, suggesting that, although NO and NO₂ have direct effects on the nitrogen metabolism of bryophytes at realistic atmospheric concentrations, they do not strongly disrupt the growth of plants. Many experimental studies of nitrogen impacts have mimicked wet deposition of nitrogen pollution through the addition of NO₃⁻ or NH₄⁺, and it is now well known that the addition of nitrogen in this way results in the induction of nitrate reductase, and in an increase in bryophyte tissue nitrogen concentrations (Lee *et al.*, 1998). Experimental additions to semi-natural communities have also demonstrated effects on the abundances of two common pleurocarps, *P. schreberi* and *Rhytidiadelphus squarrosus* (Morecroft, Sellers & Lee, 1994); however, more recent research has shown that physiological measurements, such as phosphorous metabolism enzymes and tissue nitrogen : phosphorous ratios, may be better indicators of nitrogen deposition impacts and recovery in bryophytes (Arróniz-Crespo *et al.*, 2008). Arróniz-Crespo *et al.* (2008) suggested that *Pseudoscleropodium purum* would be a good indicator of nitrogen deposition impacts and subsequent recovery as a result of its ability to withstand the phosphorous limitation stress resulting from high levels of nitrogen uptake, and thereby respond to the changes in nitrogen supply resulting from air pollution. Bryophyte transplant studies along nitrogen deposition gradients have also found significant effects of nitrogen pollution on bryophyte growth, tissue nitrogen, and community composition (Mitchell *et al.*, 2004, 2005), although different metrics of nitrogen pollution severity correlated with different responses in different species, suggesting many complexities waiting to be uncovered (Mitchell *et al.*, 2005). Broader surveys of species richness across modelled nitrogen deposition gradients have shown significant declines in species richness associated with either reduced or oxidized forms of nitrogen deposition. For example, Field *et al.* (2014) surveyed five broad habitat types across the UK, and, using multiple regression, found significant negative relationships with forms of nitrogen deposition for moss (liverworts were excluded from survey) species richness in acid grassland and sand dune communities, and in bogs and upland heaths for lichens (although spatial autocorrelation was not accounted for in the statistical models used, suggesting that the marginal *P*-values reported for the lichen species richness responses may be less reliable).

The results from surveys of species richness across pollution gradients are likely to be the cumulative outcome of both the direct and indirect effects of pollutants. Much of the research on the indirect

effects of SO₂, NO_x, and NH₃ on bryophytes has focused on the pH of species' substrates, particularly bark. As noted above, early on in cryptogam-air pollution research Gilbert (1968, 1970) examined a variety of habitats along a pollution gradient, and showed that an epiphyte's substrate was crucially important for tolerance to acidic pollutants. Other studies, including Coker (1967) and Skye (1968), have directly demonstrated how SO₂ can acidify bark and reduce its buffering capacity. Another well-researched indirect impact of air pollution on bryophytes is the increased competition from vascular plants resulting from the eutrophication caused by nitrogen deposition. Both experimental work and national-scale correlative studies have demonstrated negative relationships between nitrogen and bryophyte cover (Lee & Caporn, 1998; Carroll *et al.*, 2000) and species richness (Duprè *et al.*, 2010; Edmondson *et al.*, 2010).

The question of why epiphytic bryophytes are so sensitive remains to be definitively answered: terricolous species have clearly also been impacted by the direct and indirect effects of atmospheric pollutants (Bates, 1993), although the evidence for change at the national scale over the recent past has not been as striking as it has for epiphytes (Hill & Preston, 2014). Given that the epiphytic bryophytes so impacted are from a number of different families, the impacts cannot solely be the result of any particular phylogenetic tendency towards pollutant sensitivity but must result from some more general physiological constraint. The answer may lay in part with the typical life strategy of epiphytic bryophytes: most are classified as long- or short-lived 'shuttle' species in ecological life-strategy schemes created for bryophytes (During, 1992). It is generally assumed that these species 'track' habitat patches across the landscape, with extinction at one patch offset by the regular production of propagules allowing them to locate new habitat (Rydin, 2008). Such a strategy clearly relies on the availability of suitable regeneration niches (Grubb, 1977), and it can be postulated that the acidification of bark as a result of SO₂ (and NO_x) may have resulted in a catastrophic loss of these niches for a suite of species that rely on the continual extinction and colonization processes of a metapopulation existence (Wiklund & Rydin, 2004).

LICHEN-FEEDING LEPIDOPTERA

Evidence from biological records over time

Approximately 2500 species of Lepidoptera have been recorded in Great Britain (Waring & Townsend, 2009). These are split, for convenience rather than by phylogeny, into micro- and macro-moths. The micro-moths are the most speciose with over 1500 species, the majority of which, as their name suggests, are

very small; their size and difficulty of identification has meant they have been poorly recorded and studied in comparison to their larger relatives. For this reason, we focus our analysis of air pollution impacts on the macro-moths, for which good distribution data (Asher *et al.*, 2001; Fox *et al.*, 2010) exist for most species. The macro-moths are extremely diverse with species found in most terrestrial and some aquatic environments. The larval stages have an equally diverse range of food sources, although the majority are herbivorous. A small number feed on lichens, particularly the Lithosiinae, a group of moths in the Erebidae family commonly known as the footman moths. In the UK, there are sixteen resident footman moths (Table 3). There are an additional six species from three families (Erebidae, Geometridae, and Noctuidae) in the UK whose larvae are also lichen feeders (Table 3). There are also a small number of macro-moths that are suspected to feed on mosses, for example, barred carpet (*Martania taeniata*). However, these additional species are uncommon, relatively poorly recorded, and their early life stages poorly understood, and thus they are also excluded from our analysis.

Similar to many other taxa, macro-moths have shown significant declines in recent decades, with over two-thirds of studied species showing declines since 1968 (Conrad *et al.*, 2006). However, many of the lichen-feeding species appear to have increased, some significantly, especially the eight footman moths for which population and distribution trends have been analyzed (Conrad *et al.*, 2006; Fox *et al.*, 2006, 2013). Other species, for which data are insufficient in these national datasets to determine trends, have also been observed to be increasing. For example, *Cryphia algae* is a recent colonist previously regarded as an immigrant to the southern coast of Britain. Over the last decade, this species has become well established in the south-east and is rapidly expanding its range north and westwards, with annual records in Buckinghamshire and Berkshire over the last few years. *Lithosia quadra* was largely regarded as an immigrant moth with residency restricted to a few areas in south-western England and Wales. This species is increasingly recorded across southern counties and further inland suggesting a range expansion. *Eilema caniola* historically had a restricted distribution in the south-west of Britain with immigration to the south-east but is now resident in much of the south-east with records as far west as Oxfordshire in recent years. Increases in these lichenivorous moth species have been linked to the recovery of some lichen species following a reduction in SO₂ pollution but, as yet, there has been no study to provide evidence to support this (Fox, 2013). Indeed, despite the more direct link between lichenivorous moths and

lichen recovery as a result of changes in air pollution, most studies to date in this area have focused on the change in frequency of typical and melanistic forms of the peppered moth (*Biston betularia*) (Cook & Saccheri, 2013).

Using distribution data collected as part of the National Moth Recording Scheme (NMRS), we provide an overview of changes in the frequency of lichenivorous moths in Britain. Sufficient data were available to assess changes for 21 species in total (Table 3). This included two Red Data species and three nationally scarce species, as well as two species, *E. caniola* and *C. algae*, which have recently colonized south-eastern England. To do this, we performed a chi-squared goodness-of-fit test on the relative hectad frequencies of each species in two 20-year time periods (1960–1980 and 1993–2013). These frequencies were compared with those of all other species-hectad records in the same time periods to account for changes in recording intensity between the two time periods (see ‘Bryophytes’ section above) (Ball *et al.*, 2011; Hill & Preston, 2014; Isaac *et al.*, 2014). The relative frequency of each species was also analyzed separately for the high and low SO₂ zones (Fig. 6B) for both time periods. We also ran a Frescalo analysis (Hill, 2012) across 5-year periods from 1960 to 2013 for the high and low SO₂ zones separately, allowing us to look in greater temporal detail at species’ estimated relative frequency changes in the two pollution zones.

Out of the 21 species investigated (Table 3), only one showed a significant decline in its relative frequency of occupied hectads over time, the dew moth, *Setina irrorella* (Table 4). Chi-squared goodness-of-fit tests showed increases in frequency over time for the remaining species; this was significant for two-thirds of these species (Table 4). Within different SO₂ zones, we found an over-riding pattern of greater increases of lichenivorous macro-moths in areas of historically high pollution compared to low pollution areas. Three species showing significant increases overall showed a nonsignificant decline in low SO₂ areas, combined with significant increases in high SO₂ areas; these were *Mitochrista miniata* (Frescalo time-trend shown in Fig. 9), *Nudaria mundana* and *E. caniola*. A further ten species for which overall significant increases over time were observed showed a greater magnitude of increase in high SO₂ areas than in low SO₂ areas, and one species, *S. irrorella*, showed a highly significant decline in low SO₂ areas only, with no significant change in high SO₂ areas (Frescalo time-trend shown in Fig. 9). The remaining species showed highly significant increases across both areas, for example *Eilema griseola* and *Eilema depressa* (Frescalo time-trends shown in Fig. 9).

The overall conclusion reached from analysis of the 21 lichenivorous macro-moths reviewed here is that,

Table 3. Twenty-one lichenivorous macro-moth species for which sufficient data to analyze changes in national frequency were available

Species	Family	Distribution		Population status	
		Status	Regionality	1968–2002 (annual change rate)	
<i>Alcis jubata</i>	Geometridae: Ennominae	Local	W, N	Increasing	0.062
<i>Cleorodes lichenaria</i>	Geometridae: Ennominae	Local	W, WC, (S, N)	Declining	–0.011
<i>Thumatha senex</i>	Erebidae: Arctiinae	Local	S, C, (N)	Increasing	0.013
<i>Setina irrorella</i>	Erebidae: Arctiinae	Rare (nationally scarce)	S, NW	Unknown	NA
<i>Mitochondria miniata</i>	Erebidae: Arctiinae	Local	S, (C)	Increasing	0.040
<i>Nudaria mundana</i>	Erebidae: Arctiinae	Local	T	Increasing	0.022
<i>Atolmis rubricollis</i>	Erebidae: Arctiinae	Local	S, WC, (N)	Unknown	NA
<i>Cybosia mesomella</i>	Erebidae: Arctiinae	Local	S, (C, N)	Increasing	0.004
<i>Pelosiia muscerda</i>	Erebidae: Arctiinae	Rare (Red Data Book species)	E	Unknown	NA
<i>Eilema sororcula</i>	Erebidae: Arctiinae	Local	S	Unknown	NA
<i>Eilema griseola</i>	Erebidae: Arctiinae	Common	S, (C)	Increasing	0.063
<i>Eilema caniola</i>	Erebidae: Arctiinae	Uncommon (nationally scarce), recent colonist to SE	SW, SE	Unknown	NA
<i>Eilema pygmaeola</i>	Erebidae: Arctiinae	Rare (Red Data Book species)	SE, E	Unknown	NA
<i>Eilema complana</i>	Erebidae: Arctiinae	Local	S, C	Increasing	0.091
<i>Eilema depressa</i>	Erebidae: Arctiinae	Local	S, (C)	Increasing	0.065
<i>Eilema lurideola</i>	Erebidae: Arctiinae	Common	S, C, (N)	Increasing	0.010
<i>Lithosia quadra</i>	Erebidae: Arctiinae	Uncommon (nationally scarce), immigrant	SW	Unknown	NA
<i>Laspeyria flexula</i>	Erebidae: Boletobiinae	Local	S, (C)	Declining	–0.029
<i>Cryphia algae</i>	Noctuidae: Bryophilinae	Immigrant; recent colonist	S, (C)	Unknown	NA
<i>Cryphia domestica</i>	Noctuidae: Bryophilinae	Common	S, C, (N)	Increasing	0.051
<i>Cryphia muralis</i>	Noctuidae: Bryophilinae	Local	S	Unknown	NA

Distribution data from Waring & Townsend (2009); population status from Conrad *et al.* (2006). Species names and taxonomic position follow Agassiz, Beavan & Heckford (2013). Regionality definitions (Waring & Townsend, 2009): C, central (from the Wash northwards to Cumbria and Northumberland); E, eastern; N, northern (from Cumbria and Northumberland to Shetland); NA, not available; T, throughout; S, southern (south of the Wash); and W, western (east and west respectively of a north-south line bisecting the Isle of Wight). Regions given in parentheses indicate that the main part of the distribution is elsewhere, but that the distribution extends to this area.

Table 4. Species occurrences as a proportion of all species-hectad occurrences (multiplied by 10 000 for readability) in each 20 year time period (1960–1980 and 1993–2013) across all British hectads, and separately for hectads in low and high SO₂ areas

SO ₂ zone							
All	Low	High					
Time period							
1960–1980	1993–2013	1960–1980	1993–2013	1960–1980	1993–2013		
Total hectad occurrences							
188 333	516 923	57 359	179 351	130 974	337 572		
Species	Family						
<i>Alcis jubata</i>	Geometridae: Ennominae	4.8	5.4	14.5	14.2	0.6	0.7
<i>Cleorodes lichenaria</i>	Geometridae: Ennominae	6.4	9.1***	16.6	20.3*	1.9	3.2**
<i>Thumathia senex</i>	Erebidae: Arctiinae	7.1	10.9***	4	6.5**	8.4	13.2***
<i>Setina irrorella</i>	Erebidae: Arctiinae	1.0	0.5**	1.7	0.6***	0.7	0.4
<i>Mitochrista miniata</i>	Erebidae: Arctiinae	9.2	11.7***	10.3	10.1	8.7	12.5***
<i>Nudaria mundana</i>	Erebidae: Arctiinae	12.0	13.3	21.3	19.8	7.9	9.9*
<i>Atolmis rubricollis</i>	Erebidae: Arctiinae	3.0	14.4***	5.4	18.1***	2	12.4***
<i>Cybosia mesomella</i>	Erebidae: Arctiinae	9.8	10.0	13.2	11.3	8.2	9.3
<i>Pelossia muscerda</i>	Erebidae: Arctiinae	0.4	0.9*	0.2	0.2	0.5	1.3**
<i>Eilema sororcula</i>	Erebidae: Arctiinae	1.9	13.8***	2.3	7.8***	1.7	17***
<i>Eilema griseola</i>	Erebidae: Arctiinae	9.7	21.3***	8	15.3***	10.4	24.5***
<i>Eilema caniola</i>	Erebidae: Arctiinae	1.4	3.1***	3.5	3.1	0.5	3.1***
<i>Eilema pygmaeola</i>	Erebidae: Arctiinae	0.4	0.7	0	0.1	0.6	1
<i>Eilema complana</i>	Erebidae: Arctiinae	12.2	18.5***	7.3	10.6**	14.4	22.6***
<i>Eilema depressa</i>	Erebidae: Arctiinae	4.3	20.9***	4.4	16.7***	4.3	23.1***
<i>Eilema lurideola</i>	Erebidae: Arctiinae	29.6	30.1	25.3	26	31.5	32.3
<i>Lithosia quadra</i>	Erebidae: Arctiinae	2.8	5.7***	4.7	8.3***	2	4.4***
<i>Laspeyria flexula</i>	Erebidae: Boletobiinae	12.1	13.8***	5.1	6.7	15.1	17.6*
<i>Cryphia algae</i>	Noctuidae: Bryophilinae	0	2.0	0	0.2	0	2.9***
<i>Cryphia domestica</i>	Noctuidae: Bryophilinae	23.5	25.0***	16.9	17.6	26.4	28.9
<i>Cryphia muralis</i>	Noctuidae: Bryophilinae	3.9	7.0*	5.8	7.5	3.1	6.8***

Significance of changes between the time periods, derived from chi-squared goodness-of-fit tests: * $P < 0.05$; ** $P < 0.01$; *** $P < 0.001$.

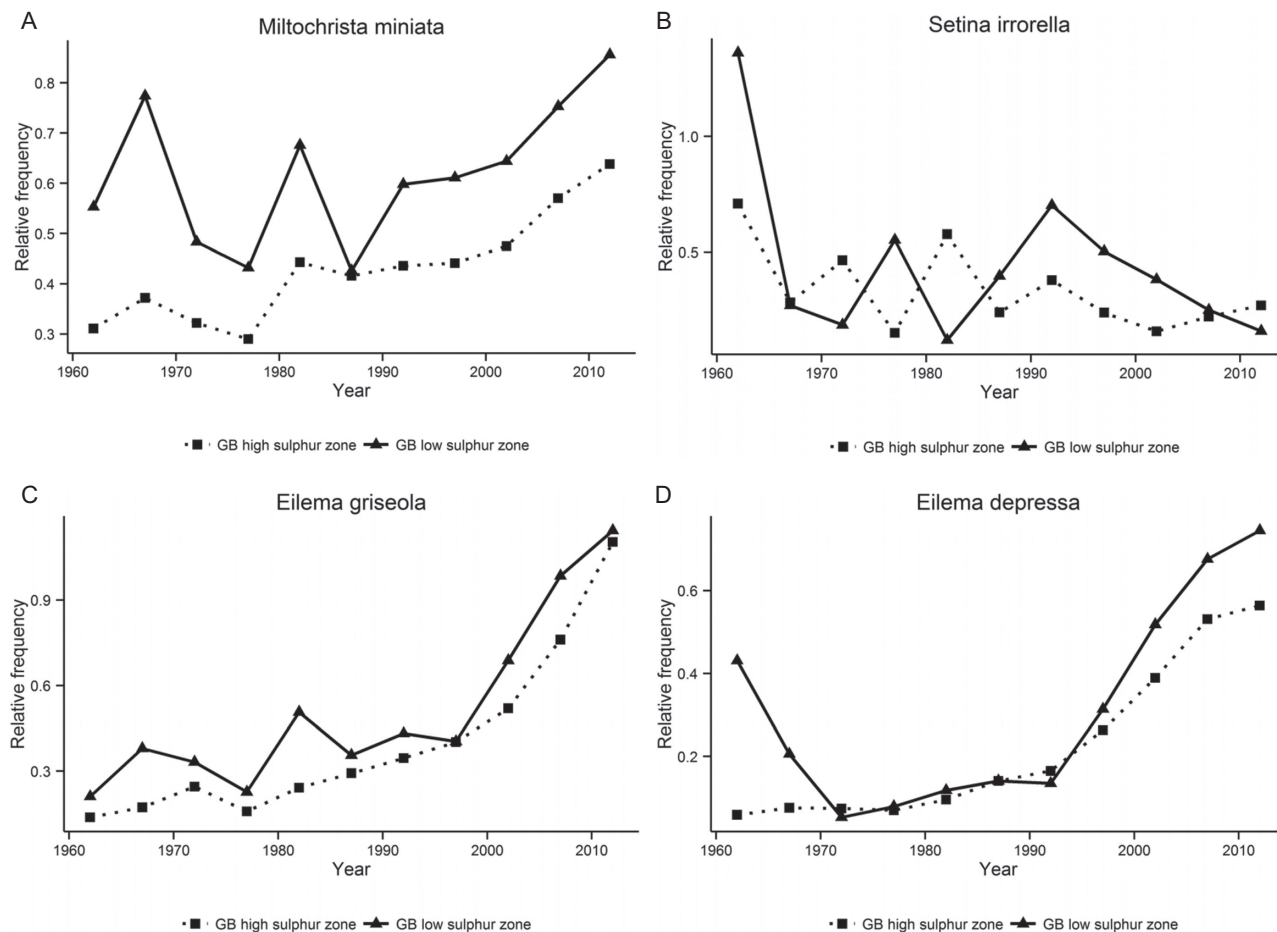


Figure 9. Frescalo time-trends for four lichenivorous moth species in Britain, calculated separately for high and low sulphur zones (see text); note the different scales. A, *Miltochrista miniata*; B, *Setina irrorella*; C, *Eilema griseola*; D, *Eilema depressa*. Frequencies are relative to the average local occupancy of a set of 'benchmark species' (Hill, 2012); frequencies above 1 indicate that a taxon has become commoner than the average benchmark species.

in Britain, most of these species appear to have increased across the recent period of air quality improvements. Only one species, *S. irrorella*, has declined significantly, and this is a moth restricted to coastal habitats in south-western Britain (Waring & Townsend, 2009) that is unlikely to have been strongly affected by historic SO_2 pollution. Increases in the frequency of most lichenivorous moths have tended to be greatest in areas previously characterized by high levels of SO_2 , suggesting that the subsequent reduction in air pollution is potentially strongly associated with an increase in these moths, paralleling responses in their lichen hosts. From our analysis, we cannot rule out other important drivers of change, such as climate and habitat change. Climate in particular may be extremely important, and may explain the increased abundance and residency of once predominantly migrant species such as *C. algae* and *L. quadra* (Sparks, Roy & Dennis, 2005); however,

positive effects of climate change are often limited by habitat loss in Lepidoptera and it is widely recognized that some species, habitat specialists in particular, can only respond positively if suitable habitat is available (Warren *et al.*, 2001). Thus, increases in lichen abundance and diversity as a result of reduced air pollution may have also provided the opportunity for some species to benefit from a warming climate.

Whether or not changing lichen diversity is a major driver in itself is difficult to determine at this stage, particularly given the bias in recording towards areas that were previously highly polluted through industrialization, and because of a lack of autecological knowledge for most of the moth species for which trends were determined. Although we do not know which lichen species, if any, are preferred by each moth species, those lichens historically abundant in polluted areas are likely to have had high levels of toxins (Pöykkö & Hyvärinen, 2003) and those species

that have recovered subsequent to the reduction in SO₂ have tended to exhibit certain growth forms, because fruticose and foliose lichens have typically been more impacted by acidic pollution (Coppins *et al.*, 2001). As well as potentially providing more easily digestible food sources, such lichens may also provide better refugia (Pöykkö, 2011) and/or better background matching for increasingly effective crypsis, thus reducing predation and leading to population increases. There is clearly a need for a more extensive analysis of available biological records data to determine the role of improved air quality in the observed increases of lichenivorous Lepidoptera, than we have had the space to provide in this overview. We expect that the increasing availability of well-resolved biological records, and environmental covariate information, will enable more in-depth analyses in the near future.

CONCLUSIONS

The improvement in the air quality of our islands has been one of the environmental success stories of the 20th Century. The tradition of biological recording in Britain and Ireland has given naturalists and ecologists a wide range of opportunities to connect the changing atmospheric pollutant loads of the past 200 years to the abundance and diversity of various taxa. These observations have informed systematic studies and sophisticated experimental work, as well as contributing original conclusions of their own through increasingly thorough recording and analyses designed to account for potential biases in records datasets. Air pollution is still a significant issue affecting our biodiversity, although the effects may now be more subtle as a result of lower levels of most pollutants. Although such effects may not be straightforward to identify at the hectad scale, the increasing trend towards recording at finer scales (Preston & Rorke, 2014) suggests that these unparalleled, partly citizen scientist-collected datasets will continue to play an hugely important role in the monitoring of our environment.

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