

1 **The impact of air pollution on terrestrial managed and natural vegetation**

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15 **Abstract**

16 Although awareness that air pollution can damage vegetation dates back at least to the 1600s, the  
17 processes and mechanisms of damage were not rigorously studied until the late 20<sup>th</sup> century. In the  
18 UK following the Industrial Revolution, urban air quality became very poor, with highly phytotoxic  
19 SO<sub>2</sub> and NO<sub>2</sub> concentrations, and remained that way until the mid-twentieth century. Since then  
20 both air quality, and our understanding of pollutants and their impacts, have greatly improved.

21 Air pollutants remain a threat to natural and managed ecosystems. Air pollution imparts four major  
22 threats to vegetation are discussed through a series of case studies. Gas phase effects by the primary  
23 emissions of SO<sub>2</sub> and NO<sub>2</sub> are discussed in the context of impacts on lichens in urban areas. Effects of  
24 wet and dry deposited acidity from sulphur and nitrogen compounds are considered with a  
25 particular focus on forest decline. Ecosystem eutrophication by nitrogen deposition focuses on  
26 heathland decline in the Netherlands, and ground level ozone at phytotoxic concentrations is  
27 discussed by considering impacts on semi-natural vegetation. We find that, although air is getting  
28 cleaner, there is much room for additional improvement, especially for the effects of eutrophication  
29 on managed and natural ecosystems.

30 **Keywords**

31 Acid deposition, ammonia, nitrogen, ozone, sulphur

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33 **1. Introduction**

34 There is a very long history of understanding that air pollution is a threat to vegetation. The  
35 medieval writer and mystic Hildegard von Bingen (1098 – 1179) noted in her book *Causae et Curae*  
36 [1] that dust within rain was believed to damage crops. In the 1600s developments in early scientific  
37 understanding did not miss the importance of pollutants from combustion processes and industry in  
38 damaging plants, including the first evidence of air pollution as a transnational concern. In the first  
39 decade of the 1600s, King James passed an act “against burning of Ling, and Heath, and other Moor-  
40 burning...” noting that “some parts even of France itself lying South west of England, did formerly  
41 make of being infested with Smoakes driven from our Maritime Coasts, which injur'd their Vines in  
42 Flower” [2].

43 However, prior to the 19<sup>th</sup> century and the Industrial Revolution, there were no air quality  
44 measurement networks, and evidence that air pollution was damaging vegetation was limited to  
45 areas in the immediate vicinity of point sources (e.g. lime kilns, charcoal production, early smelting  
46 activities). In *Sylva* [3], John Evelyn recognised the threat to vegetation through being “infected with

47 foggs and poys'nous vapours, or expos'd to sulphurous exhalations", while Fabri (1670) [4] saw  
48 volcanic emanations as a source of acidic damage to fruit. As industrialisation increased, so did  
49 reports of environmental damage: in the summer of 1794 a visitor to south Wales observed, "Nearer  
50 to Swansea, there are extensive works both of copper and iron, from the malignant influence of  
51 which every trace of vegetation in the neighbourhood has from a very short period after their  
52 erection, been totally annihilated" [5]. At this time the main phytotoxic pollutant was thought to be  
53 SO<sub>2</sub> and to a lesser extent HCl and NO<sub>2</sub>.

54 An analysis of global sources of the major gaseous pollutants by Hoesly *et al.* [6] showed the  
55 dominance of sources of SO<sub>2</sub> and NO<sub>x</sub> in Europe and North America from 1880 until 1980 (Figure 1).  
56 Thereafter, following the introduction of control measures in Europe and North America and the  
57 rapid growth of sources in Asia the main sources of SO<sub>2</sub> and NO<sub>x</sub> have been in East and South Asia  
58 (Figure 1). Early records of pollution impacts included observations from both the natural and  
59 agricultural environments. From around 1800 the dark or melanic form of some species of moths in  
60 England began to increase in frequency. On soot-covered tree trunks, darker moths were better  
61 camouflaged and thus more avoided predation than light ones [7]. Sheep fleeces became blackened  
62 by smoke [8] and in the industrialising United States bird feathers became covered in black carbon  
63 particles [9]. 'Black rain' was observed in remote areas of Scotland [10] and soot from Manchester's  
64 factories caused black snow in Scandinavia as described by the playwright Ibsen in his play *Brand* in  
65 1867. Interestingly, the latter observation shows clear evidence of long-range transport of pollutants  
66 in Europe long before the arguments of the 1970s on whether industrial emissions from the UK,  
67 Germany and France contributed to acid deposition in Scandinavia.

68 The rapid increase in both industrial and domestic SO<sub>2</sub> and NO<sub>2</sub> emissions in the early 20<sup>th</sup> century in  
69 several European countries, notably the UK, Germany and France, and in North America (Figure 1),  
70 generated regional surface concentrations sufficient to damage both crop- and wild plant species as  
71 demonstrated by the following examples. An extensive series of field investigations by Cohen and  
72 Ruston published in 1925 [11] substantiated the observations of various earlier workers on the  
73 effects of coal-smoke polluted atmosphere on city vegetation. Using a transect from the suburbs  
74 towards the centre of Leeds, Cohen and Ruston [11] found depressions in yield, various degrees of  
75 visible damage and alterations in growth habit of a number of species (e.g. *Laurus nobilis*), which  
76 increased in intensity as pollution levels increased towards the city centre. The ill effects on  
77 vegetation were positively correlated with the annual wet deposition of particulate matter and  
78 sulphur compounds, but the relative toxicity of the different components of the coal smoke was not  
79 determined. In 1928 Pettigrew [12] described extensive damage to plants and the total failure of  
80 ornamental trees and shrubs in Manchester parks as a result of atmospheric pollution, while in 1941

81 Metcalfe [13] described the pollution-induced premature shedding of leaves, flowers and buds on  
82 ornamental species (e.g. *Begonia foliosa*) in towns during periods of foggy weather.

83 Much of the earlier interest in the effects of air pollution on vegetation in the UK focused on such  
84 damage to ornamental species in urban areas, where losses were considered to be more serious  
85 from an aesthetic rather than economic viewpoint [e.g. 11, 12, 13]. However in 1952 Bleasdale [14]  
86 devised an experiment to show that a coal-smoke polluted atmosphere can considerably depress the  
87 yield of the economically important strain of the pasture grass *Lolium perenne*. When attempts  
88 were made to improve the quality of hill pastures in heavily polluted districts of East Lancashire by  
89 reseeding with this strain of *Lolium perenne*, the plants became established successfully, but soon  
90 declined in productivity and eventually disappeared. Bleasdale grew the plants in Manchester in two  
91 greenhouses: one ventilated with polluted ambient air and the other with air purified by passage  
92 through a water scrubber. Significant depressions in yield occurred in the plants in the polluted  
93 greenhouse compared with those grown in purified air. Importantly, there were no observable  
94 lesions on the leaves, something that had previously been considered an indicator of damage [15].

95 Metal pollution was a further concern, with hotspots of metal deposition recorded close to smelters  
96 in Canada, the United States and at several locations in Europe [16-18]. Effects of metal pollution on  
97 vegetation were relatively local, within 30 km of the source, but the scale of emissions and  
98 persistent nature of some of the metal species left long-term soil contamination in these areas and  
99 damage to some flora and fauna. Research was largely concerned with the possibility of metals  
100 entering the human body via ingestion of contaminated crops. Of wider concern was the use of  
101 tetraethyl lead in petrol, which had been introduced as an anti-knocking agent in many countries in  
102 the 1920s. By the middle of the twentieth century, it became apparent that lead was deposited  
103 alongside roads, with levels falling exponentially with distance from the road, but detectable up to  
104 100 m away [19]. The identification of such widespread environmental contamination by lead  
105 resulted in massive pressure to reduce the use of tetraethyl lead, leading to a ban throughout much  
106 of the world.

107 Ground level ozone became an important part of the global pollution climate in the mid-20<sup>th</sup>  
108 century. Trends in surface ozone from measurements over the last 6 decades [20] show large, long-  
109 term trends in Europe. In presenting trends at remote sites, the effects of local sources are  
110 minimised and the regional trend becomes clearer. In the analysis by Cooper *et al.* [20], annual  
111 average surface ozone concentrations increased from 20 to 40 ppb between 1950 and 2000 and  
112 changed little since then in Europe. Remote sites in North America and East Asia show increases of  
113 typically 10 ppb between 1980 and 2010, whereas the monitoring stations in the remote Pacific,

114 Antarctica, and Tasmania show quite small changes (<10ppb) between 1980 and 2010 [20]. Ozone is  
115 produced within the troposphere through photochemical degradation of volatile organic compounds  
116 in the presence of NO<sub>2</sub>. Ozone became recognised as an important regional phytotoxic compound in  
117 the 1960s, initially in California [21], but extending to much of the United States and Southern  
118 Canada and detected in Europe during the 1970s [22].

119 Air pollution issues of the mid-20<sup>th</sup> century gradually extended to ever-larger areas as global  
120 emissions of sulphur approached peaks (Figure 1). In part, these changes were due to the number  
121 and scale of sources, but also to the increasing heights of emission, with tall stacks on industrial  
122 sources of 200 m or more and large buoyant plumes of pollutant gases effectively injected towards  
123 the top of the boundary layer, promoting wide dispersion and long-range transport. These tall  
124 stacks, which were deployed to reduce ground-level pollution, essentially turned a local pollution  
125 problem into a regional one. Ecological effects of long-range transport of sulphur and nitrogen  
126 compounds began to be recognised in countries remote from the sources of the primary pollutants,  
127 initially as effects of acid deposition in Scandinavia [23] and in Scottish lochs [24]. By the end of the  
128 20<sup>th</sup> century, various national air quality laws and programmes had been implemented in many  
129 developed countries around the world. These had the effect of reducing emissions and deposition of  
130 many air pollutants, though emissions and deposition still remained higher than would be expected  
131 in the absence of anthropogenic emissions across much of the world.

132 Considering this background, we discuss four main threats for the global pollution climate in the late  
133 20<sup>th</sup> century:

- 134 1. Gas phase effects of the primary emissions of SO<sub>2</sub> and NO<sub>2</sub> reflecting their emissions in  
135 urban areas and industrial regions including substantial tracts of arable cropland in Europe,  
136 North America, East and South Asia,
- 137 2. Effects of wet and dry deposited acidity from sulphur and nitrogen compounds, notably in  
138 upland areas where inputs are dominated by wet deposition. We give particular attention to  
139 the poorly buffered geological regions throughout Northern Europe, Scandinavia, the north-  
140 eastern states of the USA and parts of Eastern Canada.
- 141 3. Ecosystem eutrophication by nitrogen deposition in both oxidised and reduced forms over  
142 much of northern Europe and large parts of eastern North America,
- 143 4. Ground level ozone present at phytotoxic concentrations over much of the arable cropland  
144 of Europe and North America, East and South Asia.

145 In addition to the four main threats to vegetation more localised effects of metals, especially heavy  
146 metals, close to smelters and other metal processing industries, are notable through the 20<sup>th</sup>

147 century, but severe cases of damage to vegetation were not regional in scale. These four main  
148 pollutant threats will be examined via a series of case studies of major pollution impacts.

## 149 **2. Gas phase effects of SO<sub>2</sub>, NO<sub>2</sub> and NH<sub>3</sub>: case study lichen decline**

150 In Europe at the beginning of the twentieth century, industrialisation and large populations of cities  
151 produced high concentrations of SO<sub>2</sub> and NO<sub>2</sub> and thus urban air quality was typically very poor. The  
152 peaks in emissions and exposure of vegetation were between 1960 and 1970 for SO<sub>2</sub> in Europe and  
153 North America thus during the middle decades of the 20<sup>th</sup> century extensive areas of cropland and  
154 semi-natural areas of Europe and North America were exposed to damaging concentrations of SO<sub>2</sub>  
155 [25]. As a consequence of air pollution legislation, concentrations of SO<sub>2</sub> declined steadily in many  
156 developed countries from about 1970. By 2015, there were no significant areas of Europe or North  
157 America experiencing concentrations of SO<sub>2</sub> at levels damaging to plants from anthropogenic  
158 sources [26]. In East Asia peak emissions were later, but since 2012 emissions of SO<sub>2</sub> in China have  
159 declined by approximately 40%. Emissions of NO<sub>2</sub> peaked around 1990 in Europe, rather later than  
160 SO<sub>2</sub> due to the rapid growth in vehicle usage, as well as industry and the slower introduction of  
161 effective controls on vehicle NO<sub>2</sub> emissions. Thus, concentrations of NO<sub>2</sub> remained high in urban  
162 and many rural areas through to the early years of the 21<sup>st</sup> century in Europe and North America  
163 [26].

164 Ammonia emissions, mainly from agricultural activities, steadily increased in all industrial countries  
165 through the 20<sup>th</sup> century [27]. Few countries have made significant reductions in emissions, the  
166 Netherlands and Denmark being notable exceptions, each of which have reduced emissions over the  
167 last two decades by approximately 50% [28, 29]. The overall global emissions of NH<sub>3</sub> continue to  
168 increase and as emissions of NH<sub>3</sub> are closely coupled to ambient temperature, it is likely that  
169 changes in climate will further increase emissions [27].

170 These broad changes in the chemical climate of industrial countries have had widespread effects on  
171 crops and natural vegetation, only some of which have been documented in detail. Here, the effects  
172 on lichens, which are particularly sensitive to atmospheric composition are used as an example of  
173 the changing chemical climate of industrial countries.

174 Lichens are rarely a dominant feature of terrestrial habitats yet they are a ubiquitous component of  
175 most habitats globally, from arctic to desert to tropical forest, and in both managed and natural  
176 ecosystems. Although dependent on the substratum for their physical attachment, lichens have no  
177 vascular root system: they receive nutrients and water directly from the atmosphere, making them  
178 highly susceptible to changing atmospheric conditions.

179 In 1866 Nylander [30] observed that 'lichen deserts' were created around Paris, often extending for  
180 considerable distances aligned to the direction of the prevailing wind. Similar patterns were  
181 observed around that time across Europe, and in the mid-twentieth century around New York City  
182 [31] as well as other parts of the U.S. [32-34]. A cause of these declines was hypothesised as  
183 concentrations of acidifying pollutants in urban air. However, there were no systematically-  
184 measured air quality data to support the observations. In the UK it took the dramatic loss of life  
185 associated with the London smog in the winter of 1952 (estimated 12,000 deaths [35]) to establish a  
186 national air quality recording system across both urban and rural areas; by the 1960s this comprised  
187 c. 1300 stations [36]. In the late 1960s, Hawksworth and Rose [37] correlated the distribution of  
188 lichen species across the UK with recorded levels of SO<sub>2</sub> from this network to create a ten-point scale  
189 of estimated SO<sub>2</sub> pollution based on species' sensitivity to air pollution. Since lichens absorb  
190 pollutants directly from the air, shrubby or hanging filamentous lichens with a large surface area  
191 such as *Usnea* species were at the top of the list of sensitive species, while crustose species  
192 associated with naturally acidic habitats such as *Lecanora conizaeoides* and leprose species of  
193 *Lepraria*, at the bottom of the list, were tolerant and becoming widespread in urban areas [37].

194 As SO<sub>2</sub> pollution declined, reactive nitrogen became the dominant atmospheric pollutant (Figure 1),  
195 both as NO<sub>x</sub> from vehicular and industrial emissions and as NH<sub>3</sub> from agricultural processes. The  
196 effect on lichen communities was dramatic, as species tolerant of nitrogen (nitrophytes) began to  
197 appear, in urban and agricultural areas. Gaseous ammonia deposited onto wet surfaces acts as a  
198 base, removing hydrogen ions from water and producing ammonium and hydroxide ions. Thus, in  
199 areas receiving high ammonia deposition, formerly common species tolerant of acid (acidophytes)  
200 became rare [38]. In areas affected by ammonia deposition, including naturally acidic moorland and  
201 heathland vegetation dominated by terricolous species of lichen, *Cladonia* died back [39, 40]. The  
202 effect of ammonia is to increase bark pH, and research in the UK showed a strong correlation  
203 between lichen communities and bark pH and ammonia concentrations [41-43]. Quantitative studies  
204 of epiphytes on a range of tree species in sites where ammonia was monitored showed that lichen  
205 communities varied with tree species as well as air quality, so that acidophytes survived longer on  
206 acid-barked trees such as oak and birch, while nitrogen-tolerant species of the *Xanthorion* alliance  
207 appeared earlier on trees with a higher bark pH such as ash or poplar (Figure 2) [41, 43-47]. This  
208 shift in lichen communities from acidophyte species to nitrophyte species was demonstrated by a  
209 resurvey of twig communities on a nature reserve in Wales after ten years which showed a loss of  
210 nitrogen-sensitive species on branches and the appearance of the *Xanthorion* [48]. A nationwide  
211 survey of sensitive and tolerant species of epiphytic macrolichens in the UK was used to test their  
212 response to measured ammonia at air pollution monitoring sites across the UK using lichen

213 frequency and bark pH as measured variables. The results showed that a loss of nitrogen-sensitive  
214 species was taking place prior to the appearance of nitrophytes, and that this was happening at c.  
215  $1\mu\text{g m}^{-3}$   $\text{NH}_3$ , well below the current critical level (concentration above which direct adverse effects  
216 are thought to occur) of  $8\mu\text{g m}^{-3}$  [49]. Since then  $1\mu\text{g m}^{-3}$  has been accepted as the critical level for  
217 lichens [50].

### 218 **3. Deposited acidity in upland areas: case study forest decline**

219 Prior to the late 1970s, concerns about acid deposition focused primarily on aquatic ecosystems.  
220 However, in the late 1970s and early 1980s, foresters and scientists began to notice widespread  
221 diebacks of forest trees in both the north-eastern US and Europe, and suspicion mounted that acid  
222 deposition was the cause. The European decline was popularly known as “Waldsterben” (forest  
223 death): it affected both hardwood and softwood trees and was characterized by extreme thinning of  
224 the crown, premature senescence, discolouration and loss of foliage, active casting of green foliage,  
225 and loss of fine root biomass [51]. The North American decline was most severe in red spruce  
226 (*Picea rubens*) in mountainous locations of New York and New England [52]. Red spruce decline  
227 symptoms were the same on both continents and included reddening of needles in the early spring,  
228 a condition usually associated with winter freezing damage.

229 Most researchers on both continents pointed the finger at air pollution as the driving force behind  
230 the forest declines. Large concentrations of acidity in orographic cloud and a windy upland climate  
231 for many forests leads to high deposition rates of the pollutants contained within the cloud water,  
232 which were shown to reduce the frost hardiness of red spruce [53]. Furthermore, Cape *et al.* [50]  
233 showed that it was the acidity and  $\text{SO}_4^{2-}$  ions rather than  $\text{NO}_3^-$  that were responsible for the observed  
234 reduction in frost hardiness. In Europe two major sets of hypotheses around the mechanisms gained  
235 prominence [51, 54, 55], The ‘top-down’ hypotheses focused on the direct impacts to leaves and  
236 needles of high levels of pollutants, particularly oxides of sulphur and nitrogen, and ozone [e.g. 51,  
237 56]. Other pollutants such as metals and organic compounds were also implicated. The ‘bottom-  
238 up’ hypotheses proposed that the primary causal element of forest decline lies in the soil, through  
239 acidification and depletion of basic cations from soil exchange sites, leading to mobilization of toxic  
240 aluminium, as well as the excess enrichment by reactive nitrogen, leading to nutrient imbalances  
241 and acidifying nitrification pulses [e.g. 57, 58]. Soluble aluminium causes a range of physiological  
242 damage, including direct mortality to fine roots and mycorrhizae, impaired ability to transport  
243 calcium and magnesium at the soil-root interface and a loss of the soil fauna that transport oxygen  
244 and nutrients to the deeper soil [59]. These two groups of hypotheses were generally not considered  
245 mutually exclusive, but which were the primary and which were the contributory factors was hotly



246 debated. Since forest decline damage was ultimately recorded across a large number of hardwood  
247 and conifer species over a wide area and range of environmental conditions in Europe, it is highly  
248 likely that the primary and secondary drivers of damage also varied. However, to our knowledge  
249 there has been no comprehensive analysis of the literature to provide an overview of the relative  
250 roles of these different mechanisms across European forests.

251 The cause of declines in red spruce in the north-eastern US was identified as foliar leaching of  
252 calcium, specifically membrane-bound calcium, causing reduced frost hardiness that could lead to  
253 winter damage [53, 60, 61]. The problem was particularly acute for red spruce because it occurred  
254 primarily in montane forests where it was exposed to cold winter temperatures and high deposition  
255 of acid rain and also to extremely acidic cloud droplets which, are very effectively scavenged by  
256 conifer needles [62]. Low calcium availability in the soil could exacerbate the problem, as indicated  
257 by a study that showed that, compared to a reference catchment, frost damage was much reduced  
258 in a catchment in which calcium was added experimentally to the soil [63]. The same study showed a  
259 major improvement in the health of sugar maple (*Acer saccharum*) in the calcium-treated  
260 catchment, underscoring that it had also been affected by acid rain in the north-eastern U.S. [64].  
261 Researchers have since reported that many other species likely show similar effects, though possibly  
262 not as severe and not as widespread because they are less common across the landscape [65].

263 In recent decades, the decline in acid deposition in the eastern US has led to a resurgence of red  
264 spruce, which is growing well throughout the region and expanding its range into lower-elevation  
265 forests [66, 67]. Similarly the International Co-operative Programme on the Assessment and  
266 Monitoring of Air Pollution Effects on Forests (ICP Forests) showed no major changes in the  
267 proportion of damaged trees in Europe overall from the mid-1990s to the early 2010s, with conifers  
268 showing an improvement in condition and broadleaves showing a decline. Much of this was due to  
269 the new inclusion of Mediterranean ecosystems, where oak have shown a strong decline in recent  
270 years, mainly attributed to drought and insect damage [68]. In 2018 and 2019 a new decline in forest  
271 condition was reported in central and northern Europe; it is thought the primary driver is a  
272 widespread, persistent drought [69].

273 As sulphur emissions began to decline in North America and Europe, research focused increasingly  
274 on nitrogen deposition. The concept of “nitrogen saturation” was advanced by Ågren and Bosatta  
275 [70] and Aber et al. [71] to indicate deposition of nitrogen exceeding the biological capacity of the  
276 ecosystem to retain it. According to these studies, this leads to a sequence of changes in ecosystem  
277 function including increased plant tissue nitrogen concentrations, increased nitrogen cycling in the  
278 soil, increased nitrification, and ultimately elevated nitrogen leaching and soil acidification. Elevated

279 nitrogen leaching was found to occur in many catchments in the eastern US where deposition was  
280 above 8 kg N ha<sup>-1</sup> y<sup>-1</sup> [72] and in many forest plots in Europe where deposition was above about 10  
281 kg N ha<sup>-1</sup> y<sup>-1</sup>, especially if in the soil pH was low and % organic nitrogen was high [73]. The nitrogen  
282 saturation theory was further refined by Lovett and Goodale [74] who noted that the impacts of  
283 excess nitrogen deposition were not necessarily sequential, but rather all could occur simultaneously  
284 and their expression was controlled by the relative strength of the vegetation and soil sinks for  
285 nitrogen in the ecosystem. The nitrogen saturation theory marked a major shift in thinking about  
286 nitrogen by forest ecologists: previously nitrogen was primarily considered a key limiting nutrient,  
287 but the nitrogen saturation theory argued that when chronic deposition levels are high, it can also  
288 have adverse effects.

289 The impact of air pollution on forest health is now considered as a part of, or an acceleration to, a  
290 combination of stresses, especially drought [75, 76]. Past management is also clearly a contributing  
291 factor: many parts of central Europe had a history of poor silviculture practices including even-aged,  
292 monocultural plantations with little genetic variability and litter-raking which removed valuable  
293 nutrients [75]. Thus the extreme acid deposition of the late 1970s may have been a ‘final straw’ for  
294 a large area of European forest already damaged and predisposed to further stress, triggering a  
295 widespread decline.

296 As pollutant emissions continue to decline, there may be a long-term legacy of damage done to soils  
297 by decades of acid deposition. Key studies show that forests where acidification is reduced can  
298 undergo major shifts in ecosystem pools and processes, including reductions in forest floor and  
299 upper mineral soil carbon and nitrogen pools and mobilization of nitrogen into deeper soil levels or  
300 export from the ecosystem [55, 64, 77, 78]. The impact and duration of legacy effects in the  
301 ecosystem for both nitrogen and sulphur are determined by the extent to which the “slow pools” in  
302 the ecosystem have been affected. These slow pools could include the available base cation pools  
303 that are replenished primarily by mineral weathering, or the stable soil organic matter pools that  
304 turn over very slowly [79, 80]. As a result, we see some impacts of acid deposition reversing quickly  
305 (e.g., lower foliar leaching of nutrients, improved health of trees and declining stream nitrate fluxes  
306 and concentrations [81-83]) while other impacts, such as the depleted base saturation of acidified  
307 soils, may take decades or centuries to recover [84]. Thus, although the forests have appeared to  
308 stabilize and even improve in some areas, reducing acid deposition has also not removed all of the  
309 stresses.

310

311 **4. Ecosystem eutrophication by nitrogen deposition: case study heathland decline and**  
312 **biodiversity losses**

313 As described above, air pollution in Europe during the second half of the 20<sup>th</sup> century is dominated  
314 by the gradual reduction in sulphur emissions and deposition and the slower decline in emissions  
315 and deposition of oxidized nitrogen. Notably, the deposition of nitrogen has been largely unchanged  
316 as emissions of NH<sub>3</sub> continued largely unabated [38]. The European hot spots for nitrogen deposition  
317 have been, and largely remain, the low countries of northern mainland Europe and the Netherlands  
318 in particular. Parts of the intensive livestock farming areas of the UK France and Denmark also  
319 receive high levels of nitrogen deposition and gas phase NH<sub>3</sub> [42].

320 The most dramatic impacts of nitrogen deposition have been large community shifts in nutrient-  
321 poor terrestrial habitats such as grassland, heathland and bogs. In the Netherlands during the  
322 1970's and 80's, large areas of heathland transitioned to grassland. This typically occurred quite  
323 rapidly at a site (one to two years as documented in one study [85]) and by 1983 80 km<sup>2</sup> of  
324 heathland in the Netherlands, nearly 20% of the total, had become dominated by grasses [86, 87].  
325 The grasses that became dominant were typically *Molinia caerulea* and *Deschampsia flexuosa*. It  
326 was observed that outbreaks of heather beetle (*Lochmaea suturalis*) caused serious damage to  
327 *Calluna vulgaris* which led to the death of heather plants and a replacement by grasses. This was  
328 first linked to eutrophication in 1977 by de Smit [88] and early experiments pointed to the existence  
329 of relationships between eutrophication, heather beetle infestation and grass encroachment [86].

330 In the 1970's nitrogen deposition in the Netherlands ranged from around 40 to 80 kg N ha<sup>-1</sup> y<sup>-1</sup> [89].  
331 Experiments demonstrated, however, that even at N addition levels much higher than ambient (200  
332 kg N ha<sup>-1</sup> yr<sup>-1</sup>), *M. caerulea* did not directly outcompete *Calluna vulgaris* [90]. Instead, fertilisation by  
333 atmospheric deposition of N increased the quality of *C. vulgaris* as food plant for heather beetles.  
334 Heather beetle infestations on *C. vulgaris* cause many individual plants to die, which opens up the  
335 canopy and allows vigorously-growing grasses to become dominant. During a heavy infestation of  
336 heather beetles little nitrogen is lost from the system: almost none is leached and only about 1 kg N  
337 ha<sup>-1</sup> is lost with dispersing insects [86]. Soils under dead *C. vulgaris* were found to have high  
338 mineralisation and large N pools and low NH<sub>4</sub><sup>+</sup> immobilisation, these conditions led to ammonium  
339 accumulation under dead heather, facilitating grass invasion [91].

340 Heather beetle larvae are able to detect and feed preferentially on the most N-rich shoots [92] and  
341 experiments showed that the beetles from fertilised plots showed higher growth rates compared to  
342 unfertilised plots and were the heaviest adults [85, 93, 94]. This was a consequence of better  
343 survival rates at larval stage and through hibernation induced by high nitrogen in the larval food

344 supply [92]. The increased survival rates led to an increase in the severity (outbreak densities can be  
345 as high as 2000 beetles m<sup>-2</sup> [95]) and frequency of beetle outbreaks [93] with the frequency of  
346 outbreaks increasing from every 20 years to every 8 years [96]. It has since been shown that  
347 population numbers are highest after long-term nitrogen addition, indicating that impacts are likely  
348 to be exacerbated over time [92].

349 The conversion of heathland to grassland is very visible, but other impacts of nitrogen deposition on  
350 biodiversity are less apparent without surveys. One of the most reported of these is wide-spread  
351 losses of plant species richness. Declines in species richness with increasing nitrogen deposition had  
352 been observed in experimental situations for many years [e.g. 97, 98]. In well-buffered prairie  
353 grasslands long-term N-addition experiments in the field showed declines in species richness as a  
354 consequence of competition for other resources [99, 100]. In 2004 it was demonstrated that the  
355 plant species richness of acidic grasslands was negatively correlated with ambient levels of nitrogen  
356 deposition across the UK. In acidic grasslands the decline equated to one plant species lost for every  
357 additional 2.5 kg N ha<sup>-1</sup> y<sup>-1</sup> [101] of long-term N deposition. Forbs in particular declined in species  
358 richness and cover, whilst cover of grasses tended to increase.

359 Since 2004 negative correlations between nitrogen deposition and plant species richness have been  
360 reported in a wide range of habitats [e.g. 102, 103] and in different parts of the world [e.g. 104, 105,  
361 106]. These changes in species richness are accompanied by changes in the chemistry of soils,  
362 microbial communities [e.g. 107] and soil processes [e.g. 108]. It is likely that changes in plant  
363 species richness and composition are driven by a combination of mechanisms including acidification,  
364 eutrophication and subsequent plant competition and interactions with secondary drivers. The  
365 relative importance of these drivers is likely influenced by habitat type, soil properties and climate as  
366 well as other variables. Such widespread changes in plant species richness are probably having  
367 impacts on invertebrate communities and higher up the food chain. There is growing evidence of  
368 these impacts [109] although this is an area where further research is needed. N deposition in some  
369 regions of the world continues to increase, but in parts of Europe it is beginning to decline [26].  
370 However, evidence from some long-term experiments indicates plant species richness and  
371 composition in many damaged areas are unlikely to recover without active management [e.g. 110,  
372 111-113].

### 373 **5. Ground-level ozone at phytotoxic concentrations**

374 Ground level ozone is formed in a series of photochemically driven reactions between oxides of  
375 nitrogen and volatile organic compounds. Unlike sources of SO<sub>2</sub> and NO<sub>2</sub>, ozone is produced within  
376 the atmosphere, through the photochemical degradation of VOC in the presence of NO<sub>2</sub> [114]. The

377 rate of production varies between the organic compounds as well as NO<sub>2</sub> mixing ratios and the VOC  
378 emitted by traffic provide some of the largest ozone production rates. Ozone production continues  
379 throughout the troposphere where the reactants and solar radiation are present and methane is a  
380 major contributor to tropospheric ozone production in remote regions [115]. Ozone is a  
381 hemispheric scale pollutant [114] among the gaseous pollutants its effects on vegetation may occur  
382 much further from the sources of the precursor pollutants than the effects of SO<sub>2</sub> and NO<sub>2</sub>. Ground  
383 level ozone pollution is an issue in much of the mid-latitudes of the northern hemisphere as well as  
384 parts of the southern hemisphere, with highest levels during summer in Southern Europe, South and  
385 East Asia and large areas of the southern states of the USA and Mexico [116]. Concentrations of  
386 ozone in the Northern Hemisphere mid-latitudes doubled between the late 19th century and 1980  
387 and have continued to increase more slowly, reaching current levels of 35–40 ppb [116]. Ozone is a  
388 toxic pollutant to both agricultural crops and semi-natural vegetation. As a powerful oxidant it exerts  
389 its effects primarily following stomatal uptake and the extent of injury is proportional to the  
390 absorbed stomatal dose [117]. Ozone impacts on crops are discussed in depth by Emberson *et al.*  
391 [118].

392 Ozone impacts on vegetation were first recognised in Los Angeles, California in the 1950s. At the  
393 time poor air quality in the region was characterised by dense smogs which led to eye irritation and  
394 had a distinctive odour. These smogs were first linked to ozone by the chemist Arie Haagen-Smit,  
395 who described the process of ozone formation by photochemical oxidation of hydrocarbons and  
396 nitrogen oxides [119]. The impacts of the smog on crops were already well described, but Haagen-  
397 Smit isolated ozone as the cause using laboratory fumigation experiments with spinach, beets,  
398 endive, oats and alfalfa. The results showed damage from ozone that was indistinguishable from  
399 that produced by the smog [119]. Whilst the impacts of ozone on crops had already been described  
400 [120] this was the first time they were linked to smog.

401 Until the early 1970s it had not been considered that ozone could ever become a problem in Britain  
402 due to the cooler climate, but then cases of extensive plant damage in the Rotterdam district of  
403 Holland occurred as a result of photochemical pollutants. Atkins, Cox and Eggleton [22]  
404 subsequently detected summertime elevated ozone levels in a rural district of Berkshire UK, remote  
405 from any large conurbations. Bell and Cox [121] followed up these measurements by recording  
406 visible ozone damage on an ozone-sensitive cultivar of tobacco at the same location and showed  
407 that the injury correlated with ambient levels.

408 For the next two decades research into the impacts of ozone on plants focussed on damage to crops,  
409 with many species identified as showing visible damage and yield reductions [122]. Forest damage in

410 the San Bernadino Mountains, California, in the early 1970s and visible signs of ozone stress in  
411 central European forests led to research into the impacts of ozone on young trees. This work led to  
412 clear evidence that ozone can affect the chlorophyll content, photosynthetic rate [123] and carbon  
413 allocation [124] of trees. Despite this, separating out the effects of ozone, acid deposition and other  
414 stressors on crown condition was a challenge. A number of regional- and continental-scale studies  
415 were devised to address this [125]. However, it was not until the 1990s that interest in impacts of  
416 ozone widened from crops and trees. In 1998 a thorough review of ozone impacts on wild species  
417 highlighted the urgent need to investigate ozone impacts widely on semi-natural vegetation [125].

418 It remains the case that less is known about ozone impacts on semi-natural plants than agricultural  
419 crops, but understanding has developed considerably over the last two decades. Understanding the  
420 mechanisms of ozone impacts in semi-natural ecosystems is complicated by the need to account for  
421 species interactions which can lead to shifts in species composition and losses of biodiversity [126].  
422 Wide variation in the responses of semi-natural species have been reported, as well as intra-specific  
423 variability in ozone sensitivity and heritable differences in ozone responses [126, 127]. These  
424 heritable differences are likely a consequence of previous exposure [126]. Legumes [128] and  
425 summer annual plants [127] have been identified as particularly sensitive to ozone, but beyond this  
426 there remains debate about whether sensitivity can be predicted from plant traits [126].

427 Predicting community responses remains a challenge. Mills et al. [129] used EUNIS (European Nature  
428 Information System) level 4 communities to predict community-wide ozone sensitivity from the  
429 published responses of individual species. They found all 54 of the EUNIS communities they  
430 considered had six or more sensitive species and were thus defined by the authors to be ozone  
431 sensitive. Grasslands had the most sensitive communities followed by heathland, scrub and tundra  
432 and mires, bogs and fens. However, Bassin et al. [130] caution that the risk to low-productivity  
433 perennial grasslands is commonly less than predicted from risk assessments based on individual  
434 species or immature mixtures in chambers.

## 435 **6. Learning from the past and looking to the future**

436 It is clear that we have come a long way in our understanding of the effects of atmospheric pollution  
437 on managed and natural vegetation. Pollution effects on plants have been observed for hundreds of  
438 years, but the identity of the pollutants responsible, the dose response relationships, and causal  
439 mechanisms were unknown until the mid-20<sup>th</sup> century.

440 This paper has reviewed the evidence for four main threats from air pollution: (1) direct phytotoxic  
441 effects from gas phase constituents, (2) indirect effects from deposition of acidifying agents, (3)

442 indirect effects from deposition of nutrients and (4) direct toxicity of ozone. The evidence suggests  
443 that each of these is not controlled by a single mechanism, nor do they operate in isolation. Rather,  
444 there are multiple pathways through which each mechanism may operate, many of which are  
445 occurring simultaneously, but to different degrees, within the same ecosystem [131, 132]. For SO<sub>2</sub>,  
446 direct phytotoxic effects appear to mostly be localized phenomena around point sources, although  
447 historically, when emissions were less regulated, direct effects were likely more widespread [11, 12].  
448 Acidifying and eutrophication effects appear to be widespread and simultaneous phenomena, but  
449 differ in relative magnitude based on a host of factors including the composition of the atmosphere.  
450 Ozone interactions are commonplace because ozone usually occurs in concert with other air  
451 pollutants such as sulphur and nitrogen oxides, which may affect plants simultaneously.

452 Although we have learned much about the possible outcomes related to increasing pollution  
453 deposition, we are only beginning to understand the reversibility of effects [79, 113]. Pollution  
454 deposition has been decreasing through much of Europe and North America since the 1980s and  
455 1990s and much more recently in parts of Asia [133]. These patterns provide an opportunity for  
456 “natural experiments” to learn about the reversibility of these effects, in addition to the few  
457 controlled experiments. Early signs indicate that “fast cycling” processes may recover fairly quickly  
458 (e.g. foliar nutrient balances, nitrate leaching, etc.), while “slow cycling” processes may take several  
459 decades or more to recover (e.g. soil N pools, base cation availability, plant communities) [55, 64, 77,  
460 78]. In some cases, an alternative stable state may be reached and recovery may not occur over  
461 timescales relevant to public decision-making (e.g. several decades) without management  
462 intervention [79, 134].

463 Looking forward, it is clear that there are multiple paths that different countries may follow either  
464 intentionally or unintentionally. Many developed nations are beginning to reduce pollution  
465 emissions and deposition through successful environmental policies, although the ultimate  
466 outcomes of many of those national policies on the environment are undetermined because the  
467 timescales of some anticipated changes are expected to take many decades and so far, insufficient  
468 time has passed. Developing nations may tread the same paths as developed nations, increasing  
469 both economic activity and air pollution emissions leading to regional reductions in biodiversity, soil  
470 acidification and eutrophication, only later to consider recovery, or they may evade the worst by  
471 learning from history. The axiom that “an ounce of prevention is worth a pound of cure” is  
472 especially germane, as these developing nations, with the right international support and domestic  
473 incentives, have the opportunity to avoid many of these adverse effects of air pollution; for example  
474 the costs of renewable energy have reached grid parity with many fossil fuels in many nations [135].  
475 Regardless, the global scientific community has made significant strides in understanding and

476 addressing the ecological effects from air pollution, though much work remains as we assess the  
477 reversibility of these effects and the transferability of these lessons to understudied ecosystems.

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#### 819 **Figure legends**

820 Figure 1. Global emissions of SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub> and NMVOC between 1750 and 2010. Adapted from Hoesly  
821 *et al.* [6].

822 Figure 2. Distribution of the nitrogen tolerant lichen *Xanthoria parietina* showing records in 10 km  
823 squares between 1951-1970 and between 2001-2020 (British Lichen Society).