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Atmospheric nitrogen compounds—issues related to agricultural systems

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Abstract

Workable pollution abatement policies and effective legislation must be based on sound science. However, despite many years of research, there are still uncertainties about the effects of atmospheric nitrogen compounds on crops and other vegetation. This paper reviews the current state of knowledge of the main compounds, focussing on the concentrations and combinations of pollutants that occur in rural areas. The sources, concentrations and effects of oxidised, reduced and organic nitrogen compounds are considered in turn, then the effects of deposited nitrogen on ecosystems are discussed. Research priorities on the effects of deposited nitrogen in Europe and the USA are compared. Finally, the review leads to a list of issues for discussion and recommendations for research.

Keywords: NO_x; NO_2; PAN; NH_y; HNO_3; Atmospheric pollution; Abatement policy

1. Introduction

Fourteen years after the National Crop Loss Assessment Network reported on the effects of air pollution on crop yields (Heck et al., 1988), ozone is still the most important gaseous air pollutant in the developed world. However, in the intervening years concern has risen about the effects of atmospheric and deposited nitrogen compounds on national and global scales (Vitousek et al., 1997; Mansfield et al., 1998) and there are several major research programmes, especially in Europe. As successful abatement policy must be based on good science this paper reviews the current state of knowledge of the effects of atmospheric nitrogen compounds at the concentrations that are found in agricultural regions. The sources, rural concentrations and effects of oxidised, reduced and organic nitrogen compounds are considered in turn, then the effects of deposited nitrogen on ecosystems are discussed. Research priorities on deposited nitrogen in Europe and the USA are compared. Finally, the review leads to a list of issues for discussion and recommendations for research.

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2. Sources, concentrations and effects of fixed atmospheric nitrogen compounds

2.1. Oxidised nitrogen compounds

Table 1 lists the main oxidised atmospheric nitrogen compounds, their sources, range of concentrations in rural areas, and effects. The two most important compounds are NO and NO_2.

The main source of NO and NO_2 is combustion of fossil fuels. In most countries, regulation of NO_x emissions has focussed on power stations and urban vehicular sources. In the USA, farm non-road diesel engines emit only about 4% of the total NO_2 (Fig. 1, EPA, 2000) but it is released in rural areas where crops are grown. In addition, the use of fertilizers increases emissions of NO from soil (Skiba et al., 1998) so agriculture makes a significant contribution to the rural NO_x budget and therefore to the ozone problem (Aneja et al., 1998).

The productivity of many ecosystems is limited by the supply of nitrogen so it is important to know whether low concentrations of NO_x might contribute to plant nutrition, possibly increasing growth. There are a few reports of positive effects of NO_2 under controlled environment conditions but most indicate either no effect or deleterious effects (Wellburn, 1990). A simple calculation suggests that at current rural concentrations, NO_2 is unlikely to act as a significant source of nitrogen for annual crops. Assuming a

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Table 1
The main oxidised atmospheric nitrogen compounds, their sources, range of concentrations in rural areas and possible effects

Sources	Compounds, long-term mean concentrations in rural areas	Environmental effects
Lightning, fuel combustion, emission from soil + photochemistry	NO (< 1 to >20 ppb) (< 1.2 to 24 μg m^{-3})	NO is a natural plant product with a signalling role. Emitted from soil and contributes to rural NO_x budgets. Suggested effects on plants at low concentrations via interactions with O_3?
	NO_2 (< 5 to 20 ppb) (9 to 38 μg m^{-3})	NO_2 may affect crop yield, interacts with SO_2 and O_3? Promotes growth at low concentrations?
	NO_x	NO_x generates tropospheric O_3.
	HNO_2 (rural 0.01–1.0 μg m^{-3})	HNO_2 generates OH radicals in atmosphere.
	HNO_3 (24 h averages < 0.4–0.8 ppb, peaks >10 ppb: < 1.0–2.0 μg m^{-3}, peaks >25 μg m^{-3})	HNO_3 alters cuticular waxes, Growth effects on perennials?
	Nitrates	Nitrates ≡ $PM_{2.5}$ affects human health

Data from: NAS (1977), Meyer and Hicks (1988), Bytnerowicz et al. (1998), Lammel and Cape (1996), Mansfield et al. (1998), McCulloch et al. (1998), and Danalatos and Glavas (1999).

rural NO_2 concentration of 5 ppb (ca. 9.4 μg m^{-3}) and a deposition velocity of 2 mm s^{-1}, deposition to a crop canopy will be around 6 kg N ha^{-1} $year^{-1}$, or 1.5 kg during a 3-month growing season. Even if this were all taken up into the leaves, it would not represent a significant addition on top of the normal rates at which nitrogenous fertilizer is used.

Most of the published research on the effects of NO and NO_2 used concentrations much higher than those that occur in rural areas (NAS, 1977; Legge et al., 1980; Wellburn, 1990; Bytnerowicz et al., 1998) so it is not relevant to most agricultural situations or to natural ecosystems. Furthermore, although there is good evidence of interactions between NO_x and pollutants such as SO_2, the evidence suggests that they occur only at concentrations well above those found in rural areas (Bytnerowicz et al., 1998). However, Mehlhorn and Wellburn (1987) suggested that NO may increase ozone injury when it is present in very low concentrations (ca. 2 ppb, 2.4 μg m^{-3}) due to production of ethylene and its interaction with ozone. Investigating this interaction is technically challenging because adding NO reduces the ozone concentration so it is difficult to produce identical ozone concentrations in different experimental treatments. In attempting to verify Mehlhorn and Wellburn's theory, Nussbaum et al. (1995) found that adding about 6.9 ppb NO lowered the ozone critical level (i.e., the exposure above which there is a significant effect) from 11,292 to 8749 ppb-h. The authors considered that the data showed no evidence to support the theory, however, they did suggest a NO-ozone interaction on wheat yield at low ozone concentrations. Later Nussbaum et al. (2000) concluded that "the effects of NO in the low ppb range are mainly due to shifts in gas phase chemistry at the leaf-atmosphere interface and that they are negligible in pollution climates dominated by ozone".

2.2. Air quality standards for NO_x

The USA has a system of primary and secondary air quality standards, the former being for the protection of human health and the latter for protection of vegetation. In the case of NO_x, the primary and secondary standards are the same, being an annual mean of 53 ppb (Federal Register, 1996). Clearly, if this standard is reliable, then it suggests that there are few agricultural or wilderness areas where there is an impact of NO_x. However, in Europe, a critical level has been set for effects of NO_x on vegetation that is much lower than the US standard. It is cited in CLAG (1996) in the following terms: "Because of new knowledge of the important phytotoxic effects of nitric oxide (NO), the present critical level is a combined value for NO plus NO_2. The critical level is 30 μg m^{-3} as an annual mean and applies to NO_x . . .for all categories of vegetation. The level should only be applied where . . .SO_2 and O_3 are close to their critical levels. —In areas where the critical level is exceeded it is believed that there are likely to be adverse direct effects of NO_x on vegetation, although the nature of the effects is uncertain".

The large difference between the USA standard and the UN ECE critical level is symptomatic of the fact that they are

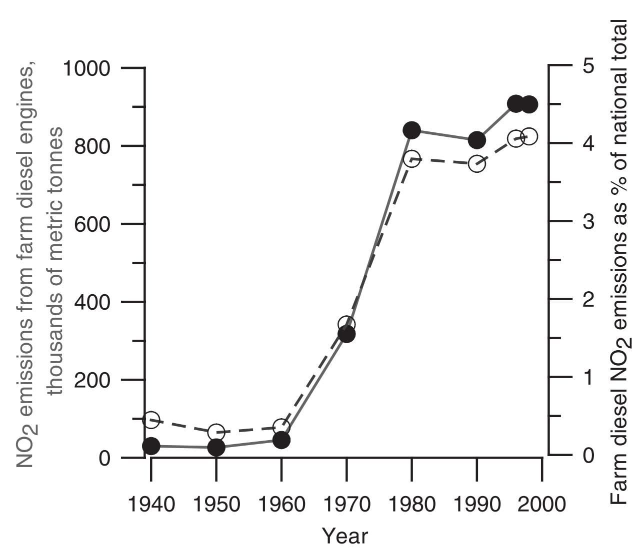

Fig. 1. Annual NO_2 emissions from farm non-road diesel engines (left hand Y axis, closed circles, solid line) and the same emissions expressed as a percentage of the national total NO_2 emissions (right hand Y axis, open circles, dashed line). Data from EPA (2000).

based on very little research and that hardly any of that research used concentrations that were in the low ppb range. Furthermore, neither deals satisfactorily with the effects of interactions between NO_x and other pollutants. The US standard ignores interactions. The critical level refers to interactions but is vague—should the critical level be applied only when *both* ozone and SO_2 are close to their critical levels or when one is close, and how close do they have to be? A further weakness of both the standard and the critical level is that it is not possible to quantify the effects of exceedences. These shortcomings make a compelling case for more research on interactions between NO, NO_2 and other pollutants at the low concentrations that occur in rural areas.

2.3. HNO_3

Concentrations of nitric acid vapour in rural areas are generally low, with 24 h averages between about 0.4 and 0.8 ppb (1.0–2.0 μg m^{-3}) but records are sparse and more data are needed for a range of climates and vegetation types. Although the concentrations are low, the deposition velocity (Vg) of HNO_3 is high. Meyer and Hicks (1988) data suggest that the Vg for HNO_3 to crops is roughly 6 times that of O_3. In their work, a mean HNO_3 concentration of 0.7 μg m^{-3} (ca. 0.28 ppb) gave deposition of 1.0 kg ha^{-1} direct to a crop canopy over a 12-week growing season. Bytnerowicz et al. (1998) reviewed research on the effects of HNO_3 but the little work that has been done used high (>50 ppb) concentrations and short exposure times. As HNO_3 is deposited on the cuticular surfaces that act as a defence against pests and diseases, further work is needed using long-term exposures and perennial species. In addition, interactions with ozone should also be investigated because they co-occur in photochemical smog (Bytnerowicz et al., 1998).

3. Reduced nitrogen compounds

Table 2 shows the sources, rural concentrations and effects of reduced nitrogen compounds. Acute exposure to

Table 2
The reduced atmospheric nitrogen compounds, their sources, range of concentrations in rural areas and possible effects

Sources	Compounds, long-term mean concentrations in rural areas	Environmental effects
Animal wastes, soil emissions, accidental leakage—e.g., refrigeration	NH_3 (0.1 to >10 ppb, >56 ppb near animal units) (0.14 to >14 μg m^{-3}, >80 μg m^{-3} near animal units)	Odours, Effects on plants—negative and positive?
		Ammonium salts ≡ $PM_{2.5}$ human health

Data from: Fangmeier et al. (1994), Bytnerowicz et al. (1998), Pitcairn et al. (1998), Sutton et al. (1998), and van der Eerden et al. (1998).

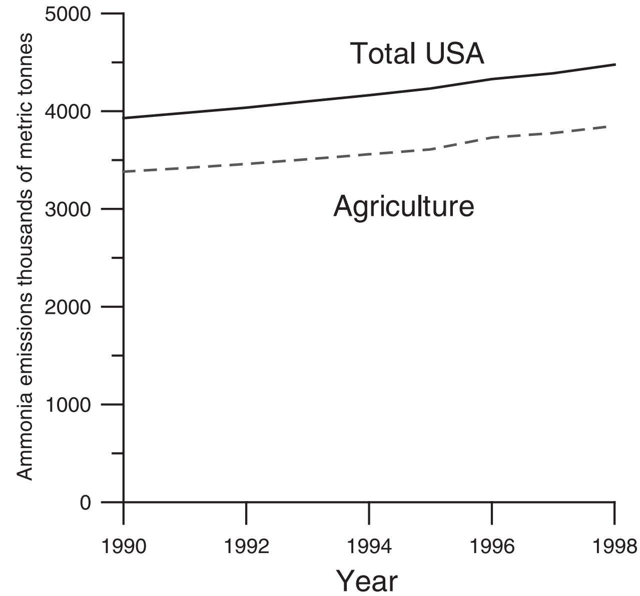

Fig. 2. Annual total ammonia emissions (solid line) and contribution from agriculture (dashed line). Data from EPA (2000).

ammonia occurs occasionally, usually as a result of leakage from refrigeration plants, and such incidents may increase with the change in refrigerants from CFCs and HCFCs to ammonia. However, agriculture is the major source of ammonia in North America and Europe. Fig. 2 shows that agriculture contributes about 90% of the atmospheric ammonia burden in the USA. The sources are animal production and emissions from slurries and manures.

Acute plant damage has been reported in the Netherlands near intensive animal units and there is evidence that ammonia at ca. 100 μg m^{-3} may reduce a plants resistance to low temperature (Fangmeier et al., 1994). Fig. 3 shows

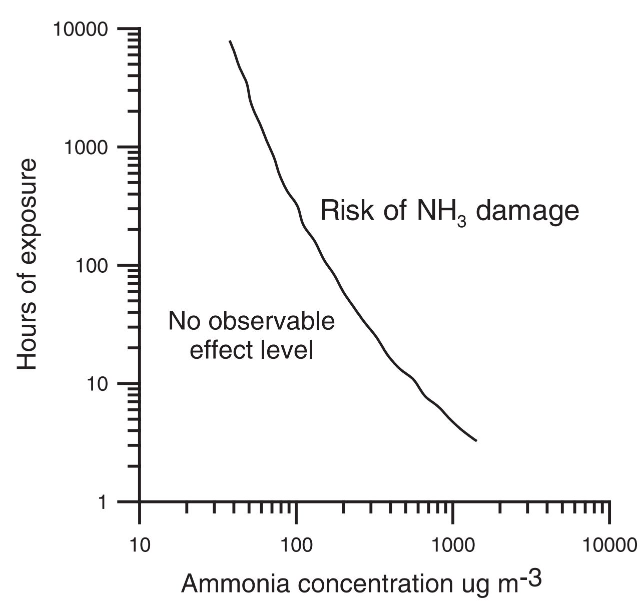

Fig. 3. Estimated No Observable Effect Level for ammonia injury to plants. Re-drawn from van der Eerden et al. (1998).

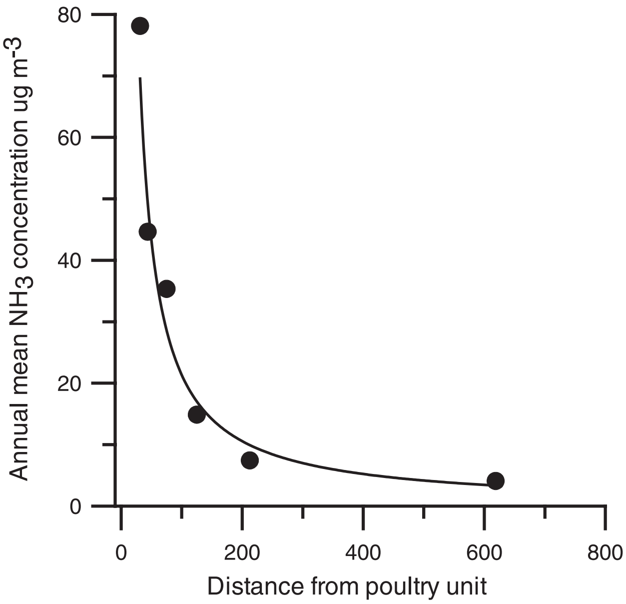

Fig. 4. Change in annual mean atmospheric ammonia concentration with distance from a poultry unit. Re-drawn from Pitcairn et al. (1998).

the estimated ‘No Observable Effect Level’ estimated by van der Eerden et al. (1998). Because of rapid deposition, the direct effects of the gas are restricted to close to source. The fall-off in concentrations with distance is illustrated in Fig. 4 by data from Pitcairn et al. (1998). In this case, the annual mean concentration was 78 μg m^{-3} at the source but fell rapidly with distance to about 4 μg m^{-3} at 600 m. The effect of the NH_3 and deposited nitrogen was to change the flora around the source, promoting the growth of grasses, and eliminating other species such as mosses and ferns. At lower concentrations, typical of those remote from sources, it is unlikely that NH_3 has either negative or positive effects on crops. However, the effects of deposited reduced nitrogen can be important (see last paragraph in Section 5).

4. Organic nitrogen compounds

There are many different organic nitrogen compounds in the atmosphere, mostly in very low concentrations. The main compounds, their sources, rural concentrations and effects are shown in Table 3. PAN is the best known and most critical of these compounds.

Peroxyacyl nitrate (PAN) is the best known of this group, mostly because of early research in California (Temple and Taylor, 1983; Grosjean, 1984) and more recent work in the Far East (Temple et al., 1998; Cape, 1997). PAN is thermally unstable (Temple et al., 1998), decomposing more rapidly at higher temperatures. Consequently, the highest concentrations, and most reports of injury, occur in urban and peri-urban areas where the pre-cursor concentrations are high. However, at lower temperatures, PAN is more stable so it persists longer in the cooler seasons and at higher latitudes. Cape and McFadyen (in press), for example, reported that at a rural site in Scotland the annual average was 0.1 to 0.15 ppb but that there were episodes up to 3 ppb for several hours due to long-range transport.

There are many reports in the literature of acute injury being caused by concentrations higher than 20 ppb but, as might be expected, there are far fewer studies using lower concentrations. However, recent work in Japan and Taiwan indicates that exposure to concentrations as low as 3–10 ppb, for a few hours, may cause acute injury (Temple et al., 1998; Cape, 1997). Thus, it is possible that effects of PAN are more widespread in the cooler rural areas than has been previously appreciated.

A complication in assessing the effects of PAN is the apparent difference in its toxicity in the field and in controlled environments. Temple et al. (1998) summarised work that suggests that plants develop symptoms at PAN concentrations 2–3 times lower in the field than in controlled environments. This was not apparently due to an interaction with ozone but it reinforces Cape’s (1997) conclusion that there is a need for long-term experiments involving low concentrations of PAN and the other photochemical oxidants.

5. Deposited nitrogen compounds and their effects on vegetation

Atmospheric gases and particles are eventually deposited on to vegetation, land and other surfaces. The range of deposition of nitrogen compounds observed in Europe and the USA, and observed environmental effects are shown in Table 4. Experience in Europe over the last 20 years has shown that deposited nitrogen has profound effects on sensitive ecosystems. These include acidification, changes in species composition and loss of diversity (Bobbink et al.,

Table 3
The main organic atmospheric nitrogen compounds, their sources, range of concentrations in rural areas and possible effects

Sources	Compounds, long-term mean concentrations in rural areas	Environmental effects
Fuel combustion + photochemistry	Peroxyacyl nitrate (PAN) annual mean < 0.2 ppb but episodes of 2–5 pbbv, maxima up to 8 ppbv	PAN involved in O_3 chemistry. Acute exposures cause visible injury. May interact with O_3. Effects of concentrations < 10 ppbv not well known.
	Other compounds related to PAN	Low concentrations, some more toxic than PAN.
	Particulate organic nitrates (PON)	Mostly very low concentrations (ng m^{-3}) effects on crops unknown.
	Amines	?
	Nitrophenols	?
	NitroPAHs	?

Data from: Temple and Taylor (1983), Grosjean (1984), Cape (1997), Temple et al. (1998), and Cape and McFadyen (in press).

Table 4
The rate of deposition on oxidised and reduced nitrogen compounds recorded in Europe and the USA, and their potential effects

Rates of deposition, wet + dry	Environmental effects
Oxidised N 0.1 to >20 kg ha^{-1} year^{-1}	Acidification. Eutrophication of N-limited habitats. Promotion of NO and N_2O emissions from soil.
Reduced N <1 to >40 kg ha^{-1} year^{-1}	

Data from: INDITE (1994), Wedin and Tilman (1990), Bobbink (1998), and Bobbink et al. (1998).

1998). Nitrogen-limited communities owe their particular assemblages of species to the low nutrient supply so when the nitrogen status is increased, aggressive nitrophilous species expand or invade and out compete those that are unable to make use of the extra nitrogen. Although nitrogen-limited communities are not usually of any great agricultural value, they often have high conservation status so their loss or degradation may be considered important in view of the world-wide concern over biodiversity. In Europe, the effects of deposited nitrogen are considered so serious that the signatories of the UN Economic Commission for Europe, Convention on Long-Range Transboundary Air Pollution have developed critical loads for the biological effects of deposited nitrogen on specific ecosystems (Table 5). The values are based on empirical evidence but values for the most sensitive ecosystems are being verified by long-term nitrogen addition experiments. Although there will undoubtedly be continued refinement of the values, there is broad agreement on the critical loads for the most sensitive ecosystems: tundra, certain shallow pools and ombrotrophic bogs. Rates of deposition of nitrogen above about 5 kg ha^{-1} year^{-1} will change the species composition of the most sensitive ecosystems. This rate of total nitrogen deposition is exceeded in many parts of Europe and the USA.

Table 5
Nitrogen critical loads for biological effects (Bobbink et al., 1996)

Ecosystem	Critical load, kg N ha^{-1} year^{-1}
Forests	
Acidic coniferous	7–20
Acidic deciduous	10–20
Calcareous	15–20
Heathland	
Lowland dry heath	15–20
Moorland	10–20
Tundra	5–15
Grassland	
Calcareous	15–35
Neutral-acid	20–30
Wetland	
Mesotrophic fen	20–35
Ombrotrophic bogs	5–10

In the USA, there are several studies that demonstrate the effects of nitrogen enrichment on species composition and diversity. For example, Wilson and Tilman (1991), Wedin and Tilman (1990) and Haddad et al. (2000) have demonstrated experimentally the effects of nitrogen on plant and insect species diversity. Some data of Wedin and Tilman (1990) are shown in Fig. 5. Baron et al. (2000) produced evidence that 3–5 kg of nitrogen deposited in lakes of the eastern frontal range of the Rockies has changed diatom communities towards domination by more eutrophic species. In California, Weiss (1990) has shown how dry deposition of nitrogen encourages grass invasion of serpentine soils leading to loss of the food plant for the checkerspot butterfly and a subsequent population crash of the insect. However, these examples are relatively rare; the major concern in the USA appears to be on nutrient cycling, acidification, the nitrogen saturation concept, watershed chemistry and greenhouse gas fluxes. Most publications are concerned with forests and there is little reference to nitrogen-sensitive ombrotrophic ecosystems or grasslands. In a major review of ecosystem responses to nitrogen excess, Fenn et al. (1998) listed ecosystem responses as: nitrate leaching and export; eutrophication of estuaries; toxicity of surface waters; foliar nutrient responses; nitrogen mineralization and nitrification; effects on soil organic matter; soil acidification; and greenhouse gas fluxes. Effects on species composition or loss of diversity in terrestrial or freshwater ecosystems were not mentioned, yet some species, notably those of ombrotrophic habitats such as mosses and lichens, are affected long before there is nitrogen saturation or detectable effects on nutrient cycling or soil acidity. Similarly, Environmental Defense (Gutt et al., 2000), which made an otherwise comprehensive

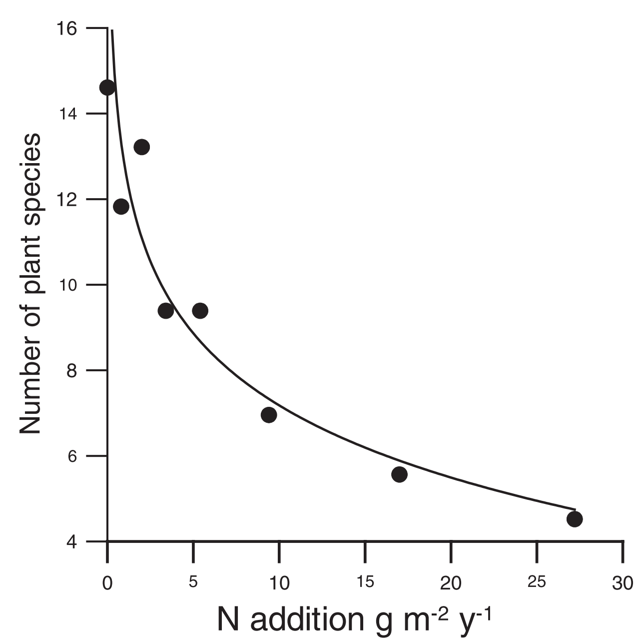

Fig. 5. Example of the effects of 12 years of nitrogen fertilizer addition on the number of plant species in a Minnesota grassland. Data from Field C, Wedin and Tilman (1990).

case for reducing NO_x pollution, did not mention loss of diversity as being an effect of enrichment. Seen from a European perspective, the danger is that important effects on species composition and diversity are probably being overlooked in the USA. Therefore, important questions are: how widespread are effects of deposited nitrogen on biodiversity of nutrient poor ecosystems in the USA and are current research programmes constructed in such as way that they will reveal effects on biodiversity?

6. Conclusions: issues and topics for research

- Assessing the effects of air pollution can only be done if there is a comprehensive record of the atmospheric concentrations of all the potentially phytotoxic pollutants in the whole range of rural environments. However, although ozone is routinely monitored, there is a lack of systematic monitoring of HNO_3, PAN, NH_y and in some places NO_x, so the pollution climate over most agricultural and wilderness areas is not sufficiently well defined to inform the research community.
- Information about the effects of low concentrations of nitrogenous pollutants and pollutant mixtures is sparse and often contradictory so air quality standards do not have a firm, scientific basis. Consequently, research is needed using realistic mixtures and a range of species. This presents a considerable challenge because effects cannot be assessed by fumigating with one pollutant at a time (Cape, 1997) and there are so many combinations that the task of working with mixtures is technically and economically daunting. Furthermore, the pollution climates and vegetation of the regions of the USA and Europe differ so profoundly that no single study in one location could provide the data to inform the regulatory process.
- There is a disparity between the USA air quality standard for NO_x and the UN ECE critical level, which is probably a reflection of the sparsity of experimental data. Although the UN ECE critical level attempts to encompass interactions and pollutant mixtures, it is vague and ill defined. Neither the USA standard nor the critical level for NO_x allows a quantitative assessment of the effects of exceedences. If such standards are to stand up to serious review and protect vegetation, then these shortcomings must be addressed.
- Interest in, and research on, the effects of deposited nitrogen has been increasing over the last 20 years, especially in Europe where some of the problems are more acute than in the USA. In Europe, the focus of research is on the effects of nitrogen in reducing biodiversity. The concerns are such that critical loads have been developed for different ecosystems. In the USA, the focus is at the ecosystem level, research on effects on biodiversity is relatively uncommon and there are no air quality standards designed to protect biodiversity from deposited nitrogen. Based on European experience, important questions for the USA are how widespread are effects of deposited nitrogen on biodiversity of nutrient poor ecosystems in the USA and are current research programmes constructed in such a way that they will reveal effects on biodiversity?

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