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Keynote Paper - Atmospheric Inputs of Nitrogen

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Introduction

The requirement for estimates of the terrestrial inputs of fixed nitrogen for Europe with a spatial resolution of between 5 km x 5 km and 50 km x 50 km places great demands on the monitoring information available and our knowledge of processes. It also provides a clear focus for the monitoring and process based research to provide both the input and an estimate of the uncertainty in inputs and its spatial variability.

For the estimates of critical loads exceedance maps for nitrogen, maps of the total inputs are of course essential. In this brief review the focus is on what is known, and can be mapped, what is not known because insufficient monitoring is in place, and lastly what is not known because the processes are not understood or for which key parameters are uncertain. In addition, because the principal applications of mapped data are to estimate critical loads exceedance, the issue of the scale dependence of deposition estimates and the extent to which current approaches can be extended to provide fine scale or land use specific resolution is considered qualitatively and to a limited extent quantitatively.

Wet Deposition

The monitoring networks for wet deposition provide good estimates of the precipitation weighted concentrations of major ions in simple terrain throughout Europe. There are areas of uncertainty, mainly as a consequence of complex terrain such as in the Alps and in the mountains of north and west Britain and Scandinavia. For relatively simple terrain the wet deposition estimates based on the product of a very spatially detailed precipitation field and the much less variable spatial pattern in weighted mean concentration are entirely adequate for the mapping procedures at 10 km x 10 km scale and upwards. The problems in wet deposition are due primarily to complex terrain. In such areas orographic effects modify the precipitation scavenging process by, for example, " seeder-feeder" effects and these lead to marked increases in wet deposition in the uplands of the UK and western Scandinavia. In the seeder-feeder effect, relatively clean precipitation from higher level cloud "washes" out more polluted cloudwater from lower level orographic (feeder) cloud. The mechanisms of orographic enhancement have been studied in detail and simple parameterization schemes have been developed to modify wet deposition maps for this effect over the UK. While intensive field studies have been made at a range of mountain locations to examine the processes and determine the variability in concentrations of major ions in precipitation, orographic cloud and wet deposition with altitude, these have generally been 2 dimensional transects through "simplified complex terrain". For example, trends in concentration and deposition have been observed along a transect over a simple ridge at Great Dun Fell in the Pennines of northern England. Larger mountains in more complex environments have been studied, as for example, on the western slopes of Ben More Assynt in Sutherland, north west Scotland. In all of the campaign studies, the effect of seeder-feeder enhancement in precipitation and concentration have been observed. The wide range of study sites for the field studies is illustrated in Figure 1 which shows the sites at which the measurements have been made. The individual profiles of concentration and deposition lie outside the scope of this brief review but have been simulated using models of the processes (Choularton *et al.* 1988). However a summary of the results of the campaigns (each of typically 4 to 6 weeks sampling of each precipitation event) and of the longer term monitoring of hill cloud and precipitation are given in Table 1.

The overall mean enhancement in concentration of scavenged feeder cloud/seeder rain of 2.3 for SO_4^{2-} from the table provides support for the simplifications made to model orographic enhancement of wet deposition at the UK scale. Such approaches are necessarily crude and do not allow for fine scale (<20 km) effects of orography on deposition patterns. The wet deposition maps provided with orographic enhancement are therefore restricted in spatial detail to a 20 km x 20 km grid and cannot be used to obtain the fine scale wet deposition inputs for exceedance estimates. (The specific effects of spatial scale averaging on exceedance estimates are considered later.)

It is a matter of importance and not a mere coincidence that the geographical areas in which complex terrain complicates the scavenging processes, and introduce extra uncertainty into input estimates, are also the areas of greatest critical levels exceedance for wet deposition.

Cloud Droplet Deposition

The importance of these processes for inputs to high altitude forests were little known or understood in the early years of acidic deposition research. The interest in processes at high elevation sites led to research on the processes of turbulent deposition of droplets onto natural surfaces (Dollard *et al.* 1983; Fowler *et al.* 1989; Gallagher *et al.* 1988). These studies demonstrated the dependence of deposition rates on droplet size and are illustrated in Figure 2 from the work of Gallagher *et al.* (1991) which showed that for the bulk of the liquid water in orographic cloud on windy uplands of Northern Europe the droplets are captured by the vegetation at rates similar to those of momentum, and can therefore be modelled from the relatively well known (and monitored) data on wind and roughness length.

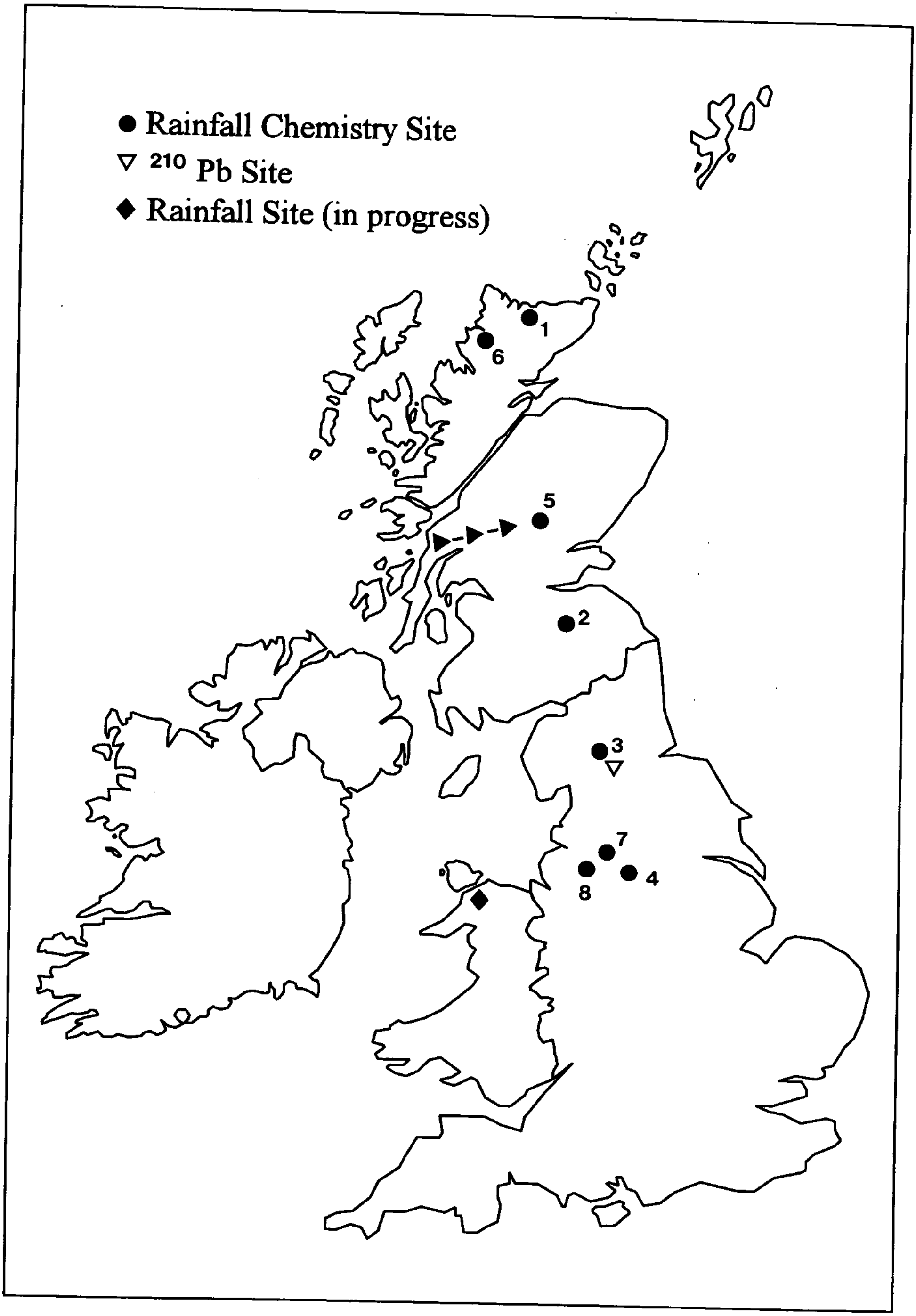

Figure 1 **Sites of orographic enhancement of wet deposition in the UK**

Table 1 Summary of results of long-term monitoring measurements and campaign studies

		Concentration ratio hill cloud/seeder rain			
Long-term measurements	Site No.	SO_4^{2-}	NO_3^-	NH_4^+	Cl^{-1}
Halladale	1	8	12.8	1.1	14.4
Dunslair	2	7.7	7.3	7	7
Great Dun Fell	3	4.51	9.72	4.05	5.68
Holme Moss	4	4.55	6.6	4.21	10.4
Short-term (campaign) measurements					
Glen Bruar	5	2.9	1.89	2.34	3.95
Assynt	6	3.56	1.53	-	3.35

		Concentration ratio scavenged feeder cloud/seeder rain			
Long-term measurements	Site No.				
Halladale	1	2.5	3.1	1.1	4.1
Great Dun Fell	3	1.46	2.8	4.6	1.58
Holme Moss	4	2.48	2.05	3.04	1.96
Short-term (campaign) measurements					
Great Dun Fell	3	3.3	3.5	4.7	4.4
Holme Moss (Saddleworth Moor)	7	1.98	2.33	2.15	1.97
Winter Hill	8	1.88	2.74	2.2	5.13
Very long-term measurements ^{210}Pb at Great Dun Fell 2.2					

To estimate the actual deposition rates of nitrogen in cloud droplets in the uplands from the above work, it is necessary to quantify the spatial variability in cloud frequency and the concentration of the NO_3^- and NH_4^+ in cloud. These quantities are not widely monitored in Europe, although there are 4 sites in Britain (Dunslair Heights, Great Dun Fell, Holm Moss and Halladale) for which more than 1 year of continuous monitoring of cloud and precipitation composition and frequency have been made. Likewise, there is a similar number of sites in Germany at moderate elevation for which similar data are available. These data show a consistent height dependence of the concentrations of major ions as illustrated in Figure 3.

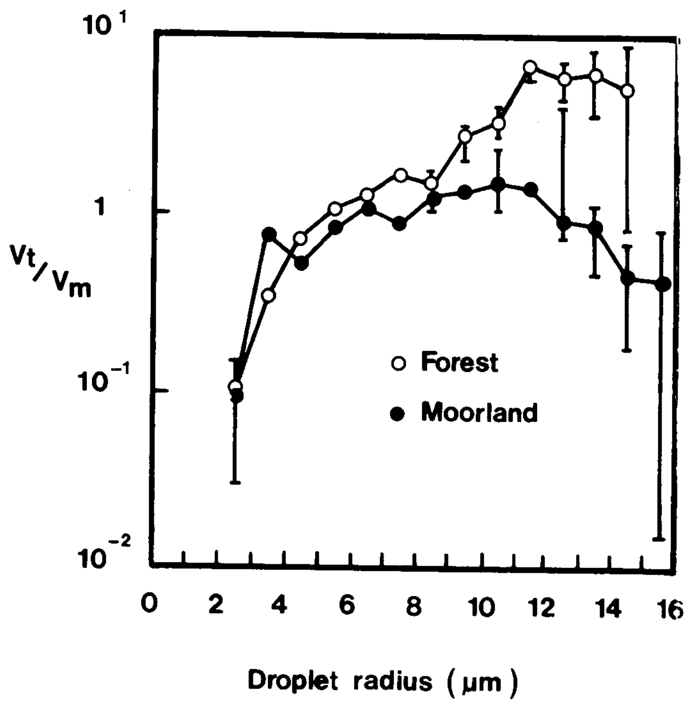

Figure 2 **Effects of droplet size in the range 2-15 m on the ratio of turbulent deposition velocity (V_t) of cloud droplets to that of momentum (V_m) (where $V_m = r_{am}^{-1}$ for forest and moorland from measurements by Gallagher *et al.* 1991).**

Extending the existing measurements to regional scales presents a problem, and to provide a consistent mapping approach for the UK, the relationship between concentrations of NO_3^- and NH_4^+ in hill cloud and those in precipitation have been used together with a map of cloud frequency (Weston, 1991) and wind velocity and land use data from national UK data bases. Estimates of the regional importance of cloud droplet deposition have not been made for other European countries.

A missing component of this area of work has been that of radiation fogs, which also contribute to the deposition budget. Key detailed fog chemistry measurements have been made in the Po Valley (Fuzzi *et al.* 1988). These have been extended to estimate fog water deposition, but only to a very limited extent and more clearly needs to be done in other countries. It may be much more important for the assessment of effects through exposure of vegetation to very large concentrations than as a contribution to the deposition budget.

Dry Deposition

Gases

The key nitrogen containing species are NO_2, NH_3 and HNO_3. Although other species including NO, HONO and PAN are exchanged at the surface, the rates of exchange are small and make only a minor contribution to ecosystem inputs. On continental and global scales the emission of NO from soils makes a major contribution to the oxidized nitrogen budget of the atmosphere and the approaches of Williams *et al.* (1992) provide a helpful if rather empirical method to estimate

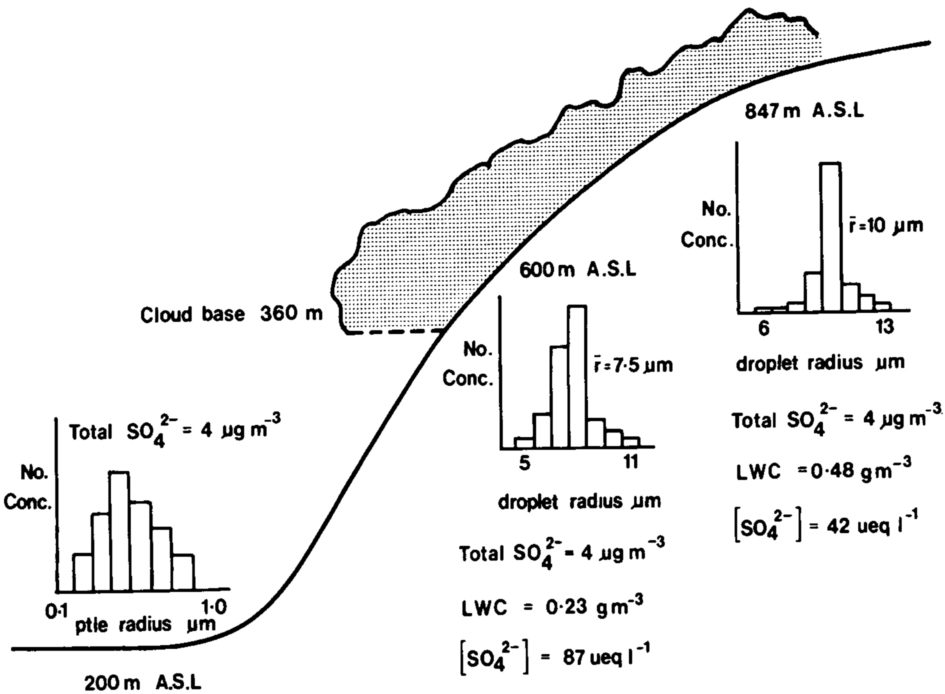

Figure 3 **An illustration of the changes at Great Dun Fell in particle size and ionic concen tration as aerosols are activated into cloud droplets and grow as they are advected up the hillside. (From Fowler *et al.* 1991)**

annual fluxes. Similar simplistic approaches by Skiba *et al.* (1992) for the UK suggest that this source contributes only *ca* 5% of the national emissions from all sources.

NO_2

A mechanistic basis for NO_2 deposition has developed from the large field measurement campaigns of the last 8 years and laboratory studies to investigate processes at the leaf level. The measurements of Hargreaves *et al.* (1992) for example show that for agricultural crops during the growing season, rates of NO_2 exchange are regulated by stomata, and no significant mesophyll (internal) resistance to deposition is present. Such measurements have been used to parameterize a big-leaf resistance model to qualify the spatial variability in NO_2 dry deposition
over regional scales (Duyzer & Fowler, 1994).

A major complication in the provision of field measurements of NO_2 dry deposition rates is chemical reaction in the lowest few metres of the atmosphere in which gradients in concentrations are used to infer fluxes. The reactions between NO, NO_2 and O_3 in the surface

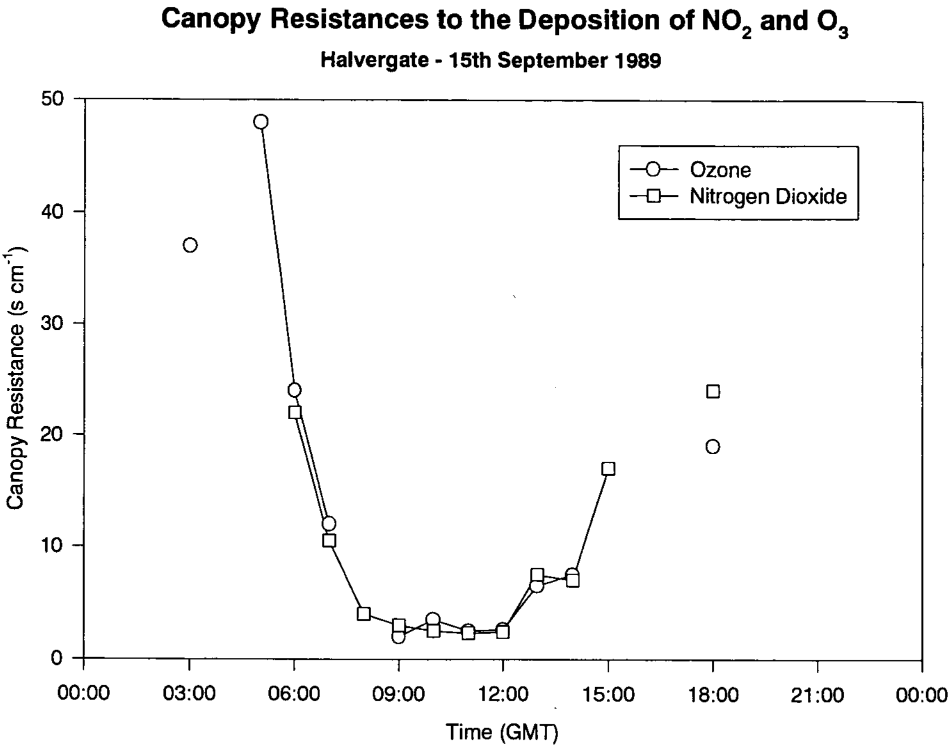

Figure 4 **Canopy resistances to the deposition of NO_2 and O_3 measured at Halvergate in East Anglia, September 1989.**

layers in the presence of upward fluxes of NO from soil microbial processes and NO_2 deposition to the ground, complicate the simple interpretation of fluxes from the gradients and require a more complex analysis. Such approaches have been developed by Kramm *et al.* (1991), Gao *et al.* (1991) and by Duyzer *et al.* (1995) among others. All approaches make considerable demands on the quality of field data and the more pragmatic approach of Duyzer *et al.* offers a realistic opportunity for making progress in the underlying science with the equipment currently available. The strong dependence of NO_2 uptake on stomatal opening lead to an assumption in

the modelling that leaf surface uptake is negligible. New measurements in winter conditions over moorland of NO_2 deposition show that small but significant rates of NO_2 deposition occur at times when stomata are closed (during cold winter weather over senescent moorland). An example of these recent NO_2 dry deposition data is shown in Figure 5.

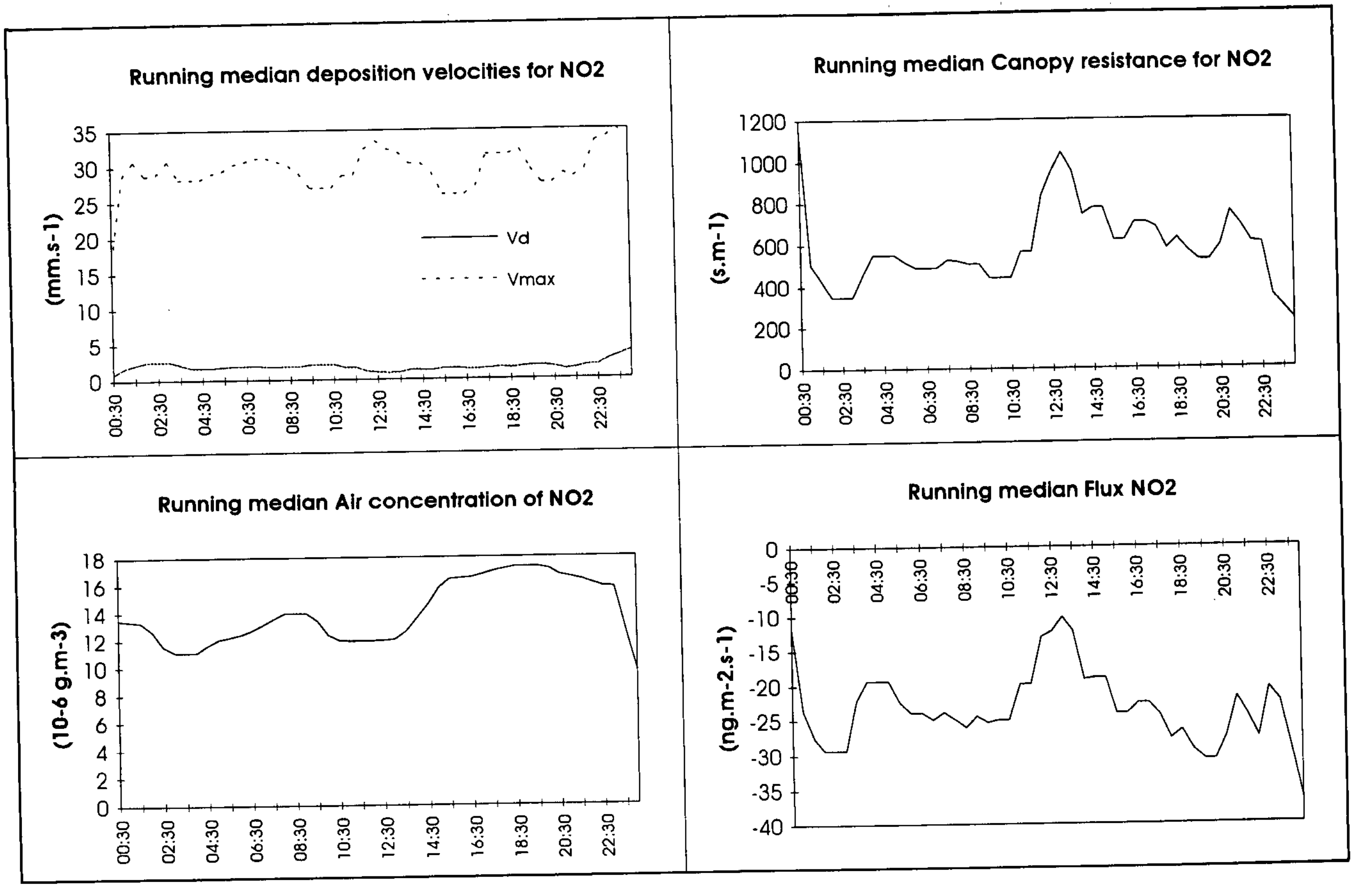

Figure 5 NO$_2$ measurements at Authencorth Moss, near Edinburgh (LIFE site) on 4 January 1995. a)Running median deposition velocities;b) Running median canopy resistance; c) Running median air concentration; d) Running median flux.

Application of the existing simple parameterization scheme for NO_2 deposition in the UK leads to an annual input flux of about 100 kt N. Unlike the similar modelling exercise for SO_2 the data are not supported by an NO_2 concentration and dry deposition network with dedicated rural monitoring stations and a number of continuous monitors. The concentration field has been provided by two intensive (6-month) campaigns of NO_2 diffusion tube measurements (1986 and 1991) throughout the UK. For the larger scale (European) estimates of concentration and dry deposition fields, the monitoring data are even more patchy, with excellent data for the Netherlands and parts of Germany and few available data for large areas of France and Spain. In the absence of monitoring data, the concentration fields provided by EMEP have generally been applied. The upgrading of NO_2 monitoring and development of a continuous flux measurement stations are clear priorities to assess oxidized N deposition in Europe.

HNO_3, HONO, PAN

The field data on deposition processes support the treatment of natural surfaces as perfect sinks for nitric acid (Muller *et al.* 1992). Climatological (or weather) data can be readily adapted with land use to map spatial variability in deposition rates. What is missing is an adequate measurement network for HNO_3 concentrations. The few monitoring stations available within the EMEP network and additional measurements from research studies show that in northern Europe, HNO_3 makes a minor contribution to oxidized nitrogen input, while in central and southern Europe, the contributions are much larger and require measurement networks to define concentration fields.

Similar arguments may be advanced for HONO and PAN, both of which are present in much larger concentrations in central and southern Europe than in western and northern Europe and Scandinavia. The measurement base is weak and is inadequate to support estimates of fluxes. For these two gases the exchange with natural surfaces also remains uncertain. Recent measurements by Kitto and Harrison (1992) show the evidence of HONO production at the ground from NO_2. However too little of the underlying mechanisms and its dependence on NO_2 concentration temperatures and other variables is available to predict annual fluxes and the importance of this process on regional scales.

NH_3

The exchange of ammonia over natural surfaces has now been studied in detail over moorland forest and agricultural land by Duyzer *et al.* (1992), Sutton *et al.* (1994) among others. The main features of the process are understood. The exchange of NH_3 is bidirectional, with emission fluxes from vegetation when ambient concentrations lie below a canopy compensation point, and fluxes are directed towards the canopy when ambient NH_3 concentrations exceed the canopy compensation point. The leaf surface sink uptake is a major factor competing with stomatal exchange to determine the net flux (Figure 6). The net NH_3 flux over natural vegetation is dominated by uptake, which at times approaches the maximum values possible (i.e. canopy resistance $r_c = 0$). However, even over these surfaces a canopy resistance is usually present with values in the range 5 to 30 s m^{-1}. These limit the deposition rates of NH_3 to semi-natural vegetation to 0.5 to 3 cm s^{-1} under most conditions. In some conditions, such surfaces may be seen to emit NH_3 to the atmosphere (Sutton *et al.*, 1994).

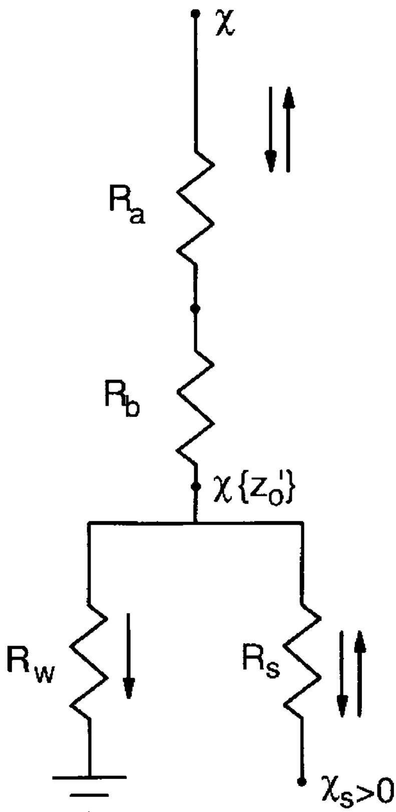

Figure 6 **Proposed resistance model of NH_3 exchange with plant canopies accounting for parallel deposition onto leaf surfaces (R_a) and exchange through stomata (R_s) with a stomatal compensation point (χ_s). The net canopy compensation point is given by $\chi\{z_o'\}$.**

For agricultural crops bidirectional exchange is a much more common feature of the data. As an example, Figure 7 shows data for measurements over a wheat crop which indicate a change in direction of the flux (from deposition to emission) when air concentration of NH_3 reaches about 1 µg NH_3 m^{-3}. A model simulating net canopy-atmosphere exchange using the canopy compensation point model χ_c

where $[\chi\{z\}/(R_a\{z\}+R_b)+\chi_S/R_S]\ \chi_c = [(R_a\{z\}+R_b)^{-1}+R_S^{-1}+R_W^{-1}]$

simulates the net exchange very well, assuming 100 µmolar NH_4^+ in intercellular fluids Figure 6 (Sutton & Fowler, 1993). These recent developments in simulating net exchange between the atmosphere and terrestrial ecosystem provide the basis for mapping annual reduced nitrogen inputs and regional scale. However, the spatial variability in several key parameters remains unknown; the temporal and spatial patterns in intercellular NH_4^+ concentration for agricultural crops, the temporal variability in leaf surface resistance and last and most important, the ambient ammonia concentrations are all required to complete this work. The approach of the Netherlands in providing a dedicated monitoring network for NH_3 concentrations provides the ideal model for other contries of Europe. Many European countries have no monitoring of NH_3 at all.

Within the UK, diffusion tube surveys by Atkins and Lee *et al.* (1992) in the UK for 1988 provide the current best national estimate of measured concentration field, but is subject to significant uncertainty, not least because there appears to be a significant over-estimate of actual NH_3 concentration using these methods. To provide the best estimate of reduced nitrogen deposition with current information, a correction factor for the diffusion tube data of 0.43 has been applied. The semi-natural vegetation has been assumed to be a consistent sink for NH_3 with a mean canopy resistance of 10 s m^{-1} and deposition velocities and fluxes have been calculated for measured wind and temperature fields and land class cover information to identify the roughness length (z_0) for each of the major land uses. The arable land is at present assumed to be ammonia neutral (that is that with reference to measured air concentration there is no net NH_3 exchange with the atmosphere over the year).

This is the area of nitrogen deposition that is most uncertain and in need of major improvement in mapping and synthesis of current data. The state of the measurements of NH_3 is similar in all other countries with the exception of the Netherlands where a monitoring network is in place. The modelling of net NH_3 exchange over regional scales is at a rudimentary stage everywhere.

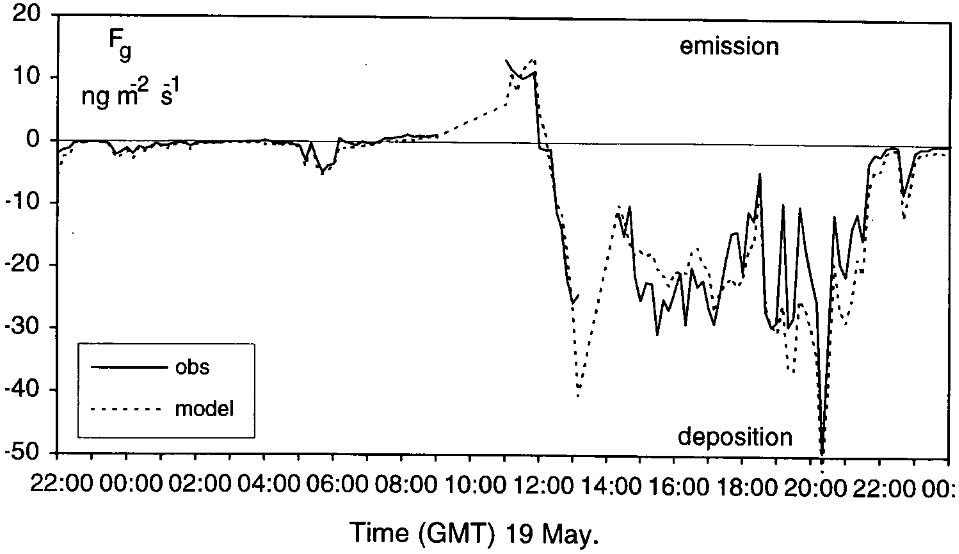

Figure 7 **Predicted flux of NH_3 compared with measurements. Model assumes 100 molar NH_4^+ in leaf tissues and R_w dependant on relative humidty. (Sutton & Fowler 1993)**

Particle Deposition

With the exception of cloud droplet deposition, the field of particle deposition has been largely ignored during the last 10 years. However, a number of issues and scientific developments have made it important to provide particle deposition inputs throughout Europe. First, base cation deposition is a very important contributor to the mass balance approach for critical loads estimates, and atmospheric base cations are deposited on forests at significant rates. Second, recent field data show that for aerodynamically rough vegetation, rates of (1 to 5 μm) particle deposition are in the range 7 to 15 mm s^{-1} (e.g. Wyers *et al.* 1995). Thus for forests and other aerodynamically rough surfaces the inputs of NH_4^+ and NO_3^- in particle form may be a significant contribution to the total input.

To provide the necessary parameters and fields for mapping, a low density network of monitoring stations for size resolved aerosol NO_3^- and NH_4^+ concentrations is necessary. The research required is essentially the size dependence of deposition velocities of aerosols over short ('grass'), medium height (e.g. cereals) and tall vegetation (forest) and urban areas. These data are also required to quantify the atmospheric life-times of aerosols by dry deposition, an issue of considerable importance in the current debate over aerosols and human health and aerosols and radiative effects.

The Effect of Deposition Grid Scale for Exceedance Estimates

The concentration fields, meteorological and land use input data used to obtain estimates of the annual input have an optimum grid scale, which varies with the different gaseous species and the mechanism of deposition. The choice of 20 km x 20 km for application in all maps is largely a consequence of the wet deposition data within which orographic enhancement has been calculated on rather simple assumptions to avoid the real complexity of orography which is currently beyond the modelling approaches that have been developed. Thus, for a 20 km x 20 km grid square in complex terrain, the actual deposition at a number of points at the same altitude may differ as a consequence of upwind topography.The average value mapped results from the enhancement of precipitation for the grid square over the interpolated coastal values. While these simplifications prevent the application of deposition data at a finer scale than 20 km x 20 km, it is possible for selected grid squares to estimate variability in actual 1 km x 1 km deposition that would be expected on the basis of current understanding of deposition processes. An exercise in quantifying these effects by Smith *et al.* (1995) showed that in the high rainfall areas of the UK, the 1 km x 1 km critical loads exceedance would be doubled by quantifying actual wet deposition variability with 20 km x 20 km grid square. Procedures to provide this detail for all grid squares have not been developed. The consequence of this finding is that current maps of critical load exceedance (for soils and acidification) underestimate the actual exceedance throughout the areas of complex terrain.

Conclusions

- Wet deposition. This is generally well estimated in simple topography with current networks, relative to dry deposition. For the UK, the network is currently at its minimum size for the mapping tasks and a few more monitoring stations are necessary.

- Uncertainty in complex terrain leads to under-estimation of deposition (and exceedance). There is a need to develop procedures to provide finer scale inputs and validate inputs and their spatial variability in complex terrain with measurements.

- Cloud deposition. Estimates appear satisfactory at a limited number of high altitude regions of northern Europe where monitoring underpins the modelling. The importance of radiation in fog to deposition is largely unknown.

- Dry deposition - NO_2. The theoretical basis for input estimates is good but there is very little validation and therefore large uncertainty and the network for NO_2 concentration measurement is not adequate. An NO_2 network and flux measurement facility is necessary.

- Dry deposition - NH_3. Most sensitive receptors show large rates of uptake and are therefore best estimated using small r_c (10-20 s m^{-1}). Uncertainty in input is very large. There is no current basis for net flux over agricultural areas. There is no network for NH_3 concentration measurement and the lifetime of NH_3 due to gas to particle conversion is also very uncertain. Much work is needed.

- Particle deposition. Particle deposition is important for forests, especially in areas remote from sources, and is poorly estimated. Rural concentration means of particle concentrations for NH_4^+, NO_3^- are not known. Process research (size dependence of aerosol Vg) and monitoring are both necessary.

- Land-use. Land-use specific inputs of nitrogen provide a more rigorous basis for Critical Loads exceedance mapping.

- Spatial scale of deposition estimates. The accumulated area of exceedance increases as the area used to estimate deposition decreases (for example, the change from 20 km x 20 km to 1 km x 1 km leads to a doubling of the area of exceedance).

- Complex terrain. The effects of complexity in the terrain due to hedgerows, small woodland patches and changes in roughness length on deposition rates remain unknown.

- Urban deposition. The deposition rates of NO_2, NH_3 and particle NO_3^- and NH_4^+ to urban surfaces are unknown and are needed to define inputs and the lifetime of pollutants in urban air.

References

Atkins, D.M.F. and Lee, D.S. (1992) The distribution of ammonia in the United Kingdom. AEA-EE-0330, AEA Technology, Harwell, Oxon.

Choularton, T.W., Gay, M.J., Jones, A., Fowler, D., Cape, J.N. and Leith, I.D. (1988) The influence of altitude on wet deposition.Comparisons between field measurements at Great Dun Fell and predictions of a seeder-feeder model. *Atmos. Environ.*, 22, 1363-1371.

Dollard, G.J., Unsworth, M.H. and Harvey, M.J. (1983) Pollutant transfer in upland regions by occult precipitation. *Nature*, 302, 241-243.

Duyzer, J.H., Verhagen, H.L.M., Westrate, J.H. and Bosveld, F.C. (1992) Measurement of the dry deposition flux of NH_3 on to coniferous forest. *Environ. Pollut.*, 75, 3-13.

Duyzer, J. H. and Fowler, D. (1994) Modelling land atmosphere exchange of gaseous oxides of nitrogen in Europe. *Tellus*, 46B, 353 - 372.

Duyzer, J.H., Deinum, G. and Baak, J. (1995) The interpretation of measurements of the surface-atmosphere exchange flux of nitrogen oxides; correction for chemical reactions. *Phil. Trans. Roy. Soc. Lond.* (In press).

Fowler, D., Baldocchi, D. and Duyzer, J.H. (1991) Inputs of trace gases, particles and cloud droplets to terrestrial surfaces. In: *Acidic deposition, its nature and impacts.* Proceedings of the Royal Society of Edinburgh Vol 97B, F.R. Last and R. Walling (Eds).

Fowler, D., Cape, J.N., Leith, I.D., Choularton, T.W., Gay, M.J. and Jones, A. (1988) The influence of altitude on rainfall composition at Great Dun Fell. *Atmos. Environ.*, 22, 1355-1362.

Fowler, D., Cape, J.N. and Unsworth, M.H. (1989) Deposition of atmospheric pollutants on forests. *Phil. Trans. R. Soc. Lond.*, B324, 247-265.

Fuzzi, S. (1988) Fog chemistry and deposition in the Po Valley, Italy. In: *Acid deposition at high elevation sites*, edited by M.H. Unsworth & D. Fowler, pp 443-452. Kluwer, Dordrecht.

Gallagher, M.W., Choularton, T.W., Morse, A.P. and Fowler, D. (1988) Measurements of the size dependence of cloud droplet deposition at a hill site. *Quart. J. Roy. Met. Soc.*, 114, 1291-1303.

Gallagher, M.W., Beswick, K., Choularton, T. W., Coe, H., Fowler, D. and Hargreaves, K.J. (1991) Measurements and modelling of cloud water deposition to moorland and forests. *Environ. Pollut.* 75, 97-107.

Gao, W., Wesely, M.L. and Lee, I.Y. (1991) A numerical study of the effects of air chemistry on fluxes of NO, NO_2 and O_3 near the surface *J. Geophys. Res.*, 96, D10, 18761-18769.

Hargreaves, K.J., Fowler, D., Storeton-West, R.L. and Duyzer, J.H. (1992) The exchange of nitric oxide, nitrogen dioxide and ozone between pasture and the atmosphere. *Environ. Pollut.* 75(1), 53-60.

Kramm, G., Muller, H., Fowler, D., Hofken, K.D., Meixner, F.X. and Schaller, E. (1991) A modified profile method for determining the vertical fluxes of NO, NO_2, ozone and HNO_3 in the atmospheric surface layer. *J Atmos. Chem.*, 13, 265-288.

Kitto, A.M.N. and Harrison, R.M. (1992) Nitrous and nitric acid measurements at sites in south-east England. *Atmos. Environ.*, 26A (2), 235-241.

Muller, H., Kramm, G., Meixner, F., Dollard, G.J., Fowler, D., and Possanzini, M. (1992) Determinations of HNO_3 dry deposition by modified Bowen ratio and aerodynamic profile techniques. *Tellus*, 45B, 346-367.

Skiba, U., Hargreaves, K.J., Fowler, D. and Smith, K.A. (1992) Fluxes of nitric and nitrous oxides from agricultural soils in a cool temperate climate. *Atmos. Environ.*, 26A(14), 2477-2488

Smith, R.I., Fowler, D. and Bull, K.R. (1995) Quantifying the scale dependence in estimates of wet and dry deposition and the implications for critical load exceedances. In: *Acid Rain Research - Do we have enough answers?* Proceedings of the s-Hertogenbosch conference, The Netherlands, October 1994, Elsevier Science, (In press).

Sutton, M.A., Asman, W.A.H. and Schjorring, J.K. (1994) Dry deposition of reduced nitrogen. *Tellus*, 46B, 225-273.

Sutton, M.A. and Fowler, D. (1993) A model for inferring bi-directional fluxes of ammonia over plant canopies. In: *Proceedings of the WMO conference on the measurement and modelling of atmospheric composition changes including pollutant transport.* WMO/GAW - 91. (Sofia 4-8 October 1993). World Meteorological Organization, Geneva. pp179-182.

Weston, K.J. (1991) Objectivity analysed cloud immersion frequencies for the United Kingdom. *Meteorl. Mag.*, 121, 108-111.

Williams, E.J., Guenther, A. and Fehsenfeld, F.C. (1992) An inventory of nitric oxide emissions from soils in the United States. *J. Geophys. Res.*, 97, 7511-7520.

Wyers G.P., Veltkamp A.C., Geusebroek M, Wayers A. and Moels J.J. (1995) Deposition of aerosol to coniferous forest. In: *Acid Rain Research: Do we have enough answers?* Proceedings of the s-Hertogensbosch Conference (9-11 October 1994). Elsevier Scientific, (in press).