PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY B BIOLOGICAL SCIENCES

Concluding Remarks

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Concluding remarks

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Introduction

The topic of this Discussion stands at the meeting point of several strands of inquiry and concern. Recent years have seen deepening interest in biogeochemical cycles, especially of carbon, nitrogen, oxygen, phosphorus and sulphur, and their modification by man, and this has been the subject of several reports by SCOPE. The effects on physical and biological targets of the perturbations we call pollution have received increasing attention. In our present theme these two areas of research come together and influence social responses to phenomena that have aroused deep public concern.

Our aim has been to evaluate scientific understanding of the ecological effects of deposited sulphur and nitrogen compounds, not to propose Governmental or industrial policies. But the continuing evolution of such policies needs to be based on the most critical possible evaluation of the scientific facts. Without this, false conclusions could all too easily lead to actions wasteful of resources and unhelpful to the environment.

In preparation for this meeting, nine issues – each prompting many questions – were brought to the attention of speakers. Here I will condense them to five specific and one general theme.

(1) How far do we understand the spatial pattern, trends and causes of wet and dry deposition of sulphur and nitrogen compounds?

(2) How far can we state dose–response relations for the effects of these depositions on plants, including figures for the costs of impaired yields?

(3) What is the nature of the interaction between depositions and soils?

(4) How far do these interactions in turn affect plant growth and yield?

(5) How far are the depositions affecting freshwater composition and ecology (including fisheries), and how does one explain the fact that nearby catchments on similar rocks, with similar acidity of deposition, differ in pH and biota?

Running through our meeting has been the sixth theme, which is one of scientific integrity. Which hypotheses have been subjected to critical test? How far have adequate models been developed? How far have rates of change really been established? How far can we distinguish local from general phenomena? How far can we predict likely futures including responses to management? And, if I may add a question of my own, how resilient are ecological systems likely to be in the face of the stresses we are discussing, and how far are changes likely to be reversible?

One crucial issue of this kind arose late in the meeting. We questioned the adequacy of our measurements of pH and the validity of the comparisons between different data sets. We heard that the problems were worst in low-ionic, poorly buffered water, and that the reference electrode was the problem, but I felt that this issue was left uneasily up in the air.

Atmospheric processes and depositions

The first session reviewed physical processes of dispersion and deposition, providing a link with the symposium held a week earlier in Oxford. We were reminded that gases and particles can be deposited directly by dry deposition or by wet deposition in rain, or through cloud and mist droplets as so-called 'occult precipitation'. Later in the meeting Dr Jacobson provided further analysis of these pathways, pointing out that gases were deposited by diffusion and adsorption at low velocities, while aerosols and fog and mist droplets ranging from 0.01 to 30 μm diameter deposited by impaction, and mist and rain in the 30–300 and 300–3000 μm diameter range by impaction and sedimentation, with high velocities. These analyses led me to ponder again the unwisdom of using a popular term like 'acid rain' to describe much broader processes of deposition. I suggest we either banish this term from the scientific literature or redefine it. And, incidentally, we were reminded that natural rain was variably acid, perhaps with a mean of pH 5 and a minimum of 4.5, not the 5.6 commonly quoted.

Four points in Mr Goldsmith's analysis of atmospheric pathways and transformations struck me particularly. First, he showed that predicted total depositions from both local and distant sources agreed broadly with measurements. Second, he stressed the difficulty of precise measurement, showing the variable quality of European records and that only a small fraction of sites recorded increases in acidity that correlated with increases in sulphur emissions between 1965 and 1975.

Mr Goldsmith's third point was that modelling of daily deposition was much more difficult and laborious than handling averages, but likely to be crucial since the phenomena were highly episodic. In Cornwall, 30% of rain falls on 8.2% of wet days: 30% of sulphur is deposited on 4.2% of deposition occasions. Nitrogen deposition also peaks on a few days. Forecasting days of potential high deposition might conceivably allow control measures, like substitution of low-sulphur fuel, to be geared to risk. But Professor Rosenqvist asserted that there was no correlation between acid rainfall episodes and acid episodes in rivers: the latter correlated instead with high rainfall regardless of its pH.

Finally, Mr Goldsmith examined chemical transformations in the atmosphere. He showed field data demonstrating how the SO_2:SO_4 ratios changed with time as a plume dispersed. A linear curve of direct oxidation from SO_2 to SO_4 and NO_x to NO_3 is commonly assumed, but field measurements indicate nonlinearity and the importance of ozone and other oxidants in the oxidation process. Professor Thrush in discussion mentioned reactions involving hydrocarbons as additional ozone sources and the importance of OH and peroxide as catalysts. The point is that emission chemistry alone is not enough, and interactions between atmospheric constituents – including a range of pollutants – are crucial. The complexities of the processes preceding deposition became evident, and it cannot be assumed that sulphur or nitrogen depositions are directly proportional to emissions, although Dr Jacobson said in discussion that proportionality did seem to emerge when the data were averaged for a broad region or periods.

Dr Fowler analysed the mechanisms of deposition on surfaces. He pointed out that, inadequate or not, we did have some direct monitoring of wet deposition whereas dry deposition could only be calculated. In the U.K. he estimated that dry deposition of sulphur would be dominant in southern England, with totals of up to 5 g m^{-2} a^{-1}, but that wet deposition at over 3 g m^{-2} a^{-1} would dominate in the Scottish Highlands, the Southern Uplands and Cumbria. He also demonstrated how deposition phenomena vary with vegetation height and texture. The

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enormous importance of stomata as sites of SO_2 and NO_x uptake in both short and tall vegetation emerged, with the corollary that when stomata are shut, or in deciduous species in winter, quite different deposition rates and patterns ensue. Very little dry deposition appeared to go directly to the soil. Taken together, this analysis implied that we should be able to indicate peak deposition points within a vegetation type, and relate this to the meteorological factors reviewed by Mr Goldsmith, so as to evaluate relative vulnerabilities within the system. I deduced from the discussion that we were not at this point yet, but that this was certainly a conceivable development.

Direct effect on plants

We heard three evaluations of the direct effects of these substances: especially SO_2 on crops and trees. At this point, as not uncommonly when biological diversity intrudes, variability and uncertainty became perplexing. In fumigation experiments reviewed by Dr Roberts this appears to stem from diverse experimental conditions and interactions between pollutants and other variables like age, growth rate, the spacing of plants, low temperature and other stresses. Another factor may be the tendency of experimenters to select genotypes they think will be sensitive. SO_2 alone certainly impairs growth of sensitive plants at concentrations above 150 μg m^{-3}: SO_2 and NO_2 in combination somewhat below this, while if ozone is also present the threshold appears lower still. In view of what came later, I was interested to see that the best model of the process of damage included effects on root growth as well as on foliage. Data for ambient SO_2 concentrations in Britain implied that effects were most likely on urban fringes and in areas like the South Pennines, known to have suffered marked ecological change following the industrial revolution.

Dr Keller stressed that in trees lack of visible symptoms is not adequate proof that the plant is healthy. Photosynthesis and growth are liable to impairment well below the threshold of visible damage: at 130 μg m^{-3} in one experiment cited. Again he stressed variability – between genotypes (such as different clones of *Picea abies*) and in relation to periodicity of pollution (where long, occasional peaks did most damage to *Pinus sylvestris*). Fumigation in a dormant period was shown to depress growth in the following growing season. Like Dr Roberts, Dr Keller indicated that a multifactorial model must be applied to trees, but his data brought out most clearly our crucial need for reliable physiological indicators of performance.

Dr Jacobson's paper provocatively questioned some of the figures and conclusions of his predecessors. He stated that acid aerosol concentrations in polluted atmospheres were several orders of magnitude too low to cause foliar injury, although the gap was narrower in mists and rain. He argued that the acidity required to cause visible phytotoxic effects was present only in the worst environmental circumstances. However, he agreed that meteorological and other factors acting at the atmosphere:leaf interface, and the interplay of pollution and plant nutrition could increase liability to damage.

It is not surprising that in discussion we spent a lot of time debating this issue of visible injury. Various sensitive indicators of impaired performance – notably tree growth rings and CO_2 uptake – were suggested. In Germany tree ring increments were said by Professor Hüttermann to indicate declining performance since the 1950s: well before the recent appearance of acute damage. Professor Rutter said that he had found impairment of growth in coniferous trees at SO_2 concentrations of 50–100 μg m^{-3} and that such effects could be cumulative. Sensitivity to acute damage causing visible injury, and liability to chronic damage and long-term performance impairment might not be closely related. Indirect effects can occur through pathogens (and pollutants can beneficially suppress these as well as let them in through stress). All in all, I concluded that we were still well short of a proper *materia medica* in this area, but that it has been a feature of recent research that as techniques have improved more subtle effects, at lower thresholds, with more evidence of complex interactions, became evident.

Soil–plant interactions

The four papers on soil–plant interactions re-emphasized the multifactorial nature of the situation. Dr Abrahamsen, with others, pointed out that deposited S and N compounds could have a fertilization as well as acidification effect. Dr Miller showed how percolation of wet deposition through tree canopies was accompanied by changes in ionic composition that vary with species and age, acidification being most pronounced in older coniferous trees. Potassium, calcium and magnesium all increase as throughput acidity rises, and Dr Miller presented the Rehfuess model relating damage to leaf cell membranes by ozone and SO_2 to accelerated calcium and magnesium leaching in the leaf which root uptake could not match, leading in turn to foliar Ca and Mg deficiency, and with feedback via reduced root growth and increased frost sensitivity aggravating the situation. He also pointed out that this abstraction of cations led to H^+ accumulation in soil, which could in time aggravate the situation through root damage.

Professor Hüttermann, presenting his and Ulrich's work, emphasized that acid deposition commonly outstripped the release of neutralizing capacity by weathering, leading to soil acidification. He agreed with Dr Miller that the acidity reached the soil through withdrawal of neutralizing ions, and Dr Abrahamsen later also stressed that the main impacts on forests were indirect, via the soil. If this acidity at the roots is not redistributed by the soil fauna an 'acidification push' especially of HNO_3 can result. In discussion, and in the poster session, we learned that lumbricid and enchytraeid worms appeared to be inactivated in acid soils, making such accumulations plausible. Root injury, diminished nitrate uptake and decomposition can intensify the situation. It seemed possible, although it was never expressly mentioned in the meeting, that there might also be effects on mycorrhiza: one of the topics we neglected. Experiments suggested that the poor survival of *Fagus sylvatica* seedlings in Germany in 1977 – the last good seed year – was related to root necrosis, with breakdown at the inner cortical surface in acid soils. Aluminium ion toxicity was proposed as an agent, acting through the replacement of calcium in cell walls especially in meristem and inhibiting Ca and Mg transport to needles in spruce, thus causing the foliar deficiency. In discussion, however, we were told of problems in the wider application of this model and of the critical importance of the species of aluminium involved, and it is clear that further research is required.

Dr Abrahamsen supported the general model that hydrogen, sulphur and nitrogen deposition lead to leaching of Ca, Mg and K, and to Al mobilization, nutrient deficiency and toxicity. His figures indicated that soil sulphur inputs and outputs probably balanced or inclined towards retention, but that only 30% of wet-deposited nitrogen left the system. The question was how far the rest is denitrified and how far it could accumulate and saturate soils. Nitrogen deposition seemed to have a real fertilization effect, but Ågren's data, which implied that a medium-type soil could be saturated in 40 years and then show higher leaching, left many questions for future research, and the discussion revealed a number of doubts about this proposition. Experiments with application of sulphuric acid indicated a fall in calcium (although not to deficiency point) and in magnesium in pine and in upper soil horizons: an observation that fitted evidence of magnesium deficiency in German forests. Data on aluminium seemed to accord with the Hüttermann–Ulrich presentation, although in discussion it became clear that actual soil concentrations of aluminium in Norway were below German levels and below the likely threshold of damage.

Dr Johnson went into detail about some of these processes. He pointed out that biological immobilization dominated the nitrate flow in soil and that whereas sulphate inputs and uptakes balanced and a fertilization effect existed in certain crop situations, in forests sulphur inputs exceeded biological demand and sulphate was retained, especially by iron and aluminium oxides. He re-emphasized that increasing acidification led to more aluminium in the leachate and more sulphate retention, up to a steady state. The question of reversibility arose, with the hypothesis of hysteresis and only slow removal of adsorbed sulphur even if inputs reduced, but the mechanisms and rates to be expected were not clear. In discussion the possible importance of reduction to H_2S emerged as yet another issue to be looked into.

Three other points struck me: that if nitrate is retained in soil (maybe because of high input and high biological fixation) it could be highly acidifying; that carbonic acid could be a powerful leaching agent in some tropical soils and that organic, especially folic, acids could also be important. The final comparisons of the Solling and North American soils rubbed in Dr Johnson's general point that these systems are so diverse that only site-specific studies examining all major ions are likely to allow the prediction of response to acid inputs. The discussion provided one further important point. Dr Abrahamsen said that Arctic and more southerly forests may differ, with nitrogen the only nutrient in short supply in the former but phosphate and potassium limiting further south.

Effects in the terrestrial environment

Looking back on these 'terrestrial' papers and discussions I was left with the view that only now were we beginning to see the ecological complexities. The diversity of situations scooped up under the broad and misleading title 'acid rain effects' is now obvious. A wide range of atmospheric components and pollutants interact to determine acidifying inputs, with some pointers to differing significances of, for example, photochemical oxidants in different areas. While prediction of effects is increasingly possible in laboratory studies designed to simplify the system, in the real environment the diversity of soils, meteorological factors, genotypes, physiologies and histories forces us back to a site-by-site approach, and general models of ecosystem response to these inputs remain far away. And we are still grappling with partial and imprecise data in many areas.

Effects of freshwater composition

I cherished a naïve hope that all would be easier when we got into the more familiar area of soils, streams and lakes, and especially fish. Instead, the parallels with the terrestrial systems were what impressed me. The global diversity in the situation was stressed by Dr Howells' demonstration of the extreme natural acidity of parts of the Amazon system and certain volcanic lakes. Dr Bache demonstrated how a 'model' rainfall might be altered in interaction with various soil waters and horizons, and stressed that prediction of pH and ionic composition of the input to a water body demanded knowledge of hydrological pathway, the pH, composition and stratification of the soil, and whether the deposition was even or peaked. Continuing the earlier story, he showed how bicarbonate removed acidity, how fulvic and humic acids could become important, how aluminium tended to be high in leachates from a podsol B horizon and how calcium, magnesium and sodium would come from a C horizon. The variations expected in drainage from fluvioglacial soils, moraine or bog with the same sulphate deposition provided a cautionary tale that re-emphasized Dr Johnson's demand for site-specific studies. And Dr Bache brought us back to the issue of expected consequences of other changes, like drainage of waterlogged soil releasing adsorbed sulphate as H_2SO_4 in place of the bacterially produced H_2S found in the reducing environment.

Dr Goldstein gave us the first lake system study. While many variables were discussed – topography, climate, snow depth and sub-ice stratification among them – I was impressed by the dominant role of soil type and especially soil depth in determining percolate acidity. From his presentation it became obvious that the reaction of catchments to changes in sulphate and nitrate deposition could only, as with soils, be judged on a site-specific basis. In discussion Dr Goldstein was pressed about how his model predicted the consequences of changes in rainfall acidity, and declined to make a general reply on the grounds that each catchment needed individual analysis and that the broad regional statements could only be statistical, and founded on a sufficient sample size. There is, I suggest, a lesson here.

Two other 'modelling' papers by Dr Christophersen and Professor Kramer demonstrated how much further in this respect limnologists have gone than their terrestrial colleagues. Dr Christophersen, like Professor Rosenqvist in discussion, had a basic model of input to freshwater in which deposition, the upper organic soil reservoir and the lower inorganic one were separated. I noted some debate over how far sulphate was the main vehicle of cation transfer, over the significance of sodium and chloride and of upward capillary flow and other details, but was struck by the emphatic statement that rainwater never reaches a brook unmodified by ionic interchange. Obvious? Of course, but it is one of those obvious facts that has been stated insufficiently in the wider literature about acidification. I was also interested in the continuing debate over the significance of snow-melt peaks, Dr Christophersen stressing their importance in removing accumulated pollutants in Spring and Dr Sorensen in discussion arguing that brief high rainfall peaks at other seasons could be equally important.

Professor Kramer's model again incorporated hydrology, soil–water interactions and hydrometeorological variables. He was able to show that field capacity on the outflow, hydraulic conductivity in the saturated zone and soil depth and acid-neutralizing capacity – especially in the soil upper layers – were the sensitive elements: the last of them clearly tying in with Dr Goldstein's paper. Unlike the latter, Professor Kramer did speculate for example that halving the hydrogen ion input might raise lake pH by only some 0.4 unit with no effect on soil pH. Under current conditions the lake he studied was changing only very slowly, with a century or so before significant alteration would be expected. In discussion, Dr Christophersen – who had already stressed that in Southern Norway sulphate and nitrate of anthropogenic origin had to be assumed in order to explain fish kills – was led to suggest further that to get fish back there had to be changes increasing the lime potential of the catchments: a significant, indeed crucial point for those of us whose responsibilities overlap from science into policy, but not one we appear able to pursue in depth at the moment.

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I was generally encouraged by the way that these three freshwater studies, with a considerable modelling component, told a comparable tale even though in many places it emphasized the need for more work and stressed the problems of generalization from a catchment to a region. Dr Clymo's presentation fitted well with this point, illustrating the natural acidity of the *Sphagnum* mosses and other acidophilous ecosystems so common on the boreal uplands, and showing that these systems can bring the pH of their waters down to 4.0 without the need to invoke acid rain (which, in discussion, was said to be likely to account only for a depression of 0.2–0.3 of a pH unit). He also demonstrated that cation exchange during plant growth and evaporation in Summer could concentrate the acidity and lower pH by 0.3–0.4 unit. Vegetational changes such as we know to have occurred around the Boreal–Atlantic transition and more recently, partly through human land use, could therefore readily alter the characteristics of upland drainage.

Dr Battarbee's presentation linked with this point in another way. It does appear that the sub-fossil diatom record in sediments can be used to indicate past fluctuations in acidity even though there is no final consensus on the best index and the record cannot discriminate between atmospheric inputs and land use changes. It was interesting that in three out of four Scottish lakes acidification, which showed no long historic trend, set in markedly in the mid- or late nineteenth century, and in the exception the 1925 change preceded afforestation of the catchment. In other parts of Britain, notably the South Pennines, we know from the peat record that there were ecological changes indicative of acidification in the mid-nineteenth century. I was struck by the evidence in discussion that while in the U.K. acidification thus appears to have begun over a century ago, its onset is more recent in Scandinavia and the Adirondacks. The diatom records are clearly valuable evidence, but the implication is that acidification has different causes in different places and that lumping all the instances as a single general phenomenon may be positively misleading when it comes to deciding on remedial action.

This point was reinforced in the subsequent papers dealing with effects on freshwater biota. Dr Kinsman showed that while substantial variations occur within and between freshwater bodies, spatially and temporally (and Sir James Beament commented on the meaninglessness of 'the pH of Windermere') a general pattern of correlation between acidity and biotic impoverishment does occur. Marginal macrophytes give way to *Sphagna*, blue green benthic algae commonly dominate below pH 5, and the phytoplankton drop out at pH 5.5 although primary productivity is said to be sustained in acid lakes down to a pH of 4.5 (so that these are not 'dead' even in a popular sense). Zooplankton and benthic invertebrate changes are broadly documented, but we are ignorant of effects on the decomposer cycle which could be vitally important and surely must be a subject for future research.

Professor Magnuson and Dr Muniz established clearly that there has been a decline in fish populations in areas of North America and Scandinavia. The first paper helpfully pointed out the three kinds of data source – spatial and temporal observation, laboratory experiments and field experiments – and confirmed that all three indicated the progressive, sequential dropping out of species with increasing acidity, with reproductive failure some 0.5 pH unit above population disappearance. Professor Magnuson also predicted that continuing acidification would move lakes now at over pH 5 to pH 4.5 while those with strong episodic acid peaks could come to this range even from a present pH around 7.0. Dr Muniz documented Scandinavian fish declines, some beginning at the start of this century, some in the 1930s and more – especially of brown trout – since the 1940s. He raised, and Dr Howells elegantly pursued, the question of cause. Her experiments and field analyses demonstrated our need to consider pH, calcium and aluminium together with altitudinal, climatic and hydrological factors and taking episodes into account as well as means. Episodes could remove a recruitment class, but we are still unclear how it is that reproductive success fails before gross physiological changes appear. Perhaps breeding areas are particularly vulnerable. Perhaps there is an analogy with the botanical situation, and fish physiologists need to look for more subtle indicators of performance.

Finally, we came to treatments. Dr Howells suggested that we could consider better management of water flow and use, evaluate land use controls, and go on applying the remedial treatment of liming as clearly the only thing likely to have an effect in the short term. Dr Nyberg showed, however, that liming has limited application especially in rivers and other toxic waters of high episodic character, and will not prevent the leaching of toxic ions. It can help protect sensitive systems while longer-term remedial measures are applied, but it is not a full cure. Dr Muniz implied that more subtle measures might become available when we understood the systems better, and were more able to correct acid inflows, while Dr Christophersen argued that reducing anthropogenic sulphur and nitrogen inputs would be essential in any event.

Conclusions

What general conclusions should we draw from this Meeting? Looking back to the original questions put to speakers I would summarize the position as follows.

1. We do have a reasonable picture of the spatial pattern, trends and causes of wet deposition of sulphur compounds, and adequate models especially of the meteorological processes involved, but the models run ahead of the measured data and we still lack good records of dry deposition, of short-term periodicity and of small-scale local variability relatable to the receiving surface. Our knowledge of the nitrogen system is substantially inferior to that of sulphur.

2. It is now obvious that we must stop talking about 'acid rain' and certainly abandon the notion that acidification is simply a matter of SO_2 or NO_x emissions. We have seen that the atmospheric reactions involve hydrocarbons, OH and peroxides, and that deposition dry and in mist can be as important as in rain.

3. We have some quantitative data on dose–response relations for direct phytotoxic effects of SO_2 and rather fewer for combinations of pollutant gases on selected plants, largely from fumigation experiments. However, we are far short of a basis for estimating subtle damage below the threshold of visible injury in the field, and need to establish more sensitive indicators of plant performance.

4. Many of the effects with which we are concerned are clearly mediated via the soil. There is an emerging consensus on a basic model involving cation – especially calcium and magnesium – withdrawal from the soil and aluminium accumulation, but the details, the role of other ions, and the scope of application of the model remain to be worked out. We need to know a great deal more about processes in the rhizosphere, and especially about the interaction between plant roots, mycorrhiza, soil faunas and soil chemistry. And we are far short of a model we can apply predictively to relate depositions, via soil changes, to plant growth and yield.

5. It is clear that the composition and structure of the receiving soil are crucial both in determining effects on plants and for acidification of drainage waters. Some sites, like

Sphagnum bogs, and some lakes are clearly naturally acid and are becoming more so as a result of natural processes, and in these situations atmospheric inputs can be a small proportion of the total acidity in the system. We have to face the fact that soils and catchments have to be characterized site by site and that regional generalizations will emerge only as statistical statements when samples are adequate. There may be the basis of a general model emerging, in which upper soil horizons in leached podsols are especially important as a source of toxic aluminium (an ion whose importance at many stages in the total process has become evident as this Meeting has progressed), but many components of this model still require to be worked out.

6. Similarly, lake systems have to be examined individually. There are indications of biotic impoverishment that correlate with high acidity, low calcium and high aluminium, and perhaps also with altitude, but we are still uncertain of causal mechanisms and potential remedies.

7. Finally, we have only begun to glimpse the timescales involved in these phenomena. It is clear that some changes, for example in freshwater catchments and forest soils, are slow and have been operating over decades if not centuries. It is clear that many are unlikely to be rapidly reversible. This is perhaps why we see no effect yet of the considerable reductions in sulphur emissions that have occurred in parts of Europe in recent years. If we are to reverse these trends we shall need better models, must accept that different problems of acidification may require different solutions, and be prepared to be patient if the responses to changed inputs of acidifying substances also take decades to manifest themselves.