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Introduction

The UK research community involved in effects of air pollutants on ecosystems was brought together in 1974 by the Natural Environment Research Council to promote liaison and co-ordination of research in a nationally-important field of science. This coincided with global interest in the issue of acid rain in Europe following the 1972 UN Stockholm conference on the Human Environment, at which Sweden presented a case study on the impact of sulphur in air and precipitation. Specifically the issue raised was that of air pollutants crossing National boundaries and the widespread damage in Scandinavia from acidic pollutants emitted by the major industrial countries of Europe, notably the UK, Germany and France. This introduction provides a brief history of major developments in Europe since 1974 in the science of air pollution effects on ecosystems, and the interactions between scientific understanding and environmental policy at the international scale.

The approach taken is chronological and represents a relatively short period, just 41 years, yet the changes in the composition of the air over Europe and over the UK in particular, have been dramatic. In the 1970s the air over the UK received 6 million tonnes of SO_2, mainly from burning coal. Annual mean concentrations of SO_2 were in the range of 10-50 $\mu g\ m^{-3}$ with surface concentrations regularly exceeding 100 μgm^{-3} in large cities, and large parts of the country were a lichen desert. Today, annual emissions of SO_2 are 250 kilotons, with concentrations in cities generally lower than 10 $\mu g\ m^{-3}$, and barely detectable in rural areas.

During the early years of CAPER, research on direct effects of SO_2 on crops and semi-natural plant communities was extensive, along with studies to quantify the deposition processes and effects of acid deposition in the UK. The range of pollutants studied was broadened to nitrogen compounds, ozone, and metals to characterise the full air pollution climate of the country, which lagged some years behind Scandinavian work in this field. Motivated primarily by observed effects, the policy responses to air pollution issues have driven large improvements in air quality and have eliminated the cause of widespread damage by sulphur compounds in the middle years of the last century.

However, there remain important air pollution issues for most developed and, especially, developing countries, where air pollution is a major cause of premature human mortality and represents a threat to food security and ecosystem resilience. Among the widespread ecological effects of transboundary air pollution are eutrophication, acidification, and biodiversity loss due to nitrogen deposition (Bobbink et al. 2010) and damage to the structure and metabolism of crops and semi-natural plant communities due to ground-level ozone(Mills et al. 2011). Atmospheric nitrogen and ozone pollution, which are both at least in part due to human perturbation of the global nitrogen cycle(Fowler et al. 2013), are proving from a policy perspective to be quite intractable. These pollutants and their impacts are the subject of the four papers in this special section.

The scientific community was well aware of the potential for air pollutants to damage plants and animals in the early 20th century. Many of the industrial cities in Europe and North America already had substantial surface concentrations of SO_2, NO_2, and particulate matter(Brimblecombe 1987). However, until the second half of the 20th century, air pollution impacts were regarded as local or national issues. What changed in the 1970s was the recognition of the scale of transboundary air pollution transport and deposition. For Sweden and Norway in particular, the amounts of sulphur deposited within their countries greatly exceeded their national emissions, and this deposited sulphur was rapidly acidifying freshwater ecosystems and acid-sensitive soils. Sweden presented a case to a United Nations

Conference on the Human Environment in 1971 arguing for a mechanism to regulate the cross-border transport and deposition of pollutants(Sweden 1972).

A development of monitoring networks, process studies, experiments, and modelling rapidly followed, which conclusively demonstrated the scale of inter-country exchange of pollutants within Europe. This international effort was co-ordinated by the European Monitoring and Assessment Programme (EMEP) which was established under the Convention for Long Range Transport of Air Pollution (CLRTAP) by the United Nations Economic Commission for Europe (UNECE) in 1979(Bull et al. 2001). The UNECE CLRTAP convention provided a framework within which emission controls were developed to reduce emissions of the major air pollutants in Europe, beginning with sulphur and extending to oxides of nitrogen, volatile organic compounds, and ammonia. Successive protocols defined emission targets for individual countries and extended the range of pollutant issues to include acidification, eutrophication, and ground- level ozone in the Gothenburg protocol of 1999.

The CAPER research community focussed on effects of acidic pollutants and ozone on agricultural crops and natural plant communities throughout the 41 years. Along with Dutch ecologists, this community has provided global leadership in the effects of atmospheric nitrogen deposition on semi-natural plant communities, with field surveys(Pitcairn et al. 2001), surface-atmosphere exchange studies (Sutton et al. 1993) and long term experiments (Phoenix et al. 2012) demonstrating the role of atmospheric nitrogen deposition on plant communities.

By 2014, these control measures have reduced emissions of sulphur in Europe by 80% from their peak values in the 1970s. Acid deposition has greatly decreased, and freshwater ecosystems throughout Europe are slowly recovering. Furthermore, the phytotoxic ambient concentrations of SO_2 in the most polluted regions of the UK, Poland, and the Czech

Republic have declined to very small values which no longer present a threat. Similarly, legislation to reduce emissions of the precursor gases for eutrophication (NOx and NH_3), and for tropospheric ozone (VOCs and NOx) were designed to address the damage by these pollutants in Europe. As a result, emissions of oxidised nitrogen and VOCs in Europe declined by approximately 50% between 1980 and 2014.

However, the scale of emission reductions has not been sufficient to prevent the widespread continuing impacts of eutrophication on ecosystems(Duprè et al. 2010). Furthermore, the emissions of NH_3 have declined by only about 20% from their peak value, and there is clear evidence from at least some plant communities that the direct vegetation effects of dry-deposited NH_3 are greater than those of wet oxidized or reduced nitrogen(Sheppard et al. 2011). Thus, the deposition of oxidised and reduced nitrogen throughout Europe remains substantially larger than the level needed to protect ecosystems from further decline, and to promote recovery.

In the case of ground-level ozone, although peak concentrations have declined appreciably following the reductions in VOC and NOx emissions, mean O_3 concentrations have increased by 20-30% since widespread monitoring began in the 1970s (Jenkin 2008). The effects of ozone are primarily driven by the absorbed flux through stomata (Mills et al. 2011) and there is little evidence that the overall leaf-surface O_3 flux has declined in Europe, with increases in mean concentrations compensating for the declines in peak values. The problem of ground level ozone is not restricted to Europe: it was first identified in North America and is now recognised as a global issue(Shindell et al. 2012).

The process for policy development in Europe which delivered very effective reductions in sulphur and acid deposition was strongly supported by science, from monitoring and assessment through to experimentation and modelling. In principle, the same mechanisms are capable of delivering continued improvement in the chemical climate, especially in the case of eutrophication, for which the European pollutants are mainly of European origin. There are complicating factors. In the case of eutrophication, there is no doubt about the primary cause, oxidized and reduced nitrogen emissions. However, the recognition that air pollutants, especially particulate matter is a major cause of premature human mortality (Dockery et al. 1993) has led to eutrophication effects on semi-natural plant communities receiving a much reduced priority in the policy agenda. Secondly, the widely recognised effects of ozone on crop and natural plant communities is a global scale issue, requiring, at least hemispheric scale reductions in VOC and NOx emissions to reduce mean concentration in the mid Northern latitudes, for which there is no international policy instrument.

The four papers in this special section of Environmental Pollution represent the current air pollution effects research focus on ozone and nitrogen deposition, two related issues and are proving from a policy perspective to be quite intractable issues. The UK CAPER research community continues to advance the underpinning science and engages closely with the user community in government departments and more widely with parallel research communities in North America and continental Europe. Increasingly these research groups will need to work closely with their equivalents in East and South Asia, where the greatest exposures to pollutants occur, and where the most promising research opportunities are to be found.

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