Deposition of Atmospheric Pollutants

Edited by
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PREFACE

The problem of "acid precipitation" has been recognized with growing concern in many industrialized countries. The incorporation of pollutants into cloud and rain elements and their transfer to the ground by "wet deposition" are dominant mechanisms leading to a self-cleansing of the troposphere but, on the other hand, to hazards to the soil, vegetation and forests. The influence of orographic and meteorological parameters and of the regional distribution of precipitation on the deposition of pollutants are insufficiently known factors.

During previous years, several projects and analyses have been initiated to improve our knowledge on the dry and wet deposition of pollutants and on the mechanisms of transport of gaseous and particulate components from the atmosphere to the ground. Research activities have been supported in different fields and it appeared not only useful but necessary to bring the different research-groups together to endorse the communication and cooperation between scientists in the related fields. A symposium was arranged in Oberursel/Taunus in November 1981 to discuss the results of experimental and theoretical work in the field of deposition and to gain a better understanding of each other's methods, experience and observations.

The proceedings presented in this volume permit a fair survey of the present-day knowledge and will be a useful tool for all working in this field.

The meeting would not have been possible without the financial support of the German Environmental Agency (Umweltbundesamt).

The preparation of the symposium and of the publication of the contributions caused a considerable amount of work. I would like to express my gratitude to Miss C. Perseke, Mr. E. Rohbock and Mrs. H. Wallenwein for their continuous assistance.

Hans-Walter Georgii

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Introduction

COMMENTS ON THE INVESTIGATION OF THE DEPOSITION OF ATMOSPHERIC POLLUTANTS

Dr. J. Pankrath

This colloquium is part of a R&D-programme which is funded by the Federal Environmental Agency in the framework of the Environmental Research Programme of the Ministry of the Interior. The research is being carried out under the leadership of Professor H.-W. Georgii of the Institute of Meteorology and Geophysics of the University of Frankfurt and is concerned with the deposition of atmospheric pollutants with special emphasis on wet deposition. From an environmental point of view the measurements of sulphates, nitrates and heavy metals in a precipitation chemistry network is of great importance, because these components pose a threat to ecology in regions well removed from industrialized areas.

We can generally speak of acid rain in this context being aware that there are other components such as heavy metals and organic compounds which are distributed over longer distances as well. Compared with other types of air pollution, acid rain is less well understood and of less immediate concern in the minds of the respective decision makers. Public awareness is now growing due to information about the menace of acid rain which, for instance, is said to be partly responsible for the damage of spruce forests also in Germany. The special problem of acid rain is its slow and insidious effect on the environment: changes are noticed only when they are irreversible.

In fact, it is misleading to believe that the use of high chimneys leads to air pollution being well mixed in the atmosphere, thus resulting in insignificant concentrations at ground levels. Moreover, while there have been major improvements, many sources of pollution are just as bad as ever. Acid rain is produced when sulphur dioxide and nitrogen oxides combine with oxygen in the air and with water vapour to form sulphuric acid and nitric acid. The SO_2 is

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derived mainly from the burning of fossil fuels that contain sulphur. Nitrogen oxides are a product of the chemical combination of oxygen and nitrogen at high furnace temperatures and in internal combustion engines. These gaseous emissions may be carried hundreds and even thousands of kilometres before the atmosphere is depleted by dry and wet deposition. The farther these emissions travel, the longer they are exposed to reactions of different pathways ending up with partly acid sulphates and nitrates. The deposition of these substances may change the chemistry of soil, making it less alkaline and more acidic; they may damage foliage that absorbs them; and they may change the chemical balance of vulnerable lakes, so that these can no longer support aquatic life.

The acidity of precipitation varies greatly from day to day but in the annual mean it is now at about a pH-value of 4.2 to 4.5 at rural sites in Germany. That means the precipitation is about 25 fold more acid than it would be if there were no air pollution but had a CO_2 content of about 340 ppm.

In the case of long range transport, the actual concentrations of air pollutants are generally small and nearly always below air quality levels. It is the long-term accumulation by deposition that is important.

The precise response of terrestrial ecosystems to acidic and other toxic substances is extremely difficult to assess and predict. For instance, serious changes have been noted in soil chemistry which are believed to be triggered by acid rain. These changes are reflected in the serious degradation of beech and spruce forests. Every environmental hazard has its own biological monitor. Forests act acid concentrators, that means acid reaching the forest ground annually is two (beech) to four (spruce) times the acid in the rain entering the forest canopy. This is so because foliage and bark capture and oxidize sulphur dioxide present in the atmosphere in very low concentrations. The resulting acid is then added to the acid aerosols already deposited in the canopy from the air. When it rains, both these acid sources are cleansed from the trees and reach the forest ground.

The increased acid input slows the breakdown of humus, and carbon and nitrogen stores in the soil are increasing. The increased nitrogen in the soil undergoes additional reactions that generate more acid, further stressing the trees, mainly by releasing inorganic aluminium into the soil solution. This aluminium is toxic to the fine roots in the soil's mineral layer, and they become damaged. The trees survive though they are severely stressed because the fine rootlets in the humus layer above are less effected by the organically complexed aluminium at that soil level. Although the trees' ultimate survival is in doubt, liming is recommended as a remedial measure to rectify damage done by the altered soil chemistries.

Despite numerous investigations, the acid rain problem is a controversial subject that encompasses scientific, economic, and political issues. The most obvious requirement is to achieve a sound scientific understanding of the problem. But this is hampered by the fact that available data and numerical calculations at the moment cannot obviate contrasting views.

There are disturbing, unresolved problems that make it difficult even to know what the true pH of rain, not influenced by anthropogenic activities, ought to be, although most scientists believe it to be at about 5.6. There are natural sources of acidifying substances emitted into the atmosphere as well as pollutants arising from combustion of fuels and other processes. Neutralizing substances present in the atmosphere also affect the acidity of rain. Because of the episodic nature of precipitation events it may require decades of information on the composition of rain before trends can be established unambiguously.

Rain contains elements important for plant growth, namely nitrogen and sulphur, as well as excessive acidity. Nitrogen and sulphur in rain are absorbed by foliage and by plant roots from the soil and are then used in support of plant productivity. Excessive acidity, however, interferes with physiological processes and can reduce growth and yield. There are, then, two opposing influences at work and the net effect seems to vary with species, stage of plant development, pattern of rainfall, soil nutrient supply and probably other factors as well. Because the biological processes are complex, it appears that long-term studies are needed to provide conclusive answers to the harmfulness of acid rain. In an analogous manner, the exact contribution of acid deposition versus natural processes in the acidification of lakes cannot be assessed conclusively.

One important question about acid deposition concerns the scale of effects that result from the deposition of substances from the atmosphere. Industrial societies now appear capable of inadvertently altering regional and even global atmospheric chemistry. Perhaps one of the most disturbing consequences of acid rain is that risks of uncertain magnitude may be imposed on the population of other nations. Being aware of this situation, the member countries of the Economic Commission for Europe (ECE) of the United Nations have signed in 1979 a convention in order to combat transboundary air pollution fluxes. A scientific co-operative programme, called EMEP, is part of this convention; the programme is to proceed with the investigation into the fundamentals of the mutual impact of other nation's emissions. The USA and Canada are carrying out a comprehensive research programme in accordance with the memorandum of intent on transboundary air pollution concluded in 1980. There are some efforts in this direction in Germany as well, in order to obtain sufficient information on which to base effective action to prevent ecology suffering from harmful impacts. But the main difficulty is not resolved as yet: it culminates in the question: how can uneffective decisions be avoided when strategies have to be developed with an imperfect knowledge of the problem. This is a fundamental issue that requires thorough discussion and analysis if the decision makers are set to national environmental policies to deal with the acid rain problem. Sufficient information does not exist on the extent of damage, on the causes, and on the transport/transformation mechanism that contribute to acid deposition. Owing to this complexity, a linear relationship between emission rates and deposition rates at specific sites cannot be expected at all.

Concluding my short glance on the acid deposition problem, I would like to invite the assembly to draw their attention to the question of public acceptance of long-term environmental and ecological risks. The research into the deposition of acid and, more general, of harmful substances can be an adequate incentive.

Dry Deposition

FIELD MEASUREMENTS OF THE DRY DEPOSITION OF SMALL PARTICLES TO GRASS

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ABSTRACT

Deposition rates determine the extent and intensity of the effects of atmospheric pollution. Previous measurements of the rate of dry deposition of small particles have suggested that there may be a systematic difference between wind tunnel and field results. Three recent field experiments on the dry deposition of lead, Aitken nuclei and 1.8 μm monodisperse particles are discussed. They give deposition velocities within a factor of three of the wind tunnel data. The larger deposition velocities suggested in some other experiments are not consistent with estimates of the atmospheric lifetime of small particles.

1. INTRODUCTION

Dry deposition is an important mechanism for the removal of certain reactive gases and large particles from the atmosphere. The deposition of smaller particles in the region 0.05 to 1 μm is of interest since this region contains a large fraction by mass of the atmospheric aerosol. In particular, the sulphate aerosol, some heavy metals, and many other products of combustion are found in this size range, and the rate of deposition influences the geographical distribution and intensity of the effects of industrial emissions on plants, soil chemistry and crop contamination.

Several investigators have studied the deposition of small particles to smooth and rough surfaces and elements of foliage in wind tunnels, (eg see Chamberlain, 1966; Clough, 1975; Möller and Schumann, 1970, Belot et al, 1976). Results are usually expressed as the deposition velocity.

$$v_g = \frac{F}{\chi(Z_r)}$$

where F is the flux density of particles to the surface, and $\chi(Z_r)$ the concentration measured at a suitable reference height, Z_r, within the boundary layer above the surface. Wind tunnel experiments give direct measurements of v_g to grass and similar surfaces, and permit deposition

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to forest to be predicted (Belot et al, 1976). For particles in the range 0.05 to 1 μm diameter the wind tunnel results indicate a minimum of v_g (eg see Fig 1) with values in the range $3x10^{-3}$ to $5x10^{-2}$ cm s^{-1} for a wide range of wind speeds and for surfaces as different as short grass and forest. Larger and smaller particles deposit more rapidly, due to increasing departure of particle trajectories from the gas flow occasioned by Brownian motion (for smaller) and inertia (for larger particles)

Field observations of the deposition velocity of particles in the 0.05 to 1 μm size range are difficult to perform and few are reported in the literature. They give more variable results and often indicate deposition velocities larger than 0.1 cm s^{-1}. Some published experiments are attempts to use micrometeorological methods. Everett et al (1979) measured the variation of the concentration of sulphate with height and deduced deposition velocities of 1 or 2 cm s^{-1}. Wesely et al (1977) found deposition velocities of 0.1 to 1 cm s^{-1} using an electrical charging device to detect particles of about 0.05 to 0.1 μm in an eddy correlation experiment.

Several trace elements commonly occur in the size region of interest, and direct measurements of their accumulation on grass or on artificial surfaces provide evidence of the deposition of small particles. Davidson and Friedlander (1978) considered the observed fluxes of lead to grass (which correspond to $v_g \sim 1$ cm s^{-1}). The lead, chiefly from motor exhaust, is predominantly sub-micron, but they explained the large v_g by the presence of small numbers of large particles. The same explanation may account for the deposition velocities to filter paper in the range 0.2 to 0.7 cm s^{-1} found by Cawse (1974) for several trace elements with submicron median diameters in the atmospheric aerosol. However the large values of deposition velocity for submicron particles observed in some field measurements have not been reconciled with the wind tunnel observations.

The large difference between some estimates of v_g obtained in field experiments and in the wind tunnel suggests that there may be an important difference between the mechanisms of deposition in the two circumstances. It has been suggested that such a difference might arise because of differences in the spectrum of turbulence in the two situations; certainly eddies of metre to kilometre scale exist in the atmosphere, while the eddie size in the wind tunnel is limited by the dimensions of the tunnel cross section and cannot usually exceed ~ 1 m. it is important to resolve this issue since the higher values of deposition velocity suggested in some field experiments would require an important change in our understanding of the lifetime and behaviour of the atmospheric aerosol.

Here we discuss the results of three recent experiments regarding the deposition of particles near the size of the minimum of the deposition velocity curve.