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ACID PRECIPITATION LITERATURE REVIEW 1986:

EMISSION ,TRANSPORT ,TRANSFORMATION AND DEPOSITION OF ACIDIC TRACE SPECIES

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SUMMARY

More and more effort is devoted to the investigation of the atmospheric distribution and chemistry of compounds derived from nitrogen oxides and ammonia, and from biogenic sulphur compounds. The annual deposition of nitrate and ammonium in southern Scandinavia is approx. 7 kg ha^{-1} as NO_3^--N and NH_4^+-N, while it is approx. 15 kg ha^{-1} as SO_4-S (Semb and Dovland, 1986).

Measurements of PAN, NO, NO_2, HNO_3, and nitrate in aerosol at a remote location in the Colorado Mountains showed that the concentration of PAN is comparable with NOx (=NO+NO_2) during the fall, and slightly less in the summer. HNO_3 and NO_3^- were usually 100 ppt of order of magnitude (Fahey et al., 1986; Parrish et al., 1986).

Biogenic sulphur contributes to the "background" sulphur in the atmosphere. Dimethylsulfide (DMS) is an important biogenic sulphur species with mainly marine sources, and about 40 TgS/yr is estimated to be emitted into the atmosphere (Ferek et al., 1986).

The emissions of SO_2 from anthropogenic sources in Northern Europe seemed to have dropped by about 20% from 1978 to 1983 (Semb and Dovland, 1986). Improved emission inventories for NOx and VOC in Europe are being established through the PHOXA-project (Stern and Builtjes, 1986) and through ongoing activity in OECD.

Studies of the dry deposition of HNO_3 indicate that below -3^0C over a snow surface, very little HNO_3 is dry deposited (Johansson and Granat, 1986). Average midday deposition velocities for North America for SO_2, sulphate and HNO_3 were calculated to be 0.8, 0.2 and 2.5 cm/s respectively (Walcek et al., 1986), while at night the values were about 50, 45 and 70% of the corresponding daytime values.

Aircraft measurements in FRG have shown that the concentration of SO_2 is high in situations with easterly winds across the FRG/DDR-border, while NOx is relatively low. In westerly winds, NOx dominates over SO_2 (Beilke et al., 1986). In central Ontario, Canada, SO_2 and NOx were

much higher in winter than in summer, while O_3 and $SO_4^=$ are much reduced. HNO_3, PAN and NO_3^- showed little seasonal change, and PAN represented about 30% of total oxidised nitrate. The molar ratio of nitrate to sulphate was 0.1-0.2 in summer, in winter 1-2 (Anlauf et al., 1986).

Both in Europe and North America there is an extensive effort put into model calculations of acid deposition. A new version of the EMEP-model was used to calculate the sulphur budget for Europe for several years (Lehmhaus et al., 1986). The country-by-country budget for sulphur is quite different from the results of their earlier versions of the model. The new calculation fits better with observations. On a national basis, 28 kt(S)/yr is emitted in Norway and 194 kt(S)/yr deposited, of that is 92 kt undecided, and Norwegian emissions, DDR, Poland, USSR and the UK all contribute approx. 7% of the total deposition of sulpur in Norway.

Some results from the calculation of ozone in episodes over Europe have been published using the PHOXA-model (Stern and Builtjes, 1986). The calculation indicates that changes in the emissions of NOx and VOC mainly will move areas of maximum ozone rather than lowering the maximum concentration. Substantial reductions in NOx- and VOC-emissions are needed to obtain a significant ozone reduction over Europe in episodes.

Oppenheimer has continued his research into the relationship between emissions and deposition of sulphur, and found that there is a linear relationship between emissions and concentration in the monthly mean measurements of SO_2 and $SO_4^=$ in the National Atmospheric Deposition Program in the U.S. (Epstein and Oppenheimer, 1986). Hidy et al. (1986) interpreted the same measurements somewhat differently.

CONTENT

ACID PRECIPITATION LITERATURE REVIEW 1986: EMISSION, TRANSPORT, TRANSFORMATION AND DEPOSITION OF ACIDIC ATMOSPHERIC TRACE SPECIES

1 TRENDS IN ACID DEPOSITION

Analysis of an ice core drilled on south Greenland showed that excess sulphate has tripled since 1900-1910, while nitrate has doubled since 1953 (Mayewski et al., 1986). Results published by Finkel et al., 1986, support these conclusions.

The concentration of $SO_4^=$ and NO_3^- in precipitation from 1900 to 1980 at Hubbard Brook, New Hampshire, was modelled by Fay et al. (1986a) using constant meteorology and an annually varying emission strength. It was found that US emissions contributed 0.8 mg/l of $SO_4^=$ in 1900 and 2.0 mg/l in 1920, remaining fairly constant afterwards, while NO_3^- increased steadily from 0.25 mg/l in 1900 to 1.3 mg/l in 1980.

Rodhe and Rood (1986) used data from the European Air Chemistry Network (EACN) to show that the nitrate concentration at most north European stations approximately doubled between the late 1950s and early 1970s, with less pronounced increase in NH_4^+. There is no further increase between 1972 and 1984 at 12 Swedish sites, while sulphate dropped by 30% for the same locations and time period.

Semb and Dovland (1986) presented data on acid deposition from Scandinavian background sites, and showed that there are changes in some areas reflecting reduced SO_2 emissions (see Figure 1).

Davies et al. (1986) claimed that long-term variations in the atmospheric circulation can affect acid deposition levels in the UK and mask the effect of changing emissions.

A historical perspective on acid deposition was presented by Schell et al. (1986) who showed data on the accumulation of many elements in peat bogs which receive their nutrients and moisture from the atmosphere. In Figure 2 is shown the results for the deposition of C, S, N and Br in a peat bog core in Pennsylvania. The major compound of a bog

is the carbon content, usually 40-60% of the dry weight. The loss of C with time is due to microbial decay processes. On the other hand, it is believed that S and N added artificially are fixed rapidly, and of the total S most would be organic bound and would accumulate in organic rich soils.

A mountaintop peat bog in Pennsylvania was analysed to give a record of the deposition of atmospheric chemical over nearly two centuries (Schell, 1986). It was found that the current sulphur accumulation rate is at least 20 times that corresponding to 1917, for total nitrogen the deposition rate now is 45 times the 1917-rate (100 kg $ha^{-1}yr^{-1}$).

In a long-term, global model calculation, Hov and Isaksen (1986) showed that due to changes in the emissions of NOx and hydrocarbons over the last decades, the gas phase conversion of SO_2 and NOx to sulphate and nitrate may have become more efficient, while the liquid phase conversion of SO_2 to sulphate is less influenced.

An application of an analytical model suitable for climatological long range dispersion problems, was described by Matthias and Lo (1987).

Methods for the detection of changes or trends in data from networks measuring SO_2 and sulphate, were discussed by Munn et al. (1986), while Terry et al. (1986) reported on a method to fill in missing values in a time series of acid rain composition.

Holdsworth (1986) claimed that nitrate in ice-cores from suitably located glaciers reflect the increased fixation of nitrogen by nuclear fireballs.

2 ACID SUBSTANCES IN THE REMOTE TROPOSPHERE ("clean air")

Aerosol exchange in the remote troposphere with the emphasis on Antarctica was reported by Hogan (1986). Ito et al. (1986a) published balloon observations of aerosol in the Antarctic troposphere, while Bodhaine et al. (1986) published aerosol mesurements at the South Pole where NOAA operates an atmospheric monitoring observatory. A nearly

continuous record of the concentration of condensation nuclei exists since 1974, and there is a maximum exceeding 100 cm^{-3} in the austral summer and a minimum of 10 cm^{-3} in the winter. Legrand and Delmas (1986) reported measurements of nitrate in Antarctic firn samples, and showed that tropospheric gaseous HNO_3 is the dominating source. H_2O_2 is an important atmospheric trace constituent, and it is thought to be the main oxidising agent for S(IV) to S(VI) transformation. Neftel et al. (1986) found H_2O_2 to be one of the dominant trace components in the ice drilled at Dye 3, Greenland and at Byrd Station in West-Antarctica. In Greenland ice there was a decrease in H_2O_2 with increasing depth, while in Antarctica a peak in H_2O_2 was found in the period 6-12000 yr before present.

Ziereis and Arnold (1986) reported the first measurements of gaseous NH_4^+ ions in the free troposphere between 4 and 8 km. The concentrations were very low (0.1 ppt and less), supporting the generally held view that ammonia is predominantly present as an aerosol.

PAN (peroxyacetyl nitrate) is an important compound in the tropospheric budget of NOx-compounds, both on a continental scale and a global scale. Penkett and Brice (1986) published clean air measurements of PAN averaging more than 300 ppt in the spring in the UK. Clean air was identified when the CFC-concentrations were lower than the Northern Hemisphere average. Singh et al. (1986) presented measurements of the global distribution of PAN, showing that the abundance decreased from 50-100 ppt in the Northern Hemisphere over marine areas, to approximately 1/10 of this value in the Southern Hemisphere atmosphere.

At Niwot Ridge in the Colorado mountains (more than 3000 m.a.s.l.) Fahey et al. (1986) found PAN to be comparable with NOx ($=NO+NO_2$) on the average during the fall and slightly less than NOx in the summer.

Measurements of HNO_3 and nitrate in aerosol at Niwot Ridge in the Colorado Mountains at more than 3000 m.a.s.l. for a 5-year period were reported by Parrish et al. (1986). The order of magnitude of the concentrations of HNO_3 and NO_3^- were usually 100 ppt. The ratio HNO_3 to NOx had a much more pronounced diurnal variation in summer than in

winter, supporting a photochemical conversion mechanism of NOx to HNO_3.

Measurements of ozone in surface air in the Southern Hemisphere were reported by Galbally et al. (1986). At the global background station at Cape Grim on Tasmania, the maximum 1-hr average concentration was less than 40 ppb, while in a more industrialized area east of Melbourne, O_3 exceeded 40 ppb between 1 and 3% of the time.

Sahara dust significantly increases the pH of rain water in the Mediterranean when precipitation falls from air masses which have passed over the desert (Loÿe-Pilot et al., 1986). In this context it is of interest to note that the concentration of mineral aerosol at Barbados during 1983 and 1984 exceeded the concentrations measured before the Sahel drought by a factor of four (Prospero and Nees, 1986).

Troposheric background concentrations of SO_2, NOx and aerosol species were measured on Bermuda by Wolff et al. (1986) in SE winds ($SO_4^=$ 1.1 $\mu g/m^{-3}$, HNO_3 41 ppt). Corkum et al. (1986) reported PAN-measurements on the north shore of Lake Erie at a rural site for 1980-1981, with monthly average daily maximum concentrations from 2-5 ppb, and daily means of 1-2 ppb, highest in summer.

Data on atmospheric deposition to remote areas were also reported by Buat-Ménard and Duce (1986), Adams et al. (1986), Altwicker et al. (1986a, 1986b), Johannes and Altwicker (1986), Laird et al. (1986) and McBean and Nikleva (1986).

2.1 ARCTIC AIR

An overview of current knowledge about arctic air pollution was published by Barrie (1986a). Glacial records indicate a marked increase in Arctic air pollution since the mid 1950s. Air chemistry observations in the Canadian Arctic were reported by Hoff and Barrie (1986).

Measurements made at Alert (82^0N and 62^0W), North-West Territory, Canada show that PAN is the dominating NOx-species in the spring (Bottenheim et al. (1986)).

Aircraft measurements of air pollution in the Norwegian Arctic have shown evidence for a vertical layering of pollution, with significant pollutant concentrations at several km altitude, with different source regions compared to pollution layers closer to the ground (Ottar et al., 1986).

2.2 BIOGENIC SULPHUR

The cycle of biogenic sulphur compounds is receiving increasing attention through ambient measurements, laboratory kinetic studies and model work. The interest in biogenic sulphur is mainly based on its contribution to background sulphur in the atmosphere (Staubes et al., 1986; Ockelmann et al., 1986; Nielsen et al., 1986; Brimblecombe and Shooter, 1986a; 1986b; Barnes et al., 1986a; Martin et al., 1986; Carroll et al., 1986; Barnes et al., 1986b; Yin et al., 1986; MacLead and Aschermann, 1986; Brown, 1986; Adewuyi and Carmichael, 1986; Dacey and Wakeham, 1986; Johnson and Harrison, 1986; Mayo et al., 1986; Amundsen et al., 1986).

Natural sulphur in the global atmospheric sulphur cycle is partly originating from H_2S-emissions from coastal marine sediments, and Aneja (1986) measured marine H_2S-fluxes of the order of 0.1 $gSm^{-2}yr^{-1}$.

The vertical distribution of dimethylsulphide in the marine atmosphere was studied by Ferek et al. (1986). Dimethylsulphide (DMS) is excreted into sea water by marine phytoplankton and then transferred across the air/sea interface into the atmospheric boundary layer. This process represents about one-half of the estimated global natural sulphur emissions. About 40 TgS/yr is estimated to be the atmospheric input of DMS, contributing significantly to SO_2 in the free troposphere.

Measurements of excess sulphate and methanesulfonate in maritime aerosol at Cape Grim since 1976 have been reported by Ayers et al. (1986), and the mean concentrations over this time period were 2.80 and 0.176 $nmole/m^3$ respectively, or about half of the values reported for 9 sites in the Atlantic and the Pacific (Parungo et al., 1986; Saltzman et al., 1986; Okita et al., 1986, Ito et al., 1986b).

A global budget of the uptake of COS by vegetation and soil was estimated by Brown and Bell (1986), who speculated that vegetation may be the major global sink for COS (2-5 Tg yr^{-1}), contrasting soil uptake of only 0.04 Tg yr^{-1}.

A simple model of the flux of sulphur from the Gulf area to the North American continent showed that the possible range is 0.25-0.04 Tg(S)/yr, including dimethyl sulfide and carbonyl sulfide (Luria et al., 1986).

3 REACTION KINETICS

A number of papers dealing with the kinetics of atmospheric sulphur and nitrogen species were published by Kok et al., 1986; Lee and Lind, 1986; Leod et al., 1986; Austin et al., 1986 and Johnston et al., 1986. Weschler et al. (1986) and Graedel et al. (1986) published model studies of reactions and chemistry of transition metal ions in atmospheric droplets and raindrops.

Chameides (1986) calculated that at night, NO_3 radicals can be formed in the gas phase and taken up in droplets at a rate which can enhance the SO_2 to sulphate conversion.

The gas-phase conversion of SO_2 to sulphate through the reaction with methylperoxy radical, has been thought to be of some importance for the atmospheric sulphur-budget. Cocks et al. (1986) found that the equilibrium

$$CH_3O_2 + SO_2 \underset{k_{-1}}{\overset{k_1}{\rightleftharpoons}} CH_3O_2SO_2$$

followed by

$$CH_3O_2SO_2 + CH_3O_2 \xrightarrow{k_2} CH_3O_2 + SO_3 + CH_3O$$

takes place with

$$\frac{k_1 \cdot k_2}{k_{-1}} \approx 1.3 \times 10^{-2} \text{ ppm}^{-2}\text{s}^{-1}$$

and it is estimated that these reactions are unimportant in the production of sulphate from SO_2 in the troposphere.

A review of the chemistry of carbonyl compounds in the atmosphere was published by Carlier et al. (1986).

Russell et al. (1986) demonstrated the importance of the NO_3-radical in nighttime atmospheric chemistry in the formation of PAN and HNO_3.

On the mountain Great Dun Fell in Cumbria in the UK, there is a research station for meteorology, cloud microphysics and atmospheric chemistry. It is operated by the Weather Service and the University of Manchester and is used by institutions like CERL, Harwell Laboratory and the School of Environmental Sciences at the University of East Anglia. Optical and microphysical characteristics of clouds enveloping Great Dun Fell, were described by Choularton et al. (1986).

Hill et al. (1986) reported on a model of the dynamics and microphysics of a hill cap-cloud. Measurements at Great Dun Fell in Cumbria, UK were used for model validation. It was found that the oxidation of SO_2 to sulphate takes place through reaction with e.g. H_2O_2 in such clouds, and that this process is oxidant limited.

The chemistry occurring during cloud formation over hills in northern England was modelled by Hough (1986).

McElroy (1986c) reported on the results of the oxidation of aqueous formic acid by O_3 and H_2O_2.

Sources of H_2O_2 in cloudwater were discussed by McElroy (1986b), who found that scavenging of gaseous HO_2 may give rise to H_2O_2 formation in cloud droplets. The same author also studied the aqueous oxidation of SO_2 by OH radical (McElroy, 1986a).

Cocks and McElroy (1986) have reviewed the role of metal ions in atmospheric droplet chemistry, and found that oxidation of SO_2 catalysed by iron may be of some importance under UK conditions.

Multiphase processes were studied by Jacob (1986) in a remote, non-precipitating tropical cloud, and by Lee et al. (1986a) for the H_2O_2 + S(IV) reaction in rainwater.

Kinetics and mechanism of oxidation of aqueous SO_2 by O_3 was studied by Hoffmann (1986).

Several approaches to model the mass and chemical composition of atmospheric aerosols containing $SO_4^=$, NO_3^-, NH_4^+ and water, were discussed by Saxena et al. (1986a).

Brimblecombe (1986) published a book on air composition and chemistry. Ozone-induced oxidation of SO_2 in clouds may be important at moderate and low acidity (Miller et al., 1986).

4 DEPOSITION PROCESSES

4.1 DRY DEPOSITION

Johansson and Granat (1986) studied the surface resistance of HNO_3 to a snow surface, and found that the resistance decreased from more than 5 s mm^{-1} at -18^0C to about 1 s mm^{-1} at -3^0C. For periods with temperatures below -2^0C, the dry deposition of HNO_3 was estimated to be at most 4% of the wet deposition of nitrate. Findings in support of such a number were reported by Cadle et al. (1986).

Peat bogs have been analysed by Rapaport and Eisenreich (1986) to estimate the atmospheric deposition of toxaphene, which is a complex mixture of polychlorinated camphene derivatives averaging 67-69% chlorine. It was widely used in the US as an insecticide following the DDT ban in 1972 until its ban in late 1982. In the years 1947-1977 $5x10^8$ kg were produced globally (approx. 1/3 of US DDT-production). Peat bogs derive all their nutrients and hydrological input from the atmosphere. It was found that toxaphene had accumulated at a rate of 0.5-9 $\mu g\ m^{-2}a^{-1}$, indicating atmospheric concentrations in the range 8-150 $\mu g\ m^{-3}$.

A review of the literature on the dry deposition of SO_2 and nitrogen oxides was published by Voldner et al. (1986), with calculated monthly deposition fields for SO_2 on a 127 x 127 km^2 grid for North America.

In the EPA/NCAR-model for acid deposition over North America, land use and meteorological data were used to estimate deposition velocities of SO_2), sulphate and HNO_3 for a 3-day springtime simulation. The average midday deposition velocities were 0.8, 0.2 and 2.5 cm/s, respectively, for SO_2 sulphate and HNO_3, while at night the values were about 50, 45 and 70% of the corresponding daytime values, respectively (Walcek et al., 1986).

Deposition velocities for HCl, HNO_3, NOx, NH_3 and H_2O_2 were reported by Dollard (1986) using gradient techniques.

The dry deposition of ammonia on selected plant species was investigated by Aneja et al. (1986) who found a deposition velocity during the day of 0.3-1.3 cm/s, one order of magnitude lower at night.

Calculations of acid deposition to forests based on yearly or regional averages overlook strongly time and space dependent deposition patterns (Wiman, 1985).

Dry deposition was reviewed by Hicks (1986). Barrie and Schemenauer (1986) reviewed the mechanisms for wet deposition of acid substances through rain and fog, but did not make an assessment of the relative importance of these processes on a regional basis. Sievering (1986) reported measurements of dry deposition rates of sulphur in clean air and high wind conditions. Other investigations of dry and wet deposition were published by Lovett and Lindberg (1986) and Brocco et al. (1986).

Dry deposition fluxes of NO_2 and SO_2 were measured by Edwards and Ogram (1986) using tunable diode laser absorption spectroscopy, while Dasch (1986) reported measurements on how deposition to vegetative surfaces decreased from the perimeter to the interior of forest canopies.

5 MEASUREMENTS

5.1 GASES AND AEROSOLS

The hydroxyl radical (OH) determines the rate of gas phase conversion of SO_2 to sulphate and NO_2 to HNO_3. Hard et al. (1986) reported measurements of OH in two 36-hour periods in 1985, with summer and autumn-maxima of 3×10^6 and 4×10^5 molecules/cm^3, respectively (0.1 and 0.02 ppt). This confirms both previous measurements and theoretical calculations.

Reiter et al. (1986) reported measurements of O_3, SO_2, NOx and other gases at several mountain sites around Garmisch-Partenkirchen for the 1978-1984 time period, among them Zugspitze. There is a late winter peak in SO_2 and NOx at all sites, with a decline in SO_2 over the years while NOx is apparently rising.

The climatology of O_3 and SO_2 was determined by Vukovich and Fishman (1986) for the eastern two-thirds of the U.S. for the summers 1977-81. Based on the monthly distribution of the diurnal maximum of O_3 and of SO_2, it was found that the centers of high SO_2 were affected by the path of anticyclones, but less so than for O_3. High pressure systems that become stationary, weaken and dissipate, have a profound effect on the distribution of O_3 and SO_2.

Cloudwater composition at Åreskutan in central Sweden was measured by Ogren and Rodhe (1986) who found 3 pptv of sum ammonia, 6 pptv of sum nitrate and 30 ng m^{-3} sulphate as a median concentration in air of Arctic origin.

Wallén (1986) used precipitation data from the BAPMon-network and other data to show regional and global distributions and trends in nitrate and sulphate over the last decade. Areas with more than 1.5-2.0 mg(S)/l of sulphate in precipitation were found over large parts of the populated continents, while values less than 0.1-0.2 mg(S)/l were found over the oceans in the Southern Hemisphere.

Altschuller (1986) published a review paper on how NOx is involved in nonurban ozone formation.

Measurements of sulphur deposition with distance from a smelter in the Flin Flon Area of Manitoba, Canada, were reported by Phillips et al. (1986). Tang et al. (1986) published measurements of the spatial pattern of sulphate and nitrate wet deposition in Ontario, Canada, 1981-1983. The spatial and temporal variation of the sulphate to nitrate ratio in precipitation in eastern North America was discussed by Summers and Barrie (1986). Some of their results are shown in Figure 3. Wet and dry deposition of sulphate and nitrate in Eastern Canada 1979-1982 was reviewed by Barrie and Sirois (1986). Barrie (1986b) discussed the relationship between Arctic haze and acid rain in North America.

Lindberg et al. (1986) found that airborne particles and vapors contributed significantly to the nutrient requirements and the pollutant load of a mixed hardwood forest in eastern U.S. Dry deposition was an important mechanism of atmospheric input to the foliar canopy, and measurement by standard bulk deposition collectors significantly underestimated the atmospheric deposition.

A review of the atmospheric chemistry, transport and deposition of nitrogen oxides was presented by Grennfelt et al. (1986) as a status report to the working group on nitrogen oxides within ECE´s convention on long-range transboundary air pollution.

NO_3 and N_2O_5 are important compounds at night, and Atkinson et al. (1986) estimated that N_2O_5 may reach 15 ppb in polluted air at night. N_2O_5 may be converted to HNO_3.

With improved communication between China and the U.S., some joint papers have appeared on air pollution in China, e.g. by Dod et al. (1986) where it is reported that in Beijing, the winter sulphate is produced locally from products of incomplete combustion. Ambient carbonaceous aerosols are derived principally from coal combustion.

Sturges and Harrison (1986) studied the forms of bromine which enter the atmosphere from the oceans. Br may catalyze SO_2 oxidation, and may influence stratospheric ozone.

Alkezweeny et al. (1986) measured ammonia in Kentucky during the summer of 1983. At the surface, NH_3 ranged from 0.04 to 5.6 $\mu g/m^3$, and the concentrations were higher at night than during the day. Also, the aloft concentrations were lower than the surface values.

In the Federal Republic of Germany, extensive flights have been carried out to determine the transboundary fluxes of SO_2 and NOx across the border between FRG and DDR (Beilke et al., 1986). In easterly winds, the concentrations of SO_2 is often high along the border between FRG and DDR, while NOx is relatively low. In westerly winds, NOx dominates over SO_2.

Summer and winter concentrations of acidic atmospheric species and their precursors were measured in central Ontario by Anlauf et al., 1986a. SO_2 and NOx were much higher in winter than summer, while O_3 and sulphate were much reduced. HNO_3, PAN and aerosol nitrate showed little seasonal change. PAN represented about 30% of total oxidised nitrate. In summer, the molar ratio of nitrate to sulphate was 0.1-0.2, in winter 1-2.

A documentation of the Cross-Appalachian Tracer Experiment (CAPTEX) is given by Ferber et al. (1986). Fluorcarbon tracers were used to simulate the long-range transport of pollutants in North America.

5.2 MULTIPHASE MEASUREMENTS

Measurements of the composition of precipitation across the Great Lake States in northern U.S., indicate that the wet deposition of total acidity in the middle and western part of the region is comparable to that of impacted sites in the Adirondacks and in regions of Scandinavia (Glass and Loucks, 1986).

Acid deposition measurements in Italy have been reported by Joannilli et al. (1986), Mosello and Tartari (1986) and Facchini et al. (1986).

Analysis of wintertime aerosols collected in a city in Yugoslavia indicate that up to 20% of SO_2 may be oxidised to sulphate through multiphase processes in winter (Bizjak et al., 1986).

Hegg and Hobbs (1986) reported analyses of the chemical composition of air and liquid water in and around cumuliform clouds in western U.S.

Airborne and ground level field studies of cloud water acidification were carried out in the fall of 1981 and the summer of 1982, for non-precipitating clouds in Ontario, Canada, and results were reported by Leaitch et al. (1986).

A short-term study of organic acids in precipitation during early summer in Wisconsin, showed that there were detectable concentrations of formate and acetate ions, with up to 56 µmol/l of the former and 33 µmol/l for the latter, and the contribution of organic acids to free acidity averaged 18.6% for samples with pH $\leq$ 5.0 (Chapman et al., 1986).

Analysis of a regional cloud and fog event in the eastern U.S. (Weathers et al., 1986) showed that the cloud- and fog water had a pH between 2.8 and 3.09, and the concentrations of nitrate and sulphate were 7-43 times greater than those for average precipitation at four eastern U.S. sites.

Papers on air pollution and acid rain in China were published by Zhao and Sun (1986a, 1986b) both working at the Research Institute of Environmental Chemistry in Bejing. SO_2 emissions from coal combustion in China are estimated to 15-18 Mt/yr as SO_2 and account for more than 90% of the total SO_2 emitted. The emissions of NOx are low because of the pattern of fossil fuel consumption.

The spatial and temporal variability of precipitation in New York State was discussed by Pagnotti and Rao (1986), and they proposed a method to account for the variability in precipitation in achieving targeted wet deposition threshold values.

For a two-year period, the chemistry of daily precipitation samples for a site in southern Indiana was analysed for the effect of seasons and synoptic storm types. The storms were classified as frontal, cyclonic, convective and other. Higher concentrations of sulphate, ammonium and hydrogen ion and lower sodium occurred in the warm

ammonium and hydrogen ion and lower sodium occurred in the warm seasons (April -September) than in the cold (October - March); nitrate, chloride and calcium concentrations were similar in both seasons. Convective and frontal storms contained the highest concentrations of ions, and cyclonic and other the lowest. Frontal storms showed significantly higher concentration of sulphate, nitrate, ammonium and hydrogen ion and lower of sodium in warm seasons than in cold, while cyclonic storms yielded significantly higher nitrate in the cold seasons (Topol et al., 1986).

In the northeastern U.S., between 10 and 15 rain episodes deposit about 60% of the annual wet sulphate. Also, wet deposition of sulphate is much larger in summer than in winter. Golomb et al. (1986) argued that targeting emission reductions to certain seasons and perhaps even dry periods before rain could be more beneficial than an all-year reduction, in order to aim at the reduction of the wet deposition of sulphate.

Saxena et al. (1986b) gave a critical assessment of existing data bases in the U.S. for acid deposition modelling studies.

Granat (1986) published measurements of deposition of acid substances in Sweden for 1983-85, with some information on the seasonal behaviour for the period 1972-1984. Nodop (1986) presented EMEP-data for the 1980-1984 time period. On a yearly basis nitrate contributes about 40% of the acidity of wet deposition, and the ratio of nitrate to excess sulphate is generally higher in winter than in summer.

Ferm et al. (1986) reported the first results from the extended measurement campaign organised by the Nordic Council of Ministers at 6 EMEP-stations in Denmark, Finland, Sweden and Norway in the fall of 1985. The concentrations of H^+, NH_4^+, NO_3^- and SO_4^{2-} in precipitation and NO_2, SO_2, total ammonium, total nitrate and particulate sulphate in air were measured. Total ammonium includes gaseous NH_3 as well as particulate NH_4^+. Total nitrate includes gaseous HNO_3 and particulate nitrate. They were both measured using impregnated filters mounted in open face filter holders. The measurements were made continuously on a 24-h basis at six stations from August 15 to October 31, 1985. High concentrations in air occur episodically at several sites almost

simultaneously. The results are evaluated using backtrajectories. From the data it is clear that NO_3^- and SO_4^{2-} are enriched in precipitation compared to NH_4^+. One possible explanation to this is the in-cloud oxidation of SO_2 and NO_2. Another contributing factor is a higher scavenging rate for SO_4^{2-} and NO_3^- that may be caused by a lower concentration decrease with altitude.

The chemical composition of radiation fog in the Po-valley in Italy was discussed by Fuzzi et al. (1986).

Analysis of bulk precipitation and bulk throughfall collected in a Danish spruce forest showed that there is an enrichment of ions in the throughfall compared to precipitation, caused by interception deposition, release and uptake within the canopy (Freiesleben et al., 1986). Throughfall chemistry was also studied by Shiba et al. (1986).

Data from two national precipitation chemistry monitoring networks show that in northeastern North America, there is a strong annual cycle in H^+, $SO_4^=$ and NH_4^+ deposition and some of the other ions although these cycles are not all in phase. The wet NO_3^- deposition contributes relatively more than $SO_4^=$ to the acidity of snow as compared to rain. Wet deposition is highly "episodic" with about 50% to 70% of the total annual deposition of $SO_4^=$ and NO_3^- accumulating in the highest 20% of the days. Over eastern North America as a whole, dry deposition is approximately equal to wet for both $SO_4^=$ and NO_3^-. Dry may exceed wet in the high emissions zone but drops to about 20% of the total deposition in more remote areas. Deposition via fog or low cloud impaction is an important input to high elevation forests (Summers et al., 1986).

Based on sulphate measurements in deposition at Brookhaven, New York, Hameed and Sperber (1986) estimated a mean sulphate scavenging coefficient of $1 \times 10^{-4} s^{-1}$, virtually invariant for rainfall rates less than 5 mm h^{-1}.

Data on the composition of marine aerosol and precipitation led Keene (1986) to conclude that assumptions involved in sea-salt corrections are not always satisfied.

Möller and Zierath (1986) compared the acidity of precipitation in Europe and North America. The fraction of basic species in European precipitation is twice as high as in North America, whereas no differences are found between Eastern and Western Europe. The nitrate share in Eastern Europe is 1/3 less than in Western Europe. The precipitation chemistry at the site Caribou in Maine, U.S., was reviewed by Artz and Dayan (1986). This site has been included in all major national precipitation chemistry networks.

Measurements on acid precipitation in western North America 1979-1985 were published by Popp et al. (1986). Lee et al. (1986b) at Brookhaven National Laboratory showed from sequential measurements of precipitation that H_2O_2 has a strong seasonal variation, the maximum concentration in summer (120 µM) being 6 times greater than in winter. H_2O_2 also exhibited a strong diurnal variation with a peak in the afternoon and a minimum after midnight. This suggests that H_2O_2 is governed by photochemical activity.

Dew samples collected nightly in rural Pennsylvania showed that the conversion of S(IV) to $SO_4^=$ occured on a time scale of hours. There was a strong diurnal variation in the deposition of HNO_3. Rain was a much more important source of chemical deposition than dew or fog (Pierson et al., 1986).

Aircraft measurements of NOx and aerosols showed at distinct maximum in aerosol particle numbers near the frontal surfaces (Banic et al., 1986). Duncan et al. (1986) reported measurements of precipitation composition at a mountain site in Washington state.

For a site in South Scotland, Cape and Fowler (1986) showed that daily pH measurements gave significantly greater acidities than monthly pH measurements, but these differences were small for months with rain-weighted pH < 5.

Bronin et al. (1986) reported measurements of the vertical distribution and transport of SO_2 and $SO_4^=$ across the western USSR border. They found that the highest SO_2 concentrations were transported with air from Central Europe.

Precipitation chemistry measurements in Alberta, Canada, revealed an average 5 yr wet sulphate deposition of 9.1 kg $ha^{-1}yr^{-1}$ (Bertram et al., 1986).

Measurements of major cations and anions collected at three sites in central Switzerland showed that between 950 and 3500 m above sea level, concentrations decreased but much less for $SO_4^=$ than for NO_3^-. The most acidic events occured in spring and summer and were characterized by increased NO_3^- (Fuhrer, 1986a). Fuhrer (1986b) published a paper on the chemistry of fogwater and estimated rates of occult deposition in an agricultural area of Central Switzerland.

Glavas (1986) reported precipitation analysis at a Mediterranean site in Greece where precipitation from air which has passed over central Europe usually is acidic while from north Africa, the precipitation is neutral or basic due to contribution of soil dust.

Fluoride measurements in precipitation in India show both anthropogenic and sea salt sources of fluoride in precipitation. The average background fluoride concentration in precipitation averaged 5 ppb (Mahadevan et al., 1986).

Gatz et al. (1986) reviewed the role of alkaline materials in precipitation chemistry. Böhm (1985), Topol (1986) and Rodda and Smith (1986) also published papers on precipitation chemistry, while papers on trace metals were published by Chan et al., 1986 and Nriagu and Davidson, 1986.

6 ANALYTICAL TECHNIQUES

NO_2 is an important atmospheric trace species and precursor for nitric acid in the atmosphere. Its concentration at remote locations can be lower than the detection limit of standard instrumentation. Schiff et al. (1986) presented measurements of NO_2 with a luminol instrument with a detection limit around 50 ppt.

Denuder tubes and several filter media for the measurement of atmospheric nitrate, sulphate and sulphur dioxide were tested by Anlauf et al. (1986b) both in the field and in the laboratory.

Automated denuder systems were described by Slanina et al. (1986). Denuders can be applied to measure NH_3, HCl, HF, HNO_3, H_2SO_4, NH_4NO_3, $(NH_4)_2SO_4$ and H_2O_2 in air with a better selectivity than with traditional filters or impinger methods.

Denuders are used to measure e.g. gaseous HNO_3 and particulate NO_3^- in the atmosphere as described by Ferm (1986), and to remove NO_2 from atmospheric samples (Adams et al., 1986).

The exent of SO_2 conversion on Membrana Nylasorb nylon filters has been investigated under field conditions in Canada (Chan et al., 1986). It was found that the observed conversion decreased with increasing SO_2 and increased with increasing relative humidity and ranged from 2-8% of SO_2.

The use of remote sensing techniques to detect air pollution was discussed by Chung (1986) who identified dust clouds from a volcanic eruption, smoke plumes from large forest fires and dust clouds from Sahara from satellite data.

Tunable diode laser absorption spectroscopy was used by Slemr et al. (1986) to measure hydrogen peroxide in the gas phase. H_2O_2 is an important species in the oxidation of SO_2 to sulphate.

A filter pack for the measurement of volatile organic acids (HCOOH, CH_3COOH), total nitrate ($HNO_3 + NO_3^-$), total ammonium ($NH_3 + NH_4^+$), HNO_2, SO_2 and $SO_4^=$, was described by Brocco et al. (1986).

7 EMISSIONS

Gschwandtner et al. (1986) presented a summary of historic emissions of SO_2 and NOx in the U.S. from 1900 to 1980, both as totals and by source category and by major fuel type.

The Sudbury smelter in Ontario, Canada emits 500-800 x 10^3 t(SO_2)/yr. It was shut down from June 1982 to March 1983. It was found that the wet deposition of sulphate in Ontario then dropped by less than 15%, the dry deposition by 10-20% (Lusis et al., 1986),

Wood burning in residential homes is a source of acid substances, woodstove flue gas condensate solutions are predominantly in the 2.8-4.2 pH range (Burnet et al., 1986).

Bowden (1986) assessed the impact of gaseous nitrogen emissions from undisturbed terrestrial ecosystems on local and global nitrogen budgets.

Emissions from power plants fuelled by peat, coal, natural gas and oil were reviewed by Häsänen et al. (1986).

Chester (1986) published a lecture on the relationship between emissions from coal combustion and atmospheric pollution.

Semb and Dovland (1986) gave information on the emissions of SO_2 in 12 countries in North-western Europe for 1978 and 1983, and showed that on the average the estimated SO_2 emissions declined by 23% during this period.

There is a fairly extensive international activity in Europe to collect emission data for SO_2, NOx and VOC in a way which is standardized as much as possible. In the PHOXA-project (Stern and Builtjes, 1986) emission data have been collected for Northern Europe, both east and west, on a grid with mesh size approx. 30x30 km^2. Within OECD, the member countries now are reporting SO_2, NOx and VOC-emissions in a 50x50 km^2 grid and with specification in source categories. This work is not reported in a final form yet.

8 MODELS, INTERPRETATION OF MEASUREMENTS

The first results from the PHOXA-project between West Germany and The Netherlands calculating photochemical oxidants over Europe using an Eulerian grid-model were presented by Stern and Builtjes (1986). The model area covers most of north and central Europe, and for an episode of a few days duration in July 1980, the results for ozone compared quite well with measured values.

In another part of the PHOXA-project, the deposition of acid substances over Europe will be calculated for a 4 weeks period in February-March 1982 using a complex 3-d model (Scherer and Scholl, 1986). The results of the PHOXA-project are primarily intended to assist in the assessment of how emission controls may influence the deposition of acid compounds and the level of ozone and other photochemical oxidants over Europe.

A 3-dimensional gridmodel for the chemistry and transport of photochemical oxidants and acid substances in a combined field of land-sea breeze and mountain-valley wind in Japan, was published by Kitada et al. (1986a). A 3-dimensional cloud chemistry model was described by Tremblay and Leighton (1986).

In a series of papers, a 3-dimensional Eulerian model of transport and chemistry of acid substances and photochemical oxidants was described and applied for North America and Japan (Carmichael et al., 1986; Cho and Carmichael, 1986; Chang et al., 1986; Shim et al., 1986; Hong and Carmichael, 1986a, b; Kitada et al., 1986b). The formation of ammonium nitrate aerosol based on the equilibrium

$$NH_{3(g)} + HNO_{3(g)} \rightleftarrows NH_4NO_{3(s\ or\ aq)}$$

where g means gas, s solid and aq aqueous phase, was calculated and evaluated against NH_3, HNO_3, particulate NH_4^+ and NO_3^- measured in Japan, and it was found that the aerosol equilibrium assumption was consistent with the observational data (Chang et al., 1986). Shim et al. (1986) found that the accuracy of the emission data is more

important than the temporal resolution of the data for the calculation of sulphur deposition over North America.

The parameterization of subgrid-scale transport and conversion of pollutants in clouds must be carried out with care to get a correct impression of the role of strong vertical exchange as opposed to a slow diffusion or slow vertical advection when averaged over large areas and long times. Hong and Carmichael (1986a) discussed a parameterization scheme for such processes in their paper.

Hong and Carmichael (1986b) coupled the STEM-II Eulerian model with the PLUVIUS cloud model to investigate sulphate production in an orographic rain event.

The formation of nitric acid and nitrate was studied in a model by Russell and Cass (1986). For California's South Coast Air Basin for August 1982, the reduction in NOx emissions gave rise to a nearly proportional decrease in total inorganic nitrate, while a reduction in NH_3 emissions would suppress aerosol nitrate formation , resulting in higher HNO_3 .

Stunder et al. (1986) used trajectory analysis to trace the sources of wet deposition at Whiteface Mountain. They found that the injection of tracer material to tag a particular source or source region, could improve the confidence in source attribution.

A chemical mechanism for use in long-range transport calculations of acid substances, was published by Lurmann et al. (1986). Giorgi (1986) described a dry deposition parameterization scheme for use in transport calculations. Merrill et al. (1986) reported on techniques of Lagrangian trajectory analysis in isentropic coordinates.

Comparing the results of a model using constant level trajectories to calculate acid deposition and another model using a combination of isentropic and constant level trajectories, showed that the latter approach was much better than the former when comparing with observations of sulphate and nitrate in precipitation (dePena et al., 1986).

Samson et al. (1986) discussed how the variation of input parameters in a Lagrangian transport model for sulphur compounds could give rise to changes in a source receptor relationship of up to 70% over a 5-yr modelling period. Shannon and Lesht (1986) estimated that 57% of NOx and 54% of SO_2 emitted south of 60^0N in North America, was deposited in the same area. Davis and Glantz (1986) applied a multilayer regional-scale model for acid deposition and compared with the results from a single-layer model and found a high correlation between average air concentrations but a poorer correlation between the predicted wet deposition of sulphate.

Fay et al. (1986b) estimated that in eastern North America, on an annual basis 60% of the sulphur emissions are deposited within the region and 40% flow out of the region. Also, US transport of sulphur to Canada is eight times the reverse flow.

Budd (1986) performed trajectory analysis to determine regional interrelationships between emissions of SO_2 and NOx, and acid deposition in the eastern US, and it was found that the largest emission regions in the eastern and midwestern U.S. were responsible for the most severe acid deposition episodes in the region which was studied.

The chemical mechanism used in the EPA/NCAR regional acid deposition model was described by Stockwell (1986).

Calculations and measurements of SO_2 and sulphate at the EMEP stations were compared for 1980 using a revised version of the EMEP-model (Lehmhaus et al., 1986). In Table 1 the calculated sulphur budget for Europe for 1984 is shown. The country-by-country sulphur budget is quite different from the results of the earlier version of the model. The new calculation compares considerably better with observations.

The nitrogen budget for the United Kingdom and north-west Europe was estimated in a report by Derwent (1986), who found that over the UK, N-deposition was largely as gaseous HNO_3 derived from domestic sources. Derwent and Nodop (1986) presented long-range calculations of acidic nitrogen in north-west Europe.

An air pollution study for Cracow in Poland was reported by Juda (1986). Measured SO_2-values up to 250 $\mu gSO_2/m^3$ were compared with predictions from a 3-d grid model.

Analysis of the monthly mean of measurements of SO_2 and sulphate in the National Atmospheric Deposition Program (NADP) in the U.S. showed a linear relationship between emissions and concentration (Epstein and Oppenheimer, 1986). In letters to Science, Hidy et al. (1986) concluded that one cannot disregard alternative explanations of sulphate behaviour in precipitation in Western U.S. for the conclusion proposed by Epstein and Oppenheimer (1986).

A model calculation of removal of soluble species by warm stratiform clouds was presented by Qin and Chameides (1986) who found that the rainout of highly soluble gases is limited by the rate at which new gaseous material can be transferred from the cloud-free air into the cloud.

A model of the redistribution and transformation of pollutants in a stratus cloud was described by Cho et al. (1986).

Fisher and Clark (1986) tried to arrive at general expressions for the removal of SO_2 in rain, based on a scale analysis of the equations of conservation for cloudwater, rainwater, SO_2 in air, and sulphate in cloud and rainwater. It was found that the removal efficiency of SO_2 is strongly non-linear with respect to the SO_2 concentration, with efficient removal at regional average SO_2 concentrations, but decreasing rapidly at higher SO_2 concentrations.

Chang (1986) presented calculations of the formation of nitrate in rain and snow, while Kleinman (1986) calculated the photochemical formation of peroxides (H_2O_2 and CH_3OOH) in the boundary layer.

Based on daily air and precipitation data from the Canadian APIOS network, scavenging ratios derived from precipitation ($mg\ l^{-1}$) and air ($\mu g\ m^{-3}$) concentrations were estimated to be 4.3×10^5 for $SO_4^=$, 4.6×10^4 for SO_2, 4.7×10^5 for NO_3^- and 8.4×10^5 for HNO_3. (Chan and Chung, 1986).

Measurements of sulphate and nitrate in an urban area in Japan were used by Kadowaki (1986) to show that droplet-phase reactions are important for the SO_2 oxidation to sulphate, while gas-phase reactions are predominant for the NO_2 oxidation to nitrate.

A French research group has published model calculations of the generation of nitric acid in a moist atmosphere during the night, and has included microphysical processes, i.e. activation and scavenging of cloud droplets. They concluded that there is a threshold of about 60% in relative humidity for the disappearance of nitrate (Chaumerliac and Rosset, 1986).

Hsu (1986) discussed two procedures to estimate areal means of deposition from point measurements - named areal-temporal correlation and krieging.

Model calculations show that there is large variability in the concentration of ammonia with height both in the gas and the cloud phases, after a simulated convective storm (Luecken and Hales, 1986). The variations were strongly dependent on storm duration.

A general mathematical framework for modelling long-term average concentration and deposition patterns for atmospheric tracers which are subject to transformation and deposition, was presented by Egbert and Baker (1986). It was shown that the joint statistics of random particle transport and transformation for a generalized particle can be used to calculate long-term mean fields for deposition and concentration.

Statistical long-range transport models have been used to estimate S-deposition only, and it is not clear that such models can be used to estimate the effect of emission reductions since the model parameters describing scavenging and chemistry are taken to be independent of the concentration of the pollutants (Venkatram, 1986a).

Inoue et al. (1986a,b) carried out a regression analysis on hourly NO-concentration data and found that the prediction schemes they developed were applicable for the prediction of concentrations up to 1 h

later. This may be of some interest in urban airsheds with very high NO-concentrations.

A review of the modelling of acid deposition was published by Hough and Eggleton (1986).

The extent of nonlinearity in the atmospheric formation of sulphate was discussed on the basis of model calculation by Saxena and Seigneur (1986), who found that nonlinearities exist over relatively large regions in the U.S. (e.g. over the Northeastern U.S.).

A popular look at the modelling of acid deposition was published by Venkatram and Karamchandani (1986), where several approaches of modelling were presented and results shown.

A statistical evaluation of the relation between precipitation chemistry and some meteorological parameters in the Netherlands, was published by Duysings et al. (1986), while Fisher (1986) investigated the effect of tall stacks on the long-range transport of pollutants.

9 MISCELLANEOUS

Lefohn and Mohnen (1986) reviewed measurements of O_3, SO_2 and NOx at rural sites in FRG to find a possible link between air pollutants and forest damage, but the data material did not allow to draw firm conclusions.

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Table 1: Sulphur budget for Europe for 1984 (Lehmhaus et al., 1986).

PERIOD OF CALCULATION FROM 84 1 1 12 TO 85 1 1 6
TOTAL (DRY+WET) DEPOSITION OF SULPHUR
HORISONTAL - EMITTERS
VERTICAL - RECEIVERS
UNIT - 1000 TONNES OF SULPHUR

	AL	AT	BE	BG	CS	DK	FI	FR	DD	DE	GR	HU	IS	IE	IT	LU	NL	NO	PL	PT	RO	ES	SE	CH	TR	SU	GB	YU	RE	IND	SUM
AL	6	0	0	4	0	0	0	0	0	0	5	0	0	0	2	0	0	0	0	0	0	0	0	0	1	0	0	1	0	9	30
AT	0	39	1	1	26	0	0	4	13	11	0	19	0	0	30	0	0	0	14	0	0	1	0	1	0	2	2	16	0	25	206
BE	0	0	63	0	2	0	0	16	4	18	0	0	0	0	1	0	4	0	1	0	0	1	0	0	0	0	10	0	0	9	147
BG	0	0	0	141	2	0	0	0	1	0	3	2	0	0	1	0	0	0	3	0	3	0	0	0	6	9	0	4	0	22	198
CS	0	10	2	2	407	1	0	5	61	20	1	58	0	0	11	0	1	0	75	0	1	1	0	0	0	5	4	12	0	28	685
DK	0	0	1	0	6	30	0	2	14	10	0	1	0	0	1	0	1	0	9	0	0	0	1	0	0	1	7	0	0	10	94
FI	0	0	1	0	4	1	60	1	8	5	0	1	0	0	0	0	1	1	9	0	0	0	6	0	0	60	4	0	0	67	229
FR	0	3	27	1	26	1	0	349	36	57	0	6	0	1	60	2	6	0	16	1	0	54	0	4	0	1	51	4	1	161	1181
DD	0	2	4	0	90	2	0	7	535	53	0	5	0	0	8	0	3	0	36	0	0	0	0	0	0	2	10	2	0	22	779
DE	0	6	23	1	61	3	0	45	122	414	0	8	0	1	36	2	13	0	29	0	0	3	1	3	0	2	35	5	0	72	918
GR	1	0	0	17	1	0	0	0	0	0	93	1	0	0	2	0	0	0	1	0	0	0	0	0	9	4	0	1	0	26	157
HU	0	5	0	4	23	0	0	1	6	3	1	226	0	0	10	0	0	0	14	0	2	1	0	0	1	3	1	30	0	21	348
IS	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	18	20
IE	0	0	0	0	1	0	0	1	1	1	0	0	0	17	0	0	0	0	1	0	0	0	0	0	0	0	11	0	0	21	56
IT	1	4	1	4	9	0	0	13	5	7	3	9	0	0	654	0	0	0	6	0	0	8	0	2	1	2	3	16	2	91	851
LU	0	0	0	0	0	0	0	2	1	2	0	0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	0	0	0	1	11
NL	0	0	14	0	3	0	0	9	6	41	0	0	0	0	1	0	37	0	2	0	0	1	0	0	0	0	15	0	0	12	152
NO	0	0	2	0	6	5	1	2	15	9	0	2	0	0	1	0	1	14	13	0	0	0	6	0	0	12	14	1	0	92	194
PL	0	5	3	3	138	3	0	6	154	32	1	40	0	0	10	0	2	0	660	0	2	1	2	0	1	18	10	10	0	45	935
PT	0	0	0	0	0	0	0	1	0	0	0	0	0	0	1	0	0	0	0	21	0	19	0	0	0	0	1	0	0	27	72
RO	0	1	0	27	18	0	0	1	9	3	2	35	0	0	6	0	0	0	23	0	36	0	0	0	4	30	1	24	0	56	273
ES	0	1	1	0	5	0	0	17	4	5	0	2	0	0	12	0	0	0	4	4	0	533	0	1	0	0	6	1	3	98	713
SE	0	1	2	1	16	10	7	3	34	15	0	4	0	0	3	0	2	4	30	0	0	1	53	0	0	21	16	2	0	104	323
CH	0	1	1	0	5	0	0	7	4	6	0	2	0	0	37	0	0	0	2	0	0	1	0	11	0	0	2	1	0	14	103
TR	0	0	0	9	1	0	0	0	1	0	6	1	0	0	1	0	0	0	1	0	0	0	0	0	147	19	0	1	0	87	275
SU	0	5	5	14	92	7	21	8	103	37	2	54	0	1	11	0	3	1	203	0	11	1	9	0	72204	22	15	0	398	3174
GB	0	0	5	0	4	1	0	12	7	11	0	1	0	4	1	0	3	0	4	0	0	4	0	0	0	0	523	0	0	61	654
YU	2	5	0	41	11	0	0	3	5	3	10	39	0	0	48	0	0	0	10	0	3	2	0	0	4	8	1	202	0	79	476
RE	4	7	45	37	96	35	19	123	175	158	50	34	0	13	278	1	34	7	156	7	4	217	27	3	61	183	398	34	10	918	3198
SUM	15	97	203	307	1053	101	110	1225	1328	922	179	552	1	401	1226	9	115	28	888	33	65	849	107	27	24325	871	1147	385	17259	516452
	AL	AT	BE	BG	CS	DK	FI	FR	DD	DE	GR	HU	IS	IE	IT	LU	NL	NO	PL	PT	RO	ES	SE	CH	TR	SU	GB	YU	RE	IND	SUM

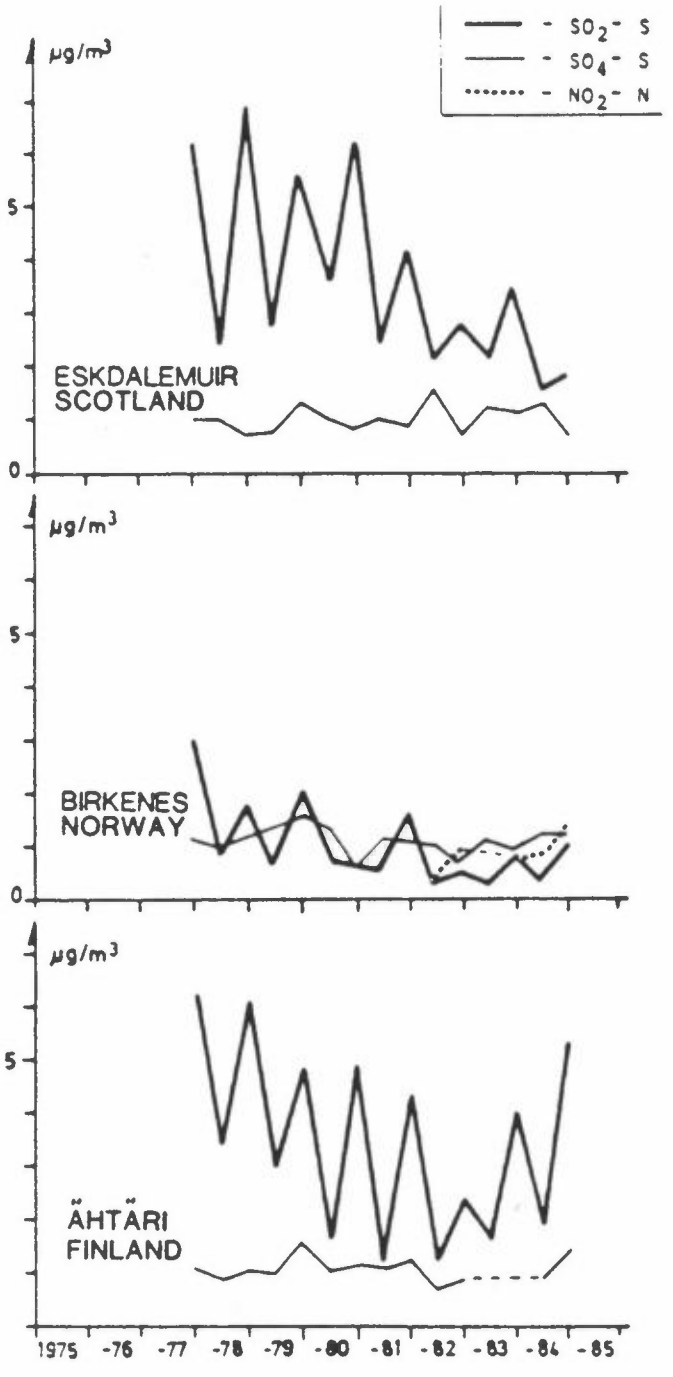

Figure 1: Mean half-yearly concentrations of sulphur dioxide and sulphate in air at 3 EMEP stations (Semb and Dovland, 1986).

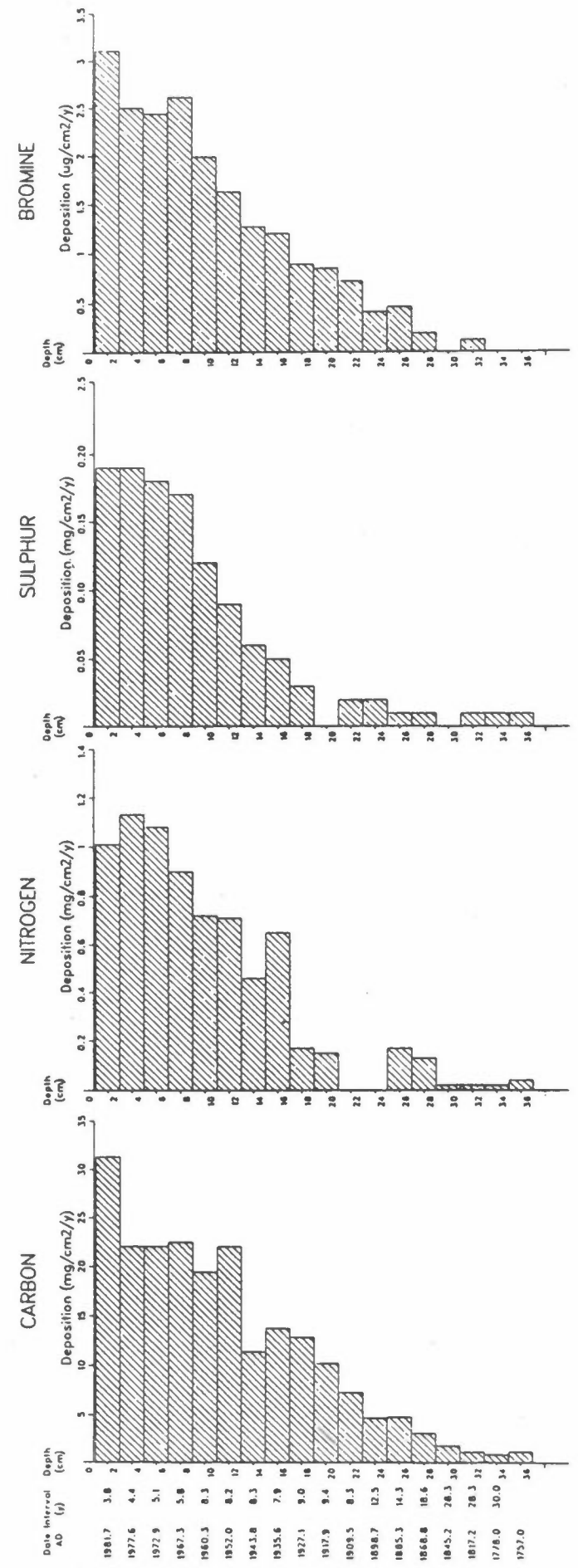

Figure 2: Net deposition of C, S, N and Br in a peat bog core collected in Pennsylvania (Schell et al., 1986).

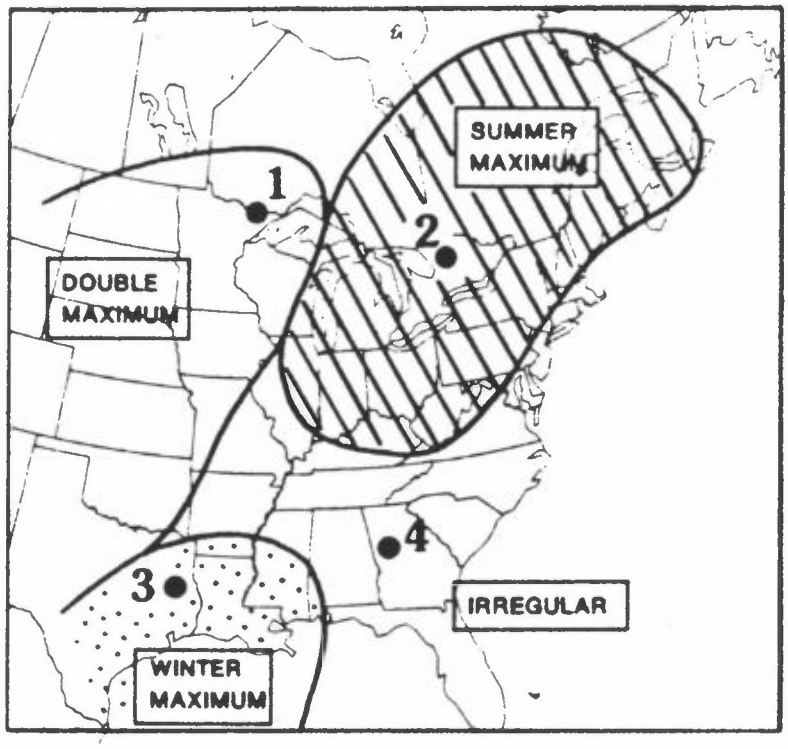

Figure 3: Regions in eastern North America which show the same general behaviour of the annual cycle of the S/N molar ratio in precipitation. The location of the sites are: 1 - Fernberg; 2 - Dorset; 3 - Forest Seed Centre; 4 - Georgia Station (Summers and Barrie, 1986).

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TITLE

ABSTRACT (max. 300 characters, 7 lines)

* Kategorier: Åpen - kan bestilles fra NILU A
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