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ACID PRECIPITATION LITERATURE
REVIEW 1987:
EMISSION, TRANSPORT, TRANSFORMATION
AND DEPOSITION OF
ACIDIC ATMOSPHERIC TRACE SPECIES

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SUMMARY

The role of ammonia emissions in acidification of soils and freshwater in Europe has received considerable attention, in particular through the work of Asman (1987). There has been at least a 2.3 factor increase in the emission of ammonia in Europe since 1870.

The chemistry and transport of NOx compounds in Europe and North America have been the subject of model calculations. Levy and Moxim (1987) found using a 3-dimensional model based on GCM predicted meteorology, that very little of North American NOx emissions reach Europe. A new model for NOx deposition in Europe has been developed in EMEP (Hov et al., 1987a). The objective is to calculate country to country budgets of species derived from the emissions of NOx, SO₂ and NH₃.

Further work has been done to deduce trends in atmospheric trace species. Thirty years of ozone measurements at rural sites in DDR indicate a 1-3%/a linear increase (Feister and Warmbt, 1987).

The concentration of SO₂ at Norwegian background stations did not change much from 1985 to 1986, while NO₂ increased. Sulphate has dropped by about 30% since the end of the 1970s (Joranger et al., 1987).

The study of nighttime chemistry of NOx compounds is receiving considerable attention, in particular reactions involving the transformation of NO₂ to NO₃, N₂O₅ and NO₃⁻ in the absence of sunlight (Nielsen and Cox, 1987).

Measurements of the dry removal of NOx show little or no removal by branches of Scots Pine trees at low NOx-concentrations (NO₂ <1-3 ppb), indicating that biological systems also are a NOx source (Johansson, 1987). Dry removal of aerosol species (like SO₄⁼, NO₃⁻ and NH₄⁺) has been shown to decline sharply from the upwind edge of a forest and downwind over the forest (Hasselrot and Grennfelt, 1987).

Analytical techniques for the measurement of H_2O_2 , $\text{HNO}_3(\text{g})$, nitrate, NH_3 and ammonium are becoming more and more reliable, making it possible to measure simultaneously the concentrations of the key species involved in acid deposition.

The NCAR acid deposition and oxidant model is now ready for use in North America. A description was published by Chang et al. (1987).

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1 INTRODUCTION

Atmospheric processes leading to acid deposition in Europe were reviewed by a COST 611 Task Force (Hov et al., 1987b).

The atmospheric behaviour of ammonia and ammonium was the topic of a doctoral thesis presented by W.A.H. Asman (1987). It contains papers published separately in the open literature on emissions, measurement of atmospheric concentrations, parameters for dry and wet removal, long-range transport model development and an estimated historical development of ammonia and ammonium deposition in Europe 1870-1980 (at least a 2.3 factor increase in ammonia emissions since 1870).

The linearity question was discussed at an international meeting in Hungary (Alcamo et al., 1987a).

Glenn Shaw (1987) speculated that there may have been a significant historic relationship between the abundance of biogenic sulphur particles and the transparency of the atmosphere. Natural sulphate particles are mainly found in the removal resistant gap between 0.1 and 1.0 μm , of the same order as the wavelengths of light. A one or two orders of magnitude increase in sea to air sulphur flux from biota would raise the optical depth of the atmosphere to levels where the atmosphere would become translucent and overclouded.

2 REMOTE TROPOSPHERE

The concentration of ozone has been measured for more than 30 years at some rural stations in the German Democratic Republic using the wet chemical method. There is a significant linear increase of 1-3%/a, with a stronger increase in the summer than in the winter (Feister and Warmbt, 1987).

The mapping of atmospheric concentration in remote parts of the troposphere of species derived from oxides of nitrogen, is scarce and the concentrations variable. Aircraft measurements east of the Asian continent showed ≈ 200 pptv of NO_y (sum of all NO_x -derived species) between 3 and 8 km over the Pacific Ocean between $30\text{--}35^\circ\text{N}$, NO was 15-35 ppt. Over the Japan Sea, up to 1000 ppt of NO_y was measured between 1 and 6 km altitude. Some NO_y at high altitude was of stratospheric origin (Kondo et al., 1987).

Airborne measurements of NO over the Eastern Pacific using several measurement techniques were reported by Hoell et al., (1987), Davis et al. (1987) and Ridley et al. (1987). Mixing ratios from <15 pptv to >100 pptv were found.

High altitude measurements of nitric acid at Mauna Loa, Hawaii, have shown that in 1984-1986 the maximum average monthly concentration during daytime was 97 pptv (August) minimum 18 pptv (February). The summer/winter ratios of NO_3^- , SO_4^{2-} and NH_4^+ were 3, 2 and 0.5, respectively (Galasyn et al., 1987).

Ice samples were obtained from Vavilov glacier (79°N , 95°E) to a depth of 11 m. Analysis by ion chromatograph showed little chemical variation; excess S content ranged from 0 to 0.32 mg/l and resulted from natural factors. Since polluted air masses are transported to such latitudes, pollutant deposition velocities must be very low there (Astratov et al., 1987).

Atmospheric aerosol particles collected at Ny-Ålesund, Svalbard in the spring of 1983 had mass size distributions with modes in the range $0.13\text{--}0.26\ \mu\text{m}$. Overall, the aerosol had properties indicative of one that had remained relatively unchanged after transport from its source region (Heintzenberg and Covert, 1987).

Mayewski et al. (1987) sampled snow near Dye 3 on South Greenland, and found that there was both anthropogenic sulphate deposited and that excess sulphate was deposited when the El Chichon cloud arrived in South Greenland.

3 REACTION KINETICS

Rate constants for some oxidations of S(IV) by radicals in aqueous solutions, were determined by Huie and Neta (1987). They found that secondary radical-radical reactions may be of considerable importance in the mechanism of sulphite oxidation.

The second-order reaction rate constant for the ozone + sulphur dioxide reaction in water, was remeasured by Nahir and Dawson (1987) and could be represented by $k = 1.23 \times 10^4 [\text{H}^+]^{-0.51} \text{ M}^{-1} \text{ s}^{-1}$. The process is of importance in low acidity regions and where the concentration of H_2O_2 is low.

The mass accommodation coefficient for HO_2 radicals on aqueous particles was measured by Mozurkewich et al. (1987) and found to be sufficiently large to be a potentially important source of H_2O_2 . Lind et al. (1987) measured the rate coefficients for aqueous phase oxidation of S(IV) by H_2O_2 , methylhydroperoxide and peroxyacetic acid.

To assess the relative importance of H_2O_2 in atmospheric SO_2 oxidation to SO_4^{2-} , prepared samples of rainwater were acidified with H_2SO_4 and treated with KMnO_4 . The O produced was removed, converted to CO_2 and cryogenically separated for $\delta^{18}\text{O}$ measurement. Results for summer samples from Illinois, New York, Michigan and North Carolina indicate that $\geq 40\%$ the SO_4^{2-} in precipitation was formed by H_2O_2 oxidation of SO_2 (Holt and Kumar, 1987).

Clarke and Radojevic (1987) studied metal catalysis in S(IV) to S(VI) transformation in water droplets and found lower rates than previously published by others.

Humidity effects on photochemical aerosol formation in a SO_2 -NO-C₃H₆-air system was studied in a laboratory experiment by Izumi et al. (1987), Ibusuki and Takeuchi (1987) arrived at expressions for the rate constants of the aqueous oxidation reactions of SO_2 catalyzed by Mn (II) and Fe (III), including the catalytic synergism between the two catalysts.

Däumer et al. (1987) found out that sulphuric acid aerosol coated with organic substances decreased the neutralization of the aerosol through the prevention of an efficient uptake of ammonia.

Intermediate reaction products in liquid phase SO_2 oxidation were discussed by Chang et al. (1987).

The degradation of carbonyl sulphide (OCS) through reaction with OH is measured by Wahner and Ravishankara (1987) and found to be of minor importance in the troposphere.

The atmospheric composition of species derived from NO_x emissions, is not completely understood. The difference between total NO_y and the sum of individual species like NO, NO_2 , HNO_3 and PAN, is often appreciable at remote locations, and is thought to be made up e.g. of alkyl nitrates (of the form RONO_2 where R is an organic radical). The formation of alkyl nitrates is discussed in a paper by Atkinson et al. (1987a).

Nitric acid is formed during the night through the reaction of NO_3 radicals with hydrocarbons. Atkinson et al. (1987b) published reaction rate coefficients of NO_3 radicals with a series of aromatic compounds.

A COST 611 workshop on tropospheric NO_x chemistry-gas phase and multi-phase aspects, was held at Risö in Denmark (Nielsen and Cox, 1987). Emphasis was put in particular on mechanisms and rates of the reaction of the nitrogen trioxide radical with different volatile organic compounds.

Nitrous acid (HNO_2) is formed at night from NO_2 in a reaction which is first order both in NO_2 and H_2O , and proceeds heterogeneously on most unpoisoned surfaces. The reaction is probably an important source of nighttime nitrous acid (Svensson et al., 1987).

The equilibrium constants for dissociation of solid ammonium chloride aerosol into gaseous HCl and NH_3 was calculated by Pio and Harrison (1987a).

There is a temperature and humidity dependent equilibrium between $\text{HCl}(\text{g}) + \text{NH}_3(\text{g})$ and $\text{NH}_4\text{Cl}(\text{p})$. At $\text{rh} > 75\text{-}85\%$, ammonium chloride exists in the liquid phase, with the dissociation constant two orders of magnitude lower at 98% rh than for solid aerosol. Ammonium chloride aqueous aerosol forms mainly in fogwater and cloud droplets and in regions with important local NH_3 sources (Pio and Harrison, 1987b).

Primary quantum yields of NO_2 photodissociation were reported by Gardner et al. (1987).

The calculation of the photolysis of O_3 to $\text{O}({}^1\text{D})$ depends on a good determination of the solar flux in the 300-320 nm wavelength region. A procedure is given by Ritter et al. (1987) to determine appropriate photolysis formation rates of $\text{O}({}^1\text{D})$, which is important as a source of hydroxyl through its reaction with water vapor.

Hydrocarbon reactivity was studied experimentally by Carter and Atkinson (1987).

A detailed chamber study of hydroxyl radical reactions with xylenes was presented by Gery et al. (1987), specifying the yield of gas and aerosol products including species like methylbenzyl nitrates.

Photochemical formation of nitric acid was measured in an outdoor smog chamber in Detroit, and a conversion rate of $10.6 \pm 1.9\% \text{ h}^{-1}$ of NO_x to HNO_3 was found on sunny days. The conversion rate was not strongly influenced by additions of extra NO_x or NMHC, while ozone was (Kelly, 1987).

4 DEPOSITION

4.1 DRY DEPOSITION

Eddy-flux measurements of particle deposition to land surface showed that $v_d = 0.6\text{-}1.0 \text{ cm/s}$ for an unstable period with mean wind of 8.7 m/s , and $0.1\text{-}0.4 \text{ cm/s}$ for a stable period with a mean wind of 6.4 m/s ($0.2 \mu\text{m}$ particles). Aerosol interception by surface roughness elements may account for the deposition at high winds (Sievering, 1987a).

Filter-pack measurements of HNO_3 at a rural site at Harwell, U.K., indicate a higher percentage of HNO_3 of total nitrate (70%) than reported elsewhere in Europe, and dry removal velocities ranging from 5-26 cm/s, significantly exceeding those of other gases (Dollard et al., 1987).

Low dry depositions were measured by Valdez et al. (1987) for SO_2 and NO_2 over snow (of the order of 0.01 cm/s). A physical-chemical model was also described (Bales et al., 1987). Another dry deposition model was described by Chameides (1987).

Using the micrometeorological gradient method, Duyzer et al. (1987) found an average dry deposition velocity of 1.9 cm/s for NH_3 and 0.2 cm/s for ammonium particles.

Using a chamber technique, Johansson (1987) found that the uptake of NO by branches of Scots Pine trees was small for concentrations ranging from about 1 ppb to 50 ppb. The deposition rate of NO was less than 0.1 mm/s. The uptake of NO_2 varied linearly with concentration. At high levels (> 10 ppb) the uptake was limited both by a stomatal and internal resistance with a marked diurnal variation. For NO_2 > 10 ppb the deposition rate per projected needle area was 1-2 mm/s during the day and 0.05-0.2 mm/s during the night. No uptake of NO_2 could be detected at low concentration (1-3 ppb), and a deposition velocity of < 0.1 cm/s was obtained.

Estimates have been made of the dry removal of O_3 and H_2O_2 over north-eastern US, and found to be 0.7-0.8 cm/s for ozone during a spring period dropping to 0.1-0.2 cm/s at night, while for H_2O_2 corresponding to 80 m height, a midday peak of 1.6-2.0 cm/s is predicted with a fall to 0.6-0.9 cm/s at night (Walcek, 1987).

Using the profile method, Nicholson and Davies (1987) measured the dry deposition of particulate sulphate over surfaces typical of much of East England. The mean deposition velocity was 0.07 cm/s.

There is an interaction between NH_3 and SO_2 in dry deposition on plant surfaces because in the first dripping at the beginning of rainstorms (initial throughfall), equivalent amounts of NH_4^+ and SO_4^- are measured (Draaijers et al., 1987). Wet surfaces strongly stimulate the dry deposition of NH_3 and SO_2 , in which case there is a synergistic effect. Dasch (1987b) measured dry deposition to surfaces in deciduous and pine canopies.

Heil et al. (1987) showed that deposition of air pollution is not evenly distributed on a small scale, but depends on the structure of the intercepting surface. Even low vegetation has a strong filtering effect. Grennfelt and Hasselrot (1987) showed in a half-year study of an edge in a pine forest that the sulphur flow by throughfall was 50% higher at the edge compared to the closed forest. Corresponding number for nitrate was 190%.

The uptake of atmospheric ammonia by leaves (of *Phaseolus Vulgaris* L.) was measured by van Hove et al. (1987) and found to increase linearly with concentration in the range 4-400 $\mu\text{g}/\text{m}^3$. Flux densities increased with light intensity. The NH_3 transport into the leaf is via stomata, transport through the cuticle is negligible.

A canopy stomatal resistance model for gaseous deposition to vegetated surfaces was developed by Baldocchi et al. (1987), who found that differences in plant species and environmental and physiological conditions can affect the canopy stomatal resistance by a factor of four.

A model was developed by Meyers and Yuen (1987) to calculate representative day and night deposition fluxes of SO_2 and O_3 .

A "big leaf" model to estimate dry deposition velocities is presented by Hicks et al. (1987), where the canopy resistance to deposition is calculated from information about leaf area index, wetness, temperature, sunshade fractions etc. An application of the model was reported by Matt et al. (1987) where the values were found to be somewhat higher than those derived from eddy correlation.

The deposition velocity of SO_2 was calculated using a higher order closure model for canopy/surface exchange. The leaf boundary-layer resistance contributes only a little to the total resistance to diffusion from the air to the sub-stomatal cavity. The minimum stomatal resistance and the light response coefficients are more important factors (Meyers, 1987).

4.2 WET DEPOSITION

Rainwater and aerosol measurements on Virginia Key in Florida gave annual washout ratios of 330 and 290 for nitrate and non-sea salt sulphate, respectively (expressed as $(\mu\text{g}/\text{kg rain})/(\mu\text{g}/\text{kg air})$) (Savoie et al., 1987).

Scavenging ratios of sulphate on the south-central Greenland ice sheet were discussed by Davidson et al. (1987). Scavenging ratios (dimensionless on a mass basis) of 100-200 in winter and 200-400 in summer were found, the greater summer values attributed to increased riming. Dry deposition was estimated to account for 10-30% of total sulphate deposition on a year-round basis.

A washout ratio of 270 (on a mass basis) was estimated for fine particles ($< 2.5 \mu\text{m}$ diameter) during January and February 1983 on Bermuda. Washout ratios for fine $\text{SO}_4^{=}$ were 2-14 times greater (Wolff et al., 1987).

5 MEASUREMENTS

5.1 MEASUREMENTS OF GASES AND AEROSOLS

Ozone measurements over African equatorial regions during the rainy season were reported by Cros et al. (1987).

Satellite observations of total ozone together with surface measurements were used by Fishman et al. (1987) to show that the space based platform can be used to track air pollution episodes over the United States.

Oxidant data collected in OECD-Europe from April-September 1985 were published in a report by Grennfelt et al. (1987a), as part of a Nordic initiative to coordinate national ozone measurements in Europe. This work will be carried further in the framework of EMEP from 1988.

Ozone and PAN measurements at rural sites in Southern Norway 1984-85 were reported by Schjoldager et al. (1987).

Perner et al. (1987) reported new measurements of tropospheric OH using long path UV absorption spectroscopy, and found concentrations ranging from $0.7-3.2 \times 10^6$ molecules/cm³. At the same time O₃, H₂O, NO, NO₂, CH₄, CO and NMHC were measured and the values used to calculate OH. The modelled OH agreed well with the observed values for NO_x > 2 ppb, for NO_x < 2 ppb, lower OH was measured than calculated.

An hourly maximum PAN concentration of about 9 ppb was measured in Creteil in Paris in October 1985 and almost 7 ppb in February 1986 by Tsalkani et al. (1987). These measurements show that PAN is an important species in the NO_y-budget particularly in cool weather.

Aircraft measurements of H₂O₂, O₃ and SO₂ over Eastern US during fall 1984 showed H₂O₂ concentrations in the range 0.2 ppbv (detection limit) to 4.1 ppbv, with less H₂O₂ near the surface than in aloft air, and with maximum just above cloud tops (Heikes et al., 1987).

Estimates of the natural background ozone concentration at rural ground stations in the US were done by Altschuller (1987a) who found the level to be 10-20 ppb in the summer months.

A survey of UK ozone distributions has been made by the UK photooxidant review group (1987), including a description of measurement methods and analysis of the contribution of UK sources to ozone in the UK.

Lidar measurements of sulphur dioxide and ozone near the ground in Southern France were reported by Ancellet et al. (1987). Measurements of particulate sulphur in and around St. Louis, Missouri were interpreted by Altschuller (1987b).

Transboundary fluxes of air pollutants to the Netherlands were measured by aircraft, and it was found that an accurate determination of both mixing depth and wind velocity was essential for the mass flux calculation (Lelieveld et al., 1987).

Daily ammonia and ammonium measurements at five different sites in the Netherlands showed a strong seasonal variation for ammonia with an early spring maximum. Monthly averaged NH_3 ranged from $0.2 \mu\text{g NH}_3/\text{m}^3$ in a coastal area during westerly winds, to $35 \mu\text{g NH}_3/\text{m}^3$ in the most intensive livestock breeding areas. Ammonia concentrations were halved after 600 m of transport downwind of an area with very large emissions. There is a strong decrease in NH_3 with height (Erisman et al., 1987).

A review was given by Quinn et al. (1987) of the atmospheric concentration of ammonia both in gas and particles and in cloud, rain and sea water. Sampling methods and analytical techniques were also described.

In January 1985 a serious smog episode occurred over most of northwest Europe. Hourly concentrations of SO_2 in excess of $1000 \mu\text{g}/\text{m}^3$ were measured e.g. in the Federal Republic of Germany. The pollution levels in the UK during the smog period were reviewed by Simpson et al. (1987) and in Hungary by Meszaros et al. (1987).

Moore (1987) reported on the conditions in which power stations contribute to high ground level SO_2 concentrations in the UK. The occurrence was often linked to subsidence in slow moving anti-cyclones.

5.2 MULTIPHASE MEASUREMENTS

Fresh snowcover in a Scottish catchment was found to be chemically heterogeneous, with even more variability in an aged snowcover (Tranter et al., 1987). The sampling of snow packs should be done under tight statistical constraints to ensure representative estimates of the solute content of the snowcover.

During April 1981 a comprehensive field experiment was carried out in a region covering roughly 110 km by 110 km around Fort Wayne, Indiana. From 43 precipitation sampling stations combined with aircraft, surface and sequential precipitation chemistry and aloft concentrations of major ions were measured during 4 storms (the OSCAR experiment). Results from the study were reported by Chapman et al. (1987).

A statistical summary and analysis of the event precipitation chemistry data from the MAP3S network in northeastern United States 1976-1983 was published by Dana and Easter (1987). There is a small decrease in sulphate, nitrate and ammonium in rain over the period. There is a summer maximum and winter minimum for sulphate and ammonium in rain, nitrate varies less over the year except at some coastal sites.

Winter rains in the northeastern United States have lower nitrate levels but higher sulphate levels than snows in the same area. The lower nitrate levels in snows are due to the generally lower precipitation depths associated with snow than with rain. The behaviour of sulphate in rain compared to snow can be explained by incloud oxidation of SO_2 to sulphate, while N species are not effectively incorporated into cloud water (Dasch, 1987a).

Seilkop and Finkelstein (1987) used precipitation data from seven nationwide chemistry networks in the US and Canada for 1980-84 to estimate that decreases as large as 15-20% were observed in sulphate and nitrate in rain between 1980 and 1983.

Flossmann et al. (1987) described a model of the processes which control wet deposition of atmospheric aerosols and pollutant gases.

~~The National Acid Precipitation Assessment Program (NAPAP) was discussed in a Science editorial (Abelson, 1987), in connection with the publication of the 4-volume report from the programme (NAPAP, 1987).~~

The wet deposition of nitrate and sulphate in Schleswig-Holstein was estimated to be $20 \text{ kg S ha}^{-1} \text{ a}^{-1}$ and $5.5 \text{ kg N ha}^{-1} \text{ a}^{-1}$ of sulphate and nitrate, respectively, at the North Sea island Sylt and in Schleswig,

12 for S and 4.7 for N in Kiel, 16 for S and 4.3 for N in Lübeck and 18 for S and 4.2 for N in Quickborn near Hamburg (Schnug and Holz, 1987).

The relationship between SO_2 emissions and sulphate in rain was discussed by Chen et al. (1987).

An EMEP workshop on data analysis and presentation was held in Cologne in June 1987 (EMEP, 1987).

The mechanisms of acid rain formation were studied by collection and analysis of rain, snow, cloud, water, aerosols and soil in Colorado (Parungo et al., 1987a).

Hales et al. (1987) discussed which criteria should be satisfied for a measurement network to qualify as a research measurement network and contributor to Federal assessments of acid deposition.

In an experimental setup, Richardson and Hightower (1987) studied the evaporation of ammonium nitrate particles. They concluded that solid ammonium nitrate particles in the atmosphere have a long lifetime and can be transported over long distances regardless of ammonia and nitric acid vapor concentrations, indicating that there is not an instantaneous equilibrium between $\text{NH}_3 + \text{HNO}_3$ and NH_4NO_3 .

The ionic composition of rain at a midcontinent site in the US between 1980 and 1984 was discussed by Wagner and Steele (1987). The composition of snow taken in April 1983 in southern France by Colin et al. (1987) was dominated by nitrate, about twice the amount of sulphate on an equivalent basis. The chemical composition of precipitation at the Sub-Antarctic Marion Island was found to be very similar to that of the surrounding ocean ($\text{Cl} > \text{Na} > \text{SO}_4^{2-} - \text{S} > \text{Mg} > \text{K} \approx \text{Ca}$) (Smith, 1987).

Carbonyl sulfide (OCS) is scavenged by clouds and precipitation. Measurements by Belviso et al. (1987) outside Paris showed that in the majority of 45 rain samples, there was a significant OCS supersaturation, and the excess OCS increases with pH.

In a winter study of air, cloud and precipitation chemistry in Ontario, Canada it was found that the nitrate/sulphate equivalent concentration ratios in cloud water, in precipitation measured by aircraft and ground precipitation were 0.7, 0.6 and 1.4, respectively. This suggests that precipitation scavenging of nitric acid below cloud base is an important process during winter (Isaac and Daum, 1987).

In a summer study in the San Gabriel mountains in Southern California in 1985 it was found that HNO_3 made up up to 73% of the total amount of airborne nitrate, of the NO_y compounds NO_2 made up 69-86% of the total, nitrate 9-15%, HNO_3 4-11%, NH_3 3-9% and NO 1-2%. PAN was not measured (Bytnerowicz et al., 1987a).

In a coastal, urban area in Israel, sulphate particles were mainly in the 0.3-0.8 μm size range, while nitrates were mostly in the μm size range, as NaNO_3 formed through heterogeneous reaction of HNO_3 and sea salt particles (Mamane and Mehler, 1987).

Continuous aircraft measurements of aerosol sulphate and light scattering in cloud interstitial air and in associated clear air at several locations in Eastern US showed essentially complete scavenging of sulphate aerosol by cloud droplets in clouds (ten Brink et al., 1987).

Cloud chamber experiments show that in haze, the heterogeneous SO_2 conversion rate is limited by the supply of condensed water, while in clouds, chemically related factors like pH and oxidant abundances limit the SO_2 conversion rate (Lamb et al., 1987).

Observational and numerical investigation of cumulus cloud scavenging, transport and chemistry in Ontario, Canada showed that the rate of pollution transformation in a cloud did not exceed a few per cent per hour (Tremblay, 1987).

For acidity and sulphate a downward trend has been indicated in monthly atmospheric precipitation samples from 12 stations within Czechoslovakia covering 9 years (Moldan et al., 1987).

Measurements of acidity of rain in the tropical forests of the Ivory Coast show a nearly alkaline pH during the dry season with influence of Sahel dust, and a pH of about 4.2 in the beginning of each humid period (Lacaux et al., 1987).

There is a poor correlation between H^+ and $SO_4^{=}$ in many western and some midwestern precipitation chemistry data sets in the US (Sievering, 1987b). It is believed to be caused mainly by SO_2 to sulphate conversion on the surface of dust particles.

Particulate nitrate in Western US exhibits a north-south gradient with the highest concentrations in the north (White and Macias, 1987).

Organic sulphur compounds were measured by Watts et al. (1987a). Hoppel (1987) calculated that at 100% relative humidity, 1 ppt of MSA (methanesulphonic acid) vapor supersaturates the atmosphere, at 10% rh 5 ppb is required. The annual injection of DMS (dimethylsulphide) to the atmosphere is estimated to be 40 ± 20 Mt S, a significant portion of the global natural S-flux of 60-90 Tg S/a.

The ionic composition of ammonium salts during an acid deposition wintertime episode in eastern Austria in 1987, was reported by Ober et al. (1987).

Trace elements associated with airborne particulate matter were reviewed by Schroeder et al. (1987).

Dissolved organic nitrogen in marine precipitation has been found to consist of dissolved free amino acids as an important class of compounds. These may be oxidized further to nitrate (Mopper and Zika, 1987).

Environmental pollution and control in China was reported by Yuanjun and Zhongxing (1987).

The size distributions of atmospheric particulate sulphate and nitrate as estimated from measurements were reviewed by Milford and Davidson (1987).

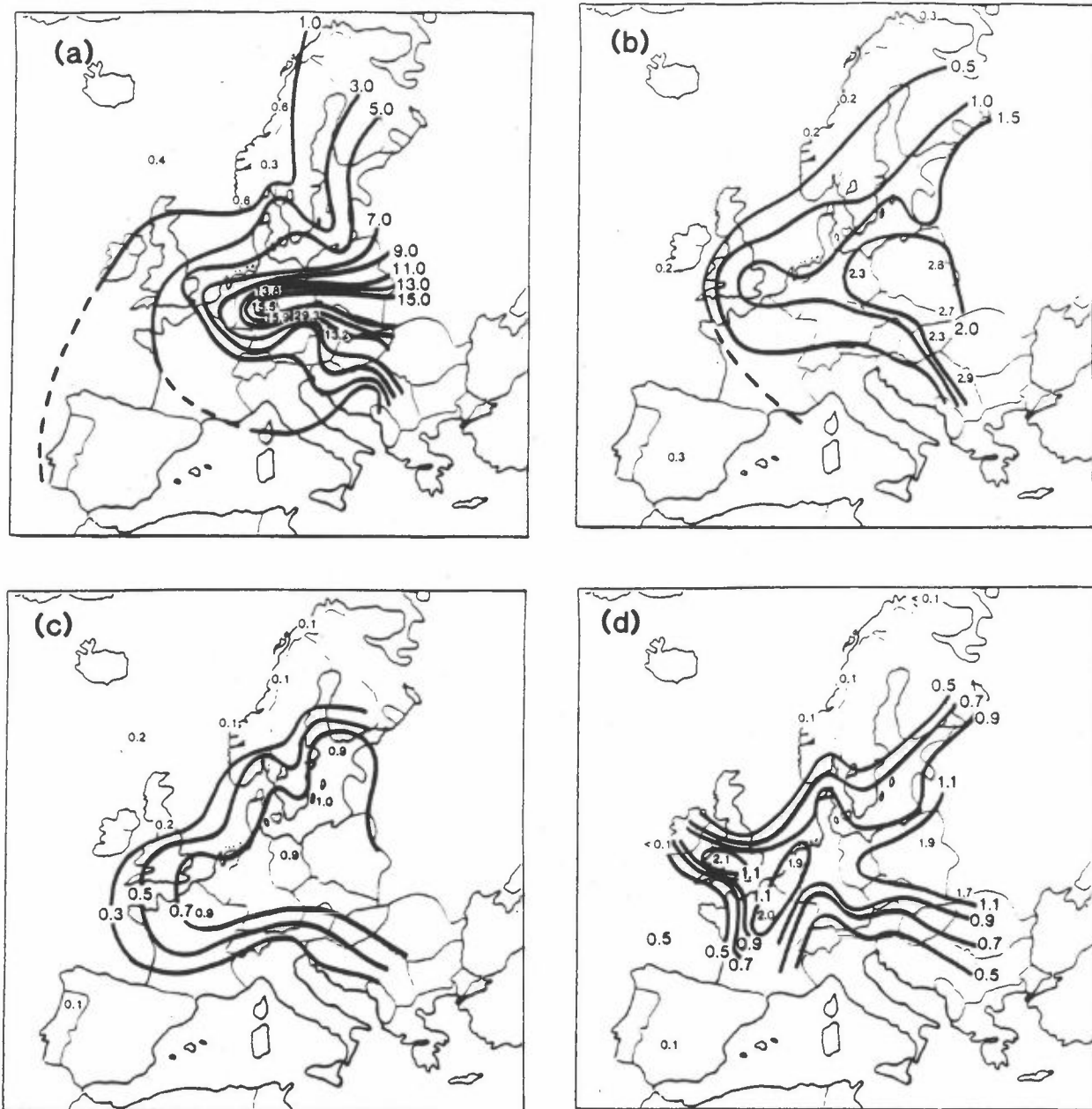


Figure 1: Annual mean concentration of sulphur dioxide in $\mu\text{g S/m}^3$ for 1985 (a), for excess sulphate in precipitation in mg S/l (b), nitrate in precipitation in mg N/l (c) and ammonium in precipitation in mg N/l (d).

Aerosol measurements at American Samoa were reported by Arimoto et al. (1987), while Dulac et al. (1987) estimated atmospheric input of trace metals to the western Mediterranean sea. Barrie et al. (1987) reported measurements of trace metals (Cd, Cr, Pb, Ni, V and Zn) in precipitation at remote locations.

The chemistry and physics of a winter stratus cloud layer were described by Daum et al. (1987). Gardner et al. (1987) measured the mass accommodation coefficient of SO_2 on water droplets and found that SO_2 transport into clean aqueous cloud and fog droplets will not be limited by interfacial mass transport.

Pyruvic acid (CH_3COCOOH) is formed through isoprene oxidation and is found predominantly as vapor, 10-20% in rainwater (up to about $0.1 \mu\text{g}/\text{l}$ measured in Brasil by Andreae et al., 1987).

Throughfall chemistry beneath sitka spruce was measured by Stevens (1987).

Galloway et al. (1987) compared the composition of acid rain in China, the US and at Katherine, Australia. Wet deposition rates of sulphur in China are 7-130 times those at Katherine, Eastern US somewhat in between.

Sulphate is the most important contributor to non-marine acid deposition in Nova Scotia, Canada, on the average 2-3 times the nitrate deposition on an equivalent basis (Underwood et al., 1987).

Hasselrot and Grennfelt (1987) studied the atmospheric deposition of air pollutants at a forest edge and at different parallel lines in the forest behind the edge. The ratios in throughfall flows between the edge and the line 50 m into the forest were 1.5 for SO_4^{2-} , 2.9 for NO_3^- , 2.7 for NH_4^+ and 3.1 for Na^+ . These results can also be generalized to hillsides, hilltops, edges between stands of different age, etc.

The composition of precipitation has been measured in a Belgian spruce forest under trees and in clearings (Coenen et al., 1987). An enrichment of acidic species and trace metals was found under the trees, mainly due to the washout of pollutants deposited on foliage surfaces in dry periods.

Cadle et al. (1987) found that the first 50% of the snowpack acidity was released in meltwater and rainwater equal to 25% of the original snowpack water content. The snowmelt and runoff were studied the winters 1982-1984 at the north tip of Michigan's lower peninsula.

Buijsman and Erisman (1987) estimated on the basis of ammonium concentration data in precipitation at 218 European measuring sites that about 2.6 Mt NH_4^+ /a is wet deposited. The estimate depends on a scarce data set in particular in the southern and eastern part of the continent and over adjacent seas (e.g. the North Sea).

Gaffney et al. (1987) questioned an uncritical focus on sulphuric acid, pH and nitric acid when acid rain effects are studied. Photochemical oxidant and acid species formation are closely interlinked, and precipitation may contain hundreds of organic compounds. Oxalate, phenols, nitrated phenols and formaldehyde-sulphur dioxide adducts are stable organic compounds found in precipitation in concentrations ranging from a few nanomol to several micromol/l, and the environmental impact of these and other trace species in rain needs to be assessed.

Rain chemistry was measured in August 1983 at Allegheny Mountain in southwestern Pennsylvania (Pierson et al., 1987). A mole ratio $\text{SO}_4^{=}/\text{NO}_3^-$ of 1.8 was found. An acid deposition budget was calculated: 47%, H_2SO_4 in rain; 23%, SO_2 dry deposition without dew; 16%, HNO_3 in rain; 11%, HNO_3 dry deposition without dew; 2%, HNO_3 and H_2SO_4 in fog and dew; 0.5%, aerosol dry deposition without dew.

SO_2 is dissolved in rime ice, and the fraction of SO_2 entrapped by the riming process is proportional to the drop supercooling (Lamb and Blumenstein, 1987). Riming is common in mountainous terrain in winter storms and can lead to significant deposition of water mass between

precipitation periods. Growth of supercooled cloud water droplets through accretion is also a primary growth mechanism of precipitation in mid-latitude storms.

Dry deposition of nitrate, ammonium and sulphate to a forest canopy and to surrogate surfaces in California (nylon filters, Petri dishes) was measured by Bytnerowicz et al. (1987b). They found good correlation of the deposition flux of NO_3^- and NH_4^+ to the foliage and to the nylon filters, but not to the Petri dishes. For sulphate the correlation was better between the flux to the foliage and to the Petri dishes than to the nylon filter.

Annual wet deposition of H^+ , NO_3^- and SO_4^{2-} in the southern part of Sierra Nevada in the U.S. was estimated to be 0.045, 3.6 and 3.9 $\text{kg ha}^{-1} \text{a}^{-1}$, respectively, based on several years' data (Stohlgren and Parsons, 1987).

The Sudbury smelters in Ontario, Canada, emitted about 900.000 t SO_2/a in the early 1980s. Still the smelters' emissions contributed little to the wet deposition of sulphate in Ontario (< 12%), and perhaps 20-30% to the dry deposition of sulphur (Tang et al., 1987a).

Primarily organic acids were found in precipitation from Katherine which is a remote, terrestrial site in northern Australia (14°S) (Likens et al., 1987).

The size distribution of insoluble particulate matter in rainwater over a rural area in the Federal Republic of Germany was studied by Schütz and Krämer (1987). They found that for particles with $r > 0.5 \mu\text{m}$, the shape of the number size distribution followed that of average urban and rural aerosol, while it is flattened out for $r < 0.5 \mu\text{m}$.

Data from 34 sites in three networks in Eastern US spanning 1978-1983 were combined into a uniform archive of daily precipitation amounts and ion concentrations. SO_4^{2-} , NO_3^- and NH_4^+ were most closely related to acidity; at six representative sites, empirical linear equations using those ions and Ca, Mg, Na, Cl and K concentrations explained about 95% of the variability in H^+ . Spatial and temporal

patterns were statistically interpolated through Kriging of yearly data; highest concentrations of H^+ , SO_4^{2-} and NO_3^- occurred along the Ohio River valley northeastward to New England, and maximum NH_4^+ concentrations from the northern Great Plains eastward across the Great Lakes. A small trend towards decreasing acidity and concentrations (but not amount deposited) was observed, but more data are required to determine if the trend is continuing (Endlich and Eynon, 1986).

In EPRI (1987) is summarized results from the Utility Acid Precipitation Study Program (UAPSP) which extended an early study of the Electric Power Research Institute. UAPSP operates 21 monitoring sites in the eastern and central United States and will continue in modified and expanded form as the Operational Evaluation Network to be run by EPRI. A cursory analysis of data from 1979 through 1985 shows remarkable constancy in annual wet deposition through the network and its subregions; wet deposition of sulphate and nitrate has been virtually constant in the Midwest, the Northeast and the Southeast, though there are subregional differences.

The UK Department of Environment (1987) review on acid deposition found that in the UK, acid deposition declined by 25% between 1981 and 1985, compared to the period 1978 to 1980. Although sulphur in acid rain declined, the levels of nitrate remained nearly constant. The highest levels of acidity were found in Cumbria and the Scottish Highlands.

Atmospheric long-range transport of heavy metals to the terrestrial environment in Norway was evaluated by Steinnes (1987).

Measurements at rural sites in Norway in 1986 were reported by Joranger et al. (1987). The highest SO_2 concentrations were measured in the southeast of Norway and in the north close to the Russian border, where influence from Nickel in USSR is likely. Sulphate, NO_2 , sum nitrate and ammonium have a north-south gradient, declining northwards. SO_2 did not change much from 1985 to 1986 while an increase was noted for NO_2 . Sulphate has dropped by about 30% since the end of the 1970s.

Nazarov et al. (1986) describe techniques used and results obtained from a series of monitoring stations along the western border of the USSR, including deposition to snow.

Spatial correlation patterns of SO_4^{2-} in snow and meltwater showed correlations for station spacing and were discussed by Belikova et al. (1986).

Inhomogeneities in snow pollution were found at two scales, 1000 km and 100 km; implications for station spacing were discussed by Vasilenko et al. (1986a).

Up to 25% of the SO_4^{2-} in spring runoff is from polluted snow; one third of that is estimated to be transported from western Europe to the USSR (Breslav et al., 1986).

SO_4^{2-} , SO_3^{2-} and S^{2-} concentrations in fresh precipitation and during storage of samples were described by Lavrinenko (1986).

An automated modelling system based on pollutant trajectories and deposition from generalized source regions, incorporating aircraft and ground-based measurements was described by Pressman et al. (1986).

The Eulerian model of the GDR predicts peak concentrations, daily and long-term average concentrations and transboundary flows of SO_2 and SO_4^{2-} (Discher et al., 1986a).

Discher et al. (1986b) compare the performance of an Eulerian and a Lagrangian multilevel transport, transformation and deposition model.

Vasilenko et al. (1986b) assessed the spatial dependence of trace metal and SO_4^{2-} snow cover pollutant measurements on point and large-area sources in the USSR.

6 ANALYTICAL TECHNIQUES

The measurement of atmospheric hydrogen peroxide is important due to its involvement in S(IV) to S(VI) oxidation. Sakugawa and Kaplan (1987) compared a cold trap method with an impinger bubbling method for the measurement of H_2O_2 and found that the impinger bubbling method always gave much higher gas concentrations (0.06-3.7 ppb) than the cold trap method (0.02-1.2 ppb), and attributed this to aqueous phase generation of peroxides on the impinger.

The enzyme fluorometric and the peroxyoxalate chemiluminescence techniques for analyzing hydrogen peroxide (H_2O_2) were compared in laboratory studies and under field conditions during the fall of 1985, and the methods agreed very well (Beltz et al., 1987).

Analytical techniques to measure biogenic sulphur emissions were discussed by MacTaggart et al. (1987).

Hartkamp and Bachhausen (1987) described a spectrophotometric determination of atmospheric H_2O_2 with a 1 ppb detection limit.

Calibration gas standards for H_2S , CH_3SH , CS_2 and SO_2 were independently prepared by two US groups and compared, and the differences found to be <12% for CH_3SH , CS_2 and SO_2 and <21% for H_2S (Farwell et al., 1987).

An analytical technique to determine hydroxymethanesulfonate in wet deposition samples was described by Ang et al. (1987).

The spatial distribution of the deposition of compounds derived from NOx emissions is quite dependent on the split between nitric acid gas and nitrate aerosols, since the dry removal is very different. Measuring techniques in routine use are able to determine the sum of nitrate and nitric acid, while the separation is more difficult to obtain. Durham et al. (1987) reported nylon denuder measurements of nitric acid and nitrate, and identified a number of problems indicating that such an instrument is not ready for monitoring networks.

Tunable diode laser absorption spectrometry (TDLAS) was described and used by Harris et al. (1987) to measure NO_2 and HNO_3 in diesel exhaust gas.

Two procedures for the calibration of an ECD (electron capture detector) for PAN were discussed by Meyrahn et al. (1987). The calibration of the ECD is a difficult part of the measurement of PAN, and standardized methods are needed.

Roberts et al. (1987) evaluated the tungsten oxide denuder tube technique for the measurement of nitric acid. Nitric acid levels measured by this technique averaged a factor of 3 higher than levels measured by a nylon filter technique.

Instrumentation and intercomparison of instrumentation for the measurement of CO, NO and OH and used during the NASA Global Tropospheric Experiment (GTE) around 1985, were reported by Beck et al. (1987).

A method to analyse for C_2 - C_6 hydrocarbons was described by Leuenberger et al. (1987).

Atmospheric acidity is linked to a range of trace gases and aerosols, and more and more analytical techniques become available for the simultaneous determination of a sequence of species at low concentrations. One such techniques is matrix isolation followed by Fourier transform infrared spectroscopy (FTIR), used to measure N_2O , CFCl_3 , CF_2Cl_2 , OCS , CS_2 , SO_2 and PAN in the Federal Republic of Germany and from aircraft (Griffith and Schuster, 1987).

The pressure drop in aerosol samplers can lead to evaporation of the collected particles. This is a serious problem in the measurement of nitric acid, nitrate, ammonium and ammonia using filter methods. A theoretical analysis was reported by Zhang and McMurry (1987).

Microscopic and diffraction methods for a qualitative identification of atmospheric aerosol particles, were described by Zwodziazk and Koniec (1987).

Results from the ninth intercomparison of analytical methods within EMEP were reported by Hanssen and Ladegård (1987).

The efficiency of Whatman 41 filterpaper as an air sampling medium was discussed intensively in Atmospheric Environment, and it was generally agreed that the filter is unsuitable for low-volume sampling but suitable for high-volume sampling (Watts et al., 1987b; Lowenthal and Rahn, 1987; Harrison, 1987; Storr and Baker, 1987).

Various sources of errors in daily and cumulative precipitation chemistry sampling were analysed by Tang et al. (1987b) for a network in Ontario, Canada. Both daily and 28 d sampling had good reproducibility, while automatic wet-only collectors may be a major source of error due to unrepresentative sampling.

A wet-only rainfall collector was described by Bridgman (1987), designed for non-freezing conditions.

Fog can in places be an important source of acid deposition. Hering et al. (1987) compared 5 types of fogwater collectors in a field intercomparison. All samples agreed for fogwater pH. Four of the samplers showed reasonable agreement for analyte concentrations. Only three of the samplers showed any agreement for liquid water content.

A wet-only an sequential fraction rain collector based on the Atmospheric Environment Service standard Canadian rain gauge, was described by Vermette and Drake (1987).

Two new ground-level water sampler designs were described by Daube et al. (1987), one active cloud water collector and one passive, collecting cloud droplets by inertial impaction on Teflon strands.

An automatic rain gauge for continuous, real time determination of rainwater chemistry has been developed at CERL in the UK (Ames et al., 1987).

7 EMISSIONS

The fraction of NO_2 in the NO_x emissions is high ($> 30\%$) for gasoline cars with air injection, a system which is in increasing use for passenger cars to meet legal restrictions on vehicle emissions of CO and hydrocarbons. Diesel vehicles also have $> 30\%$ NO_2 in their NO_x emissions, while gasoline car with 3-way catalyst had low NO_x total and $< 1\%$ NO_2 (Lenner, 1987).

Volatile organic compounds (VOC) emissions from 46 in-use passenger cars were reported by Sigsby et al. (1987). Both exhaust emissions and evaporative emissions were determined and the relative content of individual hydrocarbons specified.

An overview of the impact of motor vehicles on UK air quality was published by Williams (1987).

Isotopic ratios of lead can be used to trace the origin of long range transported air pollution, because the lead isotope ratio is specific for the origin of the ore used (Sturges and Barrie, 1987).

Middleton (1987) compared two emission inventories for the US and Canada and found that the differences in the Eastern US daily emissions were of the order of 5% for SO_2 and NO_x , 20% for VOC and 85% for NH_3 .

Carlson et al. (1987) published information about the most important emissions inventories for sulphur dioxide emissions from point sources in the Eastern United States and Southeastern Canada for 1978 and 1980.

Between 1975-1984 there is a statistically downward trend in the SO_2 emissions in 32 of the United States, the decline particularly in the Eastern and Western States (Lins, 1987).

Ammonia emissions for the UK and the rest of Europe were reported by ApSimon et al. (1987). They believed the European NH_3 emissions to have increased by 50% between 1950 and 1980. Buijsman et al. (1987) published the first gridded ammonia emission survey for Europe, specified on the EMEP grid.

European ammonia emissions were reviewed by Buijsman (1987), concluding that 6.4 Mt/a of NH_3 was emitted in 1985 with 81% from livestock wastes. The uncertainties in ammonia emission factors can have led to a 25-35% underestimation of the total emissions.

Ammonia emissions at a cement production plant were reported by Cheney and Knapp (1987).

Ferm and Christensen (1987) measured that during the first 24 h after spreading of cattle slurry to a grass ley, 41% (22.3 kg NH_3 -N/ha) of the applied NH_3 was lost, during the succeeding week 12% (6.6 kg NH_3 -N/ha) was lost. Ammonia emissions from livestock operations in the Netherlands were described by Klarenbeck (1987).

Nitric oxide (NO) is related to photochemical production of HNO_3 and the OH radical in the troposphere, and is released directly, or can be produced by nitrous oxide (N_2O) reactions. Anderson and Levine (1987) examined seasonal and diurnal emissions of NO and N_2O from agricultural sites in Virginia and Colorado as a function of soil conditions and fertilizer use. In Virginia, 2.4% of the annual NO flux occurred from November to April. Soil moisture had a large effect on emissions at both sites. NO is produced mainly by nitrification in aerobic soils, while N_2O is formed by denitrification in anaerobic soils.

Kavanaugh (1987) estimated future CO, N_2O and NOx emissions from energy combustion on a global and regional basis. It is believed that future global CO emissions from combustion will decrease because of regulation in the developed countries. NOx combustion emissions are estimated at 10.3 Tg N/a in 1960, 17.3 in 1975, 30.3-31.4 in 2000 and 45.5-47.4 in 2025.

Night-time measurements of NO at a rural site in the Federal Republic of Germany showed that there is a biological source which is increasing with temperature. During summer, the morning peak of biological NO may override the NO peak found around noon due to NO₂ photodissociation (Helas et al., 1987).

Isotope ratios ¹⁵N/¹⁴N of nitrate and ammonium in rain can provide information about precursor sources because NOx derived from natural, biological sources have a lower ¹⁵N/¹⁴N ratio than NOx derived from pollutant combustion sources. Isotope data can also provide information about the importance of lightning as a source for NOx (Heaton, 1987).

Emission of nitrogen oxides from a flooded soil fertilized with urea, has been measured by Galbally et al. (1987). The flux of NOx from the fertilized area was high when the nitrite and nitrate content of the floodwater was high. At night, ambient NO₂ was absorbed by the floodwater. Of the urea applied, 30% was lost to the atmosphere by NH₃ volatilization (as N), 15% by denitrification (as N₂), and the remainder except for minor losses of NO and N₂O, remained in the plant/soil/water system.

NOx production through lightning is estimated at 7 Mt N/a globally by Chameides et al. (1987). NO emissions from soils were estimated to be on the average 3 ng N/m² x s at a grassland site in Colorado August-November 1985 (Williams et al., 1987).

Observations of sulphate in snow from several Antarctic locations reveal four major volcanic events of the past two centuries (Agung, Krakatoa, Tambora and another which went unrecorded) (Legrand and Delmas, 1987).

One of the channels of the sun-synchronous meteorological satellites NOAA 6-9 is very sensitive to radiation from bodies in the temperature range 500-1500 K, and can detect flare plumes, ship trails, straw burning and other hot spots (Scorer, 1987).

The major source of cloud-condensation nuclei (CCN) over the oceans appears to be dimethylsulphide, produced by algae in sea water and oxidized to sulphate in the atmosphere. The Earth's albedo is sensitive to the amount of CCN present (Charlson et al., 1987).

Methanesulphonic acid (CH_3SO_3^- , MSA) is an unequivocal indicator of marine biogenic activity, and its presence in ice can be used to reconstruct the history of marine biogenic activity (Saigne and Legrand, 1987).

Biogenic H_2S emissions from Florida wetland surfaces were measured by Castro and Dierberg (1987) and found to be insignificant contributors to the sulphur burden in the atmosphere over Florida.

Utilizing isotopic signatures, Nriagu et al. (1987) found that dimethyl sulphide from boreal wetlands may account for up to 30% of the acidifying sulphur burden in the atmosphere in remote areas of Canada.

Biogenic sulphur emissions from the subantarctic and Antarctic oceans consist mainly of DMS. Atmospheric DMS concentrations of about 100 ppt, MeSH 3.6 pptv, aerosol MSA at 0.27 n mol/m^3 , non-seasalt SO_4^- at 0.31 n mol/m^3 and SO_2 at 11 pptv were reported by Berresheim (1987).

Natural sulphur emissions measured by gas chromatography using enclosure techniques and reported by a NOAA-group (Goldan et al., 1987) were lower than found in earlier, independent investigations. It was concluded that sulphur fluxes from soil make a negligible contribution to the atmospheric sulphur burden compared to anthropogenic SO_2 , and the contribution from salt water marshes is much smaller than previously thought. Additional measurements of biogenic sulphur emissions from soils and vegetation in the United States were reported by Lamb et al. (1987).

The short-term variability in biogenic sulphur emissions from a Florida marsh was studied by Cooper et al. (1987a, b) who found that the flux of H_2S varied over 4 orders of magnitude, correlated primarily with the stage of the tidal cycle. The fluxes of dimethyl sulphide, carbondisulphide and dimethyl disulphide varied by less than one order of magnitude and depended mainly on temperature.

Toon et al. (1987) published a paper on the sulphur cycle in the marine atmosphere.

8 MODELS AND INTERPRETATION OF MEASUREMENTS

A three-dimensional Eulerian regional acid deposition model to calculate episodic chemical concentrations and dry and wet deposition of acids in North America was described by Chang et al. (1987). This transport, transformation, and deposition modelling system subdivides the troposphere over the Eastern United States, Southeastern Canada, and the Western Atlantic Ocean into a six-level, 30 by 30 horizontal grid with a horizontal grid size of $80 \times 80 \text{ km}^2$. Transport and vertical diffusion of 24 trace gases and particles are calculated using temporally and spatially varying meteorology, provided by a mesoscale meteorological model.

A gas phase chemical reaction mechanism is used to simulate concentrations and chemical conversion rates for 36 species, including 14 stable organics and 11 short-lived radicals. Altitude-, latitude-, and season-dependent photolysis rates for nine reactions in the chemical mechanism are specified using a delta-Eddington radiative transfer model which includes O_2 and O_3 absorption, scattering and absorption by clouds and aerosols, Rayleigh scattering, and ground reflections. Subgrid scale vertical transport, aqueous chemical conversions, and trace gas and particle scavenging by clouds are parameterized using a one-dimensional dynamical and microphysical cloud model and a box aqueous chemistry and scavenging submodel.

The aqueous phase chemistry model includes sulphur oxidation by H_2O_2 , O_3 , trace metals, and two organic peroxides, with numerous equilibria

between all soluble trace species. Dry deposition rates for 13 compounds are computed using species-specific deposition velocities that depend on the local meteorology, season, land type, insolation, and surface wetness conditions. Emissions of SO_2 , SO_4^- , NO , NO_2 , CO , NH_3 , and 10 classes of volatile organic compounds are included in the model. Trace gases are emitted into different vertical levels of the model according to a plume rise submodel. Hourly emissions are adjusted according to season and weekday or weekend activities. This model provides a framework to examine the relative importance and sensitivity of numerous physical and chemical processes responsible for the formation and deposition of tropospheric acidity.

Walcek and Chang (1987) applied the model to assess the acid deposition to individual land types during an acid deposition episode in Northeastern US.

The source-receptor relationship is difficult to quantify in Eulerian models, contrary to the situation in Lagrangian models where it is easily derived. Hsu and Chang (1987) have developed a new approach to delineate the source-receptor relationship in an Eulerian model, where a small, unique oscillatory signal is superimposed on each emission source. The calculated concentration time series is analysed by a Fourier transform to give the amplitude-frequency spectrum, and peaks in the spectrum and their height express the contribution from a specific source to a specific receptor site. The source-receptor relationship from nonlinear atmospheric models was also studied by Kleinman (1987).

Fay et al. (1987) made a source apportionment of nitrate wet deposition at several receptors and an estimation of the nitrogen budget for Eastern North America. The model parameters were optimized by matching the model output to observed deposition at 109 precipitation sampling stations.

Fekete and Szepesi (1987) described a first generation model that includes S and N source strengths and heights, mixed layer height, regional transport and transformation, and wet and dry deposition. The model simulates annual average regional distributions of SO_2 , NO_2 ,

gaseous HNO_3 , and particulate and deposited SO_4^{2-} and NO_3^- , and is validated using 5 years of data from the Regional Background Pollution Station in Hungary.

Asman and Janssen (1987) described the choice of transformation and removal parameters in a receptor-oriented Lagrangian model for ammonia and ammonium in Europe. Derwent (1987a) presented results of a similar model for northwest Europe. Asman and Janssen (1987) found that ground-level concentrations of NH_3 are determined to a large extent by local sources, making difficult the comparison with calculated values in a long-range transport model of the EMEP-model type. Alcamo and Bartnicki (1987) presented a sensitivity analysis of the EMEP-model for sulphur, and showed i.e. that a 20% input change in transfer coefficients throughout Europe resulted in 15-22% change in total sulphur deposition.

A model for the calculation of the distribution and deposition of species derived from emissions of NOx in Europe has been developed in the framework of EMEP. Meteorological data from the numerical weather forecast model at the Norwegian Meteorological Institute in Oslo were used to calculate dry deposition, trajectories, wet removal, photolysis rates etc. Emissions of SO_2 , NOx and NH_3 were taken into account to calculate both airborne and deposited amounts of NO, NO_2 , PAN, HNO_3 , nitrate, NH_4NO_3 , NH_3 , SO_2 , sulphate and ammoniumsulphate, and the allocation to emitting countries (Hov et al., 1987a).

Levy and Moxim (1987) calculated that of 7.5 Tg N of combustion nitrogen emitted annually in the US and Canada, 2.1-3.5 Tg N is dry deposited and 1.5-2.3 Tg N wet deposited over the same continent. At most 0.2 Tg N/a reaches Europe.

Atmospheric ammonia was calculated to be significantly depleted in a convective storm event (Laulainen et al., 1987).

Fisher (1987) applied his statistical long-range transport model previously used to calculate long-term wet deposition of sulphur over Europe with some success, on ammonia. It was difficult to explain observed long-term ammonium wet deposition, and he speculated that the

emissions of ammonia could be underestimated, or ammonia does not behave like a reactive atmospheric gas.

Clark et al. (1987) found that rainfall data for sulphate and its relationship to changing SO_2 emissions cannot be properly assessed without a transport model.

The Regional acidification information and simulation (RAINS) model is an integrated model of acidification in Europe designed as a tool for evaluating control strategies. The model was described by Alcamo et al. (1987b).

The measurement of the composition of orographic cloud droplets and rain at the Great Dun Fell mountain site in Cumbria, UK, has added to the understanding of liquid phase oxidation processes and precipitation chemistry. Hill et al. (1987) published a model of sulphate deposition through washout and turbulence onto a hill like Great Dun Fell. Hough (1987) published a detailed model study of the chemistry in clouds over hills.

A reactive plume model was evaluated with power plant plume data from France (Joos et al., 1987).

Model calculations, together with the measurement of nitrate in dew at a rural site in southwest Pennsylvania, indicate that nitric acid is the main source of nitrate in dew. NO_3 and N_2O_5 deposition can also significantly contribute to dew nitrate at night, in particular at high NO_2 (Chang et al., 1987).

The substances related to acid deposition and photochemical smog are fairly short-lived as long as they are confined to the lower troposphere (below about 1-kilometer altitude). If, however, the contaminants are rapidly transported to the upper troposphere, then their atmospheric residence times grow and their range of influence expands dramatically. Although this vertical transport ameliorates some of the effects of acid rain by diluting atmospheric acids, it exacerbates global tropospheric ozone production by redistributing the necessary nitrogen catalysts. Results of recent computer simulations suggest

that thunderstorms are one means of rapid vertical transport. To test this hypothesis, several research aircraft near a midwestern US thunderstorm measured carbon monoxide, hydrocarbons, ozone, and reactive nitrogen compounds. Their concentrations were much greater in the outflow region of the storm, up to 11 kilometers in altitude, than in surrounding air. Trace gas measurements can thus be used to track the motion of air in and around a cloud. Thunderstorms may transform local air pollution problems into regional or global atmospheric chemistry problems (Dickerson et al., 1987).

Altschuller (1987c) found that multiday regional scale horizontal transport with a gradual transport of sulphate down to the surface would be consistent with cloud and ground based measurements in St. Louis, Missouri.

Popovics et al. (1987) did not find a parallel variation in SO_2 emissions and sulphate in rain when analysing 25 years' data from three European networks (EACN, OECD and EMEP). The lack of correlation was explained in parts by analytical and sampling procedure differences, and in parts by a shift from low level SO_2 sources to tall stacks.

A targetted emission reduction in Europe was discussed by Shaw (1987). Through an optimized emission reduction with attention being paid to particularly sensitive receptors, large savings can be done compared to overall cuts by fixed percentages.

Chagnon (1987) used measurements of haze, smoke, dust and visibility in Midwestern US from 1901 to 1980 to show sizable temporal fluctuations, and that the measured parameters are useful as surrogates of air pollutants in times when no direct measurements were possible. There was a general decrease in visibility from 1950 to 1970, with some improvement in very large cities after 1970.

Precipitation chemistry has been measured at Hubbard Experimental Forest in New Hampshire in the US since 1963. It is now reported a decrease in precipitation acidity ascribed to decreases in SO_2 emissions in upwind source regions (Hedin et al., 1987).

A review of atmospheric chemistry, transport and deposition of nitrogen oxides was given by Grennfelt et al. (1987b) and of models for photochemical processes by Hov (1987a).

Pilinis and Seinfeld (1987) described a model that predicts the total quantities of ammonium, chloride, nitrate and water contained in atmospheric aerosols, their physical state and their distribution among aerosol particles of different sizes. The model was based on the thermodynamic equilibrium calculation of the ammonium/chloride/nitrate/sodium/sulphate/water system. Observed aerosol conditions in California (Long Beach) were successfully predicted.

Background levels of air and precipitation quality in Europe were reviewed by Szepesi and Fekete (1987). They concluded that 69% of SO_2 and 63% of NO_2 over Europe originate from continental anthropogenic sources, 15% of sulphate and 11% of nitrate in rain over Europe are due to hemispheric background, and hemispheric background values for Europe were found to be $1.25 \mu\text{g S/m}^3$ of SO_2 , $0.80 \mu\text{g S/m}^3$ of sulphate aerosol, 0.157 mg S/l of SO_4^{2-} in precipitation and 0.04 mg N/l of NO_3^- .

Aircraft measurements of SO_2 along the US East Coast were used to estimate that the eastbound flux of SO_2 is 1 Tg (S)/a (Luria et al., 1987).

Model calculations of the episodic increase of ozone in the boundary layer over Europe were reported by Hov (1987b, c). Emissions of volatile organic compounds were found to control the episodic ozone increase.

A modelling study of atmospheric transport and boundary layer photochemistry during anticyclonic episodes in Europe was published by van Dop et al. (1987) and Selby (1987).

Global tropospheric model calculations of trends in composition as the emissions of CH_4 , NO_x and CO go up show that OH and O_3 increase with increasing global NO_x emissions, while the increase in CH_4 tends to suppress OH while ozone goes up (Isaksen and Hov, 1987).

A method to assess uncertainty in atmospheric models was described by Derwent (1987b). On the basis of the probability distribution of the individual input parameters, latin hypercube sampling can be used to calculate the probability distribution of modelled species in a very efficient way.

A method to estimate uncertainties in the calculation of ozone and other air pollutants, was published by Derwent and Hov (1987).

The use of wind roses in a statistical long-range transport model was described by Shipley et al. (1987).

Turbulent concentration fluctuations are substantial in the near-source regions of pollutant emissions, and this affects the estimation of the rate of fast, second-order atmospheric reactions, where the use of space and time averaged quantities gives a substantially different rate estimate in the plume close to the source compared to a full calculation taking into account fluctuation terms (Karamchandami and Peters, 1987).

Lamb and Hati (1987) developed a method for generating ensembles of wind fields for use in regional scale pollution models. In this way an explicit treatment of the uncertainty inherent in the specification of the atmospheric state, is possible.

The use of the Kriging method to predict mean ozone concentrations was reported by Lefohn et al. (1987). Kriging is a statistical method which at its simplest can be thought of as a way to interpolate spatial data.

A model was developed by Pilinis et al. (1987) to simulate the dynamics of multicomponent atmospheric aerosols, including formation of particles by nucleation, gas-to-particle conversion, coagulation and dry deposition. Both equilibrium and non-equilibrium aspects involving sulphate, nitrate and ammonium compounds were considered, and model predictions were compared with measurements in Los Angeles.

A combined 3-dimensional mesoscale numerical model and a model for microphysics and gas and aerosol removal by cloud droplets and raindrops, was described by Chaumerliac et al. (1987) and applied for both continental and maritime cloud simulation.

Receptor modelling techniques have been used to quantify the sources of urban aerosols, while their success has been questioned in long-range transport situations. Thurston and Lioy (1987) recommend a specific approach using both chemical mass balance (CMB) and multivariate receptor oriented models (e.g. principal component analysis) to apportion transported aerosols.

The relationship between sources and receptors in acid rain was reviewed in White (1987).

Elemental signatures in air samples in the Northeastern US were evaluated and compositions characteristic of different source regions arrived at (Dutkiewicz et al., 1987)

A tracer experiment design to follow reactive pollutant emissions over North America during long range transport and deposition, was described by Hidy (1987). Conversion and loss of nitrogen and sulphur oxides would be calculated using isotopic S- and N-compounds together with inert perfluorocarbons.

Data from the perfluorocarbon tracer experiment (CAPTEX '83) across the Appalachian Mountains were used by Haagenson et al. (1987) to show that the wind flow corresponding to the low to middle boundary layer is the most appropriate for the simulation of pollutant transport in the boundary layer. Isentropic and isosigma transport assumptions are more realistic than the isobaric assumption.

Valenta and Nguyen (1987) attempted to establish long-term trends in the wet deposition of acids and toxic heavy metals (Cd, Pb, Cu, Zn) on a regional and local scale in the FRG for 1980-1984. Free acidity in rain and snow is apparently uniformly distributed in all regions as a result of the medium range transport of the acid precursors. Heavy metal deposition and concentrations in polluted regions show a decreasing trend since 1986.

A review of the chemistry and transport of reactive nitrogen in the troposphere was given by Singh (1987), while the aquatic chemistry of acid deposition was reviewed by Stumm et al. (1987).

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REFERAT (maks. 300 anslag, 7 linjer)			

TITLE
ABSTRACT (max. 300 characters, 7 lines) More than two hundred references from the open and the grey literature in 1987 about emission, atmospheric transport, transformation and deposition of acidic trace species have been compiled and reviewed.

* Kategorier: Åpen - kan bestilles fra NILU A
 Må bestilles gjennom oppdragsgiver B
 Kan ikke utleveres C