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REVIEW OF PAPERS PUBLISHED IN 1985 ABOUT EMISSION, TRANSPORT, TRANSFORMATION AND DEPOSITION OF ATMOSPHERIC TRACE CONSTITUENTS OF IMPORTANCE FOR ACID DEPOSITION

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1 SUMMARY OF IMPORTANT SCIENTIFIC FINDINGS REPORTED IN 1985

1.1 MODELS FOR ACID DEPOSITION AND PHOTOCHEMICAL OXIDANTS

Three-dimensional grid models are being developed to describe episodes of elevated pollution lasting for a few days in the atmospheric boundary layer. This is being done for the United States and Canada at EPA and at NCAR in Boulder, for Canada and Europe through a project funded by FRG and Canada and for Europe through the so-called PHOXA-model by FRG and the Netherlands (Builtjes, 1985). Nothing is yet published in the open literature about the results from these models, but there is hope that over the next few years it will be better understood how emission control may affect acid deposition and the concentration of photochemical oxidants over Europe and North America.

The EMEP-model has been used to calculate the airborne transboundary sulphur pollution in Europe (Eliassen et al., 1985). The agreement between calculated and measured concentrations of sulphur species was reasonable when comparing 5-year averages, and in many cases monthly concentrations also compared well.

It has been shown using the extended EMEP-model with boundary layer chemistry that control of hydrocarbons was very efficient in reducing the ozone-level in southern Scandinavia in episodes with long-range transport. HC-control is much more efficient than a combined HC- and NOx-control or NOx-control alone. (Hov, 1985a). Changes in ozone influence the concentration of OH and H_2O_2 which are important for the formation of sulphate and nitrate in clean air masses.

It has been found that the concentration of hydroxyl (OH) in the atmosphere may have dropped by 25% from 1950 to 1980 due to the increase in methane and carbon monoxide in the same period (Levine et al., 1985). Changes in "clean

air" over the last 30 years may therefore have led to a substantial decline in the "clean air" transformation rate of SO_2 to sulphate and NO_2 to nitrate since OH directly in the gas-phase, and indirectly in the wet-phase due to its importance for H_2O_2 and other species acting catalytically, is rate-determining in the conversion process.

1.2 MODEL INPUT (EMISSIONS, CHEMISTRY, DEPOSITION, METEOROLOGY, MODEL FORMULATION)

In the last year or two there has been considerable progress in the understanding of wet phase chemistry and the role of radical-species to form H_2O_2 in the dry and wet phase (Giorgi and Chameides, 1985, McElroy, 1985, Seigneur and Saxena, 1985). Knowledge about liquid water content in different cloud types, cloud cover and changes, the changes in cloud water content with time and entrainment of clear air into clouds, is still quite fragmentary. The gas phase chemistry of species relevant to acid deposition is less controversial (Leone and Seinfeld, 1985, Calvert et al., 1985).

Emission inventories of NOx, SO_2, HC and NH_3 are being recorded in Europe and North America on fine grid systems (down to 25 km resolution in some cases). Nothing has been published in the open literature so far, except for an ammonia emission inventory for Europe on the EMEP grid (Buijsman et al., 1985).

Numbers for the emission of SO_2 outside Europe and North America have been published by Varhelyi (1985).

About deposition, a range of experimental investigations have been published which mainly have served to ascertain existing knowledge.

1.3 MODEL VALIDATION (MEASUREMENTS)

Model validation involves the use of measured concentrations of acid substances and their precursors, photochemical oxidants and their precursors on a regional scale, measurements of the composition of the global troposphere

and in the polar regions and detection of trends in the composition of the global troposphere.

There has in many ways been a break-through in model validation during 1985. Hydroxyl radical concentrations have been measured at sites in FRG with different levels of air pollution. Maximum observed hydroxyl concentrations ranged from less than $5x10^5$ to $6x10^6$ molecules/cm^3. The measurements for NOx concentrations above 2 ppb could be well described by model calculations using observed concentrations of ozone, NOx, hydrocarbons, aldehydes and other species as input, while for lower NOx concentrations, the model indicated that there may be yet unrecognized hydroxyl loss processes. This is the first time that it has been demonstrated through simultaneous measurements of both precursors and products that the concentration of hydroxyl which is measured is consistent with theory, at least in moderately polluted air (Hofzumahaus et al., 1985).

There has been a significant increase in the reporting of precursor- measurements, in particular of individual hydrocarbon concentrations. Measurements have been carried out both in regional air pollution over Europe and in the clean troposphere (Rudolph et al., 1984, Tille et al., 1985a, 1985b, Colbeck and Harrison, 1985a, Rudolph and Khedim, 1985 and Ehhalt et al., 1985).

The concentration of SO_2 at Norwegian rural sites is declining year by year, while sulphate in air is fairly unchanged (SFT, 1985).

Beilke et al. (1985) reported aircraft and groundbased measurements of transboundary fluxes of nitrogen- and sulphur-compounds across the boarder between FRG and GDR. On an annual basis, the net flux of S-compounds across the border is small, but on an episodic basis there are numerous serious pollution episodes with transport from the east.

Measurements aimed at understanding the relationship between forest damage and acid deposition were reported by Georgii (1985). It was found that in central parts of FRG, the frequency of fog and low-layer clouds increases with altitude in mountain areas. There was considerably higher sulphate-, nitrate- and chloride-concentration in fogwater than in rainwater.

Many important scientific findings about trends in atmospheric composition over the last years, decades and centuries have been reported in 1985. Rinsland et al. (1985) and Rinsland and Levine (1985) reported that methane has gone up by 1.1% per year in the period 1951-1981 and carbon monoxide by 2% per year over Europe in the period 1950-77. Air entrapped in ice in western Antarctica has nearly twice as high methane content today than 200 years ago (Stauffer et al., 1985). Measurements of sulphate and nitrate in firn samples from South Greenland show that both sulphate and nitrate increased by a factor of about 2 during the period 1895-1978 (Neftel et al., 1985). Firn core analysis has indicated that between 1956 and 1977 there was a 75% increase of Arctic air pollution associated with a marked increase in the emissions of SO_2 and NOx in Europe (Barrie et al., 1985).

There has been a near doubling in the concentration of ozone in clean air over Europe during the last 20-25 years (Attmannspacher, 1985).

1.4 EFFECTS AND POLICY QUESTIONS

A number of air pollution effect studies have been reported in 1985. Some of them are included in this chapter. Policy questions related to the costs, effectiveness and social and political impact of emission control, have been discussed in many papers (e.g. Kroppenstedt, 1985).

2 MODEL DEVELOPMENT

2.1 MODELS FOR AIR POLLUTION ON A REGIONAL SCALE, E.G. EUROPE

The EMEP-model has been used to calculate the airborne transboundary sulphur pollution in Europe, and compared with data covering a five-year period (Eliassen et al., 1985). The agreement between calculated and measured concentrations of sulphur species was reasonable when comparing 5-year averages, and in many cases monthly concentrations also compared well.

The extended EMEP-model with boundary layer chemistry has been applied to investigate how emission control of NOx and hydrocarbons may influence the

ozone-level in southern Scandinavia found in pollution episodes with long-range transport. It was concluded that HC-control is much more efficient than a combined HC- and NOx-control or NOx-control alone. NOx- control alone may sometimes lead to an increase in long-range transported ozone (Hov, 1985a)

The report by Fay et al. (1985) described the application of a long-term wet sulphate model for northeastern United States.

Haagenson et al. (1985) presented case studies of acid precipitation in cyclonic storms.

Methods for estimating fine particle and inhalable particle mass concentrations using relative humidity corrected light extinction coefficients from airport visual range observations, were presented in the paper by Øzkaynak et al. (1985).

A lagrangian trajectory model was presented by Saab et al. (1985) using 3-dimensional winds. The model was applied for long range transport of sulphur.

The chemical development of the constituents in a plume from a coal-fired power plant was calculated by Joos et al. (1985) taking into account both gas phase and aerosol- phase reactions.

The PHOXA-project was described by Builtjes (1985). An Eulerian 3-d model is being assembled to calculate the generation and transport of photochemical oxidants and acid substances over Europe.

A 3-d mesoscale model with microphysics has been applied by Chaumerliac (1985) to the study of the removal of gases and aerosols by cloud and raindrops.

ApSimon et al. (1985a) decribed the Lagrangian puff trajectory model MESOS, which is a long-range transport model designed to simulate transport and dispersal of radionuclides. In their second paper, ApSimon et al. (1985b) described an application of the MESOS-model.

Scire and Venkatram (1985) discussed in-cloud oxidation in relation to total wet scavenging of sulphate in convective clouds. In-cloud conversion is an important removal mechanism for SO_2 and accounts for a significant fraction of the precipitation sulphate.

Venkatram and Pleim (1985) found that when observations of annual wet deposition of sulphur were analysed using a statistical long-range transport model, the parameters that represent the conversion of SO_2 to $SO_4^=$ and the wet and dry removal of sulphur were insensitive to the concentration levels, the variation of the wet deposition field was closely related to the distribution of the sulphur emissions, and the observations demanded efficient wet scavenging of SO_2.

Calculations of SO_2-concentration and wet deposition of sulphur were reported by Ellenton et al. (1985) for eastern North America using a trajectory puff model. Annual wet deposition compared reasonably well with monitored values.

Kumar (1985) presented an Eulerian model for the scavenging of pollutants by raindrops.

Russell et al. (1985) used a mathematical model to study the fate of NOx emissions in the Los Angeles basin.

Ruff et al. (1985) compared three regional air quality models, one which represented the Eulerian class models (RTM-II), one source-oriented Lagrangian model (ENAMAP-2) and one receptor-oriented, backward trajectory model similar to the one used in EMEP (ACID). It was found that both for 6-h and annual average concentrations there was poor correlation between measured and calculated SO_2 and sulphate.

Renner et al. (1985) reported on the results of a multi-level Lagrangian atmospheric model for transport, transformation and dry and wet deposition of SO_2 and sulphate. No real situations were studied, only theoretical examples.

Misra et al. (1985) found that the scavenging ratios for SO_2, sulphate and nitrate vary considerably between precipitation events, and the introduction of constant values for scavenging ratios in models will lead to errors in the calculations.

It was shown by Vukovich et al. (1985) that if half of the ozone mass which accumulate over the United States in a high pressure situation during the summer was transported into the free troposphere, then the amount of ozone transported out of the boundary layer would approximate the amount of ozone transported downward during a tropopause fold event.

High oxidant situations in 1979 in central Japan were examined by Kurita et al. (1985) with regard to the transport mechanisms.

It was found by Seigneur and Saxena (1985) that for typical northeastern U.S. conditions, the formation of ozone was considerably reduced when calculating photochemistry in both the wet and the gas phase, compared to the dry case only. This was due to the high solubility of aldehydes which are major HO_2-radical sources in the gas phase, the scavenging of radicals by cloud droplets, and lower photolytic rates inside of clouds.

Model calculations of the effect of HC- and NOx-emission control during long-range transport of photochemical oxidants to the Netherlands, were presented by deLeeuw and van den Hout (1985). HC-emission reduction was the best way to reduce the peak values of ozone and other photochemical oxidants.

Other papers on regional models were published by Charlson and Kowalski (1985), Hanson et al. (1985), Bøhler and Isaksen (1984), Augustin and Bessemoulin (1984), Nasstrom et al., (1985), van Egmond and Kesseboom (1985) and Oppenheimer (1985).

2.2 MODELS FOR GLOBAL AIR POLLUTION

Model calculations were presented by Isaksen et al. (1985) of the global distribution of nonmethane hydrocarbons and the potential role that high

springtime concentrations of nonmethane hydrocarbons and PAN in the Arctic may play in the formation of ozone at mid and high latitude during spring.

2.3 MODELS FOR TRENDS IN THE COMPOSITION OF THE TROPOSPHERE AND OF CLIMATIC CHANGE

The paper by Ramanathan et al.(1985) examined the potential climatic effect of all known radiatively active trace gases that have been detected in the atmosphere including chlorofluorocarbons, chlorocarbons, hydrocarbons, fluorinated and brominated species, and other compounds of nitrogen and sulphur, in addition to CO_2 and O_3. A one-dimensional radiative-convective model was used to estimate trace gas effects on atmospheric and surface temperatures for three cases: (1) modern day (1980) observed concentrations were adopted, and their present trends were extrapolated 50 years into the future. These projections were based on analyses of observed trends and atmospheric residence times; (2) the preindustrial to present increase in CO_2 and other trace gases were inferred from available observations; (3) a hypothetical increase from zero to one ppbv was considered to provide insight into the radiative processes of gases not present naturally in the atmosphere. Trace gases other than CO_2 were shown to be potentially as important as CO_2 for long-term climate trends.

Model calculations by Levine et al. (1985) indicated a 25% decrease in the atmospheric concentration of hydroxyl in the 1950-80 time period due to the increase in the atmospheric concentration of methane and carbon monoxide.

3 MODEL INPUT

3.1 GAS PHASE CHEMISTRY

Leone and Seinfeld (1985) presented an evaluation of currently used reaction mechanisms in photochemical smog models. Also a comprehensive reaction scheme was given which serves as a standard. First day- generation of secondary products was discussed, and the results of the different mechanisms were usually within a factor of two.

The peroxyacetyl nitrate (PAN) forming potential of t-butene, propene, ethene, n-butane and toluene in single hydrocarbon/NO_2/synthetic air system was probed in smog chamber experiments by Glavas and Schurath (1985). Maximum yields, expressed in percent NO_2 converted to PAN, were 50% for t-butene, 33% for propene and 6% for toluene. No PAN-maxima were reached for n-butane or ethene after 5-6 h of irradiation.

Computer simulations can rationalize the observed seasonal trends in sulphates and nitrates. Recent tropospheric measurements of gaseous hydrogen peroxide show that this gas is a major oxidant leading to sulphuric acid generation in cloud water (Calvert et al., 1985).

At low NO-concentrations compared to propene, the oxidation of SO_2 to sulphate seemed to proceed through the reaction with hydroxyl. At high propene to NO-ratios, SO_2 seemed to react primarily with a ozone-propene adduct to form sulphate aerosols (Luria and Sharf, 1985).

HONO, NO_3 and N_2O_5 may play important roles in urban smog formation through nighttime formation and photolysis in early morning hours to produce OH (Killus and Whitten 1985).

The yield of SO_2 from the photo-oxidation of DMS ranged between 20 and 30% in a series of experiments reported by Hatakeyama et al. (1985).

Other papers on gas phase chemistry were published by Becker et al. (1985), Jolley and Forster (1985), Cole et al. (1985), Grosjean (1985), Shepson et al. (1985), Platt et al. (1985), Jourdain et al. (1984), Barnes et al. (1984), Lorenz et al.(1984), Schmidt et al. (1984), Bagnall and Sidebottom (1984), Moortgat and McQuigg (1984), Cox et al. (1984), Schurath and Goede (1984) and Burrows et al. (1984).

3.2 LIQUID PHASE CHEMISTRY

The reaction of aqueous hydroxyl radicals with bisulphite ion has been proposed as a significant and in some cases a primary pathway by which SO_2 is

oxidized to sulphate in tropospheric cloud. The calculations presented by McElroy (1985) showed that the reaction of the SO_5^- radical with bisulphite ion is rate determining, although the rate constant remains uncertain.

Gaseous SO_2 and aqueous peroxide were rarely simultaneously present in substantial concentrations; generally one reactant was present in great excess over the other, consistent with the occurrence of the aqueous phase reaction of peroxide with S(IV) in which one or the other species is the limiting reagent (Kelly et al. 1985).

In-cloud scavenging is a determining factor for precipitation sulphate, while it is relatively unimportant in the case of ammonium. The sub-cloud scavenging of NO_2 and SO_2 is not too significant. For HNO_3 and NH_3 it is an effective process (Meszaros and Szentimrei, 1985).

Benner et al. (1985) found H_2O_2 to be produced during incomplete combustion and led to the oxidation of SO_2 to sulphate when dissolved in water.

Other papers on liquid-phase chemistry were published by Giorgi and Chameides (1985), Chen et al. (1985), Goodman (1985), Graedel et al. (1985), Hegg (1985), Barrie (1985b), Ayers et al. (1985), Seigneur et al. (1985), Clarke et al. (1984), Raes and Janssens (1984), Winkler (1984), Ferm (1984), Rømer and Reijnders (1984), Liljestrand (1985), Overton (1985) and Marsh and McElroy (1985).

3.3 EMISSIONS OF SO_2, NOx, HC AND NH_3

In the paper by Varhelyi it was found that at present about 2.4, 4.1, 0.7 and 18.3 Tg(S)/y are emitted as SO_2 in Africa, South America, Oceania and Asia, respectively, with the greatest increase in the anthropogenic SO_2 emission during the last decade in Asia.

Jørgensen and Okholm-Hansen (1985) measured the flux of DMS, H_2S, COS, CH_3SH and CS_2 from Danish estuaries. DMS was the most important sulphur gas released from grass and algae, while mostly H_2S was released from intertidal mud flats.

A time series model for SO_2 stack emissions from coal boilers was presented by Gleit (1985).

About 230 t(SO_2)/d is emitted from Mount Erebus in Antarctica. The activity of the volcano is rather constant, and the emissions account for much of the sulphur observed in Antarctic snow (Rose et al., 1985).

Ammonia emissions were negligible from two pulverized-coal furnaces (Bauer and Andren, 1985).

The use of blended fuels did not have a significant effect on the emission of hydrocarbons, except for an increase in aldehyde emissions. Aldehyde emissions from a methanol-fueled car were roughly one order of magnitude higher than those resulting from blended fuel usage (Gabele et al., 1985).

Northern peatlands may be an important source of global atmospheric methane (Harriss et al., 1985).

Acetic and formic acids were the dominating organic acids in Los Angeles air (0.37-7.45 ppb), followed by propionic acids. Formic, acetic and benzoic acids were detected as major species in used engine oil, with negligible concentrations in new oil (Kawamura et al., 1985).

Isoprene fluxes in deciduous forest at 30^0C varied from 2500 to 8000 $\mu g/(m^2 xh)$ increasing exponentially with temperature. Alphapinene emissions from Douglas fir ranged between 9 and 1320 $\mu g/(m^2 xh)$, being influenced by the relative humidity (Lamb et al., 1985).

Børger (1985) gave a summary of a VDI-seminar held in Mannheim in March 1985 about catalytic and thermal methods for waste gas purification.

Calculations of the ammonia emissions in 27 European countries were presented by Buijsman et al. (1985) for the year 1982. The total emission amounts to 6.4 million tons of NH_3 or less. Decomposition of livestock wastes appeared to be the most important source. Less important were fertilizers and industrial sources. Calculations were also made for grid systems with grid elements of 75 x 75 km^2 at 60^0N and of 150 x 150 km^2 at 60^0N (EMEP grid).

Other papers on emissions were published by Gravenhorst (1985) and Blommers (1985).

3.4 METEOROLOGY

A 1 x 1 degree latitude-longitude digital data set of areally weighted clear sky surface albedo of snow-covered land in the middle and high latitudes of the northern Hemisphere has been computed from satellite imagery. The data set is relevant for e.g. long-range transport calculations to determine deposition velocities (Robinson and Kukla, 1985).

Solar spectral direct and diffuse irradiance and directional radiance at the surface, spectral absorption within the atmosphere and the upward reflected spectral irradiance at the top of the atmosphere, were calculated by Justus and Paris (1985).

Three subtypes of fair-weather cumulus clouds have been identified based on the nature of their interaction with the mixed-layer: forced, active and passive clouds. Mixed-layer air can be vented into the free atmosphere in active fair-weather cumulus clouds (Stull, 1985).

It was found by Brown et al. (1985) that average rainfall intensity and maximum intensity were quite dependent on the duration of an rainfall event (defined as one or more consecutive hours with at least 0.25 mm of precipitation). Hourly precipitation amounts within an event were not independent or identically distributed.

Miller and Harris (1985) reported on the Western Atlantic Ocean Experiment. Direct flow off the North American continent occurred 60% of the time during a 7-year period. Chemical data from Bermuda showed that the more the flow was from the NW and WSW, the more acidic transport took place across the Atlantic.

Sebacher et al. (1985) found that polluted, stratified layers in the lower 3000 m of the troposphere are of particular importance for the understanding of the transport from the continent over the ocean.

Henmi and Bresch (1985) published a paper on the meteorology of high sulphur episodes in the western United States.

3.5 MODEL FORMULATION, NUMERICAL METHODS

For grid sizes like those used in regional models (50 km or more), about half of the numerical schemes investigated produced artificial diffusion larger than natural diffusion. For grid sizes of 0.5 and 5 km, most numerical schemes produced numerical diffusion smaller than the natural diffusion (Sheih and Ludwig, 1985).

3.6 DEPOSITION

Sampling errors in vertical flux measurements obtained by eddy correlation methods were investigated by O'Brien (1985).

Riggan et al. (1985) found that atmospheric deposition plays a major role in the nitrate pollution of stream water in mountain watersheds of Los Angeles County, California.

Dasch (1985) found that deposition is strongly influenced by the affinity of the surface for gases and the retention characteristics of the surface for particles.

Concurrent measurements of the dry deposition of sulphate, particulate sulphur, nitrate, SO_2, O_3 and HNO_3 were made in a campaign in Illinois during September 1981 and June 1982. sulphate and particulate sulphur mean deposition velocities were about 0.3 cm/s with near zero nighttime values and daytime values up to 1 cm/s. Daytime SO_2 and HNO_3 deposition velocities up to 3 cm/s were found, suggesting near-zero vegetation canopy resistance to mass transfer of these gases (Dolske and Gatz, 1985).

Average daytime deposition velocity for HNO_3 over grass was measured by Huebert and Robert (1985) to be 2.5 cm/s with average canopy resistance almost zero.

Peak values of 1 cm/s at midday changing to less than 0.1 cm/s during the evening for the dry deposition velocity for ozone were measured over grassland by Droppo (1985).

A long-term mean deposition velocity of 0.22 cm/s was found for particulate sulphur over Illinois grassland, with peak velocities greater than 0.5 cm/s in windy conditions by Wesely et al. (1985).

Jonas and Heinemann (1985) found that the deposition velocity for 1 micrometer particles ranged from 0.2-0.4 cm/s over grassland and forested areas.

Kluczewski et al. (1985) found that the dry deposition velocity of COS is about three orders of magnitude smaller than that of SO_2, and concluded that ground removal is not an important sink for COS.

Cadle et al. (1985) measured the deposition velocity of HNO_3, SO_2 and various particulate species over snow. The deposition velocity for HNO_3 averaged 0.15 cm/s, for SO_2 0.15 cm/s above -3^0C and 0.06 cm/s below -3^0C, for Ca^{++}, Mg^{++}, Na^+, K^+ and NH_4^+ 2.1, 1.5, 0.44, 0.51 and 0.10 cm/s, respectively, with 4.4, 2.7, 1.8, 0.9 and 0.46 micrometer diameter, respectively.

Papers on deposition were also published by Speer et al. (1985), Davidson et al. (1985), Katen and Hubbe (1985), Lindberg and Lovett (1985), Davis and Wright (1985), Granat and Johansson (1985), Feely et al. (1985), Lorenz and Murphy (1985), Neumann and Hartog (1985) and Colbeck and Harrison (1985b)

4 MODEL VALIDATION. MEASUREMENTS

4.1 REGIONAL SULPHUR AND NITROGEN SPECIES, ACID PRECIPITATION

The variation in sulphur dioxide emissions from metal smelters in the western United States has been compared with the variation in sulphate concentrations in precipitation in the Rocky Mountain states. The data supported a linear relation between emissions and sulphate concentration (Oppenheimer et al., 1985a).

Belikova et al. (1984) gave maps of sulphate accumulation in snow, mean deposition rates, and snow melt sulphate concentrations for all of the USSR.

The concentrations of eight ions in precipitation were measured over one year, and it was found that the concentrations of hydrogen, sulphate and nitrate ions were several times higher at the site influenced by Tokyo-emissions than at the site more remote from Tokyo (Dokiya et al., 1985).

Shriner (1985) gave a review of the role of Oak Ridge National Laboratory's research into the effect of energy development on forested and aquatic ecosystems.

The major sulphur gas in fresh, estuarine and coastal waters that were examined by Turner and Liss (1985), was dimethyl sulphide, with lesser amounts of carbonyl sulphide and carbon disulphide. Carbonyl sulphide showed no significant seasonal variation. The flux of the gas was always from the seawater to the air.

Stratus cloudwater was monitored by Waldman et al. (1985) as it intercepted a pine forest north-east of Los Angeles. In cloudwater the nitrate/ sulphate-ratio was between 1.5 and 2 on equivalent-basis, for rainwater at the same site about 1. Deposition of sulphate, nitrate and free acidity due to intercepting clouds was estimated to be comparable to that due to rainfall at the site.

Castillo et al. (1985) could not detect with certainty that sulphate in water at Whiteface mountain dropped during the summer 1982 in winds from the west northwest, even though Canadian smelters were idle during August 1982 reducing their SO_2 emissions by about 75%.

The chemical composition of precipitation collected for a 1 yr period at four sites in Western Washington was presented by Vong et al. (1985). Highest concentrations were found near sources of pollutants and in the summer. Seasalt, metals from a copper-smelter, urban emissions, soil and acidic sulphate aerosol were the five factors that contributed the most to measured concentrations.

Beilke et al. (1985) reported aircraft and groundbased measurements of transboundary fluxes of nitrogen- and sulphur-compounds across the boarder between FRG and GDR. The emissions of SO_2 are higher in GDR than in FRG, but the prevailing winds are from the west to the east. On an annual basis, the net flux of S-compounds across the boarder is small, but there are serious pollution episodes with transport from the east.

Georgii (1985) reported that the frequency of fog and of low-layer clouds increased with altitude in mountain areas. In the Rhine-valley 40 d/y had fog, the number of fog-days increased to 120 per year at 600 m.a.s.l. and to 200 per year at 800 m.a.s.l. in the Taunus mountains. Comparative measurements of the chemical composition of fog- and rainwater at the same location showed a considerably higher sulphate-, nitrate- and chloride-concentration in fogwater. The enrichment factor was between 5 and 10.

During low inversion conditions during the morning and afternoon of several days in December 1983, 700 and more (up to 1000) $\mu g/m^3$ of SO_2 as half hourly mean values, were measured in Hamburg (Bruckman et al., 1985).

Buck (1985a) reported on measurements of particulate matter in Nordrhein-Westfalen during the severe smog-episode in January 1985. More than 500 $\mu g/m^3$ of particulate matter as 24 h-average, was measured during the episode.

Lee and Shannon (1985) discussed numerical simulations of in-cloud chemistry. There were indications of non-linearity which may not be significant over relevant time and space scales.

Møller and Schieferdecker (1985) reported that in an area in FRG with slightly increasing SO_2- emissions, the concentration of SO_2 dropped over a 10-year period from 1970 to 1980. The reason for this could probably be found in the increasing ammonia-emissions in the same area.

In rain samples stored at room temperature considerable changes occurred in ammonium and phosphate concentrations both in light and dark storage, while storage in the dark at 4^0C resulted in satisfactory sample preservation (Ridder et al., 1985).

Sisterson et al. (1985) found that when analysing precipitation samples from northeastern Illinois sites, weekly samples had less ammonium and more sulphate (and more Ca and Mg in dry seasons) than samples collected on an event basis.

In a study reported by Hoff and Gallant (1985), the SO_2 plume from Sudbury was traced with a Barringer Cospec instrument and compared with calculated trajectories.

Rømer et al. (1985) found high molar ratios of nitrate to sulphate in aircraft measurements over the Netherlands (ratios ranging between 1 and 2), while lower values were found over southern Scandinavia.

McClenny et al. (1985) applied Fourier transform infrared transmission spectroscopy to analyse filters for ammonium and sulphate in ambient aerosol particles. The method was judged to be very accurate and of good sensitivity.

A technique for the measurement of ambient concentrations of HNO_3, was described by Lindqvist (1985a). The minimum detectable concentration was 0.014 $\mu g/m^3$ for a 30 l sample. Lindqvist (1985b) also reported on a technique for monitoring ambient sulphuric acid.

Gaseous nitric acid and particulate nitrate were measured by high-volume sampling and on non-impregnated filter by Meixner et al. (1985). Measurements at Jülich, FRG from Jan. 1982 to Jan. 1984 of gaseous nitric acid and particulate nitrate were presented. There was a summer maximum in the monthly average HNO_3 concentration of about 1.5-2 ppb, while there was a midwinter minimum of a few tenths of a ppb. The nitrate aerosol concentration showed a less clear picture with respect to its annual variation. The monthly mean nitrate concentration usually ranged from 1 to 6 ppbm. The nitric acid concentration had a diurnal cycle with lower concentration during the night than in daylight. This was particularly pronounced in summer.

Anlauf et al. (1985) reported on the measurement of nitric acid, nitrate and ammonimum using tunable diode laser absorption, tungstic acid denuder tube

and a filter pack. Reasonable agreement was found, and sub ppb concentrations were measured with sufficient sensitivity under field conditions in southwestern Ontario.

It was shown by Peake et al. (1985) that PAN is the major reaction product of nitrogen oxides in cold, dry continental climatic conditions over Calgary. Total inorganic nitrate and PAN made up on the average 8% of the total of nitrogen-containing compunds.

The distortion of the concentration of rare earth elements on fine airborne particles in emissions from oil-fired power plants and refineries compared to the crustal abundance, can be used to differentiate between sources of atmospheric particulate material (Olmez and Gordon, 1985).

Elemental analysis of aerosol from various sites in the northeastern United States revealed a persistent northeastern "foreground" upon which pulses of midwestern aerosol were superimposed every few days, in response to large-scale meteorological features (Rahn and Lowenthal, 1985).

Ogren et al. (1985) found a method to separate cloud droplets from atmospheric trace gases, rain drops and submicrometer aerosol particles on the basis of their aerodynamic properties upon evaporating by dry air. The trace gases and residual particles could be analysed by in-situ methods or collected for subsequent analysis.

Hoppel et al. (1985) found that there was a rapid decay in particles less than 0.05 micrometer diameter during the first day of transport off the east coast of the United States, whereafter the concentration stayed quite unchanged. Significant changes in aerosol distribution was often associated with changes in air mass.

Hobbs and Yates (1985) presented measurements of Aitken nucleus counts, particle size distributions, light scattering and ozone from 39 transit flights across the North Atlantic Ocean. Data on "background" aerosol concentrations were given, which is of relevance for the determination of the "unknown" sulphate in Europe.

In three papers, Wiman and Ågren (1985), Wiman and Lannefors (1985) and Wiman et al. (1985) presented detailed measurements and analysis of aerosol concentration distributions and depletion in a coniferous forest.

In the two papers by Ligocki et al., (1985a, 1985b) concentration data for a number of organic compounds were presented for rainwater and in the gas phase from Portland in Oregon. It was found that there is equilibrium between rain and the atmospheric gas phase for non-reactive neutral organic compounds.

Leuenberger et al. (1985) found through air and rain analyses that phenols almost exclusively appear in the gaseous and dissolved forms. Gas scavenging is more important than particle scavenging for phenols.

Other papers on measurements of components contributing to acid deposition: Reddy et al. (1985), Schnug and Vonfranck (1985), SFT (1985), Stachurski and Zimka (1984), Steinberg et al. (1985), Tsungai et al. (1985), van Noort and Wondergem (1985), Innes et al. (1985), Warner et al. (1985), Dufour et al. (1985), Chan and Lusis (1985), Slanina (1985), Rømer (1985), Fuzzi (1985), Onderlinden and van Jaarsveld (1985), Ronneau (1985), Winkler (1985), Nguyen et al. (1985), Freyer (1985), Slanina et al. (1985), Allegrini et al. (1985), ten Brink et al. (1984a), Rømer et al. (1984), Buck (1984), Lewin et al. (1984), Serna et al. (1984), Georgii et al. (1984), Fugas et al. (1984), ten Brink et al. (1984b), Fuhrer (1984), Vierkorn-Rudolph et al. (1984), Reiter et al. (1984), Gervat et al. (1984), Elshout and Beilke (1984), Pena et al. (1985), Vandenberg and Knoerr (1985), Dayan et al. (1985), Spann and Richardson (1985), Willison et al. (1985), Mulawa and Cadle (1985), Cher (1985) and Bilonick (1985).

4.2 REGIONAL MEASUREMENTS OF PHOTOCHEMICAL OXIDANTS, HC AND NOx

A denuder technique for sampling and analysing nitrous acid at sub-ppb levels in air was described by Ferm and Sjødin (1985). Nitrous acid is a potentially important source of hydroxyl radicals after sunrise. The detection limit was as low as 0.01 ppb.

Atmospheric concentrations of nitrous acid were measured in Gothenburg by Sjødin and Ferm (1985). During the period January-May, values in the range 1-50 nmol/m^3 (or 25 ppt-1.3 ppb) were measured with the highest concentrations during the night.

Measurements of NO_2 were carried out in 26 Swedish urban areas from January 1983-March 1984. The background NO_2-concentrations ranged from 15 to 49 µg/m^3, while the highest daily mean concentrations were measured in Norrkøping and Gothenburg (211 and 183 µg/m^3, respectively) (Svanberg and Grennfelt, 1985).

The first simultaneous measurements of ammonia and nitric acid from aircraft in the free troposphere, were reported by LeBel et al. (1985). Over land ammonia and nitric acid decreased with altitude (over Virginia and Maryland), while over the North Atlantic there was an increase in ammonia with height.

In the paper by Cvitas et al. (1985), measurements of O_3, CO, NOx and SO_2 in Athens during June 1982, were reported. Up to 300 ppb of ozone was measured.

Crutzen et al. (1985) found that burning of biomass during the dry season lead to substantial emissions of pollutants like CO, NOx, N_2O and methane. Ozone was enhanced due to photochemical reactions. The biogenic organic emissions from tropical forests were important sources for CO and other secondary gases. NOx was supplied by lightning over tropical land areas, giving rise to the generation of ozone in the free troposphere over Amazonas.

Aircraft measurements over savanna regions of central South America showed that elevated ozone concentrations were found in the smoke from biomass burning, which typically could persist for weeks in the dry season and was confined to the lower 3 km of the troposphere (Delany et al., 1985).

In the paper by Hofzumahaus et al. (1985) was reported an important step forward in the understanding of atmospheric hydroxyl. Hydroxyl radical concentrations were measured by long path differential UV absorption spectroscopy at three sites in FRG with different levels of air pollution. Maximum

observed hydroxyl concentrations ranged from less than $5x10^5$ to $6x10^6$ molecules/cm^3. The measurements for NOx concentrations above 2 ppb could be well described by model calculations using observed concentrations of ozone, NOx, hydrocarbons, aldehydes and other species as input, while for lower NOx concentrations, the model indicated that there may be yet unrecognized hydroxyl loss processes.

Measurements of the vertical and horizontal distribution of nonmethane hydrocarbons over Western Europe were reported by Tille et al., 1985a, 1985b.

Measurements of nonmethane hydrocarbons in remote, rural and semi-rural areas were presented by Rudolph and Khedim (1985). Some conclusions were drawn about the impact of man-made hydrocarbons on the chemistry of the atmosphere outside urban and industrialized areas.

Measurements of natural hydrocarbons in a forested area outside Rome indicated that they do not play a major role in determining the concentration of photochemical oxidants in rural or suburban areas (Ciccioli et al., 1985).

Anlauf et al. (1985) presented concurrent measurements of atmospheric aerosol constituents, nitric acid, ammonia, PAN, O_3, SO_2, and individual C_2-C_6 hydrocarbons at a rural site in southern Ontario during June 1982. The measurements were also interpreted in terms of sources.

Colbeck and Harrison (1985a, 1985c) reported on measurements of ozone, NOx and individual hydrocarbons during a photochemical smog episode in NW England in July 1983. Up to 156 ppb as hourly average ozone concentration was measured.

Other papers about measurements were published by Helas et al. (1985), Cocks et al. (1985), Rudolph et al. (1985a), Hagele et al. (1985), Schurath et al. (1984a), Meyrahn et al. (1984), Schurath et al. (1984b), Ciccioli et al. (1984), Hjorth et al. (1984), Muller and Riedel (1984), Sjødin and Grennfelt (1984), Kessler and Platt (1984), Bamber et al. (1984), Fenger (1984), Hewitt and Harrison (1985), Martin and Barber (1985) and Roberts et al. (1985a).

4.3 MEASUREMENTS IN THE GLOBAL TROPOSPHERE EXCEPT THE POLAR REGIONS

A detector for the chemiluminescent measurement of NO in background air has been described. Vertical profiles in the atmospheric boundary layer and the free troposphere over FRG in clean air showed NO-concentrations as low as 20 ppt, which was the detection limit for a 1 min integration time (Drummond et al., 1985).

A technique to measure PAN was described by Rudolph et al. (1985b). Down to 50 ppt was measured in clean air near Jülich in FRG.

Methanesulfonic acid (MSA) is a major oxidation product of dimethylsulphide (DMS) in air, via reaction with OH radicals. MSA and non-sea-salt sulphate were significantly correlated at both Fanning and American Samoa (Maritime stations in the Pacific Ocean). The levels of MSA and non-sea-salt sulphate were higher at Fanning which is located in the biologically productive waters in the equatorial divergence zone, than at American Samoa surrounded by low productivity waters (Saltzman et al., 1985)

The mean of 52 measurements of CS_2 was 5.7 pptv. The amounts of CS_2 in the two hemispheres were statistically the same (Tucker et al., 1985).

Methyl chloride, carbon monoxide and freon-11 may be used as tracers of regional scale pollution. Methyl chloride and carbon monoxide are good indicators of slash burning, while freon-11 originates from urban pollution (Khalil and Rasmussen, 1985).

Bonsang and Lambert (1985) reported on measurements of nonmethane C_2-C_6 hydrocarbons in the Mediterranean and Red Sea in June 1982. Typical concentrations in marine atmosphere were between 0.05 and 0.2 ppb.

Global distribution was presented by Ehhalt et al. (1985) of light hydrocarbons sampled in an aircraft flight from Greenland to South America.

Non-methane hydrocarbon mixing ratios in Kenyan savannah were as low as those measured over oceans. Carbon monoxide mixing ratios were higher than marine measurements at similiar latitude (Greenberg et al., 1985).

Other papers on global measurements were published by Volz and Kley (1985), Nutmagul and Cronn (1985), Roberts et al. (1985b), Drummond and Volz (1985), Broll et al. (1984) and Rudolph et al. (1984).

4.4 MEASUREMENTS IN THE POLAR REGIONS

Measurements of sulphur pollutants from Norwegian arctic sites 1977-1983 showed persistent annual variation with a late winter-spring maximum and summer minimum. Aircraft measurements showed that there is a persistent layered structure in the troposphere with thin, a few hundred metres thick, layers of polluted air (Joranger and Ottar, 1984).

Substantial concentrations of black carbon and sulphur were found at all altitudes in the Arctic atmosphere during the March-1983 AGASP-flight campaign (Hansen and Rosen, 1984).

Similar sulphate concentrations and aerosol acidities were found over Alaska, the North Pole and over the north Atlantic during the March-1983 AGASP flight campaign (Lazrus and Ferek, 1984).

Chemical analysis of Spitsbergen snow cores have revealed a spatial pattern consistent with orographic deposition of major anthropogenic pollutants with air movements from southeast towards northwest. The concentrations of nitrate and ammonium ions were very low relative to excess sulphate. This indicates either selective removal of nitrate and ammonium during transport, or dominating emissions of sulphur dioxide relative to nitrogen oxides and been ammonia in the source regions (Semb et al., 1984).

Firn core analysis published by Barrie et al., (1985) indicated that between 1956 and 1977 there was a 75% increase of Arctic air pollution associated with a marked increase in the emissions of SO_2 and NOx in Europe.

Ottar and Pacyna (1984) used trace metals in atmospheric aerosols together with trajectory analysis to identify sources regions for Arctic pollution.

Wolff and Peel (1985) presented a review of observed historic changes in the concentration of gases, particulate matter and heavy metals in polar ice and snow.

The concentration of nonmethane hydrocarbons at Svalbard has been shown to vary with season as the concentration of sulphate, except for species of natural origin: the concentration of ethene and propene peaks during the summer, and these species are thought to be in equilibrium with the surrounding ocean waters (Hov et al., 1984).

Possible sources of Arctic air pollution at ground level must be at nearly the same temperature as found in the Arctic. Sources south of the polar front system can only contribute to the upper level Arctic pollution. The amplitude and phase of long, planetary waves are important since they determine the position of the polar front, and provide conditions for meridional transport at certain longitudes (Iversen, 1984).

Sulphate measurements on Bjørnøya and Ny Ålesund for the period 1979-1984 were analysed with respect to pollution episodes by Iversen (1985). The episodes are frequent during late winter and early spring and during early autumn. The seasonal variations in blocking and quasi-persistent poleward flows are very much related to the seasonal cycle of Arctic pollution. Large scale, quasi-stationary atmospheric flow systems determine the basic conditions for long range transport from mid- to polar- latitudes.

Other papers on atmospheric measurements in the polar regions were published by Schnell and Raatz (1984), Delmas and Legrand (1985), Rahn (1985), Barrie and Hoff (1985), Iversen and Joranger (1985), Raatz (1985), Raatz et al. (1985a, 1985b, 1985c) and Rosen and Hansen (1985).

4.5 MEASUREMENTS OF TRENDS

Measurements of sulphate and nitrate in firn samples from Dye 3, South Greenland, each sample covering one year for the period 1895-1978, showed that both sulphate and nitrate increased by a factor of about 2 during the period (Neftel et al., 1985).

Changes in visibility and the occurrence of smoke and haze were identified for eight locations in and around Illinois by Vinzani and Lamb (1985). Summer was the season that experienced the greatest 1950-80 visibility change. There was a pronounced overall decline that coincided with a marked increase in the frequency of smoke/haze. The winter visibility trends for individual stations ranged between a moderate decrease and a noticeable improvement, and were associated with strong reductions in the smoke/haze frequency.

Air entrapped in ice bubbles of cold ice in western Antarctica has nearly twice as high methane content today compared to 200 years ago (Stauffer et al., 1985).

Analysis reported by Rinsland et al. (1985) of solar absorption spectra indicated that the tropospheric content of methane has gone up by 1.1% per year in the period 1951-1981 ($1.1 \pm 0.2\%$).

From a comparison of the reults from 1950-51 and modern measurements, an average increase of 2% per year in the free tropospheric concentration of CO above Europe is estimated for 1950-77 (Rinsland and Levine, 1985).

The paper by Enting (1985) treated the deconvolution of observed air bubble concentrations into atmospheric concentrations at a specified time.

Some examples were given by Helmes and Jaenicke (1985) of how to obtain new information from the historical records of classical networks such as meteorological networks.

Neftel (1985) found that there is a degradation of H_2O_2 in the snow with time, even when temperatures are between -5 and -10^0C. For the conservation of metastable compounds in snow cover, it seems that lower temperatures, encountered in the polar regions, are required to obtain historical concentrations.

Other papers on trend measurements have been published by Lorius et al. (1985), Walker (1985) and Attmannspacher (1985)

5 EFFECTS AND POLICY QUESTIONS

5.1 EFFECTS OF POLLUTANTS

Very few references are included in this chapter.

The analysis by Adams et al. (1985) indicated that the benefits to the U.S. society of moderate (25%) ozone reductions are approximately $ 1.7 billion, while a 25% increase in ozone pollution results in costs of $ 2.1 billion (not net costs).

Other papers on effects: McLaughlin (1985) and Ballach and Brandt (1985).

5.2 POLICY QUESTIONS RELATED TO ACID DEPOSITION

Outline of a global atmospheric research programme with five broad objectives was given in the American Global Tropospheric Programme (1984): Evaluate biological sources of atmospheric trace substances, determine global distribution of trace gases and airborne particles, test photochemical theory, investigate wet and dry deposition for trace gases and aerosol particles, develop models for global tropospheric chemistry systems.

Effects of SO_2 and NOx emissions reductions were claimed to be sufficiently well known to act in favour of emission reductions (Oppenheimer et al., 1985b).

In the paper by Morrison and Rubin (1985) was described an analytical model designed to explore the consequences of acid rain control strategies for coal-fired power plants in the 31 eastern United States.

The paper by Lee et al. (1985) was a summary of an APCA International Specialty Conference.

Scientific, economic, social and environmental perspectives on acid rain issues were presented by Perhac et al. (1985).

Outline was given of the steps taken in FRG to reduce emissions of SO_2, NOx and particulate matter (Kroppenstedt, 1985).

Summary of the outcome of a symposium on NOx in Karlsruhe in 1985 was given by Issle and Weibel (1985). Papers were presented on flue gas control in power plants in Japan, USA and Europe, with emphasis on NOx.

Halbritter et al. (1985) presented calculations of the impact of different strategies of the use of fossil fuels on the concentration, deposition and long-range transport of atmospheric sulphur. The trajectory puff model MESOS was used. It was found that for FRG, more than 70% of the sulphur comes from foreign sources, furthermore that the emission reduction of SO_2 in FRG that followed the control of large boilers in 1983, will not be accompanied by a similar reduction in atmospheric concentration and ground deposition of sulphur in FRG.

The smog-situation in Nordrhein-Westfalen in January 1985 was discussed in an editorial in Staub (Buck, 1985b).

The concentration of SO_2 and particulate matter in Nordrhein-Westfalen in January 1985 led to the initiation of smog alarms. On occasions, more than 1.2 mg/m^3 of SO_2 was measured as three-hourly average concentration. The accompanying values of particulate matter reached 0.6 mg/m^3 (Kulske and Pfeffer, 1985).

Other papers on policy questions were published by Roth (1985), Kowalczyk and Tombleson (1985), Doctor et al. (1985), Liberti et al. (1984), Ellis et al. (1985)

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Literature review | acid deposition | photochemical oxidants

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About 300 papers published in 1985 or late in 1984 have been reviewed. There has been progress in the development of models for acid deposition and photochemical oxidants. Simultaneous measurements of O_3, NOx, individual hydrocarbons, aldehydes, CO and OH have lead to the conclusion that the atmospheric gas chemistry in moderately polluted air is well understood. There is an upward trend over Europe in the concentration of methane, CO and O_3.

TITLE

ABSTRACT (max. 300 characters, 7 lines)

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