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FATE OF PRODUCTS OF DEGRADATION PROCESSES: CONSEQUENCES FOR CLIMATIC CHANGE

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ABSTRACT

The end products of atmospheric degradation are not only CO₂ and H₂O but also sulfate and nitrate depending on the chemical composition of the substances which are subject to degradation processes. Atmospheric degradation has thus a direct influence on the radiative balance of the earth not only due to formation of greenhouse gases but also of aerosols. Aerosols of a diameter of 0.1 to 2 micrometer, reflect short wave sunlight very efficiently leading to a radiative forcing which is estimated to be about -0.8 watt per m² by IPCC. Aerosols also influence the radiative balance by way of cloud formation. If more aerosols are present, clouds are formed with more and smaller droplets and these clouds have a higher albedo and are more stable compared to clouds with larger droplets. Not only sulfate, but also nitrate and polar organic compounds, formed as intermediates in degradation processes, contribute to this direct and indirect aerosol effect. Estimates for the Netherlands indicate a direct effect of -4 watt m⁻² and an indirect effect of as large as -5 watt m⁻². About one third is caused by sulfates, one third by nitrates and last third by polar organic compounds. This large radiative forcing is obviously non-uniform and depends on local conditions.

INTRODUCTION

Aerosol particles (very fine "dust") have great effects on the environment. These effects range from direct effects on human health, contributions to acid deposition and eutrophication, influence on oxidant formation to radiative forcing. This forcing is negative, leading to cooling by reflecting sunlight. Assessment of these effects as well as investigations of the sources of these aerosols require sufficient knowledge on chemical composition of the aerosols.

Aerosols play an important role in the radiative balance of the earth. The greenhouse gases retain long wave radiation in the atmosphere, leading to a change in the radiative balance of about 2.5 watt per m², according to IPCC [1]. According to IPCC [1] aerosols act in the opposite way, by reflection of sunlight, with an estimated contribution, as global mean, of about 0.8 watt per m². Aerosol particles influence the radiative balance of the earth in two ways: directly by reflection of incoming solar light and indirectly by the

process of cloud formation. For the characterization of both the direct and the indirect effect characterization of physical aspects of aerosols (number of particles, particle size distribution etc.) as well as chemical composition is necessary.

"Aerosol" particles in the atmosphere, reflect the radiation [1,2] depending on their size distribution. The size of the particles determines whether shorter radiation is reflected more effectively compared to infra red. Incoming solar radiation is effectively reflected by small particles (0.1 to 2 micrometer), while particles of this size do not intercept the outgoing infrared radiation of the earth. In order to assess the cooling influence of this reflection, both the size and the chemical composition of aerosols must be known. The chemical composition influences not only scattering but determines also whether a particle will grow by the uptake of water (vapor) due to the influence of the hygroscopic compounds of the aerosol. Smaller particles, with a diameter below 0.1 micron are more abundant in the atmosphere compared to larger ones. If particles grow by the uptake of water vapor, result is the increased reflection of solar light. In areas with high relative humidity this effect leads to a factor 2.5 to 3 more reflection than calculated on the basis of "dry particle size distribution". High concentrations of ammonium nitrate, ammonium sulfate or ammonium chloride induce increased uptake of water vapor. In fact no "dry particles" occur under conditions as encountered in the Netherlands, the reason is the combination of high ammonium nitrate, ammonium sulfate and probably ammonium chloride concentrations in combination with high relative humidity.

Until now IPCC has included in the assessment of the radiative forcing of aerosols only the contribution of sulfate. As there seems to be quite large uncertainties which components are taken into account by IPCC it is perhaps useful to stress this point. IPCC gives very clear reasons why the decision was made to ignore or rather do not yet include other aerosol compounds such as nitrate or organic compounds as will be explained below. The assessments in the present IPCC reports have been carried out in the following way: Concentrations fields of sulfate (specifically of total mass of sulfate, and not total aerosol) are obtained based on model calculations. The light-reflection of this aerosol is mostly deduced on the basis of a measured relation between aerosol-light-scattering and sulfate mass concentration, assuming a size distribution of sulfate which is globally valid. The reflection of solar light can be calculated, when the height over which the sulfate is vertically distributed is known, according to a simple formula:

$$\delta_a = \int_0^H \int_{\lambda_1}^{\lambda_2} B_{sp} \times d\lambda \times dz$$

δ_a = optical thickness

H = Height of the Boundary layer

B_{sp} = Particle scattering coefficient

$\lambda_1 \dots \lambda_2$ = wave length of solar spectrum

which leads to a change in the solar flux ΔF_{direct}

$$\Delta F_{direct} = - \frac{1}{2} FT^2 \times (1-A_c) \times (1-R_s)^2 \times B \times \Delta\delta_a$$

F = Top of the atmosphere radiative flux

T = Fraction of incident light transmitted by the atm. layer above the aerosol layer

R_s = Mean surface albedo

A_c = Annual mean fractional cloud cover

B = Annual mean upward fraction of light scatter due to aerosol

$\Delta\delta_a$ = Annual mean change in aerosol optical depth due to anthropogenic aerosols.

A correction for the change in size distribution, due to the uptake of water vapor, is applied in a number of recent assessments. Apart from soot, the contribution of other constituents of aerosols has been ignored, partly because of lack of data as in the case of organic compounds but also because of measurement artifacts such as was the case for nitrate.

IPCC describes explicitly why only the mass of sulfate is taken into account: It has been assumed in the past that nitrate is primarily present in very large particles, with a diameter over 2 micron and hence would not have a contribution to radiative forcing. The last IPCC reports [1,2] mention this reason too for not including nitrate in the radiative balance next to the remark that nitrate concentrations are rather low. Recent assessments of the influence of nitrate in aerosol have shown clearly that both prepositions are probably incorrect as will be explained in chapters 2 and 3. The present measurement methodology very often underestimates the nitrate content in aerosols and also leads to wrong conclusions regarding the size distribution.

Polar organic compounds were simply overlooked until recently. The oxidation of volatile organic compounds, either emitted by anthropogenic sources or biogenically, leads not only to the formation of ozone and other oxidants, but also to carbonyls and carboxylic acids, which are fairly polar compounds and water soluble. The organic acids and especially their salts, have very low vapor pressure and will hence be present in aerosols. Initial results of measurements both in the US and Europe have already indicated that about 25 to 40% of the aerosols consist of organic compounds and it has become clear that a large fraction of this organic material is made up by polar organic acids and related compounds [7]. This material will contribute to the direct effect and indirect effect of aerosols, in the same way as nitrate and sulfate.

In a more indirect way, anthropogenic aerosol also acts as a climate modifier. Aerosol particles are the nuclei for clouds and anthropogenic aerosol particles serve as extra cloud nuclei. Clouds with increased numbers of cloud droplets are more reflective [1,3]. So if larger number of aerosols, due anthropogenic emissions, are available as condensation nuclei, clouds with a higher albedo are formed. This phenomenon is important for certain types of clouds, namely those which exist normally of very large droplets, such a maritime stratus. The addition of extra aerosol particles, due to anthropogenic emissions, in the process of formation of maritime stratus will result in clouds with a much higher albedo than the original ones. These clouds are also stabilized, as precipitation is less easily formed from clouds with small droplets. To be able to act as condensation nuclei, aerosols must meet certain requirements normally: Their size must be over a certain threshold (70 nm under conditions in the Netherlands) and they must largely consist of soluble material. The relation between droplet size distribution in clouds and number of aerosols present under conditions of cloud formation is not linear but shows saturation at higher number of particles and this

relation can be described by mean of a logarithmic or cube root mathematical expression [3]. IPCC recognizes [1,2] the possible large contribution of this so-called indirect effect but states that it is difficult to quantify the role of sulfate (again only sulfate is taken into account in the IPCC reports) that no mean value is presented but only a possible error bar extending from 0 to -1 watt per m^2 . Recent research in the Netherlands has made assessments of the local indirect effect possible.

ASSESSMENT OF THE DIRECT AND INDIRECT EFFECT

Very recently an assessment has been made of the direct and indirect effect of aerosols under conditions in the Netherlands [3], taking sulfate, nitrate and organic compounds into account. The direct effect was assessed by a year long series of integrating nephelometer measurements in combination with closure experiments. In this way the dispersion of radiation by aerosols can be measured. Nephelometer measurements are carried out quite easily, but in these instruments the air is dried and the scatter of "dry aerosol" is measured. So the found values for scattering must be converted to ambient conditions by calculating the loss of water of aerosol particles in the nephelometer [3]. This calculation has been verified by nephelometric measurements under variable conditions of humidity. To check this procedure in a second way closure experiments are carried out in which this calculated reflection based on nephelometric data is compared to values derived from measurement of size distribution and chemical composition of aerosols [11] combined with direct measurement of the solar flux on days with perfectly cloudless skies. These experiments have shown that a good relation between calculation and measurements exist and that the direct effect can indeed be calculated on the basis of nephelometric data in combination with the verification as described above. The value found for the location of ECN, at the North-Western coast of the Netherlands is on the order of about -4 watt per m^2 . Sulfate contributes less than half to this substantial forcing. This value for the direct forcing is actually a lower limit: It is assumed in these calculations that all aerosol is present in the mixing layer and that the contribution of aerosol in all other layers of the atmosphere is negligible.

The indirect effect has been investigated by means of a special cloud chamber and wind tunnel (CHIEF) which allows the study of cloud formation of ambient aerosols under conditions which are comparable to the actual conditions in the atmosphere [3]. CHIEF can generate low and constant supersaturation, typically in the order of 0.3 % relative humidity. Outside air is pumped in the installation and droplet formation is evaluated as function of ambient aerosol characteristics. The location of ECN has the advantage that in some conditions of North-Western wind directions more or less pure background air (containing less than $50 \text{ particles per cm}^3$) is available while under conditions of easterly winds strongly polluted air can be studied. Droplet size distribution of clouds formed under more or less "true background conditions" ($50 \text{ particles per cm}^3$) can in this way be compared with the size distribution of clouds generated in air which contains more aerosol. These results have been extrapolated to average conditions in the Netherlands, taking the frequency of marine stratus and other meteorological conditions into account. The

indirect effect was in the order of -5 watt per m^2 , the contribution of sulfate is slightly uncertain but could be in the order of 50%.

The total forcing by aerosols under conditions in the Netherlands is in the order of -9 watt m^{-2} , with an estimated uncertainty of 3 watt m^{-2} and this value is nearly 4 times the forcing of the sum of the radiative forcing of all greenhouse gases. These very large values for radiative forcing in conjunction with the role of the chemical composition of aerosols has placed formation of aerosols but also measurement methodology again in the center of attention.

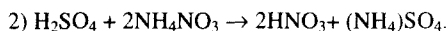
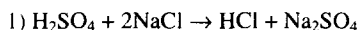
DISCUSSION OF THE RECENT RESULTS VERSUS EARLIER ESTIMATES

There are basically three reasons for the difference between these recent experimental results and the evaluation of IPCC in their latest reports based on model calculations:

1. As already mentioned, the contribution of polar organic compounds was simply overlooked. Only quite recently research has started to characterize the contribution of polar organic compounds to the direct and indirect effect. Our initial measurements indicate that the influence of organic compounds is large.
2. The classical filter methodology, used to analyze the chemical composition of aerosols, leads to a severe underestimation of the amount of ammonium nitrate in aerosols.
3. The classical assumption [1] that nitrate is only present in large particles and hence has no impact on the radiative forcing of the earth is not correct.
4. The IPCC value is a global average value and the measured values in chapter 2 are dependent on local aerosol concentrations.

ad 1. The results of recently started research on the organic compounds present in aerosol particles will provide more information about the first point, the contribution of organic aerosols. The only conclusion which can be drawn at the moment is that the influence is probably much larger than formerly estimated. If the contribution of sulfate and nitrate are known, a first order estimates of the maximum contribution of organic compounds can be made as the difference of total reflection minus the contributions of sulfate and nitrate.

ad 2. Much more information is available on point 2, errors in the measurement of ammonium nitrate by filter methods and in size distributions, as obtained by means of impactor measurements. The classical methods used for sampling of aerosols, based on the use of filters or filter packs, can lead to artifacts of losses of semi-volatile components. This has been already known for a long time and has been well documented [5,6,8,9]. These artifacts are caused by topological reactions, reactions of different species which are brought into contact with each other on the sampling filter or by evaporation of compounds such as ammonium nitrate. An overview of the most important reactions leading to errors in sampling of ammonium, sulfur and nitrogen species in aerosol are given below. The following topological reactions will influence the speciation of sampled sulfate:

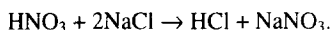


Also neutralization by ammonia can influence the speciation of sulfur compounds:

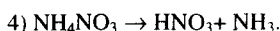


So sulfate is in principle retained in the sample but nitrate and chloride can disappear in the form of hydrochloric acid and nitric acid.

Artifacts during sampling of nitrate lead to losses as described by reaction 1) and 2).

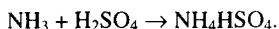


But losses caused by dissociation are also very important if ammonium nitrate is present in aerosol, as is the case at many locations:



Dissociation followed by loss in sampling procedures of nitrate and ammonium is the consequence. Dissociation is also primarily observed at longer sampling periods as well as during changes of air composition during sampling. Formation of NH_4NO_3 from HNO_3 and NH_3 can also occur but has very seldom been observed at ECN.

Artifacts while sampling ammonium are partly caused by dissociation of NH_4NO_3 according to reaction (4) as well as formation on filter:



In general, the investigations into these artifacts have led to the following conclusions:

- As long as the sampled gases and aerosols are in equilibrium with each other and with aerosol already sampled on the filter, artifacts are generally avoided. This means that if sampling periods are very short, a few hours or less, the results are much better compared to values obtained by long sampling periods (24 hours or more).
- High loading of the filter increases the possibilities of topological reactions and hence losses of nitrate and chloride after conversion to nitric acid and hydrochloric acid.
- Especially at low ambient concentrations errors by contamination become problematic. In order to avoid these errors the tendency exists to sample for longer periods and so obtain higher loadings on the filters.
- For many areas of investigations, e.g. the radiative forcing of aerosols, it is necessary to measure aerosol composition with a reasonable time resolution. The 24 hour average is not relevant but the concentration of compounds in aerosol must be known during the time that solar radiation is present.

In short, with the standard filter methods researchers are caught between two evils, blanks and sampling artifacts. In most cases people have preferred higher sampling loadings. This approach leads to unpredictable underestimates of nitrate and ammonium [6]. In Figures 1 and 2 the results of an

intercomparison with denuder methods (as explained later) and filterpacks (as applied on a large scale by US EPA) are given.

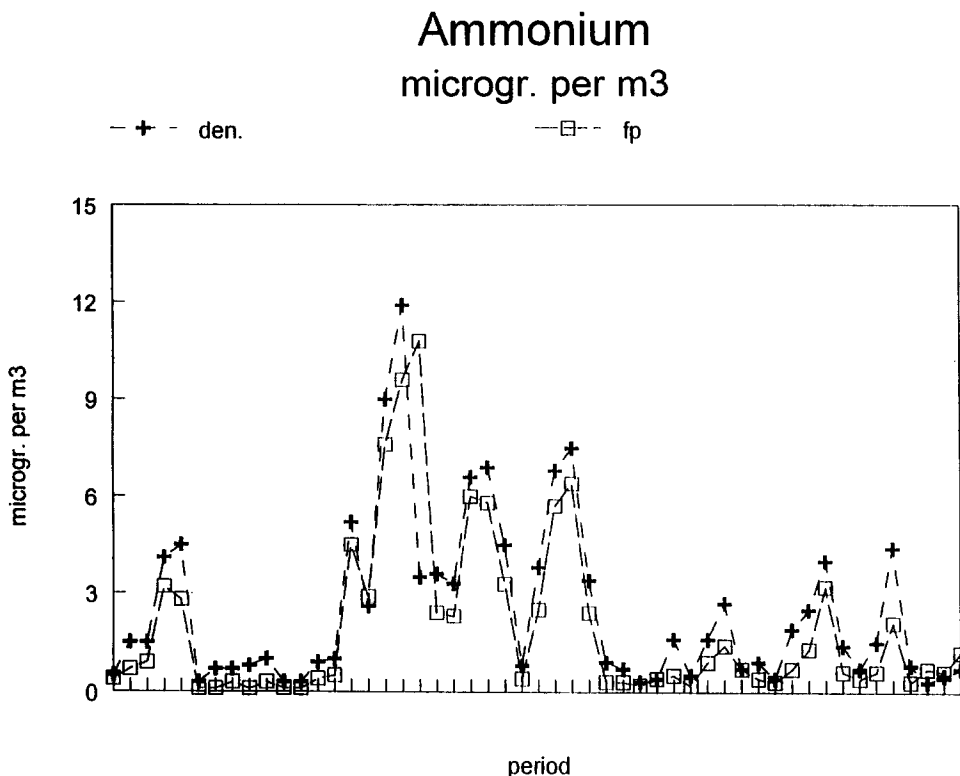


Figure 1: Intercomparison between denuder and filters for sampling of ammonium

The measurements were performed under relatively good conditions for filter measurements: The sampling period was about three hours only and the load on the filters was quite low, compared with e.g. high-volume sampling methods. Losses of ammonium and nitrate can be observed. The losses of ammonium are less striking, partially because ammonium concentrations in the Netherlands are relatively high. The losses in nitrate have a spurious character but are very often in the order of 50% or higher. This is also reflected in the results of intercomparisons for nitric acid by means of filterpacks and denuders. In this type of filterpack [10] the first filter retains particulate nitrate and the second filter samples nitric acid which either was present in the ambient air or has been liberated due to topological reactions or dissociation of ammonia present on the first filter. The intercomparison between results obtained by denuders and filterpacks in Figure 3 makes clear that the filterpack measures much higher nitric acid concentrations, caused by artifacts while measuring particulate nitrate.

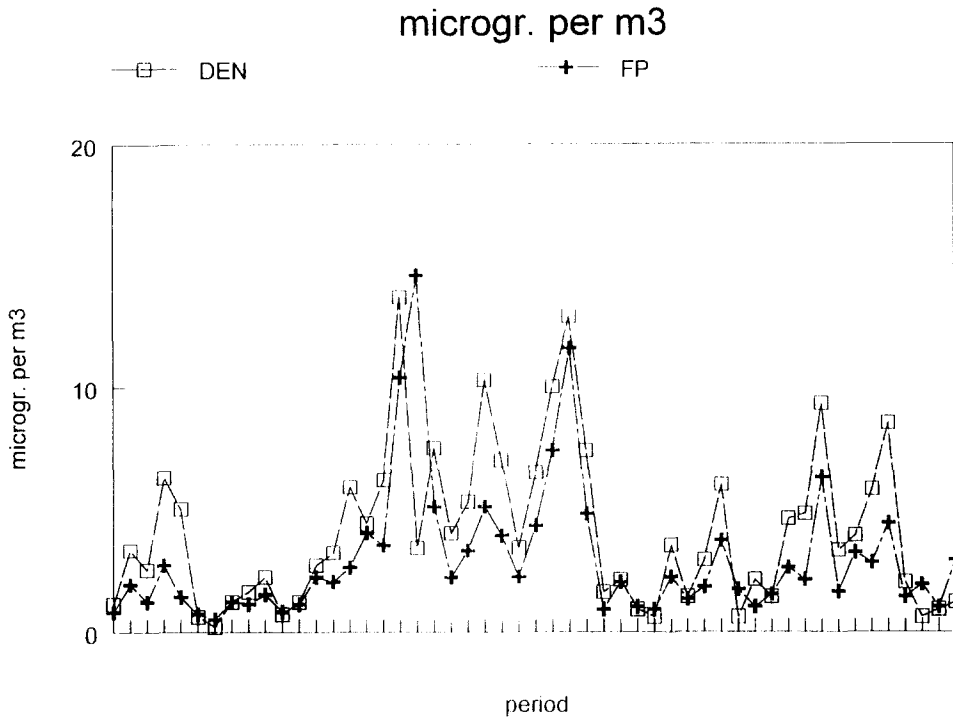


Figure 2: Intercomparison between denuder and filters for the sampling of nitrate

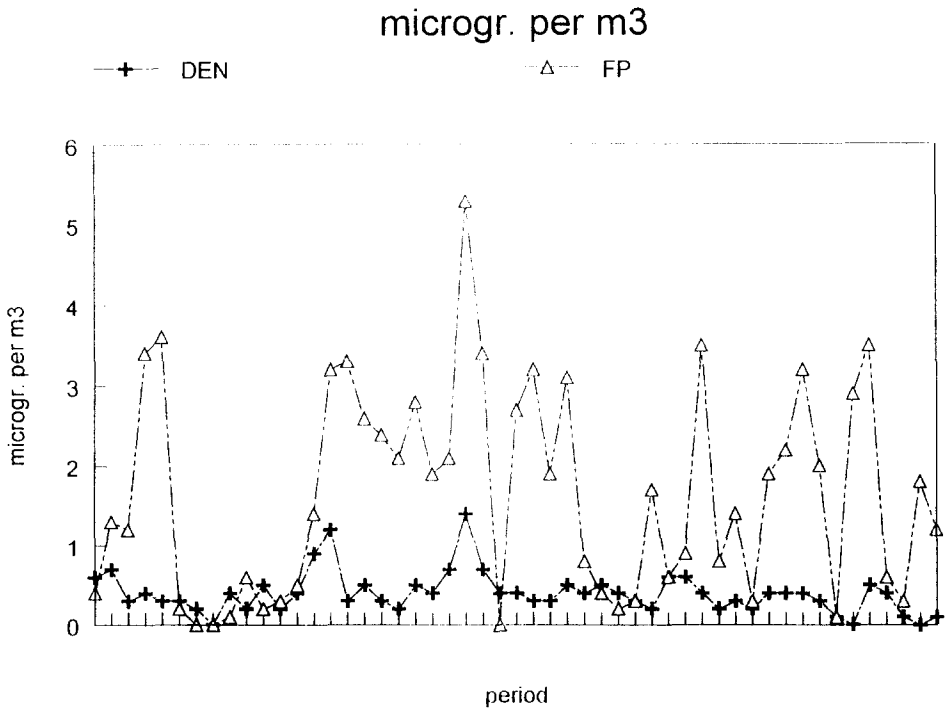
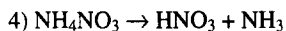


Figure 3: Intercomparison between denuder and filterpack measurements for nitric acid

ad 3. The dissociation of ammonium nitrate according to:



plays an important role in discussions about size distributions of nitrate. A good example is the latest IPCC report, where it is flatly stated that nitrate is only present in large particles and hence has no impact on the radiative forcing of the earth [1]. The concentration of nitrate in particles as a function of particle diameter has been measured using impactors. At longer sampling periods (and generally impactors are applied for sampling periods for a number of hours) dissociation of ammonium nitrate occurs in many cases. The fact that impactors operate with lowered pressure, in order to generate the required air speed for separating particles in size fractions, also promotes decomposition. It has been shown [5,11] that the pressure drop in impactors together with the instability of sampled nitrate, especially ammonium nitrate, the predominant species, leads to losses of nitrate on the plates, which correspond to smaller particle sizes and accumulates in the first stages. This phenomenon has led to the misconception that nitrate is only present in large particles. Arguments that the adiabatic temperature drop in impactors would stabilize volatile components are refuted by experimental evidence obtained at ECN and other institutes [12]. No temperature drop was measured on the plates of an Berner impactor (which was also used in this experiment) and this can be easily understood as the heat capacity of the impactor is very large compared with the possible cooling by these adiabatic processes.

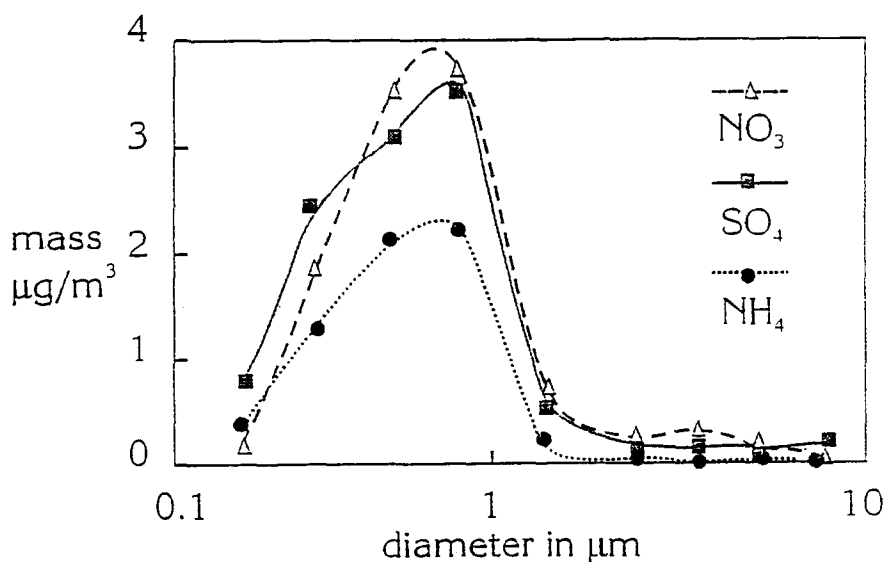


Figure 4: Size distribution for sulfate and nitrate in particles in the Netherlands

Furthermore, many measurements of the chemical composition of aerosols have been carried out near the shores of continents. Large sea-salt particles will accumulate nitrate as gaseous nitric acid is scavenged

by these particles. So if measurements are made of more or less pure sea salt aerosols, a large fraction of the nitrate is indeed found in larger particles. Measurements in the Netherlands, using the best available impactor technique with very short sampling times (30 minutes) to minimize losses, have shown that in Western Europe the same size distribution for nitrate as for sulfate is found, unless background air coming from the mid-Atlantic is sampled. A typical size spectrum for sulfate, nitrate and ammonium under condition that continental aerosol is present in the Netherlands is given in Figure 4 [11] showing that only small differences in size distribution exists for sulfate, nitrate and ammonium.

If the measurements are repeated under conditions that only "sea-salt" aerosol is present and that very low concentrations of pollutants are encountered a different size distribution is obtained, see Figure 5. Nitrate and sulfate can in particular conditions be present on large particles. The reason is that nitric acid and sulfuric acid, formed by photochemical reactions over the oceans, are scavenged on large sea-salt particles, and in this case the already mentioned processes of the displacement of chloride by nitric and sulfuric acid has caused a clear shift in the size distribution for both nitrate as well as sulfate. A large part of the mass of sulfate as well as nitrate is present in particles which are too large to effectively backscatter solar radiation.

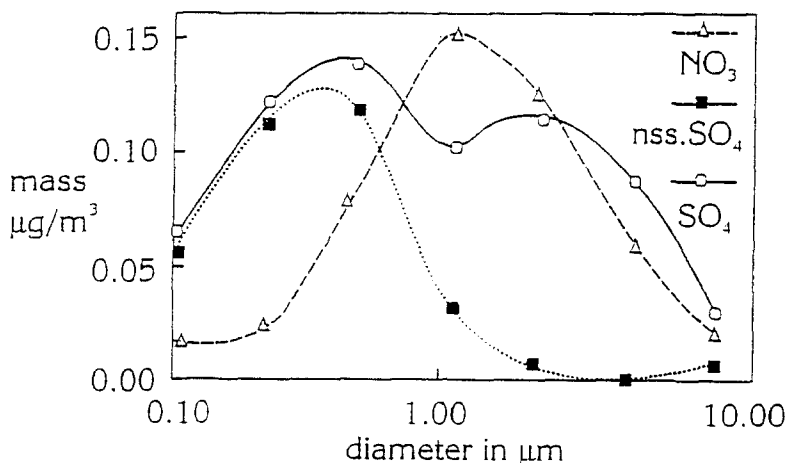


Figure 5: Size distribution for sulfate and nitrate in "sea-salt" particles in the Netherlands

It is clear that new sampling methods for the determination of the composition aerosols as function of size are urgently needed. The available methods either induce artifacts (e.g. impactors) or their sampling flow is insufficient to be used for chemical analysis (e.g. the electrical mobility analyzers). Experiments at ECN indicated that the electrical mobility analyzers does not induce artifacts by dissociation of nitrate, but its sampling flow of 1 l/min is not suitable to obtain chemical composition as function of size distribution in a reasonable time resolution, based on the present analytical methodology for sulfate, nitrate etc. Recent

measurements at other locations in Europe as well as the US make clear that much of the nitrate found over the continents is present in small particles, with very much the same size distribution as compared to sulfate.

ad 4. The contribution of sulfate to the direct and indirect effects in the Netherlands is less than half of the sum of sulfate, nitrate and organic compounds in aerosol. If the IPCC methodology, based on mass of sulfate per m^3 would be followed, a forcing of about 4 watt per m^2 would have been found for the sum of indirect and direct effect and about 1.5 watt for the direct effect only. This extrapolation can obviously not be made as the ratio of sulfate/nitrate and organic compounds will be highly variable. It is difficult to extrapolate the results in the Netherlands to global estimates due to lack of reliable measurements of the composition and size distribution of aerosols.

RECENT DEVELOPMENTS IN CHEMICAL ANALYSIS OF AEROSOL, THE SJAC SAMPLER

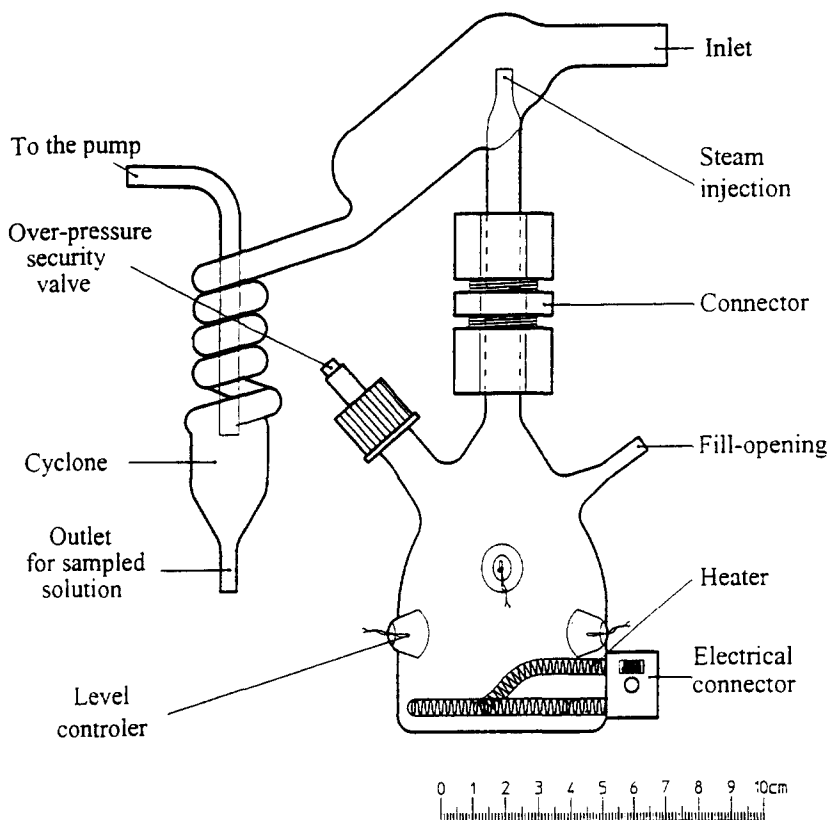


Figure 6: Steam-Jet Aerosol Collector (SJAC)

A general scheme of the Steam-Jet Aerosol Collector (SJAC) [12] is given in Figure 6. An air stream freed of water-soluble gases by passage through a wet denuder, is rapidly mixed with steam inside the mixing reservoir. The resulting high supersaturation causes aerosol particles to grow rapidly (within 0.1 sec) into droplets of at least 2 μm diameter. These droplets containing dissolved aerosol species are then collected by two cyclones in series with the cut-off diameters of less than 2 μm in diameter. The solution collected in the cyclones is constantly pumped out with a peristaltic pump and can be on- or off-line analyzed by means of ion chromatography (IC) or flow-injection analysis (FIA). Air is drawn into the system by a pump equipped with a critical orifice for 30 l/min. The actual flow rate into the sampler is 22.5 l/min because of the additional steam flow and thermal expansion.

The sampling efficiency of the sampler was determined with a back-up filter and with a DMPS (Differential Mobility Particle Sizer, TSI Model 3932). Ammonium sulfate aerosol was used as a model aerosol for these experiments. The system is capable of sampling particles quantitatively down to a few nanometers in diameter. The mass sampling efficiency of the instrument was found to be at least 99%. The detection limit of the sampler for ammonium, sulfate, nitrate and chloride ions is around 10 ng/m^3 . During measurements the sampler proved to be stable, working without any assistance for extended periods of time.

THE IMPLICATIONS OF RECENT MEASUREMENTS

Measurements of bulk compounds in aerosols in the Netherlands with this technique lead to the conclusion that the mass of nitrate in aerosol is about the same as sulfate or even slightly higher [3]. Dasgupta has applied a sampling and analysis methodology which is quite similar to the here described apparatus, to analysis of ambient aerosols in Lubbock, Texas. His results indicate the same situation, the concentration of nitrate in aerosol are higher than the sulfate concentrations at this location in the US [14]. Measurements using a SJAC sampler, carried out in South-Portugal in the framework of the ACE-2 project (Figure 7) lead again to the conclusion that even in these more remote areas considerable amounts of nitrate are present in aerosol.

Since a very limited sets of measurements, not influenced by the artifacts described here, is available, it is not possible to draw general conclusions with certainty. It seems probable though, that nitrate concentrations in aerosol are considerably higher than described in the literature. It is important to keep in mind that the most of reported values have been obtained by so-called high volume samplers. In this technique aerosol is sampled at a rate of hundreds of cube meters per hour for sampling periods of 24 hours and longer. Under these conditions large errors due to evaporation and topological reactions can be expected as function of high loadings as well as long sampling times.

The few measurements of nitrate as function of size distribution in aerosols, which have been carried out under conditions to minimize artifacts, indicate that the size distribution is different compared to results reported in the literature. This has already been pointed out above and illustrated in Figure 4. The results of impactor measurement during very short sampling periods, in order to minimize artifacts caused by

evaporation and topological reactions, indicate that the same size distribution is found for nitrate as for sulfate.

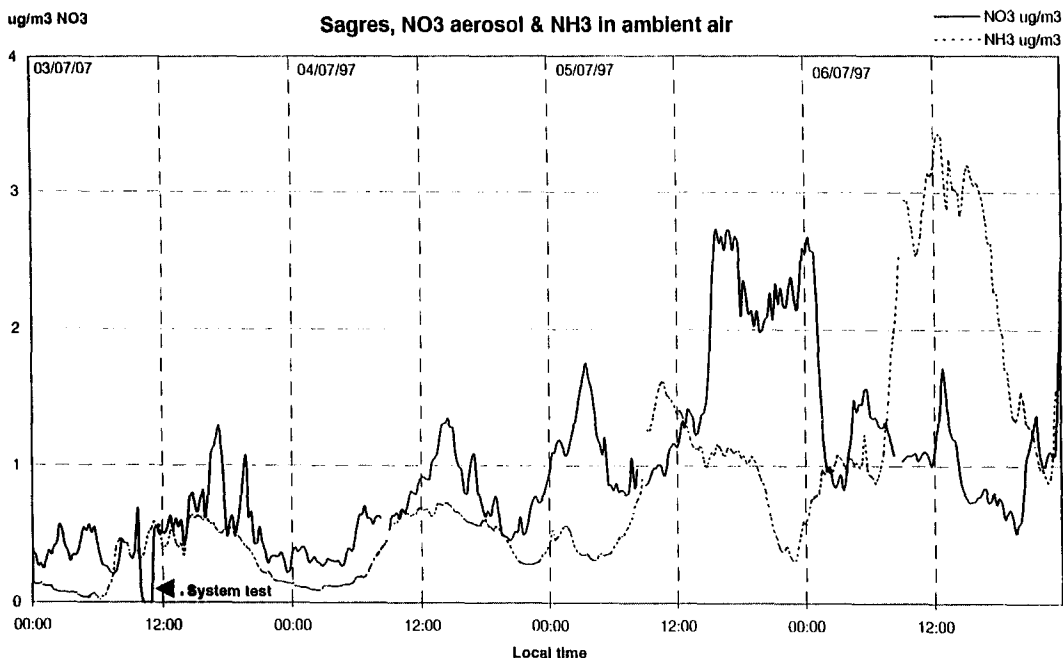


Figure 7: Measurement of nitrate in South-Portugal

ENVIRONMENTAL IMPLICATIONS OF THE RESULTS

One of the most spectacular consequences of the application of methods which avoid or minimize sampling artifacts in the analysis of semi-volatile components like ammonium and nitrate are found in the assessment of the influence of aerosols on the radiative balance. The same is probably true in general for the volatile organic compounds. As pointed out above, the size distribution of nitrate in aerosol is very often quite similar to the size distribution of sulfate. That means that one can doubt whether the conclusion of IPCC that nitrate does not play a role in the radiative balance, as it is primarily present in large particles, is still valid and tenable. For the Netherlands and surroundings the situation is quite clear as pointed out by ten Brink: The impact of nitrate is even larger than sulfate as ambient nitrate concentrations on a molar basis are higher than the sulfate concentrations. Furthermore, the high ammonium nitrate concentrations lead to stronger growth of aerosol particles due to uptake of water even at lower relative humidities and this phenomenon also contributes to the large direct effect of aerosols on the radiative balance of 4 watt m^{-2} . The contribution of nitrate is at least one third so amounting to more than 1.5 watt m^{-2} . It was also observed that organic compounds in aerosol are responsible for a contribution of about 30 %.

The same situation was found for the indirect effect: The observation that the size distribution is the same as encountered for sulfate and that the ambient concentrations are even higher, leads to the conclusion that nitrate is responsible for at least one third of the indirect effect of aerosols which is estimated to be 6 watt m^{-2} . Here the contribution of nitrate amounts to 2 watt m^{-2} and again the contribution of polar organic compounds could be in the same order of magnitude. An overview of the radiative forcing by greenhouse gases and aerosols is given in Figure 8. The relative contribution of the nitrate, ammonium, sulfate and organic substances to the composition of aerosols is given in Figure 9.

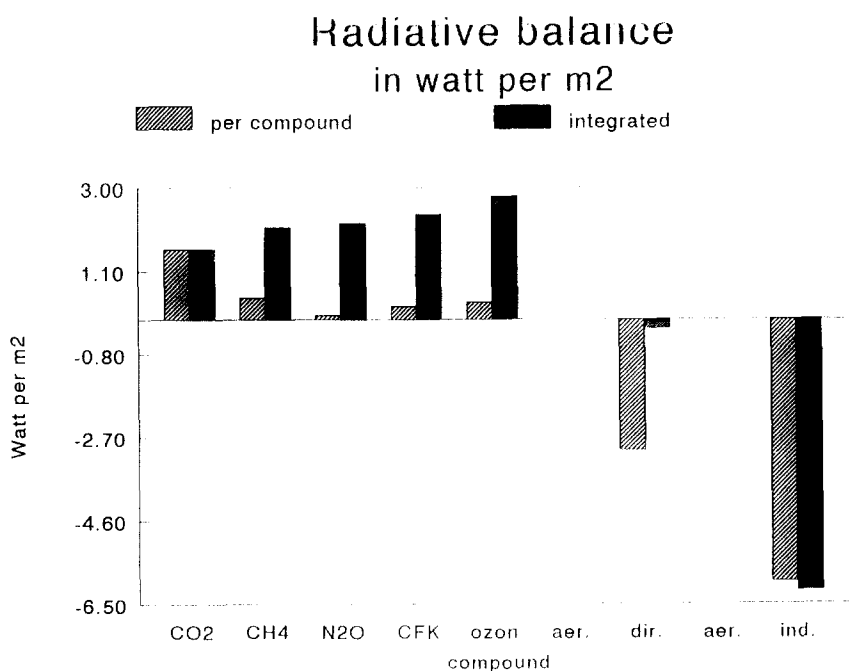


Figure 8: Overview of radiative forcing as assessed at the location of ECN

Aerosol forcing tends to show a quite different pattern compared to the forcing by greenhouse gases. In some regions the effects of aerosols may regionally compensate or overcompensate for the effects of greenhouse gases. Regional impacts of greenhouse warming may be delayed for several decades by aerosol effects. If this is so, the adaptation to climate change in these regions could be delayed as well. Furthermore, if near-term impacts of warming are expected to be lessened by aerosol effects, then there would be less political impetus for actions to mitigate long-lived greenhouse gases. Numerous predictions have been made that the climatic effect of greenhouse gases can be clearly detected in the next decade or so and if these climatic change do not occur, possibly under the influence of the forcing by aerosols, it would be difficult to persuade politicians that measures are needed. In fact this "compensation-phenomenon" could be quite environmentally dangerous. There are probably compelling reasons to take measures for the abatement of

aerosol concentrations in the future. The impact of aerosols on health, acid deposition and eutrophication will lead to measures for emission reduction and these measures could have severe consequences for the radiative balance. IPCC has based its estimates on the radiative forcing of aerosols on sulfate only and it seems fairly certain at this moment that this procedure seriously underestimates the radiative forcing of aerosols. If recent assessments of global temperatures are correct, a temperature rise has occurred in recent decades. As models have been tuned to trends in greenhouse gas concentrations in combination with temperature trends and a probably too low aerosol forcing, this means that models underestimate the climatic change and temperature rise for conditions of doubling of CO₂.

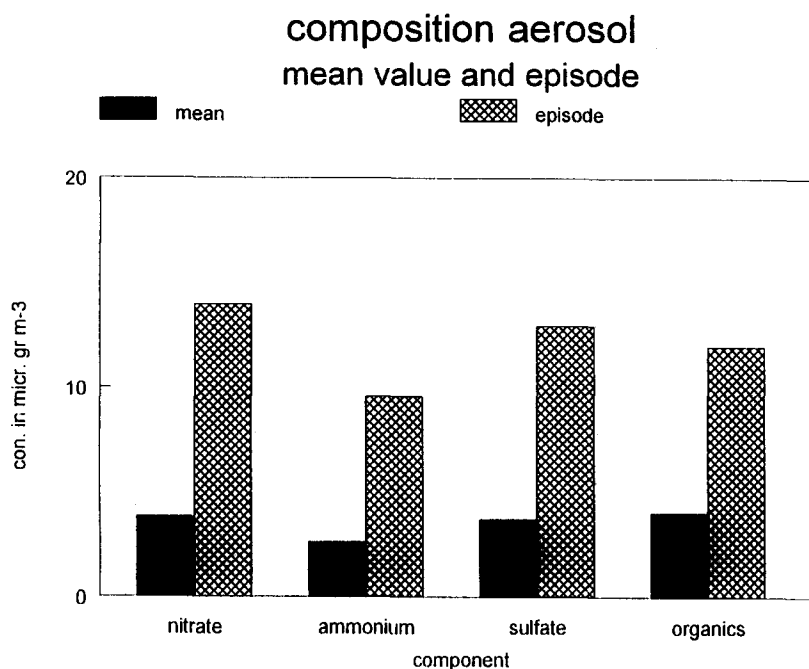


Figure 9: The relative contribution of nitrate, ammonium, sulfate and organic substances to the composition of aerosols

The products of degradation processes in the atmosphere, both of inorganic as well as organic nature, must be evaluated with care, as it is becoming clear that the formation of aerosols, as a result of these processes, could have very important environmental implications.

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