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Aerosol physico-chemical characteristics over a boreal forest determined by volatility analysis

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A thermal volatility technique was used in the boreal forest environment to examine accumulation mode (0.05–0.35 µm radius) physico-chemical properties as a function of air mass origin. Three primary aerosol species were identified in all air masses: (1) a semi-volatile organic component, (2) ammonium sulphate, and (3) a non-volatile component thought to comprise mostly of soot carbon. Under some conditions, sulphuric acid was also identified, as was sea salt. Following nucleation and growth of new particles into accumulation mode sizes, the organic fraction of accumulation mode aerosol, by mass, was observed to increase from 30%, prior to and during the nucleation event, up to 75% by the end of the growth period, indicating a substantial fraction of organic mass condensing onto newly formed particles.

Introduction

Aerosols are assumed to have a very important influence on the climate system and could possibly negate the effect of man-made greenhouse gases on the radiative balance of the atmosphere. To understand and quantify the direct (Charlson and Wigley 1994) and indirect (Twomey 1974) radiative impact of aerosols, one has to identify the source regions of new anthropogenic and natural aerosols and follow their growth, or evolution, into accumulation mode particles where they can efficiently scatter incoming solar radiation and, by acting as cloud condensation nuclei, modify cloud radiative properties. Before the impact of anthropogenic particles on climate forcing can be addressed correctly, the importance and contribution of natural aerosols must be quantified. In particular, important source regions of biogenic aerosols must be identified. Since natural emissions of non-methane hydrocarbons (NMHC) are believed to be an order of magnitude higher than anthropogenic NMHC emissions (Guenther *et al.* 1995) and are likely to contribute significantly to aerosol formation and growth, it is crucially important to understand processes leading to natural organic aerosol formation.

Over the last years a few different source regions for natural aerosol formation have been investigated: for example, the polar marine boundary layer (Wiedensohler *et al.* 1996, O'Dowd *et al.* 1997), the free troposphere (Clarke 1992), boreal forests (Mäkelä *et al.* 1997, Leaitch *et al.* 1999), and the coastal environment (O'Dowd *et al.* 1998). For the available data, it appears that two important natural sources of boundary layer aerosols are over the boreal forests, where nucleation occurs over large spatial scales (Mäkelä *et al.* 1997), and in the coastal zone, where extraordinary large nucleation rates are encountered (O'Dowd *et al.* 1999). In acknowledgement of the importance of boreal forests as a significant source of biogenic aerosols, the forested areas of Finland have provided the focus for a dedicated nucleation field experiment, comprising a consortium of European scientists aimed at elucidating the factors controlling the formation and growth of biogenic aerosols in the form of the BIOFOR experiment (Biogenic formation of atmospheric particles over a boreal forest, Kulmala *et al.* 2000).

Within the framework of this dedicated nucleation experiment, two intensive field campaigns were conducted in the Spring and Summer of 1998, and a third experiment in the Spring of 1999. The primary objectives of these experiments were to examine the role of biogenic VOC emissions in the formation and growth of particles and the effect of these species on the hygroscopic and cloud formation properties of these biogenic aerosols. In this study, we present initial results, using a volatility technique for rapid online aerosol characterisation, illustrating the physico-chemical nature of accumulation mode aerosols under different background conditions and trajectories, and compare these characteristics to those observed under a typical aerosol nucleation and growth event during the BIOFOR intensive campaigns.

Experiments and data analysis

The experimental measurement site was located at the SMEAR II station (Station for Measuring forest Ecosystem-Atmosphere Relations), Hyytiälä, southern Finland (61°51´N, 24°17´E), in a boreal forest. For a more detailed description of the forest vegetation and the site surroundings *see* Kulmala *et al.* (2000).

For the on-line characterisation of aerosol physico-chemical properties, a thermal analytic technique, comprising a coupled OPC (ASASP-X) and a fast response scanning volatility system, was deployed (O'Dowd *et al.* 1997). Before the aerosol size distribution is sized and counted by the particle counter, the aerosol is heated in scanning cycles between 30 °C and 700–800 °C every 15 minutes. From the thermal response of the aerosol number and the change in the size distribution, conclusions can be drawn about the chemical composition and the state of mixing of aerosols as a function of size. The ASASP-X measures the dry aerosol size spectrum between 0.05 µm and 1.5 µm radius, since the condensed water is evaporated after drying both in the volatility system as well as in the optical chamber in the optical particle counter. However, this drying is thought to have a negligible influence on the partitioning of volatile species. Several calibration experiments have been performed with laboratory-generated aerosols, which are commonly present in the atmosphere (Jennings and O'Dowd 1990). For example, some of the primary inorganic aerosols species posses' distinct temperature profiles, with sulphuric acid volatilisating at 100 °C, ammonium sulphate at 200 °C, sea salt at 650∞C, and soot carbon at 800 °C (O'Dowd *et al.* 1997, Jennings *et al.* 1994). Particular organic aerosol species are somewhat more difficult to differentiate, since there are often multiple organic species contributing to mass in an organic-rich aerosol population, and, furthermore, unlike most inorganic aerosol species, some VOC aerosol species do not necessary posses a step response (Becker *et al.* 1999). For internally mixed species, each species will volatilise at its characteristic volatilisation temperature leading to a dual step response for that particle size range. This effect is illustrated in O'Dowd *et al.* (1997). Additionally, laboratory studies indicate that the presence of organics does not noticeably change the volatilisation temperature of the sulphate aerosol (C. O'Dowd unpubl.).

For more information on the broader size distributions of ultra-fine, fine, and accumulation modes, a series of DMPS systems (Differential Mobility Particle System) were installed, sampling from 2, 18 and 67-m heights. However, only data from the 18 m level were included in this study.

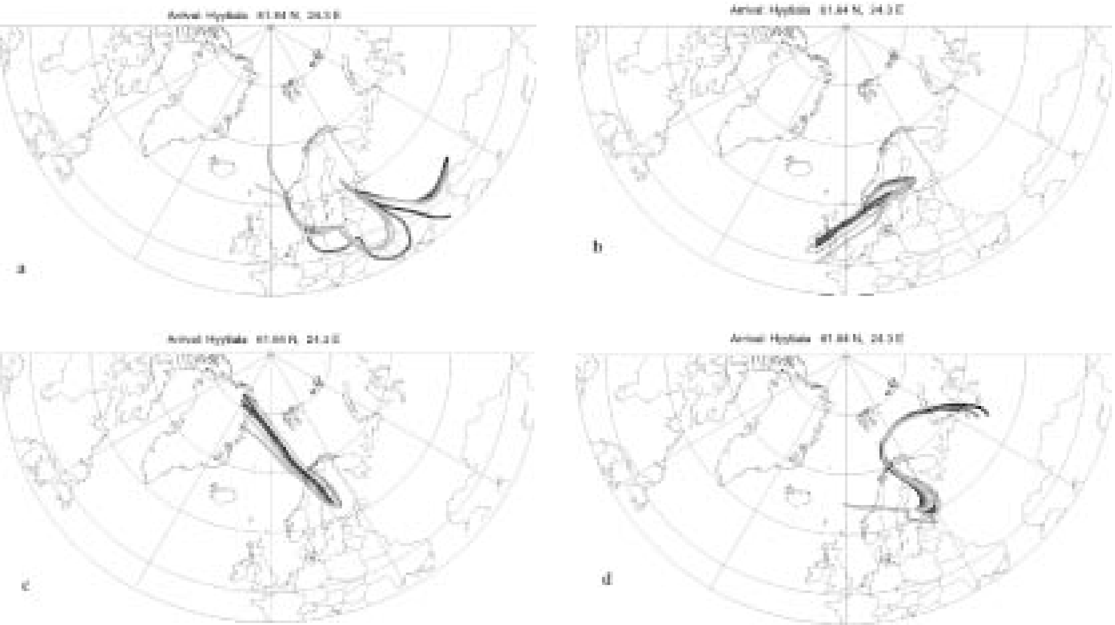

Fig. 1. Location map and typical air mass back trajectories at the Hyytiälä boreal forest SMEAR II Research Station, Southern Finland. (a) southerly back-trajectories (b) westerly back trajectories (c) northerly back-trajectories and (d) north-easterly back-trajectories.

The DMPS is capable of measuring the size distribution of the aerosols between 0.0015 µm and 0.250 µm radius with a temporal resolution of ten minutes. The DMPS installed at 2 m is an essential component of the continuous measurement program running at Hyytiälä while the 18 and 67 m installations ran only for the duration of the campaigns. All DMPS measurements used sheath flows with no additional driers etc., thus, only de-humidification processes occurring in the sample tubes reduce the particle size and the aerosols are measured close to ambient conditions.

Results

Accumulation-mode aerosol physico-chemical properties were segregated into four categories based upon air mass back trajectories. Four primary trajectory categories were identified: (1) south to southeast wind direction with trajectories from central to east Europe and primarily continental origin over the previous 5 days; (2) west to southwest with trajectories advecting in from the Atlantic and Baltic sea, but also containing passage over land; (3) air coming from the north to northwest with primarily a polar/Arctic maritime history; and (4) wind direction from the northeast also with Polar/Arctic characteristic although with a shorter fetch to the ocean. A map illustrating the location of the field measurement location and the four main (5-day) back trajectory classifications is shown in Fig. 1.

Air masses from southeast and south

One typical example day, Julian Day (JD) 120 1998, is chosen to highlight characteristics associated with this sector in which air masses have advected over polluted areas in mid-central European latitudes. For this case, accumulation mode number concentration was approximately 1600 cm^{-3}, while black carbon mass, measured by the Aethalometer, was between 800–1000 ng m^{-3}, indicating the polluted nature of the air mass.

The dry size accumulation mode number and volume spectral shapes for this case are illustrated in Fig. 2a and Fig. 3a, respectively, along with the change in spectral shape as a function of tem-

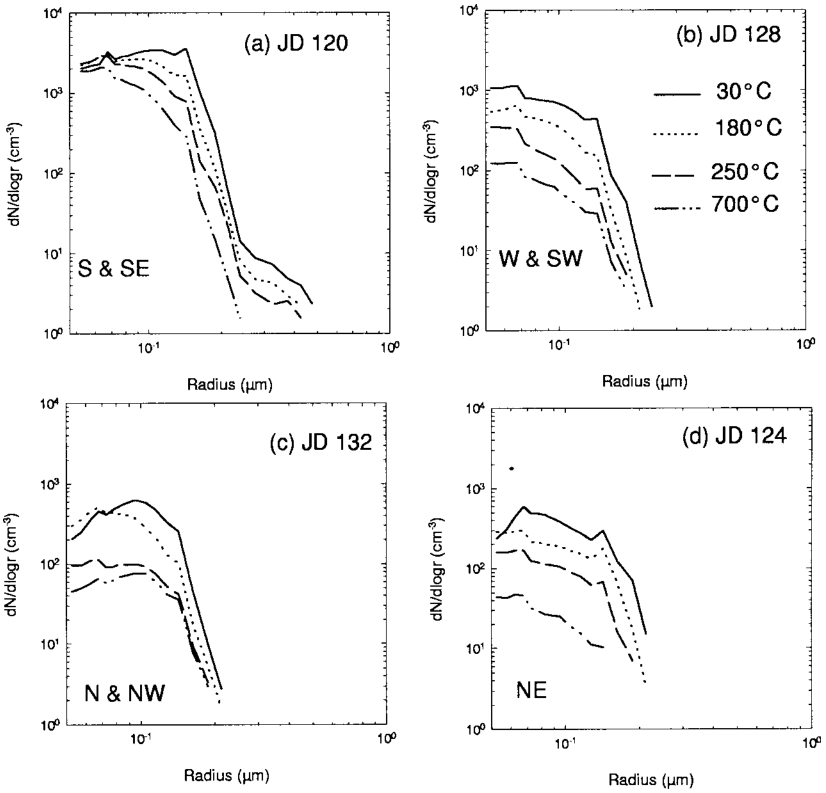

Fig. 2. Volatility number concentration spectra for aerosol occurring under different wind sector conditions. (a) south and southeast sector, (b) west and southwest wind sector, (c) north and northwest sector, and (d) north-east sector. Sizes distributions are taken at 30 °C, 180 °C, 250 °C and 700–800 °C.

perature. The shape of the distribution at dry ambient temperatures indicated a peak at approximately 0.15 μm above which the slope of the distribution falled off rapidly. A relative local minimum was also seen at about 0.05 μm. After heating the aerosol to 180 °C, a large loss in concentration, and thus mass, was observed for sizes between 0.07 and 0.2 μm, indicating a substantial fraction of quite volatile aerosol mass, possibly composed of sulphuric acid and/or organic material. On further heating to 250 °C, only a small fraction of the aerosol was volatilised over the same size range, indicating the presence of ammonium sulphate. At temperatures of 700 °C, particulate mass was lost over all sizes, leaving a noticeable residual aerosol component, typical of the elemental component of soot carbon.

Examination of the volatility temperature profiles reveals more information on the nature of aerosol volatile properties. The temperature profiles associated with this case, JD 120, are illustrated in Fig. 4. The 32 size channels of the ASASP-X were regrouped into three ranges covering sizes 0.05–0.08 μm; 0.08–0.12 μm; and 0.12–0.35 μm. It should be noted that for typically inorganic aerosol, a sharp step response is seen at specific temperatures corresponding to volatilisation of specific inorganic aerosol species. For example, a sharp response is seen at 100 °C for sulphuric acid, 200 °C for ammonium sulphate and 650 °C for sea salt (O'Dowd *et al.* 1997). Laboratory temperature profiles, for ammonium sulphate and cis-pinoic acid are also shown in one of the sub-plots in Fig. 4 for comparison with field data. The clear differences between the sharp step response of inorganic ammonium sulphate compared to the gradual response of the organic acid is evident. Pinic acid behaves in a similar manner to cis-pinoic acid but is not shown for clarity in the diagram.

The temperature profiles in this case indicated that the largest thermal response occurred for the biggest particles of size 0.12–0.35 μm. For temperatures of 30–180 °C a gradual reduction in concentration was seen in this size range as the temperature increased, indicating the dominating presence of semi-volatile organic carbon aerosol (SVOC) (Becker *et al.* 1999). If a noticeable con-

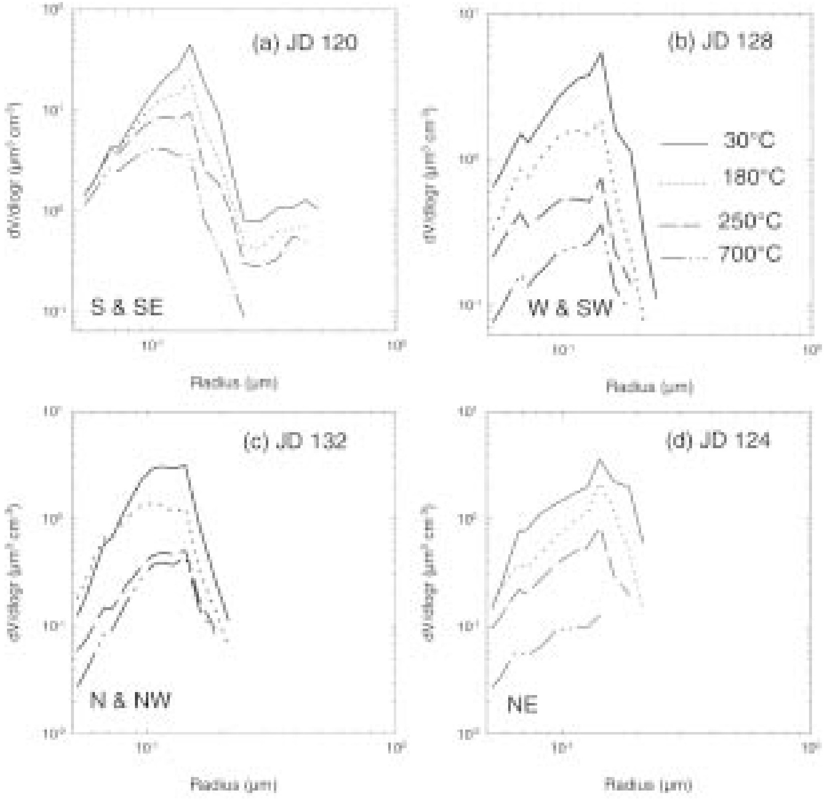

Fig. 3. Volatility volume concentration spectra for aerosol occurring under different wind-sector conditions. (a) south and southeast sector, (b) west and southwest wind sector, (c) north and northwest sector and (d) northeast sector. Sizes distributions are taken at 30 °C, 180 °C, 250 °C and 700–800 °C.

tribution from sulphuric acid had been present here, it would have been evident as a significant step response at 100 °C, however there was no such evidence. A small step function was seen at 200 °C indicating a small fraction of ammonium sulphate. A less noticeable thermal response was seen in the 0.08–0.12 µm size range, while very little change was seen at temperatures below 200 °C for the smallest sizes between 0.05–0.08 µm. In fact, at approximately 200–250 °C, a small increase was seen in the smallest size range, indicating the presence of a slightly internally-mixed aerosol as larger particles propagate into these size ranges once some of their semi-volatile mass is volatilised.

The temperature profiles indicated three aerosol groups: (I) semi-volatile organic aerosol (SVOC), (II) ammonium sulphate (AMS), and (III) non-volatile carbon component comprising the elemental carbon and highly polymerised component of soot carbon. It should be noted that in this category, there may also have been secondary organic non-volatile matter present, however, it seems unlikely that significant contributions of such secondary organic aerosol could survive temperatures of 300 °C or higher. It is possible that some of the residual aerosol remaining at the higher temperatures may have been a non-soot species, the thermal response behaved in a similar manner to soot carbon as described in previous studies (Jennings *et al.* 1994, Smith and O'Dowd 1996). Additionally, sulphuric acid has a volatilisation temperature in the SVOC volatilisation range and, when a noticeable step response is observed at 100 °C, then the contribution of sulphuric acid to this component must be considered as probably significant.

The relative contribution from each group can be derived from the loss of aerosol mass as a function of temperature. The organic fraction contribution is defined as the loss of mass between 30 °C and 180 °C when a step response at 100 °C is absent, while the ammonium sulphate contribution is defined as the loss of mass between 180 °C and 250 °C. The gradual reduction in all sizes between 250 °C and 700 °C is thought to represent the loss of non-volatile carbonaceous (NVC) component of soot carbon, while the residual remaining at 700–

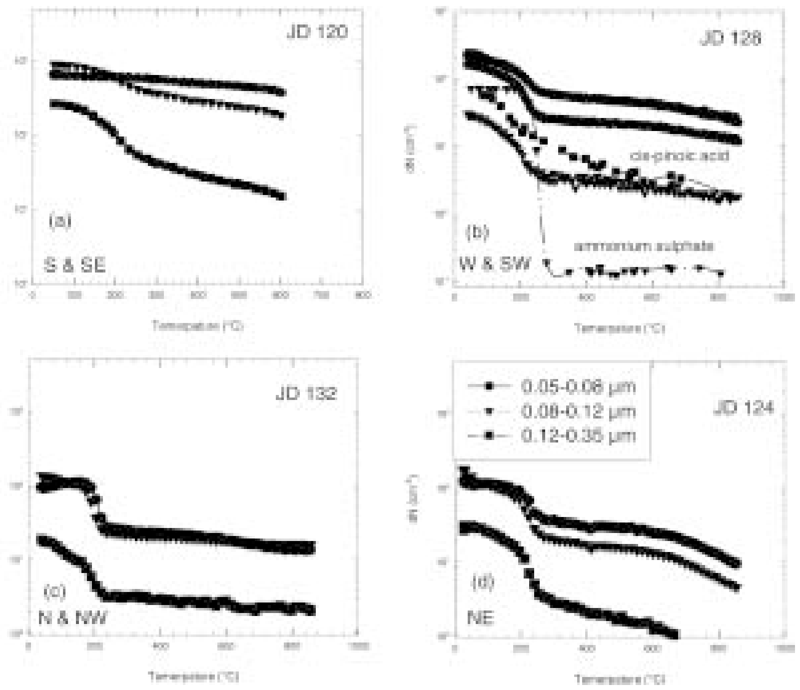

Fig. 4. Volatility temperature profiles for aerosols occurring under different wind sector conditions. (a) south and southeast sector, (b) west and southwest wind sector, (c) north and northwest sector, and (d) northeast sector. Laboratory-generated ammonium sulphate and cis-pinoic acid are shown along side field data in (b).

800 °C is thought to represent the elemental carbon component of soot carbon (EC) (Jennings *et al.* 1996, Smith and O'Dowd 1996). In Fig. 5, the aerosol was speciated in terms of concentration of SVOC, ammonium sulphate, NVC and EC, however, it should be noted that NVC and EC carbon is expected to be internally mixed and hence, the breakdown by number into separate classes is not valid. In terms of number, the total contributions from NVC and EC carbon should be treated as the soot carbon component of the aerosol. For this case, the change in spectral shape as a function of temperature indicated only partial internal mixes, indicating that we can estimate the concentration of each species for these air masses by number. In terms of number concentration, approximately 500 particles cm^{-3} behaved like SVOC aerosol, while 480 particles cm^{-3} behaved like ammonium sulphate and 620 cm^{-3} behave like soot carbon. In terms of volume, this corresponds to 3.7 µm^3 cm^{-3}, 1.8 and 3.2 µm^3 cm^{-3}, respectively, for SVOC, AMS and soot carbon.

Air masses from west to southwest

Air masses from the southwest sector typically had traveled over the North Sea and the Baltic Sea for a number of days and were considerably maritime in nature. They had also advected over southern Sweden or Denmark and consequently can, at best, be described as modified maritime air masses. Julian Day 128 is taken as representative of aerosol characteristics in this sector.

For JD 128, the total accumulation mode particle concentration was 379 cm^{-3} while soot carbon mass was of the order of 200 ng m^{-3}. The number and volumetric spectral changes as a function of temperature are illustrated in Figs. 2b and 3b, respectively, where a significant reduction in aerosol concentration at all sizes was seen for heating to 30 °C, 180 °C, 250 °C and 700 °C. The peak concentration at ambient concentrations was seen at sizes of 0.05–0.07 µm, indicating a low mode radius by comparison to the more polluted case shown previously. The spectral shape did not change with increasing temperature for this air mass type and the aerosol in all sizes displayed similar temperature profiles, indicating similar aerosol composition as a function of size. All sizes exhibited a characteristic semi-volatile organic component at temperatures below 180 °C and a significant ammonium sulphate component at 180–250 °C. It should be noted, however, that in

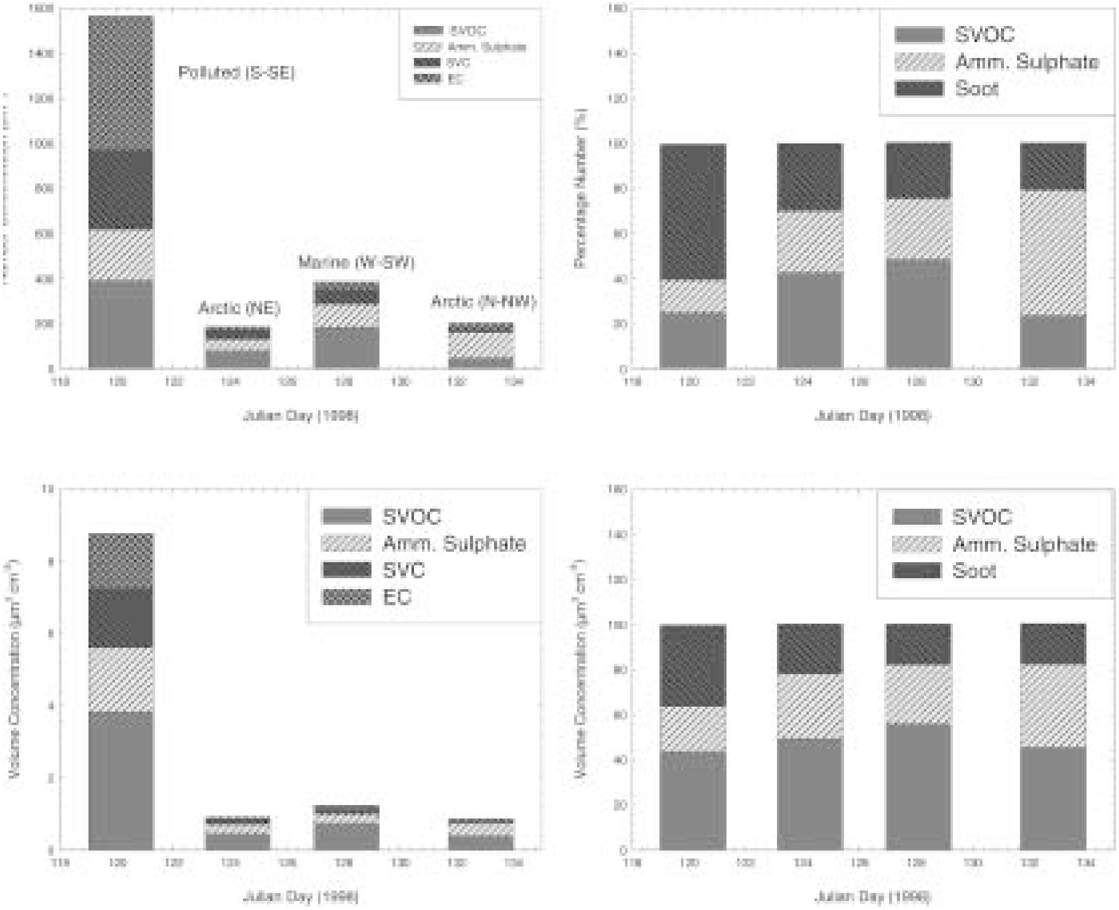

Fig. 5. (a) top left — estimated net contribution of SVOC, ammonium sulphate, and soot carbon (i.e. combined NVC and EC) to number concentrations observed under different air masses-wind sectors (b) top right — percentage contribution of SVOC, ammonium sulphate and soot carbon to total aerosol number concentration observed under different air masses. (c) bottom left — estimated net contribution of SVOC, ammonium sulphate, and soot carbon (i.e. combined NVC and EC) to volumetric concentrations observed under different air masses-wind sectors. (c) bottom right — percentage contribution of SVOC, ammonium sulphate and soot carbon to total aerosol volumetric concentration observed under different air masses. Julian Day for the selected cases is given on the x-axis.

the smallest size range between 0.05–0.08 µm, there was a slight step function at 100 °C, which suggests the presence of a small amount of sulphuric acid in this aerosol.

Relative to the total concentration, a small residue, typical of soot carbon, was seen between 250 °C and 800 °C. For this case, the lack of change in spectral shape with temperature suggests an external mixture. In terms of contributions from each species, SVOC was estimated to account for 49% (185 cm^{-3}) by concentration and 56% (0.75 $µm^3\ cm^{-3}$) by volume, while AMS was estimated to account for 26% (102 cm^{-3}) by concentration and 26% (0.25 $µm^3\ cm^{-3}$) by volume, and soot carbon 25% (100 cm^{-3}) by concentration and 18% (0.13 $µm^3\ cm^{-3}$) by volume.

Aerosol air masses from the north to northwest

Aerosols coming from the northwest were from Arctic regions and were therefore likely to be quite clean and also maritime in nature, although air Arctic air trajectories require advection over Norway and Sweden before arriving at the measurement station. Generally, the physico-chemical aerosol characteristics observed were quite similar to those observed during west-to-southwest trajectories, however, regularly a second category of thermal responses was observed under this wind sector. One typical example of aerosol characteristics in such air mass is illustrated for JD 132 where a total accumulation mode number concentration of 201 particles cm^{-3} was encountered.

For JD 132, dry aerosol size distributions (Figs. 2c and 3c) indicated a peak concentration occurring in the size range 0.1 µm. After being subjected to heating to 180 °C, not only did a reduction in concentration occur, but also there was a reduction in size, indicating an internally-mixed aerosol. A clear mode was evident indicating, possibly, a significant degree of cloud processing (O'Dowd *et al.* 1999), possibly leading to significant internal mixing. A small residue remained after heating to 800 °C, relative to the initial concentration, and, in comparison to the previous cases, the residual size distribution indicated also a more pronounced mode at sizes around 0.1 µm rather than 0.05 µm or less in the previous examples.

Examination of the temperature profiles indicated more pronounced step-responses to thermal conditioning in this case. At 100 °C, a step response was more evident for the two larger size ranges (i.e. 0.08–0.12 µm and 0.12–0.35 µm) leading to a reduction in particle concentration, while a step increase was seen in the smallest size range of 0.05–0.08 µm. Again, this behavior is indicative of internal mixing in the aerosol. This more defined response around 100 °C suggests a greater presence of sulphuric acid present in the aerosol and suggests that all the volatile mass, or concentration, over this temperature range can not be solely regarded as SVOC.

A second clear step response was evident at 200 °C, indicating the presence of ammonium sulphate in all sizes. The gradient in the concentration reduction between 250 °C and 800 °C was less steep than in previous cases. A slight step response at 600 °C was also evident, indicating that a significant fraction of the residual mass could be sea salt. This was corroborated by the shape of the residual distribution indicating a residual mode radius of the order of 0.1 µm. Additionally, soot carbon mass was amongst the lowest level observed during the campaign with concentrations of the order of 50 ng m^{-3} for this case.

Using the same criteria for aerosol speciation, we found that the more volatile fraction comprised 24% (49 cm^{-3}) of the number concentration and 46% (0.4 µm^3 cm^{-3}) of the volume concentration, while the ammonium sulphate fraction comprised 55% (113 cm^{-3}) of number and 36% (0.33 µm^3 cm^{-3}) of volume. It should be noted that we cannot strictly call the more volatile fraction SVOC since there was evidence of sulphuric acid present, so this fraction should be viewed as a combined SVOC and sulphuric acid contribution. Also, due to clear internal mixing, speciation by number is not necessarily valid in this case.

The residual fraction comprised 21% (42 cm^{-3}) of the number concentration and 18% (0.15 µm^3 cm^{-3}) of the volume concentration. Again, the presence of sea salt in the residual aerosol indicates that the residual cannot be regarded as solely soot carbon. In fact, from the change in spectral shape at the higher temperatures, combined with the temperature profiles, suggests that the residual aerosol was predominantly sea salt for this case.

Air mass origin from northeast

An example of air masses from the northeast is taken for Julian day 124, with the aerosol having a modified arctic behavior, as the originally clean arctic air passed over parts of northeast Russia and then Finland. The particular case chosen indicated a peak mode radius around 0.065 µm (Fig. 2d and 3d), while the lack of change in spectral shape with temperature indicated similar composition as a function of size along with being an externally mixed aerosol. The residual spectra at 250 °C and the spectral shape of the aerosol remaining at 800 °C suggests that this residual aerosol was predominantly soot carbon, although some sea salt also appears to have been present from the temperature profiles (Fig. 4d). The presence of soot carbon is corroborated by Aethalometer concentrations of the order of 100–200 ng m^{-3} for this case. The temperature profiles between 30–180 °C indicate SVOC while a significant fraction of ammonium sulphate was also present.

By concentration and volume, SOVC accounted for 43% (81 cm^{-3}) and 49% (0.46 µm^3 cm^{-3}), respectively, while ammonium sulphate

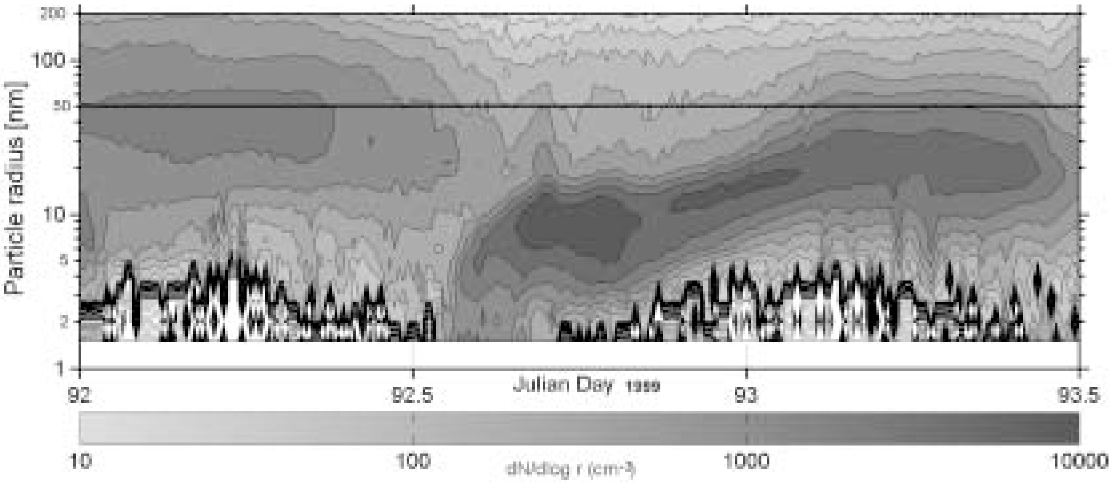

Fig. 6. DMPS spectra for one nucleation and growth event. The solid black line highlights the lower measurement limit of the volatility system.

accounted for 27% (51 cm^{-3}) and 28% (0.36 µm^3 cm^{-3}), respectively. The residual aerosol accounted for 30% (55 cm^{-3}) of the concentration and 22% (0.2 µm^3 cm^{-3}) of the volume.

Discussion

In summary, it has been difficult to distinctly and accurately differentiate between physico-chemical aerosol characteristics in all air masses arriving at Hyytiälä as a function of air mass origin, although clear differences were observed in terms of concentrations of identified species.

The most noticeable difference was the higher contribution of soot carbon to the aerosol for polluted continental air. On some occasions, sea salt was detected in the temperature response curves for marine air. In general, in all air masses, a significant fraction of the aerosol comprised a semi-volatile component, and a significant fraction of both ammonium sulphate and soot carbon. The relative contributions of each primary species, as a function of air mass origin, is displayed in Fig. 5. For all air masses, the semi-volatile organic component generally contributed the dominant fraction of aerosol mass, with ammonium sulphate typically contributing a similar, or a smaller amount. While the volatility technique can identify the presence of semi-volatile organic aerosol mass, it can not identify the actual chemical composition of this organic fraction. It should be noted also that under certain conditions, sulphuric acid may also contribute to this component, and if it contributes in a similar magnitude, it can be difficult to distinguish from organics. Laboratory studies on the thermal response of terpene derivatives such as cis-pinonic acid and pinic acid suggest that these compounds do not posses a step function response as observed for some inorganic acids such as sulphuric acid (Becker *et al.* 1999). Also, it is likely that multiple organic species will contribute to the aerosol mass, thus further making it difficult to distinguish between individual species. Nevertheless, the thermal response observed at temperatures between 30–180 °C were similar to those observed for cis-pinonic acid and pinic acid, suggesting the presence of these species in all air masses arriving at Hyytiälä. Given that these are major VOC oxidant products from biogenic forest emissions, this may not be too unlikely.

Organic contribution to accumulation mode mass during post-nucleation aerosol growth

Nucleation of new particles is quite regularly observed at the SMEAR II station in Finland. Typically these events occur in polar or Arctic air masses under northwesterly airflow and very clean conditions and result in growth to accumulation mode sizes (Kulmala *et al.* 2000).

Figure 6 shows an example of a typical nu-

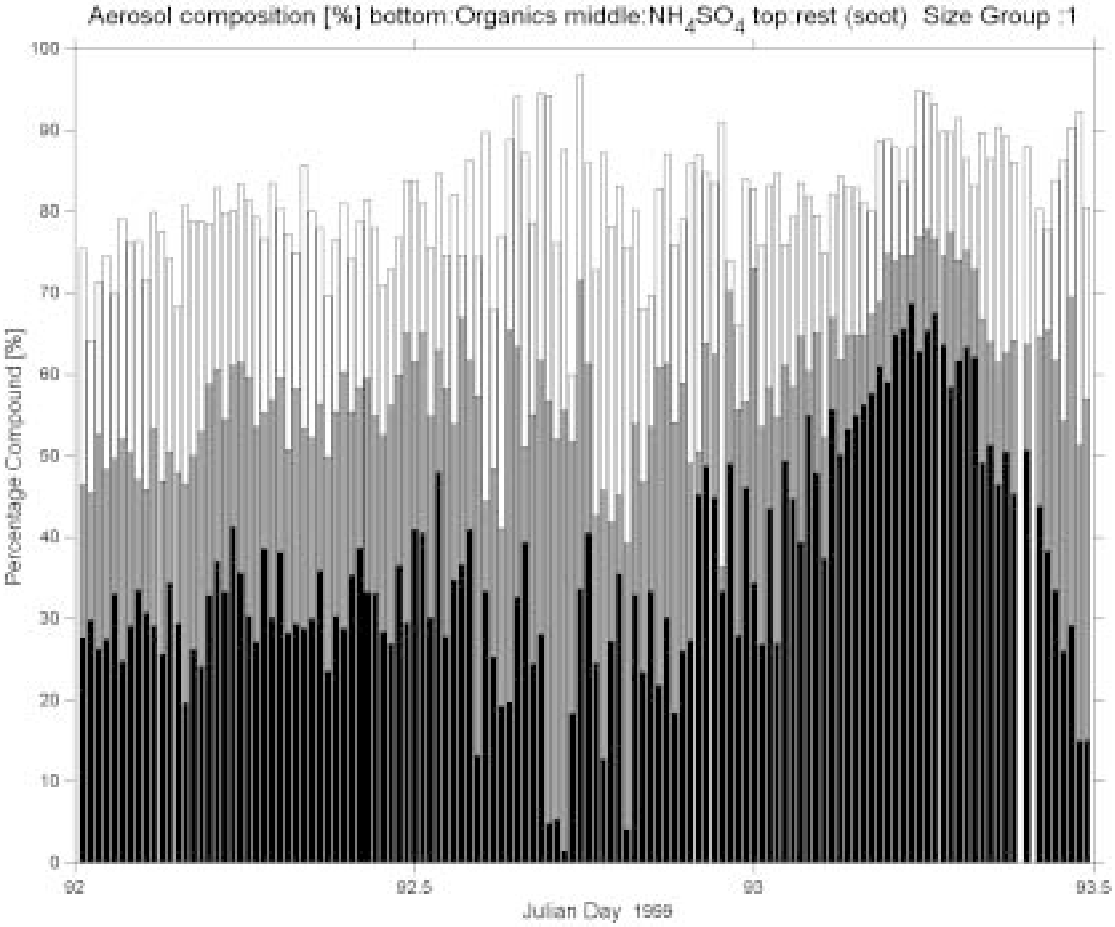

Fig. 7. Volatility-derived organic aerosol (bottom shade), sulphate aerosol (middle shade) and soot carbon aerosol (top shade) during the nucleation and growth period shown in Fig. 6.

cleation event, occurring on 2 April 1999. The size spectrum from 1.5 nm up to 250 nm, taken from the DMPS spectrometer, is shown throughout the day and into the following day. Prior to the nucleation event just after noon, a reduction in pre-existing aerosol concentration, due to dilution was seen. Just after the nucleation event, recently formed particles started to appear at sizes around 1.5 nm and proceeded to grow rapidly into sizes larger than 5 nm. After approximately 18 hours of continuous growth, a significant fraction of these particles had grown into sizes of 50 nm and greater where they can be detected by the volatility system. Taking a wind speed of 5 m s^{-1}, the spatial scale of this event was of the order of 300 km. In Fig. 7, the relative contributions of semi-volatile organic aerosol, ammonium sulphate aerosol and soot is illustrated as a function of time. Prior to and during the nucleation event, a similar contribution of each species was seen (approximately 30% of each species). However, as the recently formed aerosol grew into the accumulation mode, the organic fraction of aerosol masses dramatically increased to approximately 75%, thus indicating a very significant organic contribution to aerosol mass during the condensation growth processes following such nucleation events. The increase in semi-volatile organic aerosol occurred in all particle sizes as would be expected from condensation across the size distribution. However, the largest increase occurred for the smallest particles growing into the accumulation mode, suggesting that this organic material contributed significantly to the growth particle mode mass.

Conclusion

A thermal volatility technique was used in the boreal forest environment to examine accumulation mode aerosol physico-chemical properties as a function of air mass origin. Three dominant aerosol species were identified in all air masses: (1) a semi-volatile organic component, (2) ammonium sulphate, and (3) soot carbon. Under some conditions, sulphuric acid was also identified, as was sea salt. Due to the multiple contributions from different aerosol species, and the fact that semi-volatile organic aerosol and soot carbon posses multi-valued thermal responses, it can be difficult at times to clearly distinguish between each species using this technique. However, for specific cases, species differentiation was possible. In general, semi-volatile organic aerosol comprised between 20% and 50% of aerosol number concentration, and 45% and 55% by volume. Ammonium sulphate comprised between 15% (most polluted case) and 60% (cleanest case) by number, and 20% and 40% by volume. Soot carbon comprised 60% by number in the most polluted case and 20% in the cleanest (although the sample here was significantly influenced also by sea salt). By volume, these fractions ranged from 25% to 15% respectively. During a nucleation and growth event, the (SVOC) organic fraction of the aerosol was observed to increase from 30% prior to and during the nucleation event, and up to 75% of the total mass after the recently formed particles had grown into accumulation mode sizes, indicating a substantial fraction of organic mass condensing onto newly formed particles.

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