

Effects of air masses and synoptic weather on aerosol formation in the continental boundary layer

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ABSTRACT

Nucleation of near nm sized aerosol particles and subsequent growth to ~ 100 nm in 1–2 days has in recent years been frequently observed in the continental boundary layer at several European locations. In 1998–99, this was the focus of the BIOFOR experiment in Hyytiälä in the boreal Finnish forest. Nucleation occurred in arctic and to some extent in polar air masses, with a preference for maritime air in transition to continental air masses, and never in subtropical air. The air masses originated north of the BIOFOR experiment by paths from the southwest to northeast sector. The nucleation was also associated with cold air advection behind cold fronts, never warm air advection. This may relate to low pre-existing aerosol concentration, low cloudiness and large diurnal amplitudes in the continental boundary layer associated with cold air advection and clear skies. Arctic and polar air together with cold air advection did not always lead to nucleation. The most important limiting meteorological factors were cold front passages and high cloudiness, probably through reduced photochemistry and wet scavenging of precursor gases and new aerosol particles. The preference for nucleation to occur in arctic air masses, which seldom form in the summer, suggests a meteorological explanation for the annual cycle of nucleation, which has a minimum in summer. The connection to cold-air outbreaks suggests that the maximum in nucleation events during spring and autumn may be explained by the larger latitudinal temperature gradients and higher cyclone activity at that time of the year. Nucleation was observed on the same days over large parts (1000-km distance) of the same air mass. This suggests that the aerosol nucleation spans from the microphysical scale to the synoptic scale, perhaps connected through boundary layer and mesoscale processes.

1. Introduction

Atmospheric aerosol particles have the ability to influence the atmospheric radiation budget, and hence the climate, through the back scatter of solar light back to space, or by acting as cloud condensation nucleus (CCN) and thereby determining the cloud lifetime (Albrecht, 1989) and albedo (Twomey, 1974; Boucher and Lohman,

1995). This indirect effect is the most uncertain part of anthropogenic climate forcing (Intergovernmental Panel of Climate Change, 1996).

The number of CCNs can increase if already existing less hygroscopic particles are made more hygroscopic by condensation. However, the most powerful way to increase the CCN population is by nucleation of new particles (~ 1 nm in diameter) and growth of these to CCN size (~ 100 nm). Until a few years ago, this was only rarely observed. The reason was originally an inability to detect small enough particles, but as instruments improved, the reason was probably the lack

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of long-term measurements and that much effort was confined to the marine boundary layer (Covert et al., 1992). This turned out to be the wrong place to look for frequent aerosol nucleation. In 1996, continuous measurements of the aerosol size spectra started in Finland. This was to change the picture entirely. At present we know that nucleation, or strictly speaking the appearance of ultra-fine particles detected at a few nm, and subsequent growth to ~ 100 nm in 1–2 days, is a frequent phenomena in the continental boundary layer. The observations span from sub-arctic Lapland, over the remote boreal forest (Mäkelä et al., 1997; Kulmala et al., 1998b), suburban Helsinki (Väkevää et al., 2000), industrialized agricultural regions in Germany (Birmilli and Wiedensohler, 2000), a mountain site in southern Germany (Birmilli et al., 2000) to rural United Kingdom (Coe et al., 2000).

In 1998–99 a large research project, BIOFOR (Biological Aerosol Formation in the Boreal Forest, [HYPERLINK http://mist.helsinki.fi/projects/Bioforhomepage](http://mist.helsinki.fi/projects/Bioforhomepage) <http://mist.helsinki.fi/Biofor/index.html>) focused on this problem. BIOFOR took place in Hyytiälä ($61^{\circ}51'N$ $24^{\circ}17'E$) in the boreal forest of central southern Finland, (Fig. 1), and included three intense field campaigns. BIOFOR 1 from 11 April to 22 May in 1998, BIOFOR 2 from 17 July to 29 August in 1998, and BIOFOR 3 from 11 March to 30 April in 1999. Hyytiälä is where the continuous aerosol size spectra measurements started at the Finnish SMEAR II station (Station for Measuring forest Ecosystem–Atmospheric Relations). The continuous long term monitoring of meteorological, chemical and biological properties at this site allow us to place BIOFOR into the context of annual cycles and inter annual variability (Vesala et al., 1998). For an overview of BIOFOR, see Kulmala et al. (2001b).

The best understood path for atmospheric nucleation is binary homogeneous nucleation by H_2SO_4 and H_2O (Kulmala et al., 1998a), which was also the favorite candidate in several years. However, it has become clear that in many cases, this nucleation path is unable to explain observed nucleation. Several routes to enhance the nucleation have been suggested. (1) Nucleation around ions is energetically more favorable than homogeneous nucleation and could enhance nucleation (Turco et al., 1998). (2) A 3rd molecule could participate. When BIOFOR was planned, the



Fig. 1. Map of Finland with its neighbors. A = Tahkuse, B = Jokioinen, C = Hyytiälä, D = Halli, E = Tikkakoski and F = Värriö.

hypothesis was that oxidation products of monoterpenes or other organic compounds emitted from the forest caused the observed aerosol formation. However, Buzorius et al. (2001) found no connection between the photosynthetic activity of the forest and the aerosol formation and therefore no support for biogenic emissions of precursor gases for nucleation. Furthermore, Janson et al. (2001) was unable to find any support for a connection between nucleation and the oxidation products from monoterpenes by reaction with OH, O_3 or NO_3 . Instead, the NH_3 concentration together

with the H_2SO_4 source term showed some correlation with the maximum number concentration during particle events. This suggests that ternary nucleation of H_2O , NH_3 and H_2SO_4 caused the aerosol nucleation (Korhonen et al., 1999; Kulmala et al., 2000). Model calculations within BIOFOR arrives with similar conclusions (Kulmala et al., 2001a). (3) Nucleation is extremely sensitive to temperature and to the vapor pressure of the participating gases. A small change to higher vapor pressure and lower temperature has a strong positive non-linear effect on the nucleation rate. Therefore, temporal fluctuations as from turbulent and convective eddies (Easter and Peters, 1994) or atmospheric waves (Nilsson et al., 2000), or spatial gradients such as inversions (Nilsson and Kulmala, 1998) could enhance the nucleation by many orders of magnitude.

BIOFOR 1 and 3 were intended to match the spring maximum in nucleation events and BIOFOR 2 was to take place during the summer minimum. Already the frequency of nucleation events, on the average once per week (Kulmala et al., 1998b), which is close to the synoptic time scale, suggests a linkage to synoptic weather systems. We will evaluate the BIOFOR data with a focus on what types of air masses favor nucleation in the continental boundary layer and in what context of synoptic circulation the nucleation occurs. Furthermore, we will try to find plausible reasons for why these air masses and weather situations favor nucleation.

2. Data

2.1. Weather maps, satellite images and air mass classification

A large number of weather maps from the European Meteorological Bulletin (German Weather Service) and Berliner Wetterkarte (Institute for Meteorology, Free University of Berlin) and NOAA-14 satellite images in the visible and infrared channels were collected. These were utilized to achieve a consistent overview of the day-to-day synoptic weather patterns during the BIOFOR campaigns and to keep track of air mass movements and the positions of fronts and their cloud systems. We have used the air mass classification and surface front analysis made daily at Berliner Wetterkarte for 00 UTC (02 local Finnish

wintertime). The air mass classification recognizes arctic, polar and sub-tropical air masses, each divided into marine and continental air and transition cases in between. To everybody's confusion, this traditional definition says that arctic air masses form at the pole, while polar air masses form south of the arctic air masses.

From the daily weather maps we have established the types of air masses that were located over the experimental site, the presence of fronts, their passages over Hyytiälä and if there were conditions of warm or cold air advection behind cold fronts, especially so called cold air outbreaks. In addition, we have located the latitudinal approximate surface position of the arctic front (the border between arctic and polar air) and the polar front (the border between polar and sub-tropical air) along the 25° east longitude line, which passes close to Hyytiälä. Sondes from Hyytiälä, Jokioinen and Tikkakoski have also been used to confirm the air mass classification and front identification.

2.2. Radiosoundings

Radiosoundings were performed in order to follow changes in the synoptic weather situation and to understand the boundary layer structure during the BIOFOR field campaigns. The purpose of the soundings was also to verify the numerical weather data and to study the applicability of the soundings made at Jokioinen and Tikkakoski to Hyytiälä (Fig. 1). Sondes of the type Vaisala RS80 were launched on a regular basis by the Finnish Meteorological Institute (FMI) from Jokioinen (60°49'N 23°30'E, 104 m a.s.l., 179 km southwest of Hyytiälä) at 0 and 12 UTC and from Tikkakoski (62°24'N 25°40'E, 141 m a.s.l., 93 km northeast of Hyytiälä) at 6 and 18 UTC. The ground equipment used at both sites was the Vaisala DigiCORA II with the Loran-C windfinding system. Only significant pressure levels were recorded. More than 400 sondes from Jokioinen and Tikkakoski have been analyzed for BIOFOR.

Soundings were made at Hyytiälä only during part of BIOFOR 1 (21 sondes) and most of BIOFOR 3 (40 sondes) campaigns in cooperation with the Finnish Defence Forces. Balloons were launched on an irregular basis to match significant changes in the boundary layer and to some degree in the synoptic weather on days it was believed

nucleation would occur. The optimizing of the launches was refined for BIOFOR 3 based on what we had learned during BIOFOR 1. The sounding system was a Vaisala DigiCORA with radiotheodolite windfinding. The launch height was 145 m above the sea level. We used 300 g balloons filled with H_2 resulting in an ascent rate in the range of $2\text{--}7\text{ ms}^{-1}$, averaging 5 ms^{-1} , in the lowest 5 km. Since data were stored every 2 s, this corresponds to an average vertical resolution of 10 m. Each rawinsonde was calibrated at 0% relative humidity and indoor temperature and to ambient conditions, including wind speed and wind direction, a moment before launch.

The comparison of the soundings made at Hyttiälä and Jokioinen or Tikkakoski revealed that the wind and temperature profiles were usually quite similar. The biggest differences were encountered in association with nocturnal surface inversions, passing fronts and the onset of the break up of the nocturnal inversions. The vertical profiles of humidity can, however, differ significantly between the sounding sites. In many cases, the daytime mixed layer height and especially the temperature and humidity gradients at the mixed layer top differed considerably.

2.3. Surface weather observations

Surface weather observations made at the Kuorevesi/Halli aviation weather station ($61^{\circ}51'N$, $24^{\circ}47'E$, $h = 145\text{ m a.s.l.}$), some 20 km east of Hyttiälä were obtained (Fig. 1). The data set consists of manual synoptic observations made in 3-h intervals (00, 03, 06, 09, 12, 15, 18 and 21 UTC). The observations include cloud cover, visibility, present weather, past weather, low cloud cover, ceiling and amount of precipitation (2 observations per day). The ceiling was assessed with the help of a laser ceilometer with a maximum range of $\sim 4000\text{ m}$. In addition, a data set of daily precipitation and snow cover depth observations made with standard gauges at the FMI's Hyttiälä climatological station was compiled.

2.4. Aerosol measurements

The dry aerosol number size distributions were measured with a Differential Mobility Particle Sizer (DMPS) system in 10 min cycles at heights of 2, 18 and 67 m in Hyttiälä, which gives a

continuous view of the distribution and evolution of sub micrometer aerosol particles. The DMPS system used here actually consists of two DMPS systems. The first one includes a TSI 3025 UFPCP and a Hauke-type short DMA (Differential Mobility Analyzer). It measures particles between 3 and 20 nm in dry diameter. The second one includes a TSI3010 CPC and a Hauke-type medium DMA and measures particles between 20 and 500 nm. Aalto et al. (2001) describes this system in more details, together with other aerosol physical measurements.

The aerosol formation events days were classified by Mäkelä et al. (2000) in categories according to the Hyttiälä DMPS measurements. (1) Cases that showed a clear nucleation mode that was easily distinguishable until it had grown to at least the Aitken mode. (2) There were fewer particles formed, or some background concentration existed in the smallest DMPS channels, or the growth was less nice, than for class 1. (3) Same problems as for class 2, but much worse. In this class it was difficult to see the nucleation mode at times. We will only consider class 1 and 2 as clear nucleation days in our analysis.

We will furthermore make use of measurements at the SMEAR-I station in Värriö ($67^{\circ}48'N$, $29^{\circ}30'E$, $h = 400\text{ masl}$) (Fig. 1), with a similar DMPS system, as well as measurements at the Tahkuse observatory/University of Tartu, Estonia ($58^{\circ}31'N$, $24^{\circ}56'E$, $h = 23\text{ m a.s.l.}$) of mobility spectrum of air ions of both polarities. The air ion mobility spectrum was measured by means of a tailor-made complex multichannel air ion spectrometer (Hörrak et al., 1994; Tammet, 1995) covering the mobility range of $0.00032\text{--}3.2\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ (corresponding to a range of D_p between 80 nm–0.36 nm divided into 20 intervals).

2.5. Trajectories

We have calculated 96 h long back-trajectories with the Lagrangian Gaussian long-range trajectory and dispersion model TRADOS (Pöllänen et al., 1997), which uses the numerical meteorological data from the HIRLAM weather prediction model (Gustafson, 1993). Two sets of 96-h long back-trajectories arriving at Hyttiälä were calculated. Forecast trajectories were calculated during the field campaigns in order to help decide beforehand the measurements to be made the following

day. These trajectories were computed for 3 different pressure levels (1000, 925 and 850 hPa) arriving in 6-h intervals. Later a 2nd set of trajectories with an extended meteorological data output was calculated for data interpretation and modeling work. This set consisted of trajectories arriving at Hyytiälä in 3-h intervals and to five different pressure levels (1000, 925, 850, 700 and 500 hPa). In addition a set of trajectories arriving at the Tahkuse observatory/University of Tartu, Estonia during the BIOFOR 3 campaign in 1999 was produced.

3. Results and discussion

3.1. Case study

The 1st week of April 1999, part of the 3rd campaign, was chosen for a case study to begin to place the aerosol formation into a synoptic weather context. Two cyclones, one east of the Kola Peninsula and one over the Norwegian sea, and a high pressure over Scandinavia dominated the flow over Finland in the lower troposphere (Fig. 2). During Julian day 90 (31 March), a day without nucleation, a cold front associated with the Kola Peninsula cyclone, passed Hyytiälä and the north-westerly flow brought a maritime polar air mass and caused low visibility and precipitation from a low overcast cloud cover (Fig. 3). This was the beginning of a several-day-long cold air outbreaks over Finland and East-Europe. During Julian days 91–92 (1 April and 2 April) Finland was still covered by the maritime polar air mass as the cold air outbreak continued.

On Julian day 92, the skies cleared (Fig. 3), and nucleation occurred (Fig. 4). New particles of a few nm in size appeared around noon and seemed to grow during the rest of the day and the following night. On the next day, Julian day 93 (3 April), there was an event like burst of particles around 10 nm in the afternoon, but the continuous growth was broken. The period with a larger number of 10-nm sized particles was preceded by a period with much smaller aerosol number at all sizes and interrupted by another period with smaller aerosol numbers. This was related to the passage of a second cold front, which belonged to the cyclone over the Norwegian sea. Clouds accompanied the two periods of reduced aerosol number, the second also involved reduced visibility

(Fig. 3). Clouds or rain along the cold front may have prevented nucleation by scavenging of precursor gases or already nucleated particles or by limiting photo-chemistry below the frontal clouds. The gradual temperature decrease in the troposphere during the cold air outbreak can be clearly seen in Fig. 5, especially in the lower troposphere after the second cold front on Julian day 93.

The strong cold air advection continued behind the second cold front. The new air mass type was continental or maritime arctic air in transition to form continental air. The satellite image in Fig. 2 shows the fully developed cold air outbreak on the afternoon of Julian date 94 (4 April) at 14:40 local time with the two cold fronts over East-Europe. Since the second cold front had almost caught up with the first cold front there was now only a narrow zone of polar air in between them. The new arctic air mass covered all of Scandinavia, Finland, the Baltic countries and north-eastern Russia. The 2 cold fronts were at their southernmost stretch accompanied by bands of deep convective cumulus, and further to the east stratus and nimbostratus clouds. The cold arctic air mass north of the cold front was partly cloud free and partly covered by patches of fair weather clouds. The cumulus clouds were in some parts of the air mass organized by meso-scale circulation systems into cloud cells and cloud streets visible on the satellite images, e.g., over Finland (Fig. 2). During the three-day presence of an arctic air mass (4–6 April, Julian day 94–96) aerosol particles appeared at the lower detection limit of the DMPS system, indicating nucleation of new aerosol particles, each day around noon followed by growth to larger sizes (Fig. 4). The series of nucleation events were interrupted by an occluded warm front passage late on Julian day 96 (6 April) (Fig. 5), again with overcast clouds and precipitation (Figs. 3, 4). This is the same warm front that was approaching from the Atlantic Ocean in Fig. 2. On Julian day 97 (7 April), the warm front brought with it a new maritime polar air mass and warm air advection in which no nucleation occurred.

3.2. Nucleation in arctic and polar air masses during cold air outbreaks

Fig. 6 shows the change with time of the surface front positions at 00 GMT during BIOFOR 3. By looking at the pentagrams located at the latitude

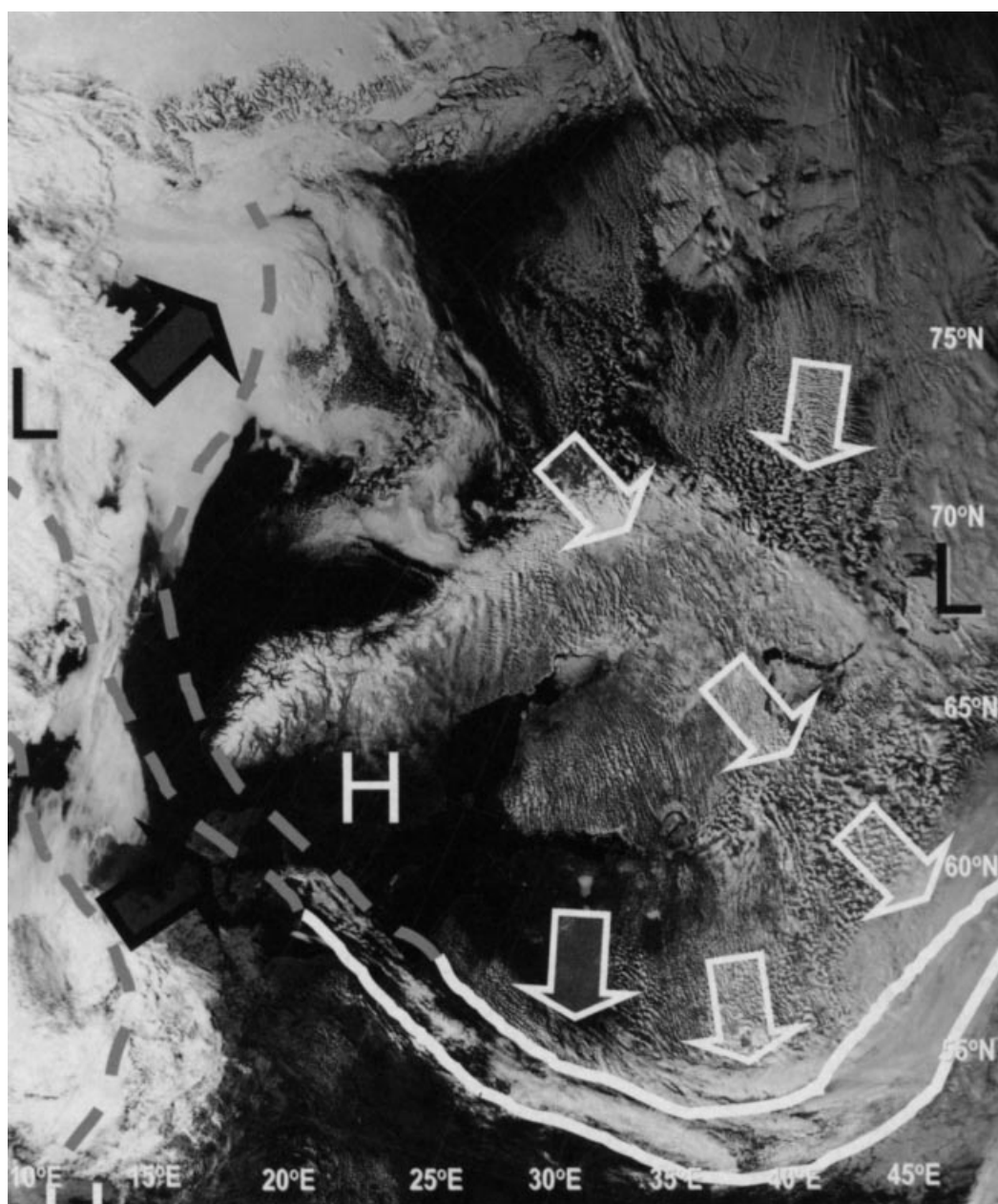


Fig. 2. NOAA-14 satellite image (from the satellite-receiving station of the University of Dundee), visible light, channel 2, from 12:40 UTC, 4 April. Cold fronts are marked with full black lines, warm fronts with dashed black lines. Cold air advection is indicated with light arrows and warm air advection with dark arrows. Positions of minimum and maximum surface pressure are marked with L and H respectively.

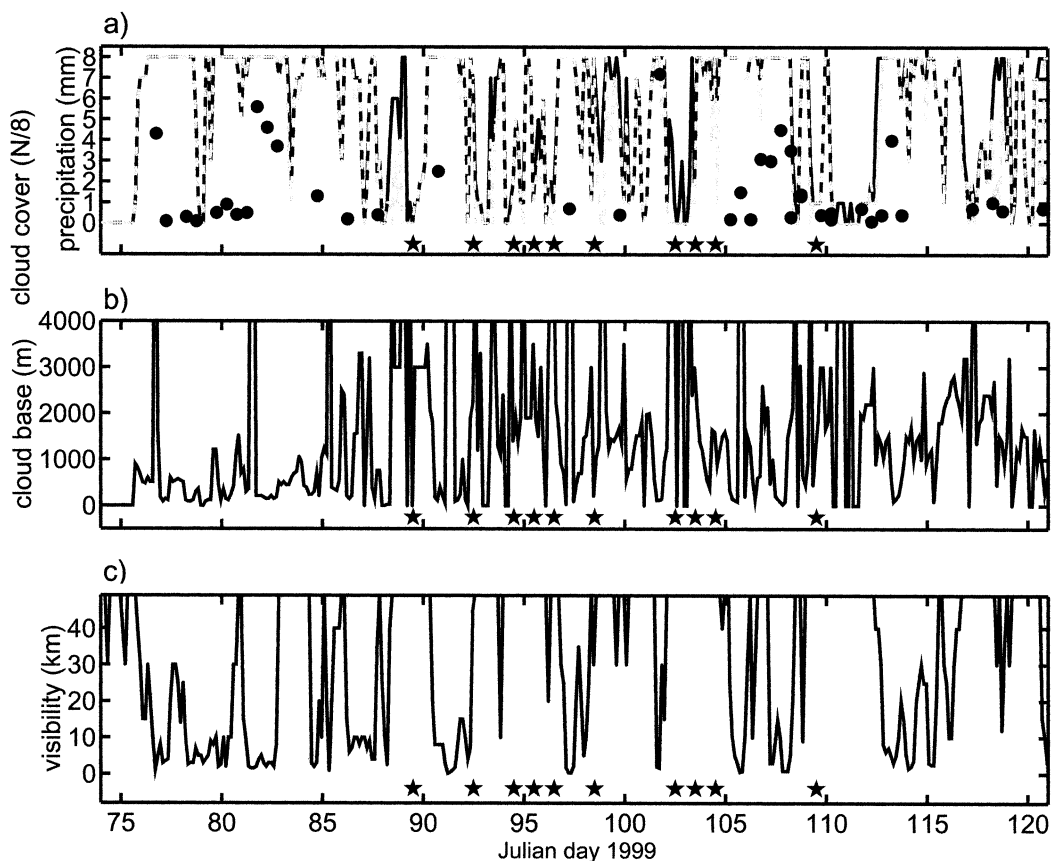


Fig. 3. (a) Low cloud cover (dashed gray line), total cloud cover (black line), precipitation (filled circles) at Halli during BIOFOR 3. Each pentagram indicates a nucleation day in Hyytiälä. (b) As 3a, but for the cloud base up to 4 km. (c) As 3a, but for visibility.

of Hyytiälä to mark each day with nucleation, it is obvious that the aerosol formation took place only in arctic or polar air masses. Furthermore, the nucleation always took place during cold air advection, most clearly during cold air outbreaks, marked by gray triangles. The period of the case study in section 3.1, Julian day 90–98 (31 March to 7 April) (Figs. 1, 2), can easily be recognized in Fig. 6.

Each front passage suggested by Fig. 6 corresponds to changes in temperature, humidity, and tropopause height and wind speed throughout the troposphere in the isopleths in Fig. 5. It is perhaps most obvious how each new air mass results in a change in temperature and to some degree humidity in Fig. 5. Especially the sequences of nucleation days, Julian day 92–96 and 102–104, occurred in

periods of cold air with low relative humidity and no stratiform clouds in the lower troposphere (Figs. 3, 5). The period following on Julian day 77 also had cold arctic air and cold air advection, but no nucleation in Hyytiälä. Unlike the later cold periods, the relative humidity was high in the lower troposphere and stratus and nimbostratus clouds caused overcast conditions and gave snow and rain (Fig. 3). However, in Tahkuse, the skies were clear and nucleation was observed there in the same air mass, see the hexagrams in Fig. 6. The wind maxima below the tropopause in Fig. 6 are probably the best indication of the front passage in the upper troposphere as they mark the jet winds accompanying each front. Letters in Fig. 5 indicates the approximate surface passages of the fronts.

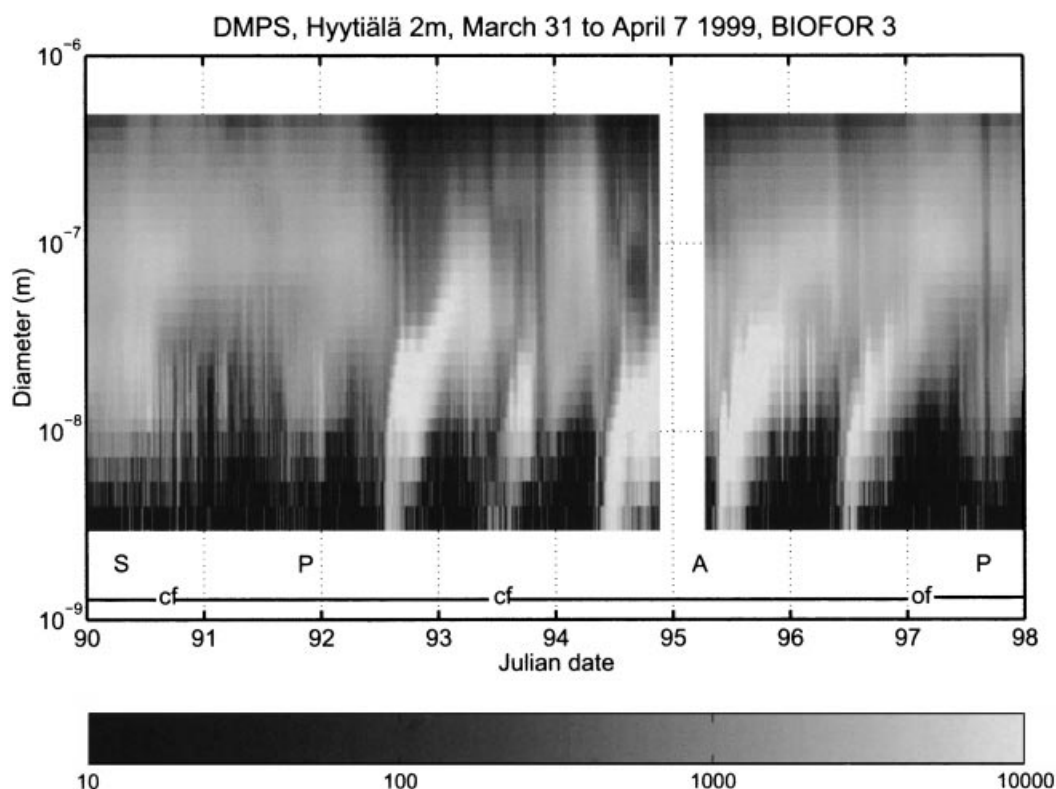


Fig. 4. Aerosol number size distribution from the DMPS system at a height of 2 m in Hyytiälä for the period 31 March to 7 April 1999 (BIOFOR 3), Julian date 90–97. Air mass types and front passages are indicated (cf = cold front, of = occluded front, S = subtropical air mass, P = polar air mass, A = arctic air mass).

In the polar air mass present from Julian day 88 and onward there is never nucleation simultaneously in Tahkuse, Hyytiälä and Värriö, but always in one of these places as the air mass moves northward and then southward again. This suggests that in a Lagrangian perspective, nucleation occur in a longer sequence than observed in each of the stations. In the following arctic air mass from Julian day 94 and onward, nucleation was observed simultaneously in Tahkuse and Hyytiälä, then in all 3 stations and finally in Hyytiälä and Värriö. These observations suggest that nucleation occurs on the synoptic scale, a microscale process somehow linked over 1000 km. There appears to be one exception in Fig. 6 to the rule that nucleation never occurred in subtropical air masses: the nucleation event in Tahkuse on Julian day 100 (10 April). However, while in most cases the 24-h air mass and front analysis were

sufficient, this is a case when the polar front moved so fast that it passed Tahkuse twice between 00 GMT on Julian day 100 and 00 GMT on Julian day 101. Other weather maps and a sequence of satellite images confirm that Tahkuse was within the polar air mass during the nucleation event on Julian day 100.

Table 1 summarizes the air mass versus nucleation statistics for Hyytiälä during the entire BIOFOR. The most striking result is that of all the clear nucleation days, 71% took place in arctic air masses, the rest in polar air masses and none in sub-tropical air. There was a preference for marine air in transition to continental air, which is natural considering the origin of these air masses, the North Atlantic and the Arctic Ocean, see Subsection 3.4 below. Furthermore, all of the clear nucleation events took place during cold air out breaks behind a cold front (in some cases an

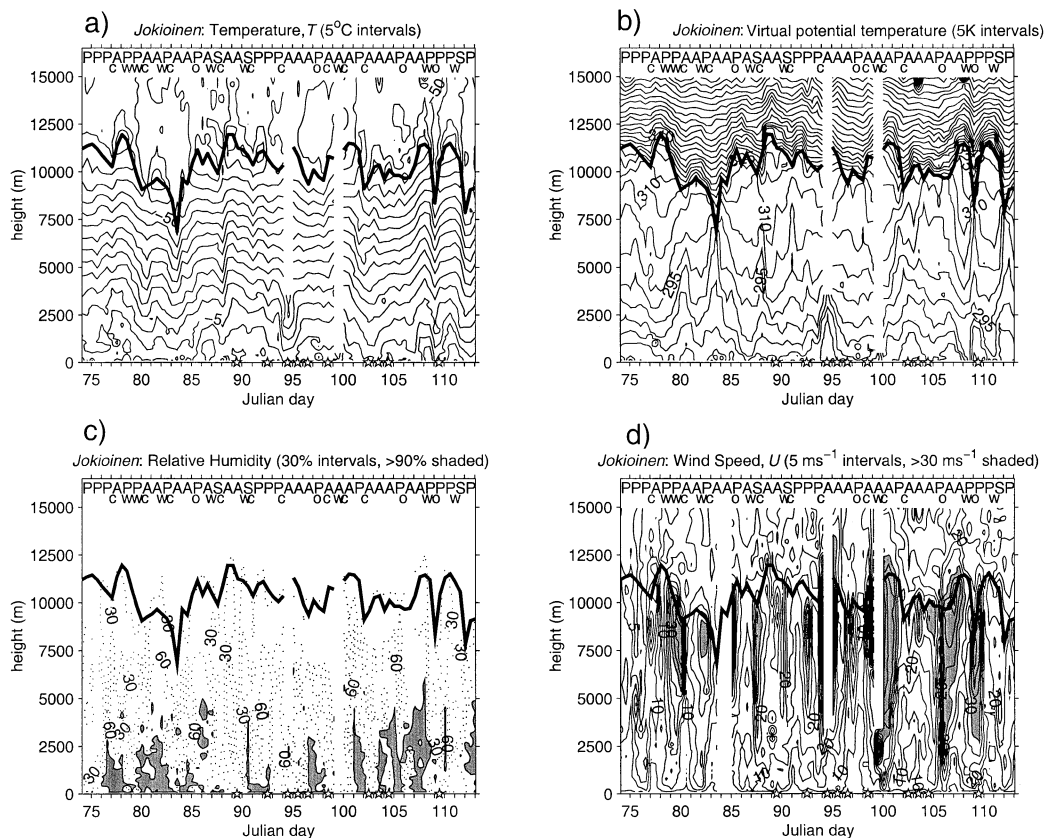


Fig. 5. (a) Isopleth figures based on sondes launched from Jokioinen during BIOFOR 3 in spring 1999, for temperature. The thick black line show the position of the tropopause. Pentagrams at the bottom axis indicate clear nucleation days. Letters below the upper axis indicate air mass (A = arctic, P = polar, S = subtropical) and the approximate time for a surface front passage at Hyytiälä (w = warm front, c = cold front, o = occluded front). (b) As 5a, but for virtual potential temperature. (c) As 5a, but for relative humidity. Shaded area indicates >90% relative humidity. (d) As 5a, but for horizontal wind speed. Shaded area indicates >30 ms⁻¹.

occluded front). This proves that the case study in Subsection 3.1 was not an exception, but the typical situation. However, the opposite was not true, as 42% of the days in arctic air masses were non-nucleation days. arctic and polar air masses were not always associated with nucleation, so obviously, there are other limiting factors. These may be of chemical or meteorological nature. It is also obvious from Fig. 6 and Table 1 that like in the case study in Subsection 3.1, cold air advection is a criterion that must be fulfilled to have nucleation. This agrees with the conclusions made by Väkevä et al. (2000). Warm air advection, even when in an arctic or polar air mass, appears to be a limiting meteorological condition, which

prevents nucleation and cold air advection appears to be favoring it. However, cold air advection does not always result in nucleation.

3.3. Effects of front passages, clouds and precipitation

Already the case study in Subsection 3.1 suggests that cold front passages in daylight, as on 3 April (Julian date 93) in Fig. 4, may disturb the evolution, which leads to nucleation. If Fig. 3 and 5 to 6 are studied together, one can see that nucleation never occurred in direct association with a front, but behind cold fronts, in some cases after occluded fronts. In general, of all front pas-

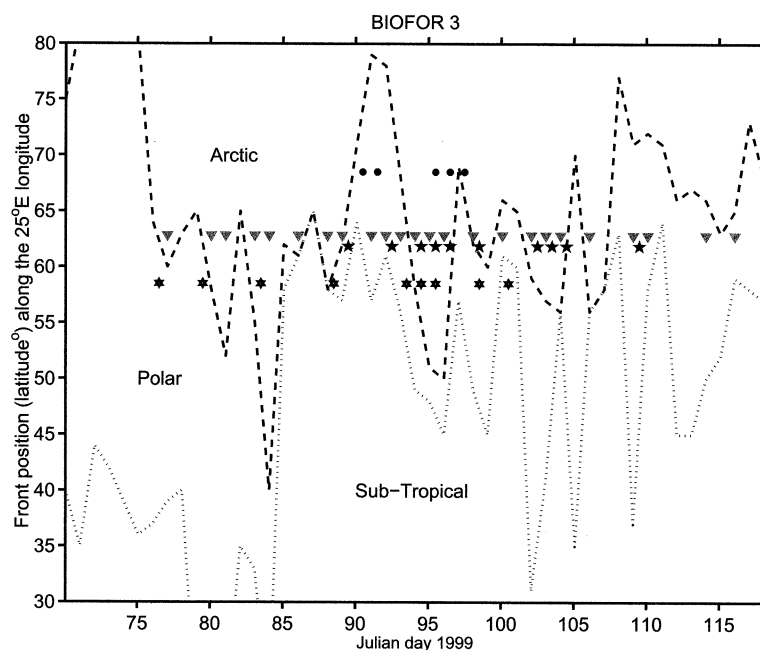


Fig. 6. The latitudinal surface position along the 25°E longitude of the polar front (dotted line) and the arctic front (dashed line) based on the Berliner Wetterkarte. Days with clear nucleation cases in Hyytiälä are marked with a pentagram at the latitude of Hyytiälä and days with cold air outbreaks at Hyytiälä are marked with gray triangles just north of the Hyytiälä latitude. Similarly, days with nucleation at Tahkuse are marked with hexagrams at the Tahkuse latitude and days with nucleation in Värriö are marked with filled circles at the Värriö latitude.

Table 1. *Statistics of air mass types and frequency of cold air outbreaks for the entire BIOFOR*

Air mass	Nucleation day	Unclear cases	Non-nucleation day
arctic	71%	22%	11%
polar	29%	65%	81%
sub-tropical	0%	13%	8%
no. of days	14	23	97
cold air outbreaks	100%	48%	20%

sages during daytime, 90% occurred on days without nucleation. As already indicated above, this suggests that the higher cloudiness and precipitation brought by a front prevent nucleation. Front passages during night usually did not prevent nucleation the following day as long as the front passage was over before sunrise. Several of the days with cold air advection but no nucleation appears to be days during which a cold front passed (Figs. 5, 6). In some cases, a cold front

appears to have interrupted a nucleation event before the new particles could grow in size. In other cases particles of several nm were observed after the front passage, an indication of an earlier nucleation in the air mass except in the direct front zone, as on Julian day 93 (3 April) in Fig. 3.

This is supported by analysis of the observed local weather. One of the factors preventing nucleation on cold air advection days is obviously cloud cover. The overall BIOFOR 3 cloud cover was close to the climatological average of the station of 5.6 and 5.5 for March and April, respectively. However, Fig. 7 shows that days with cold air advection and nucleation (full line with circles) had about half the cloud cover of cold air advection days without nucleation (dashed line with upward triangles). The cloud base and visibility were also higher during nucleation days. In addition, nucleation days had a zero frequency of precipitation, while cold air advection days without nucleation had precipitation in almost 50% of the cases, compared to about 25% for all days

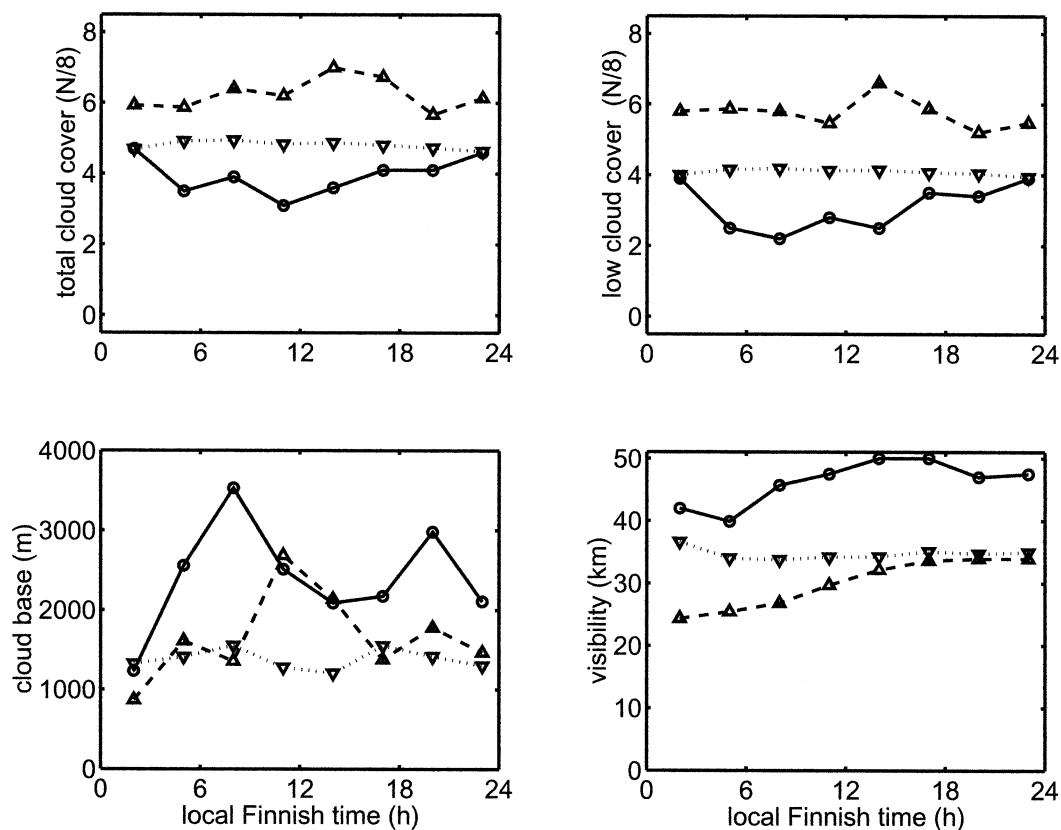


Fig. 7. Diurnal average cycles of total and low cloud cover (in octas), cloud base and visibility for days with cold air advection and nucleation (full line with circles), cold air advection without nucleation (dashed line and upward triangles), and the rest of the days (dotted line and downward triangles).

without cold air advection. In summary, while cold air advection favors nucleation, the cold front itself disturbs or prevents the nucleation. Days with front passages, overcast or precipitation were 55% of the days in arctic or polar air masses with cold air outbreaks. If these are removed it turns out that most remaining days are nucleation days (38%), with only three exceptions (7%) during the entire BIOFOR.

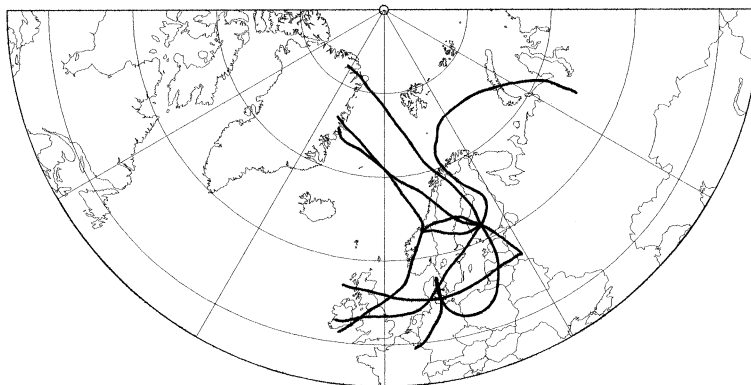
3.4. Origin of air masses

Backward trajectories arriving to Hyttiälä agree with the conclusions drawn from the air mass classification (Fig. 8), but gives in addition a picture of the route of the air masses from their formation region (usually the North Atlantic and the Arctic Ocean) to Hyttiälä. The arctic and

polar air masses during periods of cold air advection reached the experimental site from the southwest over to the north to northeast. These air masses typically originated northwest to northeast direction from the experimental site, but occasionally a polar air mass followed a cyclonic flow over Europe to reach Finland from the southwest. Even in these cases, the air mass carried much of its original characteristics when it reached Hyttiälä from the southwest, if the flow had been fast enough. This may explain why nucleation was occasionally associated with polar or arctic air, which had taken the long way and came in from the west or southwest. As can be seen from Fig. 8, there was no obvious difference in the trajectory patterns between days in cold air advection condition with and without nucleation. Owing to the large number of polar air masses, many without

96 hours long TRADOS back trajectories
Arrival: 06 UTC, 925 hPa, Hyytiälä, BIOFOR 1 + 3

a) Event days



b) Cold air advection/outbreak, no event

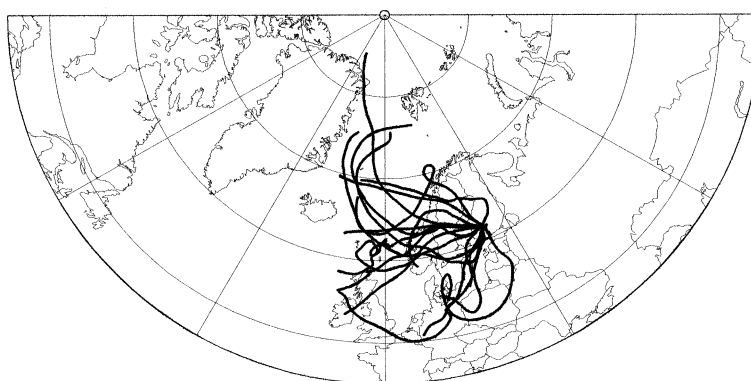


Fig. 8. (a) TRADOS Air mass back trajectories arriving to Hyytiälä at 06 UTC on the 925 hPa pressure level during days with cold air advection and type 1 (clear) particle formation event days in March–April 1999. (b) As 8a, but for days with cold air advection but no nucleation.

nucleation, trajectories came from the southwest to northeast sector in 75% of the days during the BIOFOR campaign. The trajectories themselves do not separate polar and arctic air masses, and since the air masses arrived to Hyytiälä through a wide sector, the statistical difference becomes less sharp than from the air mass characterization. However, among the trajectories arriving at 925 hPa from the southwest-north-northeast sector, 16% were during obvious nucleation days and 61% were during days, which were definitely

not nucleation days. The corresponding fractions for the south-east sector were 4 and 85% (Table 2). These results agree with those by Kulmala et al. (2000) who performed statistical trajectory analysis on nucleation events over a 20-month period.

3.5. Spatial scale of the aerosol formation

We have now established that the nucleation events are strongly favored by certain synoptic conditions, cold air outbreaks or cold advection

Table 2. *Statistics of air flow for the entire BIOFOR as revealed by four-day backward trajectories arriving at 925 hPa at Hyytiälä*

Sector of air flow to Hyytiälä	Nucleation day	Unclear cases	Non-nucleation day
southwest to northeast	16%	24%	61%
south to east	4%	12%	85%

behind cold fronts. We would therefore expect the boundary layer during the nucleation events to have certain characteristics because of two factors: (1) cold air advection over a warmer surface; (2) the often low cloudiness and therefore large amounts of solar heating of the surface below cold air masses. These factors favor large diurnal amplitudes in scale heights and fluxes, the formation of a stable nocturnal layer and convective and turbulent mixed layers during daytime with a dramatic transition during the morning. An accompanying article (Nilsson et al., 2001) is devoted to the importance of the boundary layer evolution on nucleation days, so we shall not dwell on this problem. We shall however discuss the problem we face in understanding how a microscale process such as aerosol nucleation may take place throughout large parts of a whole air mass, on the synoptic scale.

The convection during cold air advection results in Cumulus clouds, if there is enough water vapor and the mixed layer grows deep enough to reach saturation in the convective up drafts. Given certain relationships between the air flow and boundary layer scale, the convection can become organized in roll vortices or convective cells. Cumulus clouds can be seen in Fig. 2 through large parts of the arctic air mass, in particularly as cloud streets formed in the updraft regions of the roll vortices over much of Finland. Roll vortices are a secondary circulation of parallel lateral horizontal rolls with the vertical scale of a convective mixed layer, see Brown (1980) and Atkinson and Zang (1996) for comprehensive reviews. The horizontal scale of each roll is typically 4–12 km in the cross wind direction and the entire system has a horizontal scale of hundreds of kilometers. For more details on the roll vortices observed during BIOFOR, see Buzorius et al. (2001).

The measurements presented in Fig. 4 are not from a Lagrangian experiment, but from a point fixed at the surface in a moving air mass, which means that each size distribution represents a new aerosol. To interpret the evolution of the aerosol size distribution on nucleation days in Fig. 4 as there is growth from newly formed particles of a few nm to larger sizes one must assume a similar history of the aerosol over hundreds of kilometers horizontally, throughout large parts of entire air masses. It may be difficult to imagine that a microphysical process, such as aerosol nucleation and growth could be so synchronized over a meso-scale or even synoptic scale. However, given such time evolutions of aerosol size distributions as in Fig. 4, it would be an extremely unlikely coincidence that subsequently larger and larger particles are advected by random over the experimental site, not only on one day, but 50–60 days per year. The patterns of gradually increasing aerosol sizes in Fig. 4 is itself strong evidence for a similar aerosol history, almost simultaneous aerosol nucleation and similar growth, throughout a large volume of air. Even the interrupted pattern during Julian day 93 supports this, as the aerosol mode, which disappeared during the cold front passage, returned at a larger size. Furthermore, aerosol formation was observed almost simultaneous over hundreds up to a thousand km from Tahkuse, Estonia, over Hyytiälä, to Värriö, Finland, during the same cold air outbreak (Fig. 6).

Cloud streets were also observed in the same weather conditions and over a similar spatial scale. The clouds consist of individual droplets and their formation is as much a microphysical process as the aerosol nucleation. The cloud drops in convective air parcels on the vertical scale of the boundary layer form together Cumulus clouds. The Cumulus clouds are lined up by the roll vortices over the meso-scale to almost the synoptic scale of the cold air mass. The cascade of spatial scales involved is summarized in Fig. 9. We cannot see why aerosol nucleation and growth cannot span the same range of scales as that of Cumulus cloud formation. Our point is not that the roll vortices or cloud streets are necessarily involved in the aerosol nucleation, although they may be, but that they serve as a model to more easily comprehend how aerosol formation over such scales are possible. If ultrafine aerosol particles were as easily detectable with satellite borne

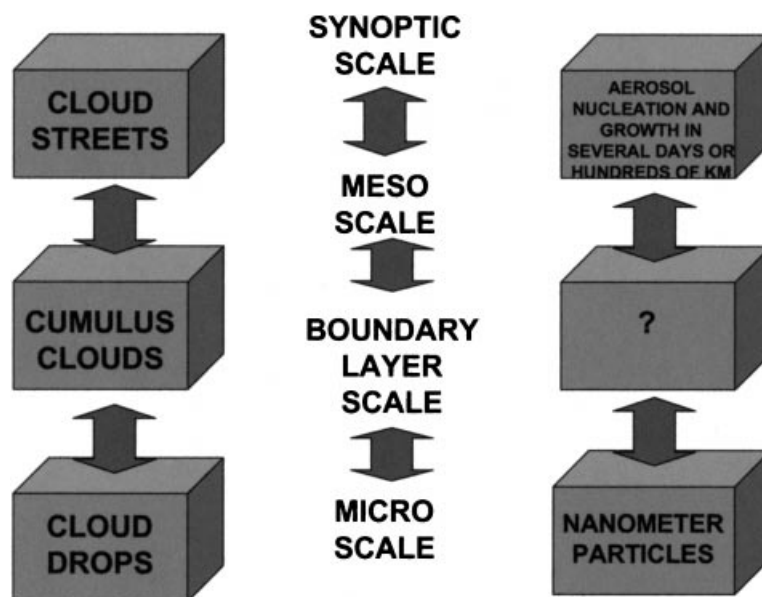


Fig. 9. To the left the cascade of spatial scales from the micro scale (cloud drops) over the boundary layer (cumulus clouds), meso scales (cloud streets) and the synoptic scale. To the right, a parallel cascade of scales for aerosol nucleation.

instruments as the cloud streets are, one might find aerosol nucleation to be as common as cloud streets during cold air advection. However, as indicated by the question mark in Fig. 9, we must still determine exactly what processes are active in between the micro scale and the synoptic scale. This will be the task of an accompanying manuscript by Nilsson et al. (2001).

The fact that aerosol formation is related to such a specific, but common, weather situation may have extensive implications. Arctic and polar air masses will be relatively clean (low pre-existing aerosol surface (Janson et al., 2001; Aalto et al., 2001)). As they are transported in over northern Europe, they will be transformed meteorologically (heated and humidified) while they are also loaded with gasses and aerosols from both natural and anthropogenic gas and aerosol emissions. If a new aerosol nucleates, at least partly from anthropogenic vapors (which would be the case if the nucleation path was ternary homogeneous nucleation by $\text{H}_2\text{SO}_4 + \text{NH}_3 + \text{H}_2\text{O}$), and if the aerosol is allowed to grow to CCN size within the same air mass (during the transformation of the air mass) this may affect the net result on total climate forcing. In the general circulation, the role of these

cold air masses is to transport cold air from the polar regions towards the equator. If this air gets more CCN and brighter clouds due to anthropogenic emissions, it will reduce the radiative heating of the air mass and perhaps allow the air to keep its colder characteristics for a longer period of time and a longer distance.

3.6. Annual variability of nucleation

If we extrapolate what we have learned about the importance of air mass type and cold air advection between the three BIOFOR campaigns over the typical annual cycle in nucleation frequency we can find a meteorological explanation for the annual cycle in the occurrence of nucleation days. Cold air outbreaks, advection of cold air behind cold fronts are a part of the latitudinal heat transport in the west wind belt in mid to high latitudes where wave disturbances, cyclones, along the polar and arctic fronts reduces the latitudinal radiation driven temperature differences. The larger latitudinal temperature differences during spring and autumn cause a larger and stronger cyclone activity than in summer and winter. In winter the polar front is on average

located further south, while in summer the arctic front does not typically exist since arctic air masses seldom form over the pole at that time (Petterssen, 1956). Altogether, this annual cycle in synoptic weather patterns and latitudinal heat exchange is itself able to explain the annual cycle in aerosol nucleation frequency with a maximum in spring and autumn and a minimum in winter and summer. Indeed, during BIOFOR 2 in July to August 1998, which had no clear nucleation days at all, arctic air never reached Hyytiälä, cold air outbreaks were uncommon as the arctic front was rarely present and the polar front was more stationary. Both during BIOFOR 1 and 3 in the springs of 1998 and 1999, especially BIOFOR 3, with numerous nucleation days, arctic air frequently reached Hyytiälä and cold air outbreaks along the arctic and polar fronts were common as can be seen in Fig. 6. The analysis of nucleation in terms of air mass classification and synoptic weather which has been made for the three BIOFOR periods should be extended to several years in order to understand the climatology of aerosol formation.

4. Summary and conclusions

We have found that nucleation occurred in arctic and to some extent in polar air masses, with a preference for air in transition from marine to continental air masses, and never in sub-tropical air. These air masses originated north of the BIOFOR experiment and reached the experimental site through flows from the southwest to northeast over the north. The nucleation was always associated with cold air advection, cold air outbreaks behind cold fronts, never warm air advection, which may be related to the typically low pre-existing aerosol concentrations, low cloudiness and large diurnal amplitudes in the continental boundary layer associated with cold air advection and clear skies.

Arctic and polar air together with cold air advection did not always result in nucleation.

There were other limiting factors, such as high cloudiness and precipitation, which must have resulted in reduced photochemistry and scavenging of the new aerosol or the precursor gases. Therefore cold front passages explain the absence of nucleation on some days with cold air advection.

The preference of nucleation for occurring in arctic air masses suggests a meteorological explanation of the annual cycle of nucleation frequency with a minimum of nucleation events in summer when the arctic air mass type seldom form in the northern hemisphere. The connection to cold air outbreaks suggests that the maximum in nucleation events during spring and autumn may be explained by the larger latitudinal temperature gradients and higher cyclone activity at that time of the year.

We suggest that the nucleation of new aerosol particles, like drop formation in the Cumulus clouds formed during cold air advection, both occurring at the microphysical scale, are connected through boundary layer processes and to the meso- and the synoptic scale. Therefore, nucleation can take place on the same day throughout large parts of these air masses. This is examined in more detail in accompanying articles (Nilsson et al., 2001; Buzorius et al., 2001).

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