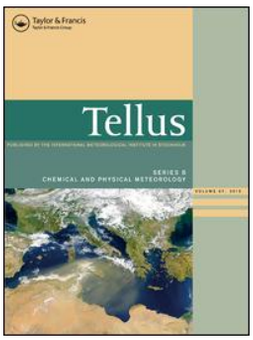

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TELLUS

Horizontal homogeneity and vertical extent of new particle formation events

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ABSTRACT

During the SATURN campaign 2002, new particle formation, i.e. the occurrence of ultrafine particles was investigated simultaneously at four ground-based measurement sites. The maximum distance between the sites was 50 km. Additionally, vertical profiles of aerosol particles from 5–10 nm have been measured by a tethered-balloon-borne system at one of the sites.

In general, two different scenarios have been found: (i) new particle formation was measured at all sites nearly in parallel with subsequent particle growth (homogeneous case) and (ii) new particle formation was observed at one to three sites irregularly (inhomogeneous case) where subsequent particle growth was often interrupted. The homogeneous case was connected with stable synoptical conditions, i.e. the region was influenced by a high pressure system. Here, the horizontal extent of the phenomenon has been estimated to be 400 km at maximum. In the vertical dimension, the ultrafine particles are well mixed within the entire boundary layer. In the inhomogeneous case the new particle formation depends mainly on the incoming solar radiation and was often interrupted due the occurrence of clouds. Thus, single point measurements are not representative for a larger region in that case.

1. Introduction

New particle formation, i.e., the occurrence of particles in the size range of a few nanometres in diameter has been observed in the atmosphere at various locations: urban, remote, marine and arctic sites (e.g. Covert et al., 1992; Wiedensohler et al., 1996; Mäkelä et al., 1997; Weber et al., 1997). Kulmala et al. (2004) reviewed more than 100 of such experimental studies from worldwide locations. Newly formed particles are too small to scatter light or act as cloud condensation nuclei. However, in polluted continental regions they may grow up to 100–200 nm in diameter within a few hours and act as cloud condensation nuclei and scatter light. Thus, they affect the regional climate (Laaksonen et al., 2005). Although these ultrafine particles occur due to nucleation and subsequent growth by condensation, the causes of these new particle formation events are still not completely understood. Some events depend obviously on mixing processes connected with the development of the planetary boundary layer (PBL). A detailed discussion of different particle formation scenarios in combination with PBL dynamics is given by Bigg (1997) and Nilsson et al. (2001a). Siebert et al. (2004) measured increased particle number concentrations between 5 and 10 nm at the inversion of the continental PBL which have been probably mixed downwards after removal of the inversion.

The processes of new particle formation as well as growth rates vary significantly between different sites and seasons (Kulmala et al., 2004). On the other hand, there should be similarities between nearby sites over homogeneous terrain with similar meteorological conditions. Therefore, simultaneous measurements at several sites are needed to understand the horizontal extent of new particle formation events. Up to now, most studies were limited to single locations and it is not clear if such nucleation events are horizontally homogeneous. The answer to this question is essential for regional and global modelling, i.e. if a single point measurement is representative for the whole grid cell of a model. Pirjola et al. (2004) showed that nucleation in many cases plays an important role and should be included in global and regional models.

Vana et al. (2004) observed new particle formation on the synoptic scale at three sites. The enhanced number concentrations of ultrafine particles were found along the trajectory of a certain air mass over distances of more than 1000 km with a time-shift of 1–2 d. Similar results have been found by Komppula et al. (2005) comparing two sites 250 km apart. They found that

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particle formation in a certain air mass type depends not only on the diurnal variation of the parameters causing the event but also on some properties carried by air mass itself. Komppula et al. (2003) investigated characteristics of new particle formation events at two sites only 6 km apart but at different heights. They showed a maximum difference in starting time of 30 min, probably caused by different turbulent mixing processes.

This study focuses on two question: Under which conditions are new particle formation events observed simultaneously at different locations (homogeneous case)? When are locally isolated events observed, i.e. the events do not appear simultaneously at different sites (inhomogeneous case)?

In order to shed light on this issue, simultaneous measurements at four research stations were performed during the SATURN ('Strahlung, vertikaler Austausch, Turbulenz und Partikel-Neubildung'; radiation, vertical exchange, turbulence and new particle formation) field campaign in 2002 (Stratmann et al., 2003). One of the stations was located in the outskirts of Leipzig, Germany, the other three stations were outside the city area. A period of three consecutive days with production of new particles and subsequent particle growth was observed at four stations indicating mesoscale events. In contrast, one case with inhomogeneous particle formation is presented.

2. Experimental

2.1. Measurement sites

During SATURN 2002 (27 May until 15 June 2002) ground based measurements of number size distributions were carried out in the city of Leipzig and at three different locations in a distance of ~10 to 50 km to the city (cf. Fig. 1):

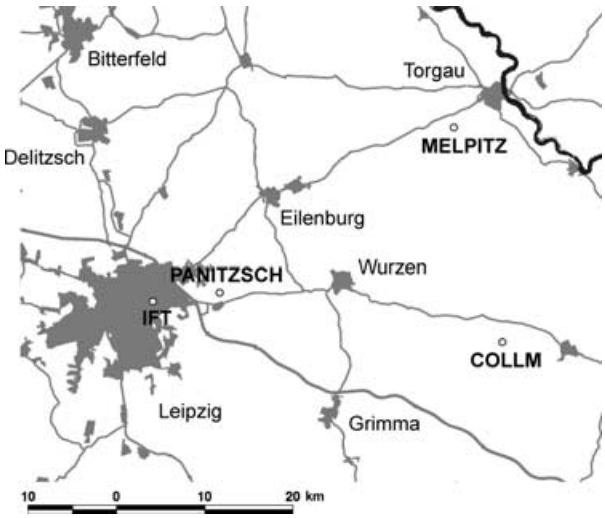

Fig. 1. Map of the measurement site during SATURN showing the location of the measurement sites Melpitz, Collm, Panitzsch and IfT.

(i) Measurements at the top of the building of the Leibniz-Institute for Tropospheric Research (IfT). The IfT-building is located in the urban area of Leipzig, considered to represent the urban background. The distance to the nearest road is approximately 200 m; more details about this station are given in Wehner and Wiedensohler (2003). The inlet was mounted approximately 3 m above the rooftop, corresponding to a height of 16 m above ground.

(ii) Panitzsch is a small village 10 km east of Leipzig (suburban), measurements were taken at the edge of a residential area, 20 m from a wheat field. The inlet was located approximately 10 m above ground, i.e. 1 m higher than the nearby housing.

(iii) Melpitz (42 km northeast of Leipzig) is the permanent research station of the IfT (Spindler et al., 2001). The measurement site is on a flat meadow, surrounded by agricultural land. The nearest major road is 1.5 km away. The inlet was located 1.5 m above the container top, i.e. 4 m above ground.

(iv) Collm is an isolated hill (45 km east of Leipzig), approx. 130 m above surrounding terrain comprising of agricultural land, small villages, and forests. The next major road is 2 km away. The inlet was mounted 1 m above the top of the building, i.e. 25 m above ground and a few metres above the treetops.

2.2. Ground-based instruments

Identical TDMPS (Twin Differential Mobility Particle Sizer, Birmili et al. (1999)) systems were used to measure particle number size distributions in the particle diameter range $3 < D_p < 800$ nm at all four measurement sites. They consist of two size spectrometers that simultaneously measure different particle size ranges at dry conditions. Ultrafine particles ($3 < D_p < 22$ nm) were measured with an Ultrafine Differential Mobility Analyzer (Hauke type) in conjunction with an Ultrafine Condensation Particle Counter (UCPC, Model 3025, TSI Inc., St. Paul, MN, USA). Particles in the range from $22 < D_p < 800$ nm were determined by means of a Differential Mobility Analyzer (Hauke type) and a CPC (TSI-Model 3010). In Melpitz, the total particle number concentration ($3 < D_p < 1000$ nm) was additionally measured using a UCPC (TSI-Model 3025). The inlets used at the four sites were commercially available low-flow PM10 inlets (Thermo Anderson, Smyrna, GA, USA).

2.3. Balloon-borne instruments

In addition to the ground-based observations tethered-balloon-borne measurements with the instrumental payload ACTOS (Airborne Cloud Turbulence Observation System) provided vertical profiles of the particle number concentration and meteorological data at the Melpitz site. ACTOS is an autonomous system, which is equipped with sensors to measure the meteorological standard parameters (three-dimensional wind vector, static air temperature and humidity) with high temporal (100 Hz

sampling frequency) and spatial resolution. Furthermore, two CPCs (TSI-model 3762) with different lower detection limits provided particle number concentration in two size ranges. From the difference, the concentration of the ultrafine particles N_{5-10} in the size range between 5 and 10 nm was derived. A more detailed description of ACTOS and the tethered balloon system can be found in Siebert et al. (2003); details of the aerosol measurements on ACTOS are described in Siebert et al. (2004) and Stratmann et al. (2003).

2.4. Ancillary data

In order to characterize similarities and differences in air mass transport routes for all four measurement sites during SATURN campaign, Back-trajectories were calculated for the sites every 3 hr using the NOAA HYSPLIT model (Draxler and Rolph, 2003). In addition, with the aid of trajectories, the horizontal extent of the new particle formation was estimated.

3. Data analysis

The process of new particle formation can be described by several parameters derived from the number size distribution. The number concentration of particles between 3 and 15 nm was used as an indicator for the new particle formation, because this size range was found to show a clear increase during new particle formation events without significant influence of anthropogenically emitted particles.

One way to parametrize the temporal behaviour of an aerosol particle mode is to fit a log-normal distribution to the ultrafine particle size range using a moment-preserving least-squares fitting algorithm (Birmili, 2001). In this work, emphasis was directed towards generating a continuous time-series of the mean diameter (*GMD*) of the nucleation mode. The growth rates (*GR*) of these particles were calculated by following the *GMD* of the nucleation mode. Additionally, based on the temporal extent of the formation burst, and on the rate of change of the observed aerosol number concentration in the nucleation mode size range (3–15 nm in diameter), the formation rate of new particles ($J3$) was estimated. Details on how these parameters are calculated can be found in Kulmala et al. (2001), Dal Maso et al. (2002, 2005).

In order to understand the new particle formation process, there is a need to characterize aerosol particle size distributions prior and during the formation event. The pre-existing particle population acts as a sink for condensable vapours present in the atmosphere, as well as for the newly formed particles. This can be quantified with the condensation sink (*CS*), which describes the loss rate [in molecules s^{-1}] of vapour to the aerosol phase existing in the atmosphere. When particle concentrations are high, the sink can be the limiting factor in the formation of new particles. All the vapour then condenses onto pre-existing particles instead of forming new ones. Additionally, if new particles are formed, they are scavenged by the pre-existing population. The effect of hygroscopic growth according to ambient humidity has been considered for calculation of the condensation sink. Parameters describing hygroscopic growth of the typical aerosol in the Leipzig area were taken from Massling et al. (2005).

Kulmala et al. (2001) proposed an analytical expression, which utilizes the condensation sink in the estimation of condensable vapour concentration (*CVC*). In practice, *CVC* is the concentration of a sample vapour (sulphuric acid in this case), which can explain the observed growth rate of nucleation mode particles during an event. Simplifications and assumptions made in the calculations can be found in Kulmala et al. (2001).

Following a work of Kulmala et al. (2001), also the condensable vapour production rate (*CVPR*) was calculated for each formation event day and each sampling site assuming a pseudo-steady state for the vapour.

4. Results and discussion

New particle formation events were observed simultaneously at the four stations on 7 d during SATURN (homogeneous case); on 3 d new particle formation was observed for 1 to 3 stations only (inhomogeneous case). Here, two case studies covering both cases are examined in detail in order to characterize the horizontal and vertical extent of the formation process.

4.1. Horizontally homogeneous new particle formation

The period between 3 June (00:00 CEST, Central European Summer Time) and 5 June (24:00 CEST) was analysed. The meteorological situation was mainly influenced by a high-pressure area over Germany yielding sunny conditions with cumulus cloud formation in the afternoon. The wind came from easterly directions (90°–130°), slightly increasing from about 3 m s^{-1} on 3 June to 7 m s^{-1} on 5 June. The ambient temperature varied between 5°C in the morning and 27°C in the afternoon.

4.1.1. Ground-based measurements. Figure 2 shows the evolution of normalized particle number size distributions between 3 and 5 June 2002 at the four stations. Each number size distribution measured by the TDMPS systems was divided by the total number concentration from 3 to 800 nm to emphasize the location of the particle size distribution maxima independent of the actual number concentration. These four contour plots have similar general features at all sites: on every day in the late morning ultrafine particles appear and grow subsequently to sizes ~80 nm until sunset. This normalized presentation demonstrates the new particle formation at the four sites nearly parallel with similar intensity.

Figure 3 shows the time-series of particle number concentration in the range of 3–15 nm in diameter (N_{3-15}) and the calculated condensation sink (*CS*) for the 3-d period. The start time of nucleation events is defined by a significant increase in N_{3-15}. Maximum values of N_{3-15} vary between the sites and from day

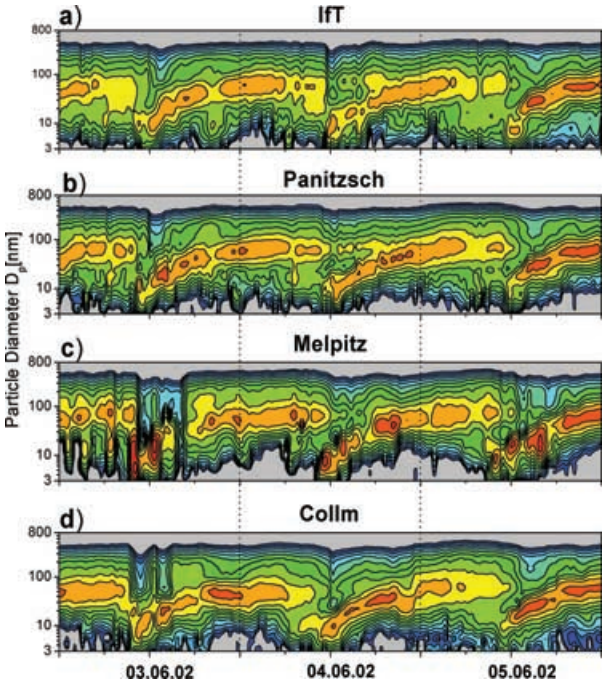

Fig. 2. Evolution of normalized number size distributions from 3 June (00:00 CEST) to 5 June (24:00 CEST), 2002 at the stations IfT, Panitzsch, Melpitz, and Collm. The colours represent particle number concentration divided by the total number concentration, grey represents relatively low concentrations and red means high concentrations.

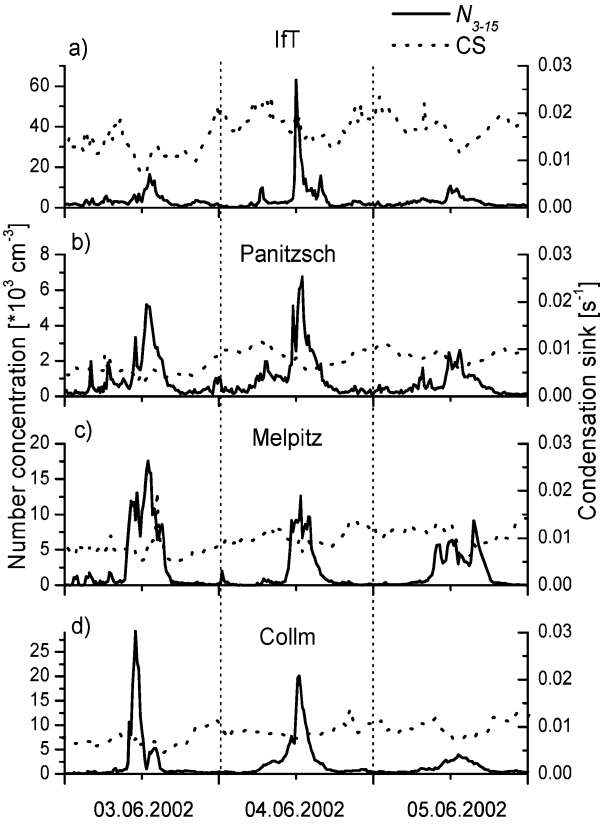

Fig. 3. Evolution of the number concentration N_{3-15} in the size range of 3–15 nm and condensation sink CS between 3 June (0:00 CEST) and 5 June (24:00 CEST), 2002 at the stations IfT, Panitzsch, Melpitz and Collm.

to day. Highest values occurred on 4 June at IfT, lowest values in Panitzsch for the whole period. Thus, new particle formation events showing similar features for all sites represented by the normalized contour plot, vary in details, such as number concentration of ultrafine particles N_{3-15} significantly.

Highest values of *CS* occur at IfT due to the additional urban particle sources leading to a higher surface area concentration, but also due to to higher precursor concentrations which can be expected in the urban air. About 2 or 3 hr before a nucleation event, often a slight decrease of *CS* is observed. Thus, there is less pre-existing surface for the molecules to condense onto. Instead, these molecules contribute to nucleation and subsequent growth of nucleation mode particles. At the other measurement sites this decrease in *CS* is not as obvious.

There is, however, not a definitive limit of the *CS*, which would automatically result in a new particle formation. Low volatile vapour concentrations are continuously produced by oxidation in the atmosphere as well as lost to existing surfaces. New particle formation has been observed to take place in both polluted conditions (Wehner et al., 2004; Mönkkönen et al., 2005) and in a pristine Antarctic environment (Koponen et al., 2003). This indicates that the increased *CS* can be compensated by a larger vapour source rate, which can vary up to four orders of magnitude between polluted and clean atmospheres (Kulmala et al., 2005).

In a more polluted surrounding, (IfT in this study), the decrease in *CS* as a result of growth of PBL and vertical mixing with cleaner air from aloft can initiate the nucleation event. The contour plots also show this process: During morning hours the maximum concentration occurs in the Aitken mode ranging between 40 and 70 nm. Between 8:00 and 10:00 CEST this mode disappears at ground level due to the break-up of the nocturnal inversion and subsequent dilution with cleaner air from higher altitudes. Therefore, the sink for condensing material and clusters is reduced and new particle formation and subsequent growth can start. At Panitzsch, Collm and Melpitz the decrease in *CS* is less obvious. The *CS* in the residual layer seems to be comparable to the values underneath the inversion in the more rural sites, i.e. reflecting both the continental background. The main reason is that at the urban site additional particles are emitted during night and morning resulting in a higher *CS* near the ground. A similar phenomenon can be observed at rural sites if cleaner (e.g. arctic) air is advected above the inversion diluting later the ground air (Nilsson et al., 2001a). However, with these measurements alone it is not possible to conclude if the nucleation occurred due to the decrease in *CS* caused by downward mixing of cleaner air or if the newly formed particles have been formed aloft and

Table 1. Overview over start time, growth rates *GR*, maximum number concentrations 3–15 nm N_{3-15}, formation rates (*J*3), condensable vapour concentration (*CVC*) and condensable vapour production rate (*CVPR*) during 3–5 June 2002.

	Start time				Growth rate [nm h^{-1}]			
Date	LE	PA	ME	CL	LE	PA	ME	CL
03.06	10:30	10:30	9:30	10:00	4.2	5.3	3.8	4.9
04.06	11:45	11:30	10:30	10:30	3.0	4.5	2.9	3.7
05.06	11:30	11:15	9:30	10:30	5.5	4.4	3.0	3.9
	Max. N_{3-15} [10^3 cm^{-3}]				Formation rate (*J*3) [cm^{-3} s^{-1}]			
Date	LE	PA	ME	CL	LE	PA	ME	CL
03.06	16.2	5.2	17.5	29.2	0.73	0.24	1.80	0.9
04.06	63.2	6.7	12.6	20.1	0.54	0.53	1.69	0.48
05.06	10.7	2.6	9.1	4.0	1.54	0.19	0.40	0.09
	CVC [10^7 molec. cm^{-3}]				*CVPR* [10^5 molec. cm^{-3} s^{-3}]			
Date	LE	PA	ME	CL	LE	PA	ME	CL
03.06	5.7	7.3	5.2	6.7	6.7	3.3	3.5	4.1
04.06	4.2	6.1	3.9	5.1	6.5	4.9	3.6	4.6
05.06	7.5	6.0	4.1	5.4	11.6	5.4	4.0	5.3

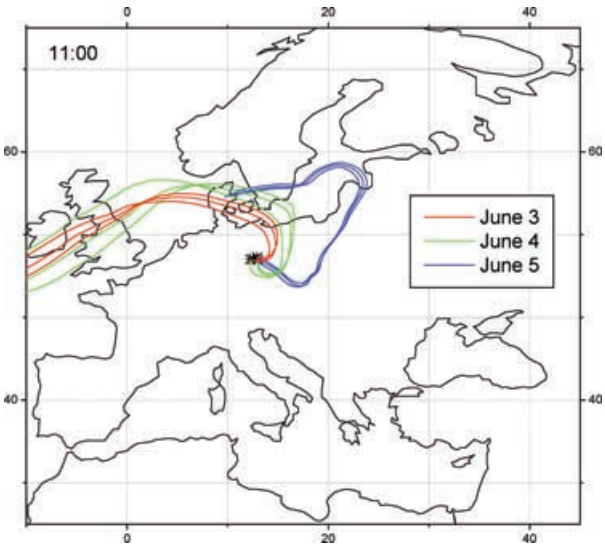

Fig. 4. 144 hr back-trajectories (HYSPLIT) for the stations Melpitz, Collm and IfT on 3, 4 and 5 June arriving at 11:00 CEST in a height of 100 m asl.

mixed downwards together with the cleaner air. The PBL development plays an important role in both cases and is one major factor controlling the occurrence of ultrafine particles near the ground.

Condensable vapour concentrations and their production rates during 3–5 June 2002 at four different sites were estimated. Table 1 summarizes the calculated parameters, when new particle formation was observed to occur spatially homogeneously. The event start varies up to 2 hr between the stations. During this selected period, the event starts always first in Melpitz and latest at the IfT site.

Figure 4 shows 144-hr back-trajectories calculated for the three stations Melpitz, Collm and IfT arriving at 11:00 CEST on the particular day [100 m above ground level (agl)]. Differences between trajectories of the three stations were negligible, also due to the spatial resolution of the trajectory model. Thus, all four stations were influenced by similar air masses. The flow from the East partly explains the time difference (30–120 min) in the onset of formation (Table 1). One reason could be the higher surface area concentration of existing particles in the urban area. This results in a higher *CS* which may suppress new particle formation. Another difference between urban and rural areas is the different diurnal temperature development, i.e. the diurnal PBL development is less pronounced in the urban areas due to the compensating character of the city. A time-shift in PBL development would probably also cause a time-shift in the new particle formation as shown also in Nilsson et al. (2001a).

Back-trajectories show a similar origin for the 3 d: on 3 and 4 June they passed more than 2 d ago the British Islands, the North Sea and part of the western Baltic Sea before turning south and spending approximately 2 d over Poland and Eastern Germany. On 5 June, the back-trajectories changed: the air mass passed the Baltic Sea before spending 4 d over Eastern Europe. Thus, on 5 June, the continental influence is more significant which might be an explanation for the general decrease of the ultrafine particle concentration.

The growth rates (*GR*, Table 1) at four sites vary from 2.9 nm h^{-1} to 5.5 nm h^{-1}, which all fall in a range typical for the Leipzig area (Birmili and Wiedensohler, 2000). Lower values were typically observed at rural sites (Melpitz and Collm), but day-to-day variability and differences between the sites were of the same order of magnitude. Formation rates of 3 nm particles (*J*3) were 0.09–1.8 cm^{-3} s^{-1}. On average, the formation rate was slightly higher in Melpitz than in other sites but the subsequent growth was slower. Thus, although the new particle formation seems to occur synchronously at the different sites, parameters describing the process such as *GR* and *J*3 may vary significantly. This might be due to local conditions such as different sources of precursor gases.

The condensable vapour concentration calculated by the model is similar for the individual sites and varies between 3.9 and 7.5*10^7 molec. cm^{-3}. The production rate for the condensable vapours (*CVPR*) at IfT was somewhat higher than at the rural sites. Due to urban pollution, the condensation sink (*CS*) to the pre-existing particle surfaces was highest at IfT. Thus, events at urban sites occur with higher vapour production leading to the

growth of small clusters to detectable sizes and later into the Aitken mode. Therefore, *CVPR* was significantly higher at IfT than at the rural sites. Values for *CVPR* at IfT were of a same order of magnitude as in Marseille and Athens (Kulmala et al., 2005).

Similarities in the growth rates can be explained by their interdependence on *CS*, *CVPR* and emissions. A source, which emits gaseous precursors as well as primary particles, increases both *CVPR* and *CS*. Vapour emissions, on the other hand, increases at the beginning *CVPR* only, but this can lead to enhanced gas-to-particle conversion thus increasing the *CS* as well. This balances the variability in the vapour concentration (*CVC*) and as a result in growth rate of nucleation mode particles (Kulmala et al., 2005).

One arising question is over which horizontal extend the phenomenon can be assumed to be homogeneous. SATURN 2002 was no Lagrangian experiment, because measurements were taken at individual points fixed at the surface in a moving air mass. Thus, the horizontal extent of new particle formation can only be estimated using different approaches: One option is taking the distance between the simultaneously observed events, in this study 40 km, for example in Nilsson et al. (2001b) 1000 km. This assumption might be used as lower estimate for cases with horizontally homogeneous terrain and also homogeneous meteorological conditions. Another way seems to be more reasonable because it considers the moving air mass, in terms of back-trajectories. They can be used to estimate, how fast an air mass was transported and from that to obtain the location of an air mass in the past. Assuming the new particle formation process being a homogeneous process on the mesoscale on days like 3–5 June back-trajectories are suitable to estimate the horizontal dimension of this phenomenon. An air parcel arriving in the evening at one of the sites contains particles which have been formed by nucleation somewhere else and grew during the transport. Assuming a mean start time of 11:00 CEST aerosol particles measured at 21:00 CEST have been travelling for 10 h. Mean air mass velocities have been estimated for a height of 100 m above surface level to 13 km h^{-1} on 3 June, 11 km h^{-1} on 4 June, and 40 km h^{-1} on 5 June, respectively. From analysing the back-trajectories one might speculate that the new particle formation observed around Leipzig is representative for a region of at least ~100 km to the east for 3 and 4 June, and of ~400 km for 5 June.

4.1.2. Vertical measurements. In addition to the ground-based measurements at the four measurement sites, vertical profiles of ultrafine particle concentrations $N_{5–10}$ in the size range between 5 and 10 nm have been measured with the balloon-borne ACTOS at the Melpitz site. This kind of measurement is available only for particular days and times. Here, selected profiles measured on 3 and 4 June will be presented, for 5 June no measurements are available.

Figure 5 shows several vertical profiles of $N_{5–10}$ (left-hand panel) and of potential temperature T_{pot} (right-hand panel) measured on 3 June between 5:35 and 11:30 CEST over Melpitz. The times represent a mean time for the respective ascent or descent, which took 10–20 min with a typical climb rate of the balloon of 1–2 m s^{-1}.

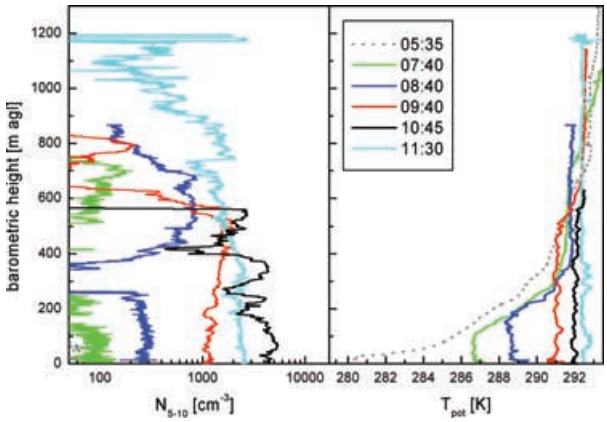

Fig. 5. Selected vertical profiles of $N_{5–10}$ in the size range of 5–10 nm (left-hand side) and potential Temperature T_{pot} (right-hand side) measured on 3 June in Melpitz. Times are in CEST and valid for the mean of profile.

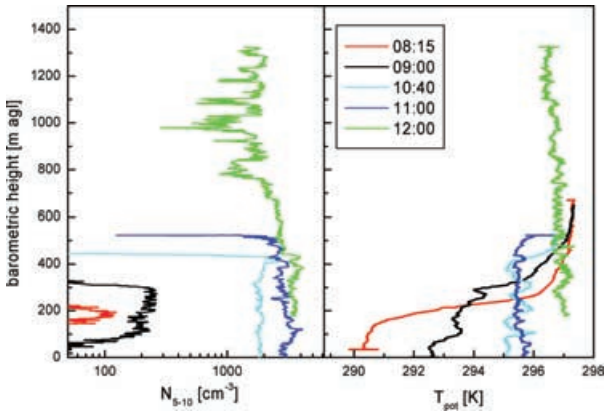

Fig. 6. Selected vertical profiles of $N_{5–10}$ in the size range of 5–10 nm (left-hand side) and potential Temperature T_{pot} (right-hand side) measured on 4 June in Melpitz. Times are in CEST and valid for the mean of profile.

During the first ascent around 5:35 CEST $N_{5–10}$ was below 50 cm^{-3}, i.e. no significant concentrations of ultrafine particles were observed within the lowest 1300 m, T_{pot} increased continuously up to a height of 700 m. The measurement at 7:40 CEST shows a temperature inversion below 300 m and $N_{5–10}$ of less than 100 cm^{-3} below the inversion but up to 200 cm^{-3} above. The following profile exhibits this development even more clearly: $N_{5–10}$ reached values up to 800 cm^{-3} above the inversion and values below 300 cm^{-3} in the lower 250 m. Thus, formation of ultrafine particles started independently at different heights because the strong inversion layer prevents mixing between ground based and overlaying air masses. $N_{5–10}$ was

significantly higher above the inversion indicating new particle formation within this layer. At 9:40 the temperature inversion broke up and a well-mixed boundary layer developed. Then, N_{5-10} was nearly constant, thus the newly formed particles from above were mixed downwards.

Figure 6 shows several vertical profiles of N_{5-10} (left-hand panel) and of potential temperature T_{pot} (right-hand panel) measured on 4 June between 8:00 and 12:00 CEST over Melpitz.

During the first measurements between 8:00 and 9:00, N_{5-10} was below 250 cm^{-3} with no ultrafine particles being observed at ground level or above the inversion layer at 300 m. The inversion layer is indicated by a strong increase of the T_{pot} below 300 m for the ascent at 08:15 and 330 m for the descent at 09:00. This temperature increase resulted in a stable stratified layer with negligible exchange with overlaying air masses. Due to technical reasons no measurements between 9:00 and 10:40 are available. Between 10:40 and 11:00 N_{5-10} increased significantly to values $\sim$2500 cm^{-3} up to a height of 450 to 500 m. This behaviour is due to the breaking of the inversion layer with subsequent efficient vertical mixing. At this stage ultrafine particles were measured also at ground level (Fig. 2). The sharp decrease of N_{5-10} indicates the actual height of the so-called mixing layer. Within the following 90 min the increase of N_{5-10} continued. At 12:00 N_{5-10} was 1000–3000 cm^{-3} in the well-mixed PBL, i.e. up to 1300 m. The higher fluctuations in the range between 800 and 1300 m indicate mixing of the PBL with cleaner air from above the inversion layer.

The conclusion from the vertical measurements is that the new particle formation may start at different heights: on 3 June it happened mainly above the inversion layer, on 4 June within a thin layer around 200 m. Thus, different PBL development and different vertical profiles of N_{5-10} may ultimately lead to similar results at the ground: appearance of ultrafine particles with subsequent growth indicated by the typical banana-shaped contour plot, but the underlying process might be different. After break-up of the inversion layer new particles were found over the entire mixing layer. This could be the result of vertical mixing from the region where nucleation and growth took place or most probably the whole process of nucleation and growth happens all over the PBL. In the upper part of the PBL fluctuation in N_{5-10} have been measured, caused by mixing with air from aloft without ultrafine particles. Thus, ultrafine particles measured at the ground were produced within the PBL.

The ground-based measurements show similarities in the shape of the contour plot, i.e. the normalized number size distributions look similar for all sites. A more detailed investigation shows significant differences in the process itself, such as nucleation, growth rates and number concentration of produced particles. Thus the process is still influenced by the different local conditions but the result looks qualitatively similar.

4.2. *Horizontally inhomogeneous new particle formation*

As an example for horizontally inhomogeneous new particle formation 11 June will be presented here. The meteorological situation in the considered area was mainly influenced by a ridge of high pressure, providing dry but mostly cloudy conditions. This cloudiness was responsible for a much lower diurnal temperature amplitude compared to the homogeneous case. The temperature varied between 10°C and 20°C.

4.2.1. Ground-based measurements. Figure 7 shows the evolution of normalized number size distributions on 11 June 2002 at the Panitzsch, Melpitz, and Collm site. Due to technical reasons, data from Panitzsch were available only from 08:00 CEST, and measurements at IfT were missing completely for this day. At the Melpitz site, the relative concentration of ultrafine particles increased at 08:00 CEST slightly and more strongly at 09:00 CEST beginning at very small diameters (4–5 nm) with subsequent particle growth. At 11:30 CEST the process of particle formation and growth seemed to be interrupted and the mean diameter of the normalized size distributions decreased. At 12:30 the process started again and the mode of newly formed particles grew. Some more shorter interruptions followed in the afternoon. Around 16:00 ultrafine particles below 10 nm were found again. Also at the Collm site new particle formation was observed, but there it started around noon with only one interruption from 15:00 to 17:00. In Panitzsch, no clear new particle formation was detected. The nucleation events at the two sites (Melpitz and Collm) showed on the first view no clear temporal correlation but the evolution after 12:00 seems to be very similar. It looks like being basically the same process at both sites which was somehow suppressed at Collm during morning.

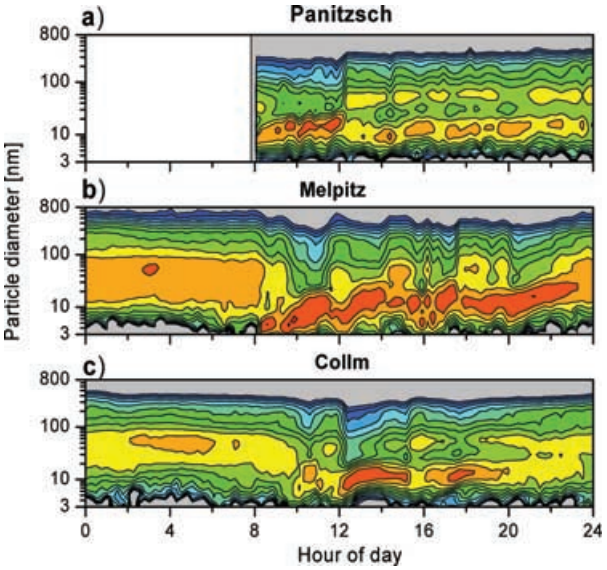

Fig. 7. Evolution of normalized number size distributions on 11 June 2002 at the stations Panitzsch, Melpitz and Collm; grey represents relatively low concentrations and red means high concentrations.

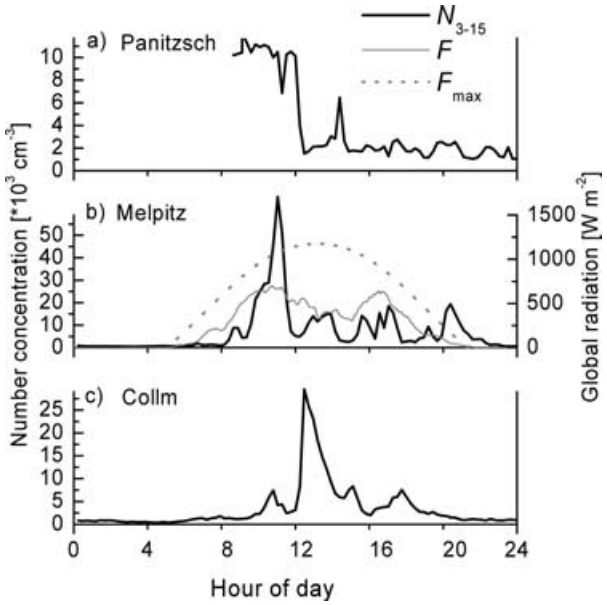

Fig. 8. Evolution of number concentration $N_{3–15}$ in the size range of 3–15 nm for the stations Panitzsch, Melpitz and Collm and global radiation F and maximum global radiation F_{max} in Melpitz on 11 June 2002.

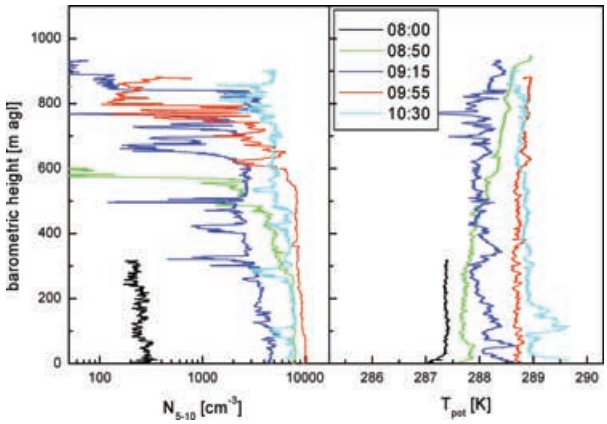

Fig. 9. Selected vertical profiles of $N_{5–10}$ in the size range of 5–10 nm (left-hand side) and potential Temperature T_{pot} (right-hand side) measured on 11 June in Melpitz. Times are in CEST and valid for the mean of profile.

This could be caused by the presence of clouds preventing the development of PBL and subsequently vertical mixing. Thus the whole process of new particle formation seemed to be ongoing in the area, but it was only occasionally observed at the ground stations. Back-trajectories identify the air mass as coming from the Atlantic Ocean, passing France and Western Germany. This air mass was more humid compared to the homogeneous case and is typical for an unstable weather situation, here represented by variations in cloud cover.

Figure 8 shows the evolution of particle number concentrations $N_{3–15}$. This presentation is suitable to emphasize absolute differences in number concentration between the sites. Additionally, the global radiation F measured in Melpitz is plotted together with the maximum global radiation F_{max} (without any atmospheric extinction or absorption). For the time period from 6:00 to 10:00 the difference between F and F_{max} was much smaller as compared with the following period indicating only minor cloud coverage. Between 10:00 and 15:00 $F \ll F_{max}$ with higher fluctuations in F, which is an indication for the occurrence of clouds. Furthermore, a slight positive correlation between F and $N_{3–15}$ was observed. That is, high cloud coverage decreased the observed formation rate.

4.2.2. Vertical measurements. Figure 9 shows the vertical profiles of $N_{5–10}$ and T_{pot} for 11 June between 08:00 and 10:30. Due to the cloud coverage the nocturnal radiative cooling is much less compared with the period of 3–5 June, therefore, the inversion layer is much less pronounced. Even for the first profile taken at 08:00 no clear inversion can be found in the profile of T_{pot}. For the same ascent $N_{5–10}$ ~300 cm^{-3} is nearly constant with height for the first 300 m. About 50 min later $N_{5–10}$ increased to values of 7000 cm^{-3}, the profile is again constant with height up to 580 m where $N_{5–10}$ sharply decreases to zero. The next two profiles (09:15 and 09:55) show a similar picture with $N_{5–10}$ ~ several 1000 cm^{-3} but the sharp jump is now around 800 m and $N_{5–10}$ shows strong fluctuations around this height due to mixing with cleaner air—with respect to ultrafine particles—from above this layer. In the last profile (10:30) no sharp decrease in $N_{5–10}$ is obvious, i.e. it is probably above the ceiling of the balloon. All profiles of T_{pot} show a near neutral stratification indicating a well-mixed PBL; no clear inversion correlates with the sharp jumps of $N_{5–10}$. Only the profile taken at 08:50 shows a slight increase of T_{pot} indicating a temporarily more stable situation.

Summarizing, for this day there is no classical PBL development observable and the momentary situation is more driven by the current cloud coverage.

5. Summary and Conclusions

Two different types of new particle formation events are shown which are typical for horizontally homogeneous and inhomogeneous nucleation events in terms of case studies out of the SATURN campaign 2002.

For the homogeneous case nucleation occurs nearly simultaneously at four measurement sites which are separated by approximately 50 km. Due to a stable synoptical situation mainly influenced by a high pressure system nucleation was observed for three consecutive days.

Vertical measurements performed at one rural site during two of the three days indicate that nucleation started at or above the nocturnal inversion layer. We may speculate that mixing processes at the inversion play an important role and initiated the nucleation events. However, after the break-up of the inversion

nucleation and growth took place in the entire mixing layer of the PBL yielding increasing number concentrations.

At the top of the PBL the concentration of ultrafine particles shows strong fluctuation between values typical for the mixing layer and zero indicating mixing of ultrafine particle-free air from above into the well-mixed PBL. From this observation we conclude that the complete nucleation event took place inside the PBL and the ultrafine particles were produced locally with subsequent growth instead of being entrained from above the PBL.

On the other hand, the local production of ultrafine particles was observed at nearly the same time at four different locations. Therefore, such new particle formation events can be seen as a large-area phenomenon. Since the typical production period of ultrafine particles was in the order of 1 hr but the subsequent growth could be observed for at least 10 hr we conclude that these events take place on a much larger area than covered by the four measurement sites. Assuming a typical wind velocity of 10–40 km h^{-1} for the 3 d the observed particles in the accumulation mode are transported with the mean wind for a distance of at least 100–400 km. Thus, the new particle formation contributes significantly to the total particle number concentration on the mesoscale. In the presented cases the newly formed particles grew to 60–80 nm and thus they might influence the mesoscale climate, because their concentration exceeds that of background aerosols significantly.

However, a few minor differences were also observed for the three rural sites compared with the more urban influenced site:

(i) Whereas at the urban site the condensation sink (*CS*) showed always a decrease before a nucleation event, *CS* in the rural areas was generally much lower and, therefore, a decrease of *CS* is not a need for a nucleation event at continental background sites.

(ii) The observed nucleation events started always earlier at the three rural sites compared with the urban site. One possible explanation for this behaviour is the compensating character of the city on the diurnal temperature development, i.e. the diurnal PBL development is less pronounced in a city compared to rural areas. Another reason could be the higher particle surface area concentration leading to higher *CS* and thus suppressing new particle formation.

For horizontally inhomogeneous conditions ultrafine particles have been found irregularly at the four stations and the subsequent particle growth was much less pronounced compared with the homogeneous case. According to the hypothesis that the observation of ultrafine particles near the ground results from downward mixing of new particles formed near the inversion layer (Nilsson et al., 2001a; Siebert et al., 2004), the occurrence of ultrafine particles is strongly connected with the PBL dynamics.

On the other hand, the local structure and dynamics of the PBL is mainly controlled by the incoming global radiation and, therefore, by the local cloud coverage. That is, advected cloud fields have a significant influence on the PBL and have the potential to suppress nucleation events in ground-based measurements. If these PBL-controlling conditions such as cloud cover are horizontally inhomogeneous, measurements at the ground will be horizontally inhomogeneous too.

We may safely conclude that under such inhomogeneous conditions the results of one-dimensional numerical models have to be interpreted with caution. In the horizontal case the process itself might be assumed to be representative for the whole area, however, individual parameters, such as nucleation rate or growth rate depend significantly on local conditions. However, these conclusions are valid for these case studies and should be validated with long term measurements.

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