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New particle formation and ultrafine charged aerosol climatology at a high altitude site in the Alps (Jungfraujoch, 3580 m a.s.l., Switzerland)

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Abstract

Aerosol nucleation is an important source of atmospheric particles which have an effect both on the climatic system and on human health. The new particle formation (NPF) process is an ubiquitous phenomenon, yet poorly understood despite the many studies performed on this topic using various approaches (observation, experimentation in smog chambers and modeling). In this work, we investigate the formation of secondary charged aerosols and its climatology at Jungfraujoch, a high altitude site in Swiss Alps (3580 m a.s.l.). Charged particles and clusters (0.5 – 1.8 nm) were measured within the EUCAARI program from April 2008 to April 2009 and allowed the detection of nucleation events. We found that the aerosol concentration, which is dominated by cluster size class, shows a strong diurnal pattern and that the aerosol size distribution and concentration are strongly influenced by the presence of clouds either during daytime or nighttime conditions. New particle formation events have been investigated and it appears that new particle formation occurs 17.5% of measured days and that the nucleation frequency is strongly linked to air mass origin and path and negatively influenced by cloud presence. In fact, we show that NPF events depend on the occurrence of high concentration VOCs air masses which allowed clusters growing by condensation of organic vapors rather than nucleation of new clusters. Furthermore, the contribution of ions to nucleation process was studied and we found that ion-mediated nucleation (IMN) contribute to 26% of the total nucleation so that ions play an important role in the new particle formation and growth at Jungfraujoch.

1 Introduction

Aerosols play an important role in the Earth radiative budget through their direct (Charlston et al., 1991) and indirect effect (Twomey, 1974). In fact, the influence of aerosols represents the highest uncertainty in understanding and modeling the climatic system and its future evolution (IPCC, 2007). One of the challenge of aerosol science is to describe precisely how new particles are formed from gaseous precursors. The formation of those secondary aerosols have been studied by many researchers but if the general mechanism is established (gas – particle conversion), predicting where and when the new particle formation will take place remains difficult in natural conditions. Long term aerosol measurements in different types of environments is still a valuable approach to better understand which conditions promote the new particle production in the atmosphere. New particle formation events have been observed in various environments (see Kulmala et al., 2004, for a review) from polluted area (Hämeri et al., 1996; Harrison et al., 2000; Woo et al., 2001; Stanier et al., 2004; Dunn et al., 2004) to clean or rural sites (Weber et al., 1997; Mäkelä et al., 1997; O'Dowd et al., 1998; Suni et al., 2008), polar areas (Weber et al., 2003; Asmi et al., 2009b) and high altitude sites (Weber et al., 1995; Venzac et al., 2007; Shaw, 2007; Nishita et al., 2008; Rodriguez et al., 2009). It has been recently proposed that nucleation was promoted at high altitude (Venzac et al., 2008). High altitude sites allows the study of aerosols in both free tropospheric conditions and boundary layer conditions. During the warmer season, the Jungfraujoch is influenced by injections of air parcels from the planetary boundary layer (PBL): Air masses from alpin valleys and the Swiss plateau from lower altitudes are transported to the site by thermal convection (Nyeki et al., 1998). During such days, pronounced diurnal cycles are measured for various aerosol parameters (Lugauer et al., 2000), since the PBL influence disappears during colder parts of the day. During winter, this effect is hardly find, and the air masses present are usually representative for the free tropospheric background conditions.

Here we discuss the physical observation of new particle formation events at a high altitude site in the Alps (Jungfraujoch, 3580 m a.s.l.) during the EUCAARI intensive observation year 2008–2009 (Kulmala et al., 2009). Ultrafine charged aerosol size distributions were measured and the influence of atmospheric conditions and air mass origins examined.

2 Description of the measurement site, data and method

Jungfraujoch is situated on the northly crest in a saddle between the mountains Mönch (4099 m a.s.l.) and Jungfrau (4158 m a.s.l.), and belong to the glacier accumulation zone. Aerosol measurements were performed at the Sphinx laboratory located on the southern side of the Jungfraujoch at 3580 m a.s.l. (46°32′51″ N, 7°59′6″ E), Switzerland, 100 m below the main crest of the Bernese Alps. Jungfraujoch is a station of the Global Atmosphere Watch (GAW) program. Therefore, among parameters, the total aerosol number concentration, light absorption and scattering coefficient at various wavelength are routinely measured at the site (Collaud Coen et al., 2007). Additionally, the size distribution between 12 and 570 nm is measured by a custom built SMPS. Meteorological parameters are monitored at the Swiss National Monitoring Network for Air Pollution (NABEL), located within a horizontal distance of 150 m, at the top of the crest (3580 m a.s.l.). The monitored parameters include thirty minute averaged concentrations of NO, NO_2, NO_x and O_3, and daily averaged concentrations of SO_2, aerosol sulfur, and particulate matter with aerodynamic diameter below 10 µm (PM_{10}).

The campaign dataset is composed of 309 days starting from the 9 April 2008 and to the 5 May 2009 with 83 days of interruption due to instrumental failures. The measurements are performed from an instrument which detects neutral particles, negative and positive atmospheric ions selected according to their electrical mobility.

2.1 The neutral aerosol and atmospheric ion spectrometer (NAIS)

The mobility distributions of atmospheric positive and negative ions are measured with a Neutral Air Ion Spectrometer developed by AIREL Ltd., Estonia (Mirme et al., 2007). This instrument provides alternatively the electrical mobility distribution of both negative and positive ions and of neutral particles in the mobility range 3.16 to 0.0013 $cm^2\ V^{-1}\ s^{-1}$. The NAIS sampling principle is based on the simultaneous selection of 21 different mobilities of atmospheric ions of each polarity (negative and positive) along two differential mobility analyzers and their subsequent simultaneous detection

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using isolated electrometers in parallel. The conversion from electrical mobility to diameter is calculated according to the Tammets inversion (Tammet, 1995). For the mean local pressure and temperature, the NAIS can collect ions in the diameter range 0.5 to 49 nm.

For the detection of neutral particles, an additional pair of unipolar charger and electrostatic post-filter were added to the inlet section after the ion offset section of the NAIS device. Once ions are filtered, neutral particle are charged by ion currents using a corona discharged and analyzed according the same process as the ion measurement mode. The role of the additional electrostatic post-filter is to cut off the concentration of corona ions used to charge the particles in the charger. Previous study defined the limit of the neutral particle detection down to 2 nm. Below this size, particles measurement are not relevant since the post-filtering process affects the sampled newly charged particles (Asmi et al., 2009a). The NAIS sampling is performed through a specific inlet (length= 300 mm , diameter= 32 mm, flow rate=10^3 cm^3 s^{-1}), directly through a window of the station. The design of the inlet was optimized to minimize diffusion losses and recombinations. The size-cuts of large charged or neutral particles sampled through the NAIS inlet are respectively 10 µm and 2 µm for wind speeds of 2 m s^{-1} and 5 m s^{-1}. As a result, few droplets could enter the inlet, except at wind speed smaller than 5 m s^{-1}. This phenomenon could lead to an overestimation of ions number concentration since impaction of droplets could lead to ions production. Furthermore to avoid freezing of inlet, we add a heating system which automatically switches on when the temperature is below 0 °C.

2.2 Data analysis

2.2.1 New particle formation events classification

A new particle formation event, as previously described by Dal Maso et al. (2005), must present four criteria: “1. A distinctly new mode of particles must appear in the size distribution, 2. the mode must start in the nucleation mode, 3. the mode must prevail over

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a time span of hours and 4. the new mode must show signs of growth". The classification of event days was performed visually using the daily contour plot of the ion size distribution evolution. Data were first categorized into three main classes : undefined, non-event and NPF event days. Since different types of NPF can be observed, event days were classified into different classes (Ia, Ib, II and Bump) according to their quality and their applicability to a growth rate analysis (Hirsikko et al., 2007):

- Ia : Continuous growth of clusters (0.4 nm) to large particle (≥ 20 nm).

- Ib : These events are not as strong as class Ia events and sometimes cluster or intermediate growth are not clearly visible on the size distribution but the growth rate calculation remains possible.

- II : A clear event is identify but the growth from clusters to large particle is not regular and the shape of the size distribution is unclear. Further analysis of the new particle formation characteristics are complex.

- Bump : A burst of clusters is detected but it is not followed by a significant growth and particle formation. Different explanation are possible such as the total consumption of the condensing vapors or a change in the air mass.

2.2.2 Growth rates estimation

The new particle formation process can be described by different steps of growth. First, the smallest particles concentration increase until a local maximum in diameter space. As the particles grow in size, the local maximum shifts to larger sizes. In the present work, a normal distribution is fitted to different size-class concentration maxima using a trust-region algorithm (Byrd et al., 1987) by minimizing the least square residue (Fig. 1). The growth rate can be deduced from the time needed to shift from the maximum of one diameter population to another as proposed by Hirsikko et al. (2005).

The charged particles growth rates were calculated for three different size classes: the cluster size (1.3 – 3 nm), intermediate size (3 – 6.8 nm) and large ion size (7 –

20 nm). GRs were computed for class Ia and Ib NPF classes. However, for some class Ib days, GRs calculation were not possible due to local pollution event, change in air masses or NPF interruption by clouds. Those days were not taken into account in the growth rate analysis. Furthermore, the effect of coagulation on the size evolution was not included in the GR analysis since its effect is negligible (Manninen et al., 2009).

2.2.3 Formation rates calculation

In order to characterize the new particle formation events, it is also important to determine the rate of particles formed during the event, i.e. the formation rate of particles which diameter is of i nm (J_i), and especially during the first steps of particle's growth, from 2 to 3 nm (Kulmala et al., 2001). The formation rate can be calculated from particle concentration in the size range from 2 to 3 nm (N_{2-3}), the growth rate of 2 nm particles which is assumed to be the same over the whole size class 1.3 – 3 nm ($GR_{1.3-3}$) and the loss of particle by coagulation scavenging of 2 nm particles on larger pre-existing particles $CoagS_2$ (Kulmala et al., 2007).

$$J_2 = \frac{dN_{2-3}}{dt} + CoagS_2 \times N_{2-3} + \frac{f}{1\,nm} GR_{1.3-3} N_{2-3} \quad (1)$$

The formation rate of charged particles in the size range of 2 – 3 nm, i.e. the charged formation rate $J_2^{\pm}$ for both polarity can be calculated from the charged aerosol size distribution obtained from the NAIS. To take into account the loss of ions by ion-ion recombination and the attachment of ions to neutral particles, two terms were added to Eq. (1) :

$$J_2^{\pm} = \frac{dN_{2-3}^{\pm}}{dt} + CoagS_2 \times N_{2-3}^{\pm} + \frac{f}{1\,nm} GR_{1.3-3} N_{2-3}^{\pm} + \alpha \times N_{2-3}^{\pm} N_{<3}^{\mp} - \beta \times N_{2-3} N_{<2}^{\pm} \quad (2)$$

where $N_{2-3}^{\pm}$ is the ion number concentration (positive or negative ions) [# cm^{-3}] in diameter range from 2 to 3 nm and $N_{<x}^{\pm}$ is the ion number concentration below x nm.

$CoagS_2$ is the condensation sink of 2 nm particles [s^{-1}], $GR_{1.3-3}$ is the growth rate for the 1.3 – 3 nm size class [$nm\,h^{-1}$]. α and β are respectively the ion-ion recombination coefficient and the ion-neutral attachment coefficient and were assumed to be equal respectively to $1.6 \times 10^{-6}\,cm^3\,s^{-1}$ and $1 \times 10^{-8}\,cm^3\,s^{-1}$ (e.g., Manninen et al., 2009). The factor f represents the fraction of the ion population in a size range from 2 to 3 nm which are activated during the growth. In this study we assumed this factor to be equal to unity. The time derivative of N_{2-3} is directly obtained from the NAIS measurements. $CoagS_2$ is derived from SMPS and NAIS data.

2.2.4 Air mass analysis

The impact of the air mass origin and trajectory on the charged aerosol background and on the potential of nucleation is studied after calculation of three days air mass back trajectories using the HYSPLIT transport and dispersion model (Draxler and Rolph, 2003). The calculation is performed over the whole measurement period every 24 h at 00:00 and 12:00 Local Time (LT). Since the air mass origin and path to the measurement site do not differ significantly between 00:00 and 12:00, only results for 00:00 will be included in our analysis.

3 Results

3.1 Ultrafine charged aerosol climatology

The median diurnal variation of total ion concentration (Fig. 2) presents a strong pattern. During the night, the concentration of ions is quite stable with a mean concentration of 678 # cm^{-3} for negative ions and 709 # cm^{-3} for positive ones. From 06:00 LT, the concentration of both polarities increase until reaching a maximum of 825 # cm^{-3} and 875 # cm^{-3} respectively for negative and positive ions at 13:00 LT. Then the concentrations decrease rapidly from 15:00 to 18:00 LT to reach the level of night time.

Night time conditions have been described to be representative of Free Tropospheric (FT) conditions at high altitude sites. Baltensperger et al. (1997) showed that diurnal variations of aerosol parameters such as surface area are due to thermally driven vertical exchange. In their article, Lugauer et al. (2000) have demonstrated that during winter the Jungfraujoch is most of time decoupled from the planetary boundary layer (PBL) and, during the summer, air from the PBL was transported to the site by thermally driven convection. Zellweger et al. (2000) was more restrictive and showed that during summertime, night time measurements with northwesterly advection are considered to represent FT conditions according to the NO_y speciation. Another study made by Forrer et al. (2000) confirms the transportation of some gases (CO) is made by thermally driven vertical transport which occur during the day time. Finally, night time measurements from 03:00 to 09:00 were considered representative of FT conditions for Jungfraujoch, (Weingartner et al., 1999) but also for other high altitude sites (Puy de Dôme, Venzac et al., 2007 and high altitude Himalaya, Venzac et al., 2008). Considering those previous works and the diurnal variation observed at Jungfraujoch, the data set has been segregated into two sub data set composed of night time measurements on one hand, and the day time measurements (from 09:00 to 18:00 LT) on the other hand.

The total ion concentration measured at night (Fig. 2) can considered as a FT ion nighttime background while the increase of ion concentration is likely due to advection of ion sources from the planetary boundary layer. Ions are classified in three different size classes: the cluster ions from 0.5 to 1.8 nm, intermediate ions from 2.1 to 6.8 nm, and large ions from 8 to 47 nm.

Those three size classes are representative of three steps in the aerosol lifetime: clusters are considered as embryos for new particle formation and growth, intermediate ions are linked to the new particle formation and growth process; (see Venzac et al., 2007, 2008), lastly large ions are the disconnected from the new particle formation event and could be linked to external incoming of polluted air parcels. Total ions concentration variations are dominated by the cluster ions concentration, which drive the observed diurnal variation. Previous studies of ion diurnal variations made by Hõrrak et al. (2003) showed that radon is the main ionizing agent responsible of the cluster ion formation in the PBL. Gäggeler et al. (1995) showed that radon concentration at Jungfraujoch is dominated by transport and not by local production since the ground is most of time covered by snow. As according to those previous results, we can assume that the diurnal variation of the cluster ions concentration is mainly due to the vertical transport of high radon concentration air masses from the planetary boundary layer during the day and to atmospheric ionization from galactic cosmic rays (GCR) during the night. Assuming that, we conclude that radon is a higher source of ions than the galactic cosmic rays (GCR) at Jungfraujoch.

A weak seasonality of total ions concentration, largely dominated by clusters, was found, opposing fall and winter (no significant difference between them) to summer (Fig. 2). In fact, summertime shows lower ions concentration than fall and winter even if the increase of ions concentration begins sooner in the daytime. This result is in agreement with the analysis performed by Weingartner et al. (1999) who pointed out that 10 – 18 nm neutral particles concentrations showed a maximum during wintertime. A closer look at the diurnal variation of cluster ions shows that summertime cluster ions are lower than winter time cluster ion concentrations especially during the 15:00–24:00 time range. Hence, it is likely that the high concentrations of larger particles updrifted at the station during summertime aroud this time of the day represent a significant condensational sink for the cluster ions.

The seasonal variation of intermediate ions is also showing a minimum for summer time. During winter, intermediate ions strongly peak between 12:00 and 15:00. This peak could be linked to the presence of clouds.

3.2 Cloud effects

Clouds were shown to have a significant effect on ion clusters and intermediate ion concentrations and on NPF occurrence (Venzac et al., 2007; Lihavainen et al., 2007). Consequently, we chose to further separate potential cloudiness conditions (RH$\geq$ 96%)

from clear sky conditions and analyze them separately to better characterized the cloud effect on aerosol. The limit value of RH used to distinguish clear sky and cloud conditions was validated on Puy de Dôme data (unpublished data). Mean ion concentration variations are reported on Fig. 3. In order to determine if differences are statistically significant between our sub-classes (nighttime/daytime, cloud/clear sky, positive/negative polarity), a T-test was performed on the data set (Table 1)

For the cluster size classes, ion concentrations decrease by a factor of 1.5 to 2.5 when the conditions change from clear sky to cloudiness (Fig. 3a). Intermediate ions concentrations are, on the contrary, increased by the presence of a cloud, especially for the negative ions. The effect of clouds on clusters and intermediate ions is in agreement with the results reported by Venzac et al. at the Puy de Dôme station (Venzac et al., 2007) and Lihavainen et al. (2007).

Concerning the large ion mode, as expected for this size, negative and positive ion population are close to equilibrium under clear sky conditions. In the presence of a cloud, the two populations are increased by a factor 1.4 (41%) and 1.2 (20%) respectively for positive and negative ions. Figure 3b is showing the average ion concentrations (as opposed to the average ion concentrations shown Fig. 3a). The clear sky concentrations are not significantly different from the median concentrations. However, the in-cloud average ion concentrations are different from median ion concentrations, indicating that intense sporadic events are taking place under cloudy conditions. From the comparison between median and average ion concentrations in cloud, we conclude that cloud conditions favor the production of sporadic high concentrations of cluster and intermediate ion positive and especially negative ions. Cluster ions are representative of clusters particles which could be activated into intermediate particles if a new particle event occur. Consequently, in the following section, we will investigate the relationship between cluster and intermediate ion concentrations and new particle formation events. First a short statistical analysis of NPF events will be performed.

3.3 New particle formation event analysis

3.3.1 Event statistics

The 309 days of data were analyzed and classified into event, non-event and undefined days (Sect. 2.2.1.). Undefined days (25 days) represent only 8.1% of the data, 73.8% of observed days were classified as non-event days (228 days) and among those days, 59% (135 days) were classified as "in cloud" conditions. NPF events were observed on 17.5% of days (54 days). Monthly statistics are shown on Fig. 4 and class occurrence of NPF event according to Hirsikko et al. (2007) are presented Table 2. New particle formation frequencies present a clear seasonality with a minimum of events during winter and maximum from spring to autumn. The maximum of events occurred in April 2009 (28.6% of observed days present an event) and the minimum during December 2008 (3.6% of observed days). At other sites where a NPF events seasonal variation could be studied, the maximal occurrence was usually observed during the spring and autumn seasons (Manninen et al., 2009), for the summer season for altitude sites (Venzac et al., 2008; Boulon et al., 2010) while the minimal occurrence frequency is always observed during winter months (Venzac et al., 2008; Manninen et al., 2009). The seasonal variation of the frequency of NPF events is opposite to the seasonal variation of ion cluster concentrations mentioned Sect. 3.1. This is a first indication that the formation of cluster ions by nucleation is not a limiting factor for NPF events at the Jungfraujoch station.

The presence/absence of cloud during NPF days was checked for all the NPF days and it appears that clouds reach the measurement site during the NPF process for only 4 days (7.1% of NPF observed days). Furthermore, when a cloud occurs during the event the nucleation/growing process is stopped. Considering the low frequency of nucleation during cloudy conditions and the fact that cloud interrupts the NPF process we conclude that the cloudy conditions inhibit the new particle formation process. This phenomenon has to be linked with 1. the lack of production of reactive species by photochemical processes enhancing the new particle formation and 2. the scavenging

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property of cloud droplets which could remove clusters or/and condensable vapors from the atmosphere (Baltensperger et al., 1998).

3.3.2 Growth rates analysis

Growth rates and formation rates were calculated for Ia and Ib events (detailed growth rates and formation rates are reported Table 3.

Differences were observed according to the event class but also, more surprisingly, according to the polarity for the formation rates. The mean growth rate for Ia and Ib for size class 1.3 – 3, 3 – 7 and 7 – 20 nm are respectively [5.3, 7.8, 5.4] and [5.0, 4.1, 5.9] nm h^{-1}. According to the type of event the growth rate variation is quite different. For Ia class, charged particle growth seem to increase more rapidly for the 3 – 7 nm size class and then slowing down in the 7 – 20 nm range, reaching a value similar to the 1.3 – 3 nm size class. This behavior of particle growth is not the same for Ib type event. In fact, the growth rate of charged particle in the size range from 3 to 7 nm is lower than the one for the two other size classes which is consistent with the Ib class definition (see Hirsikko et al., 2007). Concentration of condensable vapor and source rate were calculated from GRs values according to Dal Maso et al. (2002), detailed results are reported Table 3. Source rates at Jungfraujoch are relatively low ($5.12 \pm 3.8 \times 10^3$ cm^{-3} s^{-1}) compared to those observed in the boreal forest or in coastal environment (from 1.1 to 52×10^5 cm^{-3} s^{-1}). This difference is not surprising since boreal forest and coastal environment are known to have biological sources of condensable vapors as VOC emitted from the vegetation or iodine compounds emitted from exposed sea weed fields.

In Hyytiälä, growth rates are respectively 1.9, 3.6 and 4.2 nm h^{-1} for the size classes 1.3 – 3, 3 – 7 and 7 – 20 nm (Manninen et al., 2009). It is surprising that the Jungfraujoch GRs are higher than at a high volatile organic compound (VOC) concentration sites such as the boreal forest. On another hand, the condensational sink ($\bar{CS} = 2.6 \pm 1.3 \times 10^{-5}$ cm^{-3} s^{-1}) at Jungfraujoch is so low that even limited concentration of VOC can trigger new particle formation easily. Mean values of growth rates for mountain sites present a large scale of variation due to local biogeography. Venzac et al. (2008) have detected NPF events on Everest (5079 m a.s.l.) with an AIS during February–March 2007 and the mean GR value was estimated at $1.8\pm0.7\,\mathrm{nm\,h^{-1}}$. Mean value for puy de Dôme is $5\pm3.5\,\mathrm{nm\,h^{-1}}$ for the period between March 2006 and December 2007 (Venzac, 2008), at Mt. Norikura, Japan (2770 m a.s.l.), Nishita et al. (2008) have calculated a mean GR value of $2.6-3.1\,\mathrm{nm h^{-1}}$ between September 2001 and August–September 2002 measurement period. Shaw (2007) reports higher values of GR, $10-23\,\mathrm{nm\,h^{-1}}$, at Mt. Lemmon, Arizona (2790 m a.s.l.). Those last high values of growth rate are explained by the authors as the result of high organic vapor concentration from desert vegetation associated with high UV-A radiation. We can conclude that our growth rates values are consistent with other data from similar high altitude sites across the world. The variation of GRs with particle radius is inverse of the one usually observed (see Manninen et al., 2009), this is explained by inhomogeneities in the air masses: FT at 09:00 and PBL at 12:00 for example.

3.3.3 Charged and neutral particle formation rates: the role of ions in nucleation at Jungfraujoch

Ions could lead to significant particle formation as previously showed by Arnold (1980); Yu and Turco (2001). In order to quantify the role of ions in the nucleation process at Jungfraujoch, we compute the charged formation rate, the total particle formation rate and the ion-mediated rate for 2 nm particle.

The analysis of the 2 nm charged particle formation rates ($J_2^{\pm}$, Table 3) shows that negatively charged particle formation rate is always higher than the one of positive particles whatever the class of event (Ia or Ib). Within the accuracy of the method, the total particle formation rates J_2 (Eq. 1) were estimated from the data provided by the neutral mode of measurement of the NAIS. This was done in order to quantify the importance of the ion-mediated nucleation in this environment. We found that the median value of J_2 was less than four times bigger than the $J_2^{\pm}$ median value (Table 3). In other environment such as boreal forests, J_2 is at least one order of magnitude bigger than the charged particle formation rate (Manninen et al., 2009). This could indicate that ion-mediated nucleation is quite important at Jungfraujoch. In order to investigate the role of ions in nucleation, we have calculated the fraction of ion-mediated nucleation rate (IMN) according to Eq. (3):

$$\mathrm{IMN} = \left(\frac{\delta \times \alpha \times N_i^+ \times N_i^-}{\mathrm{CoagS}_2} \right) \times \frac{1}{N_{\mathrm{total}}} \quad (3)$$

where δ is the probability that a collision between two ions of both polarities leads to the formation of a stable neutral cluster (here we assume that $\delta = 1$), CoagS_2 is the coagulation sink for 2 nm charged particles, α is the ion-ion recombination coefficient, $N_i^{\pm}$ is the concentration of ions which size range were selected in order that the diameter of the resulting neutral diameter was in size range 2–3 nm and N_{total} is the total concentration. IMN shows a large range of values depending of the event day. It varies from 6% to 74% with a mean of 26 ± 23%. The median value of 19% is higher than the one computed by Manninen et al. (2010) for the boreal environment suggesting that IMN is an important phenomenon for new particle formation events at Jungfraujoch. The median value of J_2^+, J_2^- and J_2 at Jungfraujoch are respectively 0.19, 0.28 and 2.03 $\mathrm{cm}^{-3}\,\mathrm{s}^{-1}$. According to Manninen et al. (2009), $J_2^{\pm}$ found at Jungfraujoch is similar to the $J_2^{\pm}$ computed for many other sites in Europe, while it is the J_2 found at Jungfraujoch which is significantly lower than the J_2 found at other places in Europe. The fraction ion-mediated nucleation is hence higher than at other sites because of the lower neutral nucleation detected at this site. It is worth mentioning that our J_2 calculation is a higher limit, since neutral 3 nm particles concentrations detected with the SMPS technique were measured to be lower than the 3 nm particles detected from the NAIS. According to those results, we can assume that ion-mediated nucleation is a significant source of new particle in the troposphere as previously shown by Yu et al. (2008) for boundary layer conditions.

3.4 Air mass origin analysis

In this subsection, we investigate the impact of the air mass origin on the occurrence of the new particle formation process. Air masses were classified according to their geographical origin with a resolution of $10^{\circ} \times 10^{\circ}$. Five different classes were created: atlantic, african, nordic, eastern and western european air masses. Atlantic (68.5%) and european continental origin (respectively 6.4% and 14.2% for eastern and western Europe origin) represent the highest proportion of air mass origin ending at Jungfraujoch (89.1%) followed by african (7.5%) and polar (3.4%) air masses. We calculate a new particle formation event frequency for each air mass (Table 4) and it appears that air mass from Eastern europe (latitude:]40 N; 70 N[and longitude: [20 W and more]) have the highest probability to lead to a NPF event, while the lowest probabilities of new particle formation event are found in the air masses from western Europe (latitude: 40 N; 70 N, longitude:10 W; 20 W; if latitude < 60 N, else longitude: 0; 20 W) and polar area (latitude ≥ 70 N). This is rather different from air mass dependencies found in the boreal forest, where polar air masses were found to favor NPF event as shown by Sogacheva et al. (2005). In the boreal forest where high concentrations of condensable species are emitted from the vegetation, the absence of a preexisting condensational sink seems to be a strong condition for the occurrence of NPF events. In the case of the Jungfraujoch, which can be described as a low preexisting condensational sink environment, , the presence of condensable vapors from polluted areas (typically from Eastern Europe) seems to prevail in the occurrence of NPF events.

This hypothesis can be partly examined by studying the dependency of the charged particle size distribution on the air mass origin. Mean size distributions at different key moments of the day where calculated under clear sky conditions, in order to exclude air mass related to cloud effects (Fig. 5).

3.4.1 Non event days

We clearly observed from Fig. 5, that while the cluster mode does not experience any variation according to the air mass origin, the intermediate ion mode shows a stronger variability. During non-event days, the size distribution of charged aerosol particle (Fig. 5a) show a weak diurnal variation in all air mass types, with higher concentrations of intermediate and large ion size classes during the afternoon compared to night. This pattern is most likely due to the updraft of charged particles or ion sources (i.e. radon) form the valley by thermally driven convection.

The shape of the size distributions is quite similar from one air mass origin to the other, the more important differences are found in the population of the intermediate mode between 2 and 4 nm. In fact, in Nordic air masses we observed the most concentrated intermediate ions mode (see Table 5) whereas in air masses from eastern Europe, concentrations are the lowest. In other air masses, concentrations are similar one to the other. The situation is very different for event days.

3.4.2 Event days

During the new particle formation event days, size distributions of ions and charged particles are very different from those of non event days even at nighttime (between 03:00 and 06:00 LT, Fig. 5b). During the NPF process (between 09:00 and 12:00) the cluster mode concentration increases and its geometric mean diameter shifts to larger diameters (typically from 0.8 nm to 1 nm or more). The NPF is also characterized by a decrease of the intermediate ion concentrations which grow to sizes beyond the upper diameter bound of the instrument. The impact of NPF on the size distribution can be evaluated by comparing large ion number concentration at 15:00–18:00 for event days and non event days (Table 5).

Concentration of ions which diameter is higher than 8 nm are significantly higher when a new particle formation event has occurred except when air masses come from the nordic areas. This is probably due to the fact that NPF linked to nordic air masses

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are not type I events i.e. not intense or not complete. This result reveals that new particle formation events could increase the number concentration of particles of the free troposphere as already suggested by Sellegri et al. (2010). At the NCO-P station (5079 m a.s.l., Nepal), they have shown that new particle formation events occur with a frequency close to 50% of observed days and that upper free troposphere residual layer aerosol composition could be strongly influenced by NPF events.

A T-test was performed to determine whether there is a significant change of the 03:00–06:00 concentration when it precedes NPF events or not. The result is negative, in other terms, 03:00–06:00 cluster concentrations are not significantly different between event and non event days. This result is a second indication according to which the preexisting cluster concentration is not a driving parameter for the occurrence of a nucleation event. We conclude that nucleation occurs when condensable vapor concentrations are high enough to activate the clusters' growth. This interpretation is in agreement with the high nucleation potential of air masses from Eastern Europe which is known to be an hotspot of non-methane volatile organic compounds (Lanz et al., 2009). These observations confirms the conclusion from the study of Lehtipalo et al. (2009), in which it is shown that in some environment, NPF are driven by condensation of organics rather than by nucleation – activation of new clusters. Furthermore, this result can be linked to results obtained by Metzger et al. (2010) who show that the concentration of organic vapors plays a key role in the nucleation and growth process.

4 Conclusions

The ultrafine charged aerosol concentration variability and new particle formation events were studied using a neutral air ion spectrometer within the EUCAARI field campaign 2008–2009 at the Jungfraujoch research station in the Swiss Alps. 309 days starting from the 9 April 2008 to the 5 May 2009 were analyzed.

A strong diurnal pattern of the total ions concentration, dominated by cluster ions, was found maximum during the day and minimum during the night. This diurnal variation was related to the updraft of surface layer air parcels rich in preexisting particles and ion sources such as radon from the valley during the day. Radon was found to be likely the main cluster ion source at the station. The ultrafine charged aerosol concentration also shows a weak seasonality with minimum concentrations observed during summer. This finding is likely due to the high condensational sink due to the up-draft of large particles during this season and time of the day.

We performed a statistical analysis of events and found that event days represent 17.5% of measured days and a seasonality pattern was pointed out with maximum of event during spring and autumn and minimum during winter. This seasonality is inverse to the one of cluster ion concentrations. This is a first indication that cluster concentration is not a driving parameter for NPF events. The NPF frequency is quite low compared to other sites, including high altitude sites. The site is often under cloudy conditions, which inhibit NPF events. When the measurement site is in cloudy conditions, we showed that the cluster ion concentrations decreased by a factor 1.5 to 2.5 when the conditions change from clear sky to cloudiness. Because cloud droplets efficiently scavenge ions and particles, and presumably because photochemistry is inhibited, NPF were observed to occur only 7.1% of the time under cloudy conditions.

The ion-mediated nucleation seems to play an important role in the new particle formation process at this measurement site since ions and recombination products explain 26% of the particle formation.

The NPF are characterized by mean growth rates of [5.1, 6.0, 5.7] nm h^{-1} respectively for size class 1.3 – 3, 3 – 7 and 7 – 20 nm which are quite high compared to those found at other altitude sites (Venzac, 2008; Venzac et al., 2008). The high growth rate are attributed to low condensational sinks rather that high condensation gas sources, which where calculated to be two orders of magnitude lower than in boreal or coastal environments.

NPF were also studied as a function of air mass origins origin according to the HYSPLIT model and we found that NPF event frequency is strongly linked to the origin of the air mass but not in term of clusters concentration which remain quite stable ac-

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cording to different origins. In fact, air masses from eastern europe show to have the highest nucleating potential at Jungfraujoch. This observation can be linked to previous studies having classified those air masses as high VOC concentration air masses. This result confirms that the nucleation process at Jungfraujoch depends of the presence of condensable vapors which allowed clusters to grow rather on nucleation of new cluster particles.

Appendix A

New particle formation characteristics are reported on the table below (Table 3). Only class Ia and Ib event were studied.

A T-test was performed in order to know if the difference observed on ion concentrations between positive/negative polarity, daytime/nighttime conditions or clear sky/cloudiness conditions is significant. Null hypothesis H_0 is defined as follow : the two data sets are independent random samples from normal distributions with equal means and equal but unknown variances. If H_0 is not rejected at the 5% significance level, we assumed that there is no significant difference between the values of the mean of the two data sets (T-test= 0). If H_0 is rejected at the 5% significance level, we assumed that there is a significant difference between the values of the mean of the two data sets (T-test= 1). Results of different T-tests are reported on Table 1.

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Table 1. T-test results for "diurnal vs. FT" and "clear sky vs. cloudy" conditions for different size classes.

T-test	Jul 08	Aug 08	Sep 08	Oct 08	Nov 08	Dec 08	Jan 09	Feb 09	Mar 09	Apr 09
					Clust.					
D^{+}_{clear} vs. D^{-}_{clear}	0	1	1	1	1	1	1	1	1	1
D^{+}_{cloud} vs. D^{-}_{cloud}	1	0	0	0	0	1	0	0	0	0
D^{+}_{clear} vs. FT^{+}_{clear}	0	0	0	0	0	0	0	0	0	0
D^{-}_{clear} vs. FT^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
D^{+}_{cloud} vs. FT^{+}_{cloud}	0	0	0	0	0	1	0	0	0	0
D^{-}_{cloud} vs. FT^{-}_{cloud}	1	1	1	1	1	1	1	1	1	1
D^{+}_{clear} vs. D^{+}_{cloud}	0	0	0	0	0	0	0	0	0	0
D^{-}_{clear} vs. D^{-}_{cloud}	1	1	1	1	1	1	1	1	1	1
FT^{+}_{clear} vs. FT^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
FT^{+}_{cloud} vs. FT^{-}_{cloud}	0	1	1	1	1	1	1	1	1	1
FT^{+}_{clear} vs. FT^{+}_{cloud}	0	0	0	0	0	0	0	0	0	0
FT^{-}_{clear} vs. FT^{-}_{cloud}	1	1	1	1	0	1	1	1	1	0
					Inter.					
D^{+}_{clear} vs. D^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
D^{+}_{cloud} vs. D^{-}_{cloud}	1	1	0	1	0	1	1	0	1	0
D^{+}_{clear} vs. FT^{+}_{clear}	0	0	0	0	1	0	0	1	0	1
D^{-}_{clear} vs. FT^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
D^{+}_{cloud} vs. FT^{+}_{cloud}	1	1	1	1	1	1	1	1	1	1
D^{-}_{cloud} vs. FT^{-}_{cloud}	1	1	1	1	1	1	1	1	1	1
D^{+}_{clear} vs. D^{+}_{cloud}	1	1	1	1	1	1	1	1	1	1
D^{-}_{clear} vs. D^{-}_{cloud}	1	1	1	1	1	1	1	1	1	1
FT^{+}_{clear} vs. FT^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
FT^{+}_{cloud} vs. FT^{-}_{cloud}	1	1	1	1	1	1	1	1	1	1
FT^{+}_{clear} vs. FT^{+}_{cloud}	0	1	1	1	1	0	0	1	1	1
FT^{-}_{clear} vs. FT^{-}_{cloud}	1	1	1	1	1	0	1	1	1	1

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Table 1. Continued.

T-test	Jul 08	Aug 08	Sep 08	Oct 08	Nov 08	Dec 08	Jan 09	Feb 09	Mar 09	Apr 09
					Large					
D^{+}_{clear} vs. D^{-}_{clear}	0	0	0	0	0	0	0	0	0	0
D^{+}_{cloud} vs. D^{-}_{cloud}	0	0	0	0	0	0	0	0	0	0
D^{+}_{clear} vs. FT^{+}_{clear}	1	1	1	1	1	1	1	1	1	1
D^{-}_{clear} vs. FT^{-}_{clear}	1	1	1	1	1	1	1	1	1	1
D^{+}_{cloud} vs. FT^{+}_{cloud}	0	0	0	0	0	0	0	0	0	1
D^{-}_{cloud} vs. FT^{-}_{cloud}	0	0	0	0	0	0	0	0	0	0
D^{+}_{clear} vs. D^{+}_{cloud}	1	1	1	1	1	1	1	1	1	0
D^{-}_{clear} vs. D^{-}_{cloud}	1	1	1	1	1	1	1	1	1	0
FT^{+}_{clear} vs. FT^{-}_{clear}	0	0	0	0	0	0	0	0	0	0
FT^{+}_{cloud} vs. FT^{-}_{cloud}	1	0	0	0	0	1	1	0	1	0
FT^{+}_{clear} vs. FT^{+}_{cloud}	1	0	1	0	1	0	1	0	1	1
FT^{-}_{clear} vs. FT^{-}_{cloud}	0	0	0	0	0	0	0	0	0	0

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Table 2. Statistical data and characteristics of NPF events.

Class of NPF	Occurrence
Ia	3
Ib	14
II	17
Bump	16
Featureless	4

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Table 3. Details of NPF characteristics. Atl. is for Atlantic and EaE. for Eastern Europe.

Date	Class of event	$GR_{1.3-3}$ [nm h^{-1}]	GR_{3-7} [nm h^{-1}]	GR_{7-20} [nm h^{-1}]	J_2^+ [# cm^{-3} s^{-1}]	J_2^- [# cm^{-3} s^{-1}]	J_2 [# cm^{-3} s^{-1}]	Air mass origin
19/04/2008	Ib	6.8	9.1	6.3	0.17	0.21	2.09	–
10/07/2008	Ia	4.2	3.6	4.4	0.18	0.28	2.08	Atl.
06/08/2008	Ib	5.7	–	–	0.10	0.15	1.89	Atl.
30/08/2008	Ib	6.9	–	–	0.21	0.21	3.84	Atl.
23/09/2008	Ib	2.8	1.1	4.9	0.14	0.28	0.72	EaE.
26/09/2008	Ib	2.0	1.8	2.3	0.06	0.14	1.17	EaE.
13/11/2008	Ia	6.1	8	5.7	0.82	1.21	1.40	Atl.
14/11/2008	Ib	3.2	3.3	3.6	0.22	0.38	1.05	Atl.
26/12/2008	Ib	6.6	2.3	10.3	0.57	0.78	5.41	EaE.
20/01/2009	Ib	6.0	7.1	8.0	0.18	0.52	2.03	Atl.
19/03/2009	Ia	5.7	11.7	6.0	0.35	0.28	7.46	Atl.

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Table 4. New particle formation events according to air mass origin.

Air mass origin	NPF from air mass origin [%]	NPF frequency of the air mass [%]
Atlantic	58.3	15.4
Africa	12.5	16.2
Nordic	2.1	10
Eastern europe	20.8	33.3
Western europe	6.3	8.6

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Table 5. Comparison of ion concentration for event and non event days (Dp≥8 nm) between 15:00–18:00 LT.

Air mass origin	NPF days	Non event days
Atlantic	630	339
Africa	296	235
Western Europe	300	223
Eastern Europe	341	161
Nordic	216	594

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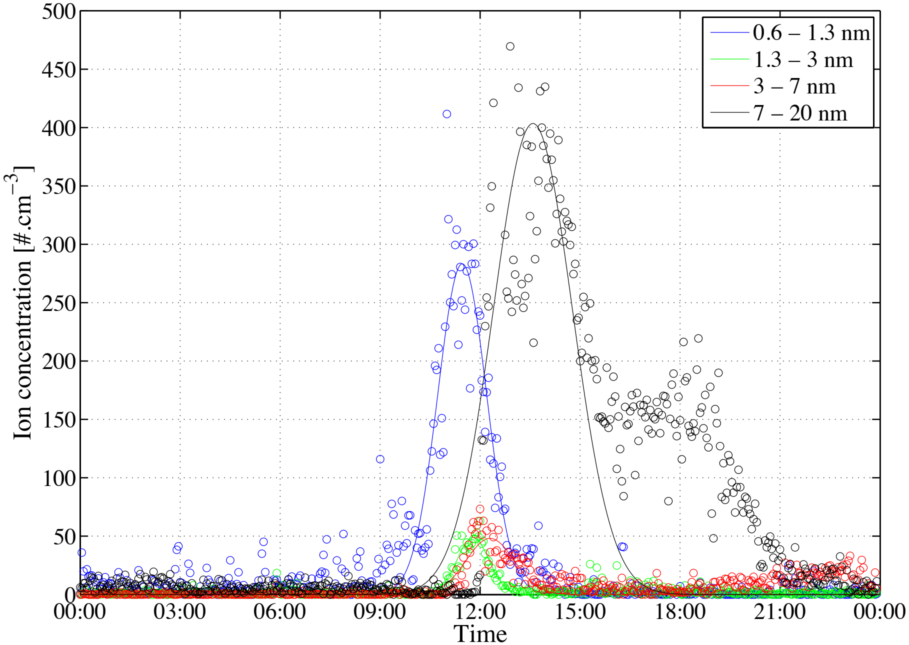

Fig. 1. Growth rates estimation fitting procedure.

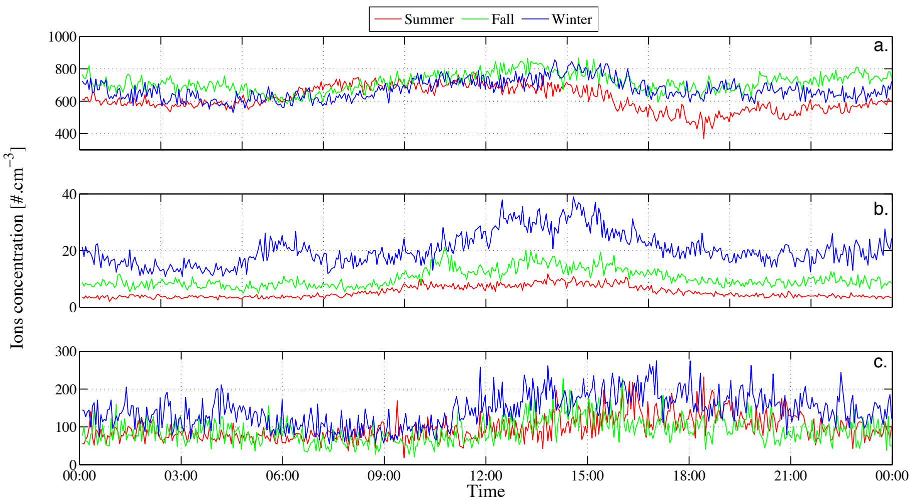

Fig. 2. Median diurnal variation of positive ion size class concentrations from July 2008 to April 2009. **(a)** Cluster ions, **(b)** Intermediate ions and **(c)** Large ions. Summer: July–September 2008, Fall: October–December 2008, Winter: January–March 2009.

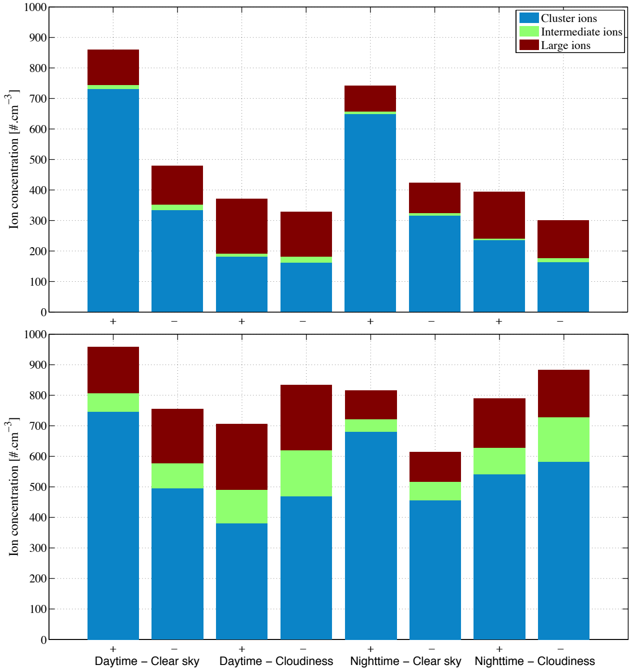

Fig. 3. Yearly median (top panel) and mean (bottom panel) concentration cluster (blue), intermediate (green) and large (red) ion concentrations from July 2008 to April 2009 under clear sky and cloudy conditions.

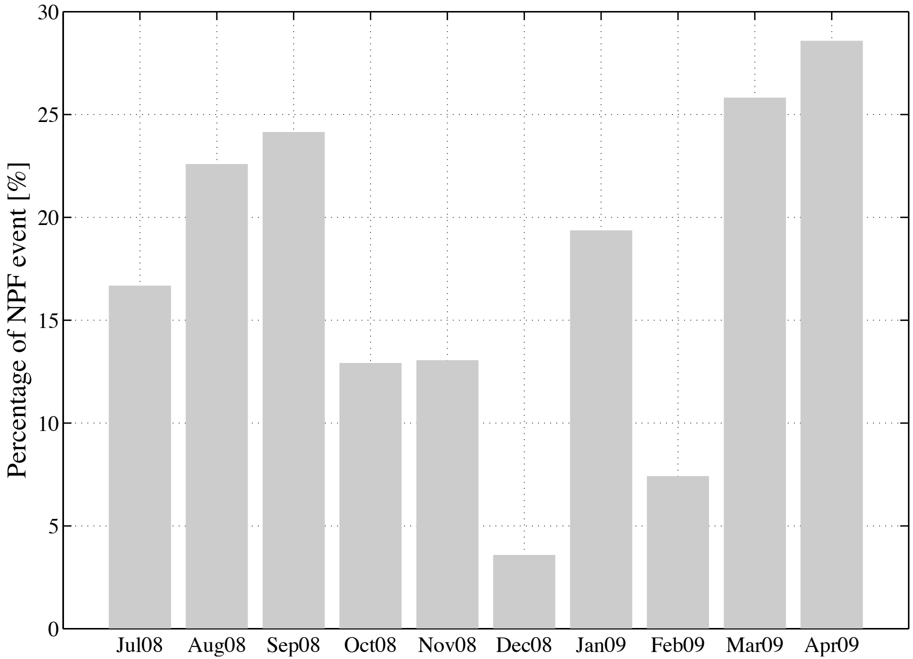

Fig. 4. Monthly frequencies of new particle formation events.

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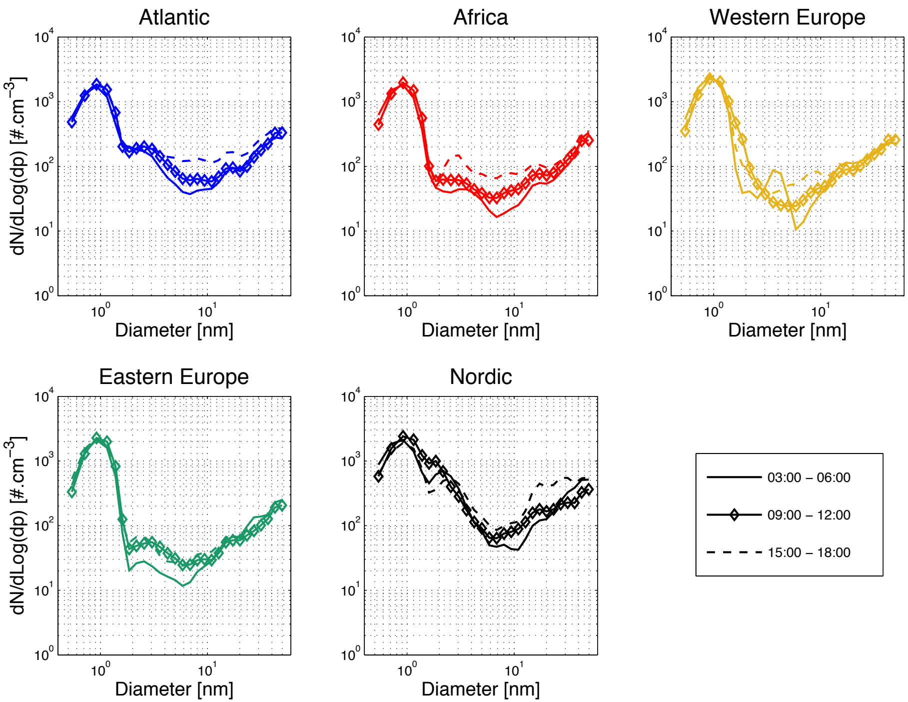

Fig. 5. Positively charged aerosol distribution of different types of air mass origin endings at Jungfraujoch at 00:00 LT for event days.

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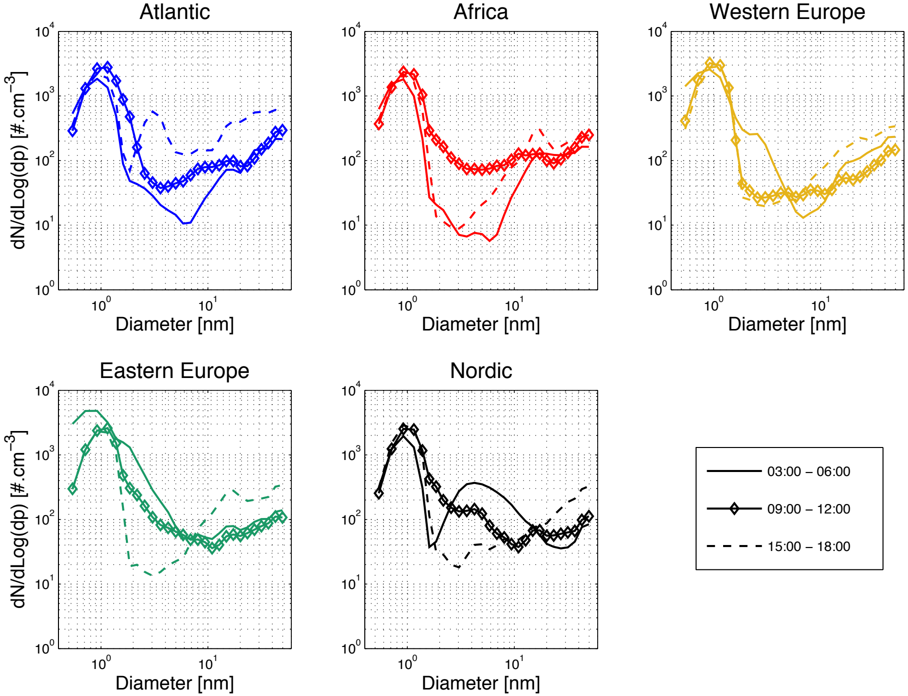

Fig. 5. Continued.

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