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Behavior of CCN to CN fraction during aging and mixing processes of atmospheric particles

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Abstract

During Spring 2003, field experiments were conducted at the summit of Puy de Dôme (1465 m a.s.l.), in Central France, mainly in the free troposphere and during cloudy conditions. During this experiment, cloud condensation nuclei (CCN) concentrations were measured with a thermal gradient CCN counter at applied supersaturations that ranged between 0.2 and 2%. Aerosol size distributions were obtained with a TSI scanning mobility particle sizer (SMPS) consisting of a DMA and a CPC, and aerosol number (CN) concentrations were derived by integrating the SMPS size spectra between 0.015 and 0.3 μm . The isentropic back trajectories were computed for the altitude of the observation site using the Hybrid Single Particle Lagrangian Integrated Trajectory (HY-SPLIT) code from NOAA. For each air mass type, the aerosol bulk chemical composition used was that previously determined from cascade impactor samples by Sellegri et al. (2003). The methodological approach of this study was, firstly, to determine the relationships between CN and CCN concentrations for various air mass types arriving on the observation site. According to their origin, air masses arriving on the observation site are classified into 4 types: continental, clean marine, modified marine and polar. The CN concentrations appear to clearly depend on the type of air mass with the highest concentrations for the continental event and the lowest ones for the marine events. Moreover, a typical range can be attributed for the CCN/CN ratio to each air mass type. Then, the objective was to observe the hygroscopic behavior of aerosol particles upon heating of the sample in a thermo-desorber. Since the heated sample can be considered as representative of the original state of the aerosol, it appears that the evolution of CN to CCN is much more affected when the air mass is modified by aging and mixing processes with the contact of various atmospheric species all along the transport pathway. This result shows indirectly but clearly the presence of an active chemical layer containing more hygroscopic properties on the aerosol particle surface that can favour the particle growth. However, the role played by organic species is not clear and more work is needed to know if organic compounds may act to increase the

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1 Introduction

It is now well recognized that clouds play an important role in the climate system. Cloud droplets are formed in the atmosphere by the presence of low supersaturation of water vapor and aerosol particles. The ability of an aerosol particle to become a cloud droplet depends on its chemical composition and physical properties and only a fraction of all particles, the cloud condensation nuclei (CCN), are able to be activated into cloud droplets under atmospheric conditions. Thus CCN have become one of the central issues in climate change. Until recently, the CCN activation spectrum was based on cloud droplet formation according to the Köhler theory for which activated droplets grow spontaneously after they have reached a critical value of the supersaturation. Under this assumption, the critical supersaturation depends upon the size and the soluble salt content of the particles. However, recent findings challenged this conceptual model. In the atmosphere, particles are most likely to be mixtures of organic and inorganic components. Even if a purely organic particle exists it is likely to collect inorganic material during cloud processing. Inversely, there is now direct (Tervahattu et al., 2002) and indirect (Husar and Shu, 1975; Gill et al., 1983; Mochida et al., 2002; Russel et al., 2002; Chuang, 2003) evidences that organic films are also probably common on inorganic atmospheric particles. Husar and Shu (1975) presented electron micrographs of Los Angeles haze particles, which suggest that the particles were coated with a non volatile film. Based on measurements of aerosol from North Atlantic arriving in Helsinki, Tervahattu et al. (2002) showed that aerosol particles were composed of a salt nucleus coated with biogenic fatty acids, generated by bubble bursting through fatty layer in the ocean. All these studies show that soluble gases, partially soluble particulate species and/or surface tension depression by organic substance can be either positive or negative artifacts for the hygroscopic growth. The main objective of the present experimental study is to determine the hygroscopic behavior of ambient

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aerosols after aging processes taking place during their atmospheric transport.

2 Experimental

For this experiment, measurements were conducted at the experimental station located at the summit of Puy de Dôme in Central France (48° N, 2° E, 1465 m a.s.l.) between 27 March and 5 April 2003. Meteorological parameters including wind speed and direction, temperature, pressure, relative humidity, radiation and gases concentration are monitored all along the year. During this season, temperatures typically vary from -10 to +10°C and westerly or northerly winds (advecting mixed continental or marine air masses) are the most frequent although southern winds can occasionally take place during short periods. The experiment was conducted mainly during cloudiness conditions, and despite its relatively low elevation, the site was located in the free troposphere according to gas and meteorological measurements.

To investigate the origin and history of the air masses for the last 3 days prior to their arrival at the observation site, the isentropic back trajectories were computed for the altitude of the observation site using the Hybrid Single Particle Lagrangian Integrated Trajectory (HY-SPLIT) code from NOAA (Draxler, 1992).

The methodological approach of the study was, firstly, to determine the relationships between aerosol number concentration (CN) and CCN concentration for various air mass types arriving on the observation site. These air-masses were classified according to their aerosol mass content and their back-trajectories. Then, the objective was to observe the hygroscopic behavior of aerosol particles upon heating in a thermo-desorber. The thermo-desorbing unit was used to volatilize particle surface up to temperatures around 220°C. For this temperature, the aim of this experiment was to observe the impact of the particle coating eventually formed during the aging process on the CCN properties.

However, in order to check that volatilization is only limited to surface components, without affecting particle mass or size, it was also necessary to analyze the aerosol

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size distribution before and after heating. Thus, the sample line was alternatively either a reference line or the heated line. The swapping of lines was operated every hour. Measurement of thermal volatilization temperatures allowed to reveal the presence of high volatility species within aerosol particles.

5 During the experiment, CCN concentrations were measured with a static thermal gradient diffusion chamber developed by University of Wyoming (model CCNC-100B) at applied supersaturations (SS) that ranged between 0.2 and 2% (Snider and Bren-
guier, 2000). CCN concentrations were measured for less than 1 min at each super-
saturation, allowing a spectrum to be obtained every 5 min. The CCN calibration was
10 previously checked by comparing the CCN concentration against measurement made
by a model 3010 TSI condensation nuclei counter, when sampling monodisperse NaCl
aerosol larger than the CCN counter's critical activation size. Aerosol size spectra mea-
surements were obtained every five minutes with a TSI scanning mobility particle sizer
(SMPS, model 3080) that included a long differential mobility analyzer (DMA, model
15 3081L) and a condensation particle counter (CPC, model 3025). The DMA operated
at ambient humidity for particles in the 0.015 to 0.7 μm mobility equivalent size range
with a sheath to aerosol flow ratio of 10. CN concentrations were derived by integrating
the DMA size spectra ($0.3 > d > 0.015 \mu\text{m}$) since aerosol particles within this size range
contribute almost 100% of the CCN. CCN and CN concentrations are then hourly av-
20 eraged. For each air mass type, the bulk chemical composition (including ionic and
elemental species, EC and OC) used for the aerosol was that previously determined
from cascade impactor samples by Sellegri et al. (2003a). Mainly, whatever the air
mass type, aerosols sampled at Puy de Dôme exhibit a high degree of external mix-
ing with carbonaceous (inorganic and organic) and ionic species associated with two
25 submicron modes centered at 0.2 and 0.5 μm in mass, respectively. Background sam-
ples are rather characterized by low nitrate and sulfate concentrations but relatively
high sodium concentrations associated with the presence of sea salt. Pollution events
which provide the most frequent aerosol type at the Puy de Dôme are rather charac-
terized by an abundance of inorganic fraction (sulfate, nitrate and ammonium) in the

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submicron range.

3 Results and discussion

3.1 Sample classification

Despite the short duration of the experiment, a large enough sample of different air masses could be observed. According to their origin, air masses arriving on the observation site were classified into 4 types: continental, marine, modified marine and polar. Figure 1 indicates the back trajectories arriving at Puy de Dôme computed for every-day at 00:00 h between 27 March and 5 April. On 27 March, a continental southern air mass arrives on the site. Then, a succession of marine air masses more or less aged and thus more or less modified are sampled from 28 March to 3 April. Their origins vary from Mediterranean Sea to Atlantic Ocean. Some traveled for less than 12 h over not very polluted regions of the continent (28–31 March; 2 April). Others were qualified of modified because aerosols are modified during transport over industrialized regions of England and Belgium before reaching Central France (1 and 3 April). Finally, a polar air mass is observed arriving on the site on 4 and 5 April.

Six-hour average concentrations of CN ($15 < d < 300$ nm), CCN (0.5% SS), and CCN/CN fractions measured at Puy de Dôme observation station between 27 March and 5 April 2003 are plotted in Fig. 2. These values were first observed by measuring concentration on the non-heated reference line. On this figure, the aerosol number concentrations appear well depending on the type of air mass with the highest concentrations for the continental event ($CN > 2000 \text{ cm}^{-3}$) and the lowest ones for the marine events ($CN < 500 \text{ cm}^{-3}$). More interesting is the CCN/CN ratio measured at 0.5% SS for which a typical range can be attributed to each air mass type: 0.3–0.4 for a continental (C) air mass, 0.4–0.7 for a clean marine (M) air mass, 0.7–0.8 for a polar (P) air mass, and 0.8–1 for a modified marine (MM) air mass. Thus, the CCN/CN computed values which are statistically different (at the 99% confidence level) for each of

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the air mass types, appear to vary significantly with the air mass origin. These results are rather consistent with previous studies for pure marine cases (Hudson et al., 1998; Hudson and Xie, 1999; Bower et al., 2000; Yum et al., 2005), in continental environment (Hämeri et al., 2001), in the Arctic environment (Yum and Hudson, 2001), or in modified marine events (Hudson and Xie, 1999; Yum et al., 2005).

In summary, the aerosol sampling is characterized by a CCN/CN ratio <0.7 when the air mass pathway over the continent is rather short (less than 12 h). For example, this is the case for the continental air mass of 27 March and the marine clean air mass of 28 March. By opposition, the aerosol sampling is rather characterized by a CCN/CN ratio >0.7 when a longer pathway over the continent allows the air mass to be modified by aging processes with the contact of various continental atmospheres. For example, this is the case for the modified marine air mass of 1 April and the polar air mass of 4 April. Thus, the process of aging and mixing among various species with different solubilities would constitute an important step in the evolution of CN to CCN. In their study on the same site, Sellegrì et al. (2003b) have suggested that the presence of inorganic species as surface coating tends to increase the hygroscopicity of the particles. On the contrary, surface coating of volatile organic species tends to decrease the hygroscopicity of the particles. Consequently, for the next part of this study, the impact of chemical surface properties of aerosols on CCN characteristics were tested on these particular young and aged air masses by using a thermo-desorbing system.

3.2 Interpretation of the thermo-desorbing experiment

By measuring volatilization temperature of natural aerosols, it is possible to know the composition of a single particle and its mixing state (internal or external, bulk or surface). Many works (O'Dowd et al., 1992; Jennings et al., 1994) have used volatility as a physical measurement to give information on the behavior of particles. For example, a volatile and hydrophobic compound can become less volatile and more hygroscopic after several transformations in the atmosphere. But the link between volatility and hygroscopicity is still difficult to establish.

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The use of an oven as thermo-desorber with temperatures around 220°C allows the volatilization of compounds such as very volatile organic carbonaceous compounds and ammonium sulfate, bisulfate and nitrate (O'Dowd et al., 1992; Hudson and Da, 1996). Other compounds such as less volatile organic species, sodium chloride, black carbon and mineral aerosols require much higher temperature to volatilize.

The heated sample can thus be considered as representative of the state of the aerosol as it was originally near its source, without coating of volatile inorganic or organic species. The corresponding reference (non-heated) sample gives the aerosol characteristics after surface transformation by processes of aging and mixing with different species during transport. No difference observed between both samples means that there is no surface coating on the particles (case of a young air mass) or the applied temperature is too low to volatilize this coating.

3.3 Behavior of CCN/CN fraction for different air masses

Figures 3–6 show the evolution of the CCN/CN fraction measured with the reference and the heated sample lines for the different air mass types arriving on the site and identified above: marine clean on 28 March (Fig. 3), continental on 27 March (Fig. 4), polar on 4 April (Fig. 5), and modified marine on 1 April (Fig. 6). For each day considered, the figure gives the temporal evolution of CN and CCN (at 0.5% SS) concentrations and CCN/CN fraction for both sampling lines, the average activated particle fraction (CCN/CN ratio) for each of the applied supersaturation (SS) that varied between 0.2 and 2%, and the average aerosol size distribution alternatively measured during the same period by the SMPS for the non-heated sample and the heated sample.

First, the comparison of both aerosol size distributions allows us to verify that the size of the particles is not altered by the thermo-desorbing experiment after they passed through the oven. It is clear from these measurements that there is no pronounced difference between these aerosol size distributions. This result well indicates that if an alteration occurs on the particle, only the particle surface seems affected. This observation confirms that the volatilization resulting from the thermo-desorbing does

not affect the bulk composition but only the surface composition of the particle.

Then, the evolution of the activated particle fraction CCN/CN as a function of supersaturation respectively observed in heated and non-heated experiments shows different features depending of the age of the air mass considered and the length of its pathway. In deed, when the air mass pathway over the continent is rather short (less than 12 h) or coming from a rather clean environment and having no time to be modified during transport, no clear difference are observed in the evolution of CN to CCN whatever the sample line. This is the case for the continental air mass of 27 March and the marine clean air mass of 28 March (Figs. 3 and 4). On the contrary, this difference is much more significant (especially for the low (<1) SS values) when a longer pathway over the continent allows the air mass to be modified by aging and mixing processes with the contact of various atmospheric species. This is the case for the modified marine air mass of 1 April and the polar air mass of 4 April. This result confirms that the process of aging and mixing among various species with different solubilities is really a key step in the evolution of CN to CCN. In the cases studied in this work, the activated particle fraction for the polar and modified marine air masses appears to be increased during atmospheric transport. This is attributed to chemical transformation of the particle surface leading to the development of surface coating of inorganic species such as nitrate and sulfate, and finally to the formation of more hygroscopic particles. However, the role played by organic species is not clear and more work is needed to know if organic compounds may act to increase the CCN concentration.

4 Conclusions

CCN to CN fractions have been measured at the Puy de Dôme observatory station for different air mass types (continental, marine clean, marine modified and polar). The aerosol number concentrations appear well depending on the type of air mass with the highest concentrations for the continental event and the lowest ones for the marine events. In the same way, the CCN/CN fraction appears to vary significantly with the air

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mass origin.

Then, we have tested the impact of surface chemical properties of aerosol particles on their CCN properties since these last ones could mainly be the result of a coating formation on the particle surface. Thus, the hygroscopic behavior of aerosol particles was observed upon heating in a thermo-desorber used to volatilize particle surface up to temperatures around 220°C. This temperature was enough high to volatilize very volatile compounds such as inorganic species or organic carbon but not too high in order to not modify the aerosol size distributions.

After analysis, it appears that when the air masses are aged enough to be modified by mixing processes with the contact of various atmospheric species, the hygroscopic properties of the aerosol particles they are transporting are also significantly modified. The observed difference is particularly significant for the low values (<1) of supersaturation which are the most frequent values available in natural conditions. These results show indirectly but clearly the presence of an active chemical layer on the aerosol particle surface that can favour the particle growth. One can easily imagine that in other circumstances other hydrophobic compounds deposited in surface could limit the capacity for a particle to be a CCN. More work is necessary to elucidate the chemical nature of the species involved in the formation of particle coating. Nevertheless, the results of this work confirm that the process of aging and mixing among various species with different solubilities is a key step in the evolution of CN to CCN.

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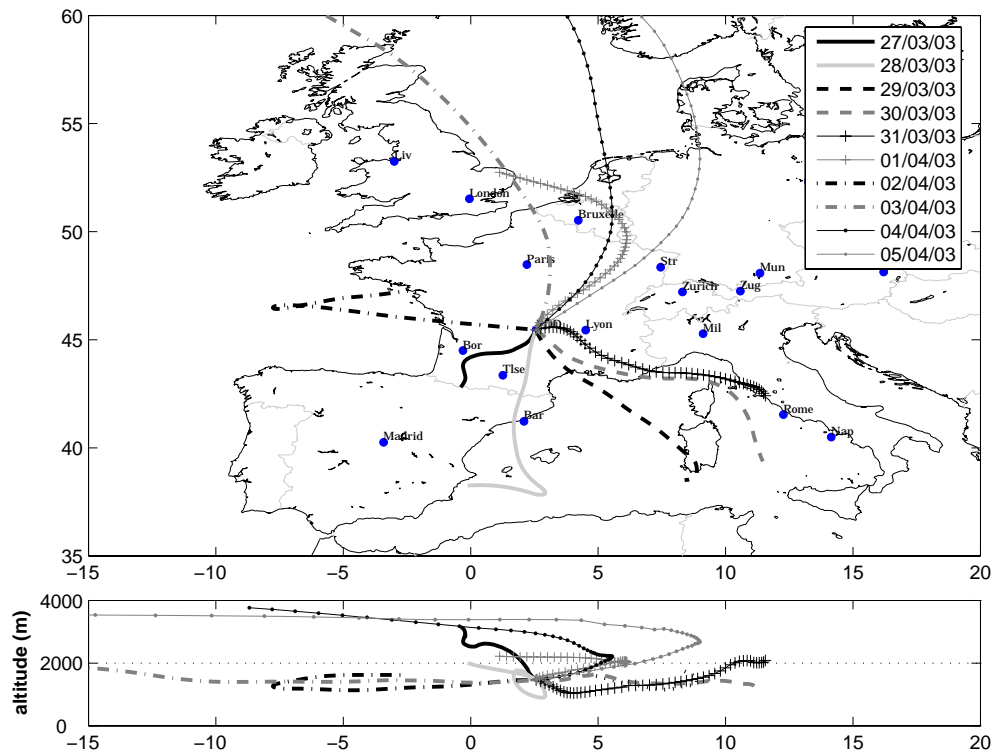


Fig. 1. Isentropic back trajectories arriving at Puy de Dôme, Central France, at 00:00 h from 27 March to 5 April.

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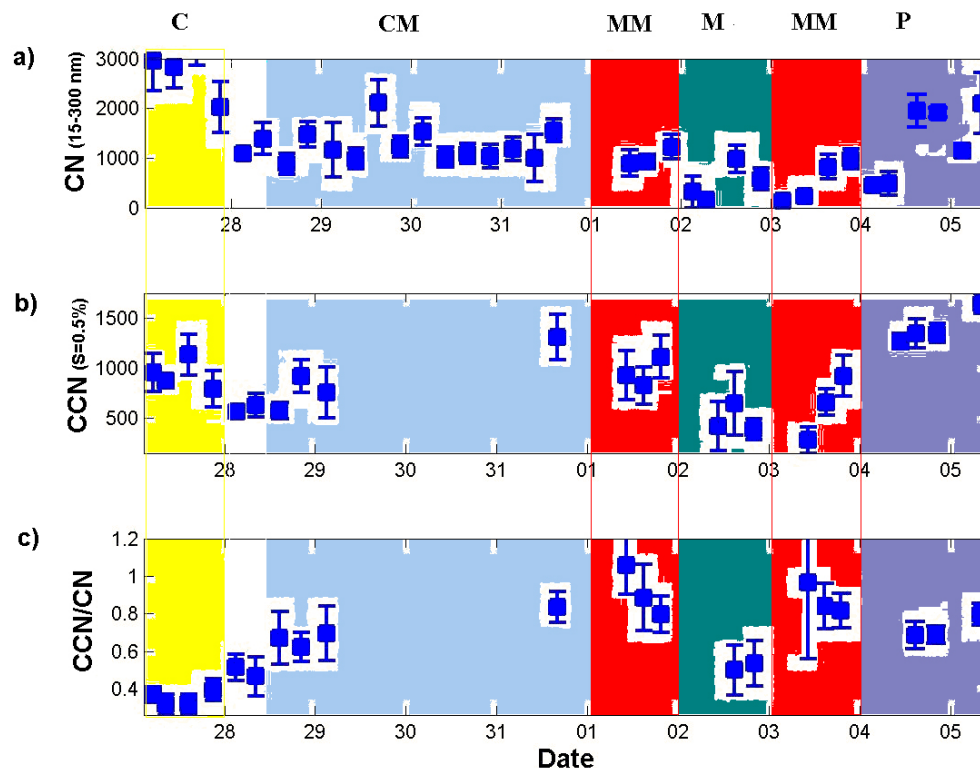


Fig. 2. Average concentrations (with sigma bars) of CN ($15 < d < 300$ nm), CCN (0.5% SS), and CCN/CN fractions at Puy de Dôme observation station between 27 March and 5 April 2003. CCN data are not available from 29 to 31 March. Sections indicate different air mass types (C: continental; CM: clean marine; MM: modified marine, M: Maritime, P: polar).

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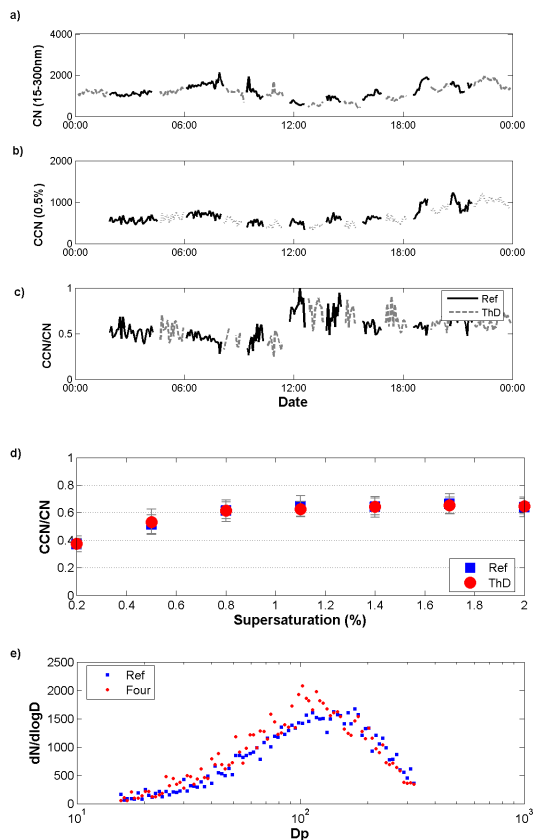


Fig. 3. Temporal evolution of CN **(a)**, CCN at 0.5% SS **(b)** and CCN/CN **(c)** for the marine clean event (28 March); **(d)** evolution of the activated particle fraction CCN/CN as a function of supersaturation respectively observed in heated (red) and reference (blue) experiments; **(e)** average aerosol size distribution alternatively measured during the same period by the SMPS for the non-heated sample (blue) and the heated sample (red).

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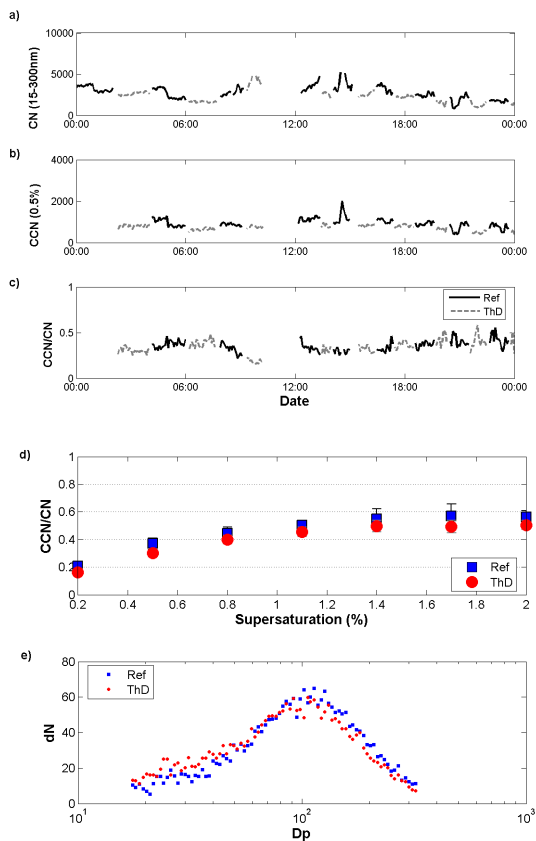


Fig. 4. Same as Fig. 3 for the continental event (27 March).

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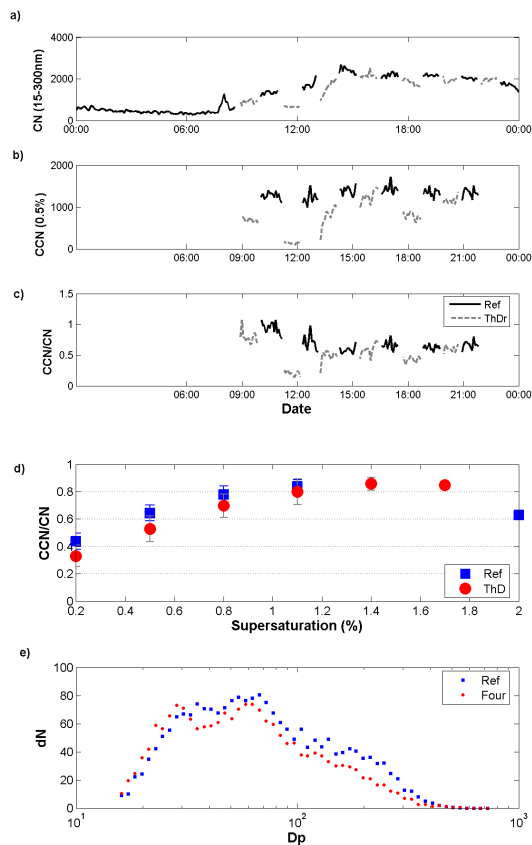


Fig. 5. Same as Fig. 3 for the polar event (4 April).

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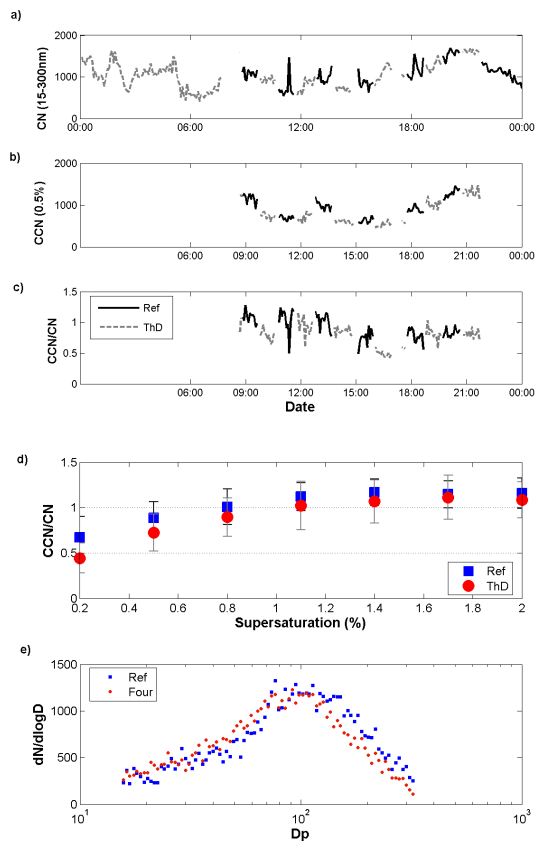


Fig. 6. Same as Fig. 3 for the marine modified event (1 April).

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