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Cn to ccn relationships and cloud microphysical properties in different air masses at a free tropospheric site

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Abstract

The fraction of aerosol particles activated to droplets (CCN) is often derived from semi-empirical relationships that commonly tend to overestimate droplet number concentration leading to major uncertainties in global climate models. One of the difficulties in relating aerosol concentration to cloud microphysics and cloud albedo lies in the necessity of working at a constant liquid water path (LWP), which is very difficult to control.

In this study we observed the relationships between aerosol number concentration (N_{CN}), cloud droplet concentration (N_d) and effective radius (R_{eff}), at the Puy de Dôme (France). A total of 20 cloud events were sampled representing a period of more than 250 h of cloud sampling. Samples are classified first according to air mass origins (Modified Marine, Continental and Polluted) and then according to their liquid water content (Thin, Medium and Thick clouds).

The CCN fraction of aerosols appears to vary significantly according to the air mass origin. It is maximum for Continental air masses and minimum for Polluted air masses. Surprisingly, the CCN fraction of Modified Marine air masses fraction is lower than the continental air mass and from expected from previous studies. The limited number of activated particles in Modified Marine air masses is most likely the result of the presence of hydrophobic organic compounds. The limited activation effect leads to a 0.5 to 1 μm increase in R_{eff} with respect to an ideal Marine case. This is significant and implies that the dR_{eff}/dN_{CN} of low-continental clouds is higher than expected.

1. Introduction

Quantification of the indirect radiative forcing of aerosols on climate through a change in cloud droplet number size distributions (better known as the Twomey effect; Twomey, 1991) is a major source of uncertainty in Global Climate Models (GCM) (Yao and Del Genio, 1999). The present uncertainty is in the range of 0 to $-1.5\,W\,m^{-2}$ and arises

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mostly from the poor representation of cloud scale processes in GCMs (Jones et al., 1994). In particular, the fraction of aerosol particles activated to droplets (CCN) is often derived from semi-empirical relationships between aerosol (or sulphate) mass or number and cloud droplet concentration (Boucher and Lohmann, 1995) or by prognostic relationships between CCN and supersaturation of water vapour (Ji and Shaw, 1998). Both approaches commonly tend to overestimate droplet number concentration (Snider and Brenguier, 2000) and do not consider possible artefacts such as the presence of soluble gases or organic material on aerosol particles (Kulmala et al., 1993; Facchini et al., 1999; Nenes et al., 2002).

The effect of increased aerosol concentration on cloud microphysics has been documented in a number of studies (Twohy et al., 1995; Bréon et al., 2002; Leaitch et al., 1992). However, one of the difficulties of relating observed changes in aerosol properties to changes in cloud microphysics and albedo is the implicit assumption that the liquid water path (cloud liquid water content) is constant. This assumption is very difficult to control and is seldom verified due to feedback mechanisms affecting cloud height (Ackerman et al., 2000; Han et al., 1998). As a consequence, the ideal methodology to relate change in cloud microphysics to change in aerosol concentration implies performing observations either at constant LWP (Feingold et al., 2003) or at constant height within clouds.

2. Measurements

In this study we observed the relationships between aerosol number concentration (N_{CN}) and cloud droplet effective radius (R_{eff}), commonly used in the parameterization of cloud optical characteristics, at the Puy de Dôme cloud station (Central France, 48° N, 2° E, 1465 m a.s.l.) during the winter and spring 2000 and 2001. A total of 20 cloud events were analyzed representing a period of more than 250 h of cloud sampling.

Measurements of total particle concentration (N_{CN}) were performed using a TSI 3010

particle counter (CPC) sampling downstream a whole air inlet. The whole air inlet has been specifically designed to allow, under cloudy conditions, the sampling of both particles and evaporated cloud droplets. The droplet number (N_d) – corresponding to the CCN number concentration N_{CCN} for LWC<0,5 g m^{-3} (Flossmann et al., 1985) – is derived from a PMS Forward Scattering Spectrometer Probe (FSSP) located inside a wind tunnel operating isocinetic conditions at 40 m s^{-1}. The cloud droplet effective radius (R_{eff}), commonly used in the parameterization of cloud optical properties (Martin et al., 1994), is computed directly from the size distribution of droplets. In addition, a particle volume monitor (PVM Gerber) provides the cloud LWC. An additional CPC TSI-3010, connected to an interstitial inlet (50% cut-off=5 μm), measures unactivated interstitial aerosol concentration (N_{INT}). All measurements are performed using a time resolution of 1 Hz. Whole air and RJI inlets as well as the wind tunnel have been described previously (Schwarzenboeck et al., 2000; Sellegri et al., 2003a) and provide an adequate sampling of clouds and aerosols. A complete description of the instrumental deployment at the site is provided by Sellegri et al. (2003b). The fraction of aerosols activated to CCNs in clouds (F_{Np}) is derived using N_d and N_{INT} :

$$F_{Np} = \frac{N_d}{N_d + N_{INT} - \Delta N} = \frac{N_d}{N_{CN}}$$

where ΔN accounts for the fraction of droplets smaller than 5 μm measured by FSSP and sampled in the RJI.

The chemical properties of aerosol particles are measured with two low pressure impactors connected to the whole air inlet. The first impactor is used for the determination of the inorganic fraction of the aerosols, the second for gravimetry, organic (OC) and elemental (EC) carbon. In order to accumulate enough material for chemical analysis, an impactor run covers the entire duration of a cloud event. Description of the analytical procedures and results from the chemical analyses are available in Sellegri et al. (2003a, b).

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3. Sample classification

Sellegri et al. (2003a) classified the air mass types at Puy de Dôme based on their aerosol mass content and analysis, and computation of back-trajectories. Based on the same cloud events than the ones studied in Sellegri et al. (2003a), and hence the same air mass classification, we observed that aerosol number concentration followed this classification with Modified Marine (MM) type showing N_{CN}<700 cm^{-3}, Continental (CL) showing 700 cm^{-3}<N_{CN}<2200 cm^{-3} and Polluted (PL) showing N_{CN}>2200 cm^{-3}. Modified Marine air masses are advected to Puy de Dôme during westerly conditions (distance to the Atlantic Ocean is 500 km) while continental air masses originate from Northern Europe. In contrast, PL air masses correspond to specific conditions where CL air masses are mixed with urban plumes from the large industrial areas of Northern France and Benelux.

The bulk chemical composition of the particles for each air mass type, derived from the sum of cascade impactor stages (Sellegri et al., 2003a), is presented in Fig. 1. As noticed by Sellegri et al. (2003a), the chemical composition differs according to the origin of the air masses. Modified Marine air masses are characterized by a high sea salt content, but also by a high fraction of organic material (56%), as compared to the other two air mass types (34% and 24% for CL and PL, respectively). Instead, polluted air masses are characterized by a higher fraction of inorganic material, and in particular NO_3^- (22% for PL as respect to 19% for CL and 7% for MM).

The averaged microphysical properties of clouds (LWC and R_{eff}) along with the averaged N_{CN} and F_{Np} for each of the air mass type are shown in Table 1. The threshold for LWC is set at 0.05 g m^{-3}. This classification shows that FNp changes with the origin of the air mass, as expected from previous studies (Leaitch et al., 1992; Gillani et al., 1995), with maximum scavenged fraction for CL air masses and minimum for PL air masses. As expected, F_{Np} is the lowest for PL air masses (CCN accounts for 33±11% of all particles >10 nm). Surprisingly, F_{Np} is larger during continental conditions (48±16%) than during MM conditions (42±14%). These values are in the range

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of those measured in previous studies (Gillani et al., 1995; Martinsson et al., 2000; Chuang et al., 2000) although direct comparison is rather difficult because very few studies considered the large variety of clouds measured in this study.

However, changes in F_{Np} are not only a result of particle properties because F_{Np} varies according to the supersaturation in cloud and therefore to the height from the cloud base. This appears in Fig. 2 where the relationship between F_{Np} and LWC is shown for the three air mass types. For clarity, an averaged F_{Np} is calculated for each 0.01 g m^{-3} interval of the LWC. This graph shows that F_{Np} is higher for CL air masses than for both PL and MM air masses regardless of the LWC. At low LWC, F_{Np} is rapidly changing with increasing LWC reaching a maximum for LWC ranging from 0.2 to 0.5 g m^{-3}. A second maximum is reached for high LWC values.

In a cloud, the adiabatic lifting of droplets leads to a linear relationship between LWC and h (Pruppacher and Klett, 1997, and reference therein). To account for the changes in F_{Np} with LWP, we assumed no entrainment of dry air into the cloud. This hypothesis is difficult to verify without continuous monitoring of the cloud base height. The process of entrainment, however, leads to a modified droplet size distribution as respect to that produced by adiabatic lifting. The spectral shape parameter k can be used to assess the variability of the droplet size distribution. This parameter is defined as (Martin et al., 1994):

$$R_{\text{eff}}^3 = kR_v^3 \tag{1}$$

where R_v is the mean volume radius of the distribution. The spectral shape parameter is equal to unity for a monodisperse distribution. The following expression of R_{eff} can be derived from Eq. (1) (Martin et al., 1994; McFarquhar and Heymsfield, 2001):

$$R_{\text{eff}} = \left(\frac{3\text{LWC}}{4\pi\rho_w k N_d}\right)^{1/3} \tag{2}$$

If k is constant, then R_{eff}^3 is linearly correlated to LWC/N_d. This is confirmed in Fig. 3 where the variations of R_{eff}^3 and LWC/N_d are compared for the 3 air mass categories

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and fitted with a linear regression. The coefficient of determination (R^2) is higher than 0.98 for all three categories. On average, the adiabatic assumption is therefore applicable at Puy de Dôme, despite turbulences and entrainment produced by the orography. The effect of entrainment is mostly seen near cloud base (i.e. at low LWC), when both F_{Np} and k show a high variability.

Under the adiabatic assumption, LWC is indicative of height from the cloud base, and we performed a second sample classification on the basis of the LWC. Three LWC classes are considered: thin clouds – sampled near the cloud base – for LWC<0.2 g m^{-3}, medium clouds with LWC values in the range of 0.2<LWC<0.4 g m^{-3}, and thick clouds where LWC>0.4 g m^{-3}. The two classifications with CN and LWC lead to 9 different categories. Thick clouds forming in MM air masses contain the lowest number of samples, accounting for less than 1 h of sampling. All other categories are represented by a sampling period of more than 2 h which reaches 32 h for the most common category (medium CL).

4. Analysis

A statistical analysis of the CCN fractions for all 9 categories is presented in Fig. 4 showing large variations among categories. The computed F_{Np} values are statistically different (at the 99% confidence level) for each one of the categories. The average aerosol partitioning F_{NP} is calculated for each category showing that the CCN fraction varies significantly with respect to both LWC and N_{CN}. As expected, the classification confirms that the CCN fraction is lower for PL air masses than for CL and MM air masses regardless of LWC. This is primarily resulting from the presence of a higher fraction of Aitken and ultrafine particles. Previous studies at Puy de Dôme showed that the activation diameter ranges from 50 to 100 nm (Gérémy et al., 2000). A large fraction of the PL particles are smaller than the activation diameter leading to a limited F_{Np}.

Unexpectedly, the scavenged fraction is smaller for MM than for CL. This is not seen

in any previous studies (Snider and Brenguier, 2000; Gillani et al., 1995; Martinsson et al., 2000). Several hypotheses can be proposed to explain this finding: 1 – a higher activation diameter in MM air masses compared to CL air masses due to higher number of smaller particles in MM air masses compared to CL air masses, 2 – presence of externally mixed particles, and 3 – limitation of the droplet activation due to the chemical composition of the particles.

We can exclude the first hypothesis: the particle size distribution as measured by cascade impactors shows that the fraction of accumulation-mode particles for CL and PL air masses is lower than for MM air masses, while the fraction of Aitken particles is higher for CL and PL (Sellegri et al., 2003a). This is confirmed by more recent monitoring of the number size distribution of aerosols performed with Differential Mobility analysers. This information coupled with the fact the particle number is lower for MM than for CL and PL, leads to the conclusion that the lower scavenged fraction during MM is not resulting from the aerosol size.

The second hypothesis (externally mixed particles) is more probable. This implies that hygroscopic marine particles are externally mixed with more hydrophobic particles. The last hypothesis is similar to the previous one except for the fact that particles are internally mixed: in both cases, hydrophobic matter would be mixed with the pure marine particles. Given the fact that the mass fraction of organic matter increases with remoteness of the air mass, it is likely that the hydrophobic particles (or their hydrophobic fraction) are (is) composed of organic material. This is consistent with Sellegri et al. (2003b) showing that, at Puy de Dôme, the organic particles are poorly scavenged in clouds.

However, the origin of the organic material during MM cases remains unclear. Recent measurements performed on the marine atmosphere revealed the presence of a large fraction of organic material (O'Dowd et al., 2004) that could explain the limited particle activation. Another hypothesis is that organic matter and/or particles are enhanced during the inland advection of marine air masses. This is consistent with the fact that the area between Puy de Dôme and the Atlantic Ocean is mostly covered by

forests and crops.

Biogenic material can either form primary particles or arise from deposition of low-volatility organic compounds onto existing aerosol surfaces. For instance, it is known that film-forming organic compounds can alter the activation process either reducing or increasing the activation diameter (Feingold and Chuang, 2003). Additional measurements, not presented in this study, show that removal of the film-forming compounds by thermo-desorption at moderate temperatures (50–70°C) can either decrease or enhance the CCN fraction. However, additional studies are necessary to show the presence of an organic film onto marine particles, and identify the film-forming organic species, their role in the process of particle activation and understand the mechanisms involved in the evolution of Marine aerosol inland.

5. Impact on cloud microphysics

The limitation of the number of particle activated during the advection of Marine air masses has direct consequences for microphysics of clouds. We can make a very crude estimate of the change in R_{eff} resulting from the modification of the original marine air mass. Investigations of the CCN fraction in Marine aerosols were performed during ACE-2 (Snider and Brenguier, 2000; Bower et al., 2000; Martinsson et al., 2000). Under similar conditions as for Puy de Dôme (droplet formation forced by orography and similar supersaturation close to 1% as deduced from modelling studies at the 2 sites – Flynn et al., 2000; Gérémy et al., 2000), the CCN fraction during a cloud event corresponded to 60% of N_{CN}. The difference in F_{Np} between Marine (ACE-2) and Modified Marine (PDD) air masses is significant, according to a Student T-test.

Such a change leads to significantly less droplets at PDD during MM air masses than expected for purely Marine air masses, and therefore a higher cloud R_{eff}. The variations of R_{eff} as a function of N_{CN} concentration at constant LWC have been computed for the three LWC classes (Fig. 5). The relationship between R_{eff} and N_d (Martin et al., 1994) is also shown on the graph for different values of LWC. This graph provides a direct

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estimate of the sensitivity of continental clouds microphysical properties to changes in aerosol number concentration. As expected, R_{eff} decreases with increasing N_{CN} at a rate that is close to that derived using a parameterisation proposed by Raga and Jones (1993) and corresponds, on average, to 2 μm between MM and PL cases. Note that the calculated dR_{eff}/dN_{CN} would have appeared much lower without the sorting of samples according to their LWC. The sensitivity of R_{eff} to change in N_{CN} (dR_{eff}/dN_{CN}) appears to be higher at low N_{CN} values. This is a direct consequence of the limited fraction of particles activated in MM air masses, leading to higher R_{eff} values.

In order to quantify the effect of activation limitation on cloud microphysics, we compared R_{eff} measured at Puy de Dôme to an estimated R_{eff} for which about 60% of CNs are activated to droplets (F_{Np}=0.6 at S=1%) as measured for a purely marine case during the ACE-2 HILLCLOUD campaign (Bower et al., 2000). The measured and calculated R_{eff} are compared at constant LWC, i.e. on the same iso-LWC lines in Fig. 5. Under this assumption, the change in R_{eff} (ΔR_{eff} [R_{eff} measured – R_{eff} calculated]) ranges from 0.5 to 1 μm (Fig. 5).

6. Conclusions

CCN fractions have been measured at the puy de Dome cloud station for a high number of in-cloud samples, classified into three different air mass types (Marine, Continental and Polluted. Marine aerosols modified by transport over the continents appear significantly less hygroscopic than in the marine atmosphere with a CCN fraction of 42%±14% (at supersaturation close to 1%), to be compared with 60% for a pure Marine case, and surprisingly less than in the continental air masses (48±16%). The limitation of the number of activated particles, and its subsequent impact on cloud microphysics, is significant. The processes leading to the hygroscopic modification of the original marine aerosol are still uncertain but are likely linked to the presence of a significant fraction of organic material. One hypothesis is that the condensation of semi-volatile organic species, possibly emitted by biogenic activity, onto the aerosol

surface limits the activation of aerosol particles in clouds. Work is in progress to confirm, or not, this hypothesis.

Compared to the theoretical Marine case, the limitation of the number of particle activated in an aged marine air mass produces an enhancement of R_{eff} in clouds from 0.5 to 1 μm involving, at the same time, a decrease in the cloud coverage compared to a pure marine ari mass. It is difficult to assess the regional extent of this effect until the elucidation of the nature of the chemical species involved in hygroscopic growth limitation.

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Table 1. Averaged values of cloud liquid water content, particle concentration (N_{CN}), scavenged aerosol fraction (F_{Np}) and Effective radius (Re) for the 3 air mass categories identified at Puy de Dôme. The uncertainties correspond to one standard deviation of the mean.

Air mass category	LWC ($g\,m^{-3}$)	N_{CN} (cm^{-3})	F_{Np}	R_{eff} (μm)
Modified Marine	0.14±0.07	525±103	0.42±0.14	6.15±1.06
Continental	0.16±0.07	1264±374	0.48±0.16	4.61±0.80
Polluted	0.26±0.15	3094±916	0.33±0.11	4.41±1.01

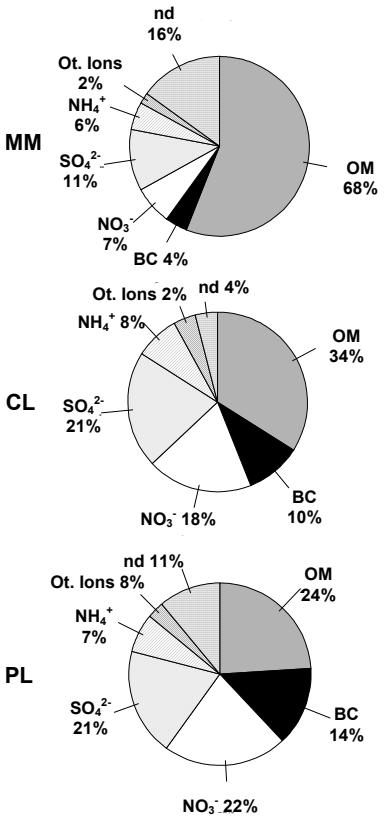

Fig. 1. Aerosol chemical composition for the 3 air masses MM, CL and PL. The composition is deduced from the work of Sellegri et al. (2003a). In this figure, OM is corrected to account for the total mass of organic matter. Other ions – Ot. Ions – is the total fraction of ions measured by ion chromatography and non determined – nd – is the difference between gravimetry and chemical analyses.

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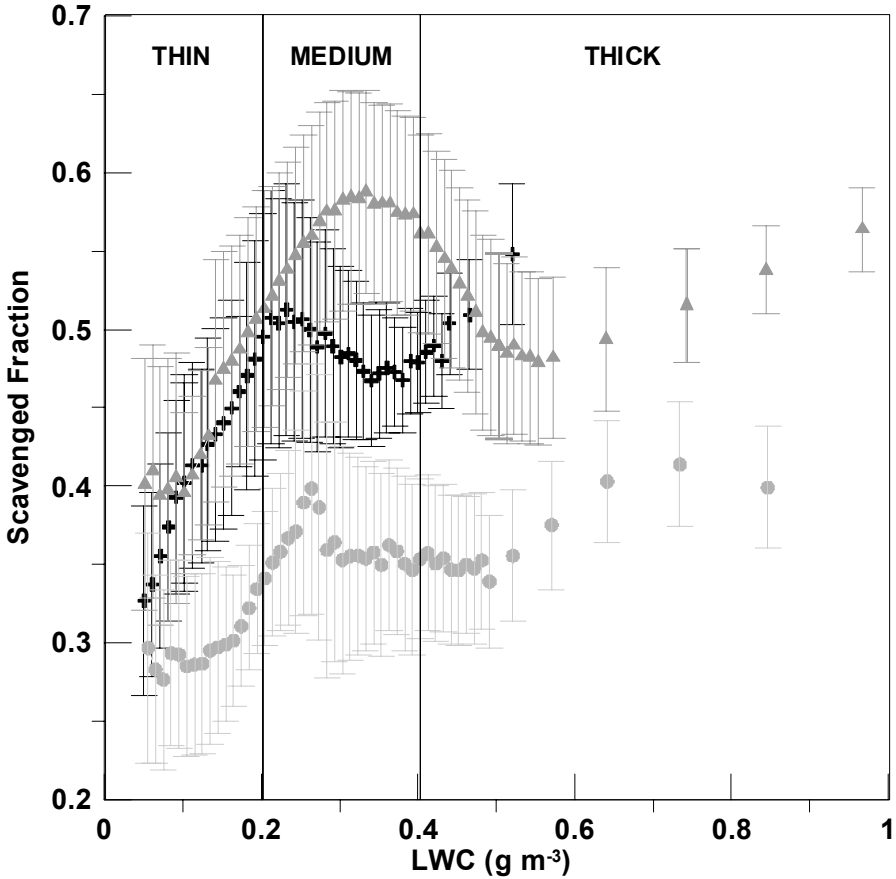

Fig. 2. Variations of the scavenged fraction of N_{CN} as a function of cloud LWC for the 3 air masses MM (cross), CL (triangle) and PL (circle) Each point represents the average N_{CN} value in a 0.01 g m^{-3} interval of the LWC. The uncertainty is the standard deviation of the mean.

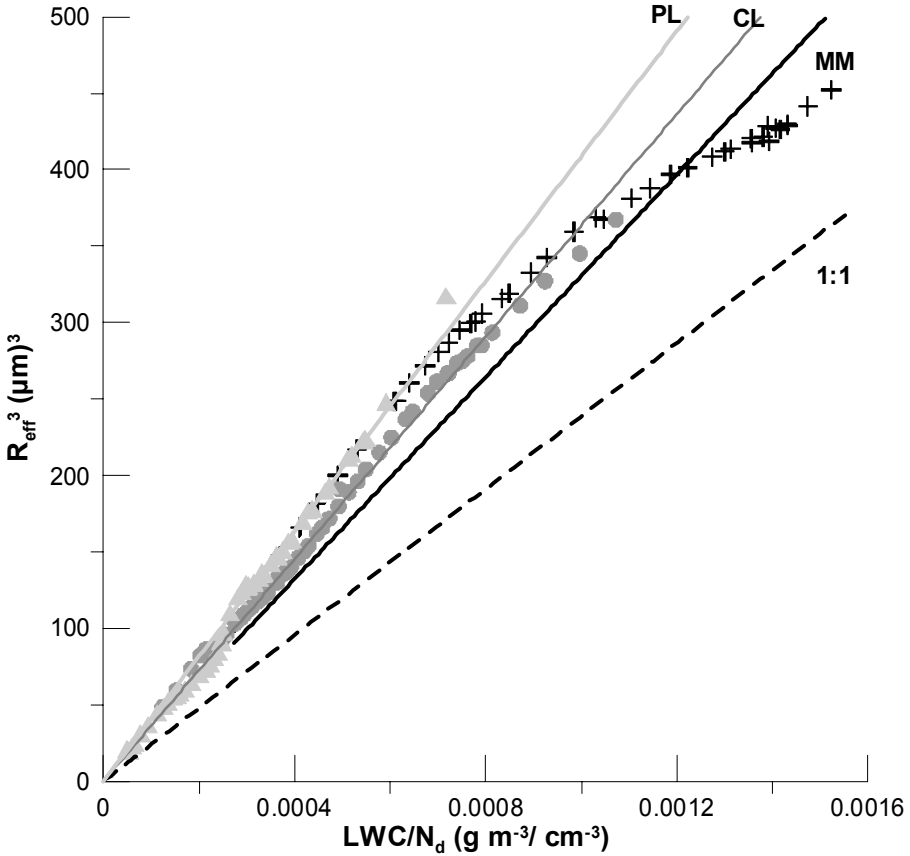

Fig. 3. Variations of R_{eff}^3 as a function of LWC/N_d for each of the three air mass type MM (cross), CL (circle) and PL (triangle). Each population is fitted with a linear regression. Each point represents the average N_{CN} value in a 0.01 g m^{-3} interval of the LWC. The solid black line shows 1:1 correspondence for monodisperse size distribution where R_{eff}=R_v.

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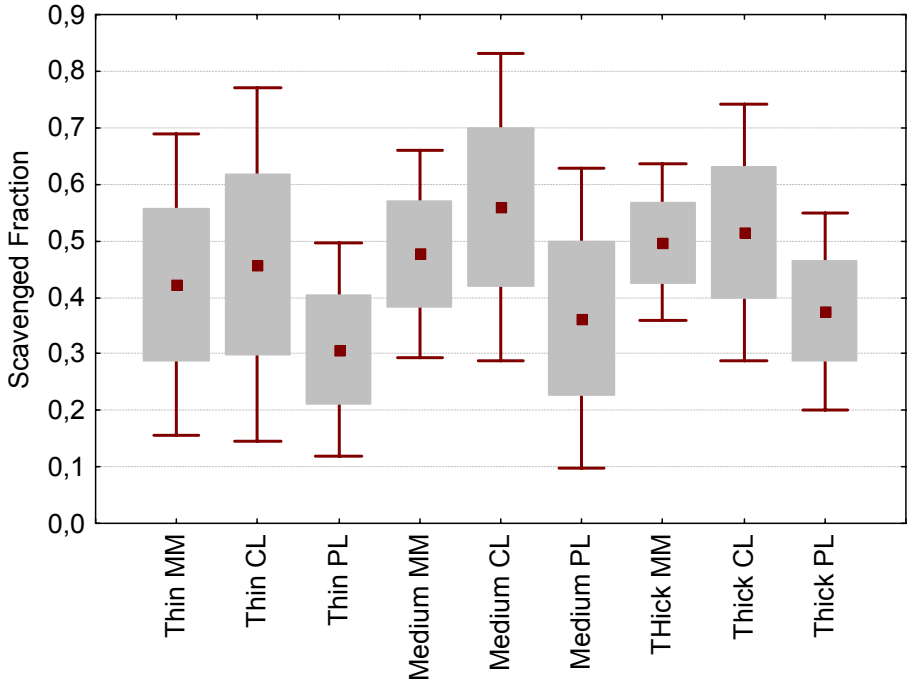

Fig. 4. Statistical analysis of the CCN fraction for the 9 classes of samples showing mean of the population, 1 standard deviation (box) and 1.96 standard deviations (whiskers), respectively. Modified Marine (1), Continental (2) and Polluted (3) and thin (A), medium-thin (B), medium-thick (C) and thick (D) clouds.

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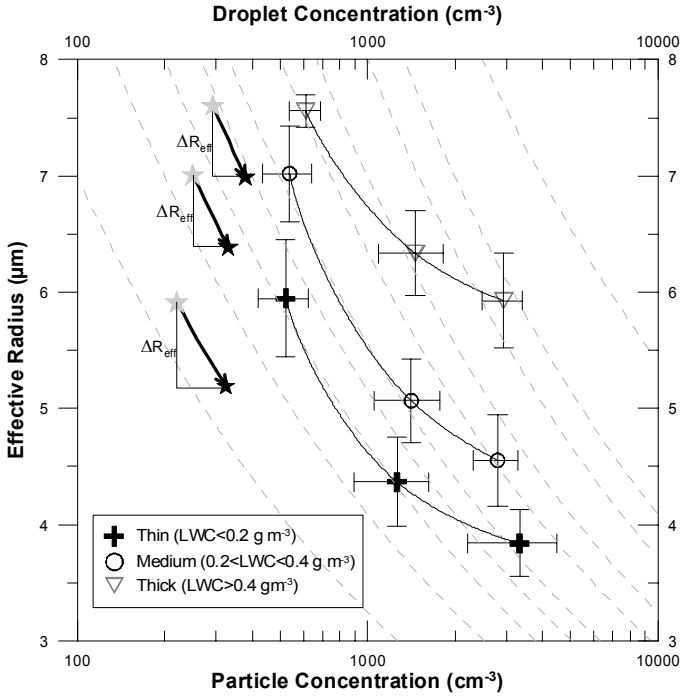

Fig. 5. Variations of R_{eff} as a function of N_{CN} concentration for the 3 classes of air mass types – Modified Marine (N_{CN}<700), Continental (700<N_{CN}<2200) and Polluted (N_{CN}>2200) – and at the 3 different range of LWC corresponding to thin (cross), medium-thin (circle), and thick (triangle) clouds. Iso-LWCs are fitted with a spline function. Grey lines are the relationship between R_{eff} and N_d at constant LWC for LWC varying from 0.05 mg m^{-3} to 2 mg m^{-3}. Grey Stars on arrow start correspond to the number of droplets measured in the marine cases while Black Stars on arrow end correspond to number of droplets calculated in the purely marine cases assuming F_{Np}=0.6. The DR_{eff} is the difference of R_{eff} between measured R_{eff} and a theoretical purely marine case (see text for details).

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