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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cn to ccn relationships and cloud microphysical properties in different air masses at a free tropospheric site" *by* R. Dupuy et al.

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Response to the Editor and Reviewers comments

Dear Editor,

We appreciate the constructive reviews from all three Reviewers that have leaded us to significantly enhance the quality of the paper, and we wish to summarise our general responses prior to providing a detailed response to all reviewers comments. In fact, important corrections have been made to the paper not only to the text but also to the structure of the data set. The original classification of samples was only based on the CN number concentration, while we now sub-divide each sample class according to air mass origins according to their back trajectories, leading to 5 aerosol classes instead of 3. As a result, conclusions regarding the relative differences in aerosol scavenging fractions according to the air mass types and corresponding consequences on cloud properties do not always hold. This version of the paper simply aims to describe a statistical analysis of scavenging properties of aerosols within different air masses at a free tropospheric site, which can occasionally be influenced by the boundary layer (PL case). A figure explaining the experimental deployment during the winter 2000 and 2001 campaigns at the puy de Dôme has been added (Figure 1), as well as a Figure explaining the statistical analysis performed prior to sample classification (Figure 2). The original version of the paper showed the bulk composition of the aerosol, for simplicity, and because it is now well known that sea salt and mineral dust are mainly found in the supermicron fraction, while carbonaceous aerosols are found in the submicron fraction. However, it is correct that the submicron part of the aerosols is the most important regarding their activation to cloud droplets. According to the referees demand, Figure 3 has been modified to show the chemical composition of the submicron part of aerosol particles for each air mass class.

Best regards,

Paolo Laj, Karine Sellegri and Regis Dupuy

Referee #1

Primary concerns :

The CCN/CN ratio is indeed governed by particle size and composition, mainly by its submicron fraction, and by cloud supersaturation. The question of submicron composition has been corrected. As for the supersaturation information, no such measurement is so far possible. A few CCN chamber measurements (Wyoming CCN chamber) were performed downstream of the WAI and were in agreement with the FSSP/CN measurements within a few percents, if the supersaturation chosen in the chamber was 1.6%. Unfortunately, the CCN chamber was operating only for a few days: what we report in this work the actual CCN/CN ratio in natural clouds and in real conditions. The liquid water content was however measured, and it is the only information available on the supersaturation that we possess. The supersaturation itself could not be known by another mean than a modelling study over the area, with the specific topography. This is not conceivable in the context of this study, where statistics are performed on 5 min averages calculated over more than 250 hours of measurements. We aimed here at comparing mean behaviours of CCN/CN ratio of different air masses, for a given LWC, hence at similar supersaturations.

1. FSSP gives accurate information on the size distribution of droplets, but it has been calibrated with the PVM prove, more adequate for the total LWC. This is now mentioned in the text.

2. RJI inlet (round jet impactor) is the interstitial inlet. It is now referred as such in the text.

3. Size range of the impactors mentioned

4. CL changed into -C

5. We have calculated the submicron part of the chemical composition of aerosol particles for different air mass classes; which we agree, is more relevant for this work.

6.”as expected” and “surprisingly” explained

7. “large variety” means that clouds were sampled in different air masses, and with different liquid water contents. It is now explained in the text.

8. The linearity of LWC/H is not relevant in this context.

9. The PVM probe is indeed used to infer the LWC measurements, hence independent of N measured by the FSSP. This is now mentioned in the text.

10. Martin et al. (1994) stipulates, based on aircraft measurements of microphysical parameters in clouds, that “where entrainment effects are small the effective radius is found to be a linear function of the volume-averaged radius in a given cloud”.

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11. Thin, medium, and thick clouds are now replaced by low, medium and high clouds.

12. For the same amount of water, hence the same LWC, a higher number of particles, found in polluted air masses, will be less activated, in proportion to the total number of particles. This is now better addressed in the text.

13. The topography around the Puy de Dôme is not perfectly symmetric, as it is part of a North-South mountain chain. However, we have been careful to compare air masses with eachother, at a given LWC, hence at a given sursaturation.

14. SMPS size distributions have been monitored during the winter 2006. However, for the statistical analysis on the total CN number concentrations since 2001, we seem to observe a gradual increase of CN concentrations and would rather not use 2006 CN concentrations to argue on results on aerosol properties obtained in 2000-2001. Aerosol size distributions were measured by mass during the winter 2000 and 2001 with cascade impactors; we now refer to these measurements for evaluating which air mass contains smaller or larger aerosols.

15. 16. The new version of the paper do not include these hypotheses.

17. The significance level for the statistical test, which is a simple t-test, is given to be 99% confidence. The nature of the test is now given in the text.

18.19.20.21.22. The original study on the impact of a limited activation of modified marine aerosols compared to pure marine aerosols have been dropped. We agree with the reviewer that this part did not contain sufficient information in order to be valid.

Referee #2

Data concerning the chemical composition of aerosol particles during the winters 2000-2001 for different air masses, and published in Sellegri et al. (2003a) are now better described regarding: 1) the instrumental set -up deployed to obtain it 2) the description of the main results

Moreover, additional calculations have been performed to show the submicron part of the chemical composition of the aerosol particles, more relevant for this work.

The size cut of the CPC TSI 3010 has been mentioned. The CPC used during ACE-II was also a CPC TSI 3010.

We have now précised that the terms “activation/CCN” were used as an approximation of what was actually measured: activation and in-cloud scavenging.

The classification into air mass types according to CN number concentration is now explained in details and illustrated by Figure 2.

RJI inlet is the interstitial inlet (Dp $<5\mu$m), it is now referred as such in the paper.

The information on the number of samples per air mass class in now added Table 4.

Referee #3

1. The paper was given for English spelling and grammar corrections to a professional company. 2. Information on experimental details relative to the field campaigns 2000-2001 are now better described and summarized in Figure 1. Key results on aerosol characteristics are also given. 3. 4. Data analysis is clarified with the addition of Figure 2 and Table 1. 5. Bulk chemical composition is now replaced by sub-micron chemical composition. 6. The original Figure 2 was simply illustrating the fact that the aerosol scavenged fraction was different from one air mass to the other, whatever the LWC. This information is in fact contained in Table 3. Figure 2 has been dropped. 7. Figure 3 is not aimed at showing differences between air mass in term of k, it aims at showing that for a given air mass, the slope k is approximately constant, which is interpreted as an indication of little entrainment in the clouds that were sampled at the puy de Dôme. 9. Section on the impact on cloud microphysics is now not anymore justified, considering the new results obtained with a more precise air mass classification.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 879, 2006.