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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud chemistry at the Puy de Dôme: variability and relationships with environmental factors" *by* A. Marinoni et al.

A. Marinoni et al.

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General Comments:

1. The consistency of the thresholds for the different air mass categories has been rechecked throughout the paper. The thresholds for classification of different kind of samples are 18 and 50 mg l-1 when we talk about solute content that take into account also the organic fraction (DOC), while 15 and 40 mg l-1 are the thresholds for the TIC (Total Ionic Content, that comprehends inorganic and organic ions). For easier understanding we actually use in all the text the solute threshold, except at page 13, where we specify the different terms.

2. Table 4 has been removed and replaced by table comparing the averaged concentrations for the 3 different air mass categories. This table include molar ratios for different species that are discussed in the text.

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Specific Comments:

1) Page 4 2nd and 3rd paragraph; we combined the 2 paragraphs.

2) Page 4/page 5; The cloud frequency in winter/spring period is much higher than during the rest of the year. In addition, cloud forms in free tropospheric air. For these reasons we limited the sampling period.

3) Page 5 (2.1; 1st paragraph); We have now added some information on the particle number concentration measured at the station. This is not an unequivocal marker for free tropospheric air but it simply shows that we are not influenced by local emissions.

4) Page 5 (2.2; 2nd paragraph); the routine measurements are specified in 2.1 section (1st paragraph): cloud water sampling is not a routine operation but is specific for this work (we specified it in the text)

5) Page 6 (2.2; last paragraph); a short description of the aerosol sampling is now provided. We also mentioned the gas-phase sampling to clearly indicate that all these measurements were performed simultaneously. Aerosol and gas phase data are already published and proper references are given.

6) Tables 1 and 2 were combined in Table 1, that has been completed to provide the reader with information on air mass origins, sampling conditions as well as aerosol sampling.

7) Page 7 (2.3.1) A table with the detection limits has been added (Table 2).

8) Page 7 (2.3.2); there is no addition of H3PO4, the sample is injected in a solution of H3PO4 (25 %) for the measurement of IC. The injection of the sample in the solution of H3PO4 allows the transformation of carbonates and hydrogeno-carbonates into CO2. Afterwards, the formed CO2 is measured in the NDIR cell. with this methods we measure only carbonates and hydrogeno-carbonates. (The organic acids lost during the acidification are not transformed into CO2). We have modified a little one sentence to clarify this point.

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9) Page 8; we have modified the paper accordingly : first by adding median values to Table 2, second by referring to median instead of averages in the discussion p. 857-860, and finally by adding a table showing average values for each of the three categories (Table 4).

10) Table 3; we added the request details. The method valid for additional mono and di-carboxilic acid was settled at the end of 2001 sampling season: 5 samples from 2001 were analysed also for better speciation of CA.

11) Page 8,2nd paragraph; the decoupling between free-troposphere and boundary layer air reflected by CPC data : the CPC number concentration is now added to the text.

12) Page 8, 3rd paragraph; the consistency of the thresholds for the different air mass categories has been rechecked throughout the paper. At this point we do not give a classification, but just a comment on the shape of frequency distribution of Total Ionic Content shown in Figure 1. The classification is subsequently derived from this graph.

13) Page 8, last paragraph; back trajectories for the SpE clearly show that high concentrations are associated with air masses originating from the heavily industrialised region of Northern France and the United Kingdom. These air masses are clearly from the free troposphere not from the boundary layer. The high concentrations are therefore not inconsistent with the statement that PDD lies in the free troposphere in winter / spring period. The concentration level is not a seasonal parameter.

14) Page 9, last paragraph; it is now specified in the text that NH3 concentrations are available in 2001 for time period corresponding to the cloud samples. High NH3 concentrations were also found during a previous campaign at PDD. Techniques used for these 2 campaigns are different and we can exclude that this is a result of local contamination. Rather, it has to be kept in mind that, regardless of their origin, the air masses reaching puy de Dôme summit have passed over forests and intense farming areas, the influence of which may not be limited to the BL.

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15) Page 10, 3.1.2; the acronym DOC is now used throughout the paper.

16) Page 11, first paragraph; we reformulated the sentence in order to be less categorical, but the correlations between BC, DOC, NCPC and NH4+ strongly suggests this hypothesis. We add information on correlation between DOC and NCPC in the text.

17) Page 11, last paragraph; we specified in the text that the contribution was to total solute content.

18) Page 12, last paragraph of 3.1.2; that is true except that photochemical reactions may not be so important during winter/spring. The H2O2 measured at PDD during CIME (Laj et al., 2001) and concurrently with this sampling was low.

19) Page 12, bottom; same comments as referee 2 and points 2: the tables are now presented with median values (that are not so strongly influenced by SpE events) and table 4 was added to specify averaged concentrations and ratios for the 3 air mass categories.

20) Page 13; as mentioned before (and this is now better indicated in the text), the NH3/NH4 measurements are discussed in the paper of Sellegri et al. 2003c. We feel that there is no reason to provide more than a reference to this previous study in the present paper. The thresholds are based on the frequency distribution. This is now more clearly stated in the text.

21) Table 5 Page 14; Tables 1 and 2 were combined: Table 1 now includes more information regarding air mass origin, sampling conditions and aerosol sampling.

22) Page 15, 3rd paragraph; the molar ratio Na/Cl in cloud water is 0.81 (very close to 0.86 of sea water): the lack of Cl in aerosol phase is due to evaporation of Cl in form of HCl and its solubilisation back in presence of liquid water. Reference to the work of Sellegri et al. (2003c) that investigate the HCl behaviour is now provided in the text.

23) Page 16, 1st paragraph; CWL, Cloud Water Loading was already defined earlier in the text (Page 14, 2nd paragraph).

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24) Page 16, Conclusions ; OK, same comment as 1: The consistency of the thresholds for the different air mass categories has been rechecked throughout the paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 849, 2004.