

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

Anonymous Referee #1

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General Comments

The paper presents very interesting data consequently I gave a good rating during the first step of the evaluation process. However looking at the paper in more detail I found a number of inconsistencies or shortcomings which really need improvements. In this outline I want to mention three examples:

The thresholds for classification of BG or ANT events are a total solute content of 18 mg/l or 50 mg/l in the abstract and 15 mg/l and 40 mg/l later on page 12 or in the conclusions, the numbers of the abstract are repeated on page 13

It is difficult to combine Table 3 and the first paragraph on page 9. Is statistical data of samples or events given in Tab. 3? Obviously #14 is included in Tab. 3, but not in the text. Please specify. Event #14 definitely is included in Fig 1.

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The whole data set is never shown in a way which would allow to check one or the other statement given in the discussion.

I still regard the data set as very interesting and I like several of the evaluations in the discussion! Nevertheless I feel uneasy about the way the data is presented.

I like the intention of the paper but it definitely needs some improvement!

Specific Comments:

Page 4, 2nd and 3rd paragraph These paragraphs could be combined and shortened .

Page 4/page 5 Sixteen cloud events have been monitored during a period of two years, but they cover "only" the period of February to May; that is fine, but a "period of two years" as stated in the introduction is misleading

Page 5 (2.1; 1st paragraph) The station lies in the free troposphere during winter/spring months; really? please show data or refer to another paper

Page 5 (2.1; 2nd paragraph) I guess that the experimental set-up described here refers to routine measurements at PDD and 2.2 gives the description of additional measurements performed within this project; please specify (e.g. in the heading)

Page 6 (2.2; last paragraph) A very brief description of the aerosol measurements has to be given here in addition to the reference to Sellegrí et al.

Tables 1 and 2 It would be interesting to combine these tables to get the information of simultaneous (or not) sampling more easily.

Page 7 (2.3.1) Add the detection limits

Page 7 (2.3.2) How does the addition of H₃PO₄ affect the concentration of organic acids? Does IC only comprise carbonates and hydrogen carbonates or are organic acids lost during acidification of the samples as well?

Page 8 I have problems to get an overview over the whole data set. As I understand

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authors averaged the samples to events and further discussion will be based on these events. But do the numbers given in Tab 3 refer to samples or events? In 2002 the number of events is equal to the number of samples anyway (disregarding #14) For me the table is not really helpful. I would consider to group the events according to BG, ANT and SpE and give min, average and max values of events for these classes. Percentiles could be omitted in that case, but please give number of events in the respective classes.

Table 3: please add a legend, Re or JNO₂ might not be straightforward to everybody, specify that samples or events are given in the table According to Table 1 10 samples have been collected in 2002 ; why do you report 15 samples for organic species determined in 2002 only? And what about fluorure ; it was not mentioned earlier, same for MSA. 78 samples for species determined during both years is in agreement with Tab. 1.

Page 8, 2nd paragraph Is the decoupling between free-tropo and boundary layer air reflected by CPC data? ; please comment

Page 8, 3rd paragraph solute range between 15 and 40 mg/l again points to an inconsistency with the classification given in the abstract ; although no classification is given here!!!

Page 8, last paragraph Which are the highly concentrated samples ; are they evenly distributed between February and May? (would be interesting compared to the statement of PDD being in the FT during winter and spring)

Page 9, last paragraph Could the elevated ammonium concentrations during ;samples be a result of contamination. It seem surprising, that ;free tropospheric samples; are influenced by agricultural activities on the regional scale.

Page 10, 3.1.2 Here the authors refer to DOC ; in the experimental section

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TOC was described.

Page 11, first paragraph On the whole I cannot see that these results clearly show that a significant fraction of DOC is not associated with long-range transport please add some explanations e.g. Why is the correlation of DOC with CPC regarded as in contrast to an influence of anthropogenic air masses.

Page 11, last paragraph Contribution of organic acids and major ions to what? TIC? (cannot be DOC as in the paragraphs above)

Page 12, last paragraph of 3.1.2 Isn't it surprising that organic acids and ammonium show a lot of similarities when the major organic acids are strongly influenced by photochemical reactions as well?

Page 12, bottom As some evaluations are based on events and some based on samples please specify that the classification was done for events (or samples). Please add how many events could be classified in the respective categories.

Page 13: Here high levels of ammonium are mentioned for polluted cases; this is in contrast to earlier findings (pages 9/10 and Fig. 2). As Tab. 3 gives an insufficient picture of the data set; it is impossible to check these findings or to get more information to better understand the text.

Generally it might be better to put this section to the beginning of the text. The thresholds were set; arbitrarily; thus this could be earlier as well.

Table 5 / Page 14 To understand whether differences in sampling time of cloud water and aerosol samples are likely to contribute to the differences of the classifications it would be nice to have a combination of Tables 1 and 2.

Page 15, 3rd paragraph If HCl accounts for the difference of 5% to 11% a rather high concentrations of HCl has to be expected. I did not (could not) compare molar ratios of sodium and chloride, but can we expect such a good correlation of sodium and chloride

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if dissolved HCl leads to a doubling of the contribution to TIC?

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Page 16, 1st paragraph Please define CWL .

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Page 16, Conclusion Please check threshold values!!

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