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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contribution of gaseous and particulate species to droplet solute composition" *by* K. Sellegri et al.

Anonymous Referee #2

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General Comments

The manuscript presents very interesting data. Unfortunately it is quite difficult to read because of several obscurities and sometimes erroneous statements (either erroneous, or I simply could not follow the text). On one hand (unnecessary) details are given (e.g. p 485, line 23 to 25, the volume was measured with gas volume meters and corrected for the pressure drop), on the other hand the description of the various inlets (one of the most important topics to understand the paper!!) is really short and confusing (both the text and Fig. 1). At the end of the introduction the authors state that they propose an experimental methodology. Thus the description of this methodology should be important. Unfortunately a few lines later they just refer to two different papers (one still in press) where the experimental deployment is discussed and only recall the characteristics, but unfortunately not detailed enough. Later on the authors

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are a bit sloppy with the use of the terms interstitial aerosol, condensed phase and particulate. Together with the poor description of the sampling inlets it is even more difficult to understand several passages of the discussion. Furthermore I miss a statement about the sampling site in the title and the abstract. You have to read until the end of the introduction to get the information that sampling was carried out at Puy de Dome. I would not recommend the differentiation between Results and Discussion. This leads to repetitions and is confusing.

As an overall rating I would recommend the publication of the manuscript, but several really important revisions have to be done.

Specific Comments

Page 484, description of the experimental set-up: As mentioned above this section has to be improved. In the following I want to outline some points: The CVI is used to sample droplets $>5\mu$m. Analysis is performed after evaporation of the droplets. But what is actually analysed? The residual particles? The intersitial aerosol sampled with a cascade impactor in the CVI ? (see page 486) on the other hand interstitial aerosol should be sampled with RJI inlet (according to page 484), but the authors do not refer to this set-up in the chemical analyses section at all. How did they sample the data given in table 6. Regarding page 496 the cascade impactor use in the CVI gives data of particles which are residues from cloud droplet evaporation. Is this a contradiction to page 486? What kind of measurements were performed in the WAI? Only mist chamber measurements? If WAI compares to CVI plus RJI gas phase concentrations have to be measured in the CVI (it might be, but I am not sure. According to page 485 no MC is installed there and according to page 486 line 22 gases are excluded in the CVI). On the other hand, to compare WAI to the sum pf CVI and RJI, some measurements of the residues (after evaporation of the cloud droplets) have to be carried out in the WAI. Again I cannot find the necessary information in the experimental section. Figure 1 does not really improve the understanding of the four sampling devices. On page 484 the authors say that the four sampling devices are clearly complementary. In the

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present version of the manuscript I have problems to agree with this statement.

Table 1: Why report the number of MC (behind RJI or WAI) and CDI samples and not aerosol (impactor) samples or CVI samples. (Later it is reported that one CVI/cascade impactor sample corresponds to an event.) Perhaps it would be better to report the number of samples in the respective tables (i.e. Tables 3 to 6). Why does the number of samples differ so much and which time period is covered by (e.g.) the MC samples (according to page 485 sampling duration was 1 to 3 hours, while event 4 lasted approx. 11 hours and only one MC sample is reported. How were the calculations made when the different sampling methods covered different time periods.

Page 488, line 19, If large measurement errors are associated to some of the result (and this does not necessarily question the importance of the measurements) it would be helpful to comment on these uncertainties in the discussion section.

Page 489, line 3, 80% of mass?

Page 489, line 23, According to Table 1 the LWC does not change so much.

Page 491, line 20 to 27, The high ammonia concentrations and the statement about the agricultural source are in contradiction to the characteristics of a free troposphere site (page 485, line 9). Furthermore the occurrence of these above average concentrations (compared to Voisin et al. 2000) together with cloud water samples being slightly acidic is surprising, however this is discussed later on.

Page 493, line 1 to 5, In case of nitric acid /nitrate it seems somehow difficult to replace Śinterstitial-phaseŠ concentrations with gas phase nitric acid only, as intersitial aerosol nitrates average much higher than gas phase nitric acid. The same applies for sulfur dioxide/sulfate during 4 of the 5 cloud events.

Page 493, line 11: condensed phase should be replaced by liquid phase (if this is correct!!) I am not sure as in line 15 in the same context a ratio of NH3/NH4 is reported. This number (62%) is different to the median value given in Table 8 (and also different

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to some result given in Voisin et al. (2000)) although the results of Table 8 are reported to be similar to Voisin et al. (2000). What is this ratio NH3/NH4? It is unlikely, that the intersitial aerosol concentrations of NH4 will make such a difference.

Table 8: What kind of fraction or ratio is given by Voisin et al. (2000) and Kasper and Puxbaum (1998)? Please specify in the legend.

Page 494, lines 18 to 28 These lines could be omitted / shortened as complete oxidation is assumed and no S(IV) traces are found in the liquid phase.

Page 495, line 26, Do the results from CVI correspond to scavenged aerosols or to the droplet residues? The droplet residues might be more than the scaveged aerosols (if chemical transformation has occurred) or less if an semivolatile aerosol particle evaporates during sampling with the CVI.

Page 496, lines 8 to11 This statement about averaging and comparison of different samples should be discussed in the experimental section.

Page 497, lines 23 to 29 The degassing of ammonia from an acidic sample is really surprising Ű I agree. Is there the possibility of a contamination?

Figure 3: I had some difficulties to recalculate the gas-originating Liq and part-originating Liq according to Table 9 . Is this just an averaging problem?

Technical Comments:

Page 492, line 8, table 6 are the interstitial aerosol concentrations, not the residues after cloud droplet evaporation, thus table 6 should be given in line 6, while data of the residual phase are given in table 4

Page 493, line 10 The text refers to table 8 although table 7 has not been mentioned yet.

Page 497, line 1 Cx=100% is not defined but obviously refers to Degassing % given in Figure 2.

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