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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Long-term observations of positive cluster ion concentration, sources and sinks at the high altitude site of the Puy de Dôme" *by* C. Rose et al.

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We thank Referee N°1 for his very helpful comments and suggestions. We feel they have greatly improved our manuscript. We have addressed the comments point by point below.

Comment 1: Cluster ion concentration evolution is not likely driven by one factor only. How cluster ion concentration changes as a function of source-rate to sink-rate ratio, where sink is from coagulation to background aerosol and ion-ion recombination? Temporally quickly varying component (radon decay) of ion production, which was mea-

sured at the site, should be deployed instead ion production derived from cluster concentration itself.

Reply 1: Indeed, the cluster ion concentration is impacted by both sources and sinks at the same time. Both the sources of ion production and the sinks are included in the balance equation that we are using, and which is a reference tool for this kind of study (e.g. Laakso et al., 2004; Hirsikko et al., 2007; Franchin, 2009). This equation also allows the simultaneous study of the three parameters of interest (concentration, source, sink), by considering the difference between sources and sinks. The use of the difference between sources and sinks has the advantage of being quantitative compared to the use of the ratio of sources to sinks. Moreover, we have shown that ionization rates derived from the balance equation were on average closed to the ionization rates derived from radon measurement and estimated GCR contribution (the difference between calculated and measured ionization rates is 0.19±2.26 cm-3s-1).

Comment 2: Is the nucleation event classification statistics different for positive and negative polarity? To my knowledge such a difference is very likely. I think that analysis of concentration variation on event vs. non-event days should be done separately for different event classes I+II and bump, and thus for different polarities, since they represent different atmospheric conditions. Especially days when particle formation is observed from larger sizes should be considered separately. Is concentration of positive and negative ions in growing nucleation mode (clearly) different at larger sizes (e.g. around 3 nm and 5 nm)? This kind of analysis could give obvious explanation why relatively slow growth of positive cluster ions can be observed while negative ions grow quicker and their cluster concentration shows less variation at the site (page 14939, lines 26-29).

Reply 2: Statistical differences between positive and negative ions were already described pages 14937-14939 of the paper. We understand this information was not found to be sufficient for the reviewer, so additional statistics on the differences between positive and negative ions are now added to the text page 14939 and in Table

3. Studying further differences between positive and negative ions according to event type would considerably complexify the paper, which is already quite long. The primary goal of this paper is to study the ion cluster concentrations and their relation to sources and sinks. The growth of different polarities and the parameters explaining these growth rates would be another complete study on its own, especially in the complex environment of a mountainous area.

Comment 3: In section 3.3.3, the authors did not have gamma radiation observations to directly estimate total ionization rate at their site. Therefore, authors have taken gamma radiation dose rate reference from other study (Laakso et al., 2004), which represents different environment and is differently influenced by ABL dynamics. Laakso et al. (2004) analyzed ion production through measurements and ion-balance equation based on two months of observation at a boundary layer site. However, the study by Laakso et al. (2004) shows hints that ground based gamma radiation dose rate is not constant throughout year which was later confirmed by e.g. Franchin (2009). Therefore, it is necessary to present references of gamma radiation dose rate in France/close to Puy de Dome (e.g. Billon et al., 2005 or more appropriate) and present sensitivity analysis based on reasonable range of gamma radiation dose rate. Non-existing knowledge of gamma radiation contribution on ionization rate lessens value of this work and may lead to wrong conclusions.

Reply 3: This is a good comment. Considering studies by Hirsikko et al. (2007) or Franchin (2009), it is clear that the external radiation contribution to the ionization rate exhibits a seasonal variation with higher values in summer and fall. Thus, even if the seasonal variation does not impact daily scale variations that can be seen between event and non-event days and which are of interest in this paper, more accurate estimations of the gamma dose rate were considered. Since no appropriate external radiation dose rate measurements were available for the Puy de Dôme (Billon et al. (2005) only gives orders of magnitude with no seasonal indication), we finally used values from a more similar site than Hyytiälä, i.e. at BEO Moussala, Bulgaria, which is

a high altitude station (2925 m a.s.l.). In Section 3.3.3, the main focus is maintained on the quickly varying component of ion production, radon, since the main purpose of the analysis is to find the link between ion production and increased ion concentration on event days. In order to discuss the contribution of external radiation (which does not show daily cycle) on ionization rate and to give more relevant numerical values of the measured ionization rate, an additional Section 3.3.4 is now proposed. When using measurements from BEO Moussala, the contribution to the ionization rate from external radiation that we finally obtain are 10.09, 9.74, 11.76 and 11.14 cm-3s-1, from winter, spring, summer and fall, respectively. These values are similar to the median value reported by Franchin (2009) for Hyytiälä (10.12 cm-3s-1) and significantly exceed ionization rates derived from radon measurements (0.94-1.57 cm-3s-1). However, the variability of the calculated ion source is very similar to the variability of the ionization rate derived from radon, indicating that external radiation contribution to the ion source is at a stable level that do not influence the ion concentration variability observed at Puy de Dôme.

Comment 4: Figs. 12 and 13 show major deviation between ionization rates based on two different methods, not similar values as authors indicate. These differences are likely to arise from non-steady state conditions and incorrect estimate of terrestrial gamma radiation contribution.

Reply 4: When considering all the seasons together, with both event and non-event days, we obtain the difference between calculated and measured ionization rates in the range 0.19$\pm$2.26 cm-3s-1. This result suggests that ionization rates based on the different methods are on average closely related. However, we must recognize that for some couples of values the differences are not negligible, and in that case imbalance between sources and sinks seem to be a plausible explanation. Thus, we finally replaced the sentence “For all seasons, the measured and the calculated ionization rates are on average in good agreement”, by “On average, the measured and the calculated ionization rates are of the same order of magnitude and display the same temporal variations at the intra-seasonal scale", . Indeed, we do believe that terrestrial gamma contribution does not drive the differences that can be observed between calculated and measured ionization rates. This is explained by the fact that the most significant variations reported in the literature (e.g. Hirsikko et al., 2007 ; Franchin, 2009) are visible at the seasonal scale and not at the daily scale. As a consequence, we assume that these variations cannot be responsible for intra-seasonal discrepancies between ionization rates derived from the two different methods.

Comment 5: What is a plausible unknown source the authors mention on page 14949, lines 25-29? Air richer with cluster ions transported from valley below the station, and thus is related to ABL dynamics?

Reply 5: When considering more relevant terrestrial gamma radiation contribution to the ionization rate, as suggested in Comment 3, ionization rates based on measurement always exceed ionization rates derived from the balance equation. In that case, if we assume to be well estimated, we are systematically missing a sink in the balance equation. The only source we are still looking for is to justify increased cluster ion concentrations on event days. The source proposed in Comment 5 does not seem to be plausible in that case since there is no reason for radon from the valley not to be transported with the clusters. Since radon concentrations are not increased on event days, we must believe that the influence of the valley in cluster ion concentration is weak.

Comment 6: I feel that manuscript analyses concentrations of both polarity cluster ions. Therefore, I suggest removing word positive from the title.

Reply 6: We agree and changed the title accordingly.

Comment 7: Page 14929, lines 6-7: " The aerosol indirect effect is still affected by the largest uncertainty among atmospheric radiative forcings (IPCC, 2007)." This sentence requires modification.

Reply 7: We agree with the fact that the wording of the sentence was not optimal, so

we changed it to: "Despite the fact that atmospheric research, and especially climate and pollution purposes, have focussed the attention of the scientific community during the last decades, the radiative forcing associated to the aerosol indirect effect is still undergoing a large uncertainty (IPCC, 2007)."

Comment 8: Page 14930, line 3: ''mobility diameter" should be ''mobility".

Reply 8: Correction was made.

Comment 9: Page 14930, lines 9-11: " while in continental areas, the variation of the small ion production is mainly driven by the variation of the radon and thoron concentrations (Laakso et al.,2004).": This is not quite correct, since evolution of small/cluster ion concentrations dependents on balance between source and sink rates.

Reply 9: It is true that the evolution of the cluster ion concentration is impacted by both sources and sinks at the same time. However, the purpose of the sentence is the cluster ion production, and not concentration.

Comment 10: Page 14930, lines 18-20: "For the ion induced nucleation mechanism, the maximum of the nucleation rate was previously found to correspond to the ionization rate (Yu and Turco, 2000)." Please modify this sentence, since it would be unphysical to have higher ion loss rate than source rate.

Reply 10: Yes, this was not clear written as so. We have changed the sentence to "For the ion induced nucleation mechanism, the ionization rate is a key entity governing the nucleation rate".

Comment 11: On page 14939, lines 12-21: the authors discuss the observed diurnal variation of cluster ion concentration in the ABL sites: Mixing of the ABL affects also on cluster ion concentration in the observation volume, not only ion source from radon decay.

Reply 11: We do agree. Indeed, besides the fact that the boundary layer dynamics impact ion sources, they can also have a direct effect on the ion concentration itself.

This suggestion was added to section 3.2.: "One should note that these dynamics of the BL can also impact the cluster concentration itself, by a concentration or dilution effect of cluster ions in the observed volume".

Comments on figures: Page 14967, Fig. 5: Data points should be more visible; Page 14971, Fig. 9: Small red and blue markers are difficult to distinguish from each other. Please find a solution to present different distributions clearly; Page 14974, Fig. 12: Indicate which season each figure represents.

Reply: All comments on figures were taken into account.

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