

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.

Anonymous Referee #1

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General comments:

The work by Rose et al. investigates aerosol particle formation, cluster ion concentration evolution, and their sources and sinks in a mountainous site in France based on 5 years of observations. Presented report of particle formation event day frequency is consisted with previous study (Boulon et al., 2011), mainly because 3/5 of time observation period is the same. However, scientific value of this work is on analysis of small/cluster ion concentration and source/sink rate temporal variation at Puy de Dôme. Maximum rate of air ion contribution into nucleation depends naturally on their source rate. Temporal evolution of ion concentration is determined by balance between

C4493

sources and sinks. This kind of research is hampered by lacking simultaneous observations of at least gamma radiation dose rate, radon decay rate, and cluster ion and background aerosol particle concentrations. The manuscript shows a number of various analyses to identify reasons behind cluster ion concentration temporal evolution. In addition to that, most of the recent studies of air ion concentration, their source/sink rates and contribution on atmospheric particle formation are from atmospheric boundary layer (ABL) sites. Puy de Dôme is one of the exceptional infrastructures which are in free troposphere much of time making analysis of cluster ion source and sink rates challenging, but more interesting.

Specific comments:

Authors analyzed differences on cluster ion concentration on event and non-event days, and have shown analyses of effect of single factors on cluster ion concentration. I suggest the authors to consider the following before concluding effect of different factors on cluster ion concentrations at their site.

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

C4494

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).

Section 3.3.3.: This section requires revision due to the following reasons:

1. 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.

2. 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.

3. 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?

Technical comments:

Page 14928, title: I feel that manuscript analyses concentrations of both polarity cluster ions. Therefore, I suggest removing word positive from the title.

C4495

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.

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

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 depends on balance between source and sink rates.

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.

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.

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.

References

Billon, S., et al., French population exposure to radon, terrestrial gamma and cosmic radiation, *Radiation Protection Dosimetry*, 113, 314-320, 2005.

Boulon, J., et al.: Investigation of nucleation events vertical extend: a long term study at two different altitude sites, *Atmos. Chem. Phys.*, 11, 5625-5639, 2011.

C4496

Franchin A., Relation between ^{222}Rn concentration and ion production rate in boreal forest, Report series in Aerosol Sciences, 105-108, 2009.

Laakso, L., et al.: Ion production rate in a boreal forest based on ion, particle and radiation measurements, *Atmos. Chem. Phys.*, 4, 1933-1943, 2004.

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C4497