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Atmospheric Chemistry and Physics Discussions

Interactive comment on "From ionising radiation to air ion formation in the lower atmosphere" *by* Xuemeng Chen et al.

Anonymous Referee #2

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The paper describes the acquisition and analysis of a long-term (3 year) data set from a boreal forest site on ionising radiation and cluster ions, attempting to ascertain relationships between them and determine factors responsible for their variation in diurnal and seasonal cycles. Both the experimental data and analysis are very extensive and provide useful insights into the role of ionising radiation from radon and progeny, and gamma radiation from the ground, in producing new cluster ions under varying meteorological and seasonal conditions, with a particular focus on new particle formation events. The work presents a significant advance in the understanding of the influence of ionising radiation on cluster ion concentrations, and infers information about the formation process itself, and hence would be a welcome and worthwhile addition to the literature in this research area. However, I feel that some minor revisions are necessary before the paper can be published. Some potential confounding factors e.g. Rn-220

(thoron) and particularly cosmic ray ionisation, data for which are not presented, are not discussed sufficiently and would benefit from being considered in the context of, and/or justified in terms of their exclusion from, the overall analysis. Also, citation of, and comparison of results with, previous work could be improved upon in the Introduction section and in discussion.

Specific comments

The paper in general would benefit from additional discussion of previous work, in comparison with results presented, but is required in particular at specific points in the manuscript:

a) In the Introduction, in particular regarding air ion mobility spectrometry, there is little citation of work undertaken outside the groups of the authors and Tammet and co-workers. Substantial though this body of work is, there are other noteworthy contributions to the literature, especially in a historical context, and I encourage the authors to expand upon this in their Introduction.

b) Section 3.3.2 describes "Variations in cluster ion concentrations in sub-size ranges" which, to a certain extent, results in a discussion in part about the ion mobility, since if ion mobility is different (either in response to atmospheric changes e.g humidity, or as a result of different polarity) then ions may be classified into different categories here. There is a reasonable body of work in the literature describing ion mobility and humidity effects which may have a bearing on results presented here, and general conclusions (positive ion concentration > negative concentration, positive ion mobility < negative mobility) are in common with several previous results.

Cosmic rays are acknowledged as a significant contributor to atmospheric ionisation and hence cluster ion formation, and briefly mentioned here but are not explicitly accounted for in this work - it is implied that some influence is folded into the 'gamma ionising capacity' data (P9 L1) but this may only account for the proportion of activity corresponding to lower-energy photons in cosmic ray showers detectable by the

spectrometer used (100-3000 keV) and would miss a substantial 'cosmic ray ionisation capacity'. If data on cosmic ray flux at the measurement site is available, this would be a very worthwhile inclusion to the paper, even if it is only discussed in a qualitative manner. For example, were there cosmic ray events or solar energetic events during the measurement period which might have influenced ionisation in a manner not accounted for in the Rn and gamma ionisation capacity data? What proportion of overall ionisation might be caused by sources other than the Rn and gamma ionisation captured in the data presented, and is this expected to be constant throughout the study or variable? This discussion might be particularly relevant given the high latitude of the measurement site.

Also, it may well be that there is not sufficient ionisation by thoron decay to justify including this as a factor in the study, and indeed this has been suggested in previous work reported from Hyytiälä (Laakso et al. (2004) ACP 4, 1933) but the possibility should at least be mentioned here, even if only to clarify it is insignificant.

I don't think Figure 1 in its current form is necessary. The concepts are adequately described in the text and the figure does not add much value to the description. If an 'overview' figure is deemed helpful by the authors, perhaps a schematic introducing the processes involved in cluster ion formation and growth due to ionisation, vapour condensation, recombination etc. would be more instructive.

Section 3.1 and Figures 2-4 show seasonal analysis, splitting the annual cycle into 4 3-month periods. However, looking at Figure 2, the 'Spring' period shows a significant change in, in particular, the gamma ionising capacity. So, for example while the average ionising capacity (gamma + Rn) is similar to Winter when taken across this period March-May as a whole (Figure 3), the ionising capacity increases significantly during this period and also the relative importance of gamma and Rn changes. It may be worth repeating some of the analyses only for, say, mid-February to end-March, to examine this specific period where Rn contributes the largest fraction to overall ionising capacity across the whole year, as I wonder whether features are being diluted and missed by

taking the average over a period in which large changes in these parameters occur.

P4 L25 Please clarify whether 'during 2003-2006' means whole-year data collection for these years, or if not, in which months data collection/use began/ended.

P6 I note also that APi-TOF may not provide direct information on the actual electrical mobility of atmospheric cluster ions, as encountered in environmental measurements using air ion spectrometers, because of the evaporation of clustering water molecules during the APi-TOF measurement.

P7 Please state the voltage scan time for the BSMA in this work (it looks like it appears in the legend to Figure 8 but should be specified in the main text as well).

P8 L11 Please clarify your justification for using only 'O horizon' soil data.

Minor technical corrections

P4 L25 "Monitoring devices for" not "The monitoring devices of the"

P5 L15 "described" not "descripted"

P8 L8 "Time-Domain Reflectometer" or "...Reflectometry (TDR) device."

P12 L10 "examine" not "exam"

P12 L22 what is meant by "shading the variability"?

P13 L9-11 move text "to the measured snow depth data" to L9: "exponential fitting ... could represent" or otherwise reword, to clarify what is relationship is being probed.

P14 L9 "blooming" not "booming"?

P20 L9 "localised" not "focalised"?

P20 L26 "level out" not "level up"

P22 L2 "support" not "supports"

P22 L19-22 the full citation now appears to be available online, please amend.

P33 L4 "0-4 cm" not "-4-0 cm"

P34 Fig 2, both y-axes "ionising" not "ioning"

P43 L3 "T was below" not "T below"

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