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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 #1

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In this paper Chen et al present a comprehensive set of atmospheric ion measurements in combination with detailed measurements of radioactivity. The heavily instrumented site used permits a very detailed analysis of the effects of ionising radiation on the concentration and mobility (size) of atmospheric ions, and how the ions are modulated by seasonal and meteorological factors. This is the most thorough analysis of its type that I have seen, and potentially makes a valuable contribution to understanding the role of ionising radiation in the variability of atmospheric ions. Some interesting differences in the contribution of ionising radiation on days when there are new particle events are also identified.

Despite the high quality of the data, there are several deficiencies in the way the paper is written, in particular, an apparent unfamiliarity with the literature in this area. The introductory material is narrow, and implies that (a) this is a topic which has hardly

been worked on and (b) there is limited understanding of how radioactivity makes ions. Since radioactivity and cosmic rays were discovered through their ionisation of the atmosphere at least a hundred years ago, these implications are incorrect. The authors should be referring to some historical material (e.g. the Irish work of Nolan and collaborators) or, at a minimum, a historical review, and the work on ion measurements from other groups in the late twentieth century, to explain how their study progresses the research area. The motivation of the paper needs to be refocused towards the work that is described, which is a far more sophisticated study of the interaction between radioactivity and the properties of atmospheric ions than the introduction suggests. The title of the paper also implies a much more basic study than is actually carried out and might be better changed to something like, "Effects of ionising radiation on ion mobility in a range of atmospheric conditions" (this is just a suggestion and should not be taken literally, but I hope you understand my point).

A second aspect of the paper that indicates lack of awareness of other studies is that the authors present their own definitions for terms that are already precisely defined. For example, they introduce a term called "ionising capacity" which to all intents and purposes appears to be identical to what the rest of the community already calls "ionisation rate", since it has the same units and even the same symbol. The only justification I can think of for bringing in this new terminology would be if the ionising capacity were a theoretical maximum amount of ionisation, given the energies and activities of the particles involved, which may not be the same as the actual ionisation rate. However, since the authors make no attempt to justify their new definition, and also, the only way to measure the "true" ionisation rate would be through detailed ion measurements of the sort made by the authors, making the distinction between "capacity" and "rate" doesn't seem particularly helpful. Some of the energy of the radioactive particles will be used for excitation and not ionisation, but I would imagine that this is a relatively small fraction, and it is not unreasonable to assume that all the energy lost by the radioactive particles goes into ionisation. I recommend that the authors remove the references to and definition of "capacity" and simply talk about ionisation rate for con-

sistency with other work. They could add a caveat mentioning energy loss to excitation if this concerns them.

The wheel is reinvented again in figure 1. Many years of work in the early twentieth century went into defining cluster ions, small ions, large ions, etc. If the authors are going to come up with their own definitions they must explain why they are needed. In particular, the distinction between primary and molecular ions seems arbitrary, since the primary ions N2+ and O2+ are technically molecular ions anyway, and the ions you call molecular ions I thought were unstable in the atmosphere and cluster immediately. Fourthly, the authors do not seem aware of the known theoretical relationships between ionisation rate and ion concentration. The paper they cite by Harrison and Carslaw (2003) contains a good introduction to this theory. The lack of theoretical awareness is particularly apparent when looking at figure 13, which seems to be a very nice demonstration of the ion balance equation in the recombination limit. I've seen similar plots before (e.g. in Aplin and Harrison, Rev Sci Instrum 2000), but this is by far the best data, however the authors do not place this work in context. They need to talk about the ion balance equation and compare their work to those of other groups; this will demonstrate the strength of their data.

The paper presents a huge amount of data is presented, in a long main text, but the conclusions are brief bordering on the obvious. The conclusions need to present a synthesis of the data and put it into context in the conclusions. It is also not clear why the results are important – for example the last sentence of the entire paper recommends that instruments that can measure sub-0.8 nm ions are needed – why? The results on new particle formation are novel and very interesting, but can you discuss their implications?

A final scientific point is that the authors do not adequately explain at the beginning of the paper that they are omitting to measure most of the cosmic rays and that they usually contribute about 20% of the ionisation rate. They are mentioned in the context of high-energy gamma radiation, but there are muons and electrons too which are not

mentioned. Later in the text, it is implied from the residuals that the cosmic ionisation rate is 3 /cc/s, and it is argued that because the "textbook" rate is 2 /cc/s, then one third of the charge created is lost before even becoming atmospheric ions. This seems totally speculative; surely it is much more likely that the fit/measurement errors and the high latitude of the Finnish measurement site and perhaps solar activity could account for the discrepancy. It would be possible to carry out a more quantitative analysis here.

Typographical/minor errors

P3 L12 "generated electric charges from . . . by the derivatives of them" does not make sense

P4 L7-21 Please revise this paragraph in line with the comments above

P5 L15 "descripted" -> "described"

P6 L17 All ion spectrometers are defined by their upper and lower mobility limits, so this sentence is meaningless. Why is the upper mobility limit 3.2 cm-2V-1s-1?

P6 L20 What are the potential consequences of the choice of mobility-diameter relationship and its parameters?

P7 L1-2 please be consistent with units.

P7 L2 how and why is only one eighth of the air sampled?

P7 L13 "programme" -> "program"

P8 L8 Please explain what the condensation sink is and its units; not everyone will be familiar with it.

P8 L6-18 this is quite a lot of meteorological detail for work which is presumably published elsewhere. Could you shorten it and cite a reference?

P8 L22 Notwithstanding the comments above, should these units include a "per unit volume"?

P9 L15 This statement is meaningless since you derive the “ionising capacity” directly from dose rate in equation 1. Please remove

P10 L19 Can you explain why marine air masses will have a low radon content; again, not all readers will necessarily know why.

P12 L10 “exam” -> “examine”

P12 section 3.2.2 Since radon emits gammas, which are included in your gamma radiation as you said earlier, can you state more clearly that you only mean alphas and betas from radon here?

P14 L8 would be better to say “growth season”

P14 L23 not sure what is meant by, “there is a hindrance”

P14 L24 what is the significance of the 1.7 nm threshold?

P15 L14-18 As it stands, this is irrelevant to the rest of the paper and should be deleted.

P15 L20-27 It is well known that negative atmospheric ions are smaller than positive ones, though this work offers more detail. Please cite the classical literature here.

P16 L16 not sure what is meant by molecular ions here.

P17 L4-6 This statement is speculative and needs to be qualified as such.

P17 L7 You need to add some words on CS here.

P17 L20 what do you mean by “unaltered gamma ionising capacity level”?

P19 L1 what do you mean by “clustering or simple charge binding?”

P19 L20 “focalised” -> “localised”

Table 3: The labels are not adequately explained – could perhaps talk about quantiles to make it clearer what you mean. Please also define STD. As discussed above, it would be better to talk about ionisation rates.

Figure 2: It would be helpful to provide some indication of the variability, for example by a shaded band around the median.

Figure 6: I don’t understand where soil temperatures fit in here, is this a typo?

Figure 7: R2 needs a superscript on the 2

Figure 11: This plot tries to convey too much information. Would it be better to present all the data together (e.g. as a median), or pick one representative year and add the rest to the supplementary information?

Figure 13: A linear fit may not be appropriate here given the theory – please revise

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