We would like to deliver our gratitude to referee #1 for the detailed and thorough review of our work. Below are our responses to the critical points presented by the referee. For each comment of the referee, our reply consists of a general response and an indication of changes in the manuscript following the guidelines.

1. 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.

   Thank you for the comment! We broadened our introduction concerning the historical works on air ion study and instrumentations used for characterising air ions. This part of the manuscript is elaborated as follows

   ‘Air ions were historically concerned in the discipline of atmospheric electricity (Israël, 1970), because their flow in the electric field of the earth-atmosphere system serves as the measureable conduction current in the atmosphere (Wilson, 1921;Tinsley, 2008;Harrison and Carslaw, 2003). The interest in atmospheric electricity could be traced back to the early 18th century when thunderstorms were suggested to be electrical phenomena (Herbert, 1997). However, only when Benjamin Franklin proposed the idea to draw electricity down from lightning in 1752, this theory was confirmed and the study of atmospheric electricity became popular (Herbert, 1997;Tinsley, 2008). Early efforts in this field were substantially invested into understanding lightning and electrification of clouds (e.g. (Canton, 1753a, b;Franklin, 1751)), even though there were reports on observations of atmospheric electricity under fair weather conditions (Canton, 1753a;Read, 1792;Bennett and Harrison, 2007). But why the air is conductive could not be explained. Meanwhile, Charles-Augustin de Coulomb observed gradual discharge of a well-insulated electroscope around 1785 and he attributed his observation to the contact of suspending particles present in the air (Walter, 2012;Angelis, 2014). This phenomenon was reproduced by Michael Faraday half a century later in 1835 (Angelis, 2014). Thanks to the further improvement of the electroscope by William Thomson and Lord Kelvin (Angelis, 2014;Flagan, 1998), Crookes (1878) could find that the discharge

rate of an electroscope decreases with a decreasing air pressure, suggesting that it is the air inside the instrument that manipulates the discharge. Yet, the reasoning remained undisclosed, until the discovery of radioactivity by Wilhelm Röntgen, Henri Becquerel and Marie and Peirre Curie enabled Julius Elster and Hans Geitel from Germany and Charles Thomson Rees Wilson from Scotland to relate the spontaneous discharge of the electroscope to ionisation of the air by radioactive sources (Angelis, 2014;Carlson and De Angelis, 2011). Therefrom, the importance of air ions in the atmosphere emerged. Contemporaneously, the interest of Joseph John Thomson, director of the Cavendish Laboratory, in the charge carriers produced by ionising radiation motivated the development of instrumentations for measuring electrical charges in gases, leading to various valuable outcomes, e.g. the cloud chamber designed by C. T. R. Wilson, as well as techniques for measuring ion mobility by Ernest Rutherford and John Zeleny and for studying gaseous ion diffusion by John Sealy Townsend (Flagan, 1998;Robotti, 2006). These works laid the theoretical and instrumental foundation for later aerosol studies. The experimental results from C. T. R. Wilson’s cloud chamber measurements in 1895 and 1899 on the influence of ionising radiation on the formation of cloud droplets brought interests of air ions into the atmospheric aerosol community. Inspired by these early works, advancements in atmospheric aerosol studies progressed both instrumentally and theoretically over the century (e.g. Tammet 1970; Nolan, 1924; Hogg, 1939; Hewitt, 1957; Tammet, 2006; Reischl, 1991; Rosell-Llompart and Fernández de la Mora, 1993; Aplin and Harrison, 2000; Mason and McDaniel, 1988; Millikan, 1923; Hinds, 1999; Tammet, 1995).

Devices employed in air ion studies comprise different types of aspiration condensers, ion mobility spectrometers (IMS) and mass spectrometers (Hirsikko et al., 2011;Tammet, 1970;Laskin et al., 2012;Cumeras et al., 2015). Notably, modern key instruments for field observations of air ions are mainly aspiration contender-based devices and mass spectrometers, such as the Gerdien counter - an integral aspiration condenser (Gerdien, 1905;Aplin and Harrison, 2000;Vojtek et al., 2006), ion spectrometers designed by Airel Ltd – single or multiple channel aspiration condensers (Tammet, 2011, 2006;Mirme et al., 2007;Manninen et al., 2009;Kulmala et al., 2016) and the atmospheric pressure interface time-of-flight mass spectrometer (APi-ToF) (Junninen et al., 2010). While aspiration condensers provide information on the concentration and mobility of charge carriers, mass spectrometers reveal mainly the chemical properties of them. The IMS, however, has a limited

application in studying ambient ions, due to difficulties in spectrum interpretation (Hirsikko et al., 2011). The purpose of these instrumentations is not only limited to air ion or air conductivity observations, but also for nano-material synthesis (Kruis et al., 1998) and for the improvement of the fundamental understanding on the relationship between mobility, mass and size (Ku and Fernández de la Mora, 2009).'

2. 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.

   We appreciate the reviewer' s comment. We refocused the motivation of our paper to better match the work we presented. It is as follows

   ' … Such deficiencies prompt the motivation of this work to examine how variations in ionising radiation are reflected in observed air ions based on ambient measurement. The aims of this study are 1) to identify the key factors responsible for the variability in ionising radiation and in observed air ion concentrations, 2) to reveal the linkage of observed air ions to variations in ionising radiation and 3) to provide an in-depth analysis on the effects of ionising radiation on air ion formation. We will first introduce factors that cause the seasonal and diurnal variability in ionising radiation and air ions, and then exposit the connection of observed air ions to variations in ionising radiation and the influence of different atmospheric conditions on this relationship. '

3. 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).

   Thank you for the comments! We decided to change our title to 'How do air ions reflect variations in ionising radiation in the lower atmosphere in a boreal forest?' to better stand for the content of our paper.

4. 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. How- ever, 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 consistency with other work. They could add a caveat mentioning energy loss to excitation if this concerns them.

Thank you for the comments! Indeed, we introduced a new term, ionising capacity. We intended to distinguish between the actual ionisation rate and the theoretical maximum ionisation rate based on calculation.

The ionisation rate is a measure of the rate of successful production of ion pairs by ionisation, which is self-evident. However, the usage of this term has been extended beyond its scope vaguely sometimes. There could be energy loss due to excitation. It seems that the reviewer has gotten this point. However, whether this energy expenditure occupies a minor share or not is something that remains unknown and needs further quantification.

Another thing that needs to be aware of is that the ionisation rate should be a universal quantity determined by natural processes, which however, cannot be measured directly yet. Our determination of the rate is based on ionising radiation measurements and the assumption that 34 eV is needed for producing one ion pair. There are uncertainties associated with measurements. Besides, how well can the 34 eV characterise the natural ionisation process at different heights of the atmosphere and at different spatial locations requires further investigation.

For the reasons stated above, we would like to keep the term, ionising

capacity, and to encourage the research community to be cautious with the term usage in describing ‘the rate of producing air ions’. We added a better description of the ionising capacity to justify its connection to and difference from the ionisation rate in the revised manuscript.

‘The ionising capacity can be viewed as a measure of the theoretical maximum ionisation rate, which however, may not well capture the true ionisation rate due to uncertainties in ionising radiation measurements, possible energy dissipation of ionising radiation in excitation and the invalidity concern associated with the use of 34 eV per production of an ion pair at near ground level in our calculation’

5. 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.

   Thank you for the comment! It is true that there exist definitions on clusters ions, small ions and large ions from historical works. For example, H. Israël (1970) wrote in the famous ‘Atmospheric electricity’ book that ‘An electron, detached from a molecular or atomic bond, cannot exist freely in air at normal temperature and pressure, but readily attaches itself to a neutral atom or molecule. However, even these molecular or atomic ions cannot remain stable in atmospheric air under normal conditions, and consequently surround themselves with a number of neutral molecules and form clusters of approximately 10-30 molecules. Only then do they reach a certain stability in the form of so-called "small ions."’

   The information in our Fig. 1 does not contradict with what H. Israël explained, rather we followed his description. H. Israël considered that a ‘small ion’ is a charged cluster that has reached certain stability threshold and he was aware of the existence of atomic and molecular ions, though they are not stable in the atmosphere to be readily clustered.

   Theoretical studies have shown that only after reaching the critical size, clusters are stable enough thermodynamically for growing bigger, which is

likely the ‘Israël threshold’ for charged clusters to be called small ions. The critical size was found to be around 1.5±0.3 nm for atmospheric nucleation (Kulmala et al., 2013). In our study, we concentrated on ions in the size range between 0.8 and 1.7 nm, which we called the cluster size range. We discussed in the paper how ions in this size range are possibly formed from smaller ions (primary ions and molecular ions) and the possible underlying dynamic processes. For this purpose, it is necessary to introduce what are primary ions, molecular ions and cluster ions, and to make a distinction between them.

The boundary between primary ions and molecular ions does not exist explicitly. It is the origin of the ions that tells what ions they are. If a charged single molecule is formed from ionisation directly, it is also a primary ion. As the reviewer wrote, primary ions N2+ and O2+ can also be classified as molecular ions intrinsically. However, if the molecule gets its charge from other charged species in the atmosphere, it is only a molecular ion, but not a primary ion. We would like to show Fig. 1 instead of only explaining in plain text to convey the message clearly to our readers that the scope of primary ions and that of molecular ions do overlap in certain aspects considering the origin of the charge. As for the stability and lifetime of molecular ions, certain big molecular ions may exist in the atmosphere, which have been demonstrated by the recent application of APi-ToF MS in field measurement (Ehn et al., 2010).

We are also aware of the possible presence of molecular ions in our cluster size range as stated in the manuscript. There is no such a clear size boundary to distinguish between molecular ions and cluster ions. To avoid misleading of readers by what we called ‘cluster ions’ in our analysis, we considered it important to demonstrate what truly distinguishes a molecular ion from a cluster ion. And this purpose is served by Fig. 1.

By taking the comment from the other reviewer into consideration, however, we added underlying processes in cluster ion formation into Fig. 1. Below is the revised figure.

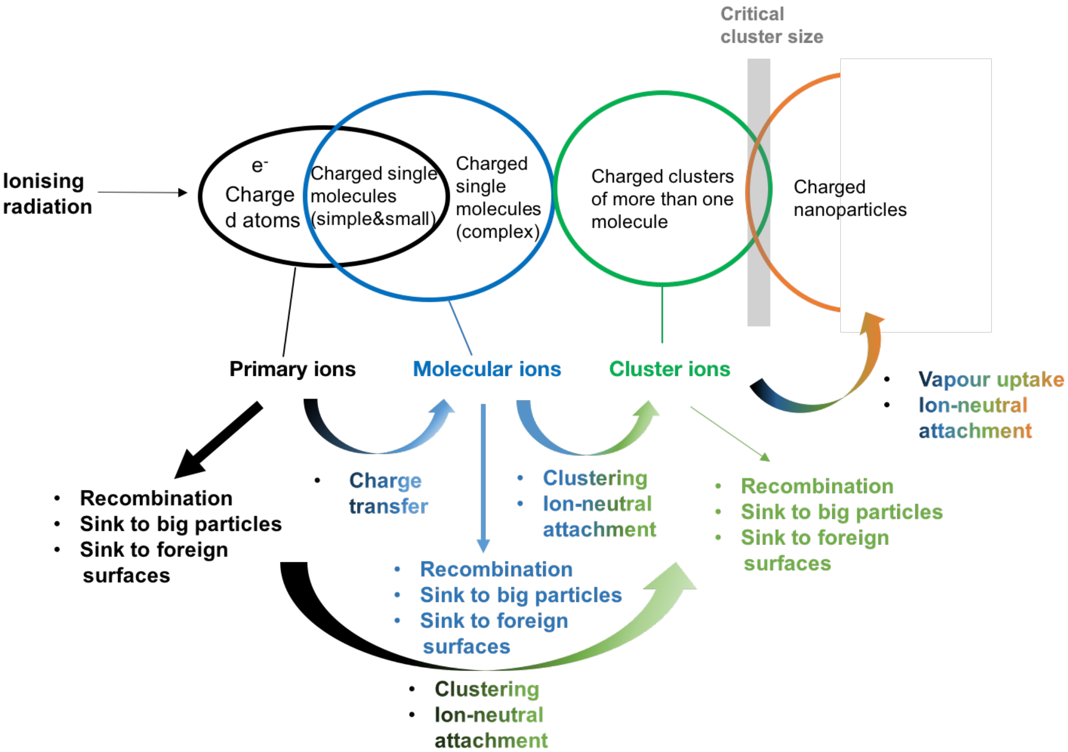


Figure 1. A schematic demonstration of the relationship between primary ions, molecular ions and cluster ions, as well as processes governing their formation and loss.

6. 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.

   We appreciate the reviewer’ s comment! The flattening out of cluster ion concentrations at higher ionising capacities in Fig. 13 is related to the fact that recombination becomes the determining process. This was said in the manuscript. However, this feature cannot be explained by the ion balance equation at the recombination limit alone. As given by Harrison and Carslaw (2003), the steady state ion concentration at the recombination limit is n = sqrt(q/α), where q is the ionising capacity in our case and α is the

recombination coefficient being about 1.6e-6 cm3s-1. However, as can be seen from *n* = sqrt(*q*/*α*), *n* will increase monotonically with an increasing *q*, but does not level out. Even with the addition of the ion-aerosol interaction term *βNn*, which can also be written as the product of the condensational sink and ion concentration – CS*n*, then *n* = sqrt(*q*/*α*+(CS/2/*α*)^2)-CS/2/*α*, the ion concentration still does not flatten out. This flattening-out could result from the synergic effect of different complicated processes, especially because we are dealing with ambient data. However, one of the reasons may be related to the availability of vapour sources or neutral clusters. But this speculation needs to be verified with further knowledge on the roles of different key chemical species participating in cluster ion and neutral cluster formation and diurnal variations in their atmospheric concentrations, which however are beyond the scope of this work.

We would like to thank the reviewer for suggesting the interesting work of Aplin and Harrison (2000). We suppose that the reviewer meant for Fig 4 in Aplin and Harrison (2000), where a relationship can be seen between the ionising radiation and air ion concentration. However, the information conveyed by this figure may not be comparable with our observation. In their work, they observed a continuous increase in the ion concentration, even if the ionising radiation showed a levelling-out. But in our case, it is the opposite that we saw suppressed cluster ion production along with a continuous increase ionising capacity.

7. 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?

We improved the conclusions according to the reviewer' s comments. We addressed the importance of our results and the reason why instruments for sub-0.8 nm ion measurement are needed. The revised conclusions are as follows

'In this work, diurnal and seasonal cycles in ionising radiation were presented and key influencing factors responsible for these features were overviewed in order to investigate how observed air ions respond to these

variations and to improve our understanding on air ion formation. To assist the analysis, a term, ionising capacity, was introduced to capture patterns in ionising radiation. The ionising capacity was determined theoretically as the potential maximum production rate of ion pairs in the atmosphere by ionising radiation, based on the assumption that an ion pair is produced upon every 34 eV energy dissipation of ionising radiation. The data used in this study were collected from ambient measurements during 2003-2006 from a boreal forest site in southern Finland. In our analysis, the accounted ionising radiation is composed of energy from alpha and beta decays of Rn-222 and accompanying gamma radiation, energy contained in the gamma radiation from terrestrial origins and gamma radiation released from the interactions between cosmic rays and air molecules. Variations in the ionising capacity were primarily related to boundary layer development, soil conditions, snow accumulation and the origin of air masses.

Although ionising radiation is known to be responsible for air ion production, patterns in the measured air ion concentration in the cluster size range (0.8-1.7 nm) did not exhibit a highly comparability to those in the ionising capacity, due to modifications of air ion properties exerted by different dynamical processes and chemical reactions during the evolution of charges in the atmosphere. Nevertheless, the connection of air ions to ionising radiation was seen for air ions in the lowest detected size band (0.8-1 nm) of the cluster size range (0.8-1.7 nm). The evolvement of these 0.8-1 nm ions with time to larger sizes in the cluster size band was also identified, affirming the primary role of ionising radiation in the production of air ions in the lower atmosphere. Yet, atmospheric conditions, such as temperature, humidity and pre-existing aerosol particles, brought complications into this relationship. By carefully constraining data to conditions of a similar meteorology, seasonality, diurnality and amount of background aerosol particles, a strong dependency of total cluster ion concentrations on the ionising capacity was identified on new particle formation (NPF) days. However, the linkage was not visible on non-event days. These observations may suggest that charges, after being born, underwent different processes on NPF days and non-event days and possibly indicate also that the transformation of newly formed charges to cluster ions occurred faster on NPF days than on non-event days. These results could help to advance our understanding on the role of ions in atmospheric new particle formation.

However, to obtain further insights into the fate of charges created by

ionising radiation in the atmosphere, i.e. ion balance, and into the role of air ions in the atmospheric new particle formation process, it is crucial to understand the transformation process of electric charges into detectable air ions. For this purpose, knowledge on the number size distribution of air ions smaller than 0.8 nm is of necessity. Additionally, theoretical understanding on the formation mechanisms of cluster ions from molecular ions needs to be deepened. Conjointly, also advancing instrumental development for the detection of sub-0.8 nm ions could be worthy of being brought onto the agenda.'

8. The results on new particle formation are novel and very interesting, but can you discuss their implications?

Thank you for the comment! The better dependency of cluster ion concentrations on the ionising capacity observed on NPF event days than that on non-event days may imply that charges on NPF event days did not undergo significant transformations after being produced via ionisation by ionising radiation to become cluster ions. Therefore, it is possible to trace the linkage between the cluster ion concentration and ionising capacity statistically. However, this connection was very poorly preserved on non-event days, possibly because after being created, charges went through a too complicated series of modifications before reaching the cluster sizes.

In other words, these results could provide us some hints on what charges have experienced in the atmosphere after formation and possibly also on the relative duration of the processes faced by newly formed charges before they become cluster ions. It could be that charges became cluster ions in a shorter time scale on NPF event days than on non-event days, which allows those cluster ions on NPF event days to retain some features in connection to ionising radiation.

These results could help advance our understanding on the role of ions in atmospheric new particle formation. Apart from this, these results could also be beneficial in model development aiming for insights into the new particle formation process.

These implications are added in the revised manuscript as follows

In the discussion of Fig. 13

' In addition, the better dependency of the cluster ion concentration on the ionising capacity observed on NPF event days than on non-event days may also imply that charges, after being produced via ionisation by ionising radiation, on NPF event days did not undergo significant transformations to become cluster ions. Therefore, the linkage between the cluster ion concentration and ionising capacity was traceable statistically. However, this connection was very poorly preserved on non-event days, possibly because after being created, charges went through a too complicated series of modifications before reaching the cluster sizes. These observations may also provide some indirect measure of the relative duration of dynamic processes faced by newly formed charges before they become cluster ions under different atmospheric conditions: it might suggest that charges became cluster ions in a shorter time scale on NPF event days than on non-event days and as a consequence, the cluster ions on NPF event days are enabled to retain some features in connection to ionising radiation.'

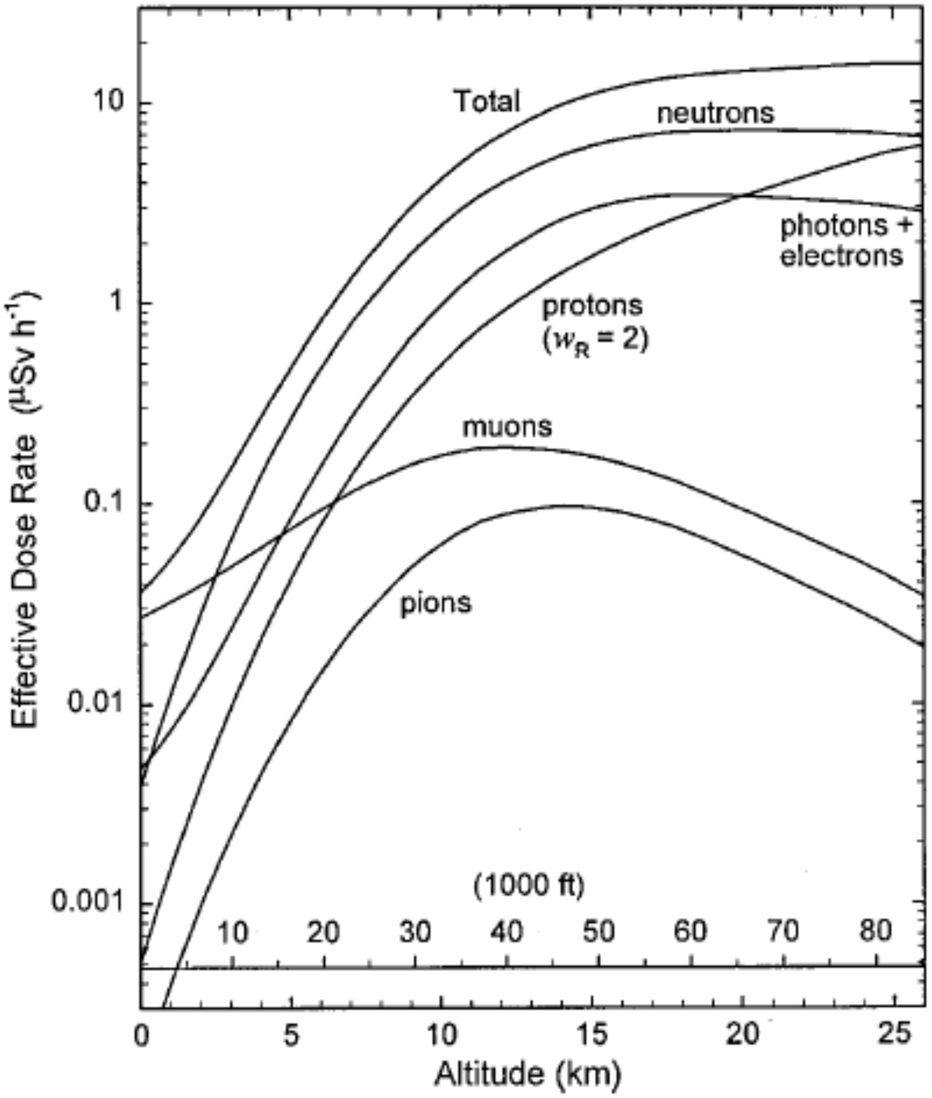
In the conclusion

'These observations may suggest that charges, after being born, underwent different processes on NPF days and non-event days and possibly indicate also that the transformation of newly formed charges to cluster ions occurred faster on NPF days than on non-event days. These results could help to advance our understanding on the role of ions in atmospheric new particle formation.'

9. 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.

We agree with the reviewer that 2 or 3 /cc/s and the text about

recombination are highly speculative. It can well fall (and most probably does fall) into the uncertainty budget. The discussion is revised.



As shown in the figure above (Goldhagen, 2000), the photon and electron contributions from cosmic ray at Hyytiälä altitude is about 10 % of the muon contribution, the same with neutrons, i.e. it is the muons that count.

The recorded total count rates by our gamma spectrometer were converted into dose rates in the air by a calibration factor obtained from an instrumental comparison to a pressurised ionisation chamber. The ionization chamber basically measured the ionization contributed by all ionizing radiation that can penetrate through the chamber wall and interact with the filled gas medium. Therefore, the gamma spectrometer could capture muons.

But for simplicity, we termed the ionising capacity derived from the dose rates as the gamma ionising capacity. This is clarified in the revised manuscript.

However, a portion of high-energy muons may not be detected well by our counting system. This is either because they do not interact enough with the detector material, or because their light production causes subsequent electrical pulses exceeding the dynamic range of the instrumentation.

Descriptions and revisions on these aspects are as follows

In introduction

‘In the case of cosmic radiation near the ground, most of the ionisation of air is due to muons, with minor contributions from neutrons, photons and electrons (Goldhagen, 2000).’

In section 2.1.1

‘The gamma spectrometer is a scintillation-type detector using a 76 mm × 76 mm NaI(Tl) as the detection medium (Laakso et al., 2004;Hirsikko et al., 2007), which is kept at the height of 1.5 m above the ground. The device employs a ratemeter to register every single pulse with a height exceeding the background level and generates the total count rate with a time resolution of 10 min. In addition, pulse height spectra over the energy range of 100-3000 keV are recorded with a separate multichannel analyser. The total gain of the detecting system is kept constant via digital spectrum stabilization using the potassium-40 gamma peak (1460 keV) as the reference. For the determination of the ionising capacity in this work, the total count rate data were used instead of the spectral information. The recorded total count rates were converted into dose rates in the air (µSv/h) by a calibration factor obtained from an instrumental comparison to a pressurised ionisation chamber. Thus, the obtained dose rates take into account ionisation by both gamma radiation and cosmic ray muons. However, a portion of high-energy cosmic ray muons may not be well detected by our counting system, possibly due to their weak interaction with the detector material or their light production leading to electrical pulses exceeding the dynamic range of the instrumentation.’

In section 2.2

‘Therefore, the data obtained by the gamma spectrometer could be considered to represent the total gamma radiation, including the terrestrial fraction, the cosmic fraction and the fraction from radon decay. In addition, the gamma spectrometer also accounts for ionising energy from muons.’

‘For conciseness and clarity, … the ionising capacity from total dose rates recorded by the gamma spectrometer as the gamma ionising capacity (Qγ).’

In section 3.2.2

‘According to Paatero et al. (2005), the constant term in the exponential fitting to the measured snow depth data, being about 3 cm^{-3} s^{-1}, could represent an approximation of the contribution by cosmic radiation to the ionising capacity. A rate of 2 cm^{-3} s^{-1} has been generally accepted as the ionisation rate of cosmic radiation for the production of small ions at sea level (Hensen and Hage, 1994). The cosmic contribution to the ionising capacity determined from our measured total dose rates with the exponential fittings was close to this value. The discrepancy comes likely from uncertainties involved in the mathematical fitting and measurement as well as possible spatial variations in the cosmic radiation ionisation rate and solar activity influence.’

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

    Thank you for pointing this out! The sentence has been rephrased as

    ‘Due to the atmospheric abundance of nitrogen (N2) and oxygen (O2), their derivatives are the initial carriers of electric charges generated from the ionisation process.’

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

    Thank you for the comment! We revised this paragraph as follows

    ‘Although it is known that ionising radiation creates ion pairs via ionisation in the atmosphere (Flagan, 1998;Harrison and Carslaw, 2003;Israël, 1970),

except for a few attempts (Hirsikko et al., 2007;Laakso et al., 2004), minor efforts have been invested in understanding the connection between ionising radiation and observed air ions in the lower atmosphere. Moreover, there is a lack of quantification on the underlying processes. Such deficiencies prompt the motivation of this work to examine how variations in ionising radiation are reflected in observed air ions based on ambient measurement. The aims of this study are 1) to identify the key factors responsible for the variability in ionising radiation and in observed air ion concentrations, 2) to reveal the linkage of observed air ions to the variations in ionising radiation and 3) to provide an in-depth analysis on the effects of ionising radiation on air ion formation. We will first introduce factors that cause the seasonal and diurnal variability in ionising radiation and air ions, and then exposit the connection of observed air ions to variations in ionising radiation and the influence of different atmospheric conditions on this relationship. To assist our analysis, we will theoretically determine the potential maximum production rate of ion pairs by ionising radiation, based on our ionising radiation measurements and an assumed average energy expenditure of 34 eV for creating an ion pair, which is termed as the ionising capacity. The ionising capacity can be viewed as a measure of the theoretical maximum ionisation rate, which however, may not well capture the true ionisation rate due to uncertainties in ionising radiation measurements, possible energy dissipation of ionising radiation in excitation and the invalidity concern associated with the use of 34 eV per production of an ion pair at near ground level in our calculation.'

12. P5 L15 "descripted" -> "described"

    Thank you for spotting out this typo! It is corrected in the revised manuscript.

13. P6 L17 All ion spectrometers are defined by their upper and lower mobility limits, so this sentence is meaningless.

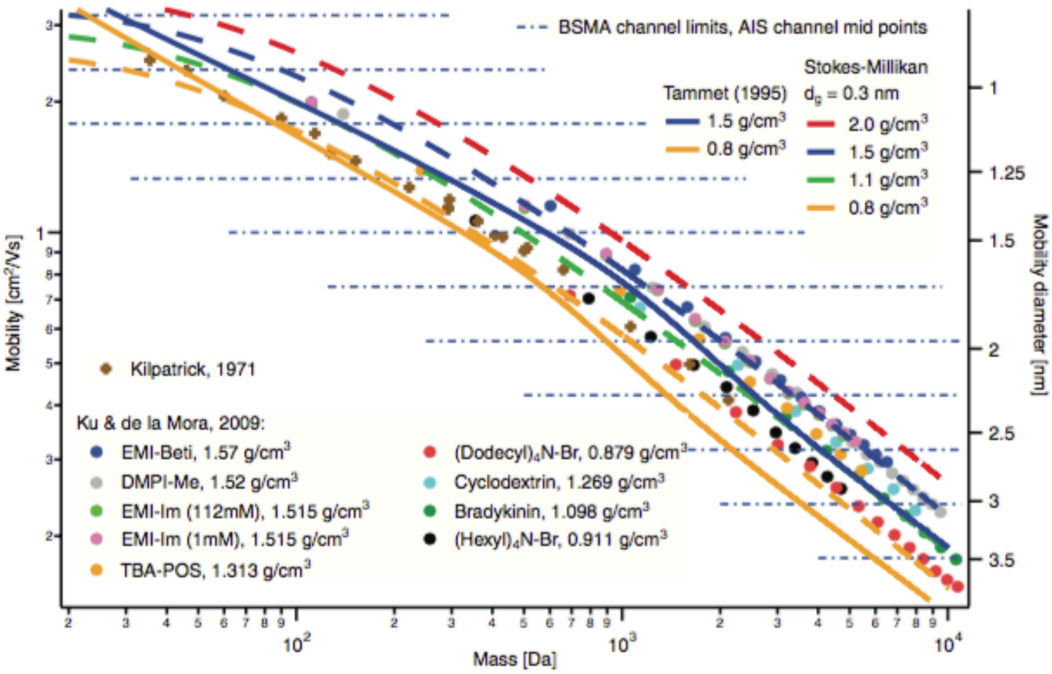
    The reviewer is right. The sentence is removed from the revised manuscript.

14. Why is the upper mobility limit 3.2 cm-2V-1s-1?

    The upper mobility limit of the instrument is determined by its configurational design, hardware limitations and transmission optimisation.

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

    Stokes-Millikan equation is a widely-accepted model for describing the relationship between electric mobility and diameter and the mobility diameter is defined based on this model. However, it has a deficiency in characterising the connection of diameter to mobility in the microscopic limit. Scientists, such as Tammet (1995) and Li and Wang (2003), have sourced the first approximation of Chapman-Enskog theory for extending the model to free molecular regime. However, they used different function forms of potentials to treat the interactions between a particle and the gas medium. In addition, also the choice of the coefficients in calculating the Cunningham slip correction factor could bring bias in the converted diameter from electric mobility (Kim et al., 2005). Moreover, the relationship could also be affected by variations in temperatures and densities of particles and the gas medium. Ehn et al. (2011) made a comparison between mobility, mobility diameter and mass diameter using different models and particle densities, as shown in the figure below. In general, the relationship between electric mobility and mass diameter agree well with those experimentally determined values. Although in this comparison, Ehn et al. did not take into account the mass correction factor proposed by Tammet (1996) in converting the electric mobility into mobility diameter, by simply using the relationship between mass diameter (dm) and mobility diameter (dp), dp = dm+0.3 nm suggested by Ku and Fernández de la Mora (2009), some clues on the possible changes in mobility diameters can be deduced.

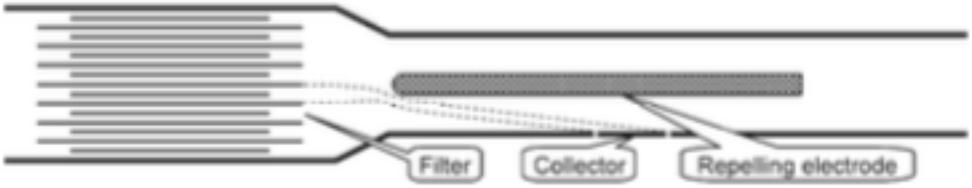


16. P7 L1-2 please be consistent with units.

It was our carelessness. The form ‘l/min’ is used in all places in the revised manuscript.

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

The instrument was designed by Hannes Tammet from the University of Tartu. A detailed description of the device can be found in Tammet (2006). Briefly, the device consists of two aspiration type plate condensers and each one is divided into 8 sections by plate electrofilters. Ions are only allowed to enter the condenser through the two central sections. However, since only the half of the condenser is used for ion collection, as shown in the figure below (taken from Tammet (2006)), therefore only one eighth of the air sucked in is considered as sample flow. The other half of the condenser that is not in use for ion collection serves the insulation purpose and allows the optimisation in transfer function determination.



18. P7 L13 “programme” -> “program”

    Thank you for pointing out this! It is corrected now.

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

    The definition of condensation sink (CS) is added into the revised manuscript, as follows

    ‘CS accounts for the loss rate of vapours due to condensational uptake by aerosol particles in the atmosphere (Kulmala et al., 2001).’

20. 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?

    Some of measurement details can be found from published research articles and some from the station’ s home page. We shortened this section in the revised manuscript as the referee suggested as follows

    ‘The snow cover depth was measured manually on a weekly basis on seven different locations at the SMEAR II station. Measurements on soil temperature and soil volumetric water content were described by Pumpanen et al. (2003) and Ilvesniemi et al. (2010). Only the organic horizon data (5 cm depth, above the mineral layer (Pumpanen et al., 2003)) were used in this work. The organic horizon is in direct contact with the atmosphere, the condition of which exerts the primary influence on radon exhalation. The ambient relative humidity and air temperature data were taken from the mast measurement at 16 m and 4.2 m, respectively. More detailed description of the mast instrumentation can be found from the home page of the measurement site (http://www.atm.helsinki.fi/SMEAR/index.php/smear-ii/measurements).’

21. P8 L22 Notwithstanding the comments above, should these units include a “per unit volume” ?

    The referee is right. We corrected the definition of the ionising capacity in the revised manuscript as follows

‘The ionising capacity (*Q*) is defined as the potential amount of ion pairs produced per unit time in a unit volume upon ionisation by ionising radiation in the atmosphere.’

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

    We used the measured ambient temperature and pressure in converting the dose rate to the ionising capacity according to eq. 1. Therefore, strictly speaking, there are small differences between the variations of ionsing radiation and the derived ionising capacity. For this reason, we would like to keep this sentence in the manuscript.

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

    Thank you for the comment! As we have explained in the introduction of this research article, radon is the decay product of radium. Radium is much more abundant in rocks and soil than in water. We added such information in the revised manuscript:

    ‘…marine air masses from Arctic and north Atlantic oceans, which favour NPF (Nilsson et al., 2001), typically have a low radon content (Chen et al., 2016). Radon comes from the radioactive decay of radium. Since marine surface water has a significantly lower radium content than the continental surface layer (Wilkening and Clements, 1975), only minor amount of radon can be collected by air masses traversing over the ocean.’

24. P12 L10 “exam” -> “examine”

    Thank you pointing this out! The correction is made in the revised manuscript.

25. 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?

    The statement is added in the revised manuscript as follows

‘In comparison with the radon ionising capacity accounting for alpha and beta emissions of radon decay, the gamma ionising capacity exhibits a simpler pattern.’

26. P14 L8 would be better to say “growth season”

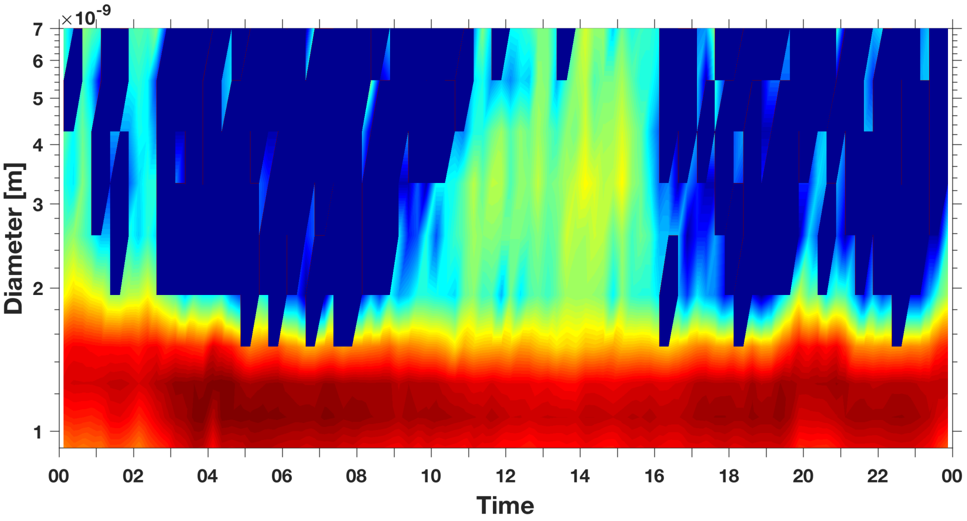
Thank you for the suggestion! We changed it to ‘growth season’ in the revised version.

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

Our apology for the unclearness. We reformulated the sentence as follows in the revised manuscript

‘In the latter case, certain removal processes of ions from the cluster size range are inhibited, which can be either the growth of cluster ions to sizes bigger than 1.7 nm or the loss of cluster ions by the attachment to bigger particles.’

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



The above figure shows a spectrum measured by the BSMA. It can be seen that on average, most ions are smaller than about 1.7 nm. Bigger ions are seen during the new particle formation event around noon. The critical cluster size was found to be 1.5±0.3 nm for atmospheric nucleation (Kulmala et al., 2013), after which further growth of a cluster in size is energetically favoured (Vehkamäki, 2006). Ions with a diameter larger than the critical

cluster size can therefore be considered as nanoparticles. In view of BSMA spectra, 1.7 nm seems to be a good approximation of the critical cluster size as there is the constant presence of ions smaller than 1.7 nm.

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

    Thank you for the comment! However, since we suggested that both changes in atmospheric conditions and vapour sources could result in the dissimilar autumn and spring patterns in the cluster ion concentration, we would like to briefly introduce what kind of differences can be expected from vapour sources. For this reason, we consider information presented on these lines are of relevance.

30. 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.

    Thank you for the comment! We elaborated the discussion and added citations to the classical literature.

    'While the positive polarity dominated the overall cluster ion concentration, more negative ions were seen in the first two sub-size ranges (0.8-1 nm and 1-1.2 nm). The former results from the electrode effect of the negatively charged earth surface, which repels negative ions in its vicinity; and it is a well-known phenomenon to the atmospheric electricity community (Israël, 1970;Wilson, 1921;Tinsley, 2008;Harrison and Carslaw, 2003). The latter agrees with observations that generally negative ions possess higher mean mobility than positive ions (Israël, 1970;Dhanorkar and Kamra, 1992;Hõrrak, 2001), i.e. on average, negative ions are of smaller sizes than positive ions.'

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

    Ionising radiation produces ion pairs by ionisation. These ion pairs are known as primary ions, which contain electrons and some small molecular ions (Fig. 1 in the manuscript). These primary ions will undergo a series of physical collisions and chemical reactions with other constituents in the air, leading to the formation of more complex molecular ions, cluster ions, larger ions and neutral species. The similar variations seen in the 0.8-1 nm ion

concentration to those in the ionising capacity could imply that the majority in this size range are probably molecular ions, which have not been heavily influenced by dynamical processes of cluster formation and therefore retained some features of primary ions.

‘The 0.8-1 nm ion concentration showed features similar to those in the ionising capacity (Figs. 9 and 3), being high in the morning and low in the afternoon. This observation possibly indicates that the dominant population in the size range of 0.8-1 nm are molecular ions, which have not been heavily influenced by dynamical processes of cluster formation and therefore retained the characteristics of primary ions.’

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

We agree with the referee that this statement of ours is speculative. However, qualification of it would require massive experimental work under carefully controlled conditions, which we consider are beyond the scope of this manuscript. We modified the text so that the message that this statement is a speculation and further work is needed for the verification is conveyed.

‘This observation might be related to the proton affinity of water molecules (H_2O), which assigns H_2O the ability to bind positive charges. The formed cations may constitute a portion of hydronium ions (H_3O^+), which are too small to be detected by the BSMA, resulting in the flattening-out of the 0.8-1 nm ion concentration. Yet, further experimental investigations are needed for the verification of this mechanism and quantification of its significance.’

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

We added explanation for the relation of CS to 0.8-1 nm ion concentrations in the revised manuscript as follows

‘As the CS can provide a measure of the condensational loss rate onto aerosol particles, the relatively high CS, as seen in Figs. S2 e and f, could be an additional reason for the lower 0.8-1 nm ion production at high radon ionising capacities.’

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

By 'unaltered gamma ionising capacity level', we intended to describe the effect of temperature on the relationship between the 0.8-1 nm ion concentration and the gamma ionising capacity. As can be seen in Fig. 10 e and f, 0.8-1 nm ion concentrations tend to decrease with an increase in temperature at a given gamma ionising capacity.

The sentence is revised as follows

'When the *T* was below 5 °C, the 0.8-1 nm ion concentration showed a linear relation with the gamma ionising capacity. In addition, more 0.8-1 nm ions also tended to appear at lower temperatures, similar to the temperature effect seen on the relationship between the 0.8-1 nm ion concentration and the radon ionising capacity.'

35. P19 L1 what do you mean by "clustering or simple charge binding?"

Certain vapours could cluster among themselves around a charge to save the charge from recombination. There are also vapours that have strong proton or electron affinity, which can steal charges from the original carriers and bind the charges on themselves. These are explained in the next 6 sentences in the same paragraph following 'This observation may be attributed to the production of certain vapours that compete with the recombination process and other sink mechanisms for electric charges either via clustering or simple charge binding'.

'This observation may be attributed to the production of certain vapours that compete with the recombination process and other sink mechanisms for electric charges either via clustering or simple charge binding. Ionising radiation can potentially free a large number of electric charges, which would 'sacrifice' themselves mostly in recombination, if not otherwise become detectable air ions. The survived electric charges take part in the formation of 0.8-1 nm ions mainly in the form of primary ions and molecular ions. Certain vapours can cluster among themselves around primary ions to form 0.8-1 nm ions. Charge-binding vapours, however, are able to take over charges from primary ions to form molecular ions. This charge transfer process may also be accompanied by chemical reactions between the vapour molecules and primary ions. Some of the molecular ions are possibly born with a size falling in the 0.8-1 nm size range.'

36. P19 L20 “focalised” -> “localised”

    Thank you for the suggestion! It is corrected in the revised manuscript.

37. 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.

    We elaborated the explanation of the labels in the caption and used 25th, 50th and 75th percentiles as notations. The revised table 3 as below

Table 1. Energy in eV m^{-3} s^{-1} deposited in the air by total environmental gamma radiation (E_γ) and by alpha and beta activities from radon-222 decay (E_{Rn}) based on the 2003-2006 data. The gamma and radon ionising capacities (Q_γ and Q_{Rn}) in cm^{-3} s^{-1} were derived from the deposited energy assuming 34 eV for the generation of one ion pair in the air. The statistical features of these data are presented by the five-number summary with two additional measures, mean and standard deviation (STD).

	Min	25th percentile	50th percentile	75th percentile	Max	Mean	STD
E_γ	0.769	242	314	338	544	295	72.3
E_{Rn} (α&β)	0.145	28.6	50.3	79.5	284	58.1	38.8
Q_γ	4.34	6.70	9.09	9.34	11.6	8.14	1.80
Q_{Rn}	0.00427	0.84	1.48	2.34	8.35	1.71	1.14

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

    We appreciate the reviewer’ s comment. We added a shaded band around

moving means to show the variability. The modified figure 2 is as follows

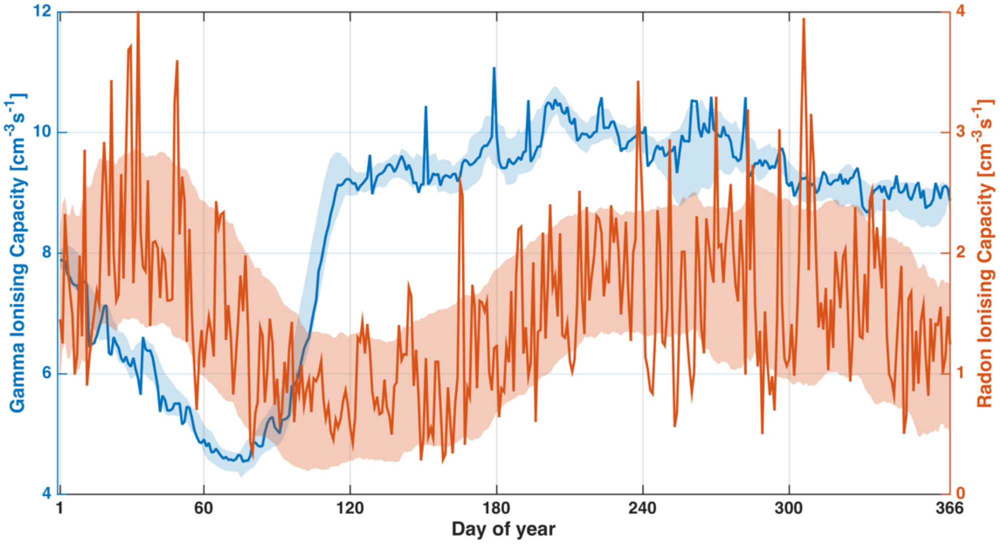
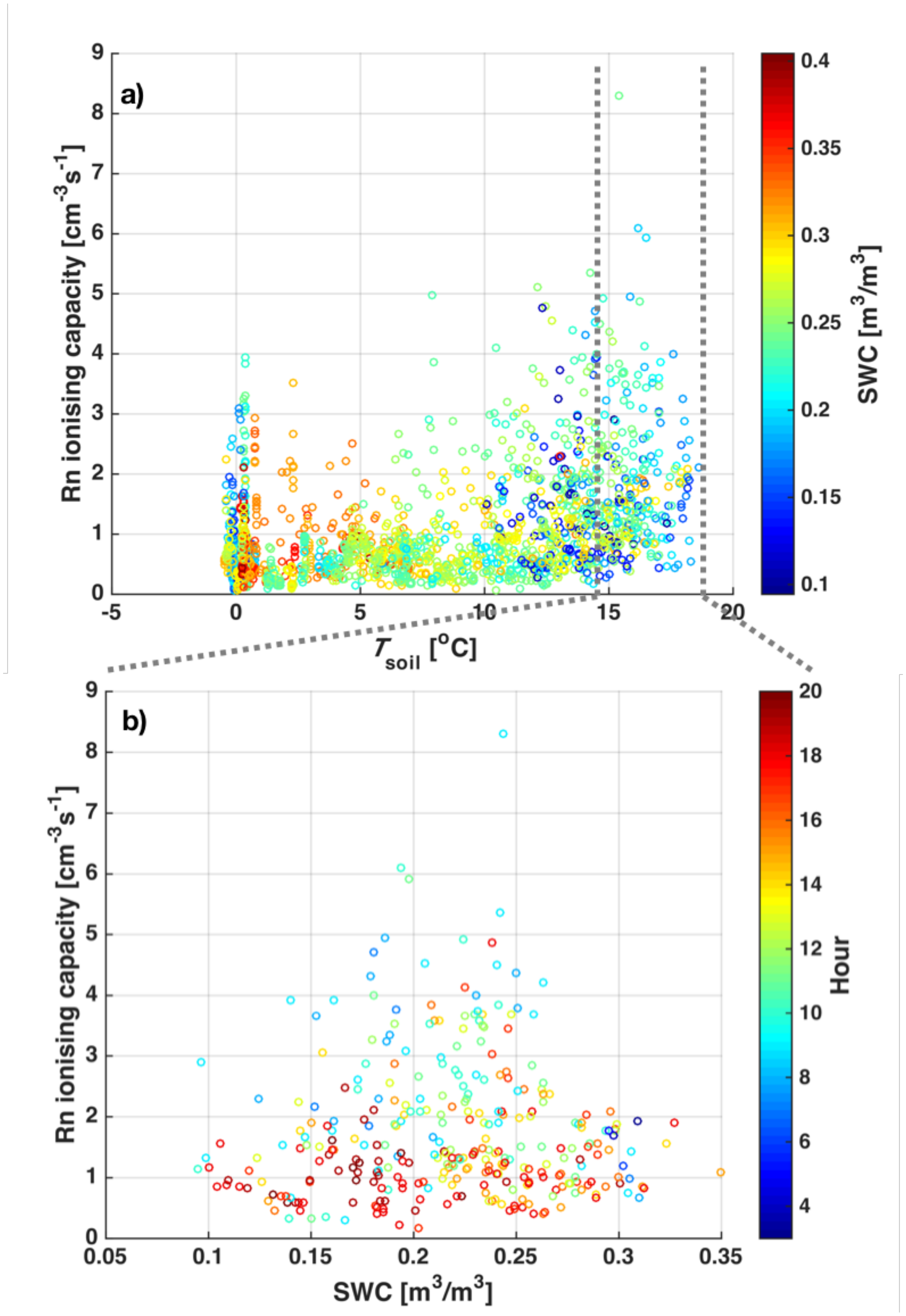


Figure 2. Seasonal patterns of radon and gamma ionising capacities as a function of day-of-year over the years 2003-2006. The radon ionising capacity was determined from the alpha and beta radioactivity associated with radon-222 decay and the gamma ionising capacity from total gamma radiation. The data are presented as daily medians. Shaded bands are dedicated to outline the variabilities, expressed by standard deviations, of the data around their means determined using the moving average method.

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

No, this is not a typo. Soil temperatures are shown on the x-axis in Fig. 6a. We apologise that the dashed line for section selection was laid on top of the x-label in Fig. 6a. It is dragged off the label now in the revised version. And as can be seen in Fig. 6a, the radon ionising capacity tended to increase with the elevated soil temperatures.



40. Figure 7: R2 needs a superscript on the 2

Thank you for spotting this out! It is corrected in the revised manuscript.

41. 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?

    Thank you for the comment! Indeed, the plot contains too much information. Here we intended to deliver the message that we could see similar diurnal behaviours of 0.8-1 nm ions, of both polarities and in different seasons, in response to variations in the ionising capacity and boundary layer dynamics; the latter has a close relation to solar radiation. Since the two polarities show similar features, we will show only one polarity (neg.) in the main article and move the other polarity (pos.) to the supplement.

    The revised Fig. 11 will be as follows

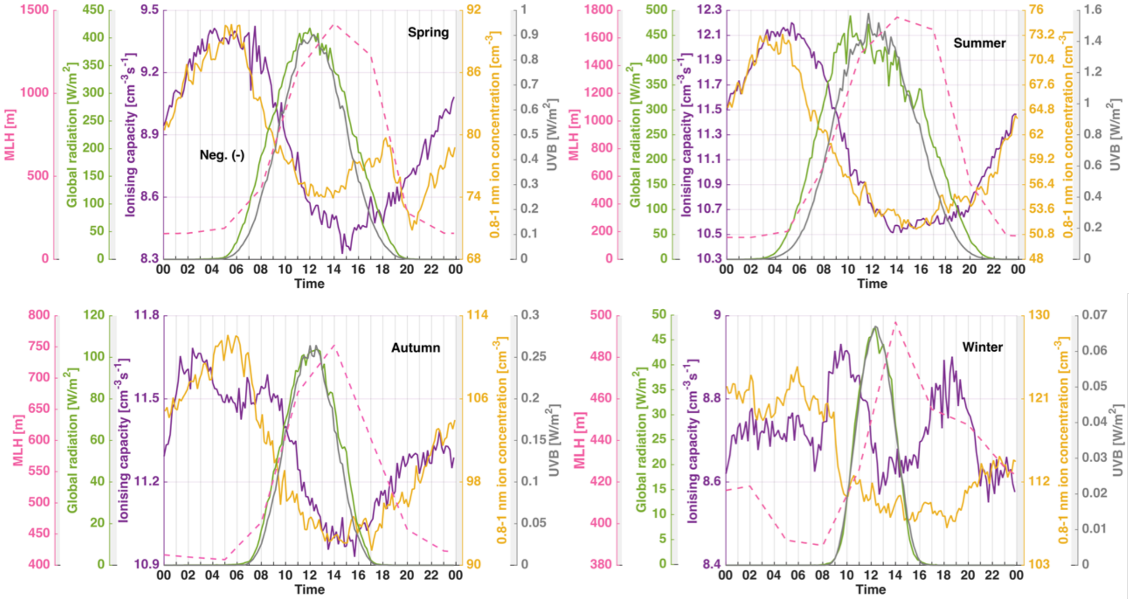


Figure 11. Diurnal patterns in median 0.8-1 nm negative ion concentrations, ionising capacities, global and UVB radiation intensities as well as modelled mixing layer heights (MLH) in different seasons over 2003-2006.

And the rest will be included in Fig. S4 as follows

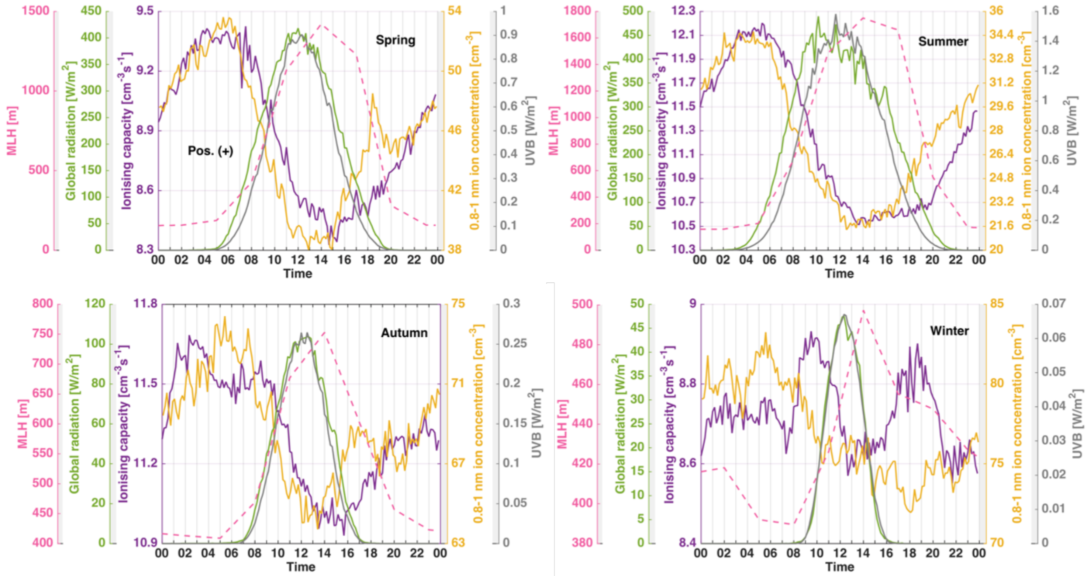


Figure S4. Diurnal patterns in median 0.8-1 nm positive ion concentrations, ionising capacities, global and UVB radiation intensities as well as modelled mixing layer heights (MLH) in different seasons over 2003-2006.

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

The reviewer is right. As it is not a linear relationship between the ionising capacity and air ion concentration, the Pearson’s correlation coefficient measure is not appropriate. We replaced them by Spearman’s rho coefficients in the revised figure. The Spearman’s rank correlation coefficient measures the dependency of the cluster ion concertation on the ionising capacity without requirements on the linearity or knowledge about the distributions of the variables.

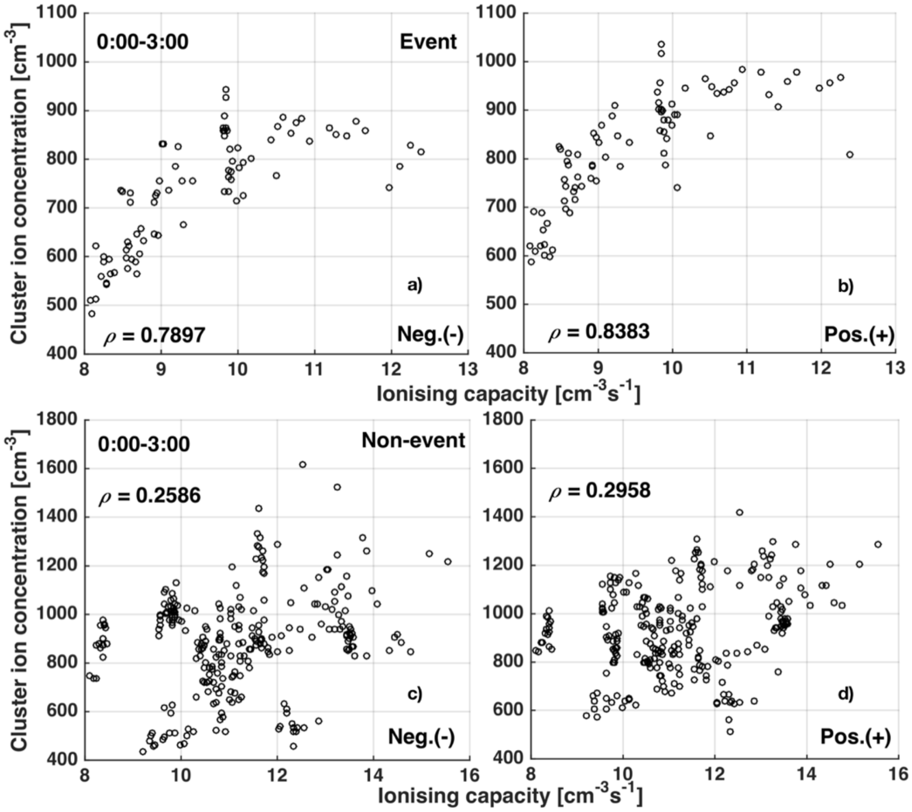


Figure 13. The cluster (0.8-1.7 nm) ion concentration as a function of the ionising capacity (radon ionising capacity + gamma ionising capacity) for selected data in the years 2003-2006, with Spearman's rank correlation coefficient (ρ). The data was constrained on a) & b) event days and c) & d) non-event days in September-December between 0:00 and 3:00 with the wind direction between 280° and 30°, while the condensation sink (CS) was below 0.002 s^{-1}. No dependence of the cluster ion concentration on the CS or hour-of-day was identified. Also the snow season was screened out. The negative polarity is shown in a) and c) and the positive in b) and d), with soil temperatures in the organic layer (-4-0 cm above the mineral soil) shown on the colour scale.

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