We would like to express our appreciation to referee #2 for the careful review of our work. The referee’ s comments are ordered by number in this document. For each critical point presented by the referee, our reply consists of a general response and an indication of changes in the manuscript following the guidelines.

1. 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.

   The reviewer is right. We added information concerning these aspects. More detailed description of these changes can be found under points 4 and 5.

2. 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.

   Thank you for the comment! We added description of other instrumentations for air ion study in the revised manuscript as follows

   ‘Devices employed for 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 de la Mora, 2009).

'

3. 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.

   Thank you for the comment! Ion mobility can indeed be influenced by ambient humidity and there are many articles written on this issue. The humidity effect is a very critical issue, for example, in interpreting spectra from ion mobility spectrometers. However, we dealt with data collected from an aspiration condenser-type instrument and the device measured the number size distribution of ambient ions. Under different humidity (Canton, 1753)conditions, an ion, e.g. a H2SO4-H2O cluster, can be classified into a different size category due to the formation of a new cluster between the original H2SO4-H2O and water molecules, known as the humidity influence. However, this is a reflection of the modification of the ambient size distribution by moisture. The data itself is still representative of the atmospheric distribution of air ions.

   We could observe some connection of the 0.8-1 nm ion concentration to the absolute humidity, with a negative proportionality. However, there was no reversed proportionality found in the adjacent size range (1-1.2 nm) in relation to the absolute humidity. This might be attributed to the fact that atmospheric clusters have too much variability in their structures and compositions and also complications from atmospheric conditions may further hinder the humidity effect from being seen.

   Some comparisons with previous results reported by other researchers are included in the revised manuscript.

   '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 a 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.'

4. Cosmic rays are acknowledged as a significant contributor to atmospheric ionisation and hence cluster ion formation, and briefly mentioned here but are not explicitly ac- counted 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 ac- counted 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 cap- tured 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.

   Thank you for the comment! Unfortunately, to our knowledge, there is no cosmic flux data available at our site.

   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 ionisation chamber basically measured the ionisation 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 also muons. But for simplicity, we termed the ionising capacity derived from the dose rates recorded by the gamma spectrometer as the gamma ionising capacity. This is clarified in the revised manuscript.

However, we are aware of the possible missing of high-energy cosmic ray muons from our counting system. Either because they do not interact enough with the detector material or their light production and subsequent electrical pulses exceeding the dynamic range of the instrumentation. This point is made addressed in the revised manuscript.

Concerning solar activities, although cosmic ray intensity is anti-correlated with them, the influence contributes only a few percentages to variations in cosmic ray intensity for 2003-2006 (Moraal and Stoker, 2010;Hensen and Hage, 1994). However, as can be seen from our results, the terrestrial activity and gamma from radon decays accounted for the major contribution in the gamma ionising capacity. Therefore, even if there are remarkable solar events during these periods, they could hardly be isolated based on our dose rate data.

Below are additions and revisions made to the manuscript

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 a 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 was 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 ($Q\gamma$).'

5. 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.

   Thank you for the comment! We added description on thoron and stated that its contribution was not considered in our study and cited Laakso et al. (2004).

   'The contributions from actinon (radon-219) and thoron (radon-220) were excluded from our study, because they are present in trace amounts naturally and have remarkably shorter half-lives (3.96 s for actinon and 55.6 s for thoron) than radon-222, which hardly permit them enough time to migrate out of the ground, especially in the case of actinon. Frozen ground and snow cover could substantially cease the transportation of thoron to the atmosphere during cold months. Even the vegetation reduces the flux of thoron from the ground to the surface air (Mattsson et al., 1996). Besides, Laakso et al. (2004) found only little contribution by thoron in the SMEAR II station to the overall radon activity.'

6. 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.

We appreciate the comment and suggestion of the reviewer. We added processes involved in cluster ion formation to Fig. 1. The original information on the demonstration of what are primary ions, molecular ions and cluster ions contained in Fig. 1 is retained. We intended to present clearly the relationship between primary ion, molecular ions and clusters ions and we think that a graphical illustration can better convey the message. We think that the suggestion of the reviewer would be a good addition to the figure and these processes are added now to describe better the underlying processes.

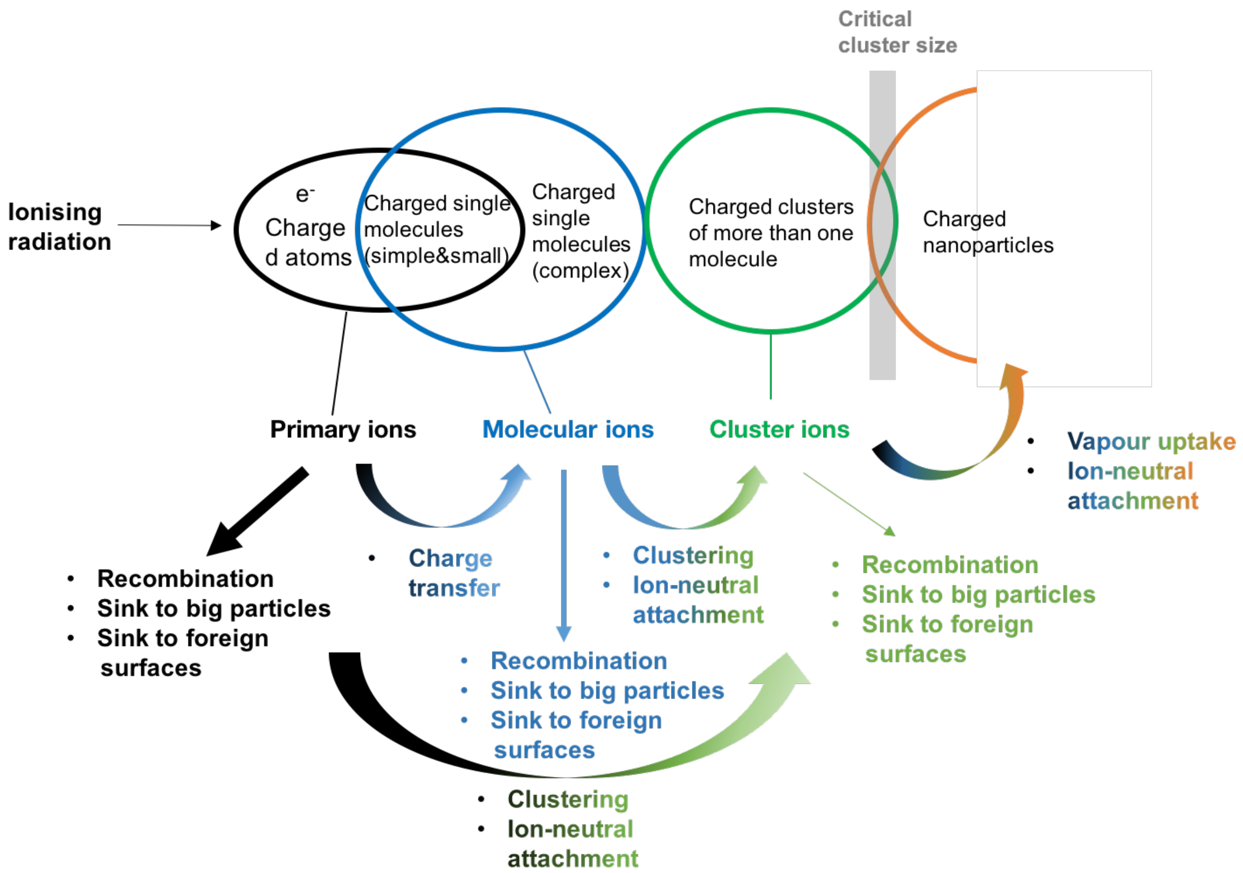
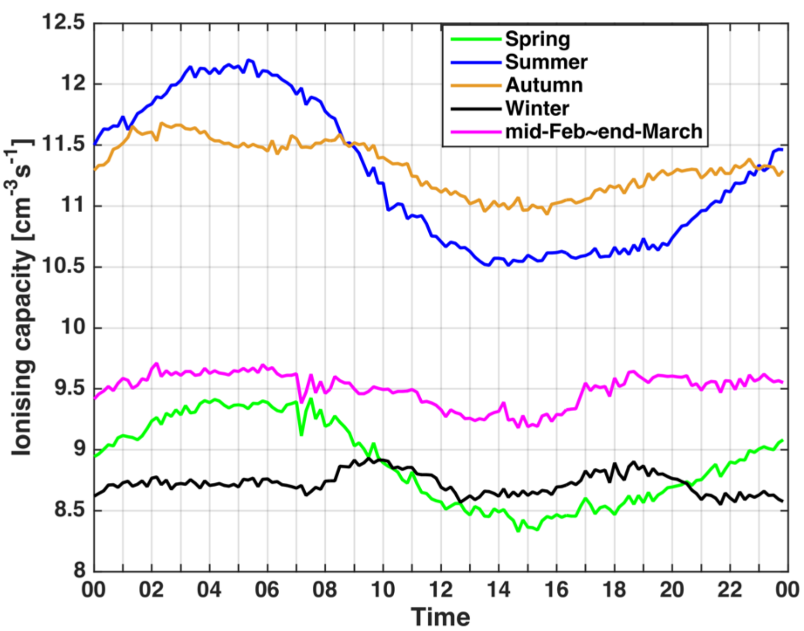
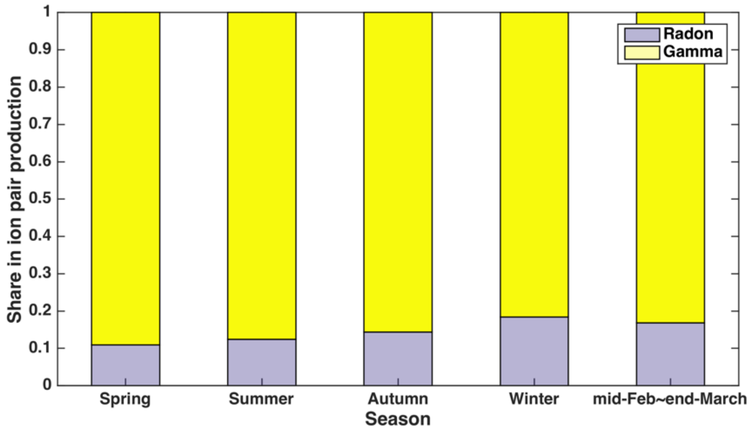


Figure 1. A schematic demonstration on the relationship between primary ions, molecular ions and cluster ions, as well as processes governing their formation and loss.

7. 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.

Thank you for the comment! The feature was blurred slightly by the 3-month average window, as can be seen from the figures below. There was a weak diurnal cycle in the min-Feb to end-March period and the ratio of medians in this period to medians in spring and in winter are 1.07 and 1.09, respectively. The contribution by radon in this mid-Feb to end-March period fell in between those in spring and winter. These hidden features are not very remarkable and, therefore, the choice of the four 3-month periods can be still considered representative.





8. 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.

   Thank you for the comment! The data were from our long-term routine measurements. There were small gaps every now and then due to maintenance activities. The ion spectrometer was introduced in 2003 and therefore there were about 2-month data missing in the beginning of 2003. But otherwise, we had almost full coverage of data over these four years. For the ionising radiation measurements, we had almost full data coverage in 2005 and 2006 for radon data and in 2003 and 2006 for total gamma. We mainly had gaps in ionising radiation measurements in the latter half of year 2004. However, gathering up the four-year data, we have a very large dataset and have data for every single day-of-year. Since we focused mostly on the general trends shown by the medians of the data, some gaps in the data could hardly bias our interpretation.

   Information about data coverages is added in the revised manuscript

   In section 2.1

   'For the study period of 2003-2006, the data availability for air ions, particles, radon and gamma was 91%, 99%, 71% and 76%, respectively, allowing the coverage of every single day-of-year by each dataset.'

9. 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.

The low pressure used in mass spectrometers could indeed cause loss of water and other loosely attached molecules on the sampled clusters. Such breaking-up of clusters is more likely to happen in mass spectrometers than in air ion spectrometers. APi-ToF is a mass spectrometer for study the mass distribution of atmospheric clusters, but it provides no direct information on mobility. Although some information about the mobility of the sample clusters can be obtained via a comparison with an ion spectrometer, like the work down by Ehn et al. (2011), the mass-mobility relationship has some dependency on the choice of model, as shown by Ehn et al.

10. 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).

    We added this information in the revised manuscript as follows

    'A full measurement cycle scanning through the mobility range for both polarities takes 10 min.'

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

    We add the reasoning for using O horizon soil data in the revised manuscript as follows

    '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.'

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

    Thank you for the correction! Changes are made accordingly

13. P5 L15 "described" not "descripted"

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

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

    Thank you for pointing this out! However, since the instrumentation for soil measurement has been described carefully by Pumpanen et al. (2003) and Ilvesniemi et al. (2010), we decided to remove the details from our paper, as suggested by the other reviewer. We add citations to those published works for readers who would like to know more about the measurement.

15. P12 L10 "examine" not "exam"

    Thank you for the correction! It is changed accordingly.

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

    We intended to say that we investigated the connection between the radon ionising capacity and RH after eliminating the influence brought by MLH, temperature and wind on the radon ionising capacity. And it is rephrased in the revised manuscript as follows

    '⋯ no clear correlation between the radon ionising capacity and RH was found based on our dataset after ruling out the variability in the radon ionising capacity brought by MLH, temperature and wind.'

17. 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.

    Thank you for pointing out this! We reformulated the sentence as follows

    '⋯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.'

18. P14 L9 "blooming" not "booming"? P20 L9 "localised" not "focalised"? P20 L26 "level out" not "level up" P22 L2 "support" not "supports"

Thank you for these suggestions! Followings are the changes made in the revised manuscript: 'booming season' to 'growth season', 'focalised' to 'localised', 'level up' to 'level out' and 'support' to 'supports'.

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

Thank you for pointing this out! The full citation is added in the revised manuscript.

20. P33 L4 "0-4 cm" not "-4-0 cm". P34 Fig 2, both y-axes "ionising" not "ioning"

Thank you for spotting these typos out! They are corrected accordingly.

21. P43 L3 "T was below" not "T below"

Thank you! It is corrected accordingly.

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22.