Author's response to referees comments on:

Ion - particle interactions during particle formation and growth at a coniferous forest site in central Europe

S. G. Gonser, F. Klein, W. Birmili, J. Größ, M. Kulmala, H. E. Manninen, A. Wiedensohler and A. Held

Both reviews are in general positive and did only demand minor revisions.
We greatly appreciate the thoughtful comments and suggestions that helped to improve the manuscript. Thank you very much for your time and effort! We trust that all suggestions have been addressed accordingly in the revised manuscript. We respond to the specific comments as follows:

Referee I (MB Enghoff)

1: Elevated growth rates of neutral particles

Referee's comment:

Six of the eight analysed events occur in the morning. From Fig. 2 I estimate that there is about a 1 hour delay between the onset of ion growth (looking at the black circles) and the growth of neutrals. In the morning concentrations of condensable gas (sulphuric acid) is increasing due to photolytic production. I therefore suggest that the observed difference in growth rate could be due to differences in condensable gas concentrations - since the ions grow earlier in the morning where less material is available for growth they grow slower than the neutrals. Looking at Table 2 it seems that the enhanced growth of neutrals is indeed only observed in the morning events (except Aug. 17 – maybe this was a cloudy day?), whereas the two noon events do not show this difference in growth rates – in fact they show the opposite behavior as predicted by theory. From Fig. 7 and 8 it also appears that the growth rate anomaly is most pronounced in the events with a large Δt. Maybe the authors have some direct measurements of condensable gas concentrations that can be used to test if this idea has merit or not, but I think it should be considered in any case.

Author's response:

The reasoning of the referee is very plausible. The lower GRs of the earlier occurring ions may be related to differences in available gas phase precursor concentrations. The exact identity of the involved gas phase components cannot be deduced from our data. Likely, sulfuric acid and oxidation products of volatile organic compounds are involved. Of these, only sulfur dioxide, being the main precursor of sulfuric acid, was measured during the campaign. Sulfur dioxide concentrations mostly showed an increase in early morning and reached peak concentrations before noon. During all considered events, the morning hours were sunny (except the 19th June), with sunrise between 5 and 6 AM local time. As the production of sulfuric acid depends strongly on photochemistry, H_2SO_4 concentrations are expected to increase strongly with sunrise. Further, oxidation products of volatile organic compounds are also expected to exhibit an increase in concentration during the morning. Therefore, in the early morning, when the first intermediate ions are formed, the precursor gases are expected to be still at relatively low concentrations. Due to less available precursor gases, the ion growth is likely to be slower compared to neutral particles which form later. Nevertheless, the ions are likely to have an accelerating influence on the later occurring neutral particles, as presented in the manuscript.

We added the following paragraph to the Discussion (p. 186 line 16):

A possible explanation for the slower ion GR could be the diurnal variation of gas phase precursors like sulfuric acid or oxidation products of volatile organic compounds (VOC). In the early morning hours, when the first intermediate ions are formed, the precursor gas concentrations are expected to be low. On sunny days, as were most of the considered event days, the precursor gas concentration will increase during the day. This is due to increasing VOC

emissions from the forest with rising air temperature, as well as due to photochemical processes leading to the formation of sulfuric acid and oxidation of VOCs. Therefore, the neutral particle growth which occurs later could be enhanced due to higher concentrations of precursor gases.

Additionally, ion-ion and ion-particle interactions enhance neutral particle GRs. As the potential precursor gases were not measured in this study, the focus to explain our observation will be on ion-particle interaction processes. Nevertheless, it is not expected that the observations can be explained fully by ion-particle interactions.

2: Estimation of ion production

Referee's comment:

Due to limitations in the NAIS the ion production rate Q is underestimated, as explained by the authors. I was wondering if it was possible to estimate the expected ionization rate, depending on if Radon measurements are available. For cosmic rays the muon flux would need to be known. If so then the ionisation can be estimated using the energy deposition (~2 MeV/(g/cm2)) and ionisation energy (34 eV/ion pair). Something similar was done by Laakso et al (ACP 4, p. 1932, 2004).

Author's response:

Unfortunately, radon concentrations were not measured during the campaign, leaving the determination of the ionization rate solely to the measurements conducted with the NAIS.

3: Efficiency of NAIS

Referee's comment:

On p. 182-183 it is described how the AIS performs better than the NAIS – if you know the difference in efficiency (preferably size-dependent), could you use this to calibrate the NAIS-measurements? It would certainly be nice to see how much of the total particle formation could be due to the ions. Maybe you could make some kind of "best" case scenario to estimate an upper limit for the ion contribution?

Author's response:

The gain of information by performing a calibration of the NAIS with AIS data is considered to be fairly small, as measurements with the AIS were performed one year after the presented NAIS measurements. Therefore, background parameters differ significantly between the two measurements. An estimation of the "real" ion formation rate, based on NAIS and AIS measurements as well as on a publication by Hõrrak et al. (2008), is given in the last paragraph of p. 183. Under the given circumstances, we don't consider a calibration to be more reliable than the already presented estimation.

4: Figure 2

Referee's comment:

Are the smoothed fits used to find the growth rates? If so I suggest splitting the fit for total and neutral particles in 2 parts since there appears to be a very steep part up to $\sim$ 7 nm followed by a more flat line.

Author's response:

The referee is right, the smoothed "fits" are used to find the growth rates. However, the "fits" are the result of smoothing the times of maximum concentration by means of a running least square polynomial smoothing filter (Savitzky and Golay, 1964). Therefore, the method does account for changes in GR with particle diameter. When inspecting the black "fit" line in Fig. 2, a change in GR somewhere around a diameter of 7 nm is also visible.

Moreover, the aim of our analysis was to compare the GRs of ions and neutral particles by applying the same method to all our data. This objective is met with this approach.

5: Technical comments

Author's response:

- The term "temporal advance" was substituted by "time difference" as proposed by the referee.

- p.183 l7: "bended" was substituted by "bent" as proposed by the referee.

Fig. 1: Why not show the lines for the 10 nm ions down to 1.5 nm like the solid lines?

Author's response:

Ion-ion, ion-particle and particle-particle interactions were calculated for particle sizes interacting with particles of the same size as well as larger sizes. Once all coefficients were calculated for one reference size, the reference size was shifted towards the next larger size. Therefore, the information on the coefficients for the interaction of larger particles with smaller ones is contained in the previously calculated coefficients of the interaction of small particles with larger ones. In other words, the interaction of a large particle of size A with a smaller particle of size B is the same as the interaction of a particle of size B with a larger particle of size A. Therefore, Fig. 1 does not include smaller diameters for 10 nm ions and particles.

Referee II (Olaf Hellmuth)

1: Page 173, line 9:

Referee's comment:

The authors are asked to explain what 'total particles' are (unlucky notion). If charged particles are just a minor part of a particle ensemble, than it is understandable that changes in the whole particle ensemble can only be seen posterior to changes in some of the minor fractions (e.g., ions) with more or less pronounced retardation.

Author's response:

A clarification of the term "total particles" was added to p. 173, line 9:

… compared to total particles (neutral + charged particles; Manninen et al., 2010).

2: Page 173, line 12:

Referee's comment:

Please explain the notion 'overcharged' and 'equilibrium charging state' (see also line 23, and page 175, lines 19-24). This is nontrivial and essential for the topic of the study.

Author's response:

The paragraph on p. 173, line 9, was extended according to the referee's suggestion:

Furthermore, the charging state of aerosol particles during NPF was observed to be frequently overcharged (Gagné et al., 2010; Iida et al., 2006; Laakso et al., 2007). The ratio of charged particle concentrations to neutral particle concentrations in a defined diameter interval is defined as the charged fraction. In a bipolar ion environment, the size-dependent charged fraction of an aerosol will eventually reach an equilibrium charging state due to ion-particle interactions (Fuchs, 1963; Wiedensohler, 1988). When the charged fraction is elevated in comparison to the equilibrium charged fraction, an aerosol is defined to be in an overcharged charging state.

3: Page 174, line 7:

Referee's comment:

Radioactive decay and cosmic radiation are considered as the main source of sub-1.6 nm ionic clusters. As these processes do not have 'event-like' character, any burst-like behaviour of intermediate and large ions must be anyhow linked to growth or neutralisation processes. It would be helpful to add a critical assessment of the explanations given by previous studies (already cited in the text) on this finding.

Author's response:

We have added a brief discussion on the occurrence of intermediate and large ions as suggested by the referee:

Elevated concentrations of intermediate and large ions are usually only present during (1) NPF events, where small ions grow due to condensation of precursor gases, (2) snow fall associated with high wind speeds, where friction processes between the snowflakes are thought to be a source for intermediate ions (Virkkula et al., 2007), and (3) rain, where the splashing of rain droplets generates intermediate ions due to the balloelectric effect (Tammet et al., 2009).

4: Page 175, line 4:

Referee's comment:

Please add manufacturers information on uncertainties of NAIS.

Author's response:

Unfortunately, the manufacturer of the NAIS does not provide any uncertainties for the instrument. Discussions of the instrument accuracy can be found in the publications by Asmi et al. (2009), Gagné et al. (2011) and Manninen et al. (2011). The uncertainties given by these publications are described on p. 175, lines 19-28.

5: Page 176, line 17-28:

Referee's comment:

The authors collocated a dataset by merging their NAIS data with data from the mobility particle size spectrometer by means of 'a linearly weighted merging algorithm'. Is it possible to add an example how this looks like? What about the total uncertainty of the resultant particle number size distribution between 2 and 680 nm underlying the calculation of the sink rates. Here, errors originating from different sources are superposed.

Author's response:

The merging method is a rather simple approach, merging the two measurements in their overlapping range by means of linearly weighting the measured concentrations. An example is depicted in Fig. R1.
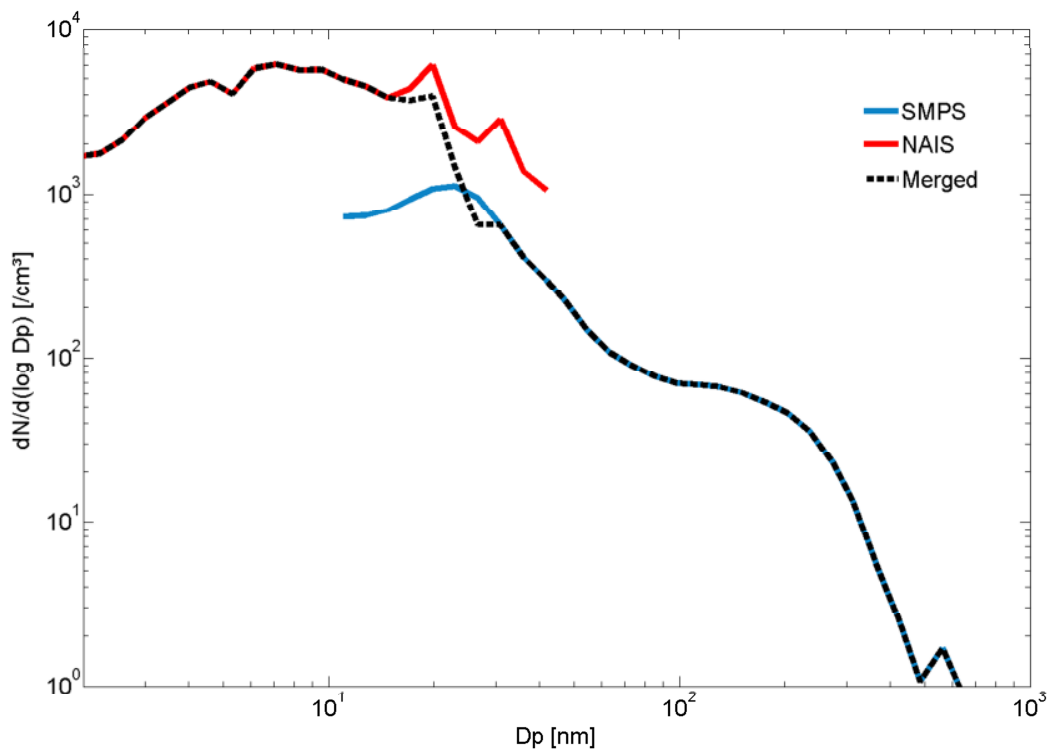
Fig. R1: Exemplary particle number size distributions measured with the NAIS (red curve) and the scanning mobility size spectrometer (blue curve); the result from merging the two measurements by means of linearly weighting between 15 and 27 nm is depicted by the black dashed curve. The number size spectrum was measured during NPF at "Waldstein" on 04 July, 2012.

For an assessment of the total uncertainty of the combined particle number size distribution, indeed, errors from different sources and instruments are superposed. The merged size distribution was used for the calculation of the coagulation and attachment sinks, which are both considered minor compared to recombination (cf. Fig. 1 in the manuscript). However, as the major scope of this study is to evaluate the influence of ion recombination on new particle formation the main basis of our calculations is the NAIS dataset. Therefore, the assessment of the error resulting from the superposed number size distributions is considered to be out of the scope of our work.

6: Page 177, Section 2.2:

Referee's comment:

The particle interactions (coagulation, ion attachment, recombination) have been 'calculated theoretically'. Here again, an uncertainty analysis would be helpful to span the corridor, inside which the 'true values' of the coefficients should be expected (maximum possible deviation from a reference value). Where do the authors get from, e.g., particle mechanic mobilities (please add unit), vdW interaction distance, or sticking probability? It is impossible to intuitively percept the overall uncertainties of the coagulation, attachment, and recombination coefficients in comparison to the corresponding size effect depicted in Fig. 1.

The equality of the recombination and attachment rates was proposed in the section, but not depicted in Fig. 1. Instead, the authors referred to an 'approximated theory' by Hõrrak et al. (2008) etc.

In view of the importance of the assigned parameters for the present approach, the qualitative description of the chosen approximations is, for my opinion, too fugacious here. The paper aims at elucidation of ion-particle interactions, an essential part of them is hidden in the underlying theory. Thus, the authors could add some additional sentences here.

Author's response:

An uncertainty analysis for the coefficients of coagulation, ion attachment and ion recombination was performed according to the suggestion of the referee. The parameters for the mechanic mobility (units have been added), sticking probability as well as the van der Waals extra distance were varied to assess the sensitivity of the coagulation coefficient on these parameters. The mechanic mobility was calculated according to Hinds (1999) and varied by 20 %, the sticking probability was varied between 1 and 0.5 and the van der Waals extra distance was doubled and reduced to 50 %, respectively. Updated Fig. 1 (black lines) shows the result of the variation of the mentioned parameters. The most pronounced effect on the coagulation coefficient is due to changes of the sticking probability.

In order to assess the possible variation of the recombination- and attachment coefficients, the electrical mobility, as provided by the NAIS, was varied for each particle / ion size. The magnitude of variation for each diameter was chosen to be the bin width of the NAIS size channels. The resulting variations of the two coefficients are now depicted in Updated Fig. 1 (yellow and green lines).

The resulting variability of the coefficients is small compared to the size effect of the single coefficients. Fig. 1 was updated accordingly:
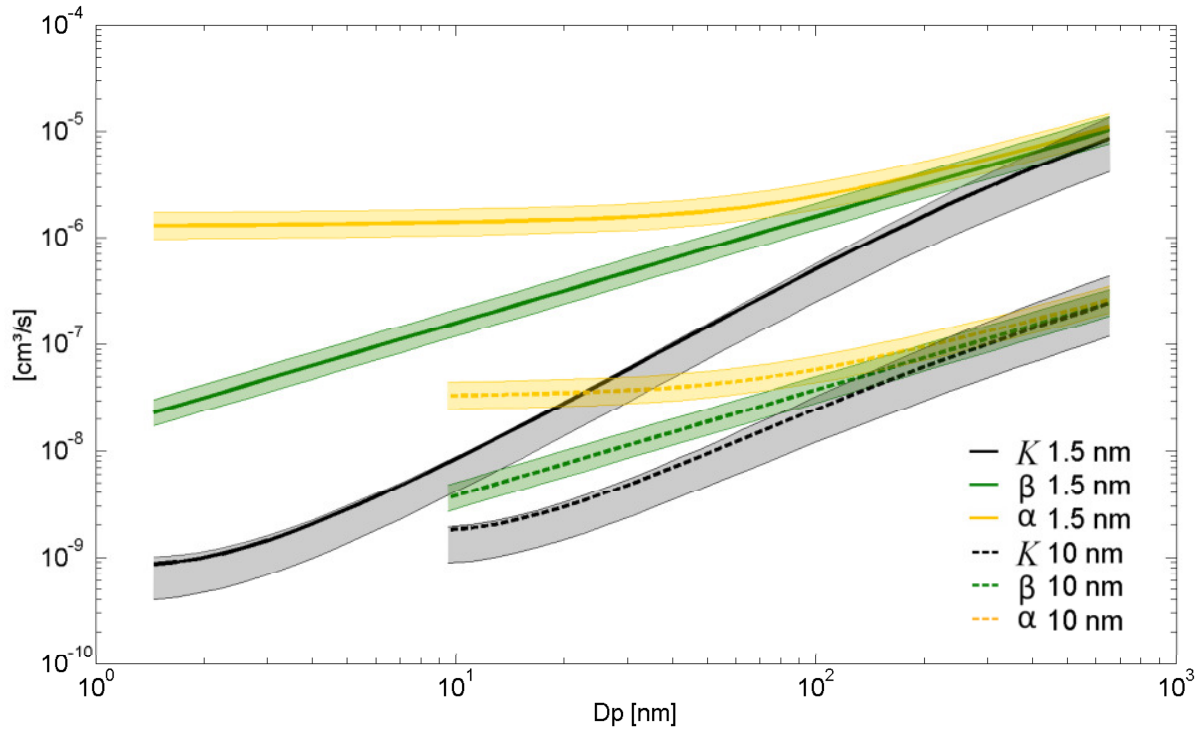
Updated Fig 1: *Coagulation coefficient K, attachment coefficient β and recombination coefficient α [cm³ s⁻¹] for small cluster ions and particles (1.5 nm, solid lines) and large ions and particles (10 nm, dashed lines) as a function of ion/particle diameter D_p. Shaded areas denote the variability of the respective coefficients, please refer to the appendix for details.*

Regarding the equality of the recombination and attachment rates, the original description was indeed somewhat misleading. Of course, the equality of the two coefficients is only given when the attachment coefficient is calculated for oppositely charged ions. In this case the attachment is denoted as recombination. Figure 1, however, shows the coefficient for ions attaching to neutral particles (β) and the coefficient for ions attaching to oppositely charged ions (α). Changes to clarify this issue have been added to the manuscript.

The sentence on p. 178 line 5 was changed to:

Considering the case of cluster ions of diameter j interacting with oppositely charged clusters of a similar diameter k,   should be comparable to the constant value of α (Hoppel and Frick, 1986).

Further, the Appendix was extended according to the suggestions of the referee:

According to Tammet and Kulmala (2005), the size dependent coagulation coefficient   is defined as

$$K_{ij} = \frac{2\pi k T (B_i + B_j)(D_i + D_j + 2h)}{1 + \gamma - \frac{0.299\gamma}{\gamma^{1.1} + 0.64} + \gamma \frac{1-p}{p}}, \qquad (A1)$$

where

$$\gamma = 2 \frac{B_i + B_j}{D_i + D_j + 2h} \sqrt{\frac{2\pi k T m_i m_j}{m_i + m_j}}. \qquad (A2)$$

Here, k is the Boltzmann constant [1.38×10^{-23} J K^{-1}], T is the absolute temperature [K], B is the particle's mechanic mobility [m N^{-1} s^{-1}] (Hinds, 1999), D is the particle diameter [m], the sticking probability p was assumed to be one, and m is the mass of an individual particle [kg]. To account for van der Waals interactions, an extra distance h = 0.115×10^{-9} m is necessary (Tammet and Kulmala, 2005). The variability of the size-dependent coagulation coefficient due to a 20 % change in B, a doubling reduction to 50 % of h and the variation of p between 0.5 and 1 is depicted as shaded areas in Fig. 1. The sticking probability exhibits the most pronounced influence on the values of K_{ij}, while the other two variables only show a minor effect.

An approximation for the size dependent attachment coefficient β_{ij} is given by Hõrrak et al. (2008):

$$\beta_{ij} = \frac{2\pi D_i k T Z_j}{e} \frac{x}{exp(x) - 1} \sqrt{1 - \frac{2}{2 + n(n-1) + D_i/(10\ nm)}}, \qquad (A3)$$

where,

$$x = \frac{ne^2}{2\pi D_i \varepsilon_0 kT}. \qquad (A4)$$

Here, Z is the electric mobility of the ion provided by the NAIS [m² V^{-1} s^{-1}], e is the elementary electric charge [1.6×10^{-19} C], n is the number of elementary charges carried by a charged particle, and ε_0 is the electric constant [8.85×10^{-12} F m^{-1}]. The variability of β_{ij} was assessed by varying Z in the range of the bin width of the respective NAIS size channel (cf. Fig. 1).

For the interaction of oppositely charged ions of size j and k, a size dependent recombination coefficient α can be described by the attachment coefficient β_{jk} (Hoppel and Frick, 1986).

Therefore, the recombination coefficient (α_{jk}) is defined equally to the attachment coefficient, but only for oppositely charged ions:

$$\alpha_{jk} = \beta_{jk}. \qquad\qquad (A5)$$

7: Page 179-180, Section 2.3:

Referee's comment:

I understand equation (1) balancing the number concentration of recombined particles, but I do not understand the 'ionisation rate' Q, which assumes that the 'ion production rate is a function of two ion sink terms only, the recombination and the attachment of ions to the present background aerosol' (see Eq.(A8)). If there would be no source of ions, then I would expect that the initially present ions in the atmosphere would have already been removed by the two sink processes. In contrast to this, one has Radon emission from the soil and CGR impacts, permanently creating new ions. What do you mean with 'ionisation rate' if there is no ionisation?

Author's response:

Eq. A8 denoted the ionization rate in steady-state equilibrium. According to the referees suggestion the equation was changed to the balance notation and the corresponding paragraph was changed.

The sentence on page 180, line 14 was changed to:

For our site, Q was calculated by means of a simplified ion equilibrium equation, assuming steady-state equilibrium (Hoppel and Frick, 1986).

The formulation and the according paragraph on page 194 were changed to:

The balance equation for the ionization rate $Q^{\pm}$ is defined as:

$$\frac{dN^{\pm}_{<1.6\,nm}}{dt} = Q^{\pm} - \sum_{jk} \alpha_{jk} N_j^+ N_k^- - \sum_{ij} \beta_{ij} p_i N_i N_j^{\pm} , \qquad\qquad (A8)$$

where size classes j and k < 1.6 nm, N_j^+ and N_k^- are positive and negative ion concentrations in size class j and k [cm^{-3}], N_i is the concentration of neutral particles in size class i [cm^{-3}] and $N^{\pm}_{<1.6\,nm}$ is the number concentration of positive or negative ions with a diameter below 1.6 nm [cm^{-3}].

8: Page 181, line 3:

Referee's comment:

Again, the uncertainty problem. How the uncertainties in the method of Yli-Juuti et al. (2011) affect the conclusions drawn in the paper on the growth process (growth of ions vs. growth of neutral particles)? Please try to quantify whatever is possible to quantify. The difficulty in the interpretation of the results is the indissoluble merging of measurements, semi-empirical evaluation methods, and model-based assumptions without accompanying uncertainty analysis for the rates of growth and particle formation.

Author's response:

The dynamic behavior of a growing particle population makes an assessment of the uncertainty of the growth rate (GR) determination very difficult. For example, tropospheric new particle formation is not occurring during one short point in time followed by the growth process but new particles are continuously formed over a rather long period. Therefore, determining GRs from atmospheric measurements gives an approximation of the true particle GR. Although we cannot consider the determined GRs to be of absolute accuracy, the relative comparability between ion and particle GRs remains reasonable. This assumption is valid, as ion and particle GRs were determined with the same method, which was applied to data recorded by a single instrument.

To account for the referees comment we altered the text to make this issue more clear:

The determination of GRs from particle/ion measurements in the troposphere are always associated with uncertainties (Yli-Juuti et al., 2011) as the growth of a particle population is a dynamic process with several influencing factors. Therefore, the result of a GR analysis from field measurements will always only result in an approximation of the true GR.

9: Page 182, line 14:

Referee's comment:

'Soil gas' is a notion to describe the air volume in the soil, containing oxygen (allowing for respiration of both plant roots and soil organisms), and carbon dioxide as a respiration product of plant roots and soil microorganisms, and is not explicitly related to radioactivity. The unit Bq m^{-3} denotes the number of radioactive decay events per unit time and unit volume, but is not a 'gas concentration'. Please revise this sentence or make clear what is meant. Is 'Waldstein' a singular place with enhanced background radioactivity?

Author's response:

The sentences were revised:

The "Waldstein" site is located in the Fichtelgebirge mountain range, NE Bavaria. The Fichtelgebirge is known for its enhanced background radioactive radiation levels. In particular,

radon is elevated, reaching soil gas activity concentrations of up to 4000 kBq m^{-3} (Kemski et al., 2001; Lüers et al., 2007).

10: Page 182, line 20, reference to Fig. 3:

Referee's comment:

Radon is permanently emanating from the ground without diurnal variation. However, boundary layer mixing at a forest site is expected to reveal at least a weak diurnal cycle with suppressed mixing during night and enhanced mixing during the day. Hence, the radon concentration should reveal a dilution-induced minimum around noon. This supports author's conclusion regarding the link between radon and small-ions concentration pattern. The plotting of percentile regions in Fig. 3 is very fine in view of the very low concentrations observed. The large differences found between NAIS and AIS measured ion concentrations (the latter unfortunately not available for the same period) underline the high importance of an uncertainty analysis (page 183). The uncertainties in the measurement process (type A vs. type B device) are of the same order as the atmospheric effects we are looking for.

Author's response:

The referee is right, the uncertainties in absolute concentrations as measured with the NAIS are substantial. However, the size information is thought to be of sufficient quality to calculate relative measures such as GR.

11: Page 183, line 20:

Referee's comment:

It seems that the authors have considered steady-state equilibrium between ion production and ion loss, which allows one to identify the loss rate with the production (ionisation) rate Q. Please make this clear at the place where the ionisation rate has been introduced. The authors derived a certain value of the ionisation rate, but at the same time suggesting that the true value is 'probably' a factor of three larger ... Maybe. It is experts suggestion, but its resilience is lower than that of calculated values.

Author's response:

The formulation/introduction of the ionization rate (Q) has been revised according to an earlier comment of the referee (please refer to comment 7 above).

In addition, the sentence in line 25, page 183 was changed to the following:

Considering these facts, Q was probably much higher than suggested by the NAIS measurements.

12: Pages 184-185, interpretation of Figure 6:

Referee's comment:

Differences between the growth rates of ions and all particles can be seen, but their significance remains obscure. One should more carefully formulate (p. 185, line 3) that the 'derived enhanced GR t compared to charged particle growth rates stands in contrast to growth theories ...'. The notion 'observation' has a great 'authority' and 'reputation' which should be reserved for quality-assured measurements being subject of uncertainty analysis (JCGM 2008). In the present case different approaches (measurements, semiempirical approaches, balancing of uncertain source terms) have been mixed, which is of course justified, but softens the explanatory power of the conclusions. I cannot see any argument calling the cited growth theories into question, I only see that without a comprehensive uncertainty analysis it is impossible to draw conclusive statements about any molecular theory employed in the present modelling approach. Moreover, with respect to ion-particle interactions I consider it already as a very great challenge to setup suitable laboratory experiments for their elucidation. The conditions in the field are much more difficult to control. It seems that at the level of nanoscale processes there are no pure 'observations', but the measurement process itself is already as complicated as the investigated atmospheric new particle formation. Thus, I suggest an 'equidistance' of both 'nanoscale observations' and 'nanoscale theories' to the 'nanoscale verity'. Perhaps, one should emphasise the tentative character of the present findings and modelling approaches and to be more carefully with respected to 'revision of theories' (an inappropriate modelling approach (by uncertain closure) does not necessarily imply a wrong theory).

Author's response:

The referee is right, the statement (p. 185, line 3) is probably somewhat misleading. Our intention was to underline the astonishing fact of the higher neutral growth rates. It was never intended to disprove the prevailing growth theories.

The paragraph was altered to:

The enhanced GR_t compared to charged particle growth rates deviates from theories for pure condensational growth, where the presence of a charge enhances the growth rates of small and intermediate ions (e.g. Yu and Turco, 2000; Yue and Chan, 1979). Therefore, condensational growth on its own cannot explain the apparent GR differences.

13: Page 186, line 17:

Referee's comment:

There is no reason to expect that the evolution of the ion concentration can be explained only by condensational growth. The appropriate description of ion–ion and ion–particle interactions is a prerequisite for the closure of the time rate of change equation of the ion concentration. The correct tuning of these terms is purely empirical, a fortiori as in the paper the specification of the 'precursor gas' does not explicitly appear. Forward integration of a model and comparison with a predicted observable

is very straightforward, but the derivation of a balance term from observed time series employing uncertain closure assumptions is not in equal measures.

Author's response:

We agree with the referee that ion and particle growth cannot be restricted to pure condensation. Ion/particle interactions are clearly needed to explain the whole process. Due to the lack of data on precursor gases we focused on the ion-particle interactions, not expecting to be able to completely explain the observed growth behavior.

According to the referee's comment the paragraph (p. 186, line 10) was extended:

… Furthermore, charged clusters clearly activate more easily and grow more quickly (e.g. Lushnikov and Kulmala, 2004; Winkler et al., 2008; Yu and Turco, 2000). Keeping this in mind and assuming no ion-ion and ion-particle interactions, the time difference between an earlier occurring ion fraction and the total particle fraction during the growth process should increase or remain constant. However, our measurements show a contrary behavior: once formed, the neutral particles grow considerably faster than the ion fraction, and eventually, the earlier occurrence of the ions vanishes completely.

A possible explanation for the slower ion GR could be the diurnal variation of gas phase precursors like sulfuric acid or oxidation products of volatile organic compounds (VOC). In the early morning hours, when the first intermediate ions are formed, the precursor gas concentrations are expected to be low. On sunny days, as were most of the considered event days, the precursor gas concentration will increase during the day. This is due to increasing VOC emissions from the forest with rising air temperature, as well as due to photochemical processes leading to the formation of sulfuric acid and oxidation of VOCs. Therefore, the neutral particle growth which occurs later could be enhanced due to higher concentrations of precursor gases.

Additionally, ion-ion and ion-particle interactions enhance neutral particle GRs. As the potential precursor gases were not measured in this study, the focus to explain our observation will be on ion-particle interaction processes. Nevertheless, it is not expected that the observations can be explained fully by ion-particle interactions.

14: Section 4, Discussion:

Referee's comment:

To my mind, this section can be shortened somewhat. Several points have been outlined previously in the manuscript.

Author's response:

The section has been revised with respect to repetitions. A few sentences were considered to be unnecessary and deleted from the manuscript (p. 186, lines 19-21 & 26-28; p. 187, lines 5-9; p. 188, lines 18-19).

15: Final comments

I found the study very interesting. The authors did a diligent work! If the authors cannot provide a comprehensive uncertainty analysis supporting their conclusions, I recommend to make the tentative character of the present findings more clear. In that sense, the revisions are minor.

Author's response:

We thank the referee for the constructive comments and suggestions which helped to improve the quality of this manuscript.

The focus of our study lies on the observation of new particle formation in the field and on the attempt to give a qualitative explanation of the observed phenomena. A comprehensive uncertainty analysis was judged to be out of the scope of our study. Therefore, the tentative character of this paper was made more clear in the response to the referee's comments no. 8 and 13 and in other passages of the text. Nevertheless, an assessment of the uncertainty underlying the coagulation, recombination and attachment coefficients was performed to reinforce the findings of our work.

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