

Dear Editor,

Thank you very much for carefully considering the referees comments and your effort to enhance the quality of the manuscript.

Your comments:

I think the following two referees' comments are not adequately addressed. The manuscript should be publishable after you properly address these in the revised manuscript.

(1) Estimation of ionization production. You acknowledged that "Q was probably much higher than suggested by the NAIS measurements." How higher is "much higher"? Could you provide a quantitative best estimation? In the abstract you mentioned that "The "Waldstein" research site is located in a spruce forest in NE Bavaria, Southern Germany, known for enhanced radon concentrations, presumably leading to elevated ionization rates." Does this imply the ionization rate at this site higher than that in Hyytiälä?

(2) Uncertainties of NAIS. You cited several previous publications about the uncertainties. Please summarize the major uncertainties in your manuscript.

In page 13, you pointed out that "it generally underestimates the ion concentrations and overestimates the total particle concentrations." Please provide more details (reasons behind these and magnitudes) and references.

Author's response

(1) In the discussion version of the manuscript we gave a quantitative best estimation of the ionization rate (page 183, line 25):

Considering these facts, Q was probably well above 3 ion pairs cm⁻³ s⁻¹ during summer 2012 at "Waldstein".

This estimation of Q had been removed from the manuscript due to a referee's comment (comment 11 of O. Hellmuth). According to the editors suggestion we restored the sentence to the original statement.

According to Hirsikko et al. (2007) the median radon activity concentration at Hyytiälä is 1.49 Bq m⁻³. On the other hand, radon concentrations were reported to vary between 20 and 70 Bq m⁻³ at the Waldstein site (Lüers et al., 2007). Measurements at Hyytiälä were conducted at a height of 6 m above ground while measurements at Waldstein were conducted at 0.2 m above ground. Therefore, the comparability of the two activity concentrations is limited due to the steep radon activity concentration gradient with measurement heights. However, we expect the radon activity concentration and ionization rate to be higher at Waldstein than at Hyytiälä.

(2) The major uncertainty of the NAIS instruments is an overestimation of the total particle concentration. This is described in detail in section 2.1 of the revised manuscript (page 4; line 25). We revised this section to make this fact more clear:

In particle mode, the recorded number size distribution is inverted by the instrument software, assuming the Fuchs-charge equilibrium of the sample prior to charging and that all classified particles are singly charged. However, if the particle population is not in charge equilibrium but either overcharged or undercharged, the NAIS will overestimate or underestimate the total particle concentrations, respectively (Kulmala et al., 2012). During NPF the charging state of aerosol particles was observed to be frequently overcharged (Gagné et al., 2010; Iida et al., 2006; Laakso et al., 2007). Additionally, an overestimation of the total particle concentration by a factor of 2-3 is a general characteristic of NAIS instruments, as was shown by an intercomparison of several NAIS instruments by Gagné et al. (2011). Therefore, total particle concentrations in this study are very likely to be overestimated.

The paragraph on page 13 was changed according to the editor's comment:

This may be partly due to the performance of the NAIS. As described in section 3.2 ion concentrations in our study are generally underestimated, while an overestimation of total particle concentrations is a typical property of NAIS instruments (Gagné et al., 2011).

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