

I think the authors of „Experimental investigation of ion-ion recombination at atmospheric conditions“ have studied a very important topic. Justified and accurate results about the ion-ion recombination characteristics are essential for understanding several atmospheric processes. Nevertheless, in the current form the manuscript contains several declarations, statements and/or results that should be justified and/or explained and/or discussed better and/or more in detail. I think these shortcomings should be eliminated.

Introduction:

The discussion about the formation of small ions in the air is supported only by one (old) reference (Smith and Spang, 1995). I do not think that this is the only available choice; neither this is not the absolutely best choice. I suggest to put a proper phrase (e.g., "model chemical composition air ions") into Google and to use/discuss other/newer references/studies (as well).

The overview of the former studies of the conditions where ion-ion recombination can be dominant sink of small ions is too vague. I suggest to put a proper phrase (e.g. "new aerosol particle formation ion driven processes") into Google and discuss some more results about the conditions where ion-ion recombination can be dominant.

It is hard to understand, what do the authors mean by "In past studies, the ion-ion recombination rate was calculated for understanding aerosol diffusion charging...". In case they want to say "... ONLY for understanding aerosol diffusion" then this is not true, e.g., consider (Tamm et al., 2006) (already referred by the authors). In case the authors want to say that the presented references (Natanson, 1960; Bates and Flannery, 1969) are the ONLY STUDIES, where the recombination rate was (remarkably) considered then this is not true, neither. I think the authors should express their meaning more clearly. Also, I wonder why the authors state "...was measured primarily for dosimetry purposes" and provide only one (old) reference (McGowan, 1965). I think the authors should present more extensive discussion about that key topic (about previous studies of ion-ion recombination) and the authors should discuss a larger number of proper references (e.g. Pageoph, 116, 1101-1113; J. Chem. Phys., 90(11), 6328-6334; Adv. At. Mol. Phys., 20, 1-40; Can. J. Chem., 47, 1711-1724; DOI: 10.1039/c2an35849b; ...).

Section 2.

Pg 3672. The authors state "The concentration of aerosol particles was below 30 per cubic cm...". What is the basis of such a statement? "Aerosol-free" can maybe sound well, but is this phrase the only proof?

Several former studies have distinguished between "initial recombination" and "volume recombination" (e.g. NUKLEONIKA 2007;52(1):7-12). Is this "initial recombination" taken into account?

How do the authors estimate the uncertainty of the obtained (raw) results, e.g. uncertainty of NAIS results?

Section 2.4

Pg 3675-3676. Is there any proof that the "dilution system" functioned just the way and only the way it was expected to function? Are the authors convinced that the dilution system did not cause any disturbing turbulence?

Section 3.1.

Pg 3676. Commonly, beta (or beta as a function of diameter) marks the sink, attributed to ambient (aerosol) particles (e.g., Tamm et al., 2006, already referred by the authors). Do the authors have any solid reason to drop the common notations?

pg 3676-3677. The equation (2) has been already solved by Israël (1970, Atmospheric Electricity, vol 1, p. 167). True, it contains a misprint.

Section 4.1.

pg 3680. "...increased approximately by a factor of 5 (from $11 \cdot 10^6$ to $2.5 \cdot 10^6$)..." How should I understand this?

section 5.

Pg 3684. I do not agree with the statement "... first study to experimentally investigate the ion-ion recombination at atmospheric conditions"; see examples of potential references to former studies above. Also, this investigation is not "...at atmospheric conditions". Yes, the authors have used air-like mixture(s), but this is not exactly "atmospheric conditions". It is still an additional question, how well the experimental conditions correspond to atmospheric conditions.

Fig.1. (b) The font for NAIS and API-TOF is very small.

Fig.2. The concentrations of positive ions behave rather differently from the ones of negative ions. For example, shortly after 6:00 the concentration of negative ions has brief but deep depression, accompanied by upward spike in the concentration of positive ions. In general, positive and negative ions should be born in pairs and recombine in pairs, therefore they should be strongly correlated. Also, the ion concentrations at Beam=0.9 are at times even lower than the ones at Beam=0.65. What are the reasons of these odds?

Fig.3. What is the reason of the large concentration fluctuations within the wide gray area ? Moreover, several blue circles (ion concentrations) are even outside this area ? Also, I think it would be useful to include a clear (graphical) example, how the authors have accomplished "...fitting the steady state balance equation..".

Fig.5. How can loss rate (constant beta) depend on recombination coefficient alpha ? Is there any unrevealed theoretical link ? The presented dependence tends to imply that these both quantities are some functions of some other unrevealed quantity (perhaps some functions of some systematic error ?). I think the authors should explain this circumstance more in detail.

Fig.6. According to the figure, the number results near 280 K are below the ones near 290 K. According to the figure, the recombination coefficient should have a strange temperature dependence: nearly constant below about 250 K, deep depression between 250 K and 280 K and slow uptrend above 280 K. Any explanation?