

Second review of Yan et al., acp-2018-187

I am happy with the replies except for the reply to my comment about line 347 (line numbers with respect to the original ACPD manuscript):

First, there seem to be three different nomenclatures used in the manuscript: Are J_{IIN} (line 346), $J_{2.5}^{\pm}$ (line 184) and J_{ion} (Figure 6, y-axis) in your definition meant to be identical? This is stated nowhere, but seems to be the case from your (very short) answer to my comment; please use identical nomenclature in all cases if it is supposed to be the same thing. Note that this nomenclature is **not** identical to the nomenclature used in Wagner et al, 2017. In Wagner et al. J_{IIN} is defined as $J_{\text{IIN}} = J^{\pm} + J_{\text{rec}}$ which is correct in my opinion, as the true ion-induced nucleation rate (i.e. “particles that overcame the nucleation barrier as ions”) is given by this expression. Therefore, if you want to discuss the “ion-induced nucleation rate (J_{IIN}) and its contribution to the total formation rate ($J_{\text{IIN}}/J_{\text{total}}$)” (l. 346-347) then you need to introduce this definition for J_{IIN} and you need to introduce and calculate J_{rec} and include it properly in your numbers for J_{IIN} and the ion-induced fraction ($J_{\text{IIN}}/J_{\text{total}}$). Note that it is not really of interest how big the charged formation rate $J_{2.5}^{\pm}$ is for 2.5 nm particles but rather the true charged nucleation rate at the critical cluster size J_{IIN} .

To illustrate the important difference: Imagine a situation where all nucleation is ion-induced (like in a Wilson cloud chamber, where neutral nucleation is completely suppressed but supersaturation is sufficient for ion-induced nucleation to take place). In a situation where the subsequent growth is slow compared to the recombination (which is typically the case), all (or almost all) the charged particles that were formed by IIN originally would recombine before reaching the size of 2.5 nm. In this case $J_{2.5}^{\pm}$ would be zero or very small compared to $J_{2.5}$ and your definition returns a value for $J_{\text{IIN}}/J_{\text{total}}$ that is zero or very small, although it should be 1!

Therefore, I do not agree with your interpretation and numbers for J_{IIN} and $J_{\text{IIN}}/J_{\text{total}}$ as presented in Section 3.5 and Figure 6.

Second, J_{total} is also not introduced in the text, I assume it is meant to be $J_{\text{total}} = J_{2.5}^{\pm} + J_{2.5}$. (which would be identical to $J_{\text{total}} = J_{\text{n,tot}} + J_{\pm} = J_{\text{n}} + J_{\text{rec}} + J_{\pm}$ in Wagner et al.)

Third, I do not agree with eq. 1 in line 176. I think it needs to include a term $-\alpha N_{2.5-3.5}^{\pm} N_{<3.5}^{\mp}$ and a term $+\beta N_{2.5-3.5}^{\pm} N_{<2.5}^{\pm}$ to reflect the gain of neutral particles from ion-ion recombination and the loss of neutral particles due to ion-neutral collisions, just symmetric to the definition of J^{\pm} . Therefore, the $J_{2.5}$ data should be reanalyzed.

Fourth, please discuss the assumed value of $\beta = 1 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ (l. 189), I don't see why this value should/could be larger than the kinetic limit for ion-molecule collisions which is around $2.4 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$ (e.g. Viggiano et al., J Phys. Chem., 1997).

Fifth, all the added references (Eisele et al., 2006; Iida et al, Lovejoy et al., 2004, Wagner et al., 2017, etc.) are not included in the list of references. Please include.

I am sorry to bring all of this up during the second round of review (and I should have noted the third and fourth point already during my first review), but this issue needs a much more thorough discussion than your 2 lines of answer to my comment about line 347.