**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):

We thank the reviewer for the valuable comments and suggestions. We provide one-to-one answers to the comments in blue below, and the respective modifications in the main text are in purple.

First, there seem to be three different nomenclatures used in the manuscript: Are JIIN (line 346), J2.5 (line 184 ) and JIon (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. JIIN is defined as JIIN = J(+/-) + Jrec 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 (JIIN) and its contribution to the total formation rate (JIIN/Jtotal)" (l. 346-347) then you need to introduce this definition for JIIN and you need to introduce and calculate Jrec and include it properly in your numbers for JIIN and the ion-induced fraction (JIIN/Jtotal). Note that it is not really of interest how big the charged formation rate J2.5(+/-) is for 2.5 nm particles but rather the true charged nucleation rate at the critical cluster size JIIN. 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 J2.5(+/-) would be zero or very small compared to J2.5 and your definition returns a value for JIIN/Jtotal that is zero or very small, although it should be 1! Therefore, I do not agree with your interpretation and numbers for JIIN and JIIN/Jtotal as presented in Section 3.5 and Figure 6.

We appreciate the reviewer's point as it will drastically improve the consistency of the paper and prevent confusions towards the nomenclature used throughout the text. We also apologize for not addressing the problem properly during the first round of review. Although we agree with the calculation presented in Wagner et al. (2017), we must base our analysis on the available atmospheric measurements and build on available atmospheric literature. Anyway, we made substantial revisions to the text, as detailed below:

First, we made it clearer what we mean by the formation rates of ions and particles obtained from our measurements (equations 1 and 2, see our response to the comments 2 and 3 below).

Second, we rewrote the beginning of section 3.5 to address the main concerns pointed out by the reviewer. It now reads:

"In order to get further insight into the importance of IIN during our measurements, we compared the formation rate of 2.5 nm ions, $J_{ION} = J_{2.5}^{\pm}$ (see Eq.2) to the total formation rate of 2.5 nm particles, $J_{TOT} = J_{2.5}$ (see Eq.1). The ratio $J_{ION}/J_{TOT}$ is equal to the charged fraction of the 2.5 nm particle formation rate. In analyzing field measurements, a similar ratio at a certain particle size (typically 2 nm) has commonly been used to estimate the contribution of ion-induced nucleation to the total nucleation rate (see Hirsikko et al. 2011, and references therein). It should be noted that $J_{ION}/J_{TOT}$ represents only a lower limit for the contribution of ion-induced nucleation, as this ratio does not take into account the potential neutralization of growing charged sub-2.5 nm particles by ion-ion recombination (e.g. Kontkanen et al., 2013; Wagner et al., 2017). At present, measuring the true contribution of ion-induced nucleation to the total nucleation rate is possible only in the CLOUD chamber (Wagner et al., 2017). We were able to calculate $J_{ION}$ and $J_{TOT}$ for 57 (out of 67) cases, and the ratio $J_{ION}/J_{TOT}$ varied from 4 to 45%, showing a clear correlation with the HOM signal (Fig. 6A)…"

Third, we rewrote the caption of Figure 6 into the following form:

"A) Charged fraction of the formation rate of 2.5 nm particles as a function of the total signal of HOM ions color-coded by the $H_2SO_4$ concentration, and (B, C and D) the differences in $J_{ION}$, $J_{TOT}$, and $J_{ION}/J_{TOT}$ between the $H_2SO_4$ – $NH_3$-involved events (S-E) and other events (O-E)."

Finally, we removed the text ", contributing up to 40% of the total nucleation rate" from the last paragraph of section 4.

Hirsikko, A., Nieminen, T., Gagne, S., Lehtipalo, K., Manninen, H. E., Ehn, M., Horrak, U., Kerminen, V.-M., Laakso, L., McMurry, P. H., Mirme, A., Mirme, S., Petäjä, T., Tammet, H., Vakkari, V., Vana, M., and Kulmala M.: Atmospheric ions and nucleation: a review of observations, Atmos. Chem. Phys., 11, 767-798, 2011.

Kontkanen, J., Lehtinen, K. E. J., Nieminen, T., Manninen, H. E., Lehtipalo, K., Kerminen, V.-M., and Kulmala, M.: Estimating the contribution of ion-ion recombination to sub-2 nm cluster concentrations from atmospheric measurements, Atmos. Chem. Phys., 13, 11391-11401, 2013.

Second, Jtotal is also not introduced in the text, I assume it is meant to be Jtotal = $J2.5^{\pm}$ + J2.5. (which would be identical to Jtotal = Jn,tot + J± = Jn + Jrec + J± in Wagner et al.)

Third, I do not agree with eq. 1 in line 176. I think it needs to include a term 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. Therefore, the J2.5 data should be reanalyzed.

We thank the reviewer for these two comments. However, there appears to be a misunderstanding, probably because we were a bit unclear in defining the quantities in equation 1. The quantity $J_{2.5}$ represents the total formation rate of 2.5 nm particles obtained from measurements, not just the neutral fraction of these particles (with some influence by recombination products). As a result, there is no need for equations 1 and 2 to be symmetric with respect to the terms representing ion-ion recombination or ion-aerosol attachment.

In order to avoid the potential confusion noted by the reviewer, we modified the sentence prior to equation 1 into the form: "The formation rate of 2.5 nm particles includes both neutral and charged particles, and it was calculated from the following equation:"

Furthermore, the sentence prior to equation 2 as modified as: “Calculating the formation rate of 2.5 nm ions, or charged particles, includes two additional terms….”.

Fourth, please discuss the assumed value of B = 1 x 10-8 cm3 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 x 10-9 cm3 s-1 (e.g. Viggiano et al., J Phys. Chem., 1997).

We thank the reviewer for his comment; however we do not see how 2.5 nm ion-neutral collision rate would be related to molecule – sulfuric acid reaction rates presented in the referenced article (e.g. Viggiano et al., J Phys. Chem., 1997) entitled Rate Constants for the Reactions of XO3 -(H2O)n (X ) C, HC, and N) and NO3 -(HNO3)n with H2SO4: Implications for Atmospheric Detection of H2SO4. Anyway, we added the following text to the end of the paragraph following equation 2:

“We consider these values as reasonable approximations, keeping in mind that the exact values of both $\alpha$ and $\beta$ depend on a number of variables, including the ambient temperature, pressure and relative humidity as well as the sizes of the colliding objects (ion-ion or ion-aerosol particle) (e.g. Hoppel, 1985; Tammet and Kulmala, 2005; Franchin et al., 2015).

Hoppel, W. A.: Ion-aerosol attachment coefficients, ion depletion, and the charge distribution on aerosols, J. Geophys. Res., 90, 5917-5923, 1985.

Franchin, A., Ehrbart, S., Leppä, J., Nieminen, T., Gagne, S., Schobesberger, S., Wimmer, D., Duplissy, J., Riccobono, F., Dunne, E. M., Rondo, L., Downard, A., Bianchi, F., Kupc, A., Tsagkogeorgas, G., Lehtipalo, K., Manninen, H. E., Almeida, J., Amorim, A., Wagner, P. E., Hansel, A., Kirkby, J., Kurten, A., Donahue, N. M., Makhmutov, V., Mathot, S., Metzger, A., Petäjä, T., Schnitzhofer, R., Sipilä, M., Stozhkov, Y., Tome, A., Kerminen, V.-M., Carslaw, K., Curtius, J., Baltensperger, U., and Kulmala, M.: Experimental investigation of ion-ion recombination under atmospheric conditions, Atmos. Chem. Phys., 15, 7203-7216, 2015.

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.

Thank you for pointing this out, we updated our reference list.

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.