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## ***Interactive comment on “Kinetic modeling of nucleation experiments involving SO<sub>2</sub> and OH: new insights into the underlying nucleation mechanisms” by H. Du and F. Yu***

**Anonymous Referee #1**

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General comments.

Actually the mechanism behind the particle formation via SO<sub>2</sub> + OH reaction is actually a hot topic; the work presented by the authors will help in the future discussion on it. Any way the authors should clarify some points before the article can be accepted for publication.

Specific comments.

Introduction, line 7-9, "in contrast, ion-mediated H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O nucleation has been shown to be able to contribute...";

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I don't agree with that, A recent work by M. Boy et al. (Atmospheric research, 2008) shows that ion mediate nucleation contribution to total particle formation in the same measuring site that the cited reference is less than 10%, further more, many other studies also indicate that ion mediated nucleation is not relevant in troposphere (Lovejoy et al., 2004 JGR , Eisele et al. JGR 2006, Kumala et al. Science, 2008). The authors should show these discrepancies in the manuscript.

#### Methods.

In Yu JPC 2007 the authors justify the use of a quasi-unary-nucleation (QUN) in base to the high concentration of water vapor in the system of interest. Here they use this QUN model to study ternary nucleation. They have used the same model in the case on ammonia-water-sulfuric acid assuming that the presence of ammonia only affects evaporation rates of the clusters. In this work they make the same assumptions, they assume that the only responsible of cluster growing is the sulfuric acid. But one of the works studied in this manuscript (Bernd et al 2008) suggest that other unknown specie other than sulfuric acid, is the responsible for cluster growing. So, how good valid are these assumptions in this case?

I found the description of the method in this section quite confusing, authors should explain eq(3) in more detail

#### Results.

##### 3.1 Kinetic study of nucleation experiments reported in Young et al (2008)

Again the test here is quite confusing, how you calculate ER2 and ER3? In figure 1c you fix the evaporation rate to ER2, why? Have you also try to fit it to ER3?, seems that the concentration of sulfuric acid determined using that fixed ER2 is then used in figure 1d to obtain again de evaporation rate, please clarify this point.

The same can be applied to section 3.2, how are calculated the ER in figure 3b?

##### 3.3 Thermochemistry: implications for the underlying nucleation mechanism. The cal-

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culated free energy lowering induced by the presence of the unknown specie in the case of young et al for dimer and trimer are 0.7 and 0.2 Kcal/mol, are this values big enough to be representative? Can be this differences induced by uncertainties in the estimation of evaporation rates or sulfuric acid concentrations instead of being caused by some unknown specie?

I don't see how the unrealistic values obtained for enthalpy can be a justification for proposing two different unknown splices participating in nucleation.

In line 29 the authors says that the chemical identities of the unknown species has not yet been resolved, but a recent work by Salonen et al (atmospheric research 2009) has studied different candidates to be this unknown specie, A comparison between the free energy stabilization calculated in that paper with the one proposed in this paper will be really interesting.

Technical comments.

2.Methods.

Lines 8 and 11 Yu 2006a should be 2006b

Figure 1(b) line for beta is missing in the legend.

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