Key point #1: Modeling seems to be incomplete and/or oversimplified.

We see how confusion has arisen over this topic as there are two different types of “modeling” described in the paper, each with a very different intention that also affects how complete they are with respect to detailed models in the literature. First is the SOA formation model (condensation of nonvolatile compounds; oligomerization of semivolatile compounds that have partitioned into the particle phase). This modeling is meant to illustrate the complexity of growth kinetics inside the flow tube, even though we are not able to directly measure it. Think of this model as a simulated growth experiment that we wish to interpret. Second is the model referred to in the title of the manuscript – which is a generic model to interpret flow tube growth data when we don’t know the detailed physicochemical processes and parameters needed to accurately calculate SOA formation. Hopefully, these concepts will become clearer in the remainder of this comment.

The goal of this study is to investigate how best to represent complex particle growth kinetics within a flow tube reactor. In a typical flow tube experiment, one measures the input and output conditions to determine a time-averaged measure of growth, which may be difficult to interpret if the growth kinetics change as particles transit through the flow tube. In this work, we use a simplified single-particle growth model for secondary organic aerosol (SOA) formation to illustrate how complex growth kinetics inside a flow tube can arise (Section 2). We then develop and assess a generic interpretive model to represent these complex growth kinetics when the details of SOA formation (chemical reactions on or inside the particle, molecular diffusion within the particle, etc.) are unknown (Section 5).
The SOA formation model used in this study is not meant to be a detailed chemical model of any specific system, but rather to simulate complex growth kinetics in a flow tube. The model we use is inspired by SOA formation from alpha-pinene ozonolysis since this system is so well studied in the literature. Accordingly, the essential features of this model are a surface-limited process (condensation of nonvolatile material), a volume-limited process (dimer formation in the particle phase that transforms partitioned semivolatile compounds into nonvolatile dimer products), and a range of volume-limited reaction pathways and rates relative to the surface-limited process (multiple semivolatile gas-phase mixing ratios and volatilities that influence their concentrations in the particle phase and hence the dimer formation reaction rate). In the revision, we will provide literature backup for this approach. Of course, a detailed model for alpha-pinene SOA would include a broader range of ozonolysis products and particle phase reactions along with relevant physicochemical properties, for example hindered molecular diffusivity in the particle phase – all of which would affect how much growth occurs due to dimer formation. However, such a detailed model is not needed to draw basic conclusions.

We will rewrite parts of the abstract, introduction, and modeling procedure sections to emphasize the above points about the SOA formation model. Again, please think of this as the simulated data we use to test the interpretive model discussed below.

A related change we will make is to replace the term “growth yield” with “growth factor”. It is still represented the same way in the modeling equations (i.e. a product “yield” for ozonolysis of the precursor compound), but we hope that the change of wording will further emphasize that we’re not trying to perform detailed modeling of a specific SOA reaction, but rather to define a parameter that empirically describes particle growth when no such detailed model exists.

The “modeling” referred to in the title is the second type of modeling, which is used to interpret flow tube data as described in Section 5. This interpretive model calculates a growth factor based on the particle diameter change between the inlet and outlet of the flow tube as simulated by the modeling procedure of Section 2. This interpretive model assumes that the growth factor is constant inside the flow tube. However, the SOA formation model (simulation) shows that even under simplified reaction conditions, the growth factor is NOT constant as particles move through the flow tube. So, the question arises (and is answered by the results): How closely does the growth factor from interpretive modeling correspond to the actual range of growth factors inside the flow tube?

Key point #2: Consolidation of figures will more effectively convey the significance of this work.

We plan just five figures for the revision. These are described below along with significance.

Figure 1 – This figure will consist of slight reformatting of Figures 1a and 1b in the original paper. This figure introduces the concept of growth factor in the context of particle growth under atmospheric conditions. Growth factor summarizes the net amount of the reacted precursor that goes into the particle phase during a specific time period – whether it irreversibly condenses, reversibly partitions, or irreversibly/reversibly forms dimers.

Figure 2 – This figure will consist of Figures 4a and 4b in the original paper, which simulates how particles are growing in the flow tube. Key points are: 1) one must use much higher mixing ratios than ambient conditions in order to obtain measurable growth over the short time period of the flow tube, and 2) growth factor changes over the time period of particles in the flow tube, illustrating the complexity of growth kinetics. This complexity arises from rapid partitioning of material to the particle phase when the
particle first enters the flow tube, and the changing rates of surface-limited vs. volume-limited growth processes as particles move through the flow tube.

Figure 3 – This figure will consist of Figure 6a in the original paper. The idea here is that the high mixing ratios of reactants in the flow tube cause the kinetics of particle growth (volume vs. surface-area limited) to be much different from that in the atmosphere. As the reactant mixing ratios increase, surface-limited growth begins to win out over volume-limited growth.

Figure 4 – This figure will consist of Figure 9 in the original paper. This figure illustrates how aerosol liquid water can enhance particle growth, and in particular shows that even just a few monolayers of water on the surface of an effloresced particles have the ability to substantially increase the growth rate.

Figure 5 – This figure will be a modified version of Figure 10 in the original paper. This figure compares the growth factor obtained from interpretive modeling of particle growth based on inlet-outlet change in particle diameter (Section 5) to the average growth factor associated with the simulated kinetics inside the flow tube (Section 2) as embodied by the Figures 2-4 in the revision. Interpretive modeling is found to be a robust way of describing particle growth when the changing growth kinetics within the flow tube are unknown. It can be used, for example, to empirically quantify the differences in particle growth due to changing precursor mixing ratios (Figure 3) or different amounts of aerosol liquid water (Figure 4).

Specific Responses to Reviewer 2 Comments

Comment: My main comments are: 1) model over-simplification, and 2) scientific significance.

Response: We hope that our general comments above about the SOA formation model explain why a simplified model is used i.e. to simulate the complexity of particle growth kinetics in a flow tube reactor. The reviewer goes on to list many additions that would be required to accurately model SOA formation by alpha-pinene ozonolysis. We agree that these are crucial for accurate modeling of this specific system. However, the goal is not model this system, but to provide a simulated dataset to show how well the interpretive model works (Figure 5 in the revision). The reviewer also makes reference to other more sophisticated SOA formation models, which we will reference when discussing the motivation for our simplified model.

We also hope that our consolidated group of figures along with specific conclusions gained from each will clarify scientific significance. The most important factor related to the scientific significance of this work is Figure 5 in the revision (Figure 10 in the original paper), which shows that the interpretive model used to determine the growth factor reproduces what is actually happening in the flow tube. We also note that the interpretive model can be applied to any system, and we will discuss this more clearly in the revision. In an actual flow tube experiment, one knows (measures) basic experimental parameters – precursor mixing ratios, residence time in the flow tube, inlet and outlet particle size distributions. These measurements plus knowledge of the gas-phase rate constant for precursor + oxidant reaction are all that are needed for the interpretive model.

Comment: I suggest the authors reduce the technicality (where possible) and length of the text (making use of a supplement), reduce the number of figures in the main
manuscript to approximately five at most.

Response: Thank you for this suggestion. We will do this as discussed in the general comments above for the five figures in revision.

Comment: Finally, noting the title, "modelling ultrafine particle growth based on flow tube reactor measurements". Please include the measurement data and a brief description of the flow tube in the manuscript.

Response: Based on reviewer comments, we will change the title of the paper in revision. A more descriptive title might be: "Representing Complex Particle Growth Kinetics Within a Flow Tube Reactor". We feel there is too much detail required to discuss actual measurement data. We will point the reader to experimental papers, and we will emphasize in this paper that the intent of this paper is to investigate the robustness of the interpretive modeling approach.

Response to specific comments: Modifications to lines 6 and 92 will be made. Figure 3 of the original paper will no longer appear in the revised paper.