

# Response to reviewer #1

We thank reviewer #1 for careful reading our manuscript and for the positive feedback as well as constructive comments on our work. Below we address first the general and then the minor comments. The reviewer's comments are in **blue**, our responses are in **black** and any additions to the manuscript are in **red**

## General comment 1

The manuscript is relatively technical and method oriented. For non-experts within the field it can be hard to understand the possibilities and limitations with the described methods. I suggest that you add more discussion about the possibilities and limitations with the described methods in the summary and conclusion section. One limitation which I think should be discussed a bit more is the uncertainties in the observations. On line 265-266 you write that you assume a 1 % uncertainty in the measured evaporation factor (EF), I think this uncertainty can be substantially bigger. How sensitive is the optimization of all parameter values to the uncertainties in the observations? I.e. what happen if the uncertainty of the measured EF is 5 %?

## Response

The question of uncertainties and their influence to the method is an interesting one. In real evaporation measurements the error may be larger than 1%. As the uncertainties vary with experimental setting and may sometimes be challenging to estimate, we concentrated here on the performance of the method in an idealized case.

In addition, we also tested the methods with data set where random noise was introduced to the EF and time. On average the EF error was 1% and  $\pm 10$  min in time for long residence time (time > 10 min data points) points. For short residence time data points (time < 10 min data points) the random error in time was 9%. For the 10 minute data point the relative error in time was +77% / -63%. The results of fitting the LLEVAP model with both schemes showed similar results as the data set with no uncertainty in the particle radius / EF and hence we decided not to include it in the manuscript. The results of this new data set are summarized in Table 1.

**Table 1:** Estimated parameter distributions and correct values for an artificial data set where random noise was introduced to the data set.

Variable	Correct values	MCMC	MCGA (Gaussian)	MCGA (uniform)
$C_{\text{sat},1}$ ( $\mu\text{g m}^{-3}$ )	0.01	0.008 – 0.016	0.007 – 0.02	0.006 – 0.014
$C_{\text{sat},2}$ ( $\mu\text{g m}^{-3}$ )	1	0.86 – 1.44	0.78 – 1.86	0.79 – 1.37
$C_{\text{sat},3}$ ( $\mu\text{g m}^{-3}$ )	1000	567 – 2488	588 – 1530	309 – 6708
$X_{\text{mole},1}$	0.5	0.48 – 0.53	0.49 – 0.56	0.48 – 0.53
$X_{\text{mole},2}$	0.4	0.38 – 0.43	0.35 – 0.42	0.38 – 0.43
$X_{\text{mole},3}$	0.1	0.06 – 0.12	0.07 – 0.11	0.07 – 0.11

## General comment 2

Now you use the term “correct” values when you talk about measured and/or literature data on particle phase density, individual compound saturation concentrations and viscosity. But can we really state that we know the correct values of all these parameters? I would avoid using the term

“correct” or at least clearly define what you mean with “correct” parameter values.

### **Response**

We have replaced the mentions of correct values to literature values when referring to literature values. We are now using the term correct value only when referring to the artificial data sets or the mass / mole fractions in mixtures 1-4 where the estimated parameters have “correct” values in the sense that they are measured or used as an input to a process model to produce the artificial data.

### **General comment 3**

It would also be good to add a short discussion about how the described methods can be used in order to suggest/design new experiments which can give better constraints on the volatility and viscosity properties of SOA particles. Recommendation of new experiments is always welcomed by experimental aerosol scientists.

### **Response**

We have added following discussion on this at the end of conclusions.

Based on the analysis shown here, various parameters can be obtained from experimental data using this method and the design of the experiments can be used to focus experiments on properties of interest. For instance, in order to distinguish between saturation concentrations of low-volatile compounds, small particles and hours long evaporation times are required. On the contrary, to distinguish between semivolatile or intermediate volatility compounds, larger particles and/or high sampling time resolution for the short evaporation time scales are needed.

### **Minor comment 1**

When I read section 3, L250-285 I start to wonder what artificial parameter values of  $C^*$  and the mole fractions did you actually use in dataset 1-4? Then later when you start to describe the results it become clear to me that these values can be found in figure 2-5. Still, I think it would be good to have a table which provide these values already in Sect. 3. In this table you can also include the best optimization values for these parameters and the 10 th and 90 th percentiles. With this table include you do actually not need all the figures.

### **Response**

We thank Reviewer #1 for this suggestion. We have added new Table 3 which shows the correct values for  $x_{\text{mole}}$  and  $C_{\text{sat}}$  for artificial data set 1-4. The old Table 3, Table 4 and Table 5 are now Table 4, Table 5 and Table 6, respectively.

### **Minor comment 2**

In Table 1 and Table 4 some field are empty, but I guess it should be saying e.g.  $X_{\text{mole,dry}}(t=0)$  “Fitted(min: 0.01 max:1)” and T(K) 293 in all fields. I suggest that you write out explicitly in each column these values, even if they are the same in all experiments/simulations.

## **Response**

Thank you for pointing this out. We have changed Table 1 and Table 4 to explicitly show what these parameters are for each data set or mixture

### **Minor comment 3**

L424-425: You use the term “non-viscous” and “viscous” when talking about mixtures with low-viscosity and high-viscosity. I would recommend that you use the late terms instead and possible specify that the particle-phase mass transfer limitation was negligible in the low-viscosity mixture and hence neglected

## **Response**

We agree with reviewer that using the term “non-viscous” is misleading as every particle has some viscosity. We have changed the text so that non-viscous was changed to “low viscosity” and viscous to “high viscosity”. We have also added an explanation of these terms to Sect. 5, line 440 in the revised version.

In the following discussion, “low viscosity” refers to particles where particle phase diffusivity is fast enough so that it doesn’t pose a limitation to the evaporation of the volatile compounds and “high viscosity” refers to particles where the particle phase diffusivity is low enough to affect the particles’ evaporation rate.