

# Advanced modeling of gas chemistry and aerosol dynamics with SSH-aerosol v2.0

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## Reply to reviewer 2

*This manuscript describes the development and updates of the model SSH-aerosol, which can be utilized to simulate the formation of secondary aerosols, and evolution of both the primary and secondary aerosols. The manuscript is well written, with the supplement/guide serving as a detailed blueprint for potential users to test and implement the model. The SSH-aerosol model serves as an interesting tool to simulate aerosol dynamics under different environmental scenarios, and would surely benefit the modelling and experimental community. I recommend the publication of this manuscript.*

*This manuscript describes the development and updates of the model SSH-aerosol, which can be utilized to simulate the formation of secondary aerosols, and evolution of both the primary and secondary aerosols. This involves simulating the gas- and particle phase heterogeneous chemistry and aerosol dynamics. Various test cases ranging from aerosol dynamic processes and gas-particle phase chemistry were simulated and presented in the manuscript to show model capabilities. The manuscript is well written, with the supplement/guide serving as a detailed blueprint for potential users to test and implement the model. The SSH-aerosol model serves as an interesting tool to simulate aerosol dynamics under different environmental scenarios and would surely benefit the modelling and experimental community. I recommend the publication of this manuscript, after the authors have addressed the following comments/suggestions.*

## General comments:

*I would recommend adding a figure outlining the different inputs and modules and their workflow. This could also serve the purpose of being a visual reference for any user trying the model.*

**Our reply:** A figure describing the workflow of the model has been added in the section about the model structure. Further-

more, a list of the different model input parameters has been added to the Appendix.

*Add figure letters (e.g. Figure4, a, b or c) to avoid confusion.*

**Our reply:** Figure letters have been added.

25

*While discussing results (e.g. L 303-306, Figure 8 etc.), the authors states what the results shows, but don't discuss why that's happening. The readers will appreciate if they get to know for e.g., why SOA yields are increasing/decreasing in the presence of NOx.*

**Our reply:**

30

*Nucleation: This section has not been discussed in depth but rather just glossed over.*

**Our reply:** To strengthen the section on ultrafine particles, more details about the treatment of coagulation have been added at the beginning of section 5.

"For coagulation, the discretized equations governing aerosol dynamics involve partition coefficients that account for the possibility that the coagulation of particles from two given size sections may produce particle sizes spanning multiple sections. The size mesh is assumed to remain fixed over time, so that these partition coefficients can be pre-computed. In previous model version, the coagulation partition coefficients were calculated using a Monte-Carlo approach. .... Because the explicit formulation has negligible computational cost compared with the Monte Carlo approach; therefore, only the explicit formulation is retained for internally-mixed particles in v2.0."

40 More details on the size distribution are also added:

"Given the strong impact of coagulation on the growth of ultrafine particles, it should always be included in their modelling. Consequently, the particle-diameter limits are held constant over the course of the simulations. Because nucleation corresponds to the formation of nanometric particles from gas precursors, the lowest particle diameter bound of the size distribution should be about 1 nm."

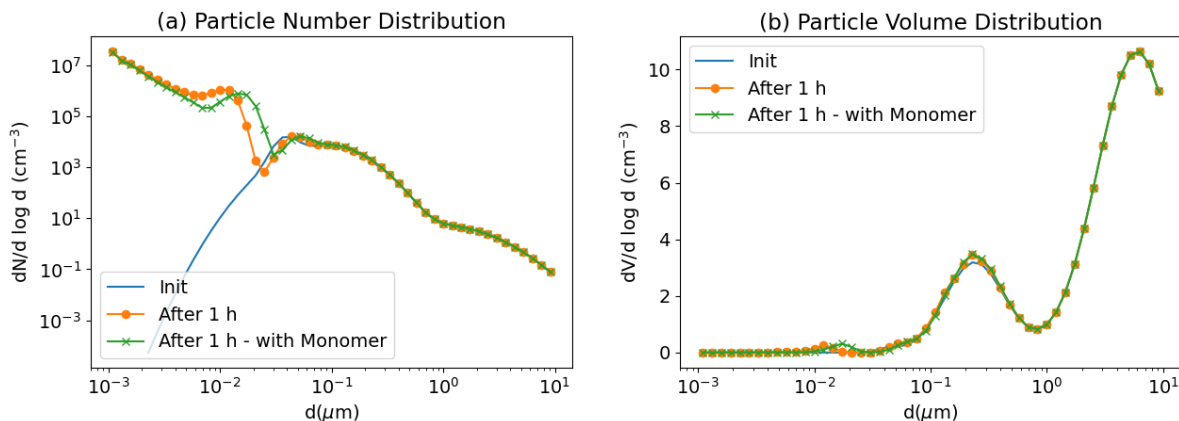
45 In the nucleation section, the following explanations are also added: "The nucleation parametrizations differ in the gas precursors (sulfuric acid, ammonia, extremely-low volatile organic compounds) and/or in the formulation of the parameterization. ", as well as the formula for organic nucleation:

$$J = scal_{org} [Organics]^{nexp_{org}}. \quad (1)$$

Nucleated particles are assigned to the first size bin, whose lower bound should be set to about 1 nm to represent freshly formed clusters. The composition of the nucleation-size bin is determined by the precursor species involved in nucleation, and it is estimated based on the properties of these precursors."

A section describing the growth of ultrafine particles has also been added. "To illustrate the growth of nucleated nanoparticles, the nucleation test case presented in Sartelet et al. (2006) is simulated. The initial distribution corresponds to the hazy

55 conditions of Seigneur et al. (1986). The sulfuric acid production rate is  $0.825 \mu\text{g m}^{-3} \text{h}^{-1}$ , the temperature is 288.15 K and the relative humidity is 60%. The particles are initially assumed to be made of 70% sulfate and 30% ammonium. The initial gas phase ammonia concentration is taken to be  $8 \mu\text{g m}^{-3}$ . The concentrations of gas-phase ammonia and particulate-phase ammonium evolve with time due to both condensation/evaporation and ternary nucleation. The simulation is run for 1 h with output every 60 s. Fig. 1 shows the size distribution of particles at the initial time and after 1 h. The nucleated particles grow under the effect of condensation of sulfuric acid and ammonia and coagulation. This growth can be accelerated by the presence of extremely low-volatility organic compounds (ELVOCs), such as the SSH-aerosol surrogate "Monomer", underlying the importance of an accurate modelling of the formation of ELVOCs for the growth of ultra-fine particles. "



**Figure 1.** Size distribution of particles in the nucleation test case. Under hazy conditions, nanoparticles nucleated from water, sulfuric acid and ammonia grow to larger size. A simulation adding ELVOCs (Monomer) in the initial conditions is performed to illustrate their effect on the growth of ultrafine particles. Number (left panel, a) and volume (right panel, b) concentrations.

*Model computational time: How much impact does different configurations (gas+SOA schemes) have on the computational time. Since this model can be couple to a 3-d model, how realistic is it to use particle phase and viscosity calculation with the coupled 3-d model since its includes discretised particle layer calculations.*

**Our reply:** A table on the model computational time has been added and discussed. While being very time consuming, accounting for diffusion inside layers is doable as a first study on considering viscosity into 3D was performed by Kim et al. (2019).

70 **Minor comments:**

*L 35: “atmospheric chambers”- I believe the correct terminology would be atmospheric smog chambers.*

**Our reply:** Yes, replaced

*L 43: “Several aerosol box models exist”. I think this line is unnecessary. Either remove it or club it with the next line.*

75 **Our reply:** The sentence has been replaced by "Several aerosol box models exist have been developed, and most represent aerosol dynamics using a sectional approach..."

*L56-57: One must be careful in formulating such descriptions. MCM is an explicit gas phase oxidation mechanism, not a SOA formation scheme. It is coupled to a SOA formation scheme and therefore should be categorized as a chemical scheme and not a SOA scheme.*

80 **Our reply:** The sentence starting by "For the formation of secondary organic aerosols,..." has been replaced by "For the formation of organic oxidation products that could be semi-volatile and partition to form secondary organic aerosols (SOAs), ...".

*L65-66: Some examples of different reduction strategies could be included.*

85 **Our reply:** Examples of reduction strategies have been added (lumping, replacing, jumping, removing).

*L89: “repertories” to repositories*

**Our reply:** "repertories" has been replaced by "directories"

90 *L 99: what kind of solver is used to deal with the ODE systems? How will it affect the computational time?*

**Our reply:** A subsection has been added in section 2 to detail the numerical implementation. A reference to this subsection is added here.

*L 110: How are the saturation vapor pressures generated? Nannolal/Evaporation/SIMPOL or another mechanism?*

95 **Our reply:** In SSH-aerosol v2.0, saturation vapor pressures are input data, they can be generated by various methods, such as those described by the reviewer. For clarity, the following sentence is added to the text: "The saturation vapor pressures are provided as model inputs and may be derived through different methods or parameterizations, including tools such as UMan-SysProp (Topping et al., 2016)."

100 *L211 – 213: does this mean that one can use 2 or more chemistry schemes at the same time? For example, one can use CB05 for estimating a reaction products conc., say X and another explicit scheme to use the [X] in their respective chemical schemes? If so, does it only work for first generation oxidation products or more complex oxidation products as well? Are these conc. profiles interpolated to the timestep of the SOA chemical scheme? L214-216: This part is unclear. Does it mean*

one can have a specific combination for each precursor + SOA scheme? E.g. alpha-pinene using MCM + beta caryophyllene  
105 using RACM2? if so how would one account for example RO2 from ap + RO2 (bcarp) reaction products from different chemical schemes, in case the resulting RO2 is not present in either scheme?

**Our reply:** For clarity this paragraph was rewritten: "Depending on the user's settings, oxidant concentrations can either be prescribed using constant input profiles, or they can be computed using existing gas-phase chemistry mechanism, such as CB05, RACM2 or MELCHIOR2. These schemes can be complemented with SOA chemical schemes describing the degradation of  
110 VOCs leading to SOA formation. To offer flexibility, the model allows users to construct customised chemical mechanisms, covering both gas-phase and SOA chemistry, by combining a chosen gas-phase mechanism with one or more SOA schemes (typically one per SOA precursor class). For VOCs already represented explicitly in the selected gas-phase mechanisms (e.g. toluene, monoterpenes), the associated SOA scheme can be added without modifying oxidant concentrations. However, some VOCs are not included in these gas-phase mechanisms, in which case the inclusion of specific reactions for radicals, oxidants  
115 as well as SOA formation may be necessary. For example, the impact of naphthalene on ozone and radical production is not represented in the CB05, RACM2 or MELCHIOR2 mechanisms."

*L223-224: There are different vapor pressure estimation methods on the UManSysProp. Which method was used?*

**Our reply:** The following was added "often using the vapor pressure estimation methods ('v0' (Myrdal and Yalkowsky, 1997)), with the boiling point estimation methods('b0' (Nannoolal et al., 2004))".

120

*L234: is this analogous to "lumped RO2" species? Is this RO2 pool rereferring to RO2 from one precursor alone or can it refer to lumped RO2 from all of MCM or a subset of precursors?*

**Our reply:** "that include RO<sub>2</sub> from all precursors" is added after RO<sub>2</sub> pool.

125 *L 236: what does "ARR" stand for?*

**Our reply:** ARR indicates it is an Arrhenius type of reaction. This has been added to the text.

*L 243: "TBRO2" is not referenced in the equation above or explained anywhere else.*

**Our reply:** This sentence has been removed.

130

*Figure 2: This makes sense since MCM doesn't yet include a fully developed peroxy radical autooxidation scheme for beta caryophyllene. It would be interesting to perform such tests with apha-pinene.*

**Our reply:** We agree with the reviewer that  $\alpha$ -pinene is a valuable reference system for exploring RO<sub>2</sub> chemistry, particularly under realistic atmospheric conditions where multiple precursors contribute.

135

*Section 3.2 My one suggestion here to improve readability would be to have a table detailing the explicit and reduced mechanism, with a shorthand representing each. For e.g near explicit mech for monoterpenes could be denoted by Expmcm+pram or something like that. It could make it easier for readability without one having to go back and forth to see what the respective*

Precursor	Mechanism name	Type	Reference
Naphthalene	H <sup>2</sup> O	Implicit	Couvidat et al. (2013)
	Expl.	Quasi-explicit	Lannuque and Sartelet (2024)
Toluene	H <sup>2</sup> O	Implicit	Couvidat et al. (2012)
	Expl.	Quasi-explicit with ipso-BPR molecular rearrangement	Lannuque et al. (2023) + Sartelet et al. (2024)
	Rdc	GENOA Reduced	Sartelet et al. (2024)
Monoterpenes	H <sup>2</sup> O	Implicit	Couvidat et al. (2012)
	Expl.	Quasi-explicit (MCM + PRAM)	Roldin et al. (2019); Wang et al. (2023)
	Rdc	GENOA Reduced	Wang et al. (2023)
$\beta$ -caryophyllene	H <sup>2</sup> O	Implicit	Couvidat et al. (2012)
	Expl.	Quasi-explicit (MCM)	-

**Table 1.** SOA chemical mechanisms used in the test cases of section 3.2

*mechanisms were.*

140 **Our reply:**

The following table (Table 1) has been added to section 3.2:

*L281: This should be right panel.*

**Our reply:** Yes, thank you.

145 *L 294: upper right panel?*

**Our reply:** Yes, modified.

*L296-298: Referring to Fig 4: these differences are barely visible in the plot. Perhaps the authors will think about providing percentage increase or decrease w.r.t the reference to indicate the difference.*

150 **Our reply:** The sentence "They are also lower than the reference with higher NO<sub>2</sub> levels in the first 1.5 hours of the simulation but higher after that, in opposition to the simulation with the near-explicit scheme." corresponds to small variations that are very meaningful and hence it has been removed.

*L 299-303: why is that?*

155 **Our reply:** For naphthalene, the difference between the concentrations simulated with the near-explicit and the H<sup>2</sup>O scheme reflect large uncertainties in the H<sup>2</sup>O scheme, which was built by (Couvidat et al., 2013) from a specific set of experiments from (Chan et al., 2009) that are not representative of the full range of atmospheric conditions. Even though these experiments were corrected from wall-loss effects of particles, they were not corrected for gas wall losses. Furthermore, the behaviour of the H<sup>2</sup>O scheme may not fully extrapolate to all experimental setups, especially flow tubes where residence times are much

160 shorter than in smog chambers. This was added in the text.

*Figure 3: what is the reference scheme? it should be mentioned in the caption. Is the related to NO<sub>2</sub> conc.?*

**Our reply:** The following has been added in the caption: (Ref. denotes the reference NO<sub>2</sub> levels, NO<sub>2</sub> x 2 the simulation with doubled NO<sub>2</sub>, and NO<sub>2</sub> / 2 the simulation with halved NO<sub>2</sub>).

165

*L 364-365: Since H<sub>2</sub>O is based on smog chamber experiments, was wall losses considered when simulating SOA for species which was compared to H<sub>2</sub>O? I suspect the SOA yields for species where H<sub>2</sub>O was used for comparison would differ if wall losses of organics would be considered.*

**Our reply:** H<sub>2</sub>O was built using smog chamber experiments after correction of wall losses for particles, but generally these  
170 corrections do not account for wall losses of gases. Wall losses for gases and particles are considered in the simulations performed in the current study, independently of the scheme used.

*L 378-380: why does the H<sub>2</sub>O mechanism overestimate the SOA mass?*

**Our reply:** Each precursor-specific H<sub>2</sub>O mechanism was developed using a limited number of chamber experiments performed  
175 under particular conditions. Its behaviour may therefore not fully extrapolate to all experimental setups, especially flow tubes where residence times are much shorter than in smog chambers. This difference in conditions may contribute to the apparent overestimation of SOA mass.

*L398: “interface the index of the layer at the interface”. what does this represent or mean? Also K<sub>bin, interface, p,i</sub> and the  
180 corresponding Mo<sub>bin,interface</sub> layer is not defined.*

**Our reply:** Quotes were missing. The phrase should be read like "interface" the index of the layer at the gas/particle interface. Layer has to be replaced by interface for the values of variables at the interface

*L 413: “f”. This needs more explanation here. For e.g., what does a value f=0.2 indicate? how is f determined?*

**Our reply:** f is a weighting factor between two iterations. The algorithm is presented into greater details in Couvidat and  
185 Sartelet (2015). Please refer to Couvidat and Sartelet (2015).

*L 430: “Compounds not affected to a specific molecular structure”. what does this mean?*

**Our reply:** The sentence was deleted.

190

*Figure 9: I assume that since this is an organic phase there would be no charged compounds would be present in the particle phase. But when the RH increases compounds in the particle phase are bound to break and form cations and anions. How will this affect the viscosity especially in the presence of acids?*

**Our reply:** Dissociation of acid in the organic phase is currently not taken. The implementation of this process will be consid-

195 ered for future version.

*Figure 9: Add panel letters to make it easy to follow. how does the c panel which is bottom left change if you have varying RH for alcohol+acid + varying RH?*

**Our reply:** Panel letters added. As there are no particular treatment for acids on viscosity, the effect of humidity will be similar  
200 to panel b. The increase of RH will lead to a faster condensation.

*L480: parameter k. Is this a function of water conc. and pH?*

**Our reply:** The text now states: "This type of reaction can account for catalysis by water (reaction rate multiplied by the activity of water) and pH (reaction rate multiplied by pH)."  
205

*L 505: Kmaxoligo , hoe is this determined?*

**Our reply:** As mentionned later, all parameter for the bulk oligomerization parameterization are coming from Couvidat et al. (2018a). In this study, the parameters were determined by running a 0D model and were fitted in order to reproduce the evolution of observed molar masses of oligomers during an experiment.  
210

*L505: amonomer, I would suggest representing this as Eta or sum(aA, monomer), both in the equation 24 and here.*

**Our reply:**  $a_{monomer}$  was replaced by  $\sum_i a_{i,monomer}$ .

*Eq. 28: is this nomenclature based on H2O or MCM or other chemical schemes?*

215 **Our reply:** This nomenclature was specifically chosen for SSH-aerosol.

*L 587: SOAP. Does the model account for the formation of salts in the particle-phase?*

**Our reply:** The text now states: "Inorganic aerosols are assumed to be thermodynamically metastable (inorganic compounds are always present in the aqueous phase of particles and no solid salts are formed) with the exception of  $\text{CaCO}_3$  that is assumed  
220 to have a very low solubility."

*Figure 13: please improve the legend. It's hard to read it in its current form.*

**Our reply:** The figure has been separated into two subfigures (see Fig. ?? in the response to comments from Reviewer 1).

225 *Nucleation: Have the authors considered to couple ACDC for more complex termolecular acids and bases?*

**Our reply:** Yes that would be interesting for a better representation of nucleation, and we will consider it in further version of the model.



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