To the editorial team,

We discovered model errors after the revision had been submitted and have made additional changes to the manuscript. The editor, Prof. Allen Bertram, and editorial support Lisa Appel had been informed of those changes, and the editor approved those edits on 2023-10-03. Thus, small portions of the text and figures would need to be updated. Lisa Appel informed us that the best option would be to apply those approved changes during the article proofing, which we now would like to do. In addition to the copy editor and typesetter comments, we have summarized the final changes below. Our comments are in **blue**, the original text in **black**, and the changes in **red**. We also request that the supplementary be replaced with the one that is attached.

Thomas Berkemeier and Hwajin Kim

**Additional changes**

**Abstract**In the proofreading version, we would like to edit the following sentence in the abstract:

“The model uses a volatility basis set (VBS) of the primary oxidation products as well as an aging rate coefficient in the gas phase, *k*age,gas, of 2.2×10-12 cm3 s-1 and an effective aging rate coefficient in the particle phase, *k*age,particle, of 2.0×10-12 cm3 s-1.”

**Page 5, line 27**  
Please adjust the sentence to the one highlighted below.

“This decrease in volatility via “bin-hopping” (Sommers et al., 2022) occurs at a rate proportional to the chemical aging rate coefficient for the gas-phase (*k*age,gas, cm3 s-1) and an effective aging rate coefficient for the particle-phase species (*k*age,particle , cm3 s-1).”

**Page 5, line 47**  
Please add the highlighted sentence after this sentence.

“The timescales and atmospheric relevance of heterogeneous oxidation in OFRs are areas of ongoing research (Zhao et al., 2019; Peng and Jimenez, 2020), but for now we opt to fit chemical aging rate coefficients for each phase. Note that surface and bulk accommodation processes are not explicitly resolved in the kinetic model and *k*age,particle acts on the concentrations of particle-phase products per unit gas volume (Sect. S1.6).”

**Page 6, line 3**  
Please adjust the values in the sentence to the ones highlighted below.

“We obtain a best model fit and a fit ensemble consisting of 1059 parameter sets for which the model’s root mean square error (RMSE) is below a threshold of 50.”

**Page 9, line 44**  
Please adjust the values in the sentence to the ones highlighted below.

“We fit *k*age,gas and *k*age,particle in the aging-VBS model to be 2.2 × 10-12 cm3 s-1 and 2.0 × 10-12 cm3 s-1respectively.”

**Page 9, line 94-107**  
Please adjust the paragraph to the one below.

“We find that the model is sensitive to *k*age,particle,and a larger rate coefficient results in higher SOSiA formation (Fig. S11a). When fitting the model with deactivated particle-phase aging (*k*age,particle = 0), model-experiment RMSE is slightly increased (Fig. S11b). Thus, the fitting process provides a weak constraint on the parameter value (Fig. S12c). The numerical value of the fitted *k*age,particle is physically reasonable as it corresponds to an effective uptake coefficient of OH molecules colliding with the particle surface in the range of 0.1–1 (Sect. S1.6). On the other hand, *k*age,gas is very influential on the model output and tightly constraint in the ensemble of model fits around a value of 2 × 10-12 cm3 s-1 (Fig. S12b). We hence postulate that multi-generational aging of in the gas phase is likely an important process for SOSiA formation, while particle-phase aging may only play a minor role under the investigated conditions.”

**Page 11, line 4**  
Please adjust the values in the sentence to the ones highlighted below.

“Figure S8 shows that the aging-VBS model used here leads to a much higher correlation between modeled and experimental values for SOSiA mass concentration compared to the same analysis with a standard-VBS model (RMSE=189 vs. 378).”

**Page 12, line 33**  
Please adjust the sentence to the one highlighted below.

“We find that the aging-VBS model is very sensitive to *k*age,gas (Fig. S12), suggesting that photochemical aging in the gas phase should be considered in these models.”

**Updated Table 1**Please adjust the *Y*SOSiA values to the ones highlighted below.

“Table 1. Summary of SOSiA mass yields (*Y*SOSiA) with aerosol sampling line corrections assuming *ρ*SOSiA = 1.07 g cm-3 for all experiments. [H2O] is the molar mixing ratio of H2O in air. For COA and [D5], the errors are the standard deviation of the data points averaged, while for *Y*SOSiA, they are calculated with error propagation. For reference, at 25 °C and 1 atm, 1 ppb of D5 is ~15 µg m-3 and one day equivalent of OHexp is ~1.3 × 1011 s cm-3 at a daily [OH]avg of 1.5 × 106 cm-3.”

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Experiment | *Y*SOSiA  (%) | [H2O]  (%) | COA  (µg m-3) | OHexp  (s cm-3) | [OH]  (cm-3) | [D5]0  (ppb) | 1 – [D5]final/[D5]0 |
| 1 | 5.4 ± 0.9 | 0.892 | 10.5 ± 0.7 | 1.73 × 1011 | 9.59 × 108 | 43.4 ± 1.3 | 0.292 |
| 2 | 4.6 ± 0.6 | 0.828 | 19.0 ± 0.6 | 1.90 × 1011 | 1.06 × 109 | 85.7 ± 2.5 | 0.316 |
| 3 | 3.1 ± 0.6 | 0.742 | 17.7 ± 0.5 | 1.26 × 1011 | 6.99 × 108 | 165.8 ± 4.5 | 0.222 |
| 4 | 18.3 ± 1.5 | 1.95 | 75.2 ± 1.9 | 4.66 × 1011 | 2.59 × 109 | 44.0 ± 1.7 | 0.606 |
| 5 | 28.0 ± 2.7 | 2.06 | 179.2 ± 3.1 | 3.80 × 1011 | 2.11 × 109 | 78.3 ± 3.2 | 0.532 |
| 6 | 25.5 ± 1.8 | 2.09 | 286.2 ± 7.1 | 3.12 × 1011 | 1.73 × 109 | 157.8 ± 3.6 | 0.464 |
| 7 | 8.0 ± 0.5 | 0.733 | 36.8 ± 1.3 | 5.76 × 1011 | 3.20 × 109 | 43.8 ± 1.3 | 0.684 |
| 8 | 17.8 ± 1.7 | 0.736 | 118.6 ± 5.6 | 4.00 × 1011 | 2.22 × 109 | 78.9 ± 3.2 | 0.550 |
| 9 | 21.1 ± 1.1 | 0.797 | 304.5 ± 2.8 | 4.19 × 1011 | 2.33 × 109 | 166.8 ± 4.1 | 0.567 |
| 10 | 38.0 ± 2.2 | 1.93 | 212.9 ± 8.1 | 9.01 × 1011 | 5.00 × 109 | 43.8 ± 1.4 | 0.835 |
| 11 | 45.6 ± 1.9 | 2.08 | 420.2 ± 3.0 | 7.78 × 1011 | 4.32 × 109 | 76.5 ± 2.2 | 0.789 |
| 12 | 52.3 ± 2.4 | 2.15 | 965.7 ± 25 | 7.39 × 1011 | 4.10 × 109 | 156.9 ± 3.9 | 0.772 |
| 13 | 4.2 ± 1.7 | 0.712 | 3.9 ± 0.3 | 8.70 × 1010 | 7.25 × 108 | 37.9 ± 1.6 | 0.160 |
| 14 | 1.7 ± 0.4 | 0.718 | 4.1 ± 0.3 | 1.09 × 1011 | 9.10 × 108 | 80.8 ± 2.3 | 0.196 |
| 15 | 1.0 ± 0.3 | 0.704 | 3.7 ± 0.7 | 8.29 × 1010 | 6.91 × 108 | 162.8 ± 4.9 | 0.153 |

**Updated Fig. 4**Please replace the previous version of Fig. 4 with the updated version.

A collage of graphs and diagrams

Description automatically generated

**Previous**

**Version**

A collage of graphs and diagrams

Description automatically generated

**Updated**

**Version**

**Updated Fig. 5**Please replace the previous version of Fig. 5 with the updated version.

A graph of a number of days

Description automatically generated with medium confidence

**Previous**

**Version**

A graph of a number of numbers and a number of days

Description automatically generated with medium confidence

**Updated**

**Version**

**Updated Fig. 6**Please replace the previous version of Fig. 6 with the updated version.

A group of graphs with different colored lines

Description automatically generated with medium confidence

**Previous**

**Version**

A group of graphs with different colored lines

Description automatically generated

**Updated**

**Version**

**Response to copy-editor comments**

**CE1**  
The color pink is correct. In Fig. 1, the molecules marked with blue and pink (now purple) are multi-functional, while red and orange are mono-functional.

**CE2**   
We confirm using “siloxanolyl” to indicate the formate is attached to a siloxanol to form an ester.

**CE3**  
Here, the slash in “VOC/NOx” means the ratio of VOC over NOx. To keep this nomenclature consistent, we request the following changes:

Page 2, line 61. Change “low-NO/low-HO2 conditions” to “low-NO/HO2 conditions”  
Page 8, line 18. Change “low-NO/low-HO2 conditions” to “low-NO/HO2 conditions”  
Page 12, line 10. Change “dilution/removal” to “dilution and/or removal”

**CE4**  
“MIR” is the maximum incremental reactivity. Since this term appears only here, the abbreviation is not necessary. We request that the sentence be modified to the following:

“By multiplying the maximum incremental reactivity with the HCHO reacted with OH, we can estimate an O3 formation potential of 8 ppt from D5 in urban air.”

**CE5**  
We the approve the changes made to Appendix A.

**CE6**  
We approve this change.

**CE7**  
The full name of “BO” is Byeonghun Oh.

**Response to typesetter comments**

**TS1**  
We have attached the revised figure files, including that for Fig. 5. However, we are unsure what compositional element of this figure had changed from the one that was submitted and request clarification.

**TS2**  
The citation for Gkatzelis et al. (2021) is below:

Gkatzelis, G. I., Coggon, M. M., McDonald, B. C., Peischl, J., Aikin, K. C., Gilman, J. B., Trainer, M., and Warneke, C.: Identifying Volatile Chemical Product Tracer Compounds in U.S. Cities, Environ. Sci. Technol., 55, 188–199, https://doi.org/10.1021/acs.est.0c05467, 2021.

**TS3**  
Yes, the “3” for “O3” and “NO3” in *k*D5+O3 and in *k*D5+NO3 should be a subscript. Similarly, the “5” for “D5” should also be a subscript. We request that instances of *k*D5+OH, *k*D5+O3, and *k*D5+NO3 be corrected to use subscripts.

**TS4**  
For the short title, please use “VOP and SOSiA from D5 + OH.”

**TS5**  
Please see the revised figures, where a higher resolution version is available.

**TS6**  
The typesetter is correct that Equation 6 was not in the previous submission. We request that the numbering of equations also be changed in the following instances:

Page 7, line 3: “With ordinary differential equations from these reactions (Reactions R7 and R8) and experimental inputs…” should be “With ordinary differential equations (Equations 6 and 7) from these reactions and experimental inputs…”  
Page 11, line 29: “Reactions R5 and R6” should be “Reactions R4 and R5.”

**TS7**  
We have checked the format of the units throughout the manuscript and affirm they are in exponential form.

**TS8**  
The citation for McLachlan et al. (2010) is below:

McLachlan, M. S., Kierkegaard, A., Hansen, K. M., van Egmond, R., Christensen, J. H., and Skjøth, C. A.: Concentrations and Fate of Decamethylcyclopentasiloxane (D5) in the Atmosphere, Environ. Sci. Technol., 44, 5365–5370, https://doi.org/10.1021/es100411w, 2010.

**TS9**  
We request that the acknowledgements and financial support disclosure be amended to the highlighted version.

“**Acknowledgements.** This work was supported by the FRIEND Project (Fine Particle Research Initiative in East Asia Considering National Differences), which is funded by the National Research Foundation (NRF) of Korea and the Ministry of Science and ICT of South Korea (2022M3G1A1020858). This work was also funded by the NRF under NRF-2021R1A2C2004365. Hyun Gu Kang is supported by the Max Planck Graduate Center with Johannes Gutenberg University Mainz. APM Engineering (Gyeonggi-do, South Korea) lent the team the PTR-MS, and the authors thank BO for maintaining it. The authors acknowledge the two anonymous reviewers for their comments, which can be found on the EGUsphere website.”

We correct the acknowledgement as follows and remove the financial support section to avoid the redundant.

“**Acknowledgements.** This work was supported by the FRIEND Project (Fine Particle Research Initiative in East Asia Considering National Differences), which is funded by the National Research Foundation (NRF) of Korea and the Ministry of Science and ICT of South Korea (2022M3G1A1020858). This work was also funded by the NRF under NRF-2021R1A2C2004365. Hyun Gu Kang is supported by the Max Planck Graduate Center with Johannes Gutenberg University Mainz. The authors thank APM Engineering (Gyeonggi-do, South Korea) for PTR-MS renting and for maintaining it. The authors acknowledge the two anonymous reviewers for their comments, which can be found on the EGUsphere website.”

“**Financial support.** This research has been supported by the National Research Foundation of Korea (grant nos. NRF-2021R1A2C2004365 and 2022M3G1A1020858).”

Remove.

**TS10**  
We have reviewed the references and affirm that the citations are appropriate.

**TS11**  
Carter et al. (1993) is not a journal publication, but rather a report. The report can be found at https://intra.engr.ucr.edu/~carter/pubs/rct1rept.pdf (last accessed: 2023-11-04).

We have corrected the citation:

Carter, W. P. L., Pierce, J. A., Malkina, I. L., Luo, D., and Long, W. D.: Environmental Chamber Studies of Maximum Incremental Reactivities of Volatile Organic Compounds, Statewide Air Pollution Research Center, University of California Riverside, 227 pp., 1993.

**TS12**  
We have corrected the citation for Chen et al. (2023):

Chen, Y., Park, Y., Kang, H. G., Jeong, J., and Kim, H.: Chemical characterization and formation of secondary organosiloxane aerosol (SOSiA) from OH oxidation of decamethylcyclopentasiloxane, Environ. Sci. Atmos., 3, 662–671, https://doi.org/10.1039/D2EA00161F, 2023.

**TS13**  
We have corrected the citation for Millet et al. (2006):

Millet, D. B., Jacob, D. J., Turquety, S., Hudman, R. C., Wu, S., Fried, A., Walega, J., Heikes, B. G., Blake, D. R., Singh, H. B., Anderson, B. E., and Clarke, A. D.: Formaldehyde distribution over North America: Implications for satellite retrievals of formaldehyde columns and isoprene emission, J. Geophys. Res.-Atmos., 111, D24S02, https://doi.org/10.1029/2005JD006853, 2006.

**TS14**  
We have corrected the citation for Seltzer et al. (2021):

Seltzer, K. M., Murphy, B. N., Pennington, E. A., Allen, C., Talgo, K., and Pye, H. O. T.: Volatile Chemical Product Enhancements to Criteria Pollutants in the United States, Environ. Sci. Technol., 56, 6905–6913, https://doi.org/10.1021/acs.est.1c04298, 2021b.

**TS15**  
This reference is an application note produced by the instrument manufacturer. The note does not have a formal list of authors and can be found at https://tsi.com/getmedia/6fe98896-89fc-45f4-822f-ab1a46c2e56a/SMPS-003appnote-A4?ext=.pdf (last accessed: 2023-11-04).

We have corrected the citation:

TSI Incorporated: Measuring Nanoparticle Size Distributions in Real-time: Key Factors for Accuracy, TSI Incorporated, Application Note SMPS-003, 7 pp., 2012.

**TS16**  
We have corrected the citation for Whelan et al. (2021):

Whelan, M. J. and Kim, J.: Application of multimedia models for understanding the environmental behavior of volatile methylsiloxanes: Fate, transport, and bioaccumulation, Integr. Environ. Assess. Manag., 18, 599–621, https://doi.org/10.1002/ieam.4507, 2021.

**Other typesetting issues**

**Graphical abstract**The image appears to have a black background.

**Page 3, line 13**The space between “*Δm*” and “(SOSiA)” is unnecessary. The same goes for “*Δm*” and “(D5)” in the line below. The same issue appears on page 4, lines 85-86.

**Page 6, line 77**The rate coefficient shown in parentheses for Eq. (R5) appears close to the reaction arrow, which is supposed to mean that VOP*i* + OH effectively removes VOP*i*. Could there be more space between the arrow and “(*k*VOPi+OH)” to prevent confusion? A similar issue can be found for Reaction R2 on page 5 and Reactions R7 and R8 on page 8.

Having the rate coefficients displayed on another line like Reaction R6 should be suitable.

**Page 12, line 66**There is a typo where “]” appears before “Abbreviations.”