Response to Reviewer #2

Zhang et al. analyze data from field measurements and chamber studies to investigate why atmospheric particle growth rates are often lower than those predicted by traditional kinetic models. They propose that nanoparticles may exist in a highly viscous phase state, resulting in limited surface-to-bulk transport and consequently reduced particle growth rates. Under this assumption, the model can successfully reproduce observations from previous field campaigns and chamber experiments. The manuscript is written very clearly and the presentation quality is outstanding.

Response: We thank Reviewer #2 for their time and effort to review our manuscript, and we highly appreciate the thorough and overall positive evaluation. Individual concerns, comments, questions, and suggestions are addressed in the following point-by-point responses, which are marked by blue font color and the label "Response" while the reviewer's comments are reproduced in black. Citations from the manuscript are displayed in indented paragraphs with changes highlighted in green font.

However, I have several major concerns regarding this work as detailed below. First, several previous studies have suggested that nanoparticles are likely to be liquid-like rather than highly viscous, which appears inconsistent with the central assumption of the manuscript. Only with indication from the modeling without experimental/observational evidence, it is hard to believe that particles would adopt a lower viscous phase state under low T. The authors do not account for coagulation nor particle wall loss, despite their potential influence on particle growth rates. These issues need to be addressed.

Response: As detailed below, we have utilized semi-empirical parameterizations of Li et al. and Kang et al. to demonstrate that SOA nanoparticles can indeed be expected exhibit high viscosity and low diffusivity under the conditions investigated in this study. Coagulation would accelerate particle growth, and wall loss could cause artifacts in the measurement of particle size distributions and condensable vapor concentrations. We have no reason to assume that the high-quality measurement data reported by Stolzenburg et al. (2025) would be subject to artifacts beyond their uncertainty estimates. For the field measurement data, Stolzenburg et al. (2025) reported "little influence of inter- and intramodal coagulation as well as diffusion-driven growth on the derived GR values making them easily comparable to our simple monodisperse growth model". For the chamber measurement data, Stolzenburg et al. (2018) reported that the "apparent growth rate of the particle size distribution does not necessarily represent the growth caused by pure condensation, as it omits coagulation and, in chamber experiments, wall losses, which both alter the particle size-distribution (6). Therefore, a systematic uncertainty of the method is estimated to be 50% (7)." Performing measurement data analyses that go deeper than the studies from the measurement data are taken would go beyond the scope of our current study, but may be addressed in follow-up studies including coagulation in KM3C provided that the necessary experimental parameters are available. This has been clarified in the revised text of our manuscript as follows.

> l. 216-220 – "Another process affecting nanoparticle growth is coagulation, which can increase measured growth rates (Cai et al., 2021, 2026). Coagulation was not explicitly treated in this study, as Stolzenburg et al. (2025) reported little influence of coagulation for the investigated field data, while Stolzenburg et al. (2018) included it into their uncertainty estimates for the investigated chamber data."

- The authors estimate extremely low diffusion coefficients (10-15 – 10-20 cm2 s-1; please show/list bulk diffusivity at different T for ambient and chamber conditions) and suggest that this is due to nanoparticles being highly viscous and nearly glassy. This is contradicting to previous work by several groups that indicate that nanoparticles should be liquid-like. a-pinene SOA formed in CLOUD was found to be viscous semisolid under low temperature

(Jarvinen et al., ACP, 16, 4423–4438, 2016); this study showed that particle viscosity is lower at lower T, which is consistent with numerous other literatures showing that particles would be more viscous under low temperature. In general, particles at room temperature are expected to be liquid for diameters below 20 nm (Cheng et al. Nat. Commun., 6, 5923, 2015). Similarly, another study has demonstrated that as SOA diameters decrease below 100 nm their glass transition temperatures decrease significantly leading to lower viscosities (Petters & Kasparoglu, Sci. Rep., 10:15170, 2020). Even if the bulk is glassy, it may have a highly mobile surface layer which is a few nanometers thick (Tian et al. Appl. Phys. Rev. 9, 011316, 2022). The fractional Stokes-Einstein relation implies that 10-20 cm2 s-1 is unlikely at higher temperature. For ambient PM in Hyytiala, bounce measurements have also indicated that small nanoparticles are less likely to bounce compared to larger particles indicating that nanoparticles are more liquid-like (Virtanen et al. Atmos. Chem. Phys., 11, 8759–8766, 2011). I understand the authors argument that more SVOC would be condensed at lower T, but it is hard to believe this effect would overwhelm temperature effect on phase state. The authors could calculate Tg based on volatility distributions presented in Fig. S3 using Tg parameterizations (even then, Tg might be suppressed for small particles).

Response: We thank the reviewer for giving us the opportunity to clarify key questions, some of which are still open and subject of ongoing investigations beyond the scope of this work. We adjusted the title of our manuscript to clarify and highlight the aspects on which our study is focused, and we extend the discussion of particle phase state and utilize state-of-the-art parameterizations to estimate molecular diffusivities as follows:

> Title – "Buffering of atmospheric nanoparticle growth by temperature-dependent shifts in molecular composition, volatility and diffusivity"

> l. 76-77 – "The diffusion properties of complex SOA mixtures have not yet been constrained with high precision, but the occurrence of different phase states and diffusivities of SOA nanoparticles in the investigated growth events is consistent with earlier studies."

> l. 83-85 – "At elevated temperatures, organic compounds need higher molecular masses and oxygen contents to reach low-enough vapor pressures for condensing and partitioning largely into the particle phase (Donahue et al., 2011; Shiraiwa et al., 2014; Bilde et al., 2015; Li et al., 2016)."

The parameterizations following Li et al. (2020) and Kang et al. (2026) are described in detail in the revised Supplementary Materials (Sect. S7). In the main text of the revised manuscript, we now added the following discussion:

> l. 95-105 – "To obtain quantitative estimates of these effects, we employ semi-empirical parameterizations of SOA diffusivity established by Li et al. (2020) and Kang et al. (2026), which relate glass transition temperatures, viscosities and self-diffusion coefficients to the volatilities of organic compounds (Sect. S7). Inserting these parameterizations in KM3C, we can calculate diffusivities for each particle layer as a function of chemical composition and time (Kang et al., 2026). As detailed in Fig. S4, the model simulations following Li et al. (2020) predict glassy solid nanoparticle phase states with glass transition temperatures (T_g) well above 300 K and diffusivities smaller than 10^{-20} cm^2 s^{-1} throughout the investigated growth events in summer and spring. Plasticizer effects of water and sulfate were taken into account (Sect. S7; Koop et al., 2011; Dette and Koop, 2015), but did not alter the modeling results substantially. The model simulations following Kang et al. (2026) in Fig. S5 predict time-dependent and radially inhomogenous semi-solid phase states with diffusivities that are initially lower in summer than in spring, and of comparable magnitude overall (10^{-20} to 10^{-18}

$cm^2\ s^{-1}$). In both scenarios, the diffusivity increases over time as the particles grow by condensation of relatively more volatile compounds (Figs. S4, S5)."

We are aware of earlier studies addressing nanosize effects on particle phase state, and we build on the results reported by Mahant et al. (2024) from which we derive ΔT_g estimates in the range of 40-50 K for the particle sizes investigated in this study. Coupled with the parameterization of Li et al. (2020), this shift is not expected to lower T_g below ambient temperatures because very high T_g values are associated with the predominantly ultra-low and extremely-low volatility compounds condensing onto nanoparticles in the investigated growth events. This information has been added to the revised manuscript as follows:

l. 105-109 – "To explore the potential influence of nanosize effects following Cheng et al. (2015), we combined the parameterization of Li et al. (2020) with data measured by Mahant et al. (2024) and find that glass transition temperatures decrease substantially ($\Delta T_g \sim 40 - 50$ K), but not below 300 K. As the nanoparticle size effect declines towards larger particle sizes (Fig. S6), condensation of higher volatile vapors offsets this change, leading to a near-constant T_g around 320 K."

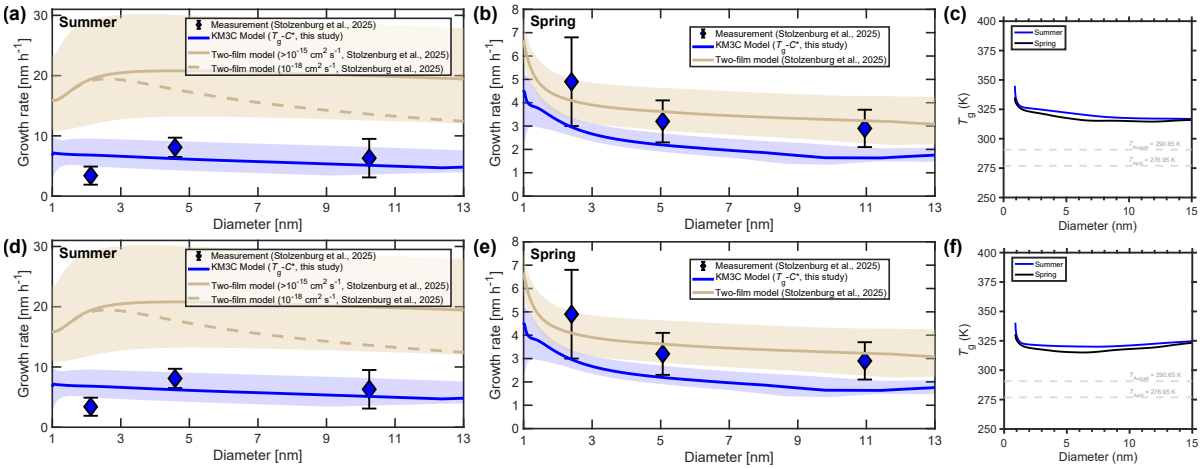

Figure S4: Observed (blue diamond markers and error bars) and modelled atmospheric nanoparticle growth rates in field experiments (Hyytiälä, Finland) in summer **(a, d)** and spring **(b, e)**, respectively. Brown line and shading represent a two-film model (Stolzenburg et al., 2025). Blue line and shading represent the KM3C model in which diffusivity is estimated following Li et al. (2020). Panels **(c)** and **(f)** show the predicted average glass transition temperature (T_g) as a function of particle size. In all the simulations shown here, T_g depression as a function of nanoparticle size following Mahant et al. (2024) is considered. In panels **(a-c)**, the water content of the SOA-H_2SO_4-water mixture is calculated using Raoult's law. In panels **(d-f)**, the water content of the mixture is calculated using an effective hygroscopicity parameter κ of approximately 0.01 as reported by Mikhailov et al. (2013) for the hygroscopic growth of ambient SOA under subsaturated conditions (RH $\sim$ 50%).

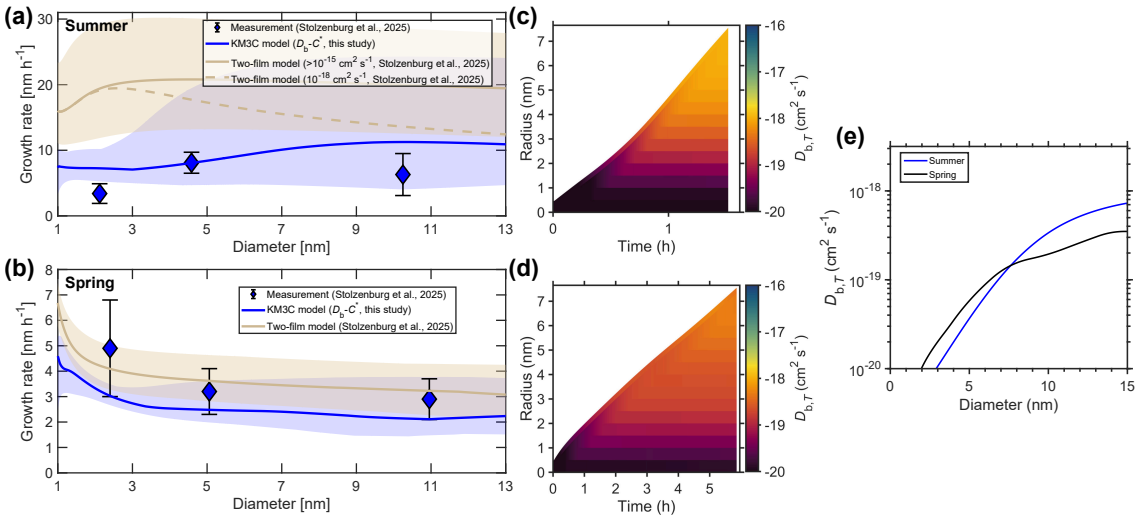

Figure S5: Observed (blue diamond markers and error bars) and modelled atmospheric nanoparticle growth rates in field experiments (Hyytiälä, Finland) in **(a)** summer and **(b)** spring, respectively. Brown line and shading represent a two-film model (Stolzenburg et al., 2025). Blue line and shading represent KM3C in this study and diffusivity is estimated following Kang et al. (2026). Panels **(c)** and **(d)** show the radial profiles of diffusivity as a function of time in summer and spring, respectively. Panel **(e)** shows the predicted average diffusivity as a function of particle size.

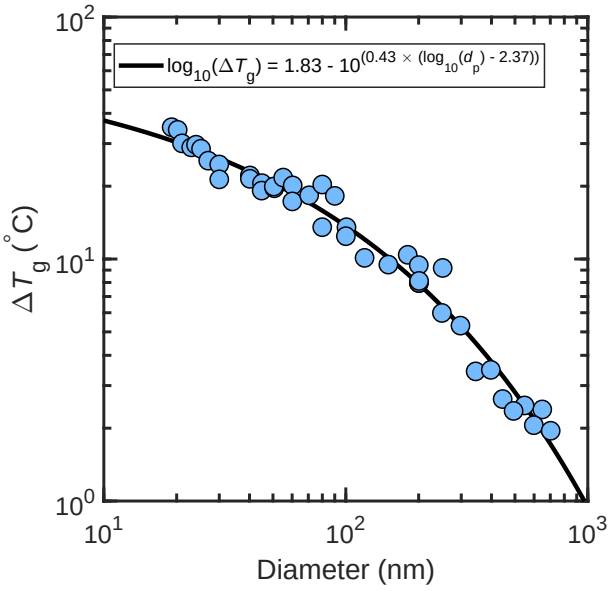

Figure S6: Glass transition temperature depression as a function of nanoparticle diameter. Black solid line represents fitting an exponential function to experimental data (blue circular markers) digitized from Mahant et al. (2024).

In the revised manuscript, we mention potential effects of enhanced mobility or quasi-liquid layers at the surface of glassy organic aerosol particles, which go beyond the scope of this study but warrant further investigation in follow-up studies:

l. 109-111 – "Additional effects such as enhanced mobility in the surface layers of solid particles (quasi-liquid layers) warrant further investigation in follow-up studies (McNeill et al., 2006; Stevenson and Wolynes, 2008; Tian et al., 2022)."

On the other hand, particle-phase chemistry has the potential to further increase viscosity and decrease diffusivity in nanoparticles, which is now also more explicitly pointed out in the extended discussion:

> l. 146-151 – "Furthermore, sunlight and atmospheric photochemistry may promote condensed-phase reactions that can enhance the formation of more highly oxygenated compounds, dimers, oligomers, and other accretion products. Such condensed-phase reaction products with high molecular mass and low volatility can influence particle composition, viscosity, and diffusivity without being reflected in gas-phase measurements of condensable organic vapor concentrations and volatility distributions (Mehra et al., 2020; Maben and Ziemann, 2023; Kenseth et al., 2023; Kang et al., 2026)."

We added a new table in the revised Supplementary Materials (Table S2) detailing the bulk diffusion coefficients used in this study:

Table S2: Diffusion coefficients used in this study at different temperatures for simulating nanoparticle growth in field and CERN CLOUD chamber studies.

	T (°C)	$D_{\mathrm{b},T}$ ($\mathrm{cm^2\,s^{-1}}$)		
		$D_{\mathrm{b},298} = 1 \times 10^{-15}$ ($\mathrm{cm^2\,s^{-1}}$)	$D_{\mathrm{b},298} = 1 \times 10^{-18}$ ($\mathrm{cm^2\,s^{-1}}$)	$D_{\mathrm{b},298} = 1 \times 10^{-20}$ ($\mathrm{cm^2\,s^{-1}}$)
Field	17.5 (summer)	-	6.59×10^{-19}	6.59×10^{-21}
	3.8 (spring)	2.91×10^{-16}	2.91×10^{-19}	-
CERN CLOUD chamber	-25	3.87×10^{-17}	3.87×10^{-20}	3.87×10^{-22}
	5	3.13×10^{-16}	3.13×10^{-19}	3.13×10^{-21}
	25	1×10^{-15}	1×10^{-18}	1×10^{-20}

Given these studies, it is questionable that the bulk diffusivity is 10-20 cm2 s-1 at higher T while Db is higher at 10-15 cm2 s-1 at lower T; Db would be lower for summer compared to spring even though temperature difference is 13 degree C (it would be helpful to compare Tg in both seasons if they could provide estimates; otherwise are there any measurements indicating difference in bounce behavior, chemical composition, or volatility distributions?). Without any additional experimental measurements or constraints, they may rather indicate that the model might be missing some critical processes (coagulation, wall loss, etc.) that might lead to this conclusion. Overall, I feel that experimental measurements of phase state/viscosity/bulk diffusivity would be necessary to solidify their results, given that the conclusion appears to be inconsistent with previous studies in many regards.

Response: We agree that uncertainties remain regarding the phase state and diffusivity of atmospheric nanoparticles and added the following statement in the revised manuscript:

> l. 113-116 – "Despite the remaining uncertainties in predicting these molecular properties, our model results clearly demonstrate that kinetic limitations related to particle-phase diffusivity of condensable organic vapors can offer a plausible, coherent, and quantitative explanation for previously unexplained observations."

In original manuscript, we had already called for measurements of particle phase state and diffusivity:

> l. 225-228 – "To further constrain key parameters and enhance the mechanistic understanding and predictability of nanoparticle growth under varying atmospheric conditions, we suggest to perform

further laboratory experiments and field observations in which particle phase state and diffusivity are determined alongside condensable vapor concentrations."

Following the suggestions of the reviewer, we added diagrams showing the evolution of T_g and D_b over time, as shown above in Figs. S4 and S5. Moreover, we emphasize which other effects may be involved and warrant further investigation:

l. 152-155 – "Our model currently assigns identical self-diffusion coefficients to the many different organic compounds lumped into each bin of decadic volatility basis sets (VBS). We note, however, that molecules in the same VBS bin may exhibit different properties such as diffusivity, effective volatility or desorption lifetime, based on their molecular structure and functionalities, even if they exhibit similar intrinsic volatilities."

l. 216-223 – "Another process affecting nanoparticle growth is coagulation, which can increase measured growth rates (Cai et al., 2021, 2026). Coagulation was not explicitly treated in this study, as Stolzenburg et al. (2025) reported little influence of coagulation for the investigated field data, while Stolzenburg et al. (2018) included it into their uncertainty estimates for the investigated chamber data. Moreover, the analysis and description of nanoparticle growth rates in atmospheric air masses can be influenced by differences between Lagrangian and Eulerian perspectives in the analysis and modeling of condensable vapor concentrations and aerosol particle size distributions. Such effects were not taken into account here but will be investigated in follow-up studies combining KM3C with regional and global 3D models of atmospheric transport and chemistry."

- I understand that ksb parameter is critical; could other unresolved miscroscopic surface processes or thermodynamics (activity coefficient or solubility?) play a role that is not treated for this parameter?

In line with previous modeling by Stolzenburg et al. (2018), we assume ideal solution behavior. Activity coefficients less than unity could make a difference for SVOC/LVOC species that do not condense at the kinetic limit. We added a statement about mixing and matrix effects to the manuscript:

l. 155-157 – "Furthermore, the properties of organic aerosol mixtures may deviate from linear combinations of properties of individual species. Fully resolving SOA nanoparticle growth may thus require a detailed accounting of specific molecular identities and their corresponding matrix interactions."

- The authors only treat condensation of SVOCs and do not consider coagulation and the impact that this may have on growth rates. Coagulation is an essential process in nanoparticle growth (Vehkamäki & Riipinen, Chem. Soc. Rev., 41, 5160-5173, 2012.) which may increase growth rates of nanoparticles significantly which might also depend on particle phase state (Nguyen et al. Environ. Sci.: Atmos., 2026, 6, 152). If the model does not treat coagulation, can you at least estimate (quantitatively if possible) or discuss the impact of coagulation on the results?

Response: In the current study, we do not consider coagulation, in line with previous investigations and model simulations building on the same data (Stolzenburg et al., 2018, 2022, 2025). Stolzenburg et al. (2025) state in their paper that "meteorological conditions are stable during regional NPF events and the pre-existing sink, as well as the overall nucleation rates, are moderate, resulting in little influence of inter- and intramodal coagulation as

well as diffusion-driven growth on the derived GR values making them easily comparable to our simple monodisperse growth model". In Stolzenburg et al. (2018), the effect of the omission of coagulation are accounted into a systematic uncertainty of 50 %.

- Coagulation could also lead to an apparent decrease in growth rates if nanoparticles are being scavenged by larger background particles. For chamber experiments there may also be losses of nanoparticles to chamber walls. Have the authors considered these mechanisms to explain the apparent low growth rates and could the omission of these mechanisms from their model be the reason for the apparent low diffusion coefficients that they estimate in their work?

Response: All simulations carried out for this work prescribe the gas-phase concentrations of condensable vapors at fixed, time-dependent values based on measurements. In line with related earlier studies Stolzenburg et al. (2018, 2025), particle coagulation and wall losses are thus not expected to have effects that would go beyond the reported uncertainties of measurement and model results.

- It is unclear how the growth rates are calculated in both the experimental measurements and in the model. Could this be clarified? Do the growth rates change over time?

Response: The measured growth rates are taken from Stolzenburg et al. (2025), who give a detailed description about the calculation of their growth rates in Stolzenburg et al. (2018) and Stolzenburg et al. (2025). In brief, for the Hyytiälä field measurements, "growth rates were estimated with the appearance time and maximum concentration methods (Stolzenburg et al., 2023). Only measurements with an agreement within a factor of 2 between maximum concentration and appearance time method are used for the in-depth analysis". For the CLOUD experiment, growth rate was estimated by the appearance-time method. Specifically, the maximum concentration method defines the representative time (t) as when the particle number concentration within a specific size bin peaks (Kulmala et al., 2013), typically determined by fitting a Gaussian function to each size channel's signal. The resulting time shift as a function of particle size reflects particle growth. The appearance time method uses the time (t_m) when the concentration reaches a specific percentage ($m\%$) of its maximum during an new particle formation event (Lehtipalo et al., 2014).

With regard to simulated growth rates, we have added the following information to the Supplementary Material of the revised manuscript (Sect. S6):

> SI l. 107-112 – "In both, KM3C used in this study and the two-film model used in Stolzenburg et al. (2025), the diameter growth rate of the particle (GR = dd_p/dt) was calculated from the $d_p(t)$ trajectory of a monodisperse particle population. The growth rate within a size interval is determined from the time required for a monodisperse particle to grow from one end of the size interval to the other:"

$$GR_{1.5\text{–}3\,\mathrm{nm}} = \frac{\Delta d_p}{\Delta t_{1.5\text{–}3\,\mathrm{nm}}} = \frac{1.5\,\mathrm{nm}}{\Delta t_{1.5\text{–}3\,\mathrm{nm}}}$$

$$GR_{3\text{–}7\,\mathrm{nm}} = \frac{\Delta d_p}{\Delta t_{3\text{–}7\,\mathrm{nm}}} = \frac{4\,\mathrm{nm}}{\Delta t_{3\text{–}7\,\mathrm{nm}}}$$

- How did you consider particle and vapor wall loss in modeling CLOUD chamber studies? These processes might

be temperature dependent (or not?). Do wall loss impact modeled or measured growth rates?

Response: As described above, all simulations carried out for this work prescribe the gas-phase concentrations of condensable vapors based on measurements. Thus, particles can be treated independently and the particle number concentration, and thus particle losses, have no effect on the modeling result. Particle and vapor wall losses were considered in the measurements of growth rates and gas phase OOMs concentration as described in Stolzenburg et al. (2018), which we would like to quote here:

(1) For particle wall losses: "Particle growth rate measurements were performed with the appearance time method." "However, this apparent growth rate of the particle size distribution does not necessarily represent the growth caused by pure condensation, as it omits coagulation and, in chamber experiments, wall losses, which both alter the particle size-distribution. Therefore, a systematic uncertainty of the method is estimated to be 50% (Lehtipalo et al., 2014)."

(2) For vapor wall losses: (2.1) To estimate the concentration of a HOM species measured by nitrate-CI-APi-ToF (NO3-CIMS), "sampling line losses SL_{HOM_i} are estimated assuming laminar flow diffusional losses in the sampling lines with a diffusion coefficient of HOMs scaling with the molecular mass M_i of the compound via $D[\mathrm{cm^2 s^{-1}}] = 0.31 \times M_i^{-1/3}$ at 278 K, determined from wall loss measurements in the CLOUD chamber. As the sampling lines of the nitrate-CI are thermally insulated, for other experiment temperatures $D \propto (T/278\mathrm{K})^{1.75}$ is assumed." (2.2) To estimate oxidized organics using PTR3-ToF (PTR3), Stolzenburg et al. (2018) applied a temperature dependent loss-correction for the sampling line losses, "which is split up into three sections: losses at the sampling line within the CLOUD chamber at chamber temperature T, as well as losses occurring at the sampling line outside the chamber at room temperature (as it was not thermally insulated) and losses within the PTR3 instrument heated to 37 °C."

- What is surface coverage during the simulation? Given very long desorption lifetime shown in Fig. S6, the coverage seems to be close to 1 (correct?). In this case, would you need to consider multilayer adsorption?

Response: Here we list the average surface coverages in the simulations of summer (both high diffusivity and low diffusivity) and spring cases over the size intervals of d = 1.5-3 nm and 3-7 nm. Even in the low diffusivity case, the surface coverage is much less than 1, therefore, a multilayer adsorption scheme is not needed in this study. We plan to look further into this topic in future studies.

	Size intervals	Surface coverage
Summer ($D_b = 10^{-20}$ cm^2 s^{-1})	1.5–3 nm	2.3×10^{-3}
	3–7 nm	1.3×10^{-3}
Summer ($D_b = 10^{-15}$ cm^2 s^{-1})	1.5–3 nm	3.0×10^{-4}
	3–7 nm	1.4×10^{-4}
Spring ($D_b = 10^{-15}$ cm^2 s^{-1})	1.5–3 nm	9.9×10^{-5}
	3–7 nm	4.6×10^{-5}

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