General Comments on the Revision of this Manuscript

After working on and careful proof reading of our manuscript in response to the points made by the two referees (below), we made additional changes to the manuscript outlined here.

We realized that our abstract does not provide any quantitative measure of the E_{des}^0 parameterization we have developed and insights for SOA formation. Since these are key findings, we have changed the abstract to reflect this fact. The revised abstract follows here and is still within the 250 word limit:

Abstract

Adsorption and desorption of gases on liquid or solid substrates are involved in multiphase processes and heterogeneous chemical reactions. The desorption energy (E_{des}^0), which depends on the intermolecular forces between adsorbate and substrate, determines the residence time of chemical species at interfaces. We show how E_{des}^0 and temperature influence the net uptake or release of gas species, the rates of surface-bulk exchange and surface or bulk reactions, and the equilibration timescales of gas-particle partitioning. Using literature data, we derive a parameterization to estimate E_{des}^0 for a wide range of chemical species based on the molecular mass, polarizability, and oxygen to carbon ratio of the desorbing species independent of substrate-specific properties, which is possible because of the dominant role of the desorbing species' properties. Correlations between E_{des}^0 and the enthalpies of vaporization and solvation are rooted in molecular interactions. The relation between E_{des}^0 and desorption kinetics reflects the key role of interfacial exchange in multiphase processes. For small molecules and semi-volatile organics (VOC, IVOC, SVOC), E_{des}^0 values around 10 - 100 kJ mol^{-1} correspond to desorption lifetimes around nanoseconds to days at room temperature. Even higher values up to years are obtained at low temperatures and for low volatile organic compounds (LVOC, ELVOC/ULVOC) relevant for secondary organic aerosols (SOA). Implications are discussed for SOA formation, gas-particle partitioning, organic phase changes, and indoor surface chemistry. We expect these insights to advance the mechanistic and kinetic understanding of multiphase processes in atmospheric and environmental physical chemistry, aerosol science, materials science, and chemical engineering.

We realized that we missed some seminal articles on PSC chemistry we wish to add to our introduction.

On line 71, we add:

(Peter, 1997; Koop et al., 1997; Muller et al., 1997; Carslaw et al., 1997)

In addition, on line 199, we mention these more explicitly by adding:

"Ground-breaking work driving much of the developments of the kinetic concepts introduced above was directed at halogen activation on stratospheric aerosol and polar stratospheric clouds (Tabazadeh et al., 1994; Peter, 1997; Carslaw et al., 1997; Hanson et al., 1994; Hanson and Lovejoy, 1995)."

We realized that we did not specify the parameter units to be used in our E_{des}^0 parameterization equations. Furthermore, we also need to state explicitly how to apply the parameterization in case of inorganic species with C = 0. We have added the following statements:

We add on line 819:

"where E_{des}^0 is in units kJ mol^{-1} and α is in units 10^{-24} cm^3."

We add on line 830:

"where M is in units g mol^{-1}. The linear regression is shown as a red line in Fig. 7."

We add on line 838:

"For application of inorganic gas species with C = 0, $O:C$ should be set to zero."

In response to the referees' raised points, we also saw the need to better highlight the relation of E_{des}^0 and τ_{des} to other molecular properties. Thus, we now show their relation to vapor pressure and widely used categories in secondary organic aerosol volatility basis set (SOA-VBS) approaches. In section "Gas-Particle Partitioning" (line 961) now called "Gas-Particle Partitioning of Secondary Organic Aerosol", we change and add the following text and figures:

Below Eq. (2), we change the sentence on line 1032:

"Figure 14 displays E_{des}^0 values for the data set of SOA oxidation products as a function of molar mass and its dependence on $O:C$ and polarizability."

to

"Figure 14 displays E_{des}^0 values for the molecular corridor data of SOA precursors gases and oxidation products from Fig. 13 as a function of molar mass and its dependence on $O:C$ and polarizability, following the molecular corridor approach using molar mass as the primary parameter characterizing a the physicochemical properties of a molecule (Shiraiwa et al., 2014)."

After line 1046, we add a new paragraph and discussion of new Figs. 15 and S10, and Table 2.:

"Figure 15 relates saturation vapor pressures (p^0) at 298 K, estimated using the EVAPORATION model (Compernolle et al., 2011), to estimated E_{des}^0, derived using Eq. 14, for selected compounds relevant for atmospheric chemistry and SOA molecular corridor data from Fig. 13 (Shiraiwa et al., 2014). The top axis of Fig. 15 shows the corresponding desorption lifetimes τ_{des} at 298 K using the Frenkel equation (Eq. 1) with a pre-exponential factor A_{des} of 10^{13} s^{-1}. The linear behavior in semi-logarithmic space (black solid line) reflects an exponential relation between p^0 and E_{des}^0 and a linear relation between p^0 and τ_{des}. Similar relationships are known for p^0 and ΔH_{vap} (Epstein et al., 2010), which underscores the correlation of these two quantities as observed in Fig. 11. We find, however, a steeper slope of p^0 with E_{des}^0 than previously found for p^0 and ΔH_{vap}, which suggests $E_{\mathrm{des}}^0 < \Delta H_{\mathrm{vap}}$ for large, oxygenated molecules with low vapor pressures. Note, however, that those gas species are lacking in our datasets. The relation of

E_{des}^0 and ΔH_{vap} and its consequences for gas-particle partitioning of SOA will be further investigated in follow-up studies.

Table 2 summarizes characteristic values of p^0, E_{des}^0, and τ_{des} for the categories of a volatility basis set (VBS) widely used for the description of SOA: intermediate volatility organic compounds (IVOC), semi-volatile organic compounds (SVOC), low volatility compounds (LVOC), extremely low volatility compounds (ELVOC), and ultra-low volatility compounds (ULVOC) (Schervish and Donahue, 2020; Donahue et al., 2009). We obtain characteristic desorption lifetimes of nanoseconds to milliseconds for VOC, milliseconds to hours for IVOC, and seconds to months for SVOC, respectively. For LVOC, ELVOC, and ULVOC we obtain τ_{des} values in the range of minutes to years and millennia. The latter, however, have to be considered as rough estimates, because the amounts of data available for parameterizing p^0 and E_{des}^0 at low volatility are very sparse. Note that these VBS categories were originally defined in terms of saturation mass concentration C^0. To express the categories in terms of p^0, we applied a constant conversion factor of 10^{-10} atm m^3 µg^{-1}, i.e., assuming a molar mass of 244 g mol^{-1}, for general orientation. For applications in which keeping the exact definition is required, a more nuanced conversion to vapor pressures could be achieved by parameterizing typical values of molar mass as a function of vapor pressure along the SOA molecular corridor. We provide an analogous figure displaying saturation mass concentrations in the Supplement (Fig. S10)."
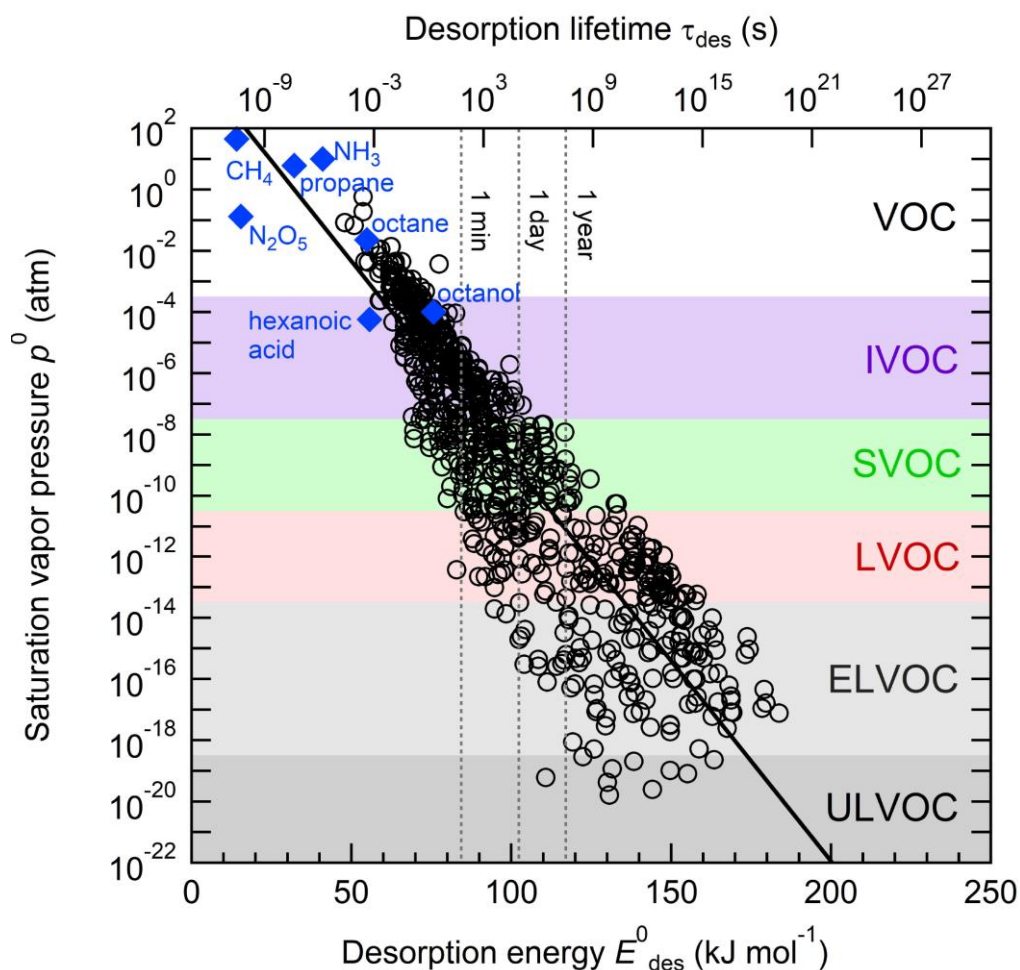
Figure 15. Characteristic desorption energies (E_{des}^0), desorption lifetimes (τ_{des}), and saturation vapor pressures (p^0) at 298 K for secondary organic aerosol (SOA) components and other selected compounds of atmospheric relevance. The blue markers show experimental literature data of E_{des}^0 and p^0. The black markers correspond to the molecular corridor data of SOA formation displayed in Fig. 13 (Shiraiwa et al., 2014), for which p^0 was estimated with the EVAPORATION model (Compernolle et al., 2011) and E_{des}^0 was estimated using Eq. 14. The black solid line represents an exponential fit to the SOA molecular corridor data. Blue markers show experimental data for selected other compounds of atmospheric relevance. Color shadings indicate widely used categories of SOA volatility basis set (VBS): volatile organic compounds (VOC), semi-volatile organic compounds (SVOC), low volatility organic compounds (LVOC), extremely-low volatility organic compounds (ELVOC) and ultra-low volatility organic compounds (ULVOC) (Schervish and Donahue, 2020; Donahue et al., 2009).
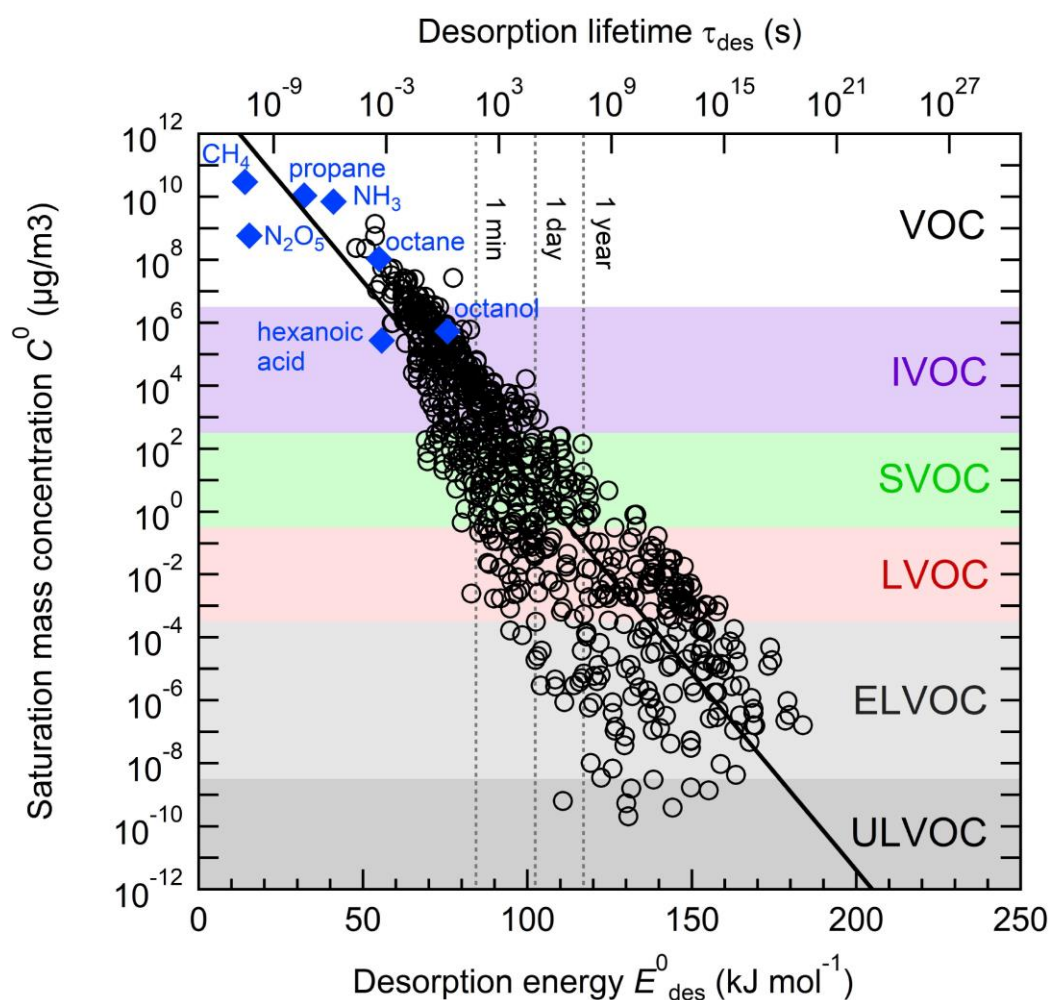

Figure S10. Characteristic desorption energies (E_{des}^0), desorption lifetimes (τ_{des}), and saturation mass concentration (C^0) at 298 K for secondary organic aerosol (SOA) components and other selected compounds of atmospheric relevance. The blue markers show experimental literature data of E_{des}^0 and C^0. The black markers correspond to the molecular corridor data of SOA

formation displayed in Fig. 13 (Shiraiwa et al., 2014), for which C^0 was estimated with the EVAPORATION model (Compernolle et al., 2011) and a constant conversion factor of 10^{-10} atm m^3 µg-1 (see text) and E_{des}^0 was estimated using Eq. 14. The black solid line represents an exponential fit to the SOA molecular corridor data. Blue markers show experimental data for selected other compounds of atmospheric relevance. Color shadings indicate widely used categories of SOA volatility basis set (VBS): volatile organic compounds (VOC), semi-volatile organic compounds (SVOC), low volatility organic compounds (LVOC), extremely-low volatility organic compounds (ELVOC) and ultra-low volatility organic compounds (ULVOC) (Schervish and Donahue, 2020; Donahue et al., 2009).

Table 2. Characteristic estimates of desorption energies (E_{des}^0) and desorption lifetimes (τ_{des}, at 298 K) for widely used categories of secondary organic aerosol volatility basis sets (SOA-VBS): volatile organic compounds (VOC), intermediate-volatile (IVOC), semi-volatile (SVOC), low-volatile (LVOC), and extremely/ultra-low volatile (ELVOC/ULVOC).

Category	Volatility p^0 (atm)	Desorption Energy E_{des}^0 (kJ mol^{-1})	Desorption Lifetime τ_{des} (298 K)
VOC	$\gtrsim 10^{-3}$	$\lesssim 60$	nanoseconds to milliseconds
IVOC	$\sim 10^{-5}$	~ 80	milliseconds to hours
SVOC	$\sim 10^{-9}$	~ 100	seconds to months
LVOC	$\sim 10^{-12}$	~ 120	minutes to centennia
ELVOC/ULVOC	$\lesssim 10^{-14}$	$\gtrsim 140$	days to millennia

Referee #1:

The manuscript entitled, "Desorption Lifetimes and Activation Energies Influencing Gas-Surface Interactions and Multiphase Kinetics," by Knopf et al., details the importance and impact of accurately measuring or deriving desorption energies to describe trace gas uptake and reaction. The authors first give an excellent overview of the meaning of desorption and how desorption energies are needed to accurately model multiphase phase kinetics either using simplified resistor models or more sophisticated kinetics simulations. They show this by changing desorption rates (i.e. by temperature) in K2-Surf simulations of reactive uptake (Figs. 1-5). The authors then compile an exhaustive list of previously measured desorption energies, from experiment and theory, for various gases onto solid and liquid interfaces. From this large data set the authors proceed to develop correlations between molecular properties (polarizability, O:C, MW, relative permittivity) and E_des. The author's objective is to develop simple ways that E_des can be easily estimated from molecular properties.

Overall, the manuscript is well written and easy to follow. The amount of data considered and compiled from prior literature is impressive and a great service to the community.

We thank the referee for their review and appreciation of our work.

There are a number of comments that the authors should address in their revision.

Eq. (3) is the normalized loss rate due to a surface reaction. Thus, shouldn't the denominator be the sum of desorption and reaction? Same question for Eq. (4), should the denominator be the sum of desorption and surface-to-bulk transfer?

We respectfully disagree. Eqs. 3 and 4 are correct. We refer to the derived equation of the Pöschl-Rudich-Ammann kinetic flux-based model (Pöschl et al., 2007)(Eqs. 115-121). Indeed, in normalized form, as derived from the flux-based balance at the surface, we obtain:

$$\gamma = \alpha_S \frac{k_s + k_{sb,net}}{k_s + k_{sb,net} + k_{des}} \ .$$

$k_{sb,net}$ is the net transfer rate coefficient of surface to bulk transfer lumping together the elementary surface to bulk transfer at the interface and the net flux due to reaction and diffusion in the bulk. When rearranging into the resistor formulation, we obtain:

$$\frac{1}{\gamma} = \frac{1}{\alpha_S}\left(1 + \frac{k_{des}}{k_s + k_{sb,net}}\right)$$

Separating out surface accommodation and separating reaction and diffusion and surface to bulk transfer, yields:

$$\frac{1}{\gamma} = \frac{1}{\alpha_S} + \frac{1}{\alpha_S \frac{k_S}{k_{des}} + \cfrac{1}{\cfrac{1}{\alpha_S \frac{k_{sb}}{k_{des}}} + \cfrac{1}{\alpha_S \frac{k_{sb}}{k_{des}} \frac{\sqrt{k_b D_b}}{k_{bs}}}}}$$

Using $\Gamma_s = \alpha_S \frac{k_s}{k_{des}}$ and $\Gamma_{sb} = \alpha_s \frac{k_{sb}}{k_{des}}$, as given in Eq. (3) and (4), respectively, and

$$\Gamma_b = \alpha_S \frac{k_{sb}}{k_{des}} \frac{\sqrt{k_b D_b}}{k_{bs}},$$

yields

$$\frac{1}{\gamma} = \frac{1}{\alpha_S} + \frac{1}{\Gamma_s + \cfrac{1}{\frac{1}{\Gamma_{sb}} + \frac{1}{\Gamma_b}}},$$

consistent with Eq. (2). To avoid complicating the introductory section too much, we prefer to not retrace this already published derivation. However, we have added some additional information to the text to easier recognize the derivation in previous published work (Pöschl et al., 2007).

Page 8 line 157. I think an additional sentence is needed to make clear from a physical perspective why accommodation, desorption and surface reaction are intertwined quantities?

We realize that the wording "intertwined" is imprecise and confusing and suggest omitting this description. Within the kinetics concept, each of α_s, k_{des}, k_{sb}, k_s, etc. are considered elementary reaction steps which are not coupled to each other. It is rather that bulk accommodation and overall uptake are depending on each of these in different ways, such that measurement of the uptake coefficient as a function of some environmental variables does often not sufficiently constrain the values of the elementary steps.

To clarify we suggest changing the original sentence to (line 186):

"More recently, kinetic multilayer model analyses of measured uptake coefficients for OH radicals on levoglucosan substrates (Arangio et al., 2015) and the heterogeneous reaction of ozone with shikimic acid (Berkemeier et al., 2016; Steimer et al., 2015) and oleic acid aerosol (Berkemeier et al., 2021) demonstrated the complex dependency of the reactive uptake coefficient on the elementary steps, such as surface accommodation, desorption, surface reaction, and bulk diffusion by virtue of Eq. (3-5). The range of experimental conditions covered in these (and many other) experiments was not sufficient to constrain the associated coefficients unambiguously."

Page 10 line 196. Given the confusing terminology used in the field, I think a few clarifying sentences are needed to link surface accommodation with thermal accommodation.

We will add additional information on line 234, partly based on our recently published article on adsorption and desorption that includes a more detailed discussion (Knopf and Ammann, 2021):

"As mentioned above, in the atmospheric sciences this is often expressed with the surface accommodation coefficient α_s (Kolb et al., 2010), operationally defined as the probability that a gas kinetic collision leads to adsorption. The adsorbed molecules may be considered an ideal

2D gas, meaning that the molecules have equilibrated with the surface in terms of the degrees of freedom perpendicular to the surface but may still retain some kinetic energy parallel to the surface. Alternatively, the adsorbed molecules may be considered an ideal 2D lattice gas, where the degrees of freedom in the horizontal plane are restricted to vibrations. Also, other models describing intermediate situations have been suggested (Savara et al., 2009; Campbell et al., 2016; Kisliuk, 1957). Here, we use α_s to describe the rate of adsorption into either adsorbed state. The term thermal accommodation coefficient, α_t, is commonly used for the case where the adsorbed molecule is fully thermally equilibrated with the substrate, thus close to the case of the ideal 2D lattice gas."

Page 16 line 337 and Page 25 line 547. The authors include only short paragraphs about liquid substrates. I agree with the authors that despite some key differences between solid and liquid interfaces the formulations developed in the manuscript nevertheless remain useful. However, for clarity I do think that the authors need to expand this discussion of liquids a bit to include not only experimental measurements but also theoretical concepts such as interfacial thickness and solvation energies derived from potential of mean force (PMF) calculations in MD simulations. Recently, for example, desorption and solvation rates/dynamics are directly obtained using these PMF. For example, see, Cruzeiro, V.W.D., et al. Uptake of N2O5 by aqueous aerosol unveiled using chemically accurate many-body potentials. Nat Commun 13, 1266 (2022). https://doi.org/10.1038/s41467-022-28697-8 and Mirza Galib, David T. Limmer, Reactive uptake of N2O5 by atmospheric aerosol is dominated by interfacial processes. Science 371, 921-925(2021)

Maybe the text up to this point has not made it clear enough, but so far, in the whole formulation of the thermodynamic and kinetic concepts, no explicit assumption has been introduced about the phase of the substrate. Even the term '2D ideal lattice gas' is not specifically referring to a substrate with a crystalline (solid) surface. The 'lattice gas' statistics may also be applied to sorption of molecules to moving substrate molecules and also independent of the dimensionality, e.g., for sorption on proteins (Hill, 1986). While partition functions may deviate in detail, with the simplifying assumptions considered here, the general expressions remain valid. We will make this clearer in the discussion in the preceding sections.

To further reflect this, we revise this last paragraph of the section with (line 386):

"In the formulation of the kinetic and thermodynamic concepts and expressions, we have not made an explicit assumption about the physical state of the condensed phase - solid, liquid, crystalline or amorphous. Lattice gas statistics can be applied generally in different dimensions and has been used for liquids, sorption of ions to proteins or polymer wires (Hill, 1986). In spite of the simplifying assumptions, we use the equations summarized above and derived in more detail in (Knopf and Ammann, 2021) for all substrates, including liquids."

And at the end of the same paragraph, we add (line 397):

"The manifestation of the change in surface tension convolutes the complex response of structure and dynamics at a liquid interface to an adsorbing molecule (Brini et al., 2017). Depending on the polarity of the adsorbate, the structural features of the interface may then also deviate significantly from that of an adsorbate on a solid surface, as exemplified in recent theory

work by Cruzeiro et al. (2022) and Galib and Limmer (2021) for the interaction of N_2O_5 with water."

On page 27, after discussion of the HCl case, we add:

Line 594: "A comparable situation as for HCl has been documented through the MB technique for N_2O_5 (Shaloski et al., 2017). Later high-level theory work established the interaction of this important trace gas with the hydrogen bonding network of water that then subsequently controls hydrolysis (Cruzeiro et al., 2022; Galib and Limmer, 2021)."

In the last paragraph of this section, when discussing the molecular dimension of the interfacial layer, we add, line 613:

"The extension of the interface depends on the type of solutes and adsorbates present, as molecules with larger hydrophobic moieties or when charges are present at the adsorbate interacting with solute ions, which may establish a larger interfacial thickness (Brini et al., 2017; Zhao et al., 2020)."

Page 20 line 438. Space between ")A" is needed.

This has been corrected.

Page 31. The example results shown in Fig. 5 for the uptake of a non-reactive species into water is confusing. The equilibration timescale above E_des > 30 kJ/mol seems entirely dominated by desorption rather than the rate at which the trace gas diffuses below the interface, which should be very fast? What is assumed about the rate coefficient for surface-to-bulk transfer in this example? In other words what is assumed about the mass accommodation coefficient in this example? I believe these details are needed for the reader to assess the actual meaning of the simulations shown in Fig. 5.

Thank you very much for bringing to our attention that the simulations shown in Fig. 5 were difficult to follow. In these calculations, we assume fully liquid particles with a bulk diffusion coefficient of water. In KM-SUB, the surface-to-bulk transfer is parameterized as diffusional transport from the sorption layer to the first bulk layer. As we assume a surface accommodation of unity, in the simulations, the mass accommodation coefficient is close to unity except in the calculations where the surface becomes physically saturated.

During revision, we found a unit conversion error in the calculations for Fig. 5 and thus repeated the simulations. To address the reviewer's question about surface vs. bulk partitioning, we now show both, surface and particle phase equilibration time scales. We also included smaller E_{des}^0 values of 10-20 kJ mol^{-1} and parameterized the bulk diffusion coefficient as a function of temperature. The updated figure is given below.
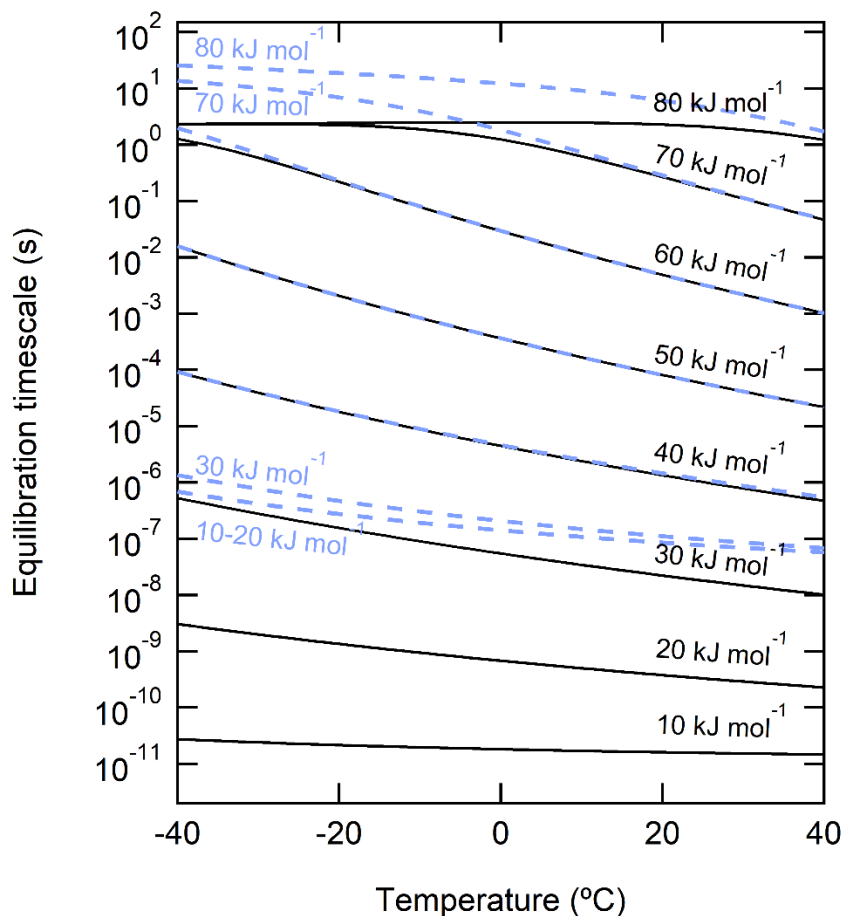
Figure 5. Equilibration timescale of non-reactive uptake of gas molecules onto the surface (solid black lines) and into the particle phase (blue dashed lines) of liquid particles with a diameter of 100 nm for different desorption energies. Gas-phase mixing ratio is fixed to be 1 ppb.

The reviewer is correct that in these calculations, the equilibration timescale at high E_{des}^0 is dominated by the timescale of partitioning to the surface, rather than partitioning to the bulk. This is in part because the Henry's law solubility coefficient was not varied with E_{des}^0 in this simulation. While we think that in reality, solvation and desorption energies are somewhat correlated (cf. Fig. 12), solubility also crucially depends on the specific solvent-solute interactions. Hence, for simplicity, we chose to keep solubility constant in this sensitivity study. The simulations can be thought of as showing a range of molecules with increasing size but limited water solubility partitioning into aqueous particles.

We have revised the discussion of the case scenario described by Fig. 5, lines 733 to 772:

"Non-reactive gas uptake into liquids. To demonstrate the effect of τ_{des} on the equilibration timescale of non-reactive gas uptake by a liquid substrate, the kinetic multi-layer model for aerosol surface and bulk chemistry (KM-SUB) (Shiraiwa et al., 2010) was applied (Fig. 5). We simulate non-reactive uptake of species X with a constant gas-phase concentration of 1 ppb into a particle with 100 nm diameter that initially contains no amount of X. The Henry's law constant

of X was set to be 1×10^{-5} mol cm^{-3} atm^{-1} at 298 K and its temperature dependence was considered using the van't Hoff equation with a solvation enthalpy of 20 kJ mol^{-1}; these values are chosen to be comparable with ozone solvation into water (Sander, 2015, 2023). The temperature dependence of the Henry's law constant is shown in Fig. S1. The particle is assumed to be liquid with a temperature-dependent bulk diffusion coefficient following the parameterization of Zobrist *et al.* (2011) for pure water, which varies from $2\times10^{-5} - 2\times10^{-6}$ cm^2 s^{-1} in this temperature range. E_{des}^0 values in the range of 10 – 80 kJ mol^{-1} were used, and the temperature dependence of τ_{des} was considered using the Frenkel equation (see Eq. (1) and Fig. 1). Here, X can be regarded as a small molecule with moderate water solubility such as ozone for the simulations at low E_{des}^0, or a carboxylic acid with similar water solubility (e.g., nonanoic acid) for the simulations at high E_{des}^0. The equilibration time is defined as the time after which the surface and particle bulk concentrations deviate by less than a factor of 1/e from their equilibrium or steady-state value.

The simulations show that equilibration times can vary over many orders of magnitude in the investigated range of E_{des}^0 (Fig. 5). For E_{des}^0 < 30 kJ mol^{-1}, the timescales of surface equilibration (black solid lines) are shorter than the timescale of bulk equilibration (blue dashed lines). The convergence of the blue lines at low E_{des}^0 (< 30 kJ mol^{-1}) reflects the kinetic limitation of gas-particle equilibration by diffusion inside the particle bulk (2×10^{-7} s^{-1}; (Shiraiwa et al., 2011)). At higher E_{des}^0, the increase of desorption lifetime leads to the increase of the equilibration times, as a larger amount of X is needed to saturate the surface; in fact, at E_{des}^0 ≥ 15 kJ mol^{-1}, the majority of molecules reside on the surface and the partitioning is governed by the surface processes in this simulation.

In the range of E_{des}^0 around 40 to 60 kJ mol^{-1}, surface and bulk equilibration times coincide, as the simulated 100 nm particles are well-mixed and non-reactive uptake is limited by interfacial transport from the gas phase. The flattening and convergence of the black lines at E_{des}^0 > 60 kJ mol^{-1} reflects the kinetic limitation of gas-particle equilibration by interfacial transport (surface adsorption and surface-bulk exchange) if the surface gets fully covered by the adsorbate. The bulk equilibration (blue lines) and thus also the overall gas-particle equilibration time still increase for E_{des}^0 > 60 kJ mol^{-1} with decreasing temperature, because interfacial transport is slowed by the high surface propensity of X and its full surface coverage. Note that the slowing of bulk equilibration time as a consequence of sorption layer coverage is a direct consequence of using a Langmuir adsorption model. In case of multilayer adsorption and bulk condensation, especially at high E_{des}^0, results may differ, which will be explored in follow-up studies (see also Sect. "Gas-particle Partitioning of Secondary Organic Aerosol"). Also note that the increased surface propensity of X with increasing E_{des}^0 is not a general rule, but a consequence of the fixed Henry's law solubility coefficient in this sensitivity study."

Page 44 line 983. Fig. 14 cited in the text should be Fig. 15

This is correct. We refer to the individual SOA oxidation products. For those molecules shown in Figs. 13 and 14, we derive E_{des}^0 and T_g using discussed parameterizations. This section and Fig. 15 have now been moved to the Appendix A1.

Page 44 line 968. I do not think that the correlation between the glass transition temperature and E_des is robust and physically defensible. There are many papers (see below*) now showing that the mobility of molecules at glass surfaces can be quite different (i.e. faster) than those molecules in the glass interior. Since desorption is sensitive to the fine details of the interface, which are clearly more complex for a glass, I do not think discussion on page 44 and the associated Fig. 15 is justified. Unless the authors can make a stronger case, I recommend this entire discussion be removed from the manuscript. *(Zhang and Z. Fakhraai, Decoupling of surface diffusion and relaxation dynamics of molecular glasses, Proceedings of the National Academy of Sciences, 2017, 114, 4915-4919. Sikorski, C. Gutt, Y. Chushkin, M. Lippmann and H. Franz, Dynamics at the Liquid-Vapor Interface of a Supercooled Organic Glass FormerPhysical Review Letters, 2010, 105, 215701.Tian, Q. Xu, H. Zhang, R. D. Priestley and B. Zuo, Surface dynamics of glasses, Applied Physics Reviews, 2022, 9.)

We appreciate the referee's point that the mobility of molecules at glass surfaces can be different than those molecules in the glass interior. Before responding directly to this point, we would like to clarify that in our discussion we do not imply a causal relationship between E_{des}^0 and T_g, but wanted to point out this observational evidence.

The reported enhanced surface mobility is observed for a few representative material systems, i.e., two typical amorphous polymers of polystyrene and poly(methyl methacrylate) (Tian et al., 2022). Thus, it has yet to be seen if enhanced surface mobility would also be applicable for atmospheric glassy SOA particles, which are highly complex multicomponent mixtures that are very different from simply polymers. Having said this, we agree with the referee that the difference in mobility on the surface compared to the bulk is most likely also relevant for atmospheric organic matter. Looking at the strong correlation between E_{des}^0 and T_g given in Fig. 15, it seems intuitive, that molecules with high E_{des}^0, thus interacting strongly with molecules of the same kind at the surface and in the bulk, also exhibit reduced dynamics in their own condensed phase (or in a mixture of similar molecules) and thus high viscosity. Considering this effect, this would still yield the observed correlation.

There is a reasonable physical argument on the positive correlation between E_{des}^0 and T_g. Based on Eq. (16), E_{des} should positively depend on molar mass and $O:C$ ratio. Shiraiwa *et al.* (2017) has shown that T_g can also be parameterized as a function of molar mass and $O:C$ ratio. As both E_{des}^0 and T_g depend linearly on the same two molecular properties, it is sensible to expect a positive correlation between E_{des}^0 and T_g. As shown in Fig. 15, we observe the expected trend that higher molar mass leads to higher E_{des}^0 and T_g, while T_g is modulated stronger by the $O:C$ ratio compared to E_{des}^0. While this analysis serves as empirical and observational evidence, the theoretical and physical basis is yet to be established, as the reviewer points out. Keeping in mind that T_g is typically considered a bulk property and enhanced surface mobility should still scale with the strength of molecular interaction, here associated with E_{des}^0, as pointed out above, the observed correlation is still meaningful for advancing our understanding of interfacial processes.

For these reasons, we would like to keep this discussion in the manuscript. However, to give it less emphasis, we move this section to the Appendix and add the valuable points mentioned by the reviewer.

We have moved the section "Glass transition" to Appendix A1 (line 1179). In addition, we have made the following changes:

In the "Summary and Conclusions" section we refer to the new Appendix section:

Line 1144 we change:

"Lastly, we outlined the correlation of glass transition points with E_{des}^0 which adds another layer of complexity when modeling multiphase chemical reactions (through the potential of viscous phase states)."

To

"Furthermore, in the Appendix A1 we outline the correlation of glass transition points with E_{des}^0 which adds another layer of complexity when modeling multiphase chemical reactions (through the potential of viscous phase states)."

We make the following changes to the text in the Appendix A1:

Line 1193:

"Recent studies have shown that a glassy surface can be much more dynamic with lower viscosity than anticipated based on T_g and bulk viscosity (Tian et al., 2022; Zhang and Fakhraai, 2017; Sikorski et al., 2010). The enhanced surface mobility, however, is mostly shown by two typical amorphous polymers of polystyrene and poly(methyl methacrylate) (Tian et al., 2022). Though it is likely that also enhanced mobility on the surface compared to the bulk is relevant for atmospheric organic matter, further studies are necessary to assess if this is applicable to atmospheric glassy SOA particles, which are highly complex multicomponent mixtures that are very different from polymers."

Line 1203:

"Since both of our parameterizations of E_{des}^0 and T_g depend on molar mass and $O:C$ ratio, it is reasonable to expect we can now construct a positive relationship between E_{des}^0 and T_g of the SOA oxidation products."

Line 1222:

"The correlation between E_{des}^0 and T_g serves as empirical and observational evidence. The theoretical and physical basis is yet to be established. It does not account for the potentially enhanced mobility on the surface of glassy matter (Tian et al., 2022; Zhang and Fakhraai, 2017; Sikorski et al., 2010). One would expect that surface mobility would similarly scale with the strength of intermolecular interactions. Molecules with high E_{des}^0 interact strongly with molecules of the same kind at the surface and in the bulk, are expected to also exhibit reduced dynamics in their own condensed phase (or in a mixture of similar molecules) and thus high viscosity. The presented correlation observed is meaningful for advancing our understanding of interfacial processes and supports further investigations."

Appendix. I believe that a list of acronym definitions (near the tables) would be helpful for a reader who doesn't want to search through the text for these. These could be placed and the beginning of the Appendix or as foot notes to the tables.

We add the following nomenclature to the Appendix.

A2. Nomenclature

τ_{des}	desorption lifetime
k_{des}	first-order desorption rate coefficient
A_{des}	pre-exponential factor
E_{des}^0	desorption energy with the energy reference of the gas molecule at rest at $T = 0$ K
γ	uptake coefficient
α_{s}	surface accommodation coefficient
Γ_{b}	normalized loss rate in the bulk-phase induced by solubility, diffusion and reaction
Γ_{sb}	normalized rate of surface to bulk transfer
Γ_{s}	normalized loss rate due to surface reaction
k_{s}	first-order rate coefficient of chemical reaction at the surface
k_{sb}	first-order rate coefficient for the transfer of molecules from the surface into the bulk (solvation)
k_{bs}	first-order rate coefficient for the transfer of molecules from the bulk to the surface
α_{b}	bulk accommodation coefficient
ΔG_{ads}^0	Gibbs free energy change of adsorption
ΔH_{ads}^0	standard enthalpy change of adsorption
ΔS_{ads}^0	standard entropy change of adsorption
κ	transmission coefficient
$(N_{\text{TS}}/\mathcal{A})^0$	standard concentration of molecules in the TS
$(N_{\text{ads}}/\mathcal{A})^0$	standard concentration of adsorbed molecules
$q_{\text{TS}}^{0\prime}$	standard partition functions for the TS
q_{ads}^0	standard partition functions for the adsorbate
q_{TS}'	partition functions for the TS
q_{ads}	partition functions for the adsorbate
M	molar mass
ΔH_{vap}	enthalpy of vaporization
α	polarizability
μ	dipole moment
$O{:}C$	oxygen to carbon ratio
ΔH_{sol}	enthalpy of solvation
ε_{r}	relative permittivity of the substrate
TPD	temperature programmed desorption
TDS	thermal desorption spectroscopy

TG-DSC	thermal gravimetry with differential scanning calorimetry
KN	Knudsen cell
MB	molecular beams
IGC	inverse gas chromatography
VM	vacuum microbalance
DRIFT	diffuse reflectance infrared fourier transform spectroscopy
FTIR	Fourier transform infrared spectroscopy
KU	kinetic uptake
VS	vibrational spectroscopy
ST	surface tension
MD	molecular dynamics
DFT	density functional theory
MC	Monte Carlo
GCMC	grand canonical Monte Carlo
ECT	embedded cluster theory
DAM	dipped adcluster model

Referee #2:

The authors build up on (their) previous work (Knopf and Ammann, 2021), explaining desorption lifetime and desorption energy and its importance for a molecular based approach for describing trace gas uptake and surface reaction on aerosol particles. Here, they provide an overview over the different experimental and modeling approaches to determine desorption energies. In addition, they survey and compile an impressive list of desorption energies and use these data and calculations to come up with a parametrization based on compound mass, polarizability and O:C ratio.

The manuscript is well written and it will certainly serve the community by providing not only the compilation of desorption energies but also illustrating its relevance for a molecular understanding of reactive uptake.

We thank the referee for their review and this positive note.

I would like the authors to consider a few comments below for the revision of the manuscript.

(1) As a non-specialist in this particular field, after reading the introduction, I am still struggling to understand the role of the surface accommodation coefficient, αS. As the (the authors) Knopf and Amman (2021) write: "In the context of atmospheric sciences, adsorption is commonly described by the surface accommodation coefficient, which is the probability that a molecule undergoing a gas kinetic collision is adsorbed at the surface". Hence, in the case of a "simple" physisorption, this probability need to somehow relate to the desorption lifetime as the authors explain starting in line 87: "In case of physisorption, Edes is equal to the negative value of the

enthalpy of adsorption with a correction for the change in degree of freedom between gas and adsorbed phase". I certainly will benefit from an expanded explanation on why we cannot then set the accommodation coefficient equal to one very generally, once we base the desorption process on the Frenkel equation (eq. 1). For example in equations 3, and 4 there remains this molecular interpretation of the terms Γs and Γsb with the first-order desorption rate based on the desorption energy, but there is also this unexplained (in terms of molecular properties) surface accommodation term. Furthermore, what follows for the surface accommodation coefficient when we assume reversible adsorption (line 252)?

There might be some misunderstanding in interpreting the phenomenon of adsorption and the kinetic treatment of desorption. The surface accommodation coefficient, α_s, is the key parameter to describe the adsorption process, and in terms of definition and value is independent of the formulation of the desorption process via the Frenkel equation (characteristic for activated desorption). The adsorption rate (and thus , α_s) does also not contain the binding energy of the adsorbed state that is contained in the Frenkel equation.

Γ_s and Γ_{sb} express the normalized loss rates due to surface reaction and surface to bulk transfer, driven by k_s and k_{sb} (the rate coefficients of the corresponding elementary processes), respectively. The normalization with the gas kinetic collision rate has to account for the probability for a gas molecule colliding with the surface to be accommodated at the surface. Hence α_s, is present in equations (3) and (4).

In (Knopf and Ammann, 2021) we derive the adsorption rate from transition state theory similar to that for desorption. By relating it to the Hertz-Knudsen equation, we then obtain the corresponding expressions for α_s, which is operationally defined by the ratio of the adsorption rate to the collision rate. It depends on the presence of an energy barrier (but not E_{des}^0, as mentioned above), the transmission coefficient (defining the probability with which an activated complex proceeds from the transition state to the adsorbed state), and the ratio of the partition functions of the transition state to those of the gas phase state (i.e., is the transition state more or less constrained than the gas phase state) (Eq. 129 in (Knopf and Ammann, 2021). Any of these parameters is typically not readily available independently in experiments, but eventually the overall α_s may be obtained from experiments (especially using molecular beam techniques). Even though α_s may often be close to 1, it may also be smaller.

In our derivations, we always assume reversible physisorption as a precursor to any further process including irreversible reactions – chemisorption would be expressed as a reaction following reversible physisorption as well.

We note that the statement (line 87) "In case of physisorption, E_{des}^0 is equal to the negative value of the enthalpy of adsorption with a correction for the change in degree of freedom between gas and adsorbed phase" is not mentioned in connection with the adsorption rate but only with the desorption rate that contains E_{des}^0 and this correction in the Arrhenius term.

The reviewer seems to refer to the coupling between adsorption and desorption under equilibrium conditions. Indeed, the adsorption equilibrium constant, i.e., the ratio of adsorption and desorption rates, could then, of course in principle, be related to the ratio of the accommodation coefficient and the desorption rate coefficient. This can indeed be used to obtain constraints on either of these parameters if the other two can be obtained from an experiment (Bartels-Rausch et al., 2005; Tabazadeh and Turco, 1993; Donaldson et al., 2012)

and we have devoted a separate section in (Knopf and Ammann, 2021) on this topic. However, we prefer to not enter this topic as this manuscript focuses on the desorption rates. Though, we have made slight text changes throughout the manuscript to make above distinction between adsorption phenomena and desorption kinetics clearer.

(2) At the end of section 3.2. "Gas adsorption by solid surfaces" the authors correctly discuss that most often atmospheric particles may have a condensed aqueous solution on its surface. They also state that in these cases one should consider the uptake process as proceeding on liquid substrates. However, the high vapor pressure of the relevant liquids does not allow easily to measure desorption kinetics (line 542). The authors suggest to use nevertheless the same concept although they admit (line 563) here the hydrogen bonding network is of particular importance and this may depend on the solutes being present. May be the authors could come back to this problem in their conclusion section?

Thank you for pointing this out. Indeed, this lack of knowledge of desorption energies for liquids with high vapor pressure and in the presence of solutes in the bulk and at the interface should be listed as a further research need. We now added this need on line 1154:

- Desorption kinetic measurements involving liquids with high vapor pressure are needed. Furthermore, the role of solutes in aqueous solutions on the hydrogen bonding network and in turn on the desorption process is not well understood. For example, adsorbates with hydrophilic functional groups exert greater E_{des}^0. Systematic examination of desorption kinetics as a function of varying solute concentration and gas species $O{:}C$ and dipole moment are needed to improve our understanding of adsorption and desorption processes on liquid surfaces.

(3) Same section "Gas adsorption by ice": I recommend to cite the review by Huthwelker et al. (2006) here for those who are interested in experimental techniques and available data and a discussion with a different focus.

Thank you for pointing out this review article (Huthwelker et al., 2006). It will be included on line 544.

(4) line 793 ff: I suggest to have similar figures for the different substrates (ice, water, aqueous, solids) like Fig.8 in the SI using the parametrization of eq. 16 to show that there are no significant physical state of substrate specific differences.

We have added a new supplemental figure that similar to Fig. 8 for solid, ice, and liquid substrates individually plotted:
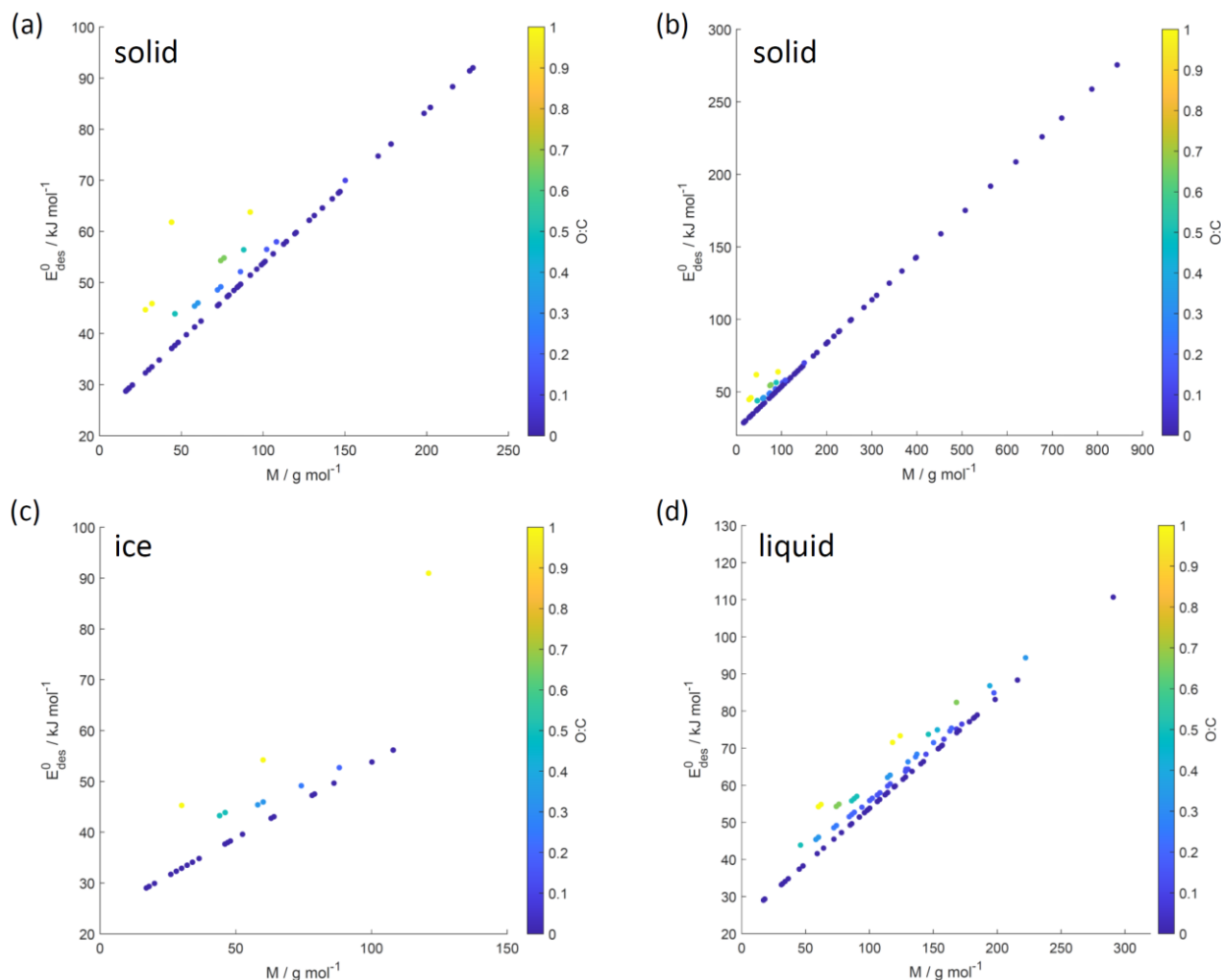
Figure S4. E_{des}^0 values derived from the new parameterization (Eq. (16)) applying the training dataset of gas species with molar mass (M) and $O{:}C$, the latter coded as symbol color described by the color bar, similar to Fig. 8. Panels (a) and (b) show data for solid substrates where (a) is an enlarged view of (b). Panel (c) shows data for ice substrates and panel (d) represents the case of liquid substrates. Note that three gas species with $O{:}C > 1$ (CO_2, formic acid, and peroxyacetyl nitrate) are included in these plots as having $O{:}C = 1$ to allow for better visualization of entire data set.

We will add a reference to this figure in the main text on line 836:

"Figure S4 shows E_{des}^0 values derived from the training data set separated by different substrate types corroborating the correlation displayed in Fig. 8."

(5) line 800 ff, Correlation desorption energy enthalpy of vaporization: Could it be that for the atmospherically very relevant liquid substrates, a parametrization based on this correlation including O:C (Fig. 11(c)) is as good as eq. (16) in particular for substances with O:C > 1? In this context: the statement in line 821 that Edes and ΔHsolv are better correlated than Edes and ΔHvap may be not true if using O:C for a parametrization as well.

For clarification purposes, we split our response in two parts:

i) The parameterization given by Eq. (16), i.e., E_{des}^0 as a function of molar weight and $O:C$ is different from a correlation between E_{des}^0 and the enthalpy of vaporization, ΔH_{vap} (Fig. 11c). We do not discuss $O:C$ values of substrates but only for gas species. We feel the reviewer meant that the correlation is better for gas species with larger $O:C$ ratios.

We provide a linear regression parameterization for E_{des}^0 as a function of ΔH_{vap} and $O:C$. This results in:

$$E_{des}^0(\Delta H_{vap}, O:C) = 5.0711 + 0.8247\Delta H_{vap} + 26.1822(O:C)$$

with an R^2 = 0.56 and RMSE = 13.0. Hence, the regression quality is similar to the parameterization given by Eq. (16).

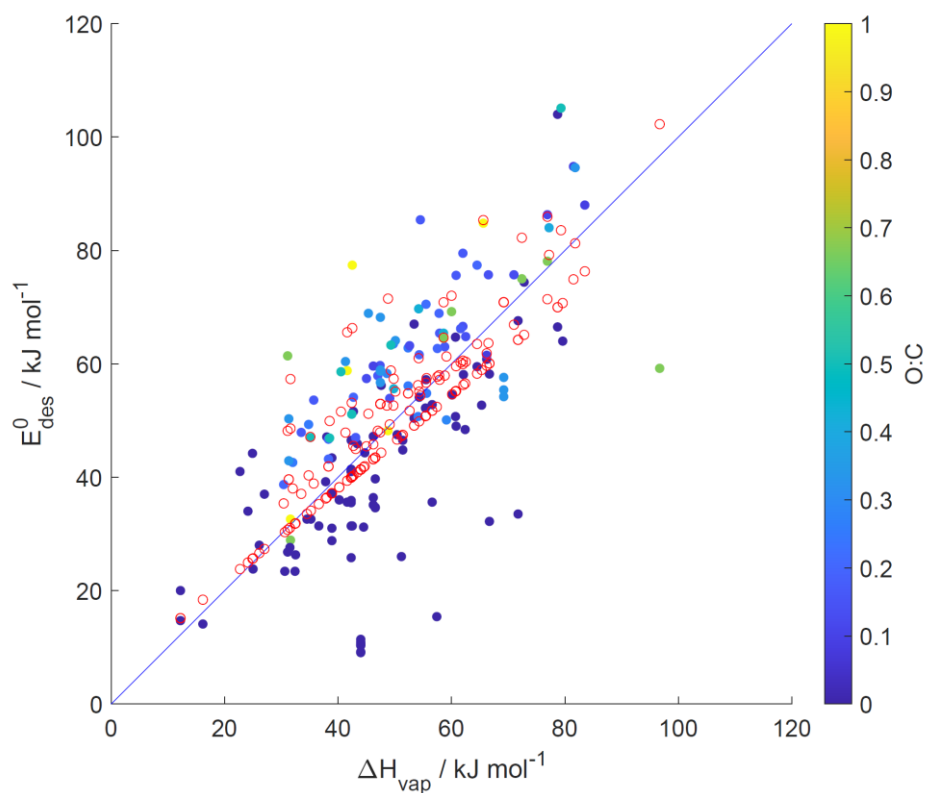**Figure S7.** Same as Fig. 11c. Red open circles depict the linear regression model applying the training data set: $E_{des}^0(\Delta H_{vap}, O:C) = 5.0711 + 0.8247\Delta H_{vap} + 26.1822(O:C)$ with an R^2 = 0.56 and RMSE = 13.0.

We add on line 893:

"Figure S7 provides a linear regression model for the case of liquid substrates."

ii) The statement on line 896 is indeed confusing, if one only looks at the liquid case. Originally this was meant to compare the E_{des}^0 correlation with ΔH_{solv} to all cases of E_{des}^0 correlation with ΔH_{vap}. This is misleading. We provide a linear regression parameterization for E_{des}^0 a function of ΔH_{solv} and O:C. This results in

$$E_{des}^0(\Delta H_{solv}, O{:}C) = 16.5830 + 0.6923\Delta H_{solv} + 9.6772(O{:}C)$$

with an R^2 = 0.39 and RMSE = 14.3. The lower R^2 value indicates that the linear regression using ΔH_{solv} results in a slightly less representative parameterization model compared to the case of using ΔH_{vap}.

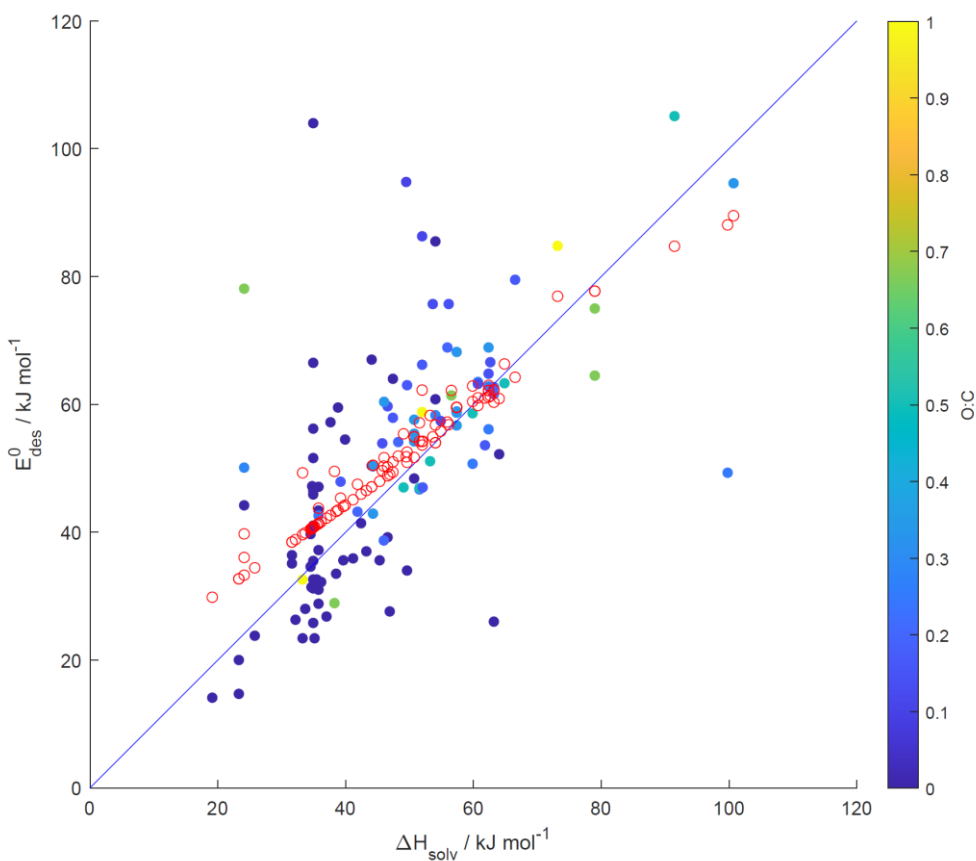**Figure S8.** Same as Fig. 12. Red open circles depict the linear regression model applying the training data set: $E_{des}^0(\Delta H_{solv}, O{:}C) = 16.5830 + 0.6923\Delta H_{solv} + 9.6772(O{:}C)$ with an R^2 = 0.39 and RMSE = 14.3.

We change the statement on line 909:

"Thus, E_{des}^0 and ΔH_{solv} values are much closer correlated than E_{des}^0 and ΔH_{vap} (Fig. 11)."

To

"In the case of liquid substrates, the correlation between E_{des}^0 and ΔH_{solv} is comparable to that observed between E_{des}^0 and ΔH_{vap} (Fig. 11c). Figure S8 provides a linear regression model for the data shown in Fig. 12."

(6) line 968 ff: "glass transition": I do not feel that the correlation between glass transition temperature and Edes goes much beyond that both correlate with molecular mass. I think this section deviates very much from the more solid molecular picture of the other sections and should be omitted.

Referee #1 voiced a similar criticism. However, as in response to Referee #1, we feel that our empirical observation of the positive correlation between E_{des}^0 and T_{g} is worthwhile reporting. We also would like to clarify that in our discussion we do not imply a causal relationship between E_{des}^0 and T_{g} but wanted to point out this observational evidence.

There is a reasonable physical argument on the positive correlation between E_{des}^0 and T_{g}. Based on Eq. (16), E_{des} should positively depend on molar mass and $O\!:\!C$ ratio. Shiraiwa *et al.* (2017) has shown that T_{g} can also be parameterized as a function of molar mass and $O\!:\!C$ ratio. As both E_{des}^0 and T_{g} depend linearly on the same two molecular properties, it is sensible to expect a positive correlation between E_{des}^0 and T_{g}. As shown in Fig. 15, we observe the expected trend that higher molar mass leads to higher E_{des}^0 and T_{g}, while T_{g} is modulated stronger by the $O\!:\!C$ ratio compared to E_{des}^0. While this analysis serves as empirical and observational evidence, the theoretical and physical basis is yet to be established. Hence we feel that the observed correlation is still meaningful for advancing our understanding of interfacial processes.

For these reasons, we would like to keep this discussion in the manuscript. However, to give it less emphasis, we move this section to the Appendix.

We have moved the section "Glass transition" to Appendix A1 (line 1179). In addition, we have made the following changes:

In the "Summary and Conclusions" section we refer to the new Appendix section:

Line 1144 we change:

"Lastly, we outlined the correlation of glass transition points with E_{des}^0 which adds another layer of complexity when modeling multiphase chemical reactions (through the potential of viscous phase states)."

To

"Furthermore, in the Appendix A1 we outline the correlation of glass transition points with E_{des}^0 which adds another layer of complexity when modeling multiphase chemical reactions (through the potential of viscous phase states)."

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