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:

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

Using $\Gamma_s = \alpha_S \dfrac{k_s}{k_{des}}$ and $\Gamma_{sb} = \alpha_s \dfrac{k_{sb}}{k_{des}}$, 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

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

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:

"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, 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:

"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."

"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:

"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:

"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 will be 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.
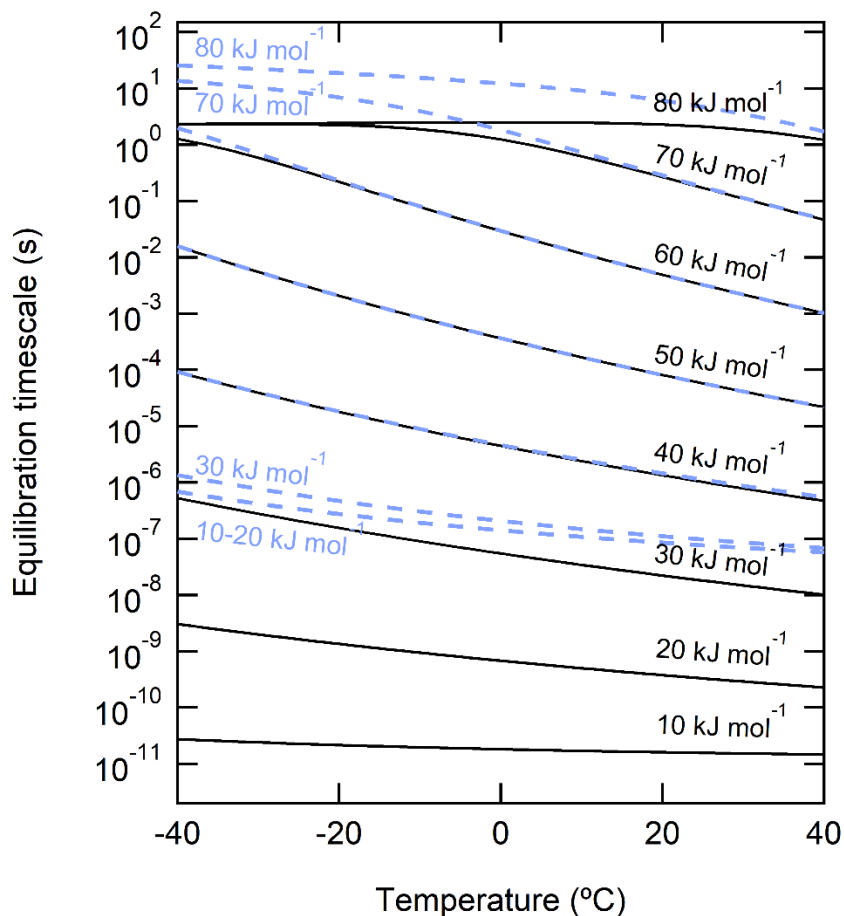
Figure 5. Equilibration timescale of non-reactive uptake of gas molecules onto the surface (solid black lines) and into the particle phase (blue dashed lines) of liquid particles with a diameter of 100 nm for different desorption energies. Gas-phase mixing ratio is fixed to be 1 ppb.

The reviewer is correct that in these calculations, the equilibration timescale at high E^0_{des} 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^0_{des} 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:

"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 will be 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 will move the section "Glass transition" to Appendix A1. In addition, we will make the following changes:

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

"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:

"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."

"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."

"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
$q_\text{TS}^{\prime}$	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

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