

Authors' response to Anonymous Referee #2

on review of *“Modeling organic aerosol over Central Europe: uncertainties linked to different chemical mechanisms, parameterizations, and boundary conditions”*

by Lukáš Bartík et al. (ecusphere-2025-167)

Dear Anonymous Referee #2,

We sincerely thank you for the time and effort you dedicated to reviewing our manuscript, and for your constructive and insightful comments. Please find below our detailed, point-by-point responses (in black) to the comments you provided (in blue).

The study presents some key challenges in accurately modeling organic aerosol concentrations over Central Europe. By using an advanced regional model along with the different approaches for atmospheric aging of organic matter, it identifies the importance of including explicit SVOC and IVOC parameterizations in emission inventories to achieve higher agreement with observations. Moreover, it points out that the share between primary and secondary organic aerosol considered for the boundary conditions is a critical choice for accuracy. This study explores a region that has not been the main focus of air quality research in Europe and would therefore be a useful reference for the community. Before I can suggest it for final publication, there are some major and minor issues that should be first addressed. You can find them below :

Major comments:

- What I found missing was a comprehensive round-up in Section 4 regarding the rate of improvement among the different model setups that were tested. For example, it is mentioned in Section 3 that the first sensitivity experiment provided increased improvement for the winter period, but the second one did so for the summer period. However different location types were taken into account as well (Rural/Urban), and it was not made clear whether one particular setup is more suitable depending on those conditions. The authors should expand the conclusions in order to clearly present which model configuration is the most important option depending on location type and season.

The conclusions have been revised to more clearly summarize the relative performance of the tested model setups by season and station type, as requested. They now indicate that the CVa configuration—combining the 1.5-D VBS scheme with aging from all OA sources and the inclusion of IVOC/SVOC emissions—provided the best overall model performance, particularly during winter. It is also noted that, under this configuration, rural stations were generally better predicted than urban stations in both seasons. The revised text further explains that adding OA to the boundary conditions had the strongest influence during summer, especially at rural sites. Together, these points clarify which configurations are most suitable depending on season and location.

Minor comments:

- Line 12: “the accuracy of modeled organic carbon concentrations improving by up to 100 %”. This sentence should be rephrased as the improvement corresponds to the FAC2 metric specifically, and can otherwise be misleading.

To avoid potential misinterpretation, we have removed the quantitative information and now refer only to “*significant improvements*” during summer at some monitoring sites.

- Line 47: The authors should consider presenting the saturation concentrations of LVOCs and SVOCs in a similar format as those for the IVOCs.

We have standardized the numerical format of all effective saturation concentrations to match the format used for IVOCs in the original manuscript.

- Line 55: “ both the original 1.5-D VBS and its various modifications”. What do these modifications alter in the original 1.5-D VBS framework?

In general, these modifications differ from the original 1.5-D VBS scheme (Koo et al., 2014) in the number of basis sets used and/or in the physical parameters that characterize them. For example, the implementation presented by Woody et al. (2016), used in our CAMx simulations, introduced five basis sets instead of four, while other studies, such as Jiang et al. (2019), further divided these into more source-specific sets and updated key parameters based on smog chamber data. These changes allow for a more detailed and source-resolved representation of OA formation and aging. We have briefly clarified this point in the revised manuscript by specifying that the modifications involve changes to the number of basis sets, the physical parameters that define them, or both.

- Line 121: What was the reasoning behind using 2 different chemical mechanisms in your simulations, since one is more comprehensive than the other?

Our motivation for this investigation was fundamentally practical. Given that both families of gas-phase mechanisms—the Statewide Air Pollution Research Center (SAPRC) mechanisms and Carbon Bond (CB) mechanisms—are widely used in the modeling community, we aimed to examine how differences in gas-phase chemistry formulations affect secondary organic aerosol (SOA) production, while keeping the SOA treatment unchanged.

To clarify this, we have added a new short paragraph in Sect. 2.1 of the revised manuscript explaining our rationale for using both mechanisms. Additionally, we note in this paragraph that their use also reflects practical constraints in the available configurations of the CAMx model: only certain combinations of gas-phase mechanisms and OA modules are supported without requiring modifications to the model code, which we sought to avoid.

- Lines 127–128: Is the aerosol size distribution in the CAMx model bimodal too? Are there any kinetic limitations taken into account for gas diffusion?

Yes, CAMx considers fine and coarse modes (0–2.5 μm and 2.5–10 μm , respectively). The coarse fraction is treated as completely inert, so organic aerosol (both primary and secondary) as well as secondary inorganic aerosol are expected to reside within the fine fraction. Moreover, all aqueous-phase chemistry occurs within the fine mode. Gas diffusion within the water condensed on aerosols is not explicitly considered; it is assumed that the gas concentration becomes uniform within the aqueous phase after absorption. Due to the sufficiently large size of aerosol particles, and thus the large corresponding aqueous spheres, kinematic effects for gas transport to the surface are also neglected.

We would also like to mention that, based on the recommendation of Referee #1, we have expanded the penultimate paragraph of Sect. 2.1 in the revised manuscript to provide a clearer explanation of how aerosols are represented in the model simulations.

- Lines 150–152: Does the REZZO inventory have a different spatiotemporal resolution than CAMS? What do you mean by 'temporal disaggregation and speciation'?

Yes, while the CAMS inventory has spatial resolution of $0.05^\circ \times 0.1^\circ$ corresponding to an area of 5 km x 10 km, the REZZO emission data offers detailed information on individual point sources as well as emission from so called “*basic territorial units*” which often corresponds to small areas of a few 100 m x 100 m (up to a few 1 km). Transport emissions from the ATEM dataset are also defined for individual roads (and interpolated to the model grid using FUME) so have also higher resolution than CAMS. We have made this clear in the revised text.

Temporal disaggregation means how the annual emissions are decomposed into hourly emission data. This is done using temporal factors describing the evolution of emissions from different activity sectors throughout the year, during the week and during the day. Speciation refers to the decomposition of total non-methane VOC emissions into individual gas-phase compounds (such as ethane, ethene, acetaldehyde, etc.) or families of compounds, such as higher aldehydes and higher ketones. It also includes the decomposition of emitted fine particulate matter into black carbon, primary organic matter, and other fine inert material. We have added this information in the revised text.

- [Section 2.3: I recommend that the authors change the titles of subsections 2.3.1 & 2.3.2 in order to make clear what was changed in each sensitivity analysis.](#)

We have extended these titles to be more specific, explicitly mentioning what was changed in each of the sensitivity analyses.

- [Table 1: Why was the VBS framework not combined with the SAPRC07TC chemical mechanism at all for the CVb and CVa experiments?](#)

Our approach was to establish a reference case using the SOAP mechanism and CB6r5 gas-phase chemistry without IVOC and SVOC emissions, and then systematically examine the sensitivity to changes in these settings—namely, by adding I/SVOC emissions, altering the gas-phase chemical mechanism, or switching the OA module.

As we explain in the newly inserted paragraph in Sect. 2.1 of the revised manuscript, when designing the experiments described in Sect. 2.4, we considered only those combinations of gas-phase mechanisms and OA modules that are directly supported by the CAMx model. Other pairings would have required additional modifications to the model code, which we sought to avoid.

Since the combination of SAPRC07TC with the 1.5-D VBS framework is not implemented in the CAMx version we used, it was not included in our experimental setup. We have clarified this rationale in the revised manuscript, and we hope this explanation sufficiently addresses the referee’s concern.

- [Lines 223 & 224: Some of the factors for SOA redistribution in Tables S3 & S4 are higher in the winter than in the summer. Since typically SOA formation peaks in the warmer periods, is this something you expect?](#)

We agree with the referee that SOA formation typically peaks during warmer periods, and we appreciate this insightful observation. However, the values presented in Tables S4 and S5 in the revised Supplement (formerly, Tables S3 and S4) are seasonal redistribution factors derived as normalized fractions — calculated independently for each season and for each reference experiment. Specifically, for each season, we divided the domain-averaged mean concentration of each surrogate SOA species by the seasonal total SOA concentration (i.e., the sum of all SOA surrogate species in that experiment and season).

These values thus represent the relative composition of SOA among its surrogate species for a given season, not the absolute concentration of SOA. Therefore, comparing values across rows (e.g., winter vs. summer) does not reflect seasonal variations in total SOA formation, but rather how SOA is distributed among the individual surrogate species within each season.

We acknowledge that the previous version of the manuscript may have left some room for ambiguity in this regard. To address this, we have revised the relevant paragraph (see Sect. 2.4.2 in the revised manuscript) to clarify both the purpose and normalized nature of these factors.

- Lines 224 & 225: Similarly, some of the factors for POA redistribution in Table S5 are higher in the summer than in the winter. Since, typically during the colder period POA emissions peak due to combustion generated for heating demands, is this something you expect?

We thank the referee for this comment, which parallels the previous one on SOA. The values in Table S6 of the revised Supplement (formerly Table S5) are likewise seasonal redistribution factors derived as normalized fractions. They represent the relative composition of POA among its surrogate species in each season and do not reflect the absolute amounts of those species. We have clarified this point in the revised manuscript (see Sect. 2.4.2).

- Section 2.4: What is the frequency of the model output regarding aerosol concentrations? Does it produce average (i.e. daily) or instantaneous values?

CAMx offers hourly averaged outputs for all species, both gas-phase and aerosol (i.e. these are mean concentrations over each hour, not instantaneous values).

Here, we would also like to inform the referee that, in addition to the changes discussed with Referee #1 regarding Sect. 2.5, 'Validation', in the revised manuscript (formerly Sect. 2.4), we revised the final two paragraphs of this section to improve clarity and precision. In the penultimate paragraph of the original version, we incorrectly implied that meteorological variables were not measured at the Prague–Suchdol station. In the revised manuscript, we clarify that Prague–Suchdol is an air quality station where accompanying meteorological measurements are indeed conducted. However, the relevant meteorological data from this site were not included in the dataset provided to us, which was limited to professional meteorological stations. For this reason, we used data from the nearby Prague–Kbely station—a professional station—which we selected as a representative site for Prague–Suchdol.

In the final paragraph, we clarified the rationale for comparing daily OC concentrations—namely, to ensure at least consistency in the duration of sampling periods across all the stations, as the longest period among them was 24 hours (at the Prague–Suchdol station), even though the sampling windows differ between sites. We also specified that CAMx was configured to output hourly averaged concentrations, which allowed us to construct daily model outputs that matched the sampling periods at each station. Additionally, we made clear that for the meteorological evaluation, only the mean daily modeled and observed values from the days with available OC measurements were used, with each daily value constructed to follow the station-specific 24-hour sampling windows.

As such, the answer to your question is now explicitly stated in the revised manuscript.

Importantly, we would like to point out that in the case of the analysis for the Prague–Suchdol and Košetice stations, we made a mistake by comparing the measured daily OC concentrations with the modeled daily concentrations derived uniformly from 00:00 UTC of the relevant day. We apologize for this oversight. We have recalculated the validation results for these two stations using the correct time windows and included the updated values in the

relevant tables in the revised Supplement (Tables S10 and S12). We also revised the figures displaying the mean daily OC concentrations (Figs. S4 and S6 in the revised Supplement) and the differences between modeled and observed values at these two stations (Figs. 6 and 10 in the revised manuscript) to reflect these corrections. We made a similar mistake in the analysis of meteorological variables at these two stations. This has now been corrected to follow the procedure described in the revised manuscript, and the corresponding table (Table S8) and figures (Figs. 2 and S1) have been updated accordingly. Based on these updated analyses, we have made several corresponding revisions to the text describing the results to ensure consistency with the corrected values.

- Lines 245 & 246: Were daily values extrapolated for the validation in this case, or was only the model output corresponding to the measurement dates used?

See our response above.

- Line 279: “the model typically tends to overestimate them more or less”. It would look better if a more accurate measure for the overestimation was stated here.

We rephrased this sentence to remove ambiguous statements and it now simply writes: *“Regarding the mean daily wind speeds, the model has the tendency to overestimate them ...”*

- Figures 2 & 3: The authors should consider adding a legend with information about what a positive/negative difference corresponds to, similarly to how Figures 6 & 7 are presented.

We have added y-axis labels to all relevant figures (Figs. 2, 3, 6, 7, 10, 11 in the revised manuscript, and Figs. S1–S7 in the revised Supplement). Additionally, we have added descriptive titles to these figures to clarify the meaning of the symbols used in the y-axis labels. We hope these changes enhance the clarity and readability of the figures.

- Lines 291–292: The underestimation of both temperature and relative humidity over the domain, also plays a role in that result. It would be important to point that out.

We agree that the underestimation of both temperature and relative humidity may influence the modeled aerosol concentrations. To address this point, we have added the following as the final paragraph of Sect. 3.1 in the revised manuscript:

“Finally, it is also worth noting that the model biases in the other studied meteorological conditions may influence the modeled aerosol concentrations. For example, the lower modeled temperatures lead to an underestimation of gas-phase reaction rates due to their temperature dependence, but they may also enhance gas-to-particle partitioning. The negative bias in the modeled relative humidity compared to the observations affects particle size and density, as both are influenced by the aerosol water content determined by the local humidity...”

- Line 328: Does that mean that the concentrations in the CVb & CVa experiments are double those of the CSnl experiment?

To clarify this point, we have revised the paragraph in the manuscript to explicitly include the relative increases in mean seasonal POA concentrations in the CVb and CVa experiments compared to CSnl. Specifically, we now indicate that, compared to CSnl, the mean seasonal POA concentrations in CVb are higher on average by a factor of 1.65 and 1.74 in the winter and summer seasons, respectively, and in CVa by a factor of 1.66 and 1.80. These values are close to a doubling of concentrations, although still somewhat lower on average.

- Line 329: What do these “similarities” refer to?

In the original version of the manuscript, the sentence *“These similarities result from the scaling of SVOC emissions using both POA for most anthropogenic sources and NMVOC emissions for diesel and gasoline vehicles...”* was intended to refer specifically to the spatial similarities between the distributions of the mean seasonal impacts in CVb and CVa and the mean seasonal POA concentrations in CSnI. That is, the observed resemblance in spatial patterns results from the way SVOC emissions were scaled.

To improve clarity, we have rephrased the sentence to make the intended reference more explicit and to better integrate it into the discussion of results. The revised sentence now reads: *“This resemblance in spatial patterns can be explained by the way SVOC emissions were scaled: POA emissions were used for most anthropogenic sources, while NMVOC emissions were used for diesel and gasoline vehicles, whose spatial distributions closely match those of POA emissions from the same vehicle categories.”*

- Figure 5: The relative increase in SOA concentrations by the CVb experiment is much more drastic (1 order of magnitude compared to CSnI) in winter than in summer, around the Po Valley. How can this be explained?

The more pronounced increase in SOA concentrations in CVb during winter (compared to CSnI), particularly in the Po Valley, is due to higher wintertime emissions of IVOCs and SVOCs from residential biomass burning. These emissions are significantly reduced in summer, leading to a smaller SOA response.

We have added this explanation to the third paragraph of Sect.3.2.1, directly after the discussion of the CVb results, with the sentence: *“The relatively smaller summer increase, compared to the winter increase, over regions such as the Po Valley, the Czech Republic, and the Pannonian Basin can be attributed to the seasonal reduction in IVOC and SVOC emissions from residential biomass burning.”*

- Line 361: Please change ‘he’ to ‘they’ when referring to a study.

Changed.

- Lines 362–365: If CSnI is taken as the reference experiment in this sensitivity, why is the comparison here with the results of Meroni et al., (2017) made with the CSwI experiment?

Although CSnI is the reference experiment for the sensitivity analysis, the comparison with the experiment by Meroni et al. (2017) was made using the CSwI experiment, as their model configuration is more consistent with that of CSwI. Specifically, they used SOAP to represent OA chemistry, the CB05 mechanism (Yarwood et al., 2005) for gas-phase chemistry, and included IVOC emissions. In particular, the inclusion of IVOC emissions is a key feature of CSwI but is not present in CSnI. Therefore, CSwI provides a more appropriate basis for comparison. We have revised the corresponding paragraph in the manuscript to clarify this rationale more explicitly.

- Lines 375 & 376: In Table 1 it states that this particular experiment did include SVOC emissions. Please clarify.

We acknowledge that the original sentence did not clearly convey our intended message and may have caused confusion regarding the inclusion of SVOC emissions. Specifically, we intended to emphasize that our assumption about the absence of SVOC emissions in the

emission inventories used to prepare input emission data for our experiments (REZZO, CAMS-REG-v4.2, and ATEM; see Sect. 2.3) is only partially accurate. We have revised the sentence accordingly to clarify this point. To further address your comment, we confirm that SVOC emissions were included in the CVa experiment, as indicated in Table 1.

- [Section 3.2.2: For the 2nd paragraph please provide percentages as a measure for the differences. The authors should also consider changing the order of paragraphs 3 & 4 to match the order of presented results by figures and tables.](#)

In the revised manuscript, we have expanded and clarified the former second paragraph by including mean percentage differences (MPDs) between the modeled and observed daily OC concentrations, thereby providing a clearer quantitative comparison. We have also reordered the paragraphs as suggested to align with the order of figures and tables and have slightly modified them accordingly.

- [Line 416: Shouldn't Tables S9 and S10 be referenced here, instead of S4 and S6?](#)

You are correct — this was a mistake. Tables S9 and S10 from the original Supplement should have been referenced there. In the revised manuscript, we have corrected the reference accordingly. It now points to Tables S10 and S11 in the revised Supplement (formerly Tables S9 and S10 in the original Supplement).

- [Lines 474–478: Is this behavior driven solely by the fractions of PAP0 and PAP1? And if so, is it expected? Are the impacts of PFP0, PFP1 and PFP2 not as important?](#)

Upon review, we found that the original statement lacked a clear reference to which simulations were being compared, and we apologize for this oversight. In the revised manuscript, we have reformulated the comparison to explicitly state that the increase in POA impacts is evaluated relative to Sp0s100 and Vp0s100, depending on the OA scheme used.

We have also expanded the explanation to clarify that the observed differences are not solely driven by PAP0 and PAP1, but rather by the redistribution of POA at the boundaries of the model domain among the POA surrogate species with different volatilities (Table S6), and by the treatment of volatility in the OA schemes. Specifically, in the VBS-driven simulations (Vp50s50 and Vp100s0), only a portion of the POA (PAP0 and PFP0) is treated as purely non-volatile. The remaining portion of this added POA can partly evaporate inside the model domain, and part of the evaporated material can be further aged to form SOA. In contrast, in the SOAP-driven simulations (Sp50s50 and Sp100s0), the same total POA as in Vp50s50 and Vp100s0, respectively, enters the model domain from the boundaries, but it is treated entirely as non-volatile, preventing both evaporation and subsequent aging.

- [Line 494: “likely linked to changes in other pollutant\(s\)”. Could you provide some examples?](#)

We agree that further clarification was needed and have revised the text accordingly. In the updated manuscript, we now explicitly state that the impacts on SOA concentrations can be directly attributable to changes in the chemical composition of the chemical boundary conditions (CBCs), which were the only factor varied in the relevant simulations (i.e., Sp100s0 vs. CSwl and Vp100s0 vs. CVb). These changes may affect both the oxidative environment — through species such as ozone, nitrogen oxides, carbon monoxide, and the hydroxyl radical — and the availability of direct SOA precursors, including toluene, xylene, and isoprene. This clarification has been added in the results section and supported by references to Sect. 2.3 in the revised manuscript and Tables S1–S2 in the revised Supplement, where the full CBC composition is documented.

- Line 497: Shouldn't it be 'Sp0s100' instead of 'Sp100s0' ? And similarly, shouldn't it be 'Vp0s100' instead of 'Vp50s000'?

The reference to Sp100s0 is correct, as it was used intentionally to compare the impact on SOA in Sp0s100 with the impact on POA in Sp100s0. However, the second case involved a typographical error — Vp50s000 should indeed have been Vp100s0. We have corrected this in the revised manuscript and also split the sentence into two for clarity.

- Lines 500 & 501: Why would the evaporation of POA be relevant for the p0s100 scenarios?

In response to this comment, we see no reason for the evaporation of POA to be relevant in the p0s100 scenarios. However, upon closer inspection of the results, we found that the statement preceding the reasoning discussed here — *“The patterns of these distributions, along with the values of Δ SOA, indicate that the increase in the mean seasonal impacts as the proportion of SOA in OA at boundaries increases is consistently higher during both seasons in experiments where SOAP handles OA chemistry.”* — is valid only in terms of domain-averaged values. The spatial distributions reveal more complexity, and the statement does not hold generally across the domain. Therefore, we have removed both the original statement and its associated reasoning from the original manuscript and did not retain them in the revised version.

- Lines 501–505: Is this behavior driven solely by the fractions of PAS0/PBS0 (VBS module) and SOA4/SOPB (SOAP module)? Are the impacts of others not as important?

Upon closer inspection of the results, we found that the original statements in the part of the paragraph referenced by the referee — describing the seasonal contrasts in the mean seasonal impacts on SOA concentrations in Sp50s50, Vp50s50, Sp0s100, and Vp0s100 — did not fully capture the spatial variability across the domain. Although the domain-averaged values of these impacts are higher during the summers in all four simulations, the mean seasonal impacts are not consistently higher across the entire domain. In some areas localized within the southeastern part of the domain, the impacts are actually higher during the winters — by up to $0.1 \mu\text{g m}^{-3}$ in Sp50s50 and Vp50s50, and by up to $0.5 \mu\text{g m}^{-3}$ in Sp0s100 and Vp0s100. To address this, we have revised the relevant text in the original manuscript, introducing a new paragraph in the revised version to more accurately reflect this spatial complexity.

Regarding the origin of this behavior, we clarify that it is not solely driven by the fractions of PAS0/PBS0 (in the VBS module) or SOA4/SOPB (in the SOAP module). Rather, it is driven by a combination of the seasonal variation in the mean monthly OA concentrations at the boundaries, which affects all four simulations, and the redistribution of these concentrations among (1) the SOA surrogate species in Sp0s100 and Vp0s100, and (2) both the POA and SOA surrogate species in Sp50s50 and Vp50s50. This clarification has also been added to the new paragraph in the revised manuscript.

- Table S2: What is the size distribution of organic matter, black carbon and sulfate? Did it have to change for the mapping? Where there any CBCs considered for ammonium, nitrate and mineral cations besides sodium, since they are also treated by ISORROPIA? Was dust considered in a bulk only state without any particular chemical composition?

All organic matter present in the EAC4 data was considered as fine aerosol, so it was entirely included in CAMx's fine aerosol bin (where POA/SOA belong). Sulphates were also considered as fine aerosol only; the same applies to black carbon. This means that we did not need to consider any modifications in size distributions. Ammonium and nitrates were not

included in this reanalysis. Mineral cations were explicitly mapped only for sea salt, which was treated as a mixture of sodium, chloride, sulphate, and magnesium. Dust was therefore taken into account as bulk matter without chemical speciation.

- **Table S3: How are the 6 SOA surrogate species allocated? In Appendix A it is mentioned that SOAP considers 3 species (2+1) from anthropogenic and another 3 (2+1) from biogenic. Can you specify which is used for what?**

We have added a footnote to the relevant table (Table S4 in the revised Supplement), which provides an explanation of the six surrogate SOA species used in SOAP.

- **Tables S3, S4 and S5: The authors should consider inserting footnotes to explain what all the species abbreviations refer to.**

We have added footnotes to the relevant tables (Tables S4–S6 in the revised Supplement), which provide explanations for all the surrogate SOA and POA species abbreviations used.

- **Table S6: If I understood correctly, the measurements from the Prague-Schudol station correspond to PM₁₀. This should be clarified in the footnotes.**

We have added a new footnote to Table S7 in the revised Supplement (formerly Table S6), which clearly states that measurements at the Prague–Suchdol station represent OC within the PM₁₀ fraction, while measurements at all other stations represent OC within the PM_{2.5} fraction.

- **Table S9: What do the last 3 columns correspond to?**

Table S10 in the revised Supplement (formerly Table S9) presents statistical analysis of the mean daily OC concentrations predicted by the individual experiments of the first sensitivity analysis at the Prague–Suchdol and Košetice stations for the individual seasons. The last three columns present analogous statistical analysis—using the same set of statistical metrics (MB, RMSE, NMSE, IOA, FAC2)—for the Košetice station, but evaluated over time periods corresponding to the winter campaign phases in the Třinecko (Tř-w) and Kladensko (KI-w) areas, and the summer campaign phase in the Kladensko (KI-s) area, respectively.

To improve clarity, we have revised the caption of Table S10 in the Supplement to explicitly describe the meaning of these columns. As a result, we have also removed the three corresponding footnotes, which are now redundant.

- **Figure S3: I believe that this figure is redundant and can be omitted, as this is more general information that can be easily accessed in statistics books.**

We agree that this information is generally available in statistics textbooks and have therefore decided to remove the figure from the Supplement.

References:

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