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Dear Editors and reviewers,

Many thanks for your attention to our manuscript (egusphere-2023-1649) “Organosulfate Produced from Consumption of SO_3 Speeds up Sulfuric Acid-Dimethylamine Atmospheric Nucleation”. We appreciate the time and effort that the editors and reviewers spent on providing constructive feedback for our paper. We have carefully considered the reviewer’s comments and questions and have addressed all their issues. Below, we provide response to the reviewer’s comment, followed by a summary of the main revisions made in the manuscript and SI.

Best regards,

Shi Yin

Referee #1

Reviewer’s comment:

The authors addressed all comment and suggestions. The paper is nicely written and the conclusions are sound. I am happy to recommend the publication of the manuscript in ACP.

Author reply:
Thanks the reviewer very much for his/her comments and efforts to improve our article.

Referee #2

Reviewer’s comment:

I apologize that my comment on the steady state concentration of SO_3 was not clear enough. I try to explain it better now. Jayne et al. (J. Phys. Chem. A 1997, 101, 10000-10011) report the reaction rate constant involving two water molecules. One water is the facilitator molecule that catalyzes the gas phase reaction. This is discussed in Jayne et al.,1997 and Hazra and Sinha, 2011. In the atmosphere sulfuric acid concentration is governed by the sulfuric acid formation and loss rates given by:
R1 $SO_2 + OH + O_2 \rightarrow SO_3 + HO_2$ $k = 1.3E\text{-}12\ cm^3\ molecule^{-1}\ s^{-1}$
R2 $SO_3 + 2\ H_2O \rightarrow H_2SO_4$
R3 $H_2SO_4 \rightarrow$ aerosol
For reaction R2 the rate constant is given by Jayne et al., as $3.90 \times 10^{-41} \exp(6830.6/T)[H_2O]^2$, which is at T= 298: $k=3.5E\text{-}31\ cm^6 s^{-1}$. For condensation of H_2SO_4 on aerosol we assume a CS of $0.01\ s^{-1}$.

The sulfuric acid concentration at steady state can be calculated by:
$d[H_2SO_4]/dt = k2\ [SO_3][H_2O]^2 - CS\ [H_2SO_4] = 0$
This yields for sulfuric acid: $[H_2SO_4] = k2[SO_3][H_2O]^2/CS$ Eq.1
rearrange for SO_3: $[SO_3] = CS\ [H_2SO_4]/(k2\ [H_2O]^2)$ Eq.2
Eq.1: Assuming $[H_2O]$=2E17 cm^{-3} and $[SO_3]$ = 1E05 cm^{-3} yields $[H_2SO_4]$ = 1.4E11 cm^{-3}
Eq. 2: Assuming $[H_2O]$=2E17 cm^{-3} and $[H_2SO_4]$ = 1E07 cm^{-3} yields $[SO_3]$ = 7 cm^{-3}
The authors use the reaction equilibrium to demonstrate, that an assumed concentration of $[SO_3]$=1E05 cm^{-3} is reasonable. They look at the reaction $H_2O + SO_3 \rightarrow H_2SO_4$ and calculate the equilibrium constant from QC data to determine a relation between SO_3 and H_2SO_4. However, the authors use the Gibbs free energy of activation barrier instead of the Gibbs free energy of formation of H_2SO_4. Thus, their determination of SO_3 or H_2SO_4 is wrong. Furthermore, the authors cite a few references to corroborate their assumption for $[SO_3]$ = 1E05 cm^{-3}.
Li et al., JACS 2018 does a calculation similar to that given above. They get $[SO_3]$ = 1E05 cm^{-3} under the assumption of $[H_2O]$ = 1E15 cm^3. Such a water vapor concentration in the atmosphere is rather low and can only be reached at 10% RH and -20°C. In the lower atmosphere such conditions occur only at special locations and SO_2 concentrations might be rather low there. Thus, this value is not representative for most of the atmosphere.
The other papers do not give any reference, why they assume $[SO_3]$ = 1E05 cm^3. Liu et al., Zhong et al. and Tsona Tchinda et al., have all co-authors in common to Li et al. Thus, it seems that they just took over this value from each other. This does not strongly support this value.
The authors also mention Yao et al, PNAS 2020. As calculated above those authors should measure sulfuric acid higher than 1.0E11 cm^{-3}, while they measure only about 1E07 cm^{-3}. In Figure 3 the authors assume that there is $[SO_3]$ = [SA] = 1E05 cm^{-3}. If one uses above steady state equations for the formation of sulfuric acid this is impossible. If [SA] is as low as 1E05 cm^3, then $[SO_3]$ is much lower and GAS would correspondingly also be lower than what the authors assumed. Thus, Figures 3 is only a theoretical exercise but not at all relevant for ambient conditions.

Author reply:

Firstly, we would like to thank again for the reviewer's time, effort and constructive feedback spending on our paper. We agree that the reviewer's consideration and question are very important and noteworthy. We believe that the key point to the summarized issue is whether the concentrations of SO_3 and gas-phase organosulfate (glycolic acid sulfate, GAS) in the atmosphere considered in our work are reasonable. At present, there are indeed not many reports on direct observation data of SO_3 and gas-phase organosulfates in the atmosphere. Actually, we also hope to have the opportunity to draw more scientists' attention to the important atmospheric active species, SO_3 and gas-phase organosulfates, through this work, so that more observational data can be reported. To the best of our ability, we found a few direct observational reports of atmospheric SO_3 and gas-phase organosulfates, summarized as follows.

(1) Yao et al. report results from two field measurements in urban Beijing during winter and summer 2019, using a nitrate-CI-APi-LTOF (chemical ionization-atmospheric pressure interface-long-time-of-flight) mass spectrometer to detect atmospheric SO_3 and H_2SO_4 (Yao et al., 2020). They found the concentration

of SO_3 was in the range between ~ 5.0 × 10^3 to ~ 1.9 × 10^6 molecules cm^{-3}, and the gas-phase H_2SO_4 was between ~ 4.5 × 10^5 to ~ 9.0 × 10^6 molecules cm^{-3} during the same period.

(2) Ehn et al. have made the first observation of organosulfate (glycolic acid sulfate, GAS) in the gas phase, and reported its ion concentration in the gas phase (6.7 molecules cm^{-3}) in the Finnish boreal forest (Ehn et al., 2010). In the same observation, the gas phase H_2SO_4 (SA) ion concentration was identified to be 242.7 molecules cm^{-3}, which is about 40 times larger than that of GAS.

(3) Le Breton et al. identified and measured 17 sulfur-containing organics (SCOs, including organosulfates, organosulfonates and nitrooxy organosulfates, GAS is one of them) at a regional site 40 km north-west of Beijing (Le Breton et al., 2018). They successfully identified a persistent gas-phase presence of SCOs in the ambient air. The mean contribution from gas-phase SCO to total SCO was found up to be 11.6 %, ~ 23 ng m^{-3} (approximately equivalent to ~ 10^6 - 10^7 molecules cm^{-3}), which suggests that a significant amount of SCO is always present in the gas phase. If we assume that the GAS accounts for ~ 1% of the total gas-phase 17 SCOs concentration, its gas-phase concentration can be roughly estimated to be 10^5 molecules cm^{-3}.

(4) Ye et al. detected abundant oxygenated organic compounds containing two to five oxygen atoms, using an iodide chemical ionization time-of-flight mass spectrometer installed with a Filter Inlet for Gases and AEROsols (FIGAERO-I-CIMS) at Guangzhou in southern China, during the autumn of 2018 (Ye et al., 2021). They detected the ion $C_2H_3SO_6^-$ with a diurnal peak in the afternoon in both gas phase and particle phase, which ion was attributed to GAS. GAS was the main organosulfate they observed.

In our manuscript, we considered the concentration of GA in the range of 1.11 × 10^7 – 2.72 × 10^9 molecules cm^{-3} according to the values of series field observations (Mochizuki et al., 2019; Miyazaki et al., 2014; Stieger et al., 2021; Mochizuki et al., 2017) and the concentration of SO_3 in the range of 10^4 – 10^6 molecules cm^{-3} according to Yao et al.'s field observations (Yao et al., 2020). According to the estimation of thermodynamic equilibrium of chemical reaction, the concentration of GAS in the range of 2.14 × 10^2 – 5.24 × 10^6 molecules cm^{-3} were considered and discussed in Figure 3, Table S1, and Figure S9, respectively, in our revised manuscript.

Thanks again for the reviewer's carefully explanation of dynamic equilibrium calculation. It is clear and we agree that the calculation is right. We apologize for the mistake of using Gibbs free energy of activation barrier instead of Gibbs free energy of the formation of H_2SO_4. We have double checked that the right Gibbs free energy of the formation was used in the calculation of GAS equilibrium concentration in our manuscript. For reaction of H_2O + SO_3 → H_2SO_4; $K_{\mathrm{H2O\,+\,SO3}} = \frac{[\mathrm{H2SO4}]}{[\mathrm{H2O}][\mathrm{SO_3}]} = e^{\frac{-\Delta G}{RT}}$; [$H_2SO_4$] = $K_{H2O\,+\,SO3}$ [H_2O][SO_3], the

Gibbs formation free energy of sulfuric acid is ~ 12 kcal mol^{-1} (Torrent-Sucarrat et al., 2012). Assuming SO_3 ~ 10^5 molecules cm^{-3}, H_2O ~ 2 x 10^{17} molecules cm^{-3}, sulfuric acid is estimated to ~ 5 x 10^{11} molecules cm^{-3}. Obviously, this result is inconsistent with the observations of Yao et al. (SO_3, ~ 5.0 × 10^3 to ~ 1.9 × 10^6 molecules cm^{-3}; H_2SO_4, ~ 4.5 × 10^5 to ~ 9.0 × 10^6 molecules cm^{-3}). Our understanding is that the concentration of the reactive specie in the atmosphere may be difficult to accurately estimate from one or a few chemical reactions, due to the complexity of real environments. We think this also reminds us that when studying species whose observed concentrations are uncertain, it may be necessary to estimate a wider concentration range to discuss its atmospheric physical and chemical effects.

Overall, we understand and agree with the reviewer's questions and concerns. We agree that the exact concentration range of SO_3 and gas-phase organosulfate are still uncertain in the actual atmosphere, due to the lack of sufficient observational data. It is possible, under some environments and conditions, their concentrations may be low and they can be ignored. In this work, we try to theoretically discuss the neglected atmospheric chemical reactions in the atmosphere that may produce secondary products that contribute significantly to the new particle formation in the atmosphere, based on limited observational data and under certain reasonable conditions. We hope that through our theoretical work, scientists may pay more attention to the secondary products of gas phase reactions, such as organosulfates. More relevant observational studies can be reported, thus providing the possibility to explore new and more complex formation mechanisms of atmospheric new particles.

Following sentences were modified and added in our revised manuscript.

"We use the reactant concentrations of [GA] = 1.11 × 10^7-2.72 × 10^9 molecules cm^{-3} according to the values of some field observations (Mochizuki et al., 2019; Miyazaki et al., 2014; Stieger et al., 2021; Mochizuki et al., 2017). Considering field measurements (Yao et al., 2020) and theoretical investigations (Tan et al., 2022; Zhong et al., 2019; Liu et al., 2019; Tsona Tchinda et al., 2022; Li et al., 2018) of atmospheric SO_3, its concentration is assumed to be 10^5 molecules cm^{-3} here. Based on the above equations, the estimated concentration of the reaction product, GAS, is about 2.14 × 10^3-5.24 × 10^5 molecules cm^{-3}, and GASA is about 2.30 × 10^{-6}-5.62× 10^{-4} molecules cm^{-3}. Thus, a range of concentration for GAS, from 10^3 to 10^5 molecules cm^{-3} as shown in Table S1, is selected for the discussion in this work." *in line 121 page 5 were modified to* "We use the reactant concentrations of [GA] = 1.11 × 10^7-2.72 × 10^9 molecules cm^{-3} according to the values of some field observations (Mochizuki et al., 2019; Miyazaki et al., 2014; Stieger et al., 2021; Mochizuki et al., 2017). Considering atmospheric SO_3 field measurements (Yao et al., 2020), its concentration is considered in the range of 10^4 - 10^6 molecules cm^{-3}. Based on the above equations, the estimated concentration of the reaction product, GAS, is about 2.14 × 10^2-5.24 × 10^6 molecules cm^{-3}, and GASA is about 2.30 × 10^{-7}-5.62× 10^{-3} molecules cm^{-3}. Thus, a range of concentration for GAS, from 10^2 to 10^6 molecules cm^{-3}, is selected for the discussion in this work (Figure 3, Table S1, and Figure S9)."

"[SO_3] = 10^4 and 10^6 molecules cm^{-3} are also considered and compared with the results shown in Figure 3a (as displayed in Figure S9). In the case of [SO_3] = 10^4 molecules cm^{-3}, it is worth noting that the cluster formation rate of GAS-SA-DMA system slightly increases with the increasing [GAS] compared to that of GA-SA-DMA system with corresponding [GA], which $J_{\text{GAS-SA-DMA}}$ reaches twice the value of $J_{\text{GA-SA-DMA}}$. For [SO_3] = 10^6 molecules cm^{-3}, the trend of this difference becomes relatively obvious, and $J_{\text{GAS-SA-DMA}}$ grows up to 2 orders of magnitude higher than $J_{\text{GA-SA-DMA}}$." *were added in line 288 page 11.*

"Le Breton et al. identified and measured 17 sulfur-containing organics (including organosulfates and GAS is one of them) at a regional site 40 km north-west of Beijing (Le Breton et al., 2018). They successfully identified a persistent gas-phase presence of organosulfates in the ambient air. The mean contribution from gas-phase sulfur-containing organics to total was found up to be 11.6 %, ~23 ng m^{-3}. Ye et al. also detected the ion $C_2H_3SO_6^-$ with a diurnal peak in the afternoon in both gas phase and particle phase, which ion was attributed to GAS, at Guangzhou in southern China during the autumn of 2018 (Ye et al., 2021)." *were added in line 201 page 7.*

"Note that the GAS concentration we discussed in this work are estimated from limited observational data of SO_3 and GA in the atmosphere. The actual atmospherics concentration of GAS still requires a large number of field observations to achieve more in-depth research." *were added in line 480 page 19.*

The authors also claim that nucleation rates at 1.2 and 3 nm are almost similar. This is very strange. They use the Kerminen-Kulmala equation and give the used parameter values. It seems that they used the condensation sink in units of s-1. This does not conform to the units in the formula and most probably led to this strange result.
Alltogether, I think the authors did not convincingly show that their assumed concentration of SO3 is reasonable and supports their further conclusions. Also the nucleation rate comparison with Mount Tai is flawed because of a wrong calculation. The paper cannot be accepted as is.

Author reply:

Thanks the reviewer for pointing out this mistake. We found the original published literatures of the revised Kerminen-Kulmala equation (Anttila et al., 2010;Lehtinen et al., 2007) and correctted the caculation as below.

$$J_x = J_1 \cdot \exp[-\gamma \cdot d_1 \cdot \frac{CS(d_1)}{GR}]$$

$$\gamma = \frac{1}{m+1}[(\frac{d_x}{d_1})^{m+1} - 1]$$

For typical atmospheric aerosols, the value of m can be set to -1.9. If we choose d_1 = 1.3 nm, d_x = 3 nm, so that $J_{1.3}$ corresponds to the “nucleation rate” at 1.3 nm and J_3 to the “nucleation rate” at 3 nm, and set m = -1.9 corresponding to typical Hyytiälä event-day conditions, we have $\gamma \approx 0.5$. GR was measured to be 3.1 nm·h^{-1} for 3.0 nm size particles during the observed events (Riipinen et al., 2007). The coagulation sink in Hyytiälä is at the level of 10^{-3} s^{-1} (Olenius et al., 2013; Dal Maso et al., 2008). Thus we can roughly get the approximate relationship $J_3 \approx 0.5\ J_{1.3}$. Considering this different value between J_3 and $J_{1.3}$, Figure 7 was amended and the related description and discussion were revised in page 16 to 18 in our modified manuscript as below.

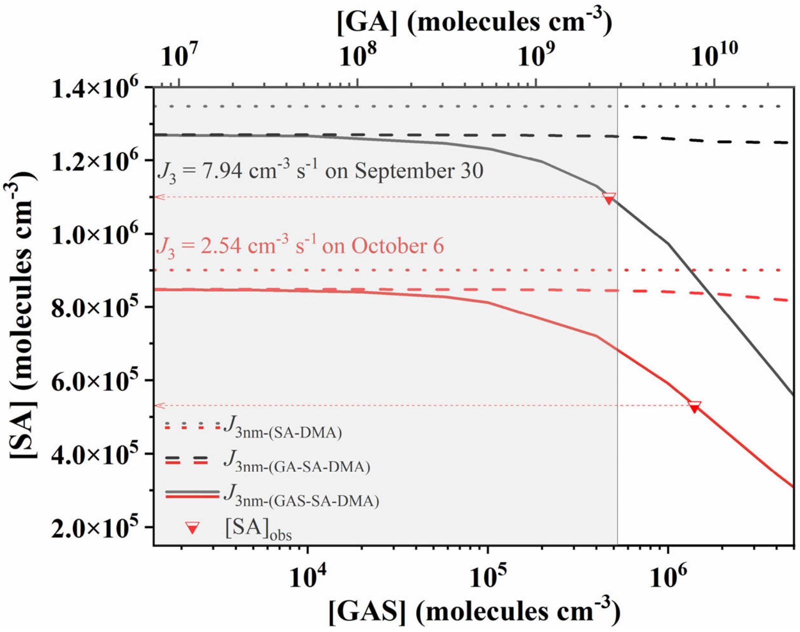

Figure 7. Required atmospheric concentrations of gas-phase precursors for pathways SA-DMA, GA-SA-DMA, and GAS-SA-DMA to reach the observed cluster formation rates (J_3) on 30 September 2014 (black lines) and 6 October 2014 (red lines) observed at Mt. Tai in China. [DMA] was set to be 10^8 molecules cm^{-3}. Dotted red lines pointing from inverted triangles to arrows represent the observed [SA] at September 30 and 6 October 2014, respectively. The shaded area represents the globally observed [GA] and corresponding [GAS]. Simulated $J_{3nm-(SA-DMA)}$, $J_{3nm-(GA-SA-DMA)}$, and $J_{3nm-(GAS-SA-DMA)}$ are represented by dotted lines, dashed lines, and solid lines, individually. Observation data of [GA] and particle formation rates (J_3) come from ref. (Mochizuki et al., 2017) and ref. (Lv et al., 2018), respectively.

“In Figure 7, we plotted the cluster formation rates for pathway SA-DMA, GA-SA-DMA and GAS-SA-DMA, individually. Note that the calculated cluster formation rates via ACDC simulation in this work are at a cluster size about ~ 1.3 nm. The observed cluster formation rates (J_3) at Mt. Tai were measured at 3 nm. According to the revised Kerminen−Kulmala equation (Anttila et al., 2010; Lehtinen et al., 2007), the cluster formation rate $J_3 \approx 0.5\ J_{1.3}$ (see details in the SI). Hence, $J_{3nm-(SA-DMA)}$, $J_{3nm-(GA-SA-DMA)}$ and $J_{3nm-(GAS-SA-DMA)}$ presented in Figure 7 were calculated from 0.5 times their associated cluster formation rates (J_{SA-DMA}, $J_{GA-SA-DMA}$ and $J_{GAS-SA-DMA}$, respectively) obtained via the ACDC simulations. Here we can see the needed concentration of SA for binary SA-DMA (dotted lines) is clearly higher than those of for ternary GA-SA-DMA (dashed lines) and GAS-SA-DMA (solid lines) system at the condition of the same formation rates. The needed [SA] for

GAS-SA-DMA system is obviously lower than that of for GA-SA-DMA, and markedly reduces with [GAS] increase. In contrast, although [GA] is higher than [GAS] in Figure 7, the variation of needed [SA] for GA-SA-DMA system is minor with the increase of [GA]. These results imply that the influence of GAS on the SA-DMA system is stronger than that of GA, and the ternary GAS-SA-DMA mechanism provides a new pathway for the NPF events with the condition of relatively low [SA] observed at Mt. Tai. The shaded area shown in Figure 7 represents the globally observed [GA] as well as corresponding [GAS]. The [SA] at Mt. Tai on September 30, 2014 is observed at 1.09×10^6 molecules cm^{-3} (the top red line pointing from an inverted triangle to the left arrow). If the new particles at Mt. Tai on September 30, 2014 are presumed to be produced from the pure SA-DMA system with the typical [DMA] of 10^8 molecules cm^{-3}, the concentration of SA around ~ 1.35×10^6 molecules cm^{-3} is needed (the black dotted line), which is quite higher than the observed [SA]. To reach the observed $J_{\text{September 30}}$ (7.94 $cm^{-3}s^{-1}$), the required [GAS] relevant to the observed [SA] on September 30, 2014, is ~ 4.70×10^5 molecules cm^{-3}, in the shaded area as shown in Figure 7. This indicates that the ternary GAS-SA-DMA nucleation mechanism corresponds well with the observed records of [SA] and NPF events. As for the GA-SA-DMA pathway, the required [GA] and [SA] are presented by black/red dashed lines. It is very clear that the GA-SA-DMA ternary system is not sufficient enough to unravel the observed NPF at Mt. Tai, for that the corresponding [GA] to the observed [SA] is beyond the boundary of shaded area. For another example, if the new particles on October 6, 2014 are assumed to be generated from pure SA-DMA system, the required [SA] is estimated to be ~ 9.01×10^5 molecules cm^{-3} (the red dotted line), which is also too high for the observed [SA] (5.3×10^5 molecules cm^{-3}, the bottom red line pointing from inverted triangle to the left arrow). Although GAS can speed up the SA-DMA driven NPF (the red line), to reach the observed $J_{\text{October 6}}$ (2.54 $cm^{-3}s^{-1}$), a fairly high concentration of GAS is required, which is out of the shaded area. This suggests that there may be other potential enhanced mechanisms for atmospheric new particle formation, which require more in-depth researches. These results and analyses suggest the GAS-SA-DMA nucleation mechanism is possible to explain the field observation on atmospheric SA-involved particles at Mt. Tai, while the binary SA-DMA nucleation is incompatible with the observed new particle formation rates. Therefore, it can be concluded that the GAS produced from the chemical reaction of GA and SO_3 could play an important role to speed up the SA-DMA driven NPF events at Mt. Tai. In the light of the deficiency of field observation of GAS in the gas phase, the further detection of GAS is still needed."

The following sentences were added in our revised supporting information.

"According to the revised Kerminen−Kulmala equation (Anttila et al., 2010;Lehtinen et al., 2007), cluster formation rates for 3.0 nm clusters ($J_{3.0}$) relate to those for 1.3 nm clusters ($J_{1.3}$) by

$$J_x = J_1 \cdot \exp[-\gamma \cdot d_1 \cdot \frac{CS(d_1)}{GR}]$$

$$\gamma = \frac{1}{m+1}[(\frac{d_x}{d_1})^{m+1} - 1]$$

where GR is the initial cluster growth rate from 1.0 to 3.0 nm, CS represents condensation sink of clusters by preexisting particles and γ is calculated as the function of d. For typical atmospheric aerosols, the value of m can be set to -1.9. If we choose d_1 = 1.3 nm, d_x = 3 nm, so that $J_{1.3}$ corresponds to the "nucleation rate" at 1.3 nm and J_3 to the "nucleation rate" at 3 nm, and set m = -1.9 corresponding to typical Hyytiälä event-day conditions, we have $\gamma \approx 0.5$. GR was measured to be 3.1 nm·h^{-1} for 3.0 nm size particles during the observed events (Riipinen et al., 2007). The coagulation sink in Hyytiälä is at the level of 10^{-3} s^{-1} (Olenius et al., 2013; Dal Maso et al., 2008). Thus we can roughly get the approximate relationship $J_3 \approx 0.5 J_{1.3}$." *were added in page S4 line 61 in our modified SI.*

Finally, please give us an opportunity to briefly explain our research purposes and ideas again. In current theoretical study we aim to reveal the potential molecular formation mechanisms of organosulfates and their potential impacts on the new particle formation, which are still much less understood. In the complex interplay of atmospheric chemistry, the role of glycolic acid sulfate in the formation of new particles has emerged as a topic of considerable importance (Long et al., 2022). Recently, Yang et al. also presented a new feasible route for the formation of organosulfates via the gas phase reactions of acetaldehyde with sulfuric acid catalyzed by dimethylamine (Yang et al., 2023). We hope more scientists will pay attention to the secondary products of gas phase reactions, such as organosulfates, through our work, and more relevant observational data can be reported. We think it will be helpful for exploring new and more complex formation mechanisms of atmospheric new particles.

We appreciate the referee's comments and questions for clarifying and improving our manuscript.

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