Dear Anonymous Referee,

Thank you for your thorough review of the manuscript. We have taken all of the your comments into consideration and revised the manuscript accordingly. All the changes have been highlighted in the revised manuscript. Our detailed responses, including a point-by-point response to the review and a list of all relevant changes, are as follows:

Q1: Line 70-71: What kind of models here? Is this overestimation a common problem for all models, all seasons or years? Is it related to problems in inventory or chemistry? How different do these models treat the formation of sulfate? The authors conclude in the abstract that this study provides a way to analyze the overestimation. I don't think this is clear yet.

A: Yes, the overestimation was a common problem. We described it in detail in line 77-84:

"Some models have reported that they failed to reproduce SO₂ and sulfate, particularly underestimating sulfate and overestimating SO₂ over China (Buchard et al., 2014; Hong et al., 2017a; Wei et al., 2019; Cheng et al., 2016). These are mainly caused by the uncertainties in meteorological conditions (Sun et al., 2016) and emission inventories (Ma et al., 2018; Hong et al., 2017b; Sha et al., 2019a;), as well as unclear and/or inaccurate physical and chemical mechanisms associated with air pollutants (He and Zhang, 2014; He et al., 2015; Georgiou et al., 2018; Sha et al., 2019b). The inadequate inclusion or lack of cloud chemistry of SO₂ consumption simulations was one of the main causes (Ge et al., 2021; Cheng et al., 2016). "

Q2: 2.3.1 and Sect. 3.1: For hourly model-observation comparisons, it is better to show them in time series instead of scatter plots in Fig. 2 so we can exam the model performance of catching cloud processing.

A: A time series might be clear than the scattered plots. We choose to use the scatter plots due to the factor that there are a lot of missing H₂O₂ and sulfate observations at Mount Tai. Those selected from the observations have to meet two conditions: 1) there are clouds over the Mount Tai from satellite image, 2) there are observations.

However, we have added the time series plots of O_3 and SO_2 both for simulation and observation for 2018 as an example (Fig. S1).



Figure S1. Time series of the simulated and observed O3 and SO2

Q3: Given the low R values of 0.06-0.4 and the clear difference in means (Table 3): the statements in Lines 185, 190, and 193 seemed inappropriate.

A: We have deleted the sentence in line 185 in the revised manuscript, and rewritten these sentences in line 217-229:

"Some reasons might contribute to the underestimations. Firstly, the latitude of the observed site at Mount Tai is 1483 meters, which may be in the boundary layer during the day time and in the free atmosphere during the night time in summer (Zhu et al., 2022). Therefore, the diurnal variation of the boundary layer affects the threedimensional concentration distribution of oxidants and aerosols (Zhao et al., 2013; Peng et al., 2021), and influences the development of cloud formation. Secondly, there exists model bias due to the difficulties to represent the complex topography of Mount Tai and the cloud physics. Thirdly, the cloud chemistry in CUACE lacks the pathway for TMI-catalyzed oxidation and NO₂-catalyzed oxidation as well as some other newly discovered oxidation mechanisms, which can lead to the bias in SO₂ and sulfate. Fourthly, typical measurement systems for ambient aerosols easily misinterpret organosulfur (mainly in the presence of hydroxy-methane sulfonate (HMS)) as inorganic sulfate, thus leading to a positive observational bias, e.g., mean bias during winter haze in Beijing is 20% (Moch et al., 2018; Song et al., 2019)." Q4: Line 178-193: The model underpredicts the sulfate concentrations at Mt. Tai a lot (Table 3). The authors explain this as the incomplete model representation of other in-cloud pathways. What in-cloud pathways are missing in the model scheme? To what extent the underestimated O₃ and H₂O₂ affect the in-cloud production of sulfate? More importantly, what are the aerosol history of the observations? Can aerosol pathways, e.g., the Mn-catalyzed oxidation [W Wang et al., 2021] or the H₂O₂ oxidation [Liu et al., 2020], be the main reason of the underestimation?

A: Yes. The cloud chemistry mechanism in CUACE has the pathways for the oxidation of SO₂ by H_2O_2 and O_3 , but lacks the TMI-catalyzed mechanism and NO₂-catalyzed mechanism as well as other newly discovered oxidation mechanisms. The result shows H_2O_2 is the main oxidant for the conversion of SO₂ to sulfate. Meanwhile, we have rewritten the reasons for the underestimation of sulfate in line 214-226 as described in Q3.

Q5: Line 198-207: I am quite confused about what was stated here. This part needs to be rewritten. The increase in atmospheric oxidation and decrease of SO₂ over years is not simulated by the model.

A: The trend is simulated by the model, and we have rewritten this paragraph in line 230-238:

"Another interesting point that is simulated correctly by the model is the increasing trend of H_2O_2 and the decreasing trend of SO₂ from 2015 to 2018. The observed and simulated mean values of H_2O_2 are 26.5 and 16.8 µM in CP-1 in 2015, to 46.9 and 32.4 µM in CP-2 in 2018, respectively. For SO₂, the observed and simulated mean values are 2.2 and 2.3 µg/m³ in CP-1 in 2015, to 0.6 and 0.6 µg/m³ in CP-2 in 2018, respectively in Table 3. Both the observations and simulations show clearly the increasing trend of H_2O_2 and the decreasing trend of SO₂ from 2015 to 2018. This conclusion is consistent with the trends of other observational studies (Shen et al., 2012; Li et al., 2020b; Ren et al., 2009; Ye et al., 2021). The SO₂ decreasing and H_2O_2 and O₃ increasing have been tightly attributed to the national SO₂ and particulate emission control measures since 2013 (Lu et al., 2020; Fan et al., 2021)."

Q6: Line 208-215: The analysis here is too brief. Please enrich to help readers understand. For the cloud liquid water, what are the observations and why the authors claim that the simulations are consistent with the observations? The simulations overestimate the cloud fraction, why and does it matter? Why do the cloud liquid water contents in Fig. 3 and 4 look different for 8:00 LST on the same dates? The sentence from Line 212-215 is long and grammatically unacceptable.

A: Yes, we have rewritten this paragraph in line 239-245:

"Figure 4 shows the RTCLD of SO2 and simulated liquid water contents at 2:00 and

8:00 LST on both June 24 and June 25 in CP-1 at Mount Tai. The column cloud and the liquid water contents which are consistent with the cloud images indicate that there is cloud with sufficient water vapor in and around the vicinity of Mount Tai (Fig. 3). The SO₂ consumption rate (RTCLD(SO₂)) distribution is consistent with the liquid water distribution at all four times (Fig. 4). The SO₂ depletion rate is above 80% at Mount Tai which is compatible to the observation (Li et al., 2020). All of these indicate that the model can capture the SO₂ consumption in the cloudy environment."

Q7: Line 216-218: The statistical values shown in this section do not sufficiently support this summary. The authors can compare their model performance to other model studies with similar comparisons to prove the goodness of the simulations here. Observational uncertainties should also be considered.

A: Yes, you are right. We have rewritten this paragraph in line 246-250:

"In summary, SO₂, H₂O₂, O₃ and sulfate concentration are in the same order of the observations, and the mean values of SO₂ are close to the observed in the cloud chemistry comparison. WRF/CUACE is also able to simulate the decreasing trend of SO₂ and the increasing trends of O₃ and H₂O₂ with year. Therefore, the cloud chemistry mechanism in WRF/CUACE is relatively reasonable to reproduce the cloud chemistry for the gaseous pollutant SO₂, sulfate and the important oxidants of H₂O₂ and O₃."

In terms of model performance, the answer to Q10 has included the other model studies with similar comparisons.

Q8: Line 225-227: I don' t observe this from Table 4. Maybe remove this sentence to avoid over-interpretation.

A: We have deleted this sentence.

Q9: Line 237-238: This is not true. After cloud evaporation, aerosol remains and can be reactivated again in the next cloud cycle. The authors need to consider the history of surface aerosol and the time scale of cloud processing.

A: We have deleted this sentence, and rewritten this paragraph in line 265-277:

"Figure 5 shows the satellite cloud images, the column cloud and the liquid water content simulated for the maturation and dissipation stages (19-22 Dec.) of the HPE. The satellite image shows that the cloud coverage region is mainly in the southwest of China besides SCB on the 19th, covering most of eastern China including NCP, YRD, PRD and SCB on the 20th and the 21st, and then moving eastward outside of China on the 22nd (Fig. 4 a1-d1). The cloud distribution fits well with the satellite images (Fig. 4

a2-d2). The column liquid water distribution also moves from west to east as the episode developed (Fig. 5 a3-d3), but is located more southern part of eastern China than that of the clouds. In SCB and YRD, the liquid water content is more abundant, reaching over 100.0 g/m², than that in PRD, only up to 10.0 g/m². NCP has the least liquid water content in the four regions, especially in Beijing, Tianjin and northwestern part of Hebei Province ranged $0.001 \sim 0.01$ g/m², mostly due to the dry environment and partly due to the overestimated temperature and underestimated humidity in Table 4. Above all, CUACE not only effectively simulates pollution but also provides a relatively reasonable meteorological background basis for cloud chemistry in the heavy pollution periods."

Q10: Line 249-252: How close? Please be specific. Comparing to other model studies for PM_5 , O_3 and SO_2 in those regions, is this model performance a good one (i.e., within a factor of two and similar means over the month)? It was concluded that the model captures well the variability of the pollutant concentrations. Do you mean spatial variability or temporal variability? Some of the R values in Table 5 are low.

A: Yes, we have rewritten this paragraph in line 280-303:

"The hourly PM_{2.5}, O₃ and SO₂ concentrations simulated in four regions are compared with the observations (Table 5). Most of the simulations are within a factor of two of the observations (figure omitted), and the mean values of the three pollutants in the four regions are close to the observations in DEC and HPE-2. It is indicated that the model captures the variability of PM_{2.5}, O₃ and SO₂ concentrations for both DEC and HPE in NCP, YRD, PRD and SCB. During HPE-2, the difference of mean values of SO₂ ranged from -7.6 to 10.4 μ g/m³, of O₃ ranged from -22 to 23.3 μ g/m³, and of PM_{2.5} ranged from -156.5 to 48.8 μ g/m³. During DEC, the difference of mean values of SO₂ ranged from -21.5 to -1.2 μ g/m³, of O₃ ranged from 1.1 to 7.7 μ g/m³, and of PM_{2.5} ranged from -71.3 to 1.3 μ g/m³. For PM_{2.5}, In the NCP, the R of HPE is 0.84, which is higher than the 0.39 of DEC in PRD. In the NCP, the R of DEC is 0.62, which is higher than the 0.30 of HPE. The R is high for both DEC and HPE in YRD, with the value of 0.73 and 0.70. The differences of R between DEC and HPE are small in YRD and SCB. For SO₂, the model simulations are better for HPE in the three regions of NCP, YRD and SCB, than that for DEC. The Rs of HPE and DEC are 0.60 and 0.48 in NCP, 0.61 and 0.45 in YRD, and 0.49 and 0.19 in SCB, respectively. The correlations between observations and simulations for HPE and DEC in PRD are not significantly different, with R of 0.32 and 0.39, respectively.

The ability of CUACE to simulate SO_2 , O_3 and sulfate concentrations have been validated in many previous research applications (Zhou et al., 2021; Zhang et al., 2021). Compared with the PM_{2.5} concentrations simulated by WRF-CUACE used by Ke et al. (2020), the correlation is 0.41~0.85 in NCP, and 0.64~0.74 in YRD. The ability of other

atmospheric models in China has the same performance such as NACRMS, and the correlation is about 0.68 for fine particulate matter in NCP during haze period (Wang et al., 2014).

The overall performance of the pollutants that can be routinely observed from the surface network have been evaluated. Then, the following part of this paper will focus on assessing the effects of cloud chemical processes."

Q11: 3: Please provide the sample size for the four regions in Tables 4-8 in Sect. 2. For the whole-month comparisons of hourly SO₂ and PM_{2.5} concentrations, I imagine some sites might not be represented well in the model. This should be discussed in Sect. 3 when presenting the modeled cloud contributions.

A: Yes. The surface observations used for the analysis in Sect. 3 are all hourly data from 55 city sites from the China National Environmental Monitoring Centre. They are mostly located in the urban area. Usually, there are several observation sites for a city. We then average the data from all the sites by excluding some obviously abnormal at one time and use the averaged data to presently the city.

For the whole-month comparisons of hourly SO_2 and $PM_{2.5}$ concentrations, some sites are not represented well in the model. Therefore, we have added discussion of that in Sect. 3 in line 401-405:

"The statistical metrics of SO_2 and $PM_{2.5}$ hourly concentrations in 55 representative cities with and without cloud chemistry in the model were analyzed. The results indicate that most of the sites are improved with cloud chemistry in the SO_2 concentration simulation and 42 of the 55 cities are with the increasing R. In the $PM_{2.5}$ simulation, the correlations also are improved in some cities after the presence of cloud chemistry.

Q12: 3.2.3 and Sect. 3.3: Are the simulations here consistent with other' s results? Comparisons to other studies should be added. For example, Aerosol surface pathways have been widely suggested in model studies for the sulfate formation [Li et al., 2018; T T Wang et al., 2022; and references therein]. Wang et al. showed in-cloud oxidation can only contribute a few percent of the surface sulfate mass in NCP [T T Wang et al., 2022; 2021]. Without implementing those mechanisms, the matches with the ground observations of the sulfate or PM2.5 mass in the model possibly means an overestimation of sulfate herein.

A: Yes. We compared with other studies, and have added in manuscript.

In line 309-319:

"Figure 6 is the mean sulfate concentration for DEC and HPE-2 for SO₂ and sulfate. The high and low centers of monthly mean SO₂ and sulfate concentrations of CUACE in December 2016 are coincided with the yearly average of the same year by Gao et al. (2021), in the SCB and NCP. For NCP, the mean sulfate concentration in Figure 6b is comparable to that by Wang et al. (2021) and Wang et al. (2022) in December 2016 as both of which increase from northwest to southeast almost in the same magnitude. The sulfate concentrations are low on a monthly basis and high at the pollution maturity stage compared to the average of several pollution processes studied by Wang et al. (2022) in December 2016. The simulation of sulfate concentration is relatively reasonable in NCP. For SCB, sulfate concentrations are compatible to the observed in winter time in 2015 by Kong et al. (2020). The sulfate concentration in Guangzhou is almost twice of the observations formed from aqueous-phase reactions in Zhou et al. (2020)."

In line 324-329:

"Ge et al. (2021) have evaluated the effects of in-cloud aqueous-phase chemistry on SO_2 oxidation in the Community Earth System Model version 2 (CESM2). They found the results incorporating detailed cloud aqueous-phase chemistry greatly reduced SO_2 overestimation, which reduced by 0.1-10 ppb in China, and more than 10 ppb in some regions in Winter. This finding is consistent with the results demonstrated in Figure 7 of this study, where SO_2 concentrations are depleted by 0.1-10 ppb in China."

In line 344-349:

"Sulfate formation rates by H_2O_2 oxidation under winter haze conditions range from 10 to 1000 µg/m³/s (Fig. S2), which is close to the range of 10 to 100 µg/m³/s tested by Wang et al. (2022) in several pollution episodes in December 2016. The heaviest and longest duration pollution episode that had a large number of clouds and high liquid water content (Fig. 5) on December 19-21, 2016, which are very favorable for the occurrence of in-cloud oxidation processes. Therefore, the in-cloud oxidation in this study is relatively reasonable."



Figure 8. The mean sulfate concentration for DEC (a, c) and HPE-2 (b, d) for SO₂ (c, e) and sulfate (a, b).



Figure S2. Sulfate formation rates by H₂O₂ oxidation in cloud in HPE-2.

Q13: Line 263-265, 272-273, 282-292: The in-cloud contributions here are all simulated quantities, for which the authors need to bring up the comparisons to specific observations (not the whole region) to justify their conclusions. For example, in Line 282-284, the cloud processing can lead to up to 225 μ g/m³ of

sulfate, which seems extremely high. I am wondering for that specific time (21:00 LST on 20 December), what the observed PM5 concentrations are in SCB or Hangzhou Bay. If the model performance isn't very good at that time, the conclusions might not be correct. I think the current manuscript was written in an over-quantitative way, which need to be revised with more careful analysis.

A: We agree that 225 μ g/m³ sulfate production by cloud chemistry was too high. We have deleted the high ranges of sulfate production 150-225 μ g/m³, which was estimated from the plot scale bar. Actual increase by cloud chemistry was not that large (Fig. 9). However, we do find that the observed PM_{2.5} concentrations up to 350 μ g/m³ at 14:00 on the 20th and 236 μ g/m³ at 21:00 on the 20th in Chengdu in SCB, up to 76 μ g/m³ at 14:00 on the 20th and 77 μ g/m³ at 21:00 on the 20th in Hangzhou in YRD (Fig.S3), were very high, partially supporting the cloud production of sulfate production at these specific times. At the same time, our model results showed that sulfate increases by cloud chemistry during these time periods were 10-20 μ g/m³ at 20-30 μ g/m³ 14:00 and 21:00 on 20th at Chengdu, 20-60 μ g/m³ and 30-60 μ g/m³ at Hangzhou.

Unfortunately, we cannot find enough observations related to cloud chemistry in all four regions. For this reason, we can only compare SO_2 , O_3 and $PM_{2.5}$ with routine observations. As for sulfate, we can only compare it at Mount Tai and with other studies as described in Q12 and Ge et al. (2021).



Figure S3. time series of PM_{2.5} concentrations in Chengdu and Hangzhou in SCB. S0-S8 represent several observation sites in a city



Figure 9. The differences in surface sulfate concentrations between with and without cloud chemistry at 21:00 LST on 20 Dec. (a), at 17:00 LST on 21 Dec. (b), and at 12:00 LST on 22 Dec. (c) (Units: $\mu g/m^3$).

Technical remarks:

1). Line 50: "a Mount site" or a mountain site?

A: Yes. We have corrected a mountain site in line 53.

2). Line 76: Define "CMA" here not in Line 163.

A: Yes. We have deleted.

3). Line 137-140: Awkward sentence. Please rewrite.

A: Yes. We have rewritten this sentence in line 155-157:

"Mount Tai with an altitude of 1483 meter, located in central Shandong Province, is the highest point of the North China Plain. It is an ideal observation site for cloud chemistry observation (Li et al., 2017; Li et al., 2020a; Li et al., 2020b)."

4). Line 141: Two "with". Please rewrite.

A: Yes. We have rewritten this sentence in line 161-162:

"WRF/CUACE is set up with two-domain nesting for the evaluation, and the Riguan Peak is the central point (Fig. 1a)."

5). Line 142: Units for 100×104 and 88×94?

A: Units for 100×104 and 88×94 are grids numbers.

Line 148-151: Awkward sentence. Please rewrite.

A: Yes. We have rewritten this sentence in line 166-168:

"The time period of December 2016 was selected to assess the regional contribution of cloud chemistry to SO₂ and sulfate in CUACE as a typical heavy pollution episode occurred from 16 to 21, covering most part of east China with the highest hourly $PM_{2.5}$ concentration exceeding 1100 µg m⁻³,"

6). Line 159-161: Usually full terms go first with abbreviations in parentheses.

A: Yes. We have corrected in line 179-181.

7). Line 164: I think you mean "air pollution" here.

A: Yes. We have corrected in line 184.

8). Line 167-169: Are those cities? PRD, YRD, NCP, and SCB have been defined previously.

A: Yes. Those are cities.We have deleted in line 186.

9). Line 169: "elements" should be "parameters".

A: Yes. We have corrected in line 186.

10). Line 174: "by five sectors of power..." should be "from power, industry, ... and agriculture sectors"

A: Yes. We have corrected in line 191.

11). Line 175: Why 2017?

A: The most recent emissions source we have were for the year of 2017.

12). Line 194-195: This is an incomplete sentence.

A: Yes. We have rewritten the paragraph and this sentence has been removed.

13). Line 228: Add a "," after "wind speed". Change "previous researches" to be "previous findings".

A: Yes. We have corrected in line 266-267.

14). Line 230: Delete "proposed by Emery et al."

A: Yes. We have deleted.

15). Line 232: What is very small? Wind speed?

A: Yes. We have rewritten this sentence in line 262-264.

"The RSME of wind speed and the wind speed for HPE is smaller than that of DEC, which indicates that the model can relatively reasonably capture the static condition."

16). Line 240-242: Awkward sentence. Please rewrite. Also, the following paragraph is redundant. That information can be merged into the analysis.

A: Yes. We have rewritten this part in line 270-277.

"The column liquid water distribution also moves from west to east as the episode developed (Fig. 5 a3-d3), but is located more southern part of eastern China than that of the clouds. In SCB and YRD, the liquid water content is more abundant, reaching over 100.0 g/m², than that in PRD, only up to 10.0 g/m². NCP has the least liquid water content in the four regions, especially in Beijing, Tianjin and northwestern part of Hebei Province ranged 0.001-0.01 g/m², mostly due to the dry environment and partly due to the overestimated temperature and underestimated humidity in Table 4. Above all, CUACE not only effectively simulates pollution but also provides a relatively reasonable meteorological background basis for cloud chemistry in the heavy pollution periods."

17). Sect. 3.2.1 and 3.2.2 can be combined. "Pollutants Evaluation" sounds strange.

A: Yes. We have changed Pollutants Evaluation to Chemical evaluation.

18). Line 247: Delete "also".

A: Yes. We have deleted.

19). Line 248: Delete "figure omitted".

A: Yes. We have deleted.

20). Overall, Sect. **3** is poorly written and wordy. Please revise the whole section for English.

A: Yes. We have rewritten Sect. 3, and all the changes have been highlighted in the revised manuscript.

21). Line 340: Add the year and month to the dates.

A: Yes. We have defined the days of the pollution stages in line 305-308:

"The regional impacts of cloud chemical processes on surface SO_2 and sulfate are analyzed for DEC and for HPE. The pollution episode (HPE) is investigated with respect to the developing stage HPE-1 (Dec. 16-18, 2016), the maturity stage HPE-2 (Dec. 19-21, 2016) and to the dissipation stage HPE-3 (Dec. 22, 2016) for the four pollution regions of NCP, YRD, PRD and SCB."

22). Tables 3-8. I believe the results in the tables are mean concentrations or values. Please clarify.

A: Yes. We have clarified the observed mean and simulated mean.

23). The figure caption for Fig. 1 isn't clear and has incorrect punctuation.

A: Yes. We have redrawn the diagram and corrected the punctuation.

24). The color bars are missing in Fig. 2.

A: Yes. The color of the dots in Fig. 2 represents the density, and the red color is the high density area. We have rewritten the figure caption for Fig. 2.

25). Please check the roles of the publisher and update the figures and captions accordingly (https://www.atmospheric-chemistry-andphysics.net/submission.html#figurestables). The terms of FY-2G cloud in Fig. 3 are redundant. Color bars can be combined for each of the two panels. The dates in the figure caption can be marked in the graph instead. Add descriptions about what the cloud image show (cloud fraction?) and what the triangle is. The font size in a3 and b3 is should be the same as others. Check the unit of liquid water content in Fig. 4. It is different from Figs. 3 and 5. It is confusing about the red triangle in a3 and b3 (real color in terms of simulated liquid water content?). Similar to Fig. 3, color bars in Figs. 4, 5, and 8 are repeated unnecessarily. The repeated legends in Figs. 10 and 11, the unnecessary frames in Figs. 6-8 and 10 make the graphs look ugly. The figure captions in Figs. 6-8, 10, and 11 and all table captions need to be revised for English. Please clarify that there are the mean values or concentrations listed in the tables not median or something else.

A: Yes. We have added descriptions about the triangle, removed unnecessary color bars, and marked the dates in figure 3 and 4. We have adjusted the font name and font size in figure 3, 5, and others. We removed the unnecessary frames in figure 6-8 and 10. We have clarified the mean values in some tables. We have checked all captions and adjusted.

26). Table 8: "sellected" should be "selected". It is better to not use abbreviation as "the whole Dec."

Yes. We have changed the table 8 to figure 11, and we didn't use the whole Dec..

The references newly added are listed as follows:

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pollutants during winter over the Yangtze River Delta, Atmos. Environ., 206, 170-182, https://doi.org/10.1016/j.atmosenv.2019.03.006, 2019a.

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- Wang, T., Liu, M., Liu, M., Song, Y., Xu, Z., Shang, F., Huang, X., Liao, W., Wang, W., Ge, M., Cao, J., Hu, J., Tang, G., Pan, Y., Hu, M., and Zhu, T.: Sulfate Formation Apportionment during Winter Haze Events in North China, Environ. Sci. Technol., 56(12), 7771-7778, https://doi.org/10.1021/acs.est.2c02533, 2022.
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