We thank the referee for the careful reviews and suggestions. Following is our response to the comments:

- ***Referee #2:***

General comments: The manuscript presents novel data regarding IVOC emission factors for a gasoline/E10 Chinese vehicle, that meets China V standard. Methods are sound, the language is cogent and very easy to follow. The presentation of the results is very clear and the main findings are thoroughly discussed and compared to previous literature, considering differences and similarities with US-based data. As the paper entails important implications for both the scientific community and policymakers, I recommend final publication after minor revisions. The following comments are mostly aimed to improve the readability, interpretability and usefulness of the study for future work.

Specific comments

1. To facilitate the use of your new data in modeling studies using the Volatility Basis Set (VBS) scheme, I would recommend to present the volatility distribution data also in terms of saturation concentration bins, in a similar way to Zhao et al. [2016] (Figure 4). Also, it would be convenient if you can report a Table, maybe in the SI, reporting the mass fraction distribution of organics for each saturation concentration bin (e.g. Table S5 in Zhao et al. [2016]). These values are usually a key input for the VBS schemes in state-of-the-art numerical models. In

addition to this, I would suggest to report the median IVOC-to-THC ratio in the abstract as well, as that is key information for modelers.

Response: Thank you for your comment. We have presented the volatility distribution data both in figure and table in the manuscript. The revision was as follows: "Considering the similarity of volatility distribution for different conditions and the importance of the volatility distribution in model input for SOA simulation, Figure S6 and Table S3 present the volatility distribution of SVOC and IVOC emissions from the tested China V gasoline vehicle, using effective saturation concentration (C*) as classification: IVOCs (C*=300-3×10^6 μg·m^{-3}), SVOCs (C*=0.3-300 μg·m^{-3}). IVOCs are the dominant part of the low volatility organics (IVOCs+SVOCs), with a median contribution of ~95%."

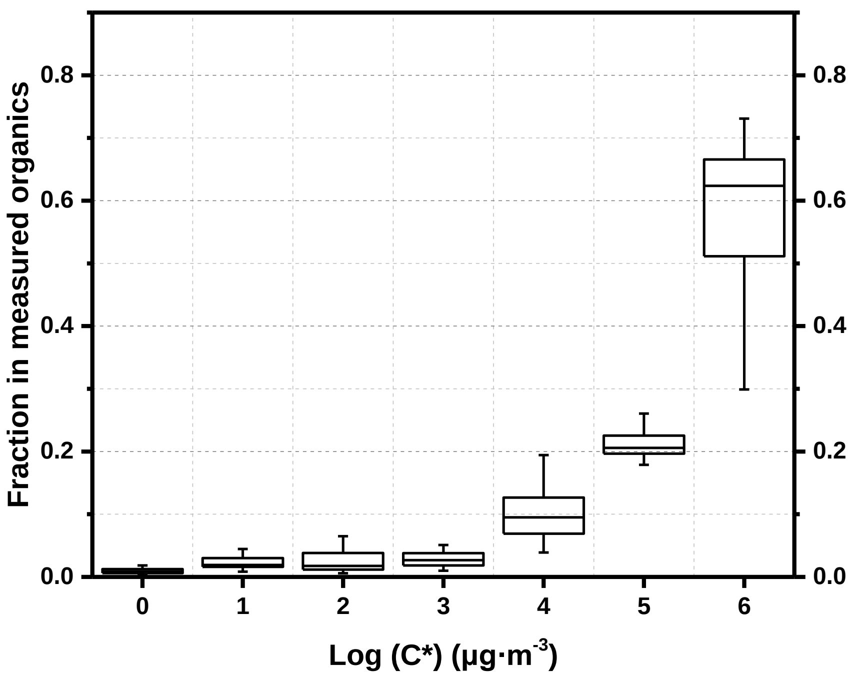

Figure S6. Volatility distribution of organics measured by GC/MS of adsorbent tubes collected during all the tests for the tested China V gasoline vehicle. The boxes represent the 75th and 25th percentiles, with the centerline being the median. The whiskers are the highest and lowest values.

Table S3. Median volatility distribution of IVOCs, SVOCs obtained by GC-MS analysis of Tenax tubes, C* 100 to 106 μg m^{-3}) as a function of effective saturation concentration (C*, μg·m^{-3}) at 298 K.

Log (C*)	**50th**
0	0.009
1	0.019
2	0.018
3	0.027
4	0.095
5	0.206
6	0.624

2. In the "atmospheric implications" section, I would suggest to at least mention the possible limitations of the study, and maybe possible future directions. One example could be the fact that only one vehicle was tested (China V), and different values might be obtained for different vehicles (even vehicles that meet the China V emission standard), implying that the total uncertainty associated with the estimated emission factors might be larger. Also, when discussing why your estimate of total IVOC emissions in China is conservative (lines 476-480), can you report what is the current percentage of vehicles that meet the China V standard in the Chinese car fleet? This would help the reader understanding the extent of the implications of the assumption made in estimating that the total IVOC emissions in China

are 30 Gg.

Response: Thank you for your comment. The representativeness of singular tested car will cause uncertainties which restricts the future model simulation. Therefore, we mentioned repeatedly in the manuscript that our result is only based on the tested China V car. We also compared our results with US vehicles in different controlling stages to verify the representativeness of our tested vehicle. Though some uncertainty may exist, the tested car still has its representativeness. More importantly, the aim of this study is to compare the IVOCs emissions under different conditions so as to provide effective suggestions for developing new technologies to reduce pollution from vehicles and making controlling policies in future vehicular management. For this reason, we think singular tested vehicle can consistently evaluate the effects of different factors on IVOC emissions.

We agree with the reviewer that possible limitations of the studies and the current situation of Chinese gasoline vehicles should be mentioned in the implication part.

The Atmospheric Implications part has been altered accordingly. More details could be found as follows (line 542-550):

“Though we have discussed the influences of different operating conditions on IVOC emissions and SOA formation for the tested China V gasoline vehicle, due to the singular vehicle tests of our study, more research i.e. vehicles meeting different emission standards, different engines should be performed both to testify the accuracy of our research and to get a full understanding of the IVOC emission inventory for Chinese gasoline vehicles. Furthermore, advanced measurement techniques e.g. GC×GC-MS and chemical ionization mass spectrometry (CIMS) should be used to obtain a comprehensive molecular-level picture of the total organics so as to reduce the uncertainties associated with the measurements and models."

We have also included the current percentage of vehicles that meet the China V standard in the Chinese car fleet into the manuscript according to the reviewer's comment. Details could be found in the revised manuscript (line 518-528).

"Till the end of 2018, the total vehicle population in China reached 0.327 billion, with automobiles contributing 61% (0.24 billion). Of all the automobiles, gasoline-fueled car took the dominant (88.1%)…. According to the statistics from the Ministry of Ecology and Environment, only 30.9% of the vehicles in 2018 meet the standards of China V. Indeed, higher percentage of pre-China V e.g. China I-IV standard cars will cause more IVOCs emission. In addition, the IVOC/NMHC ratio of diesel vehicles could be much higher than that of the gasoline vehicles (Zhao et al., 2016, 2015). This may also lead to an underestimation."

3. In Section 3.3, you mention several times that recent Chinese regulations failed in controlling PM emissions (and IVOC emissions as well), whereas they were effective for NOx and THC, according to your data. Can you expand on that? Which regulations did they implement? Why do you think they were ineffective for PM and IVOCs but effective for NOx and THC? Maybe some additional references might help – Expanding the discussion on this point can be useful to guide policymaking.

Response: Thank your comment.

The reason why we get this conclusion is that roughly the standard of China V is comparable to US LEV-2 vehicles. While NO_x and THC EFs fall into the range of US LEV-2 vehicle, PM and IVOC EFs lie in the range of pre-LEV and LEV-1. Therefore, in our opinion, compared with NO_x and THC, the effectiveness of the PM and IVOCs control is (at least) not as good as that for NO_x and THC.

We agree with the reviewer that expansion of the discussion will be more useful both for readers and for policy making. The manuscript has been altered as following (line 323-343):

"In addition, we compared our results with that from European vehicles, and found that the NO_x and THC EFs for the tested vehicle were lower than Euro 5 gasoline vehicle, while the PM EF was higher (Fontaras et al., 2014). This suggests that compared with US and

European vehicles, the stringent emission implemented by Chinese government have been effective at controlling NOx and THC, but might be inefficient to PM emissions. For past 30 years, Chinese government has adopted a series of emission control policies and measures for light-duty vehicles, including implementation of emission standards for new vehicles promotion of sustainable transportation and alternative fuel vehicles, and traffic management programs (Wu et al., 2017; Zhang et al., 2014). Wu et al. (2017) summarizes the implementation of the vehicle control policies in China, which shows the control for the vehicular pollutants is becoming stricter step by step. For example, the NO_x emission standard changed from 0.15 $g \cdot km^{-1}$ to 0.035 $g \cdot km^{-1}$ while the standard changed from China III to China VI. Different from NOx and THC which has been controlled since China III, only when in 2017, China V standard first introduced the control of PM into the emission control scope. Yang et al. (2020)investigated the effects of gasoline upgrade policy on migrating the PM pollution in China and found that there's no much space for significantly reducing the PM concentration by simply improving the gasoline quality. Therefore, for PM control, more policies i.e. developing cleaner alternatives to fossil fuels, replacing traditional vehicles with new-energy and building developed public transport system should be done."

References:

Fontaras, G., Franco, V., Dilara, P., Martini, G., and Manfredi, U.: Development and review of Euro 5 passenger car emission factors based on experimental results over various driving cycles, Science of The Total Environment, 468-469, 1034-1042, https://doi.org/10.1016/j.scitotenv.2013.09.043, 2014.

Wu, Y., Zhang, S., Hao, J., Liu, H., Wu, X., Hu, J., Walsh, M. P., Wallington, T. J., Zhang, K. M., and Stevanovic, S.: On-road vehicle emissions and their control in China: A review and outlook, Science of The Total Environment, 574, 332-349, 2017.

Yang, G., Zhang, Y., and Li, X.: Impact of gasoline upgrade policy on particulate matter pollution in China, Journal of Cleaner Production, 262, 121336, 10.1016/j.jclepro.2020.121336, 2020.

Zhang, S., Wu, Y., Wu, X., Li, M., Ge, Y., Liang, B., Xu, Y., Zhou, Y., Liu, H., Fu, L., and Hao, J.: Historic and future trends of vehicle emissions in Beijing, 1998–2020: A policy assessment for the most stringent vehicle emission control program in China, Atmospheric Environment, 89, 216-229, https://doi.org/10.1016/j.atmosenv.2013.12.002, 2014.

4. Some claims in the introduction can be better substantiated by referencing previous literature. E.g. lines 58-59 "A large discrepancy remains between modeled and measured SOA. One possible reason is missing SOA precursors." Two recent modeling works that discussed these two points are Giani et al. [2019] in Europe and [Huang et al., 2020] in China, and I suggest to add a citation to strengthen your claims. In the introduction, I would also stress the point that understanding and characterizing IVOC emissions, as well as their volatility distributions, is crucial for improving numerical models that aim to predict OA.

References

Giani, P., A. Balzarini, G. Pirovano, S. Gilardoni, M. Paglione, C.

Colombi, V. L. Gianelle, C. A. Belis, V. Poluzzi, and G. Lonati (2019), Influence of semi-and intermediatevolatile organic compounds (S/IVOC) parameterizations, volatility distributions and aging schemes on organic aerosol modelling in winter conditions, Atmospheric environment, 213, 11-24.

Huang, L., Q. Wang, Y. Wang, C. Emery, A. Zhu, Y. Zhu, S. Yin, G. Yarwood, K. Zhang, and L. Li (2020), Simulation of secondary organic aerosol over the Yangtze River Delta region: The impacts from the emissions of intermediate volatility organic compounds and the SOA modeling framework, Atmospheric Environment, 118079.

Zhao, Y., N. T. Nguyen, A. A. Presto, C. J. Hennigan, A. A. May, and A. L. Robinson (2016), Intermediate volatility organic compound emissions from on-road gasoline vehicles and small off-road gasoline engines, Environmental science & technology, 50(8), 4554-4563.

Response: Thank you for your comment. We have modified the manuscript according to the reviewer's comment. Details can be found as following (line 71-82):

"Recent model studies have shown that adding IVOC emissions into different models will greatly improve the SOA simulation results. For example, Giani et al. (2019) found a considerable OA enhancement in Po Valley (Northern Italy) when applying new S/IVOCs emission estimates and the new volatility distributions into CAMx, in which the improvement in SOA mainly due to the revised IVOC emissions. Huang et al. (2020) found a similar enhancement in SOA simulation for Yangtze River Delta (Southeast China) region when adding IVOC emissions into CAMx. They also show the importance of volatility distribution and emission parameterization for the model simulation. Therefore, understanding and characterizing IVOC emissions, as well as their volatility distributions, is crucial for improving numerical models that aim to predict OA."

Reference:

Giani, P., Balzarini, A., Pirovano, G., Gilardoni, S., Paglione, M., Colombi, C., Gianelle, V. L., Belis, C. A., Poluzzi, V., and Lonati, G.: Influence of semi- and intermediate-volatile organic compounds (S/IVOC) parameterizations, volatility distributions and aging schemes on organic aerosol modelling in winter conditions, Atmospheric Environment, 213, 11-24, 10.1016/j.atmosenv.2019.05.061, 2019.

Huang, G., Liu, Y., Shao, M., Li, Y., Chen, Q., Zheng, Y., Wu, Z., Liu, Y., Wu, Y., Hu, M., Li, X., Lu, S., Wang, C., Liu, J., Zheng, M., and Zhu, T.: Potentially Important Contribution of Gas-Phase Oxidation of Naphthalene and Methylnaphthalene to Secondary Organic Aerosol during Haze Events in Beijing, Environmental Science & Technology, 53, 1235-1244, 10.1021/acs.est.8b04523, 2019.

Huang, L., Wang, Q., Wang, Y., Emery, C., Zhu, A., Zhu, Y., Yin, S., Yarwood, G., Zhang, K., and Li, L.: Simulation of secondary organic aerosol over the Yangtze River Delta region: The impacts from the emissions of intermediate volatility organic compounds and the SOA modeling framework, Atmospheric Environment, 118079, 10.1016/j.atmosenv.2020.118079, 2020.

5. I am a little skeptical about the parametrization presented in Section 3.5, which seems somewhat arbitrary. Does the logarithmic curve have some sort of physical insight or is it based only on the shape of the calculated curve? Why not using something like k-exp(. . .) as in the

actual model used to derive that curve (Equation in Section 3.4), also because you're claiming that after 24h SOA/POA is approximately constant? The other concern that I have is that there are a lot of parameters to be estimated (9 in high-NOx conditions), which might cause overfitting to your data, thus losing generalizability. Is it a specific reason why you're using so many parameters? Is there a way of having a simpler parametrizations with similar fit performance? If so, a simpler model (i.e. with less parameters) should be preferred. I would suggest that at least you should better justify your choices for the proposed parametrization in Section 3.5. I believe that Section 3.5 can be largely improved, either by better substantiating your choices or performing some further calculations (that might exceed the scope of the paper, though).

Response: We thank the reviewer for the comment.

We agree with the reviewer that the introduction of more parameters will bring more uncertainties. The logarithmic curve was chosen due to the shape and the fitting results. There might not be accurate physical meaning of these parameters or we should study it deeper in future studies. Actually, in this part, we just try to establish some empirical formulas to help understanding the SOA formation of the gaseous precursors, especially the missing IVOCs. As previous studies have shown that the SOA yields for different IVOCs and VOCs depend strongly on the OA loading under high NO_x conditions, which can have significant influences on the model simulation results. In order to get a comprehensive understanding of these effects, we introduce these parameters. According to these seemingly complicated parameters, we could roughly estimate the SOA formation based on the OA concentration and the photochemical age which might, at least, provide a way for us to calculate the SOA formation.

More research should be done to testify the effectiveness and representativeness of these empirical equations. In fact, we are now expanding our research to do more sensitive analysis of the SOA formation using CMAQ coupled with VBS which might answer the reviewer's comment. This work is now in progress and might be submitted to ACP in several months.

6. What are the dots in Figure 5? Please explain in the caption. (I'm assuming is the SOA/POA ratio to be read on the right scale?)

Response: Thank you for your comment. We have added the description of the blue dots in figure caption which can be found as follows: "The blue circles represent the SOA-to-POA ratio after 48 h of photooxidation (right axis)".