Response to reviewer #1

In urban areas IVOCs are among the most important and yet least understood precursors to secondary organic aerosols. Tunnel test is a widely adopted approach to characterize vehicle emissions of air pollutants as it can obtain results representing real-world emissions with a large number of driving vehicles involved. This study is the first tunnel test conducted to characterize vehicular emission of IVOCs. As the field campaign was carried out in a very busy urban tunnel with traffic flows of over 30,000 vehicles per day, the results from this study are valuable and implicative for the emission reduction of IVOCs, which is of great importance for SOA to further alleviate air pollution due to fine particles in urban areas. The data quality is good with available QA/QC procedures, and the manuscript is well written and organized. I recommend its publication in the form of "measurement report" in ACP after addressing the following comments.

Specific comments:

Lines 75-79: the sentences are a little confusing, rewrite it.

Response: We have rewritten the sentences as:

"However, driving conditions significantly influence vehicular IVOCs emissions (Drozd et al., 2018; Tang et al., 2021), therefore emissions of IVOCs under real-world driving conditions may be quite different from that measured with chassis dynamometers." (*Line 75-78*)

Lines 88-91: "For this reason, IVOC emission factors derived from vehicle tests in the US have been used to update China's emission inventories with the inclusion of IVOCs (Liu et al., 2017). It is unknown whether the borrowed emission factors could well reflect the vehicular emissions of IVOCs in China.", consider to change as "As IVOC emission factors derived ……(Liu et al., 2017), it is unknown……".

Response: Thanks. Revised as suggested. (*Line 87-89)*

Lines 125-126: "classify the vehicle types", change to "classify vehicles into different fuel types".

Response: Revised as suggested. (*Line 124-125*)

Lines 129-131: "…a gas chromatography / mass selective detector (GC/MSD; Agilent, 7890

GC/5975 MSD, USA) with a capillary column (Agilent, HP-5MS, 30 m × 0.25 mm ×0.25 μm).” Should be “…a gas chromatography / mass selective detector (GC/MSD; 7890 GC/5975 MSD, Agilent Technologies, USA) with a capillary column (HP-5MS, 30 m × 0.25 mm × 0.25 μm, Agilent Technologies, USA).”

Response: Revised as suggested. (*Line 128-130)*

Line 143: the retention time: the retention times; of n-alkane: of a n-alkane

Response: Revised as suggested. (*Line 142*)

Line 148: in the 11 bins.

Response: Revised as suggested. (*Line 147-148*)

Line 169: “occurs” à “occurred”.

Response: Revised as suggested. (*Line 168*)

Lines 171-172: “IVOCs detected in the second tube only accounted for 2.6 ± 1.4% of the total in the two tubes, indicating no breakthrough during the sampling”: as 2.6% detected in the second tube, why no breakthrough?

Response: We have revised “no breakthrough” as “negligible breakthrough”. (*Line 171*)

Line 181: of a given species.

Response: Revised as suggested. (*Line 180*)

Line 192: 76.3% on average.

Response: Revised as suggested. (*Line 191*)

Line 193: a percentage of: an average percentage of.

Response: Revised as suggested. (*Line 192-193*)

Line 204: change “diesel” to “DVs”.

Response: Revised as suggested. (*Line 203*)

Line 217 “http://www.mee.gov.cn/”: it just directs to the web site of MEE, but not to the specific documents with the relevant information. Try to be more specific or you need cite other appropriate references.

Response: Revised as suggested. The following link directly provides proportion of China III or lower emission standard diesel vehicles in 2019. (http://www.mee.gov.cn/xxgk2018/xxgk/xxgk13/202012/t20201201_810776.html) (*Line 216*)

Lines 225-226: "GVs still share a much larger portion than the China V and VI ones in the on-road fleets (http://www.mee.gov.cn/)." Change to "……ones in China's on-road fleets (http://www.mee.gov.cn/....)", as mentioned above, the website address should be more specific so that the readers can easily find information about fleet compositions in China.

Response: Revised as suggested. (*Line 225-226*)

Lines 231-233: "The speciated IVOCs consist of n-alkanes, b-alkanes and PAHs. Naphthalene dominated the quantified PAHs, accounting for 56.82 ± 1.21% of total PAHs emissions." change to "Among the speciated IVOCs (Table S1), naphthalene dominated the quantified PAHs, accounting for 56.82 ± 1.21% of total PAHs emissions."

Response: Revised as suggested. (*Line 231-232*)

Line 243: bins.

Response: Revised as suggested. (*Line 243*)

Lines 244-245: "The mass ratios of IVOCs in each bin to the n-alkane in the same bin ranges 9.0-15.8 (Table S2). As n-alkanes are more easily and routinely quantified, the relationships of…" rewrite to "The mass ratios of IVOCs to the n-alkane in the bins ranged 9.0-15.8 (Table S2). As n-alkanes can be more easily and routinely quantified, the relationships of…".

Response: Revised as suggested. (*Line 243-245*)

Line 248: consider changing to "as the results here were obtained for a fleet dominated by GVs"

Response: Revised as suggested. (*Line 247*)

Line 274: "totalled": "totaled"

Response: Revised as suggested. (*Line 274*)

Line 291: "ontained in a tunnel": "obtained from this study in a tunnel"

Response: Revised as suggested. (*Line 294-295*)

4. Conclusions and implications: in this part the authors present many emission estimates and percentages, some of which cannot be directly figured out by the readers. Better added some explanations in the supporting information about these.

Response: Thank you for the suggestions. As suggested, to make it more informative to readers, we have added explanations in the Supporting Information as below:

Text S4

Estimations of IVOCs emission

Firstly, we used the mileage-based EF_{IVOCs} and the average vehicle fleet composition observed in tunnel to calculate IVOCs emissions percentage of DVs and GVs (Table S4). Then, as showed in Table S4, the fuel-based EF_{IVOCs} and fuel consumptions in China in 2019 (http://www.mee.gov.cn/hjzl/sthjzk/ydyhjgl/) were used to estimate IVOCs emissions from diesel- and gasoline-fueled engines.

Table S4. Estimations of IVOCs emission from on-road DVs and GVs and from diesel- and gasoline-fueled engines.

	Diesel vehicles	Gasoline vehicles
Mileage-based EF (mg km^{-1})	62.79±18.37	13.95±1.13
Fleet composition	5%	95%
IVOCs emission percentages	19.1%	80.9%
	Diesel-fueled engines	**Gasoline-fueled engines**
Fuel-based EFs (mg kg^{-1})	984.9±288.2	239.5±19.5
Fuel consumptions (Tg)	150	120
IVOCs emissions (Gg)	147.7	28.74

Line 306: "revealed complex and different results", it should be more specific.

Response: Thank you for the suggestions. As suggested, we added some sentences in revised manuscript:

"our tunnel tests for on-road fleet revealed that although the ratios of IVOCs-to-POA and IVOCs-to-NMHCs were comparable to that from previous chassis dynamometer tests, no significant positive correlations were found between IVOCs and POA or NMHCs in our tunnel measurements." (*Line 308-311*)

Line 339: “on-road diesels are comparable to the non-road diesel engines” change to “on-road diesel vehicles are comparable to that for non-road diesel engines”.

Response: Revised as suggested. (*Line 344-345*).

Response to reviewer #2

General Comments:

In general the manuscript is well constructed and easy to follow. To my understanding, less previous studies were focus on investigation of real-world IVOC fleet emissions, this study showed that there are still some knowledge gaps between the real-world situation and laboratory-based results of vehicular IVOCs emissions. However, besides showing the measurement results and some relation analysis, there is a lack of in-depth data analysis and discussion, which could be attributed to less supporting data from other sources (only concurrent VOCs results were used, not even showed). Overall, the manuscript is recommended to be published in the form of "measurement report" after necessary revision.

Specific Comments:

Line 196-197: Here the Emission Factors (EF_{IVOCs}) for both GVs (13.29 ± 5.08 mg km^{-1} veh^{-1}) and DVs (21.40 ± 5.01 mg km^{-1} veh^{-1}) were determined from Equation (1). Similar parameters were estimated by Equation (2) with different outcomes showed in Line 205-206 (13.95 ± 1.13 mg km^{-1} veh^{-1} for GVs and 62.79 ± 18.37 mg km^{-1} veh^{-1} for DVs). It is unclear that how to obtain these values directly from Equation (1). To my understanding, the only affecting variable would be the vehicle count, but in this case it is difficult to tell how much of EF_{IVOCs} are attributed to GVs and DVs, respectively. Please explain.

Response: Thank you for the comments. There might be some misunderstanding. Equation (1) was used to calculate the fleet-average emission factor of IVOCs (EF_{IVOCs}), which ranged from 13.29 ± 5.08 mg km^{-1} veh^{-1} to 21.40 ± 5.01 mg km^{-1} veh^{-1}. Based on equation (2) (Ho et al., 2007; Kramer et al., 2020), the EF_{IVOCs} of GVs and DVs could be derived via linear regression as 13.95 ± 1.13 mg km^{-1} veh^{-1} and 62.79 ± 18.37 mg km^{-1} veh^{-1}, respectively. We have also modified this part of writing as below:

"Based on above equation (1), fleet-average EF_{IVOCs} (GVs + DVs) ranged from 13.29 ± 5.08 mg km^{-1} veh^{-1} to 21.40 ± 5.01 mg km^{-1} veh^{-1}, with an average of 16.77 ± 0.89 mg km^{-1} veh^{-1} (Average ± 95% C.I.)." (*Line 195-197*)

References:

Ho, K. F., Ho, S. S. H., Cheng, Y., Lee, S. C., and Yu, J. Z.: Real-world emission factors of fifteen carbonyl compounds measured in a Hong Kong tunnel. Atmos. Environ., 41, 1747-1758, https://doi.org/10.1016/j.atmosenv.2006.10.027, 2007.

Kramer, L. J., Crilley, L. R., Adams, T. J., Ball, S. M., Pope, F. D., and Bloss, W. J.: Nitrous acid (HONO) emissions under real-world driving conditions from vehicles in a UK road tunnel. Atmos. Chem. Phys., 20, 5231-5248, https://doi.org/10.5194/acp-20-5231-2020, 2020.

Line 289-290: It is mentioned that there is a difference between the SOA_{IVOCs}-to-SOA_{VOCs} ratio for DVs and GVs. Is it possible to differentiate this from your data set (say utilizing the principle of Equation (2))?

Response: Thank you for the comments. According to the reviewer's suggestions, we tried to use the principle of Equation (2) to resolve the SOA_{IVOCs}-to-SOA_{VOCs} ratios for DVs and GVs, which was shown in the following figure. Although the correlation between SOA_{IVOCs}-to-SOA_{VOCs} ratios and DVs fraction was not significant, SOA_{IVOCs}-to-SOA_{VOCs} ratio of DVs (54.9) did present much higher value than that of GVs (6.82). We also added this result to our revised manuscript as below:

"Furthermore, we also resolved the SOA_{IVOCs}-to-SOA_{VOCs} ratios for DVs and GVs via liner regression (Text S3). As shown in Fig. S7, although the correlation between SOA_{IVOCs}-to-SOA_{VOCs} ratios and DV fractions was not significant, the DVs did present much higher average SOA_{IVOCs}-to-SOA_{VOCs} ratio (54.9) than that of GVs (6.82)." (*Line 290-294*).

"**Text S3**

Linear regression analysis of diesel vehicles fraction and SOA_{IVOCs}-to-SOA_{VOCs} ratio

$$R = R_{DVs} \times \alpha + R_{GVs} (1 - \alpha)$$

where R represents the fleet average SOA_{IVOCs}-to-SOA_{VOCs} ratio calculated during the campaign. R_{DVs} and R_{GVs} are the SOA_{IVOCs}-to-SOA_{VOCs} ratios for DVs and GVs, respectively. α is the fraction of DVs in the total IVOCs-emitting vehicles traveling through the tunnel." *(Supporting information)*

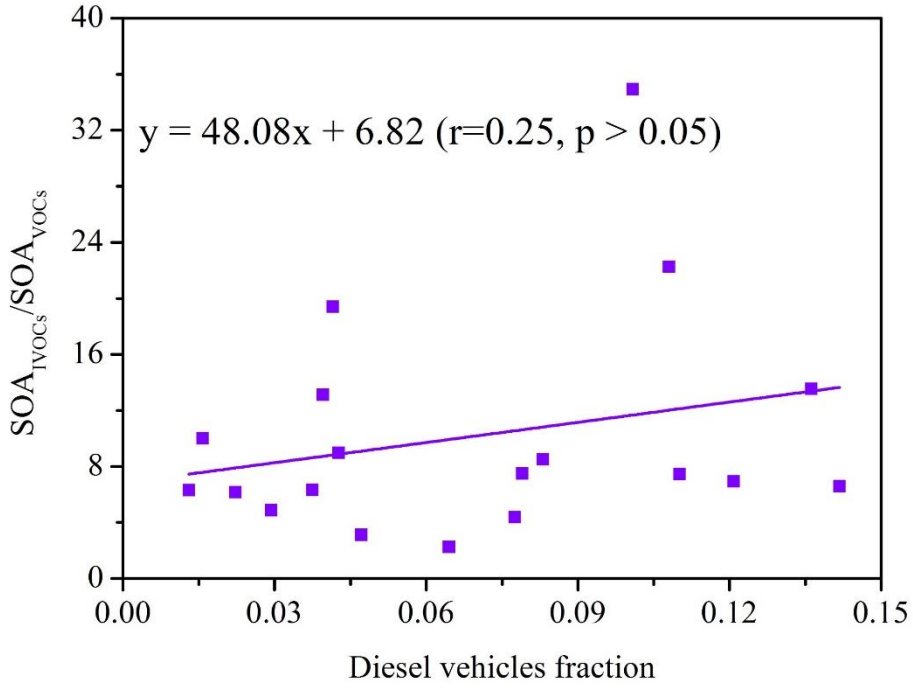

Figure S7. Linear regression analysis of diesel vehicles fraction and SOA_{IVOCs}-to-SOA_{VOCs} ratio