

Response to Reviewers

Reviewer #1

The manuscript provides a unique and valuable dataset on vehicular VOC emissions from the Tibetan Plateau, highlighting the significant role of low atmospheric pressure in enhancing evaporative emissions at high altitudes. This addresses a critical knowledge gap, impacting emission inventories and mitigation strategies. The study's comprehensive methodology is commendable. Addressing the detailed comments, especially regarding the sampling strategy's fit with EF calculations and the comprehensive validation of source apportionment, will significantly strengthen the manuscript and its impact.

Reply: Thank you for your valuable and insightful comments to improve the manuscript. We have carefully considered the comments and revised the manuscript thoroughly and substantially, to address these comments. In the following, please find our detailed responses for the comments. Referee comments are given in *black italics*, and our responses and changes in the manuscript in blue and red, respectively.

Comments:

1.Lines 74-77: This approach of sampling "accumulated air masses" seems to contradict the standard method for calculating fuel-based emission factors (EF) using simultaneously measured CO and CO₂ (Eq. 1), which typically assumes a well-mixed plume representing instantaneous emissions. Please provide a more detailed and rigorous explanation of how the sampling strategy (capturing accumulated air via piston effect) aligns with the EF calculation method. This might involve discussing the length of the tunnels, travel speed, and how "accumulation" truly translates to the average emission.

Reply: Thank you for your insightful comment and we apologize for the lack of clarity in our original description. Upon reviewing your concerns, we would like to clarify that our sampling strategy is indeed consistent with the standard approach for calculating fuel-based emission factors (EFs) in tunnel studies. Both approaches assume that in a one-way tunnel (7/10 tunnels), the vehicle-emitted gases accumulate

at the end of the tunnel, as validated by our real-time CO₂/CO monitoring. Consequently, the air at the tail end represents a well-mixed emissions plume from all vehicles in the tunnel.

To ensure our sampling strategy was representative of vehicle emissions within the tunnel environment, we took the following measures: we selected tunnels that were as long as possible, conducted sampling near the rear section (starting at approximately two-thirds of the tunnel length), maintained consistent vehicle speed during sampling, standardized sampling duration to 1 minute per sample, and repeated sampling multiple times to ensure representativeness. For the three bidirectional tunnels, we adjusted our sampling strategy by collecting air samples at the midpoint of the tunnel rather than at the rear, in order to reduce the interference caused by opposing airflows and ensure a representative mixture of emissions for both directions. This midpoint sampling strategy helps to minimize spatial gradients and turbulence near the entrances and exits, as recommended by prior tunnel sampling protocols.

We have revised the manuscript to provide a more detailed and rigorous description of the sampling methodology, as follows:

Section 2.1 Line 61-63: Following the criteria of representative altitude, we specifically chose ten tunnels located between Lhasa and Nyingchi, two major cities in Tibet autonomous region, China (Fig S1). We prioritized selecting one-way tunnels, as well as the longest available tunnels.

Section 2.2 Line 78-85: In the one-way tunnels, the online data (i.e., CO₂ and CO) showed a noticeable piston effect (Fig. S3) (Chung and Chung, 2007), with concentrations gradually increasing towards the end of the tunnel. The air at the tunnel's tail end was assumed to represent a well-mixed plume from emissions of all vehicles in the tunnel (Hwang et al., 2023; Gillies et al., 2001). Therefore, in these tunnels, offline sampling was initiated in the rear section and lasted approximately 1 minute to capture the accumulated air masses. Additionally, three tunnels in our study had bidirectional traffics, where the piston effect was less pronounced due to opposing flows. For these

cases, sampling was conducted at the tunnel midpoints to ensure representative mixing of emissions from both directions. Background concentrations of VOCs were determined at the Yangbajing background site during the same field campaign of STEP (July-August, 2022) (Tao et al., 2024).

2. Please clarify what “59 species, including those common to this work and other research endeavors” (Line 126) precisely means. Is this a consistent subset used for comparison across studies?

Reply: Thank you for raising this important point for clarification. Yes, this is a consistent subset used for standardized comparison across studies. We have revised the text to enhance clarity.

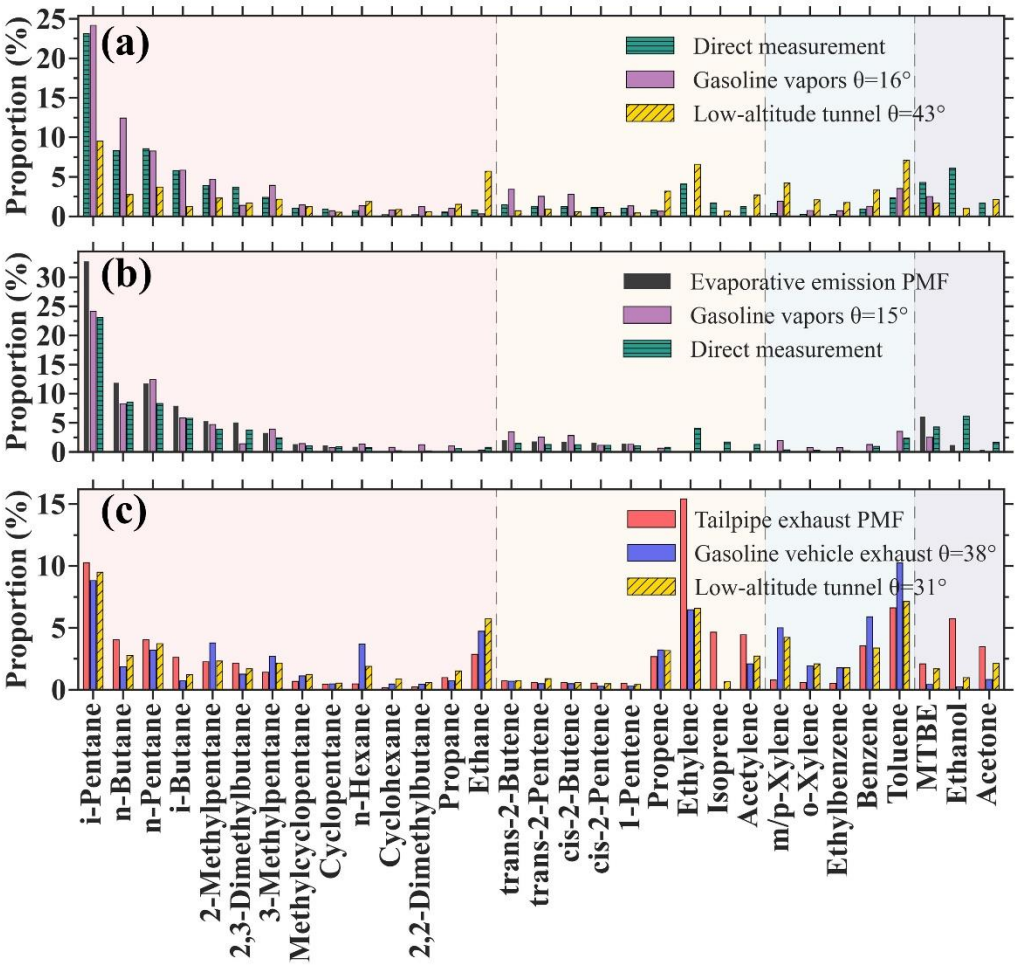
Figure 1. Line 134-136: The solid line in (a) represents a subset of 59 VOC species that overlapped with species reported in key low-altitude tunnel studies (e.g., Ho et al., 2009; Chiang et al., 2007).

We have also supplemented relevant content in Section 3.1 to enhance its clarity.

Section 3.1 line 125-126: For cross-study comparison, a consistent subset of 59 VOC species, commonly detected in both our study and low-altitude tunnel studies (e.g., Ho et al., 2009), was selected to ensure comparability in EF and ER calculations.

3. The observation that PMF-resolved tailpipe exhaust (Factor 3) shows “relatively poor similarity” (38°) with chassis dynamometer-tested gasoline vehicle exhaust (Figure 3c, Table 1). The authors attributed this potentially to “the influence of diesel vehicles, as well as potential influences from other sources.” Can the authors quantify the likely contribution of diesel vehicles in these tunnels or explain why their influence leads to such a discrepancy? Were diesel vehicles included in the comparison dynamometer data?

Reply: We sincerely apologize for the typographical error in Figure 3c, where the similarity angle was mislabeled as 41° instead of the correct 38° (as presented in Table 1). This has been corrected in the revised manuscript. Below, we address your insightful query about the potential influence of diesel vehicles on our results.



Our inference of potential influences from diesel vehicles is based on two key observations. First, during sampling, diesel trucks were occasionally observed passing through the tunnels. Second, diesel exhaust exhibits a distinctly different VOC profile compared to gasoline emissions, typically characterized by higher proportions of heavier alkanes and aromatic compounds (Wang et al., 2022; Schauer et al., 2002; Chen et al., 2025; Zhao et al., 2022), which was also detected in our measured profiles. Although our current data does not allow for a quantification of the contribution from diesel emissions, we posit that their overall impact is likely limited. This assessment is supported by the excellent consistency ($\theta = 15^\circ$) observed between our direct

measurements and the characteristic profile of gasoline vapors. Given the anticipated minor influence of diesel vehicles, we did not incorporate diesel-specific dynamometer data into our comparative analysis.

4. *The presented average EFs and ERs come with relatively high standard deviations (e.g., EF of $3.3 \pm 3.1 \text{ ug}\cdot\text{kgfuel}^{-1}$, ER of $87 \pm 92 \text{ ppb/ppm}$). Does it reflect differences in vehicle types, driving conditions within tunnels, or other factors? How does this high variability impact the statistical significance of the observed altitude trends?*

Reply: Thank you for this insightful question regarding the high variability in our reported EFs and ERs. We agree that the substantial standard deviations reflect the complex interplay of multiple factors inherent in real-world tunnel studies, including altitude variations, tunnel characteristics, vehicle types, driving conditions, and environmental parameters. Such heterogeneity is intrinsic to field-based measurements and aligns with prior studies (Zhai et al., 2020; Zhang et al., 2024).

Despite this variability, our extensive sampling strategy (n=46 valid samples across 10 tunnels) provided sufficiently statistical power to identify significant emission enhancements at high altitudes. Specifically, EFs and ERs were substantially elevated—by factors of 1.9 to 3.9—compared to low-altitude sites. Moreover, key evaporative species such as butanes and pentanes exhibited a clear monotonic rise with altitudes and contributed 20~50% to total VOCs emissions, underscoring the role of low-pressure-enhanced evaporation. These findings confirm that altitude-dependent emission patterns dominate over variability.

5. *Comparisons to low-altitude tunnels in Hong Kong, Taiwan, Tianjin, Henan, and Haikou are valuable. However, fleet compositions, fuel standards, and driving conditions can vary significantly across these regions and study years. Briefly acknowledge these potential differences and how they might affect direct comparisons.*

126 Reply: Thank you for this valuable suggestion. We fully agree that vehicle fleet
127 composition, fuel quality, and driving conditions vary among different cities and time
128 periods, and such differences can influence VOC emission characteristics. To address
129 this concern, we have revised the text in section 3.4 “Source apportionment of VOCs
130 in plateau tunnels” (Lines 253-256) to acknowledge these differences.

131 While variations in vehicle fleet composition, fuel quality, and driving conditions
132 across different cities and time periods can significantly influence VOC emission
133 characteristics in low-altitude studies, our high-altitude tunnel measurements
134 consistently demonstrate systematically elevated evaporative emissions.

136 6. The absence of an altitude-specific distribution for the CO/CO₂ ratio is interesting
137 given theoretical expectations. While attributed to “other factors”, please elaborate on
138 this, potentially with supporting evidence explaining why altitude isn't the dominant
139 influence.

140 Reply: Thank you for this thoughtful comment. We agree that reduced oxygen
141 concentrations at high altitudes was expected to decrease combustion efficiency and
142 elevate the CO/CO₂ ratios. However, our measurements did not reveal a clear altitude-
143 specific trend in CO/CO₂ ratios, which exhibited a broad range of 5.1 to 11 ppbv/ppmv
144 across all tunnels. This variability is consistent with observations from low-altitude
145 tunnel studies, such as Shing Mun Tunnel in Hong Kong (15 ppbv/ppmv), North 3rd
146 Ring Tunnel in Zhengzhou (4.4 ppbv/ppmv), Guy Môquet Tunnel in Paris (8.44
147 ppbv/ppmv), and Gubrist Tunnel in Switzerland (9.19 ppbv/ppmv). This suggests that
148 non-altitude factors exert considerable influence in real-world settings, thereby
149 obscuring any clear signal attributable to altitude alone.

150 As you suggested, we have revised the manuscript to incorporate this explanation.
151 The revised paragraph is as follows: (Line 175-181)

152 “Moreover, the CO/CO₂ ratio, as an indicator of engine combustion efficiency
153 (Vollmer et al., 2007; Ammoura et al., 2014; Hu et al., 2019), did not appear a

discernible altitude-specific distribution across our dataset (Fig. S8), with average values ranging from 5.1 to 11 ppbv/ppmv. This range is comparable to values reported in low-altitude tunnel studies, spanning 4.4–15 ppbv/ppmv across cities in Asia and Europe (Cui et al., 2018; Liu et al., 2022; Ammoura et al., 2014; Legreid et al., 2007). Although reduced oxygen at higher elevations may theoretically impair combustion efficiency and increase the CO/CO₂ ratio, other factors, such as vehicle type, engine operation conditions, and tunnel ventilation may obscure the effect of altitude alone.”

7. Please clarify what “Direct measurement” refers to in Table 1. Is it the average source profile from all tunnel measurements?

Reply: Thank you for the helpful comment. “Direct measurement” in Table 1 refers to the average source profile derived from all tunnel measurements conducted in this study. To avoid confusion, we have clarified this point by adding the following explanation to the title of Table 1 (Line 212-213):

“Direct measurement refers to the average VOC source profile based on all tunnel samples measured in this study.”

8. Please provide a clearer “good consistency” threshold from the literature (e.g., <20° or <25°) when discussing the 38° for tailpipe exhaust, to better contextualize the PMF factor.

Reply: Thank you for the suggestion. The threshold ranges for profile similarity (θ angle) were actually provided in the Materials and Methods section under “Source profiles similarity analysis” (Line 105-107), where we stated that θ angles of 15°-30° indicate “good consistency” based on previous literature (Wang et al., 2024). A θ angle of 38° thus falls into the “many similarities” category (30°-50°). To improve clarity, we have now added a citation to the relevant literature at the point in the Results and Discussions section where the 38° value is mentioned, to help readers better interpret the classification.

Reviewer #2

This pioneering study reveals unexpectedly high VOC emissions from vehicles on the Tibetan Plateau, with evaporative sources dominating due to low-pressure enhancement—novel and rigorously validated findings. The experimental design (multi-tunnel mobile measurements) and analytical rigor (PMF/NNLS source apportionment) are exceptional. While the proposal for electric vehicle (EV) adoption in Tibet offers a promising pathway for emission reduction, it overlooks severe battery efficiency decay in low-temperature high-altitude environments, weakening policy relevance. I recommend softening the emphasis on this aspect.

Reply: We sincerely thank you for taking the time to review our paper. In the following, we address your comments point by point and revise the manuscript accordingly. For clarity, we list the original comments below in *black italics*, and our responses and changes in the manuscript in blue and red, respectively.

We agree that low temperatures and high-altitude conditions can negatively impact battery performance, including reduced mileage range, charging inefficiency, and cold-start limitations. These environmental constraints could affect the practicality of EV deployment in the Tibetan Plateau. In response, we have revised the section 3.5 to acknowledge these challenges while maintaining a balanced policy outlook. We have also removed the recommendation for large-scale EV adoption from the Abstract. Revised paragraph in section 3.5 Implications and perspectives: (Line 287-295).

One promising approach for Tibet is the strategic promotion of electric vehicles (EVs), supported by China's mature EVs industry and Tibet's abundant renewable energy resources. The region's installed capacity of hydropower and solar energy exceeds 10 million kilowatts (National Energy Administration, 2024), with ongoing development of large-scale solar and wind projects. However, local electricity demand remains low due to sparse population and limited industry, resulting in surplus energy transmitted eastward via the 'West-East Electricity Transmission' project (Xinhua News Agency, 2024). Promoting EVs could absorb this surplus, alleviating grid strain. Despite this,

the deployment of EVs in Tibet faces specific challenges, particularly due to the region's low-temperature environments. These conditions necessitate advancements in battery technology, such as the development of solid-state batteries with improved thermal resilience, as well as altitude-adaptive battery management systems.

Specific comments:

1. Line 38-39: This study emphasizes the importance of non-tailpipe emissions such as evaporative emissions. Therefore, it is necessary to add some descriptions about non-tailpipe emissions. Do non-tailpipe emissions only include the evaporation of fuel and solvents? Furthermore, what does "solvents" refer to? Is it windshield wiper fluid, or automotive surface/interior coatings? How are they emitted? Are they emitted intentionally by humans, or are they continuously emitted like fuel? The literature support provided here may not be sufficient.

Reply: Thank you for raising this important point. In our study, “non-tailpipe emissions” specifically refer to evaporative emissions related to fuels and fuel-related additives, which are continuously or intermittently released through mechanisms such as running loss (during engine operation), hot soak (after engine shutdown), permeation, and diurnal breathing. In this context, “solvents” primarily refer to low-volatility hydrocarbons blended into fuel formulations, rather than materials like windshield washer fluids or interior coatings.

To address this ambiguity, we have revised the relevant paragraph in the Introduction to clarify the scope of non-tailpipe emissions, define what is meant by “solvents,” and explain how such emissions occur. We have also added additional literature references to support this clarification. The revised sentence is as follows: (Line 37-40)

“The former refers to gases emitted from engine systems due to incomplete combustion or unburned fuel (Zhang et al., 2024), while the latter mainly consists of evaporative emissions from fuels and fuel-related additives, released through processes

such as running loss, hot soak, permeation, and diurnal breathing (Zhang et al., 2024; Pierson et al., 1999; Yue et al., 2017; Man et al., 2020; Liu et al., 2015; Harrison et al., 2021).”

2. Line 50: Do the terms Qinghai-Tibet plateau and Tibetan Plateau used in this article convey the same meaning or are there any differences between them?

Reply: Thank you for pointing this out. The terms “Qinghai-Tibet Plateau” and “Tibetan Plateau” are commonly used interchangeably in the literature and refer to the same geographical region. However, to avoid confusion and maintain consistency throughout the manuscript, we have unified the terminology and now consistently use “Tibetan Plateau” in the revised version of the manuscript.

3. Line 53-56 This part seems more like a statement of conclusion, and it is not recommended to place it in section 1 Introduction. It is suggested to revise it.

Reply: Thank you for your constructive suggestion. We agree that the sentences in Lines 53–56 are more suitable for the Results and Discussions or Conclusion sections, rather than the Introduction. Accordingly, we have revised this part of the text to avoid making definitive conclusions too early. The revised paragraph now focuses on the motivation and objectives of the study, while the findings are reserved for later sections.

The revised text in introduction (Line 53–58):

“Within the framework of the second scientific expedition and research program of the Tibetan Plateau (STEP) (Yao et al., 2012; Ye et al., 2023), we conducted vehicular emission measurements in 10 tunnels across the Tibetan Plateau, spanning an altitude range of nearly 3000 m. This unique natural setting enabled us to investigate how vehicular emission characteristics respond to changing elevation, with a particular emphasis on the role of low atmospheric pressure. The study aims to enhance the current understanding of VOC emissions from vehicles in high altitude regions, which remain poorly characterized in existing literature.”

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269 *4. Line 60 and many others: Either use Table x / Figure x or Tab. x / Fig. x. Unify*
270 *throughout the manuscript according the journal's format requirements.*

271 *Reply: We have modified it according to the journal format requirements and*
272 *unified the format throughout the paper.*

273

274 *5. Line 73: This statement states that analysis will be conducted a week after sampling.*
275 *Considering that the entire mobile observation spans a long distance, how many days*
276 *did the sampling last, and are all samples taken at the same time in the same tunnel?*

277 *Reply: Thank you for your insightful question. During the mobile measurements,*
278 *which covered a total distance of approximately 2000 km across the Tibetan Plateau,*
279 *sampling was not conducted simultaneously in all tunnels; instead, it was performed*
280 *sequentially over 10 days. Following sample collection, all VOC canisters were*
281 *transported from Tibet to our laboratory in Guangzhou, a process that took*
282 *approximately one week. We initiated the GC-FID/MS analysis immediately upon*
283 *receipt of the samples, ensuring that all samples were analyzed within one week. To*
284 *enhance clarity in the manuscript, we have revised the relevant sentence in Section 2.2,*
285 *and add more descriptions in SI Text 1 about the test.*

286 *Section 2.2 Line 75-78: The online instruments were pre-calibrated to minimize random*
287 *errors. The VOC samples, collected over a 10-day sampling campaign across multiple*
288 *tunnels, were analyzed within one week after transportation to our laboratory using a*
289 *gas chromatography-flame ionization detection/mass spectrometry (GC-FID/MS)*
290 *system (Text S1), ensuring minimal pre-analysis storage time.*

291

292 *SI Text 1 Line 27-32: “Sampling was conducted over a 10-day period, during which we*
293 *covered a distance of approximately 2000 km. The samples were collected at various*
294 *tunnels along the route, and to minimize the effects of external variables, each tunnel*
295 *was tested in both directions over 4–6 rounds, with a 2-hour window for each test. After*
296 *completing the sampling, the samples were transported to our laboratory in*

approximately one week before commencing the analysis.”

6. Line 74-75: *The piston effect is generally aimed at vehicles traveling in the same direction within the same tunnel. The author mentioned sampling at the rear of the tunnel. Has this study measured tunnels with vehicles traveling in both directions simultaneously? If so, how are these types of tunnels sampled? In addition, please also check the correctness of the references cited in the literature (Chung and Chung, 2007).*

Reply: Thank you for this insightful question. Yes, among the 46 valid samples in our study, three canisters were collected in tunnels with vehicles traveling simultaneously in both directions (bidirectional tunnels). For these specific cases, we adjusted our sampling strategy by collecting air samples at the midpoint of the tunnel rather than at the rear, in order to reduce the interference caused by opposing airflows and ensure a representative mixture of emissions from both directions. This midpoint sampling strategy helps to minimize spatial gradients and turbulence near the entrances and exits, as recommended by prior tunnel sampling protocols.

We have revised the Materials and Methods section to clearly describe this adjustment in our sampling strategy. The revised paragraph is as follows (Line 78-85):

In the one-way tunnels, the online data (i.e., CO₂ and CO) showed a noticeable piston effect (Fig. S3) (Chung and Chung, 2007), with concentrations gradually increasing towards the end of the tunnel. The air at the tunnel’s tail end was assumed to represent a well-mixed plume from emissions of all vehicles in the tunnel (Hwang et al., 2023; Gillies et al., 2001). Therefore, in these tunnels, offline sampling was initiated in the rear section and lasted approximately 1 minute to capture the accumulated air masses. Additionally, three tunnels in our study had bidirectional traffic, where the piston effect was less pronounced due to opposing flows. For these cases, sampling was conducted at the tunnel midpoint to ensure representative mixing of emissions from both directions. Background concentrations of VOCs were determined at the Yangbajing background site during the same field campaign of STEP (July-August, 2022) (Tao et al., 2024).

Regarding the reference to Chung and Chung (2007), we have re-checked the citation and confirm that it correctly refers to a study that includes modeling and discussion of pollutant dispersion mechanisms in traffic tunnels, including the piston effect. The citation has also been double-checked for accuracy and relevance.

7. Line 119-122: *When comparing plateau vs. plain EF/ER, the authors selected only two tunnel examples (Hong Kong 50m and Taiwan 330m). Please include additional examples to enhance representativeness.*

Reply: Thank you for this valuable suggestion. We fully agree that including more low-altitude tunnel studies would enhance the representativeness of the comparison. However, the calculation of emission factors (EF) and emission ratios (ER) in our study relies not only on speciated VOCs data, but also requires corresponding CO and CO₂ measurements to normalize emissions relative to fuel combustion. To date, we were only able to identify two published tunnel studies (Shing Mun Tunnel in Hong Kong and Chung-Liao Tunnel in Taiwan) that provide all the necessary co-measured data (VOCs, CO, and CO₂) in a format that allows consistent EF/ER calculations. Unfortunately, other tunnel studies either do not include CO/CO₂ data or do not report them in sufficient detail for this purpose.

8. Line 137: *Inconsistent spacing around “±” symbols.*

Reply: We have modified in the revised manuscript.

9. Line 137: *Replacing "contributing to" with "accounting for".*

Reply: We have replaced "contributing to" with "accounting for".

Reviewer #3

Hereby I offer only one comment to complement other reviewer's comments on a rather important aspect of the paper. It is not true as the authors stated that "However, as far as we know, the influence of pressure on evaporative emissions has not been documented, posing a challenge to our comprehension of vehicular emissions in high-

altitude regions."

In fact, this effect of evaporative emissions on altitude has been well documented, e.g., in MOVES model by the US EPA (US EPA, 2024, p20, Equation 3-6). Therein, the effect is clearly considered, i.e., "Tank vapor generated depends on the rise in fuel tank temperature (F), ethanol content (vol. percent), Reid vapor pressure (RVP, psi) and altitude". And there is also a table comparing model parameters appropriate for Denver, a city that is ~1700 meters above sea level versus those for at sea level. Therefore, it is crucial for the authors to put their study in the context of what is already known, by changing the statement above to reflect the state of the science, and, more importantly to reconcile the measurement inferred altitude effect with those documented in the literature.

References

USEPA, 2024, *Evaporative Emissions from Onroad Vehicles in MOVES5*, November 2024, EPA-420-R-24-014, <https://nepis.epa.gov/Exe/ZyPDF.cgi?Dockkey=P101CTZI.pdf> (last accessed 8/10/2025).

Reply: Thank you for your critical and insightful comment. We sincerely apologize for the inaccuracy in our original statement regarding the documentation of altitude effects on evaporative emissions. As you correctly pointed out, the MOVES model (USEPA, 2024) indeed incorporates the effects of altitude (pressure) on evaporative emissions. However, empirical measurements under real-world high-altitude conditions remain scarce. In response to your feedback, We have added the suggested reference (USEPA, 2024) as well as another relevant study (Wang et al., 2018) to the reference list. We have also revised the relevant statement in the manuscript to more accurately reflect the current state of knowledge, while highlighting the unique contribution of our study. The amended text now reads:

Section 1 Line 45-52: The evaporation of fuels and solvents is an equilibrium process involving hydrocarbon molecules transitioning between the gas and liquid

phases, governed primarily by temperature and pressure conditions (Huang et al., 2022). Elevated temperatures and low pressures theoretically facilitate fuel evaporation (Huo et al., 2024; Wang et al., 2018). However, in plateau regions, low atmospheric pressure often coincides with cooler temperatures, resulting in competing influences that are not yet fully quantified. Computational models such as MOVES model estimate that tank vapor generation at 1,700 meters above sea level is approximately 1.4 times that at sea level, indicating potentially significant altitude-enhanced emissions (US EPA, 2024). They also underscore the critical need for empirical validation under real-world, high-altitude conditions, which remain severely limited.

In addition, we have updated the section 3.1 to better place our study in the context of existing knowledge on the subject.

Section 3.1 Line 128-132: When comparing the identical compositions, the average EF in plateau tunnels is 1.9 times higher than that in the Shing Mun tunnel in Hong Kong (50 m a.s.l.) (Ho et al., 2009). The determined ER is 3.9 and 1.9 times higher than those in the Shing Mun tunnel and the Chung-Liao tunnel in Taiwan (330 m a.s.l.) (Chiang et al., 2007), respectively. These results are significantly higher than the 1.4 times increase in fuel evaporation observed in Denver (1,700 m a.s.l.) relative to sea level (US EPA, 2024).

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