**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 $CO_2$/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 belows:

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., $CO_2$ 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.

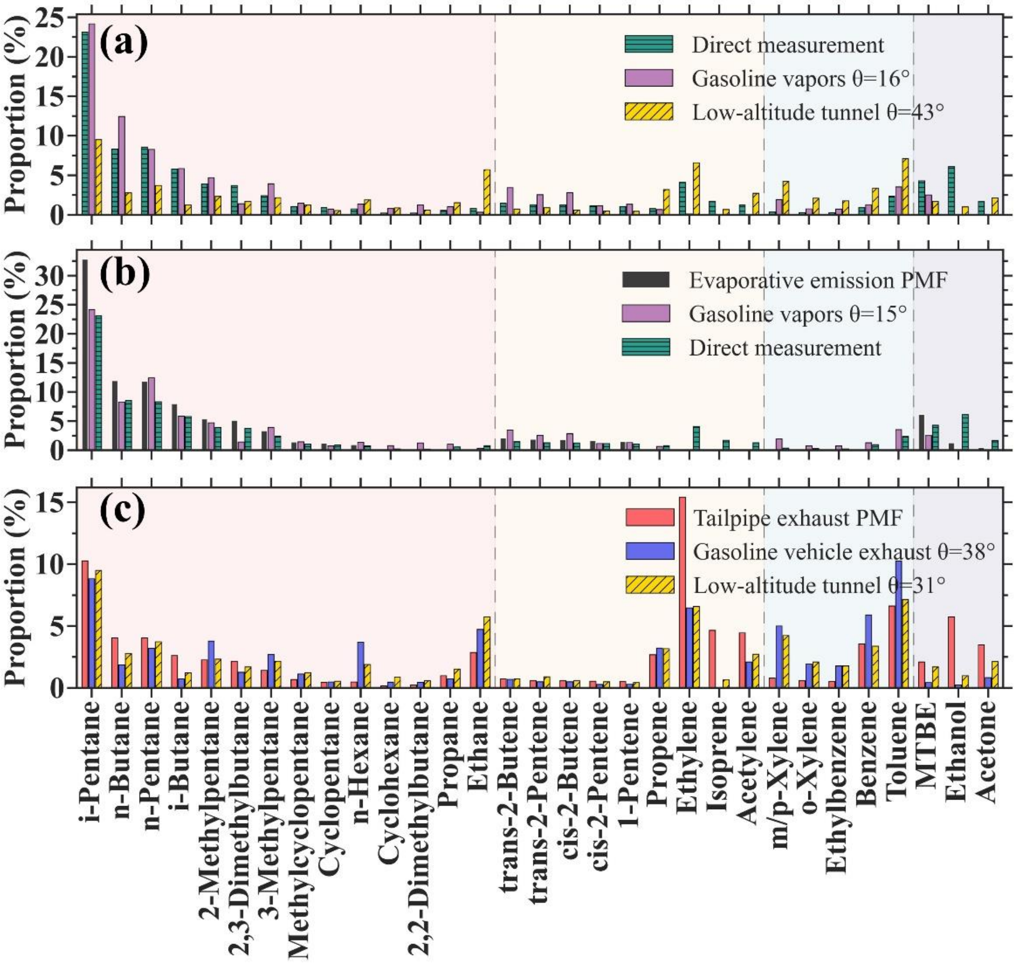
Figure 1. Line 134-136: The solid line in (a) represents a subset of 59 VOC species that overlapped with species reported in key low-altitude tunnel studies (e.g., Ho et al., 2009; Chiang et al., 2007).

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" ($38^{\circ}$) 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 (θ = 15°) 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 ± 3.1 ug·kgfuel⁻¹, ER of 87 ± 92 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.*

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

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

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

Reply: Thank you for this thoughtful comment. We agree that reduced oxygen concentrations at high altitudes was expected to decrease combustion efficiency and elevate the $CO/CO_2$ ratios. However, our measurements did not reveal a clear altitude-specific trend in $CO/CO_2$ ratios, which exhibited a broad range of 5.1 to 11 ppbv/ppmv across all tunnels. This variability is consistent with observations from low-altitude tunnel studies, such as Shing Mun Tunnel in Hong Kong (15 ppbv/ppmv), North 3rd Ring Tunnel in Zhengzhou (4.4 ppbv/ppmv), Guy Môquet Tunnel in Paris (8.44 ppbv/ppmv), and Gubrist Tunnel in Switzerland (9.19 ppbv/ppmv). This suggests that non-altitude factors exert considerable influence in real-world settings, thereby obscuring any clear signal attributable to altitude alone.

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

“Moreover, the $CO/CO_2$ ratio, as an indicator of engine combustion efficiency (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 $CO/CO_2$ 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.

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