Response to RC2: 'Comment on egusphere-2025-4818', Anonymous Referee #2, 21 Dec 2025

The Measurement report "Altitudinal Shift of Ozone Regimes in a Mountainous Background Region" by Yang et al. describes a dataset of observations of ozone and its precursors along an altitudinal gradient on a mountain in south-east China. The measurements cover a period of several months, from March to August and are quite extensive. The authors do not limit themselves to show the observations but also present some analysis of the results, which is commendable. The dataset is novel, in the sense that observations from this part of the world are still sparse, and the conclusions are generally well supported by the data. The manuscript fits the scope of the journal, and I do not see any major issue with it. I recommend publication after the authors have addressed the comments below.

Response: We sincerely thank Reviewer 2 for their thorough and constructive evaluation of our manuscript, "Altitudinal Shift of Ozone Regimes in a Mountainous Background Region" (egusphere-2025-4818). We are grateful for the positive assessment and the specific comments, which have helped us to improve the clarity and robustness of our work. Below, we provide a point-by-point response to each comment. All suggested revisions will be incorporated into the revised manuscript.

Section 2.1: the mountain-foot and mountain-top sites are described as being close to touristic areas, with shops and restaurants. I assume these involve some, although perhaps small, anthropogenic emissions which may affect the interpretation of the results. Also a "eco-friendly vehicle route" is mentioned, but it is not clear what it means, and whether it implies traffic emissions. Can the authors clarify and comment? Were data filtered for local emissions?

Response: Thank you for raising this important point regarding potential local anthropogenic influences near our monitoring sites. We appreciate the opportunity to clarify the measures taken to assess and ensure the integrity of our data with respect to regional background signals.

During data analysis, we systematically evaluated whether low-intensity local activities (such as shuttle traffic and nearby dining facilities) could meaningfully affect the observed vertical gradients. Our assessment included:

1. Comparative analysis: No systematic differences were found in pollutant concentrations between operational and non-operational hours of tourist facilities, or between high- and low-visitation periods.

2. Wind-based filtering: Removing periods with low wind speeds (<1 m s^{-1}) or persistent wind directions from potential local sources did not alter the observed altitudinal trends in O$_3$ or its precursors.

3. Correlation tests: Short-term fluctuations in NO$_x$ and CO showed negligible correlation with tourist activity indicators, but remained strongly associated with regional meteorological drivers (e.g., boundary layer height, humidity).

Based on these analyses, we conclude that while the sites are located near minimal tourist infrastructure, the

influence of these local sources on the reported regional vertical gradients is negligible in both statistical and practical terms. This is due to:

·The very small scale and low emission intensity of the nearby facilities,

·The relatively large distance and topographic separation between sources and monitors,

·The generally good dispersion conditions in this mountainous region.

To provide full transparency, we added a brief statement in Section 2.1 of the revised manuscript describing the local setting and summarizing the outcome of this impact assessment. (line 120-132 in the revised manuscript)

Section 2.3: it is not clear to me why the Random Forest modelling was used. The results reported in Section 3.2 are interesting, but they could easily be obtained by a simple covariance matrix. Can the authors explain why they decided to use a machine learning algorithm? What kind of additional information is being obtained compared to a multivariable correlation plot?

Response: This is a valid question. We chose Random Forest (RF) with SHAP analysis over traditional correlation for several substantive reasons, which provide deeper mechanistic insight:

1. Capturing Non-Linear and Interaction Effects: Ozone formation involves complex, non-linear interactions among precursors and meteorology (e.g., the NO_x-VOC-O_3 relationship, humidity effects on radical chemistry). A covariance matrix or linear correlation analysis primarily captures linear relationships and can miss these critical non-linearities and interaction effects. RF is a non-parametric method adept at modeling such complex, non-linear dependencies without prior assumptions about the functional form.

2. Robust Feature Importance Ranking: While correlation indicates the strength of a linear relationship, it can be confounded by multicollinearity among predictors (e.g., temperature, radiation, and certain VOCs are often correlated). RF's permutation-based importance measure and SHAP values are more robust in such settings, as they evaluate the contribution of a feature by considering its interaction with all others within the model's prediction framework.

3. SHAP for Interpretable Local and Global Insights: The SHAP framework, built upon the RF model, provides significant advantages over a static correlation plot:

1) Global Explanation: It quantifies the average marginal contribution of each feature to the model output across the entire dataset, which is a more direct measure of "importance" for prediction than correlation.

2) Local Explanation: It reveals the direction and magnitude of a feature's effect conditional on its specific value. For example, it shows that high RH suppresses O_3, but the suppression effect has a specific threshold (~80-90%), and its magnitude varies with altitude. A simple correlation coefficient cannot elucidate this conditional, value-dependent behavior.

3) Interaction Visualization: Dependence plots effectively visualize the non-linear response of O_3 to a single feature while accounting for the average effect of all other features.

In summary, RF-SHAP moves beyond identifying co-variation to quantifying predictive importance and mapping non-linear, conditional responses. This allows us to make stronger, more mechanistic statements about "dominant controls". We added a concise justification along these lines in the revised Section 2.3. (line 181-190 in the revised manuscript)

Section 2.5: it is stated that the OBM box model is constrained to the observations. Was this the case only when calculating the ozone production pathways (Fig. 6), or also when calculating the EKMA curves? I think some caution must be applied when interpreting the results of a constrained box-model for long-lived species such as O3. If O3 is constrained itself that could led to errors especially when considering the loss rates. Moreover, I don't think you can reliably calculate EKMA curves using a model constrained to O3. The authors should clarify these questions and discuss these points. All the necessary caveats about using box models should be explicitly mentioned in the text.

Response: We thank the reviewer for this critical observation. We realize our original description was unclear and potentially misleading. We have revised the methodology section to explicitly clarify our modeling approach for different analyses.

For calculating O_3 production and loss pathways (Figure 6), we did not constrain O_3 itself in the model. Instead, we constrained only the measured precursors (NO, NO_2, CO, VOCs) and meteorological parameters. This allowed the model to calculate O_3 production and loss rates independently. We have clarified this in the revised Methods section:

"For quantifying O_3 production and loss pathways, the model was constrained only by measured precursor concentrations and meteorological parameters; O_3 itself was left unconstrained. This allowed the model to independently simulate gross chemical production and destruction rates. The discrepancy between modeled and observed O_3 then provided insight into the net effect of physical transport processes (R_{trans})." (line 241-245 in the revised manuscript)

For EKMA curve calculations, O_3 was similarly unconstrained and allowed to evolve freely based on the perturbed precursor scenarios. We have added clarification:

"For sensitivity analyses and EKMA isopleth construction, baseline conditions were defined using campaign-averaged diurnal profiles of precursors. Model scenarios were then performed with NO_x and VOC concentrations systematically varied from 0% to 200% of baseline in 10% increments. Each simulation included a 72-hour spin-up to reach pseudo-steady state. Maximum daily O_3 concentrations from each scenario were used to generate EKMA isopleths. This unconstrained approach ensures that the O_3 response genuinely

reflects photochemical sensitivity to precursor changes, free from artificial constraints imposed by observations" (line 260-266 in the revised manuscript)

We have also added a comprehensive discussion of box model limitations:

"Several caveats should be considered when interpreting OBM-MCM results: (1) The 0-D framework cannot resolve spatial heterogeneity or explicitly simulate advection, potentially leading to misattribution between local chemistry and transport; (2) the well-mixed assumption may not hold in complex terrain with strong vertical gradients; (3) unmeasured reactive intermediates rely on model-calculated steady-state concentrations, which may deviate from actual values; (4) heterogeneous chemistry on aerosol surfaces is not accounted for, though it may be significant under high-humidity conditions; and (5) deposition velocities are not explicitly calculated but are instead lumped into the residual term. Despite these limitations, the OBM-MCM remains valuable for diagnosing the relative contributions of chemical versus physical processes and identifying photochemical regimes, provided results are interpreted within these constraints." (line 545-553 in the revised manuscript)

The authors also make the assumption that the difference between calculated and measured ozone is due to transport (line 194 and following); they also mention that dilution is included in the box model, while deposition is not mentioned. I think it is a stretch to assume that Rtrans is all advected ozone. First of all, it is unclear how would dilution work within a constrained box model, where the concentrations of key species are fixed. Second, vertical transport and deposition cannot be discounted, especially in a mountain environment where uphill and downhill air movements occur on a daily basis. Finally, the MCM is extensive but far from complete: a perhaps significant fraction of photochemically generated ozone is not accounted for by the current chemistry in the MCM, which would lead to overestimating the role of transport. The authors should explain better and in more detail how they are using the model. clarify their procedures and discuss how the potential errors introduced by their assumptions affect their results.

Response: We appreciate the reviewer's important observations regarding our modeling approach. We acknowledge that our initial presentation oversimplified the interpretation of Rtrans and the methodology and discussion sections to address these concerns have been substantially revised.

First, regarding the dilution process in our constrained box model: The dilution term in our OBM-MCM framework operates on unconstrained intermediate species (particularly radicals like OH, HO_2, and RO_2) while maintaining fixed concentrations for measured species. Specifically, we applied a diurnally varying dilution rate (kdil) based on boundary layer height evolution, which affects only the calculated concentrations of unmeasured reactive intermediates. This approach follows established protocols (Xue et al., 2014; Chen et

al., 2020). We have clarified this in the revised Methods section:

"The model accounted for boundary layer dilution via a time-dependent mixing rate coefficient (kdil) that varied with planetary boundary layer height, following the approach of Xue et al. (2014). This dilution term, which ranged from 0.1 h^{-1} at night to 0.5 h^{-1} at midday, was applied only to unconstrained radical species (OH, HO_2, RO_2) and intermediate products, while measured species (NO, NO_2, CO, VOCs) were constrained to observational data." (line 215-219 in the revised manuscript)

Second, we fully agree that R_{trans} represents a composite of multiple processes beyond horizontal advection. We have revised our interpretation to explicitly acknowledge this:

"……and R_{trans} represents the net flux due to all non-photochemical processes. These include: (1) horizontal advection of O_3 and its precursors, (2) vertical exchange through mountain–valley circulations and entrainment from the free troposphere, (3) dry deposition to surfaces, and (4) any chemical pathways not included in MCM v3.3.1. In mountainous regions, thermally driven slope flows can make vertical transport particularly significant(Henne et al., 2005; Wagner et al., 2014). Positive R_{trans} values indicate net O_3 import via transport exceeding deposition losses, whereas negative values imply net export or deposition exceeding import." (line 235-241 in the revised manuscript)

Third, regarding MCM completeness and potential underestimation of photochemical O_3 production, we have added a discussion of this limitation:

"Although MCM v3.3.1 is among the most comprehensive chemical mechanisms available, it may not capture all O_3-forming routes-particularly those involving unmeasured VOC species or complex oxidation products. Recent studies suggest that MCM may underestimate O_3 production by 10–20% in some environments due to omitted chemistry of larger alkanes, sesquiterpenes, and oxygenated VOCs(Wang et al., 2018; Jenkin et al., 2019)." (line 249-253 in the revised manuscript)

A few minor points:

*) lines 257-263. It does not look to me as if temperature, humidity and wind have "statistically insignificant" differences. In fact they all seem quite different by looking at figure 4. Please rephrase or clarify.

Response: The Reviewer is correct. The phrase "without statistically significant magnitudes" in the original text (referring to the diurnal amplitudes at mountainside and mountaintop) is ambiguous and potentially misleading. Our intention was to convey that the shape of the diurnal cycles for T, RH, and WS at mountainside and mountaintop are very similar (flat), in contrast to the strong diurnal cycle at the mountain foot. However, as Figure 4 shows, there are clear absolute value differences between mountainside and mountaintop (e.g., T is ~2°C lower at the top). We rephrased this section for clarity: "Meteorological parameters at both mountainside and mountaintop exhibited similarly attenuated diurnal variations, characterized by minimal daytime increases in temperature and slight decreases in wind speed and relative humidity (Fig. 4A-C). The

diurnal temperature ranges (difference between daytime and nighttime) were 6.8% and 10.8% for the mountainside and mountaintop, respectively (Figure 4A). Corresponding diurnal changes in wind speed were -2.0% and -0.4% (Figure 4B), and for relative humidity, -1.3% and -3.2% (Figure 4C). Contrastively, significant diurnal variations were observed in temperature, wind speed and relative humidity at mountain foot, with amplitudes of 16%, 47% and -11%, respectively (Figure 4A-C). " (line 348-354 in the revised manuscript)

*) it would be interesting to connect the O3 diurnal cycles at different altitudes with the different OH reactivities. How do the cycles of VOCs and NOx shape the diurnal O3 cycle?

Response: We thank the reviewer for this insightful suggestion. We have added a brief discussion to explicitly link the observed diurnal O_3 patterns to altitude-dependent OH reactivity and the diurnal cycles of VOCs and NO_x. The additions include: The observed nonuniform diurnal variations in O_3 and its precursors across elevational gradients reflect emergent phenomena driven by elevation-dependent shifts in precursor sources, spatially heterogeneous photochemical modulation, and complex mountain boundary-layer dynamics. This can be mechanistically interpreted through the interplay between altitude-dependent OH reactivity and precursor diurnal dynamics. At the mountain foot, elevated OH reactivity (4.4 s^{-1}), driven by higher concentrations of NO_x and VOCs, enhances daytime O_3 destruction via NO titration and peroxyl radical termination, resulting in a unimodal O_3 peak in the early afternoon followed by rapid depletion. In contrast, the lower OH reactivity at mountainside (1.5 s^{-1}) and mountaintop (2.0 s^{-1}) reflects a shift towards increasing VOC-sensitive chemistry, where isoprene and other reactive VOCs dominate OH reactivity despite their low mixing ratios. This facilitates net O_3 production during daytime at higher elevations, coupled with nocturnal accumulation due to suppressed titration and stable boundary layers. The diurnal patterns of VOCs and NO_x further modulate O_3 cycles. At the mountain foot, bimodal peaks in VOCs and NO_x-synchronized with tourist activity-intensify O_3 titration and peroxy radical conversion to nitrates, thereby suppressing daytime O_3. At mountainside and mountaintop, weaker diurnal variations in precursors allow photochemical production to dominate, with O_3 levels sustained by transport from lower elevations and in-situ chemistry under low-NO_x conditions. These findings underscore how altitude-specific OH reactivity and precursor dynamics jointly govern the diurnal evolution of O_3 in mountainous background regions. (line 373-389 in the revised manuscript)

*) isoprene dominates reactivity at the mountaintop but the model analysis indicates anthropogenic VOCs, namely aromatics, as the main contributors to RIR (line 377 and figure 8). The authors should expand the discussion on this point.

Response: We thank the reviewer for this insightful observation. We have now expanded the discussion in

Section 3.3 to explicitly address the apparent discrepancy between isoprene-dominated OH reactivity and anthropogenic-VOC-dominated RIR at high elevations. The added text clarifies that: **1.** OH reactivity reflects the total potential for radical consumption and atmospheric oxidation capacity, where isoprene-due to its high per-carbon reactivity and biogenic abundance-dominates even at low mixing ratios. **2.** RIR measures the sensitivity of net O_3 production to precursor changes under local chemical conditions. At the mountaintop, low NO_x levels shift the regime toward increasing VOC-sensitive chemistry, where anthropogenic hydrocarbons (especially aromatics) become more effective per molecule in promoting O_3 formation via longer radical chains and higher ozone yield per carbon. This distinction highlights how different metrics capture complementary aspects of VOC chemistry: OH reactivity indicates overall oxidative potential, while RIR identifies key precursors for targeted O_3 control. The expanded discussion strengthens the mechanistic interpretation of our findings and was included in the revised manuscript.

Insert the following paragraph:

The contrasting roles of isoprene and anthropogenic hydrocarbons revealed by OH reactivity and RIR metrics underscore the distinction between oxidative capacity and O_3 production sensitivity under low-NO_x conditions. Isoprene dominated OH reactivity at the mountaintop (31%) due to its high per-carbon reaction rate with OH, reflecting its strong influence on local radical cycling and overall atmospheric oxidation potential. However, its RIR contribution remained modest because isoprene-driven O_3 formation is highly nonlinear and often saturated under low-NO_x regimes, where the availability of NO_x becomes the limiting factor for converting peroxy radicals to NO_2. In contrast, anthropogenic hydrocarbons-particularly aromatics-exhibited higher RIR values because they generate more multi-step oxidation pathways and longer-lived peroxy radicals, thereby enhancing the O_3 yield per carbon under low-NO_x conditions. This decoupling between OH reactivity and RIR emphasizes that while biogenic VOCs control the radical budget, anthropogenic VOCs can exert a disproportionate leverage on net O_3 production in high-altitude, low-NO_x environments. Thus, emission control strategies targeting anthropogenic hydrocarbons may yield greater O_3 reduction benefits per unit mass, even in regions where biogenic species dominate local reactivity. (line 497-510 in the revised manuscript)

*) the measurements cover the period March-August, yet there is not mention of possible seasonal effects. Are there any differences beween spring and summer data? If so, they should be discussed. Otherwise, it should be said that there are no differences.

Response: We thank the referee for raising this important point regarding potential seasonal effects. In response, we have conducted a comparative analysis of the data partitioned into spring (March–May) and summer (June–August).

The analysis reveals clear seasonal differences in O_3 and precursor concentrations. For instance, as shown in the figure S7, at the mountaintop, mean O_3 was ~50.4 ppb in spring compared to ~33.1 ppb in summer.

Similarly, VOC concentrations were notably higher in spring (e.g., mountain foot: ~16.9 ppb in spring vs. ~9.6 ppb in summer). Differences in NO_x and CO were less pronounced and not consistently directional across sites.
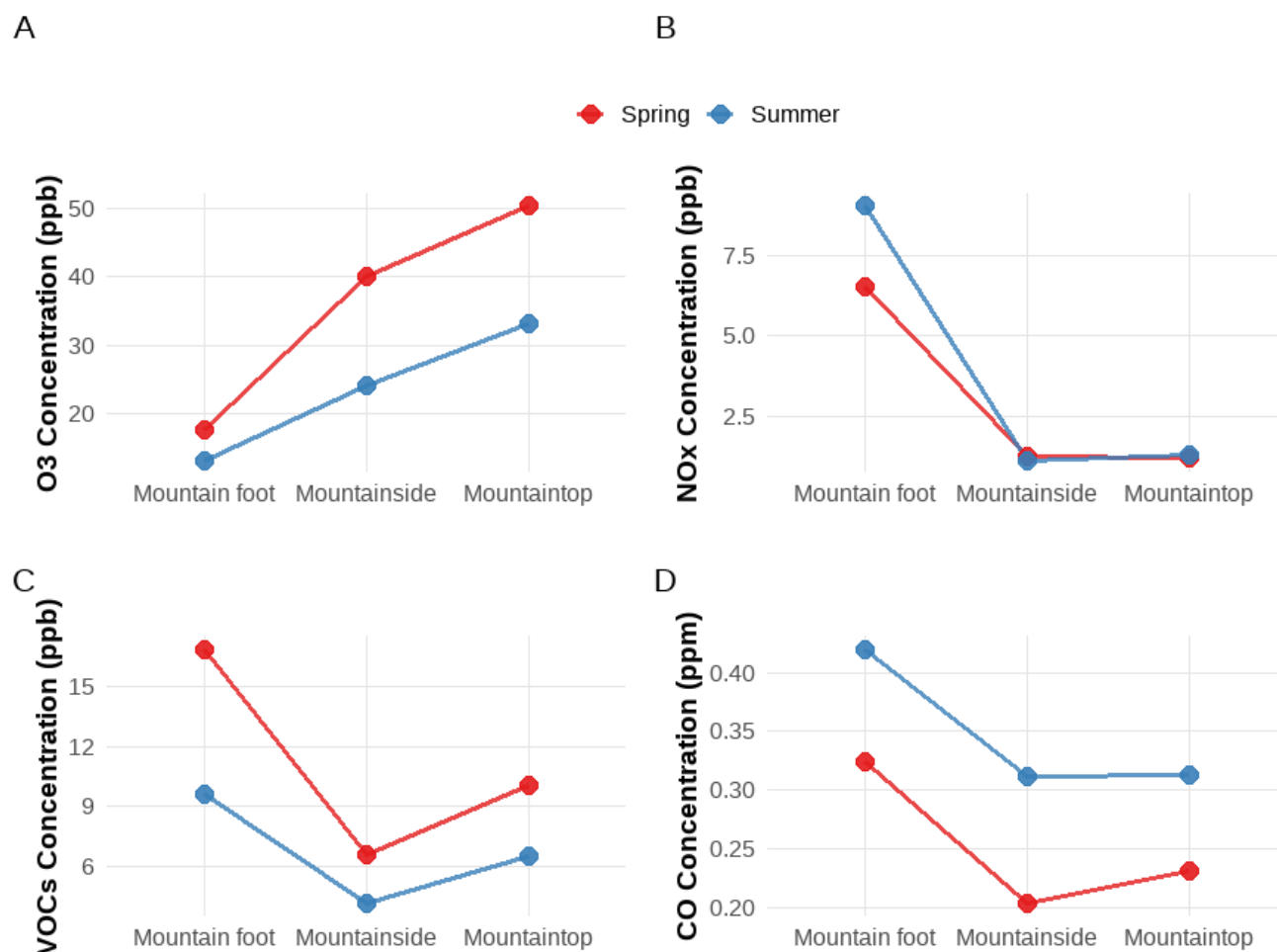

Figure S7. Seasonal and altitudinal variations in air pollutant concentrations across a mountain gradient.

Crucially, however, these seasonal variations do not alter the central altitudinal gradients and mechanistic insights that are the focus of this study. The strong positive O_3 gradient with elevation (from ~15–17 ppb at the foot to ~33–50 ppb at the summit), the shift in dominant controls from NO_x/RH at lower elevations to temperature/VOCs at higher elevations, the transition from net O_3 destruction at the foot to net production aloft, and the NO_x-limited regime with increasing VOC sensitivity at height—all these key patterns remain robust and consistent in both spring and summer datasets.

Given that the primary objective of this manuscript is to elucidate altitude-dependent mechanisms rather than to detail seasonal cycles, and because the altitudinal signals are strong and coherent across seasons, we have chosen to present the campaign-mean results in the main text. This provides a clearer and more consolidated view of the altitude effects central to our work. A detailed seasonal comparison, including relevant figures (line 102-105 in the revised Supplementary Material) and discussion (line 334-346 in the revised manuscript),

was provided in the revised manuscript and supplementary material.

We appreciate the reviewer's comment, which has allowed us to examine the data more deeply and to confirm that our main conclusions hold across both seasons.

*) this is just a suggestion, but it would be interesting to see figure 3 with the stations sorted by altitude, in addition to sorted by concentrations.

Response: We thank the Reviewer for this constructive suggestion. Sorting by altitude would provide an additional valuable perspective on the relationship between elevation and background O_3. In the revised manuscript, we added a new supplementary figure (Figure S4) presenting the same global site comparison but with stations sorted by altitude.

The revised figure does not reveal a simple monotonic trend of O_3 with altitude. This is scientifically reasonable because surface O_3 concentrations at mountain sites are influenced by a combination of factors beyond elevation alone, including:

· Regional pollution background and long-range transport,

· Local photochemical regimes (which depend on precursor emissions, solar radiation, and temperature),

· Meteorological conditions (e.g., boundary-layer height, humidity, and synoptic patterns), and

· Site-specific characteristics (e.g., proximity to emission sources, vegetation type, and exposure to free-tropospheric air).

In fact, our study also notes (Section 3.1) that the summit O_3 concentration at Mt. Fanjing (40.2 ppb), while higher than at the foot (14.8 ppb), is still lower than at some mid-elevation sites in heavily polluted regions (e.g., Mt. Tai in the North China Plain). This further illustrates that altitude alone cannot explain the global distribution of mountain-site O_3; regional pollution levels and local photochemistry play equally important roles. Thus, the absence of a clear altitude-based trend in Figure S4 underscores the complexity of O_3 spatial variability and highlights the need for region-specific analyses-such as the one presented in this work-to understand O_3 formation and transport in mountainous background areas. The altitude-sorted version of Figure S4 has been included in the Supplementary Material (line 78-82 in the revised Supplementary Material), and a brief explanatory note has been added to the main text (line 290-309 in the revised manuscript). We believe this addition enriches the discussion and thank the reviewer for raising this interesting point.
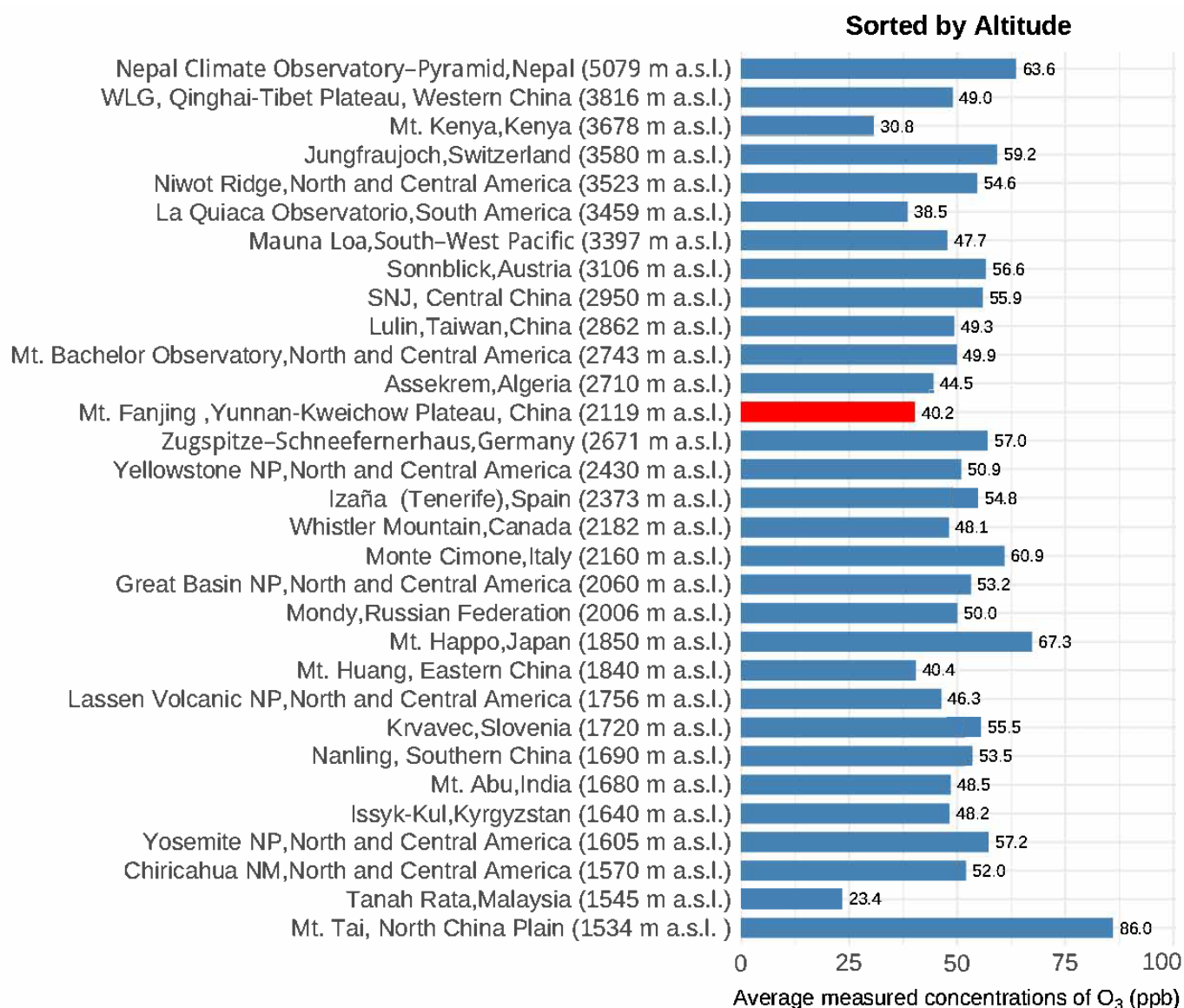
Sorted by Altitude

Site	O₃ (ppb)
Nepal Climate Observatory–Pyramid, Nepal (5079 m a.s.l.)	63.6
WLG, Qinghai-Tibet Plateau, Western China (3816 m a.s.l.)	49.0
Mt. Kenya, Kenya (3678 m a.s.l.)	30.8
Jungfraujoch, Switzerland (3580 m a.s.l.)	59.2
Niwot Ridge, North and Central America (3523 m a.s.l.)	54.6
La Quiaca Observatorio, South America (3459 m a.s.l.)	38.5
Mauna Loa, South–West Pacific (3397 m a.s.l.)	47.7
Sonnblick, Austria (3106 m a.s.l.)	56.6
SNJ, Central China (2950 m a.s.l.)	55.9
Lulin, Taiwan, China (2862 m a.s.l.)	49.3
Mt. Bachelor Observatory, North and Central America (2743 m a.s.l.)	49.9
Assekrem, Algeria (2710 m a.s.l.)	44.5
Mt. Fanjing, Yunnan-Kweichow Plateau, China (2119 m a.s.l.)	40.2
Zugspitze–Schneefernerhaus, Germany (2671 m a.s.l.)	57.0
Yellowstone NP, North and Central America (2430 m a.s.l.)	50.9
Izaña (Tenerife), Spain (2373 m a.s.l.)	54.8
Whistler Mountain, Canada (2182 m a.s.l.)	48.1
Monte Cimone, Italy (2160 m a.s.l.)	60.9
Great Basin NP, North and Central America (2060 m a.s.l.)	53.2
Mondy, Russian Federation (2006 m a.s.l.)	50.0
Mt. Happo, Japan (1850 m a.s.l.)	67.3
Mt. Huang, Eastern China (1840 m a.s.l.)	40.4
Lassen Volcanic NP, North and Central America (1756 m a.s.l.)	46.3
Krvavec, Slovenia (1720 m a.s.l.)	55.5
Nanling, Southern China (1690 m a.s.l.)	53.5
Mt. Abu, India (1680 m a.s.l.)	48.5
Issyk-Kul, Kyrgyzstan (1640 m a.s.l.)	48.2
Yosemite NP, North and Central America (1605 m a.s.l.)	57.2
Chiricahua NM, North and Central America (1570 m a.s.l.)	52.0
Tanah Rata, Malaysia (1545 m a.s.l.)	23.4
Mt. Tai, North China Plain (1534 m a.s.l.)	86.0

Average measured concentrations of O₃ (ppb)

Figure S4. Comparison of O_3 concentrations at Mt. Fanjing summit (2024, March–August) with those observed at mountain sites across the globe. The data is organized in descending order of Altitude, with each site listed alongside its elevation in meters above sea level (m a.s.l.). All average values are reported in ppb. Data were compiled from published literature (Okamoto and Tanimoto, 2016; Li et al., 2007; Lyu et al., 2021; Xu et al., 2016; Gong et al., 2018; Sun et al., 2016).

Once again, we express our gratitude to the Reviewer for their time and valuable insights, which have significantly strengthened our manuscript. We believe our revisions will fully address all points raised. We look forward to the manuscript being considered for publication in EGUsphere.

Reference:

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