Response to Comments from Reviewer

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Title: " Measurement report: Measurement report: Impacts of Thermodynamic and Dynamic Processes on the Vertical Distribution of Carbonaceous Aerosols: lessons from in-situ observations at eastern foothills of LiuPan Mountains, Loess Plateau"

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In the response, the content in **black** font represents the original text from the manuscript, the content in **blue** font indicates additions or revisions, and the content in **red** font is the response to the reviewers' questions and suggestions.

Dear Editors and Reviewers:

Thank you for your and the reviewers' comments concerning our manuscript entitled "Impacts of Thermodynamic and Dynamic Processes on the Vertical Distribution of Carbonaceous Aerosols: lessons from in-situ observations at eastern foothills of LiuPan Mountains, Loess Plateau" (https://doi.org/10.5194/egusphere-2025-3254). Those comments are all very valuable, greatly assisting us in revising and improving our paper, as well as providing important guidance for our research. We have carefully studied the comments and made corrections accordingly, which can be viewed in the revised version of our manuscript. The main corrections in the paper and the responses to the reviewer's comments are provided below:

Major Concerns:

1. The definitions of the terms "UVPM", "BC", and "IRBC" in this study are not clear and thus might influence the interpretation of data and cause confusion for the readers. The terms are also used inconsistently throughout the manuscript, raising concerns of logical conflict in the study. In Line 20, UVPM seems to be the acronym for "ultraviolet particulate matter" while the other term "ultraviolet absorbing particulate matter" also

appears to be the full form of UVPM in Line 605. More details are described as separate bullet points listed below.

Response: We sincerely appreciate this constructive suggestion, which has substantially improved the clarity and consistency of our manuscript. In response, we have carefully revised the relevant sections. Specifically, the concentration measured at 880 nm is now uniformly defined as equivalent black carbon (eBC), and all previous instances of "BC" or "IRBC" have been corrected accordingly. In addition, to avoid any potential ambiguity, and following the official documentation of the MA350 instrument (URL: https://aethlabs.com/products/ma300, last accessed: 13 November 2025), we have standardized the terminology for the species measured at 375 nm by defining them as ultraviolet-absorbing particulate matter (UVPM).

The average near-surface concentrations of equivalent black carbon (eBC) and ultraviolet-absorbing particulate matter (UVPM) in Pingliang were 0.84 μg m^{-3} and 1.24 μg m^{-3}, respectively.

2. Lines 159 – 161: Is black carbon (BC) concentration measured at wavelength 880 nm denoted as BC or IRBC? From the context of the whole manuscript, it seems that BC represents the particle concentration measured at wavelengths 470 nm, 528 nm, 625 nm, and 880 nm, while IRBC represents only the particle concentration measured at wavelength 880 nm, and the particle concentration measured at wavelength 375 is denoted as UVPM. Please ensure consistency throughout the manuscript and avoid confusion for the readers.

Response: Thank you for pointing out the deficiencies in this section. We have implemented detailed revisions regarding this issue in the manuscript: the concentration observed at 880 nm (IR) is now defined as equivalent BC (denoted as "eBC"), while measurements at 375 nm (UV) represent ultraviolet-absorbing particulate matter (UVPM). We have corrected all instances of "BC", "IRBC" and "UVPM" in the text.

3. Lines 161 – 163: Where does this definition of UVPM come from? Please provide relevant citations or explicitly mention that it is specifically defined by this study as the

term "ultraviolet particulate matter" and "ultraviolet absorbing particulate matter" are not yet a standardized term in the literature. Please also provide the reason for using this term to convince the readers if using this term is necessary to be created as opposed to using the more commonly used brown carbon or dust aerosol when it comes to absorption. Based on the context of the whole manuscript, the term UVPM seems to be the total absorption contributed from dust, brown carbon, and black carbon at the wavelength 375 nm, but it would be great if this could be clarified by the authors.

Response: Thank you for your suggestion. The observation period corresponds to the local summer season. Except for one special dust event, the air quality in this region was generally good, with no evident dust pollution. According to the instrument's official specifications and existing studies (Zhao et al., 2023a), the material detected at the 375 nm wavelength includes both primary organic carbon and black carbon directly emitted from sources such as biomass burning and coal combustion, as well as secondary organic carbon formed through various atmospheric chemical reactions. Therefore, in this study, we refer to this component as ultraviolet-absorbing particulate matter (UVPM). This information is derived from Section 1.2, paragraph 2 of the official manual for the MicroAeth® MA350 aethalometer, available at: https://aethlabs.com/products/ma300. The specific revisions are as follows:

The instrument employs five laser wavelengths: 375 nm (UV), 470 nm (Blue), 528 nm (Green), 625 nm (Red), and 880 nm (IR). Carbon concentrations measured at 880 nm are considered to represent the equivalent black carbon concentration (denoted as eBC) (Zhao et al., 2023a), whereas those measured at 375 nm correspond to ultraviolet-absorbing particulate matter (UVPM), which includes primary brown carbon and black carbon emitted from sources such as biomass burning and coal combustion, as well as secondary brown carbon formed through atmospheric oxidation processes (The relevant introduction/details regarding the substance measured at a wavelength of 375 nm can be referenced in the official manual of the MicroAeth® MA Series MA300 instrument. URL: https://aethlabs.com/products/ma300, last accessed: 13 November 2025).

Reference

Zhao, S. P., He, J. J., Dong, L. X., Qi, S. F., Yin, D. Y., Chen, J. B., Yu, Y. Contrasting Vertical Circulation between Severe and Light Air Pollution inside a Deep Basin Results from the Collaborative Experiment of 3D Boundary-Layer Meteorology and Pollution at the Sichuan Basin (BLMP-SCB). Bull. Am. Meteorol. Soc., 104(2), E411-E434, http://doi.org/10.1175/BAMS-D-22-0150.1, 2023a.

4. Lines 176 – 178: If only the absorption at wavelength 880 nm is considered as black carbon according to the statement in the previous section (Lines 159 – 161), why are the absorptions at multiple wavelengths (470 nm, 528 nm, and 625 nm) used for the calculation of light absorption coefficient of black carbon? Please revise the corresponding statements to avoid conflicts of logic throughout the manuscript.

Response: Thank you for pointing out this error. This was a typographical error on our part. The absorption coefficient calculated here should be the light absorption coefficient of carbonaceous aerosol at different wavelengths. We have corrected the content in the manuscript accordingly:

$$b_{Abs} = MAC(\lambda) \times [C] \quad (1),$$

where MAC(λ) denotes the mass absorption cross-section of carbonaceous aerosols at specific wavelengths. For the MicroAeth® series instruments, the MAC values at 375 nm, 470 nm, 528 nm, 625 nm, and 880 nm are 24.07 $m^2\ g^{-1}$, 19.07 $m^2\ g^{-1}$, 17.03 $m^2\ g^{-1}$, 14.09 $m^2\ g^{-1}$, and 10.12 $m^2\ g^{-1}$, respectively (Zhao et al., 2023a). The [C] represents the concentration of carbonaceous aerosol at different wavelengths.

Additionally, the light absorption coefficient of BC can be expressed as:

$$b_{Abs}(\lambda) = k \times \lambda^{-AAE} \quad (2).$$

In Eq. (2), k is a wavelength-independent constant, and AAE represents the Absorption Ångström Exponent, which characterizes the wavelength dependence of light absorption of carbonaceous aerosol (Ångström, 1929). A higher AAE value signifies that the aerosol absorption capacity decreases more rapidly with increasing wavelength.

By combining the b_{Abs} values derived from Eq. (1) with Eq. (2), vertical profiles of AAE for carbonaceous aerosols were obtained.

Reference:

Ångström, A., 1929. On the atmospheric transmission of sun radiation and on dust in the air. Geogr. Ann. 11, 156–166.

Minor Concerns:

1. Lines 156 – 158: Does this instrument "MicroAeth® MA350" attribute all light absorption to black carbon? How do the authors quantitatively distinguish black carbon from other light-absorbing aerosols? What is the corresponding uncertainty for this method to quantify black carbon?

Response: Thank you for your question, which allows us to clarify our methodology. (1) In this study, the MicroAeth® MA350 aethalometer was utilized, which provides measurements of carbonaceous aerosols at five different wavelengths using an optical method. Specifically, the aerosol concentration detected at the wavelength of 880 nm is defined as equivalent black carbon (eBC), while the concentration at 375 nm is designated as ultraviolet-absorbing particulate matter (UVPM). The technical specifications of the MicroAeth® MA350 monitor are elaborated in Section 1.2 (second paragraph) of the official user manual, which is available at the following link: https://aethlabs.com/products/ma300. To summarize, the light absorption discussed in this study refers to that of carbonaceous aerosols, which consist of both eBC and brown carbon (BrC). (2) A two-component model was applied in this work to quantitatively distinguish the light absorption contributions from eBC and UVPM, respectively. Detailed methodological descriptions of the two-component model have been supplemented in Text 4 of the supplementary materials.

Text 4: Two-component model

Referring to the studies by Chen et al. (2015), we used a two-component model to separate the light absorption coefficients of eBC and UVPM, calculated using the following formulas:

$$b_{Abs,\lambda} = b_{Abs,\lambda,eBC} + b_{Abs,\lambda,UVPM} = A_1 \times \lambda^{-AAE_{eBC}} + A_2 \times \lambda^{-AAE_{UVPM}} \quad (3).$$

Where AAE_{eBC} and AAE_{UVPM} are the Absorption Ångström Exponents (AAE) for eBC and UVPM, respectively. In the calculation process, eBC is typically assumed to be composed of pure carbon, thus AAE_{eBC} is approximated as 1.0 (Chow et al., 2018). The values of A_1, A_2, and AAE_{UVPM} in the equation are then determined through a least-squares fitting method.

References

Chen, L.W.A., Chow, J.C., Wang, X.L., Robles, J.A., Sumlin, B.J., Lowenthal, D.H., Zimmermann, R., Watson, J.G. Multi-wavelength optical measurement to enhance thermal/optical analysis for carbonaceous aerosol. Atmos. Meas. Tech., 8(1), 451-461, https://doi.org/10.5194/amt-8-451-2015, 2015.

Chow, J.C., Watson, J.G., Green, M.C., Wang, X.L., Chen, L.-W.A., Trimble, D.L., Cropper, P.M., Kohl, S.D., Gronstal, S.B. Separation of brown carbon from black carbon for IMPROVE and CSN PM2.5 samples, Air Waste Manag. Assoc., 68(5), 494-510, https://doi.org/10.1080/10962247.2018.1426653, 2018.

(3) Since the uncertainty here arises from the measurement process of the instrument, it is classified as a Type A uncertainty. The Type A uncertainty is calculated as follows: $unc = \frac{\sigma_x}{\sqrt{n}} = \sqrt{\frac{\sum_{i=1}^{n}(x_i-\bar{x})^2}{n\,(n-1)}}$, where σ_x represents the standard deviation of multiple observations, n denotes the number of observations, $\bar{x}$ is the mean of the observations.

We used multiple measurements obtained during a single observation period, before the tethered balloon was launched, as the observational data to estimate the uncertainty of the black carbon instrument in measuring carbonaceous aerosols. The calculated uncertainty is 0.01793 μg m^{-3}.

2. Line 183: Please cite the source of Equation 2.

Response: Thank you for your suggestion. We have added the reference (or: citation)

for this formula in the manuscript. The specific revision is as follows:

Additionally, the light absorption coefficient of carbonaceous aerosols can be expressed as:

$$b_{Abs}(\lambda) = k \times \lambda^{-AAE} \quad (2).$$

In Eq. (2), k is a wavelength-independent constant, and AAE represents the Absorption Ångström Exponent, which characterizes the wavelength dependence of light absorption of carbonaceous aerosols (Ångström, 1929).

Reference

Ångström, A., 1929. On the atmospheric transmission of sun radiation and on dust in the air. Geogr. Ann. 11, 156–166.

3. Line 200: Please cite the source of Equation 4.

Response: We appreciate your suggestion. The citation for this formula has now been included in the manuscript. The specific changes are detailed below:

Specific humidity (q, g g^{-1}), defined as the ratio of water vapor mass to the total mass of moist air (water vapor plus dry air), is calculated using Eq. (4) (Gutzler, 1992),

$$q = \frac{\varepsilon \times e}{P - 0.378 \times e} \quad (4)$$

Reference

Gutzler, D. S. Climatic variability of temperature and humidity over the tropical western Pacific. Geophys. Res. Lett., 19(15), 1595-1598, https://doi.org/10.1029/92GL01579, 1992.

4. Line 203: Please cite the source of Equation 5.

Response: We have adopted your suggestion. The reference for this equation has been incorporated into the main body of the manuscript. The modification is shown below:

Specific humidity (q, g g^{-1}), defined as the ratio of water vapor mass to the total mass of moist air (water vapor plus dry air), is calculated using Eq. (4) (Gutzler, 1992),

$$q = \frac{\varepsilon \times e}{P - 0.378 \times e} \quad (4)$$

where $\varepsilon = 0.622$, e represents vapor pressure, and P denotes atmospheric pressure, with

vapor pressure, e being derived from Eq. (5) through relative humidity (RH) and air temperature (T, °C) (Gutzler, 1992).

$$e = 6.105 \times RH \times \exp\left(\frac{17.7 \times T}{237.7+T}\right) \quad (5).$$

Reference

Gutzler, D. S. Climatic variability of temperature and humidity over the tropical western Pacific. Geophys. Res. Lett., 19(15), 1595-1598, https://doi.org/10.1029/92GL01579, 1992.

5. Lines 207 – 208: The definition of the term "planetary boundary layer (PBL)" should be provided when the term first appears in the text.

Response: Thank you for your suggestion. We have improved the introduction of the PBL in the manuscript to address this issue. The revised content is as follows:

Planetary boundary layer is the part of the atmosphere closest to the planet's surface, accounting for approximately 10% – 20% of the troposphere. It is the lowest layer of the troposphere directly influenced by surface forcing, with a response time of less than one hour, playing a critical role in the dispersion and transport of air pollutants. Within the PBL, turbulent mixing processes homogenize air temperature and humidity, resulting in relatively uniform distributions of these properties.

6. Line 213: Please provide citations to support this statement "various methodologies exist for determining PBL height (PBLH)" or revise the text by simply describing the approaches used in this study. It is recommended to provide citations while stating that various methodologies exist.

Response: Thank you for your suggestion. We have revised the text at this point as you recommended. The revised content is as follows:

PBL height (PBLH) refers to the thickness of the layer most significantly affected by the surface. In other words, it is the vertical extent that surface turbulent motion (caused by surface heating, friction, or topography, etc.) can effectively influence (Emeis et al., 2008; Seibert et al., 2000). This study employs two established methodologies for determining the PBLH: the potential temperature gradient method and the parcel

method (Holzworth, 1964; Zhang et al., 2020). The potential temperature gradient method was utilized for calculating the PBLH during the nighttime and early morning hours (20:00, 23:00, 05:00, and 08:00 LT), while the parcel method was applied specifically for the daytime periods (11:00, 14:00, and 17:00 LT). Detailed computational procedures for both approaches are summarized in Supplementary Table S1.

7. Line 227: Please cite the source of Equation 6.

Response: We appreciate your suggestion. The citation for this formula has now been included in the manuscript. The specific changes are detailed below:

To quantify the relative contributions of potential temperature gradient, mechanical turbulence index, horizontal wind speed, and vertical wind speed to UVPM variations at different altitudes, we employed a random forest regression algorithm. The model generated training subsets via bootstrap sampling, with random feature subsets selected for optimal splitting at each decision tree node. Observations were categorized into daytime (08:00, 11:00, 14:00, 17:00 LT) and nighttime (20:00, 23:00, 05:00 LT) periods to compare the dominant mechanisms governing aerosol vertical distribution. The mechanical turbulence index was calculated using Eq. (6) (Zhao et al., 2023a).

$$V_{TKE} = 0.5 \times \sqrt{\overline{u}^2 + \overline{v}^2 + \overline{w}^2} \quad (6).$$

Reference

Zhao, S. P., He, J. J., Dong, L. X., Qi, S. F., Yin, D. Y., Chen, J. B., Yu, Y. Contrasting Vertical Circulation between Severe and Light Air Pollution inside a Deep Basin Results from the Collaborative Experiment of 3D Boundary-Layer Meteorology and Pollution at the Sichuan Basin (BLMP-SCB). Bull. Am. Meteorol. Soc., 104(2), E411-E434, http://doi.org/10.1175/BAMS-D-22-0150.1, 2023a.

8. Lines 245 – 248: It would be great if this qualitative statement can be turned into a table for a quantitative comparison, which would make the argument more convincing and make this work more valuable and academically citable.

Response: Thank you for your suggestion. We have added Table 1 in the corresponding section of the manuscript, which makes the comparison in the main text clearer.

Table 1 Statistics of near-surface eBC concentrations in main city.

City (Research region)	eBC concentrations (μg m^{-3})	References
Pingliang	0.8 (average)	This study
Beijing	2.0 ~ 6.0	Yang et al., 2022
Shanghai	6.0 ~ 10.5	Wang et al., 2021a
Nanjing	3.2 (average)	Shi et al., 2021
Chengdu	5.0 ~ 8.0	Zhao et al., 2023a
Shenzhen	1.8 ~ 2.5	Wu et al., 2021
Hengshui	5.2 (average)	Ran et al., 2016
Beibu Gulf region	2.3 ~ 4.0	Yang et al., 2023
Lanzhou	3.0 ~ 6.0	Guan et al., 2022

9. Lines 253 – 254: Without showing the quantitative data for direct comparisons of emission inventories of air pollutants from daily human activities, it would be safer to use a more conservative statement like "probably lower" to ensure scientific rigor and avoid controversy.

Response: Thank you for your suggestion. I have revised the content in the manuscript accordingly, and the modified content is as follows:

This discrepancy can be attributed to several factors. Firstly, Pingliang is a relatively small city with a permanent population of fewer than 2 million, whereas the aforementioned cities have much larger urban populations. Consequently, the total amount of air pollutants generated from daily human activities in Pingliang may be comparatively lower than those in the aforementioned cities.

10. Line 288: It seems that only diurnal variations of IRBC rather than BC are shown.

Response: Thank you for your suggestion. We have already addressed similar issues consistently throughout the manuscript. We have defined the concentration measured at the 880 nm wavelength (IR) as equivalent Black Carbon (eBC) and have made corresponding revisions throughout the entire text.

3.1.2 Diurnal variations of eBC and UVPM profiles

Figures S3 and S4 respectively depict the trends and correlations of the ascent and descent profiles for eBC and UVPM. The results indicate that the ascent and descent profiles at 5:00, 20:00, and 23:00 in this study exhibit similar trends, and thus they can both be treated as independent observational profiles for analysis. For observations at 8:00, 11:00, 14:00 and 17:00, only the ascent data were used for analysis. Figure 2 shows the averaged profiles for all sampling periods during the observation campaign, the red solid line and its shaded envelope denote the mean and standard deviation of eBC, while the blue solid line and its shaded envelope denote the mean and standard deviation of UVPM. Because eBC mass concentration in the upper atmosphere is extremely low (below the instrument's normal limit of detection), optical measurements often yield negative results, which are corrected when the ONA method is used for data quality control. Furthermore, owing to the relatively high wind speeds in the upper atmosphere, our measurements in the early afternoon were typically unable to reach the PBLH.

11. Line 290: Are the profiles for BC or IRBC?

Response: Thank you for your suggestion. We have clarified this issue consistently throughout the revised manuscript. The concentration measured at 880 nm has been defined as equivalent black carbon (eBC), and all relevant terms have been updated accordingly across the entire text.

12. Line 295: The term IRBC is used here while referring to BC mentioned earlier (Line 290). It seems that IRBC only represents black carbon (BC) measured at wavelength 880 nm while the absorption measured at wavelengths 470nm, 528nm, 625 nm, and 880 nm are all considered as black carbon in this study. If this is the case, please revise the definition in earlier sections of this manuscript for consistency.

Response: Thank you for your suggestion. We have addressed similar issues consistently throughout the manuscript. The concentrations measured at wavelengths of 375 nm, 470 nm, 528 nm, and 625 nm correspond to carbonaceous aerosols,

including black carbon (BC), brown carbon (BrC). In this study, to distinguish carbonaceous aerosols measured at different wavelengths, the concentration measured at 375 nm is referred to as ultraviolet-absorbing particulate matter (UVPM), while the concentration measured at 880 nm is defined as equivalent black carbon (eBC). Corresponding revisions have been made throughout the manuscript. The specific changes are as follows:

In Figure 2, the red solid line and its shaded envelope denote the mean and standard deviation of eBC, while the blue solid line and its shaded envelope denote the mean and standard deviation of UVPM.

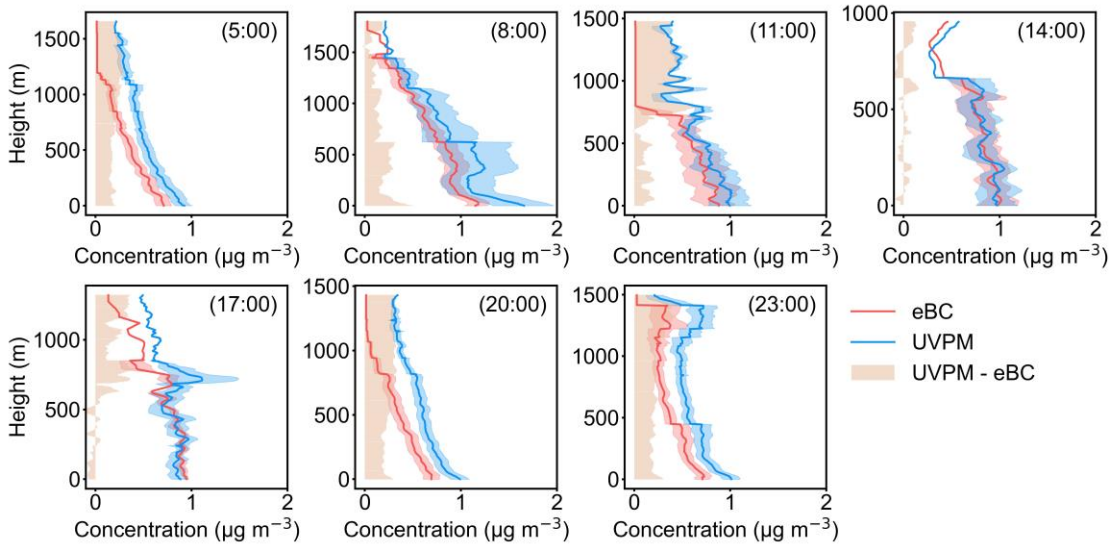

Figure 2. Averaged diurnal variation in profiles of the eBC and UVPM concentration during the field campaign. The red and blue lines represent eBC and UVPM concentrations, respectively, while the red and blue shaded areas denote the standard deviation of eBC and UVPM, respectively. The light orange shaded area represents the difference between UVPM and eBC.

13. Line 300: Are the profiles for BC or IRBC?

Response: Thank you for your suggestion. We have already addressed similar issues consistently throughout the manuscript. We have defined the concentration measured at the 880 nm wavelength as equivalent Black Carbon (eBC) and have made corresponding revisions throughout the entire text. The specific modification at this point is as follows:

Vertical profiles of eBC and UVPM concentrations reveal a consistent decrease with increasing altitude, with the most pronounced gradient observed in the near-surface layer.

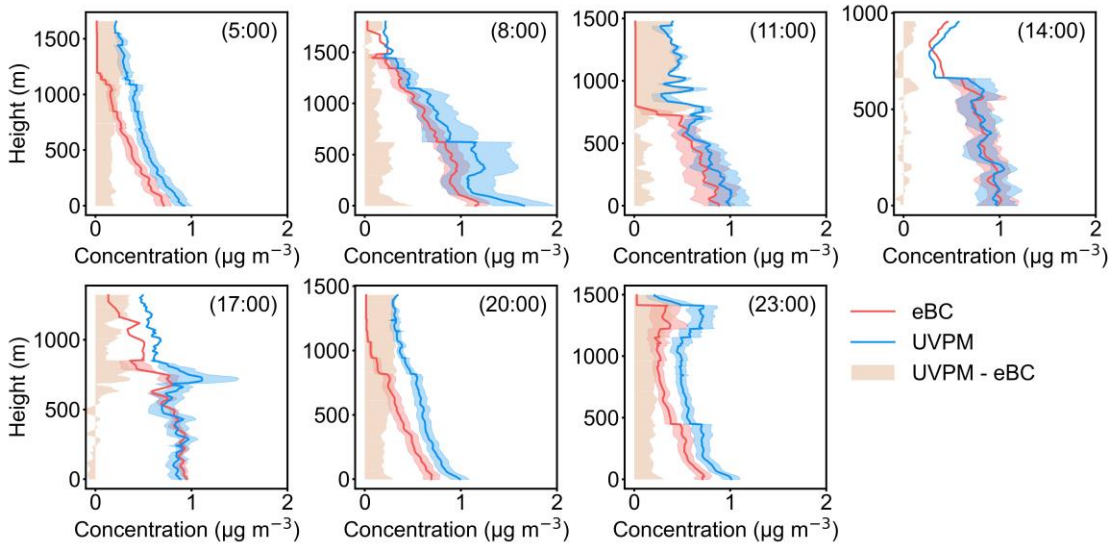

Figure 2. Averaged diurnal variation in profiles of the eBC and UVPM concentration during the field campaign. The red and blue lines represent eBC and UVPM concentrations, respectively, while the red and blue shaded areas denote the standard deviation of eBC and UVPM, respectively. The light orange shaded area represents the difference between UVPM and eBC.

14. Line 304: Is the BC here meant to be IRBC?

Response: Thank you for your suggestion. We have already addressed similar issues consistently throughout the manuscript. We have defined the concentration measured at the 880 nm wavelength as equivalent Black Carbon (eBC) and have made corresponding revisions throughout the entire text. The specific modification at this point is as follows:

Vertical profiles of eBC and UVPM concentrations reveal a consistent decrease with increasing altitude, with the most pronounced gradient observed in the near-surface layer. A comparative analysis of their vertical profiles reveals that during periods of weak convective activity, such as early morning and nighttime (i.e., 05:00, 20:00, and 23:00 LT), UVPM concentrations aloft generally exceed those of eBC (the significance test of the UVPM–eBC differences is provided in Figure S5), while near the surface the

two species show much smaller differences.

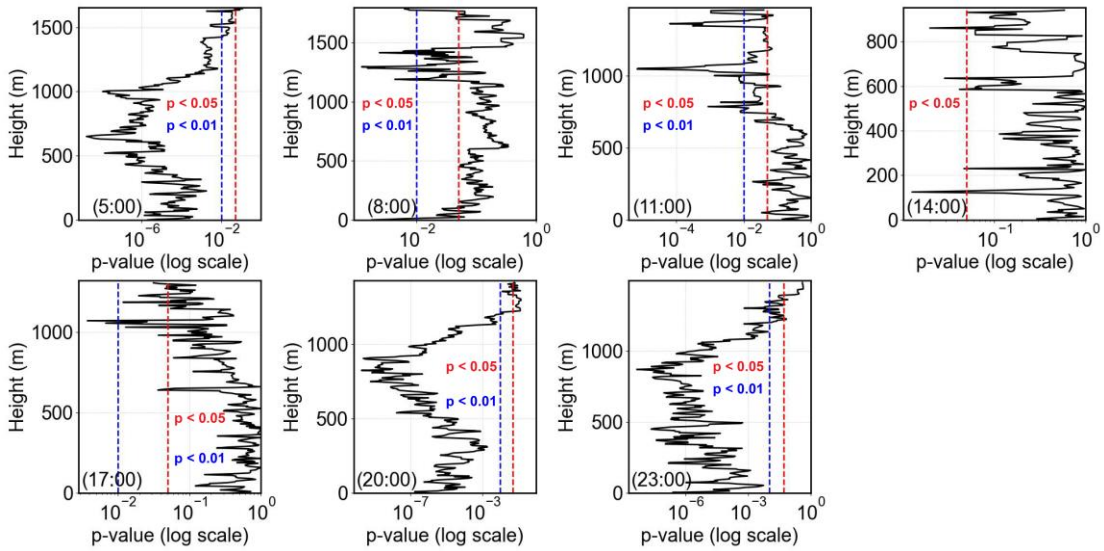

Figure S5. Significance test of the UVPM–eBC differences during different hours of the day. The red and blue dashed lines in the figure indicate p-values of 0.05 and 0.01, respectively.

15. Line 305: Is the UVPM-to-BC here meant to be UVPM-to-IRBC?

Response: Thank you for your suggestion. The term IR BC (Infrared Black Carbon) used here refers to the concentration measured at the 880 nm wavelength, which is defined as equivalent Black Carbon (eBC). We have revised all related terminology throughout the manuscript. The specific modification at this point is as follows:

Vertical profiles of eBC and UVPM concentrations reveal a consistent decrease with increasing altitude, with the most pronounced gradient observed in the near-surface layer. A comparative analysis of their vertical profiles reveals that during periods of weak convective activity, such as early morning and nighttime (i.e., 05:00, 20:00, and 23:00 LT), UVPM concentrations aloft generally exceed those of eBC (the significance test of the UVPM–eBC differences is provided in Figure S5), while near the surface the two species show much smaller differences.

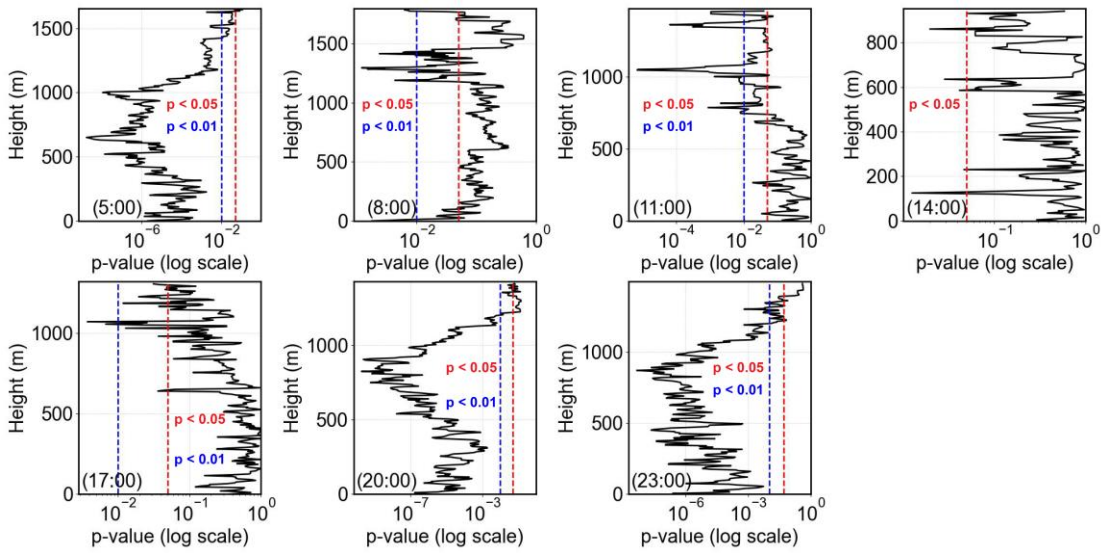

Figure S5. Significance test of the UVPM–eBC differences during different hours of the day. The red and blue dashed lines in the figure indicate p-values of 0.05 and 0.01, respectively.

16. Line 316 – 317: Could this change due to vertical mixing and dominance of black carbon? The disparity between UVPM and IRBC should not disappear at lower altitudes if the pollutants considered as UVPM mixed downward during the convection do not include black carbon. If UVPM is considered as the total absorption of dust, brown carbon, and black carbon, this reduction of disparity might indicate that the contribution of dust and organics (brown carbon) becomes less important, and IRBC is dominant during this period.

Response: Thank you for your question and suggestion. We have revised this section and added supporting evidence. According to the mean diurnal variations in vertical distribution of the $UVPM_{sec}$/UVPM ratio, the larger nighttime difference between UVPM and eBC at higher altitudes is primarily attributable to the increased contribution of $UVPM_{sec}$. The relevant discussion is presented below:

Vertical profiles of eBC and UVPM concentrations reveal a consistent decrease with increasing altitude, with the most pronounced gradient observed in the near-surface layer. A comparative analysis of their vertical profiles reveals that during periods of weak convective activity, such as early morning and nighttime (i.e., 05:00, 20:00, and 23:00 LT), UVPM concentrations aloft generally exceed those of eBC (the significance test of the UVPM–eBC differences is provided in Figure S5), while near the surface the

two species show much smaller differences. To further investigate the underlying causes, we estimated the concentration of secondary UVPM ($UVPM_{sec}$) using the least-squares method described by Wu et al. (2016), based on the ratio of UVPM to eBC. Figure 3 presents the mean diurnal variations in vertical distributions of the ratios of $UVPM_{sec}$ to total UVPM, and of $UVPM_{sec}$ to eBC. From 17:00 to 05:00 LT, the $UVPM_{sec}$/UVPM and $UVPM_{sec}$/eBC ratios aloft increase markedly, indicating that the elevated UVPM during this period is more strongly influenced by secondary sources. As time progresses, the region characterized by high ratios gradually approaches the ground surface and eventually disappears by 08:00 LT.

The enhanced contribution of secondary sources aloft may be attributed to the increasingly stable stratification after 17:00 LT, which suppresses vertical mixing and inhibits the upward transport of both eBC and UVPM. In contrast, gaseous precursors of $UVPM_{sec}$ (i.e., VOCs) can accumulate above the PBL. Under relatively clean atmospheric conditions with limited condensation sinks, nocturnal chemistry dominated by NO_3^- radicals, together with low ambient temperatures, favors the formation of secondary organic aerosols through gas-to-particle partitioning and temperature-dependent condensation (Han & Jang, 2023; Kuang et al., 2025; Kulmala, 2022; Morgan et al., 2009; Wang et al., 2023; Zhao et al., 2023b). After sunrise (around 06:00 LT in summer at this site), enhanced solar radiation promotes convective mixing and weakens the nocturnal inversion at the top of the PBL. Consequently, $UVPM_{sec}$-enriched air masses retained within the residual layer are entrained downward into the growing daytime PBL, thereby reducing the concentration gradient between UVPM and eBC within the PBL (Zhao et al., 2023a). In addition, increased human activities and strengthened thermal convection after sunrise lead to larger primary emissions throughout the atmospheric column, causing carbonaceous aerosols to be increasingly dominated by primary components. Because the PBL is still developing at 08:00 LT, the near-surface concentrations of UVPM and eBC are higher than those observed at 05:00 LT. As thermal convection intensifies, the decline in near-surface UVPM and eBC slows between 11:00 and 14:00 LT. Between 14:00 and 17:00 LT, strong

convective mixing results in an approximately uniform vertical distribution of both eBC and UVPM from the surface up to ~800 m.

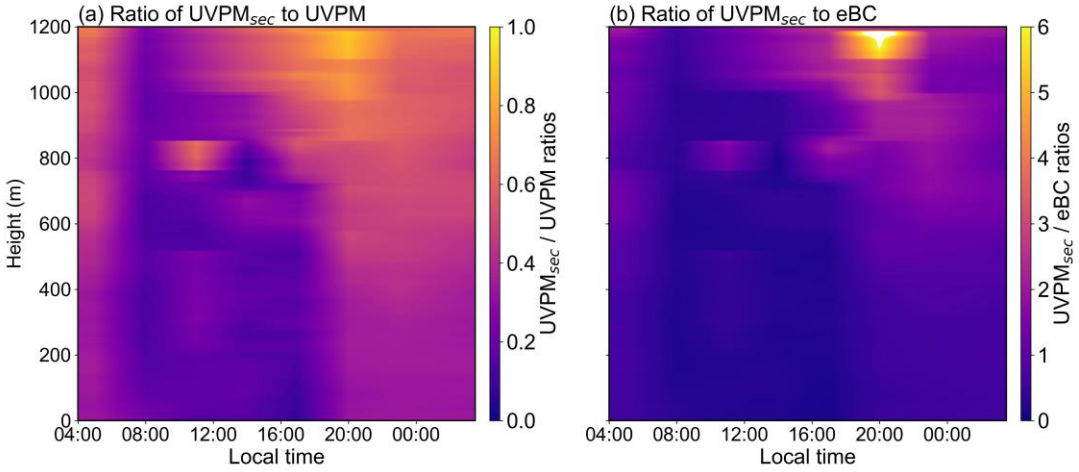

Figure 3. Mean diurnal variations in vertical distribution of a) $UVPM_{sec}$/UVPM and b) $UVPM_{sec}$/eBC ratios during the field campaign.

References

Han, S., Jang, M. Modeling daytime and nighttime secondary organic aerosol formation via multiphase reactions of biogenic hydrocarbons. Atmos. Chem. Phys., 23(2), 1209-1226, http://doi.org/10.5194/acp-23-1209-2023, 2023.

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S. J., Brown, S. S., Zhang, Y. H. Increased night-time oxidation over China despite widespread decrease across the globe. Nat. Geosci., 16: 217-223, https://doi.org/10.1038/s41561-022-01122-x, 2023.

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17. Lines 321 – 324: This observation also indicates that the UVPM defined in this study is probably the overall contribution of black carbon, brown carbon, and dust aerosol. Please clarify the definition.

Response: Thank you for raising this question. The definition of UVPM has been revised in Section 2.1.2 of the updated manuscript. According to Section 1.2 of the official manual for the MicroAeth® MA series MA350 instrument, substances measured at a wavelength of 375 nm are defined as UVPM. Since the observation period corresponds to the local summer season, except for a specific dust event, the regional air quality was generally good without significant dust pollution. Based on the instrument manual (URL: https://aethlabs.com/products/ma300) and previous studies (Zhao et al., 2023a), UVPM in this study comprises black carbon (BC) and brown carbon. The relevant discussion is presented below:

The instrument employs five laser wavelengths: 375 nm (UV), 470 nm (Blue), 528 nm (Green), 625 nm (Red), and 880 nm (IR). Carbon concentrations measured at 880 nm are considered to represent the equivalent black carbon concentration (denoted as eBC) (Zhao et al., 2023a), whereas those measured at 375 nm correspond to ultraviolet-absorbing particulate matter (UVPM), which includes primary brown carbon and black carbon emitted from sources such as biomass burning and coal combustion, as well as

secondary brown carbon formed through atmospheric oxidation processes (The relevant introduction/details regarding the substance measured at a wavelength of 375 nm can be referenced in the official manual of the MicroAeth® MA Series MA300 instrument. URL: https://aethlabs.com/products/ma300, last accessed: 13 November 2025).

Reference

Zhao, S. P., He, J. J., Dong, L. X., Qi, S. F., Yin, D. Y., Chen, J. B., Yu, Y. Contrasting Vertical Circulation between Severe and Light Air Pollution inside a Deep Basin Results from the Collaborative Experiment of 3D Boundary-Layer Meteorology and Pollution at the Sichuan Basin (BLMP-SCB). Bull. Am. Meteorol. Soc., 104(2), E411-E434, http://doi.org/10.1175/BAMS-D-22-0150.1, 2023a.

18. The term "LT" is used in Lines 225 – 226 while the term "LST" is used in other parts of the manuscript. Please explicitly distinguish the difference between the two terms. Otherwise, please choose one and stick to it to ensure consistency and avoid confusion.

Response: Thank you for pointing out this inconsistency. We agree that using different abbreviations for the same concept may cause confusion. The term "LT" used in lines 225–226 refers to "Local time"; however, its usage was inconsistent with other parts of the manuscript. To ensure uniformity throughout the paper, we have revised all instances of "LST" to "LT".

19. Figure 3: It seems that IRBC rather than BC is shown in the figure. Please update the caption and revise relevant statements in the main text.

Response: Thank you for your suggestion. We have already addressed similar issues consistently throughout the manuscript. We have defined the concentration measured at the 880 nm wavelength as equivalent Black Carbon (eBC) and have made corresponding revisions throughout the entire text. The specific modification at this point is as follows (Due to adjustments made to the figures within the manuscript, this illustration has been renumbered as "Figure 4."):

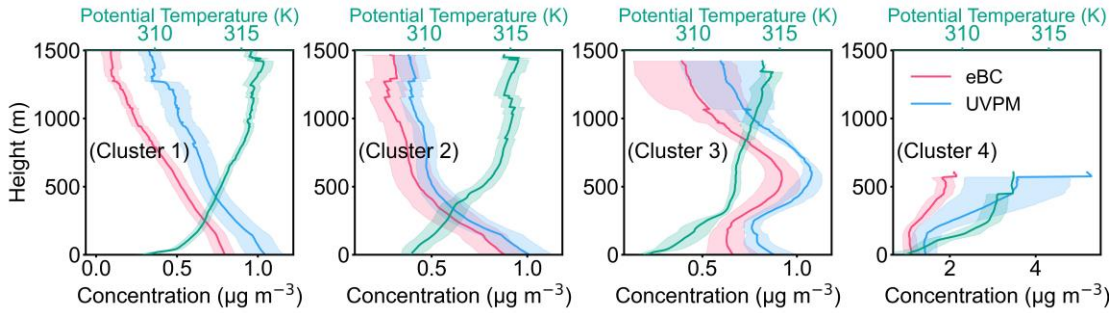

Figure 4. Cluster analysis of UVPM and eBC concentration profiles during the field campaign. The blue, red, and green lines represent the profiles of UVPM, eBC, and potential temperature, respectively, and the shaded areas in the corresponding colors denote their standard deviations.

20. Section 3.1.3: It seems that the BC used in this section should be IRBC. Please confirm.

Response: Thank you for your suggestion. We have already addressed similar issues consistently throughout the manuscript. We have defined the concentration measured at the 880 nm wavelength as equivalent Black Carbon (eBC) and have made corresponding revisions throughout the entire text.

21. Lines 364 – 366: How are the weather conditions defined based on what parameters and what data? Please provide relevant information.

Response: Thank you for your comment. We have added detailed information about the environmental conditions in the Supplementary Material. The specific content is provided below:

We further selected observations from representative pollution events to compare AAE under different environmental conditions (Figure 5) (Detailed explanations regarding the selection of pollution events are provided in Text 3 of the supplementary material).

Text 3: Selection guidelines for pollution incidents

1. Diesel vehicle emissions: During the 20:00 observation on July 17, 2024, a diesel harvester was operating approximately 50 m from our observation site and ceased work after about half an hour. Consequently, we define this specific observation as being affected by diesel vehicle emissions. For comparison, we selected the 23:00

observation—which has the shortest temporal difference—as the corresponding non-diesel vehicle emission observation.

2. Dust Pollution: A dust event occurred throughout the entire day of July 21, 2024. We defined the 8:00 observation, when the dust was most severe (PM_{10} concentration in Pingliang urban area was 350 μg m^{-3} at that time), as the dust pollution observation. Since the dust event lasted for a long duration, we chose the observation taken at the same time on the previous day (8:00 on July 20, 2024) for comparison.

The relevant information mentioned above is now reflected using shading and text annotations in the new Figure S1.

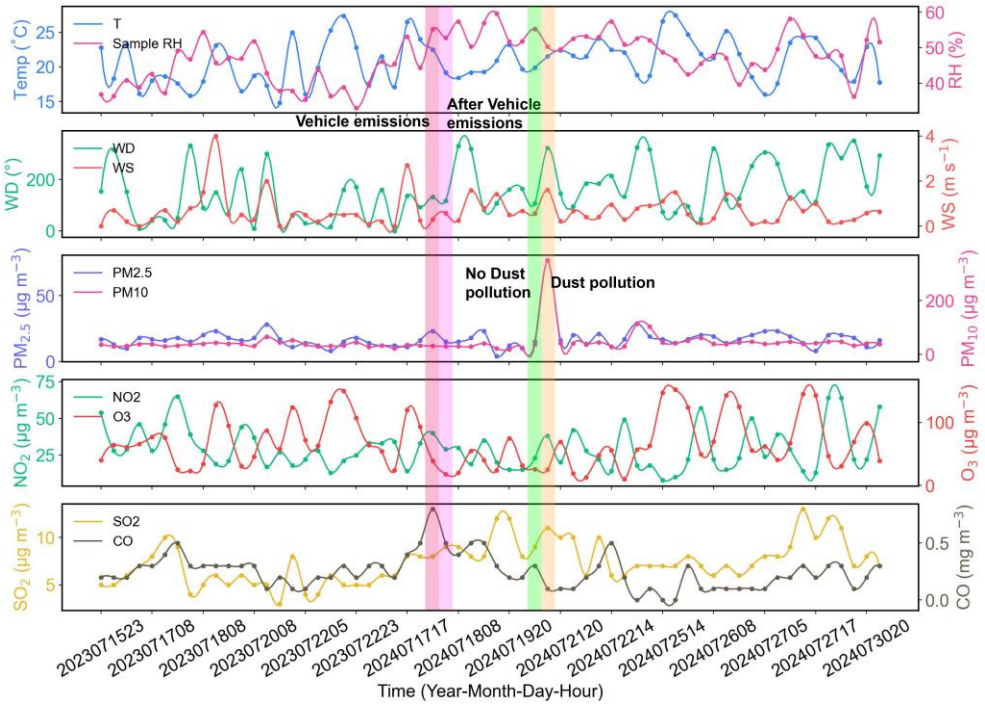

Figure S1. Temporal variations of meteorological parameters, including air temperature (T), relative humidity (RH), wind direction (WD), wind speed (WS), and air pollutants, including particulate matter with aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) and than 10 μm (PM_{10}), nitrogen dioxide (NO_2), ozone (O_3), sulfur dioxide (SO_2), and carbon monoxide (CO), were observed during the monitoring period. The shaded areas in the figure represent the during and after vehicle emissions, no dust pollution, and dust pollution at 20:00 on July 17, 23:00 on July 17, 2024, 08:00 on July 20, 2024, 08:00 on July 21, 2024, respectively.

22. Lines 375 – 377: Please provide citations of previous studies.

Response: Thank you for pointing out this issue. We have added citations to previous studies in the manuscript. The revised content is as follows:

Previous studies commonly employ a two-component model to differentiate the Absorption Ångström Exponent of eBC (AAE_{eBC}) and BrC (AAE_{UVPM}), fixing AAE_{eBC} at 1 and thereby deriving AAE_{UVPM} (Figure S9, the calculation methodology is provided in Text 4 of the supplementary material) (Chen et al., 2015; Chow et al., 2018). Elevated values of AAE_{UVPM} are typically indicative of biomass burning and secondary aging processes (Olson et al., 2015; Gombi et al., 2025).

References

Chen, L.W.A., Chow, J.C., Wang, X.L., Robles, J.A., Sumlin, B.J., Lowenthal, D.H., Zimmermann, R., Watson, J.G. Multi-wavelength optical measurement to enhance thermal/optical analysis for carbonaceous aerosol. Atmos. Meas. Tech., 8(1), 451-461, https://doi.org/10.5194/amt-8-451-2015, 2015.

Chow, J.C., Watson, J.G., Green, M.C., Wang, X.L., Chen, L.-W.A., Trimble, D.L., Cropper, P.M., Kohl, S.D., Gronstal, S.B. Separation of brown carbon from black carbon for IMPROVE and CSN PM2.5 samples, Air Waste Manag. Assoc., 68(5), 494-510, https://doi.org/10.1080/10962247.2018.1426653, 2018.

Gombi, C., Rahman, A., Hodovány, S. et al. A demonstrative study of a novel approach for spectral based source apportionment of ambient aerosols. Sci. Rep., 15, 19501, https://doi.org/10.1038/s41598-025-04022-3, 2025.

Olson, M. R., Victiria, G. M., Robinson, M. A., Rooy, P. V., Dietenberger, M. A., Bergin, M., Schauer, J. J. Investigation of black and brown carbon multiple-wavelength-dependent light absorption from biomass and fossil fuel combustion source emissions. J. Geophys. Res.-Atmos., 120(13): 6682-6697, https://doi.org/10.1002/2014JD022970, 2015.

23. Lines 407 – 408 and Lines 414 – 416: The potential-temperature gradient method is stated to yield accurate estimates at night and in the early morning in Lines 407 – 408. However, the potential-temperature gradient method is stated to overshoot the inversion top in Lines 414 – 416, which seems to be contradictory with the earlier statement of accurate estimates. Please revise the statements to clarify and avoid confusion.

Response: Thank you for pointing out this issue. As our study does not involve the

$PBLH_C$, we have removed the related statements from the manuscript.

24. Figure 5: Why are the PBLH (Parcel) lines only shown in some of the panels in the figure while the PBLH (Gradient) lines are shown in all panels? Please clarify in the revision.

Response: Due to adjustments made to the figures within the manuscript, this illustration has been renumbered as Figure 6. Based on Holzworth (1964)'s definition of the Parcel Method as: "The height at which a dry adiabatic line extending upward from the maximum surface temperature intersects the most recently observed temperature profile," it is more suitable for calculating the PBLH (Planetary boundary layer height) in unstable atmospheric conditions during the daytime. However, this method cannot calculate PBLH under stable nocturnal stratification conditions. Consequently, the PBLH calculated by the Parcel Method is not displayed in the figures representing the nighttime periods. Furthermore, during the 14:00 observation period, the observation height did not reach the actual PBLH, and therefore, the PBLH calculated by the Parcel Method is also not shown. Although the potential temperature gradient method can be used to calculate PBLH at every observation time, its results are more reliable under stable stratification, which is why it was used in this study to identify the PBLH for the (20:00, 23:00, 05:00, and 08:00) time slots. This explanation can be found in the Section 2.3 and first paragraph of Section 3.2 of the manuscript. In addition, we revised Figure 6 by retaining only the PBLH derived from the observational profiles. The specific content is as follows:

In Section 2.3:

Planetary boundary layer is the part of the atmosphere closest to the planet's surface, accounting for approximately 10% – 20% of the troposphere. It is the lowest layer of the troposphere directly influenced by surface forcing, with a response time of less than one hour, playing a critical role in the dispersion and transport of air pollutants. Within the PBL, turbulent mixing processes homogenize air temperature and humidity, resulting in relatively uniform distributions of these properties. PBL height (PBLH)

refers to the thickness of the layer most significantly affected by the surface. In other words, it is the vertical extent that surface turbulent motion (caused by surface heating, friction, or topography, etc.) can effectively influence (Emeis et al., 2008; Seibert et al., 2000). This study employs two established methodologies for determining the PBLH: the potential temperature gradient method and the parcel method (Holzworth, 1964; Zhang et al., 2020). The potential temperature gradient method was utilized for calculating the PBLH during the nighttime and early morning hours (20:00, 23:00, 05:00, and 08:00 LT), while the parcel method was applied specifically for the daytime periods (11:00, 14:00, and 17:00 LT). Detailed computational procedures for both approaches are summarized in Supplementary Table S1.

In Section 3.2:

The parcel method is well suited to convective conditions but cannot accurately resolve PBLH during early morning and nighttime (Holzworth, 1964), whereas it performs reliably under strong daytime convection. Using this approach, PBLH at 11:00 and 17:00 LT were 508 m and 450 m, respectively; at 14:00, measurement ceilings did not reach PBLH. By contrast, the potential temperature gradient method yields accurate estimates at night and in the early morning: PBLH at 05:00, 08:00, 20:00, and 23:00 LT were 260 m, 181 m, 260 m, and 223 m, respectively.

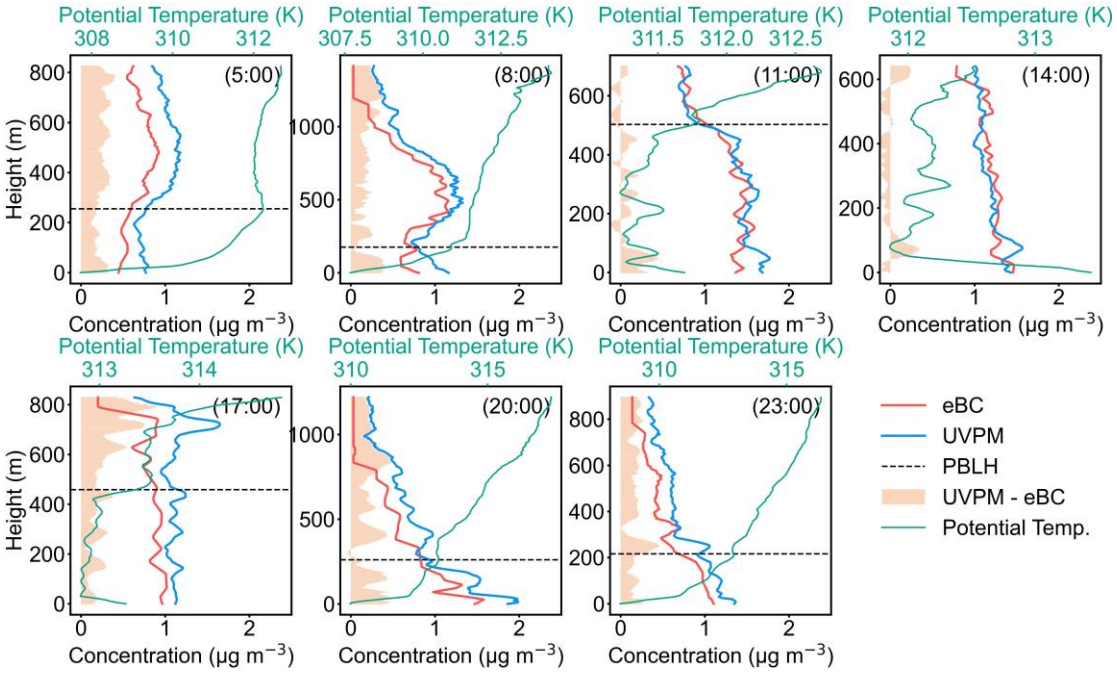

Figure 6. Diurnal variations in profiles of eBC, UVPM and potential temperature on July 27, 2024. The red line represents eBC, the blue line represents UVPM, and the green line represents the potential temperature profile. The light orange shaded area represents the difference between UVPM and eBC. The black dashed lines represent the PBLH.

References

Holzworth, G. C. Estimates of mean maximum mixing depths in the contiguous united states. Mon. Weather Rev., 92(5), 235-242, http://doi.org/10.1175/ 1520-0493(1964)092<0235:EOMMMD>2.3.CO;2, 1964.

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25. Lines 504 – 505: Point sources have continuous emissions. How can they be linked to high concentrations only infrequently or attributed to the cause of infrequently occurred high concentrations under the same wind direction? Please provide supporting information or data to strengthen this statement.

Response: Thank you for your valuable comment. We have carefully reviewed the manuscript content. Given that our observation site is located in a rural area with no stationary point sources. Sources of pollution nearby, the occasional high pollution concentrations observed under the same wind direction are likely associated with the non-scheduled production activities of local residents. Consequently, pollutants are not continuously transported to the observation site under a constant wind direction. We have revised the term "local point sources" to the more appropriate term "local and sporadic emissions" throughout the manuscript. The specific modification regarding this observation is as follows:

Southeasterly winds occasionally lead to relatively high UVPM values at both 100 m and 500 m altitude, although this occurrence is less frequent, which is consistent with the results of backward trajectories. We hypothesize that this variability may be associated with local and sporadic emissions from residents in the vicinity, as the observation site is located in a rural area with no stationary point sources. However, the specific cause cannot be definitively determined by the CPF alone. We will continue to

analyze this phenomenon in subsequent observational studies to provide a clearer explanation.

26. Lines 567 – 568: What about 14:00?

Response: Thank you for your suggestion. The reason that the 14:00 observation period was not included in this description is that the number of soundings collected at 14:00 during the campaign was insufficient (or limited), and none of them reached the corresponding PBLH for that time slot. This has already been mentioned in section 3.2 of the manuscript. The original text is as follows:

At 14:00 LT tethered-balloon sampling did not reach the PBL top, but observations within the PBL show uniform aerosol distributions, preventing a direct assessment of PBLH effects on concentration profiles.

27. Lines 570 – 572: Please double check the logic of this statement. It seems to be meant to contrast between thermodynamic and dynamic processes while the thermodynamic process is mentioned twice here.

Response: Thank you for pointing out this issue. Here, we made a writing error; the original intent should have been: "Consequently, the vertical distribution of carbonaceous aerosols within the daytime PBL is primarily governed by thermodynamic processes, in contrast to the combined dynamic and thermodynamic control that dominates within the nocturnal residual layer." We have made the correction in the manuscript, and the revised content is as follows:

Consequently, the vertical distribution of carbonaceous aerosols within the daytime PBL is primarily governed by thermodynamic processes, in contrast to the combined dynamic and thermodynamic control that dominates within the nocturnal residual layer.

28. Figure S2: Is there a specific definition of the term "conventional air pollutants"? It would be great if the caption could be specifically listing the air pollutants shown in this figure.

Response: Thank you for your comment. We have merged Figures S1 and S2 into a

single figure and have rewritten the caption. The revised content is as follows:

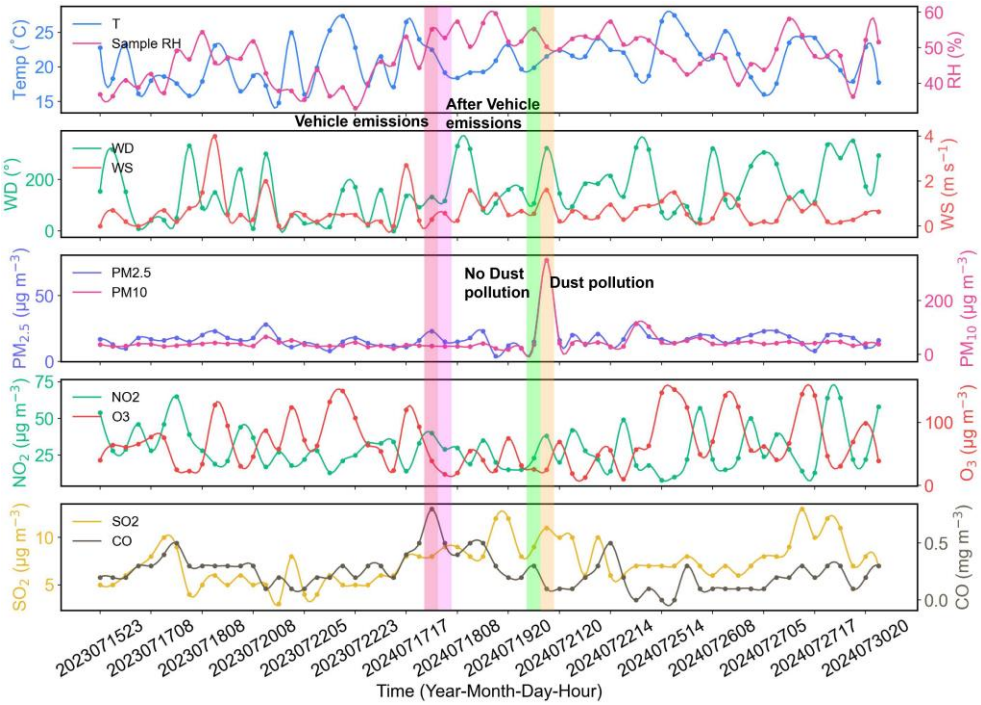

Figure S1. Temporal variations of meteorological parameters, including air temperature (T), relative humidity (RH), wind direction (WD), wind speed (WS), and air pollutants, including particulate matter with aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$) and than 10 μm (PM_{10}), nitrogen dioxide (NO_2), ozone (O_3), sulfur dioxide (SO_2), and carbon monoxide (CO), were observed during the monitoring period. The shaded areas in the figure represent the during and after vehicle emissions, no dust pollution, and dust pollution at 20:00 on July 17, 23:00 on July 17, 2024, 08:00 on July 20, 2024, 08:00 on July 21, 2024, respectively.

29. Figures S3 and S4: The terms "ascent" and "descent" are used in the main text and figure caption while the terms "rising" and "landing" are used in the figure legend, which are not consistent with each other. It would be great if the terminology could be consistent throughout the work.

Response: Thank you for your suggestion. We have revised this issue, and the modified content is as follows:

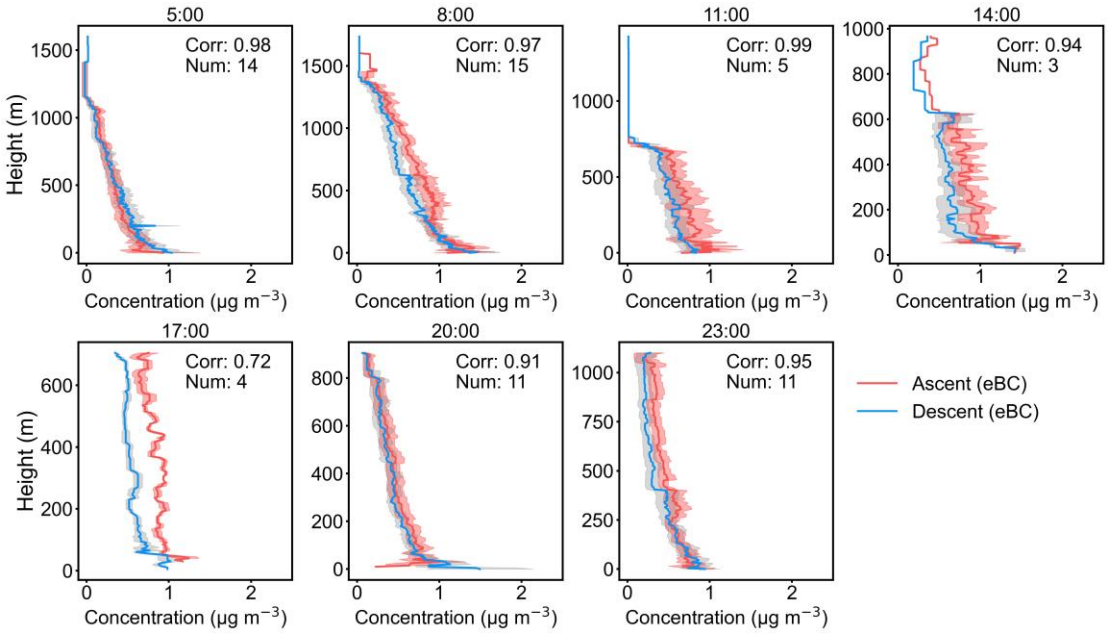

Figure S3. Comparison of equivalent black carbon (eBC) vertical profiles during ascent with those during descent of the tethered balloon.

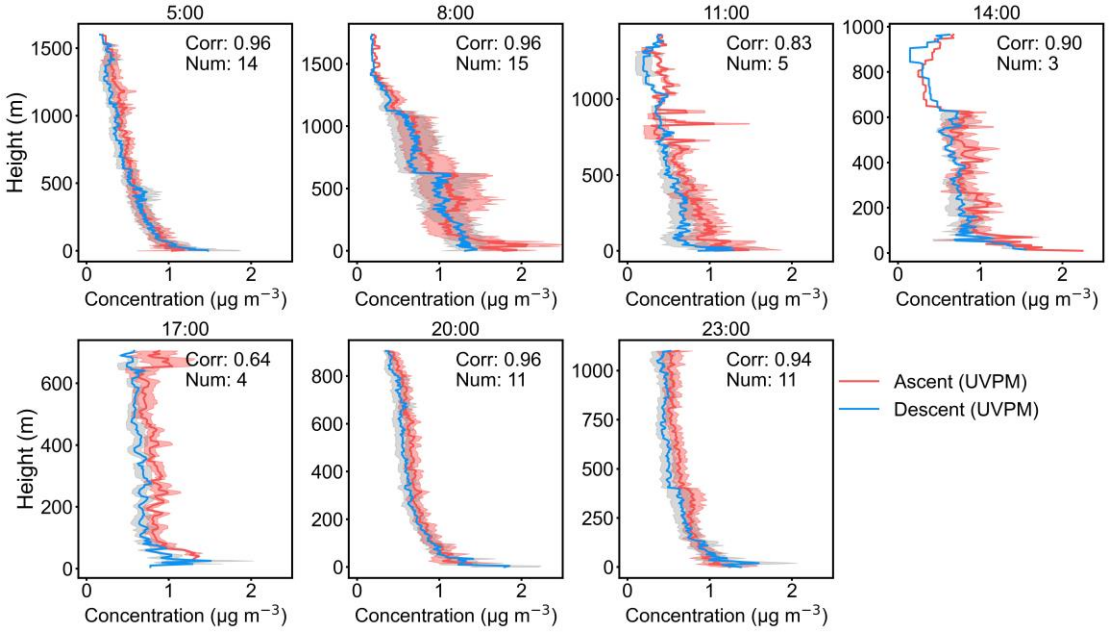

Figure S4. Comparison of ultraviolet-absorbing particulate matter (UVPM) vertical profiles during ascent with those during descent of the tethered balloon.

Technical Comments:

1. Line 23: The parameter "VTKE" seems to be a typographical error of "V_{TKE}".

Response: Thank you for pointing out this shortcoming. We have made the corresponding revisions in the manuscript. The modified content is as follows:

A comparison of the vertical profiles of eBC, UVPM, V_{TKE} (mechanical turbulence), and potential temperature showed that during the early morning and nighttime, when convective activity was weak, UVPM concentrations in the upper atmosphere were higher than those of eBC.

2. Line 413: The word "and" in front of the word "often" seems to be a typographical error and should be deleted.

Response: Thank you for pointing out this shortcoming. Since this study does not involve PBLHc, this part has been removed from the manuscript.

3. Line 637: The word "understanded" seems to be a typographical error of "understood".

Response: Thank you for pointing out this error. We have made the correction in the manuscript. The revised content is as follows:

Furthermore, the filed campaign was conducted at only a site, and thus the feedback between aerosols and the PBL meteorology cannot be fully understood at whole Loess Plateau.