Response to Referees' Comments

Response to Reviewer #2:

The manuscript addresses a critical and challenging issue in aerosol effects on weather and climate, i.e. the influence of aerosol vertical structure on boundary layer stability and height. The two-year concurrent observations from micropulse lidar, sunphotometer, and radiosonde were employed to provide direct and quantitative evidence on the aerosol radiative effects on the boundary layer development. The contrasting effects of different aerosol vertical structures identified by this study are important to know and call on a better representation of aerosol vertical profile in numerical models for future aerosol effect assessment. The paper is well written overall, and its scientific merit is clear. I recommend its publication with ACP, while I also have comments below for the authors to address.

Response: We appreciate the reviewer's positive and constructive comments on our work. All of the comments and concerns raised by the referee have been carefully considered and incorporated into this revision. Our detailed responses to the reviewer's questions and comments are listed below.

1. Figure 2, are the data here from three cases or composites from all available observations? I have the similar question for Figs. 3-7 as well. Please clarify the data source and sampling range in the figure captions.

Response: We clarify that the results are averaged from all available observations during the study period. We added the number of samples in the panels or in the descriptions of the figures. The latter are carefully checked to assure that the data sources are clearly stated.

2. Is R^* in Figure 3 for linear regression and R^* in Figure 5 for the inverse fitting?

Better to use different symbols for different types of regression.

Response: Revised, per your suggestion.

3. As shown in Figure 5, the weakly absorbing aerosols can also suppress PBLH. I assume it is caused by the reduction in solar radiation reaching the surface and the consequent suppression in surface latent/sensible heat fluxes. Since the authors have performed the radiation transfer simulations, I'm wondering if they can illustrate the importance of atmospheric heating and surface cooling for PBL development when absorbing aerosols are present.

Response: Thanks for raising this point. We clarify that both absorbing and weakly absorbing aerosols reduce the radiation reaching the lower atmosphere and the surface, thus suppressing the PBLH. Both decreasing and inverse aerosol structures can cool the surface and suppress sensible heat, thereby stabilizing the PBL. Despite the different aerosol structures, aerosols cause a notable stabilizing effect near the surface.

On the other hand, for the decreasing with height structure, the abundance of aerosols near the surface generates a stronger aerosol heating rate in the lower PBL than in the upper PBL. Such aerosol radiative forcing decreases the potential temperature gradient $(d\theta/dz)$ in the middle to upper PBL, further strengthening vertical convection in the middle to upper PBL. The opposite aerosol effects on PBL stability weakens the aerosol-PBL feedback, as is shown in our study. For the inverse aerosol structure, the significant heating effect in the upper PBL facilitates the formation of a temperature inversion and further increases the stability and suppresses the PBLH. The notable increase in stability lead to the strong, positive aerosol feedback, as is demonstrated in Figure R1 (the revised Figure 9).

Turbulent fluxes and eddies in the PBL can spread out and redistribute the radiative effects induced by aerosols. We need to resort to numerical simulations to quantify the aerosol impacts on the PBL. Since the aerosol vertical distribution

is still poorly represented in numerical models, improvements in model simulations are warranted to better understand the aerosol-PBL interaction quantitatively. This is an ongoing study by our team that will be presented in a future paper.

A detailed discussion has been incorporated into the revised Section 3.3 and Section 4.

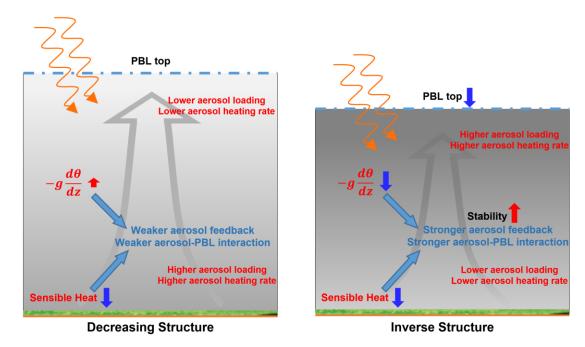


Figure R1. Schematic diagrams describing aerosol-PBL interactions when decreasing and inverse aerosol structures are present. The blue dash-dotted line indicates the top of the PBL. Orange curved arrows indicate solar radiation. The background grey arrow shows the vertical transport of humidity, aerosols, and heat. The background greyscale indicates the pollution level.

4. L285-286, the sentence is hard to follow. What do you mean by "significant heating in the different parts of PBL"?

Response: We revised this sentence as follows:

"Figure 7 shows that the vertical distributions of the heating rate differ drastically

among the different aerosol structures."

5. Since the authors possess ample observations data, can you show the occurrence/frequency of each aerosol vertical structure within PBL (decreasing, inverse, and well-mixed)? It is interesting to know the relative importance of those three structures in the real atmosphere. Moreover, can you sort out what factors determine those distributions within PBL?

Response: Per your guidance, we present the number of samples for each aerosol structure during the study period in the revised Section 3.1.

"The number of samples and percentages of decreasing, well-mixed, and increasing aerosol structures are 998 (51%), 611 (32%), and 330 (17%), respectively."

The decreasing structure is more frequent during the afternoon, partly due to the entrainment process. Through the development of a PBL, entrainment brings dry and clean air from the free atmosphere into the PBL, diluting the aerosol loading in the upper PBL. Note that multiple entangled factors can contribute to the formation of different aerosol structures within the PBL, including synoptic patterns, new particle formation, vertical turbulence, horizontal transport, entrainment rates, to name a few. The complexity of this issue is an important reason for the poor representation of the aerosol vertical distribution in numerical models.

This discussion has been incorporated into the revised Section 3.2.

6. L309-315 and Figure 9. What is the physical/chemical mechanism of the negative feedback, i.e. stable PBL leads to less aerosol formation? The color of big red arrow in the upper part of Figure should be changed to blue, as it is about negative feedback.

Response: A low PBLH and high stability increase the aerosol loading. This

mechanism is straightforward and clear. We investigate the opposite effect: how

aerosols affect the PBL via their feedbacks. If the aerosol heating effect is much

stronger on near surface than upper PBL, aerosol can decrease the stability in PBL,

and cause the negative feedback. Negative feedback partly offset the PBL's impact

on the aerosol loading. The large variations in the impact of aerosols on stability

and PBL development lead to different magnitudes in the aerosol-PBL interaction.

This discussion has been incorporated into the revised Section 3.3. We also use

a revised figure (Figure R1) to replace the previous Figure 9 to better demonstrate

the interactions.

7. *L334*, it should be Wang et al. 2013.

Response: Revised.

8. Please remove "conclusive" from the title, as it is a very subjective word.

Response: Per your comment, we changed it to "significant".

Response to Reviewer #1:

Aerosol-planetary boundary layer (PBL) interaction is proposed as an important mechanism to stabilize the atmosphere and exacerbate surface air pollution. Attempts to analyze aerosol-PBL interaction by using observation data are rare and worth encouraging. Thus, I recommend a minor revision before publication. The detail comments or suggestions are shown below:

Response: We appreciate the reviewer's positive and constructive comments on our work. All of the comments and concerns raised by the referee have been carefully considered and incorporated into this revision. Our detailed responses to the reviewer's questions and comments are listed below.

1. My main concern about this study is how to get cause-effect from correlations. As we know, PBL has a strong impact on surface aerosol concentration and aerosol vertical profile (forward effect). Compared to that, the impact of aerosol on PBL (reverse effect) can be treated as a perturbation. Thus, it is hard to get the contribution of reverse effect only. For example, Line 265 to 272. It is claimed that "In general, there are stronger correlations between PBLH and PM2.5 under inverse aerosols structure. This phenomenon indicates that the absorbing aerosol could play a more important role in the inverse aerosol structure." Let's imagine that the decreasing and inverse profile are formed by specific PBL structure, we may get a similar relationship between PBL height and PM2.5 in Figure 5. Moreover, it is possible that the correlations are caused by some other factors, simultaneously, like the front process or precipitation.

Response: Indeed, the PBL and aerosols mutually affect each other. While it is challenging to differentiate their respective impacts on each other, we have used the MPL-derived PBLH and in situ PM_{2.5} data to show their correlation, which can indicate the overall intensity of aerosol-PBL interaction, but cannot represent the feedback intensity. Since we considered only cloud-free cases, rainfall would

not affect the correlations. But many other factors do affect the PBLH- PM_{2.5} correlations, as well as the aerosol-PBL interaction. Therefore, the correlations cannot explain the causality and aerosol feedback loop. The correlations between PBLH and PM_{2.5} provide hints about the differences in aerosol-PBL interactions for different aerosol structures. Using SBDART constrained by ample observations, we investigated the vertical profiles of radiative forcing induced by aerosols and its impacts on atmospheric stability.

A detailed discussion has been incorporated into the revised Section 3.2.

2. I don't quite understand the role of Figure 6 and the corresponding part of the manuscript. It seems that Figure 6 does not support the topic directly and may be considered to be moved to SI.

Response: We apologize for not clearly describing and elaborating on the role of Figure 6 in making an important point.

In general, the inverse structure is characterized by higher aerosol loading and a lower PBLH, whereas the decreasing structure is characterized by light pollution and a well-developed PBL. For the inverse structure, the lower PBLH growth rate, along with high aerosol loading, can be explained by the strong aerosol-PBL interaction. The diurnal variations in aerosols and the PBL are controlled by many factors. The strong aerosol-stability interaction generates an unfavorable condition for aerosol vertical dissipation in the vertical, so surface aerosol loading can continuously accumulate due to emissions.

The discussion has been incorporated into the revised Section 3.2.

3. More quantitative analysis is needed in the Results part. I can barely find the detail of quantitative discussion figures, especially in 3.3. I'm not sure if Figure 7 is a specific case, a statistic scenario or just a diagram? It seems there are too many

diagrams in the manuscript.

Response: Figure 7 presents statistical results derived from all available measurements (same as Figure 6). We revised the figure description to avoid any misunderstanding. Based on observations, we calculate the aerosol radiative forcing by SBDART for all cases. We can then obtain averaged diurnal cycles of the vertical profile of aerosol radiative forcing for different aerosol structures.

4. It might be helpful to show some statistical information and meteorological condition information. For example, the occurrence/frequency of each aerosol vertical structure within PBL. Does it occur in specific seasons or weather conditions?

Response: We added the number of AEC profiles for different aerosol structures during the study period in the revised Section 3.1.

"The number of samples and percentages of decreasing, well-mixed, and increasing aerosol structures are 998 (51%), 611 (32%), and 330 (17%), respectively."

All three types of profiles commonly occur in the real atmosphere. The decreasing structure is more frequent during the afternoon, partly due to the entrainment at the top of a PBL. Through the PBL development, entrainment brings dry and clean air from the free atmosphere into the PBL, diluting the aerosol loading in the upper PBL.

Multiple entangled factors are related to the formation of different aerosol structures within the PBL, including synoptic patterns, new particle formation, vertical turbulence, horizontal transport, entrainment rates, to name a few. The complexity of this issue is an important reason for the poor representation of aerosol vertical distributions in numerical models. We will further investigate this issue, which will be presented in a future paper.

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2	lower-atmosphere stability and its critical role in aerosol-PBL	
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Abstract. Aerosol-planetary boundary layer (PBL) interaction was proposed as an important mechanism to stabilize the atmosphere and exacerbate surface air pollution. Despite the tremendous progress made in understanding this process, its magnitude and significance still bearhave large uncertainties and vary largely with aerosol distribution and meteorological conditions. In this study, we particularly focus on the role of aerosol vertical distribution on thermodynamic stability and PBL development by jointly using the micropulse lidar, sun photometersunphotometer, and radiosonde measurements overtaken in Beijing. Despite complex the complexity of aerosol vertical distributions, the cloud-free aerosol structures can be <u>largely</u> classified into three types: well-mixed, decreasing with height, and the inversed. Under these different aerosol vertical inverse structures, the. The aerosol-PBL relationships relationship and the diurnal cycles of the PBLHPBL height and PM_{2.5} associated with these different aerosol vertical structures show distinct eharacters.characteristics. The vertical distribution of aerosol radiative forcing differs drastically among the three types with strong heating in the lower, midmiddle, and upper PBL, respectively. Such a discrepancy in heating rate affects the atmospheric buoyancy and stability differently in the three distinct aerosol structures. Absorbing aerosolaerosols have athe weak effect of stabilizing the low-lower atmosphere under the decreasing structure than under the inverse structure. As a result, the aerosol-PBL interaction can be strengthened by the inverse aerosol structure, and can be potentially neutralized by the decreasing structure. Moreover, aerosols can both enhance and suppress the PBL stability, leading to both positive and negative feedback loops. This study attempts to improve our understanding of the aerosol-PBL interaction,

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- 49 which showsshowing the importance of the observation observational constraint of
- 50 aerosol vertical distribution for simulating thethis interaction and consequent feedbacks.

1. Introduction

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Aerosols eriticallyhave a critical impact on the Earth's climate through 52 aerosol-cloud interactions (ACI) and aerosol-radiation interactions (ARI), and). They 53 also continue to contribute toward the considerable uncertainty to quantifications and 54 55 interpretations of in quantifying and interpreting the Earth's changing radiation 56 budget and hydrological cycles (Charlson et al., 1992; Ackerman et al., 2004; Boucher 57 et al., 2013; Z. Li et al., 2011, 2017a; J. Guo et al., 2017; 2018, 2019a). Despite the great advances made in observationthe past decades in observational and modeling 58 studies of the aerosol effects in the past decades, it is still a challenge to accurately 59 quantify theaerosol effects on the climate system due to inadequate understanding of 60 61 some mechanisms and strong variations in aerosol type, loading, and vertical distribution (Haywood and Boucher, 2000; <u>Jacobson et al., 2001;</u> Carslaw et al., 2013; 62 <u>J.</u> Huang et al., 2015; <u>J.</u> Guo et al., 2016a; <u>Z.</u> Li et al., 2016; <u>Wang Wei</u> et al., <u>2019</u>2019a, 63 64 2019b). Aerosols are known tocan interact with thermodynamic stability through ARI 65 (Atwater, 1971; Bond et al., 2013). Absorbing aerosols can stabilize the atmosphere 66 (Ramanathan et al., 2001; Y. Wang et al., 2013; Ding et al., 2016), whereas they) and may also enhance convection and precipitation under certain conditions (Menon et al., 67 68 2002; <u>Z.</u> Li et al., 2017). Thermodynamic stability in the planetary boundary layer (PBL) dictates the PBL 69 development (Garret 1994; Stull, 1988; W. Zhang et al., 2018), thereby dominating the 70 vertical dissipation of surface pollutants to some degrees degree. Aerosols, in turn, have 71 important feedbacks on the stability in the PBL, depending on the aerosol properties, 72

especially those of the light absorption absorbing aerosols (e.g., black, organic, and brown carbon). However, due to large uncertainties in aerosol radiative forcing, it remains a challenge to quantify the impact of aerosols on thermodynamic stability and PBL development. Conventionally, increasing the aerosol absorption tends to stabilize the atmosphere, leading to a reduced PBL height (PBLH). A more stable atmosphere and lower PBLH will, in turn, increase the surface aerosol loading, which is the wellestablished positive feedback loop in the aerosol-PBL interaction (e.g., H. Wang et al., 2015; Ding et al., 2016; Petäjä et al., 2016; Dong et al., 2017; Zou et al., 2017; Q. Huang et al., 2018; Z. Wang et al., 2018. H. Wang et al., 2019. However, such a positive feedback loop may not be real for all situations and is subject to confounding factors such as aerosol type, aerosol vertical distribution, soil moisture, and PBL regime (J. Guo et al., 20192019b; Lou et al., 2019). Geiß et al. (2017) reported the ambiguous relationships relationship between surface aerosol loading and PBLH, while our previous study revealed weak correlations between surface pollutants and the PBLH in mountainous or clean regions (Su et al., 2018). A recent study by Lou et al. (2019) shows showed that aerosol has an even aerosols have a positive correlation with the PBLH under the stable PBL conditions, indicating the importance of thermodynamic conditions in the PBL-really matters. Among others, numerical models are one of the viable methods used to determine the aerosol impacts on stability and PBL (e.g., J. Wang et al., -2014; Ding et al., 2016; Y. Wang et al., 2018; Zhou et al., 2018; Wang et al., 2014). The aerosol optical depthsdepth (AOD), a measure of aerosol columnar loading, is usually taken into

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account in model simulations. However, the aerosol vertical distribution in models is generally prescribed and may differ largelygreatly from the real situation, which highly varies in the PBL and is closely linked to the significant uncertainties in aerosol radiative effects. With observational constraints, the role of aerosol vertical distribution distributions in aerosol-PBL interactions warrants a further investigation.

Coincidently, we have ample Ample observational datasets over for Beijing are available, including aerosol vertical distribution distributions derived from lidar, optical

available, including aerosol vertical distributiondistributions derived from lidar, optical properties derived from sun photometer, the sunphotometer, profiles of meteorological variables from radiosonde (RS), as well as and surface PM_{2.5} and meteorological parameters. Based on these measurements, a radiative transfer model is used to simulate the vertical profiles of aerosol radiative forcing that are employed to investigate the impact of aerosols on the buoyancy in the lower atmosphere.

The paper is structured as follows: Section 2 introduces the datasets and methods used. The Section 3 presents analyses of aerosol-PBL interaction interactions under different aerosol vertical structures are presented in Section 3. Section 54 discusses the results with a brief summary.

2. Data and Method

2.1. Site description

In this study, weWe utilized data from multiple sources in Beijing, a megacity located atin the North China Plain. As one of the most densely populated and well-urbanized regions in the world, Beijing is a polluted region with high concentrations of

absorbing aerosols (Y. Zhang et al., 2019). The micropulse lidar (MPL) located atin Beijing was operated continuously by Peking University (39.99°N, 116.31°E) from 118 MarMarch 2016 to DeeDecember 2018, with a temporal resolution of 15s15 s and a 119 vertical resolution of 15m15 m. Due to incomplete laser pulses correction pulse 120 corrections, the near-surface lidar blind zones for lidar zone is ~0.15 km. Background 122 subtraction, saturation, after-pulse, overlap, and range corrections are applied to raw 123 MPL data to calculate the normalized signals (Yang et al., 2013; Su et al., 2018). The 2017a). MPL data on raining days are excluded. Level 1.5 AOD and single-124 scattering albedos (SSA) are employed at multiple wavelengths (i.e., 0.44/, 0.5/, 0.67/, 0.87², and 1.02μm02 μm) from the Beijing RADI (40°N, 116.38°E) Aerosol Robotic 126 Network (AERONET) site duringfrom 2011-to 2018, at under cloud-free conditions (Holben et al., 1998; Zhang et al., 2017; Smirnov et al., 2000; <u>Y. Zhang et al., 2017</u>). 128 The radiosonde (RS) station (39.80°N, 116.47°E) ofin Beijing is, operated by the China 129 130 Meteorological Administration, which is ~25 km from the MPL site. The variables 131 observed at the RS station include the meteorologymeteorological data and profiles of 132 water vapor, temperature, pressure, and wind. The vertical resolution of the RS is altitude dependent and generally less than 8 m (J. Guo et al., 2016b; W. Zhang et al., 133 134 2018). The RS is routinely launched at 0800 Local Time (LT) and 2000 LT for each day, and is also is launched at 1400 LT in the summer (June-July-August). The RS 135 136 measurements are collected during 2011-2018. To reduce small-scale biasbiases and to obtain a stablepicture of the regional variation of particulate matter with the diameter smaller than 2.5 µm (PM2.5), we acquire mean PM2.5 data from twenty 138

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environmental monitoring stations <u>located</u> within 20 km from the lidar site, including one station <u>ofat the</u> Beijing Embassy of <u>the</u> United States. <u>The Figure 1 shows the</u> topography of Beijing <u>is presented in Figure 1</u>. The green square indicates the MPL site, and the yellow triangle indicates the AERONET station. The brown star represents the <u>radiosonde</u> (RS) station, and the red pink dots represent the PM_{2.5} sites.

2.2. Statistical analysis methods

Here the The statistical significance is tested by two independent statistical methods, namely, the least—squares regression and the Kendall' tau (MK) test (Mann, 1945; Kendall, 1975)—; J. Li et al., 2016). Least—squares regression typically assumes a Gaussian data distribution in the trend analysis, whereas the MK test is a nonparametric test without any assumed functional form. The latter is more suitable for data that do not follow a certain distribution. To improve the robustness of the analysis, a relationship is considered to be-significant when the confidence level is above 99% for both the least—squares regression and the MK test. Hereafter, "significant" indicates that the correlation is statistically significant at the 99% confidence level.

In this study, we We primarily use the linear—fit method to build the relationships between different parameters, and the The Pearson correlation coefficient derived from the linear regression analysis measures the degree to which the data fit a linear relationship. However, following our recent work (Su et al., 2018), the inverse fitting ([f(x) = A/x + B)] is used to establish the relationship between PBLH and PM_{2.5}. During this time, the The magnitude of the correlation coefficient (R^{\dagger}) is designed to

measure the degree to which the data fit an inverse relationship. Since the relationship

between the PBLH and PM_{2.5} is non-linear, the inverse fitting is more suitable to characterizebetter characterizes this relationship.

2.3. PBLH and buoyancy derived from RS

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The RS vertical resolution varies according to the balloon ascending rate, and. The RS is recorded records measurements every 1.2s2 s, which represents an approximate vertical resolution of 5–8 m. Prior to the retrieval of the PBLH, we further resample the radiosonde RS data to achieve a vertical resolution of 5–hPa with linear interpolation. We follow a well-established method developed by Liu and Liang (2010) to derive the PBLH based on the profiles of the potential temperature gradient that takes into account of different stability conditions. In this study, we only focus on PBLPBLs driven by buoyancy, and thus, the PBLso PBLs driven by the low-level jets will be are excluded using the RS-derived wind profiles from radiosonde (Liu and Liang, 2010; Miao et al., 2018).

The static stability <u>inof the</u> atmosphere is determined by the buoyancy force, which
can be expressed as (Wallace and Hobbs, 2006);)

$$B = \frac{d^2z}{dt^2} = \frac{T' - T}{T}g = -g\Delta z \frac{1}{\theta} \frac{d\theta}{dz} \quad , \tag{1}$$

where z is the height of the air parcel, and t indicates the time. t' represents the temperature of the parcel—and, t' represents the temperature of the environment, and t' is the virtual potential temperature of the environment. For a certain An atmospheric layer, the atmosphere is identified as a is convective condition when if the buoyancy

is above zero, but is identified as a __and_stable condition when the buoyancy is below zero. If the buoyancy is near zero, the atmosphere is under a neutral condition. Based

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on the identification method for PBL typestype (Liu and Liang, 2010; W. Zhang et al., 2018), we present profiles of buoyancy forcing for a-stable, neutral, and convective PBLPBLs (Figure 2a). Clearly, the Results shown are averages from 3069 radiosonde measurements, of which 438 cases are convective PBLs, 714 cases are neutral PBLs, and 1916 cases are stable PBLs. The strongest upward or downward forcing occurs near the surface. Figures 2b-c further show the height—dependent correlation coefficients between buoyancy and PBLH/PM2.5 with an interpolation window of 0.2km. Noted 2 km. Note that the PBLH and surface PM2.5 are fixed for the entire column, and the buoyancy is height-dependent. Due to the insufficient development of the PBL, we do not use RS data at 0800 LT here. To exclude the impact induced by the dragging effects of rainfall, we only use the consider cases without precipitation within the past 24 hours. Strong upward buoyancy can uplift the PBLH and mitigate the surface pollutants, especially in the lowlower atmosphere. Thus, we integrate the buoyancy forcing within the lowest 1km km (red line in Figure Figures 2b-c), which is defined as the lowlower-atmosphere buoyancy (LAB). As shown in Figure Figures 3ab, the LAB shows strong negative correlations with and PM2.5 but positive correlations with are negatively correlated, and LAB and PBLH. The are positively correlated. LAB also has a significant negative correlations with absorbing aerosol optical depth. It could (Figure 3c). This may be partly caused by due to the stabilizing effect of absorbing aerosolaerosols on the atmosphere, which is widely reported in many previous studies (H. Wang et al., 2015; Ding et al., 2016; Petäjä et al., 2016; Dong et al., 2017; Z. Li et al., 20172017b; X. Huang et al., 2018).

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2.4. PBLH and aerosol extinction coefficient derived from MPL

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MPL data from Beijing were used to retrieve the PBLH during the daytime (0800-206 _1900 LT). MultipleMany methods have been developed for retrieving the PBLH from 207 MPL measurements, such ase.g., the signal threshold (Melfi et al., 1985), the maximum 208 209 of the signal variance (Hooper and Eloranta, 1986), the minimum of the signal profile derivative (Flamant et al., 1997), and the wavelet transform (Cohn and Angevine, 2000; 210 Davis et al., 2000; Su et al., 2017/2017b; Chu et al., 2019). To derive the PBLH from 211 MPL data, we adopted previous well-established approaches with several refinements, 212 213 which hashave already been validated by long-term data overcollected at the Southern Great Plains (ARM SGP) site (Sawyer and Li, 2013; Su et al., 20192020). 214 215 Initially, we'We initially identify the local maximum positions (range: 0.25-4km-4 km) in the covariance transform function collocated with a signal gradient larger than 216 a certain threshold. We further estimated the shot noise (σ) induced by background light 217 218 and dark current currents for each profile, and then set the certain threshold as – 3σ. The 219 initial PBLH retrieval (0800LTat 0800 LT) is constrained by the PBLH value derived from the morning RS sounding. Then, the following PBLHs would beare retrieved 220 using a stability-dependent model based on continuity. The boundary Boundary layer 221 222 clouds are identified to diagnose the PBLH for cloudy cases. Figure 3d presents the comparison of summertime PBLH results derived from MPL and RS at 1400 LT, and 223 the showing good agreement is reasonably good (R = 0.79). 224

Multiple studies have provided a well-established algorithm to retrieve the vertical profiles of aerosol extinction coefficient (AEC) from MPL (egdata (e.g., Fernald, 1984;

Klett, 1985; Liu et al., 2012). Then, the The Klett method is further applied for retrievingused to retrieve extinction profiles (Klett, 1985). The column-averaged extinction-to-backscatter ratio (the so-called lidar ratio) is an important parameter in the retrieval processes process and is constrained using AERONET-derived AOD at 0.5 µm derived from AERONET-5 µm. The AEC is assumed to be equal within the blind zone. The overall uncertainties from the overlap function, the lidar ratio, the effects of multiple scattering, and noises are estimated to noise fall within athe range of 20–30% in the retrieval processes process (He et al., 2006).

2.5. Estimation of the impacts of aerosols on buoyancy

To illustrate theshow vertical profiles of aerosol radiative forcing, the Santa Barbara DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998) was used to simulate the atmospheric heating rate (dT/dt) induced by aerosolaerosols (Liu et al., 2012; Dong et al., 2017). The integrated integrated aerosol inputs include AODs, and SSAs (i.e., at 0.44, 0.67, 0.87, and 1.02μm02 μm) retrieved from AERONET measurements, as well as the and AEC profiles at 0.5μm5 μm obtained from the MPL. We also use the MODISModerate Resolution Imaging Spectroradiometer surface reflectance reflectances as the an additional inputs input (https://modis.gsfc.nasa.gov/data/dataprod/mod09.php). —We further use heating raterates induced by aerosols to estimate the impacts impact of aerosols on buoyancy.

Theoretically, the rate of change in buoyancy for a certain layer can be sexpressed as:

$$\frac{dB}{dt} = \frac{d}{dt} \left(\frac{T_0 - \Gamma_d \Delta z - T}{T} g \right) = \frac{\left(\frac{dT_0}{dt} - \frac{dT}{dt} \right) T + \frac{dT}{dt} (\Gamma_d - \Gamma) \Delta z}{T^2} g \quad , \tag{2}$$

where most parameters are defined in the same way as in Eq. (1), and Γ_d (Γ) represents the dry adiabatic lapse rate (environmental lapse rate). We will-primarily focus on the <u>rate of</u> change <u>rate</u> in buoyancy during <u>the</u> noontime <u>period</u> (1100-1500LT_ 1500 LT), when the PBL is well developed, and aerosol radiative forcing is strong. The <u>rate of change rate</u> in buoyancy (dB/dt) induced by aerosols is largely determined by the aerosol heating rate, which can be produced by the radiative transfer model. Additional inputs include the environmental lapse rate and temperature, which are obtained from noontime RS soundings in the summer. For other times, the environmental lapse rate and temperature are obtained from MERRA-2 reanalysis data, which assimilates coarse-resolution RS observationobservations (Rienecker et al., 2011). In this way, we can estimate dB/dt induced by aerosols with a primary focus on the daytime. Noted Note that the errors in MERRA-2 data would lead to the uncertainties in the estimated dB estimated dB/dt. A 1-3 K uncertainties uncertainty in MERRA-2 temperaturetemperatures (Gelaro et al., 2017) lead leads to 1-3% relative biases in the estimated dB/dt. Considering the large variation in dB/dt under for different aerosol structures, the biases resulting from MERRA-2 data are not a very serious issue.

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3. Results

3.1. Classification of different aerosol structure scenarios

By altering the adiabatic heating rate of the atmosphere, the aerosol vertical

distribution is of great importance to the PBL. Based on cloud-free AEC profiles in the PBL, aerosol vertical structures can be classified into three types: well-mixed, decreasing with height, and its inverse, increasing with height. If AEC varies by less than 20% within the lowest 80% of the PBL, it is considered a well-mixed structure. For the other cases, a decreasing structure indicates a peak in AEC near the surface, and the inverse structure indicates a peak in AEC in the middle or upper PBL.

To investigate the vertical variation of the PBL, the evolution of the PBLH has been to be taken into account. Following previous studies (Kuang et al., 2017; Ferrero et al., 2014), the: Kuang et al., 2017), vertical profiles were normalized by introducing a standardized height (H_s), which was calculated as follows:

 $H_{S} = \frac{z - PBLH}{PBLH} , \qquad (3)$

where z is the height above the ground, and H_s is 0 at the PBL top and -1 at ground

level. Then, Figure 4 shows the normalized vertical profiles of AEC during derived from

283 MPL data for different aerosol structures around noontime. The number of samples and

284 percentages of decreasing, well-mixed, and increasing aerosol structures are shown in

285 Figure 4998 (51%), 611 (32%), and 330 (17%), respectively. Since a temperature

inversion located at the PBL top traps moisture and aerosols, there is a sharp decrease

in the AEC <u>profile</u> from the PBL upper boundary to the free atmosphere. The Variations

288 <u>in the</u> aerosol vertical distribution largely varies depending depend on different

conditions, but share similar features underamong the different aerosol structure

290 patterns. Despite complex aerosol vertical distributions, these three types of profiles

can account for most of the cloud-free cases.

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3.2. PBLH and PM_{2.5} under different aerosol structure scenarios

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Absorbing aerosols trendtend to have a positive feedback with the PBLH, while aerosols and the aerosol vertical distribution plays a critical role in this process. We investigate the relationship between MPL-derived PBLH and PM_{2.5} for absorbing (daily average SSA ≤ 0.85) or weakly absorbing (daily average SSA > 0.9) aerosols under inverse/declining_for increasing/decreasing_aerosol structures during 0900-1900 LT (Figure 5). The PBLH-PM_{2.5} relationships can represent the intensity of the aerosol-PBL interaction. In general, there are stronger correlations between PBLH and PM_{2.5} underfor the inverse aerosolsaerosol structure. Under such structure, the PBLH-PM2.5 correlation also remains considerably stronger for absorbing than weakly absorbing cases. This is likely caused by substantial heating in the upper PBL, which would facilitatefacilitating the formation of a temperature inversion and further increase increasing the stability inof the PBL. While, under declining aerosols For the decreasing aerosol structure, the aerosols may not significantly redistribute the adiabatic energy. This phenomenon indicates Hence, the PBLH-PM2.5 correlation is relatively weak. Significant PBLH-PM2.5 correlations are found for both absorbing and weakly absorbing cases, indicating that the absorbing aerosol could scattering aerosols may also play a more an important role in the aerosol-PBL interaction, especially for the inverse aerosol structure. The Figure 6 presents the averaged diurnal cycles of AEC, PBLH, and PM_{2.5} for different aerosol vertical structures-are presented in Figure 6, classified based on the measurements made in Beijing.average AEC profiles during noontime. High humidity cases (surface relative humidity > 90%) and strong wind cases (wind speed > 5m s⁻¹) are excluded. Theoretically Here, both AEC and PBLH are derived from MPL data. Data are collected on 371 available days, of which 191 days have decreasing aerosol structures, 122 days have well-mixed aerosol structures, and 58 days have inverse aerosol structures. Multiple entangled factors can contribute to the formation of different aerosol structures within the PBL, including synoptic patterns, new particle formation, vertical turbulence, horizontal transport, entrainment rates, to name a few. In general, the inverse structure is characterized by higher aerosol loadings and lower PBLHs, whereas the decreasing structure is characterized by light pollution and a welldeveloped PBL. In theory, PM2.5 should generally decrease with increasing PBLH in the morning and forenoon due to the dilution effect. This situation is demonstrated clearly for decreasing aerosol structures. However, PM_{2.5} continuously grows during the daytime underwhen an inverse aerosol structures structure is present, regardless of the PBLH diurnal cycle. DespiteEven though many factors control the diurnal variations of aerosolin aerosols and the PBL are controlled by many factors, the strong aerosol-stability interaction may be an underlying scheme that further enhances generates an unfavorable condition for the vertical dissipation of aerosols, so the surface aerosol loading during the daytimecan continuously accumulate due to emissions. The correlations and statistical results concerning the PBLH and PM_{2.5} provide hints about the differences in aerosol-PBL interactions for different aerosol structures. However, these results cannot explain the feedback loop and causality. Therefore, we

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further use the SBDART model with the constraint of ample observations to investigate

the vertical profiles of radiative forcing induced by aerosols and its impacts on
atmospheric stability.

3.3. Aerosol radiative forcing for different aerosol structures

Following the description in Section 2.5, we calculate the statistical means of aerosol radiative forcing in the vertical for decreasing, well-mixed, and inverse aerosol structures, derived from the cases. The presented in Figure 6. Figure 7 shows that the vertical distributions of the heating rate differ drastically with significant heating inamong the different parts of PBL. This is caused by aerosol structures. For the inverse aerosol structure scenario, aerosols cause substantial heating in the upper PBL, which would facilitate facilitating the formation of a temperature inversion and further increase the stability in the PBL. Nonetheless, under the declining aerosol structure, the abundant aerosols—increasing the stability in the PBL. For the decreasing aerosol structure scenario, the abundance of aerosols at the bottom of PBL can cause a heating effect inheats the lower PBL, and hence, so can potentially enhance the convection in the PBL.

There are considerable differences in heating rate among the three distinct aerosol structures (Figure 8), which affects the atmospheric buoyancy and stability differently. On average, aerosols generally suppress buoyancy in the lower atmosphere. Such an effect is quite notable for the-inverse structure and is insignificant for the-decreasing structure with large standard deviations. Absorbing aerosol-isaerosols-are not very helpful for stabilizing lower-the-lower atmosphere under the-when a decreasing aerosol

structure is present, but playsthey play an important role under thewhen an inverse aerosol structure is present. As such, we expected expect the strongest aerosol-PBL interactions interaction to occur for absorbing aerosol cases under the when an inverse aerosol structure, which is present, consistent with the results shown in Figure 4. It should be noted that there are large variations in the impact of aerosol on buoyancy. Under an inverse structure, aerosol overwhelmingly enhance the stability in lowatmosphere, whereas, under decreasing structure, aerosols have the potential to either enhance or suppress the low-atmosphere stability depending on different cases 5. Figure 9 illustrates the shows schematic diagramdiagrams of the interactions between aerosols, stability, and the PBL-when decreasing/inverse aerosol structures are present. Overall, aerosol both decreasing and inverse aerosol structures can cool the surface and suppress sensible heat, thus stabilizing the PBL. In both cases, aerosols have notable stabilizing effects near the surface. When a decreasing aerosol structure is present, abundant aerosols near the surface generate a stronger aerosol heating rate in the lower PBL than in the upper PBL. Such aerosol radiative forcing lowers the potential temperature gradient ($d\theta/dz$) in the middle and upper PBL and can further strengthen vertical structure critically affects the convection in the middle and upper PBL. The opposite aerosol effects on PBL stability lead to a relatively weak aerosol feedback and a relatively weak aerosol-PBL interaction. The When an inverse aerosol structure is present, the significant heating effect on the upper PBL facilitates the formation of temperature inversion and further increases the

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stability and aerosol loading in the near surface. Therefore, the inverse aerosol structure

may strengthen the aerosol PBL interaction. Meanwhile, the aerosol PBL interaction can be potentially neutralized by the decreasing structure. Moreover, aerosols can both enhance and suppress the PBL stability depending on different conditions, and lead to both suppresses the PBLH. The notable increase in stability lead to the strong, positive and negative feedback loops (Figure 9), aerosol feedback.

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Highly variable aerosol vertical distributions cause large variations in the impact of aerosol on stability, and thus, exert important and highly variable influences on the aerosol-PBL interactions. Although aerosol stabilize PBL for majority cases, aerosol also can suppress the stability in low-atmosphere when aerosol heating effect is much stronger on the near surface than upper PBL, and further lead to a potential negative feedback loop. The positive feedback loop leads to strong aerosol-PBL interactions, while the whereas negative feedback loop partly offset PBL's impacts on leads to weak aerosol loading-PBL interactions. It explains the paradox of the impact of the PBL on different correlations between PBLH and surface pollutants, since its magnitude, significance, and even sign reportedly varies or even reverses (Quan et al., 2013; Tang et al., 2015; Geiß et al., 2017; Su et al., 2018).

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4. Summary and Discussion

Based on integrated aerosol and meteorological measurements overmade in Beijing, the aerosol-PBL interaction is assessed underfor different aerosol vertical structures, which are i.e., decreasing, well-mixed, and inversely increasing with height-respectively. The aerosol-PBL relationships and the diurnal cycles of PBLH and PM_{2.5}

show distinct characteristics among the different aerosol vertical patterns. For the decreasing aerosol structure, PM2.5 decreases in the morning and forenoon with relatively large PBLH growth rates. In this situation, absorbing aerosol is aerosols are not very helpful in stabilizing low-the lower atmosphere. Under For the inverse aerosol structure, PM_{2.5} continuously grows during the daytime with relativerelatively low PBLH growth raterates. This phenomenon could be a sign of thea strong aerosol-PBL interaction. The aerosol radiative forcing in the vertical-scale for decreasing, wellmixed, and inverse aerosol structures differ drastically with strong heating in the lower, midmiddle, and upper PBL₃ respectively. Such a difference in heating rate affects the atmospheric buoyancy and stability differently in the three distinct aerosol structures. Numerous studies used various models Turbulent fluxes and eddies in the PBL would spread out and redistribute the radiative effects induced by aerosols. Needed are numerical models to simulatequantify the aerosol-PBL interactions interaction and consequent feedbacks (e.g., Y. Wang et al., -2013; Ding et al., 2016; Z. Wang et al., 2018; Zhou et al., 2018; Wang et al., 2014). Aerosol vertical distribution highly varies in-distributions greatly vary on both temporal and vertical scales, and critically affect the aerosol radiative forcing. Nonethelesseffects. However, the aerosol vertical distribution usually is still poorly represented in numerical models, partly due to a lack of observational constraints. This study reveals the important role of the aerosol vertical distribution in the aerosol-PBL interactions interaction, which should be carefully taken into account in both observational analysis analyses and model simulations.

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In this This study, we use used column-averaged aerosol properties from

AERONET. However, the vertical variations of single scattering albedoin SSA and 424 425 aerosol type remains unknown, which can induceinducing uncertainties in the estimation of aerosol effects. In the future, we plan to use aircraft data from field 426 427 campaigns to better account for its influences for the influence of different types of 428 aerosols of with different properties. 429 430 Data availability. The hourlyHourly PM2.5 data are released by the Ministry of of Environmental Protection China 431 (http://113.108.142.147:20035/emcpublish). The MERRA-2 reanalysis data are 432 433 publicly available at https://disc.gsfc.nasa.gov/datasets?keywords=merra%202&page=1. The_AERONET 434 435 data are publicly available at https://aeronet.gsfc.nasa.gov. The meteorological Meteorological data are provided by the data center of the China Meteorological 436 437 Administration (data link: http://data.cma.cn/en). 438 Author contribution. T.S. and Z.L. conceptualized this study. T.S. carried out the 439 440 analysis, with comments from other co-authors. C. L., J. L., and W. T. carried out the MPL observationobservations. J. G. provided auxiliary data. W. H., C. S., W. T., J. W., 441 and J. G. provided useful suggestions for the discussionstudy. T. S. and Z. L. interpreted 442 the data and wrote the manuscript with contributions from all co-authors. 443

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711 Figures

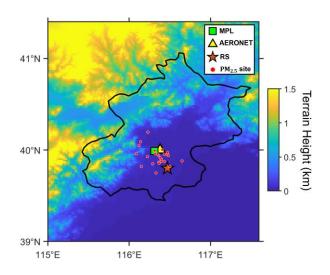


Figure 1. Topography condition of Beijing. The green square indicates the MPL site, and the yellow triangle indicates the AERONET station. The brown star represents shows the radiosonde (RS) station, and the red pink dots represents how the

716 PM_{2.5} sites.

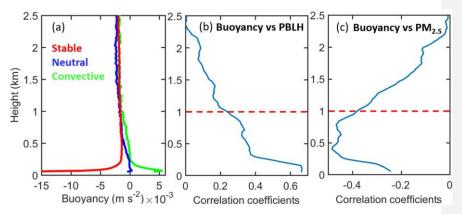


Figure 2. (a) Vertical Averaged vertical profiles of buoyancy forcing underin stable, neutral, and convective PBLPBLs. (b) Height-dependent correlation coefficients between buoyancy and PBLH. (c) Height-dependent correlation coefficients between buoyancy and surface PM_{2.5}. Noted Note that the PBLH and surface PM_{2.5} are fixed for the entire column, and the buoyancy is height-dependent. The buoyancy within low-in the lower atmosphere ((< 1 km) exerts has the most important impact on the PBLH and surface PM_{2.5}. The buoyancy and PBLH are derived calculated from RS measurements made at 1400 LT and 2000 LT from 2011 to 2018.

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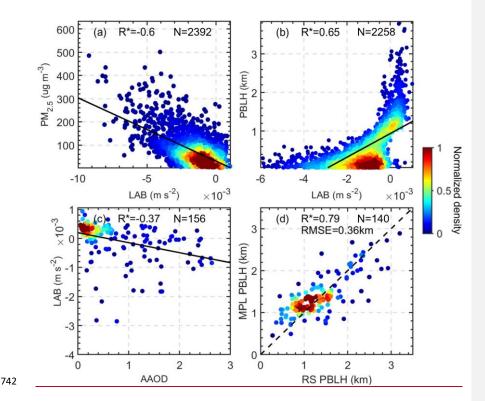


Figure 3. (a) The relationships relationship between low lower-atmosphere buoyancy (LAB) and PM_{2.5}. (b) The relationships relationship between LAB and PBLH. (c) The relationships relationship between absorbing aerosol optical depth (AAOD) and LAB. In (a, b, c), the LAB and PBLH are derived from RS measurements made at 1400 LT and 2000 LT, and AAOD is derived from AERONET measurements. The black solid lines indicate the best-fit lines from linear regressions regression. (d) Comparison of PBLHs derived from the MPL and RS at 1400 LT. Here Each panel gives the correlation coefficients (R), sample number (N), and in the following analysis, root-mean-square error (RMSE). R with asterisks an asterisk indicates that the correlation is statistically

significant at the 99% confidence level. The color-shaded dots indicate the normalized sample density.

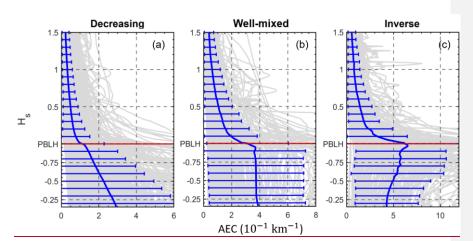


Figure 4. The normalized Normalized vertical profiles of aerosol extinction coefficients (AEC) under for (a) decreasing, (b) well-mixed, and (c) increasing (i.e., inverse) aerosol structures. The redRed line marks mark the position of the PBLH, the solid blue lines represent the average profile of corresponding profiles, and the error-bars represent the standard deviations.

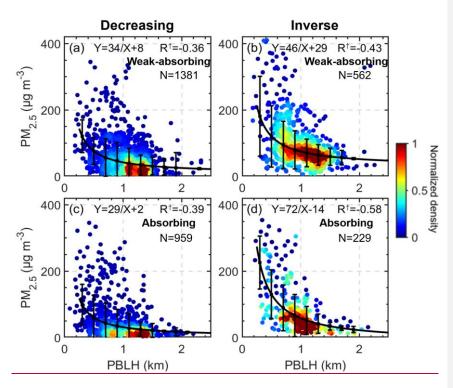


Figure 5. _The relationship between MPL-derived PBLH and PM_{2.5} for (a) weak-weakly absorbing and (c) absorbing under aerosols for the decreasing aerosols structure. The relationship between MPL-derived PBLH and PM_{2.5} for (b) weak-weakly absorbing and (d) absorbing under inverse aerosols for the increasing (i.e., inverse) aerosol structure. The blackBlack lines represent the inverse fitfits, and the whiskers indicate the standard deviations. The detailed fitting functions and number of samples are given at the top of in each panel, along with the correlation coefficient (R^{\dagger}) for the inverse fit.

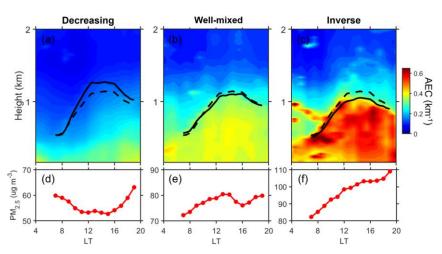


Figure 6. DiurnalThe averaged diurnal variations in AEC for (a) decreasing, (b) well-mixed, and (c) <u>increasing (i.e., inverse) aerosol</u> structures. The solidSolid black lines indicate the <u>corresponding PBLH averaged</u> diurnal cycles. The dashed black line represents of MPL-derived PBLH under the different aerosol structures. Dashed black lines represent the mean <u>MPL-derived PBLH</u> diurnal <u>eyelecycles</u>. (d, e, f) The <u>corresponding averaged</u> diurnal variations in <u>surface PM2.5 under the different aerosol structures</u>.

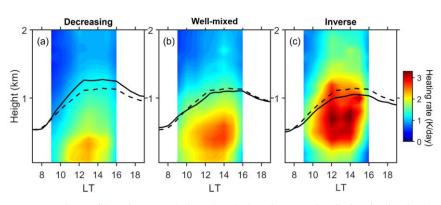


Figure 7. The profiles of averaged diurnal variations in aerosol radiative forcing in the vertical for (a) decreasing, (b) well-mixed, and (c) increasing (i.e., inverse) structures of aerosol loading. The solidSolid black lines indicate the corresponding PBLH mean diurnal cycles. The dashed of MPL-derived PBLH under different aerosol structures.

Dashed black line represents lines represent the mean MPL-derived PBLH diurnal eyelecycles.

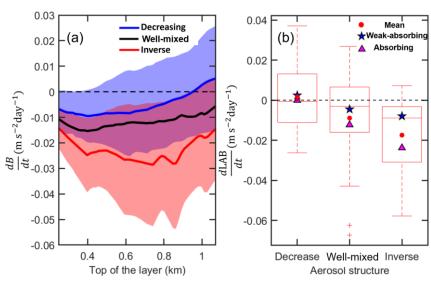
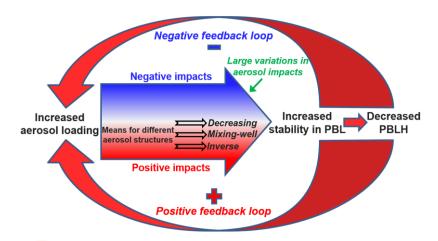


Figure 8. (a) The <u>rate of change rate-in buoyancy</u> (dB/dt) in a <u>certain-layer of the</u> lowest atmosphere <u>underfor</u> decreasing (blue), well-mixed (black), and inverse (red) aerosol structures during noontime. The bottom of the layer is the surface, and the rate of change in buoyancy is subjected to the top of the layer. The shaded areas show the standard <u>deviation deviations</u> of the rate of change <u>rate-in buoyancy</u>. (b) Box-and-whisker plots showing 10th, 25th, 50th, 75th, and 90th percentile values of <u>the rate of change rate-in LAB</u> (buoyancy within lowest <u>1km1 km</u>) during noontime. <u>The redRed</u> dots indicate the mean values, <u>while theand</u> blue stars and pink triangles show <u>the means for weak-weakly</u> absorbing (SSA > 0.9) and absorbing (SSA < 0.85) cases.

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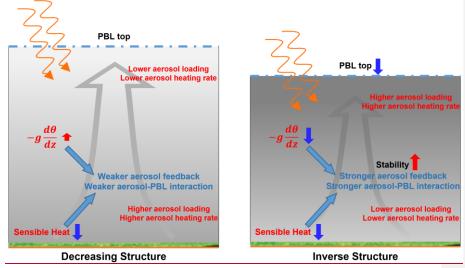


Figure 9. Schematic diagram showing the diagrams describing aerosol-PBL interactions between aerosols, stability, when decreasing and the PBL that comprise positive impacts (red arrows) and negative impacts (blue arrows). The aerosol impacts on stability largely vary with meteorology and aerosol conditions, and the means of aerosol impacts on stability for different inverse aerosol structures are indicated impresent. The blue dash-dotted line indicates the top of the PBL. Orange curved arrows indicate solar radiation. The background grey arrow sketches the vertical transport of

834	humidity, aerosols, and heat. The background greyscale indicates the diagram pollution
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