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## Revealing the Coupling Process between Aerosol, PBL, and Cloud: Identification and Mechanisms

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Abstract. Aerosols are mainly situated within the planetary boundary layer (PBL), affecting radiation, atmospheric stability, clouds, and precipitation. Aerosols, the PBL, and clouds may be coupled, influencing each other's development and further influencing meteorology and the atmospheric environment. The impacts and their interactions may be significant enough to affect weather and climate, depending on their states and coupling relationships. This study aims at understanding the fundamental processes governing the interactions between boundary-layer clouds and aerosols. By examining the development of boundary-layer clouds and aerosol transport in the context of surface-cloud coupling, we attempt to advance the understanding of their impacts on the development of convective clouds. To this end, we have developed algorithms for identifying the PBL height and cloud-surface coupling and have used them to investigate the fundamental mechanisms of aerosol-PBL-cloud coupling and their interactions, furthering our understanding of their joint effects on the development of convective clouds and air quality.

Keywords: Aerosol, PBL, cloud, cloud-surface coupling

#### **INTRODUCTION**

Aerosols exert important influences on Earth's climate through aerosol-radiation interactions (ARI) and aerosolcloud interactions (ACI). Despite extensive studies, these two effects still suffer from large uncertainties<sup>1, 2</sup>. The uncertainties are largely caused by insufficient understanding of integrated aerosol radiative effects and drastic variations in aerosol loading and properties at both temporal and spatial scales<sup>3, 4</sup>. As a key part of ARI, aerosols interact with the PBL, impacting lower-tropospheric thermodynamics<sup>2, 5</sup>. ARI has been recognized as one of the key mechanisms in modulating atmospheric stability and air pollution concentrations<sup>6, 7</sup>. Specifically, aerosols can regulate surface sensible heat fluxes, thus suppressing the PBL. The reduction in PBL height (PBLH) further facilitates air pollution accumulation, triggering severe air pollution events at the surface. These mechanisms constitute the well-recognized positive interaction<sup>2, 5</sup>.

It is unclear how convective clouds are entangled with entrainment altered by aerosols in the PBL. PBL entrainment, a process representing the turbulent exchange of air masses and heat fluxes between the free atmosphere and the PBL, can dictate PBL development and cloud evolution<sup>8</sup>. Because turbulent fluxes associated with entrainment cannot be solved mathematically, the interaction between entrainment and aerosols remains a fundamental challenge in a multi-scale chaotic system. The evolution of entrainment in the context of ARI has been poorly understood, warranting further investigation. Moreover, the parameterization of entrainment largely relies on a linear scheme, assuming a linear relationship between entrainment fluxes and surface fluxes<sup>9,10</sup>. In reality, the linear scheme may not reflect the responses of entrainment to aerosols, responses that are nonlinear and depend on aerosol properties and vertical distributions.

Convective clouds constitute another major uncertainty for ARI because they exert critical effects on surface heat fluxes, atmospheric thermodynamics, and PBL structures<sup>11</sup>. Due to cloud radiative effects, clouds interact with

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PBL thermodynamics and can change the dynamic framework of ARI. The cloud shading effect and cloud-top radiative cooling can change the PBL growth rate and phase transition, leading to drastic changes in ARI. Meanwhile, aerosols, especially the absorbing type, can affect cloud development by changing surface buoyancy and lower-atmospheric stability.

While the coupling process of marine clouds has been extensively investigated since the 1980s<sup>12-13</sup>, a robust approach to determine cloud-surface coupling over land is lacking. Using comprehensive field observations collected at the U.S. Department of Energy's Atmospheric Radiation Measurement Program's Southern Great Plains site, we have developed new methods to determine the long-term PBLH and cloud-surface coupling. They are instrumental in investigating the coupling process and interactions between aerosols, the PBL, and clouds. The entrainment process, ARI, and ACI are comprehensively analysed under different meteorological conditions. We have carried out a series of studies to illuminate the mechanism underlying these entangled interactions at the process level, summarized here to place the pertinent issues in context.

#### DISCUSSION

#### Identifications of PBL and PBL-cloud coupling over land

As the first step of our study, we have developed advanced methods to retrieve the PBLH and cloud-surface coupling from lidar and meteorological data. Reliable determination of the PBLH and cloud-surface coupling are challenging. Our methods have proven to be superior to many existing ones relying on lidar measurements, making it feasible to investigate integrated interactions between aerosols, clouds, and the PBL from the perspective of cloud-surface coupling.

Detailed techniques of the PBLH method have been presented in our previous study<sup>14</sup>, whose principles are briefly introduced here. Given the rapid change in the PBL over land, radiosondes cannot track the diurnal development of the PBL. We thus resort to ground-based lidar (e.g., the micropulse lidar, or MPL) as the primary tool to retrieve the PBLH. As a standard product of the MPL, backscatter profiles can be used to continuously track the development of the PBL at high temporal and vertical resolutions<sup>15</sup>. However, most previous approaches have been designed for well-mixed aerosol structures inside the PBL that change in harmony with the evolution of atmospheric thermodynamics. The validity of the first assumption depends on atmospheric stability, while the second implies that there is no residual aerosol layer decoupled with the PBL. To overcome these common limitations of traditional lidar-based approaches used to detect the PBLH, our method combines lidar-measured aerosol backscatter with a stability-dependent model of the PBLH temporal variation (DTDS). The new method can track the diurnal variations of the PBLH better than conventional lidar-based approaches<sup>14</sup>.

Figure 1 shows that the PBLH "calibrated" by morning sounding data is effective in circumventing the adverse impact of the decoupled aerosol residual layer. Evaluations of this new method show much better tracking of diurnal PBLH variations, with significantly smaller biases under various environmental conditions.



FIGURE 1. Daily backscatter profiles for (a) stable, (b) neutral, and (c) convective boundary layer cases. Backscatter is presented as a normalized signal on a log scale in arbitrary units. Black lines mark the PBLH retrieved from our new method (DTDS). Black dots indicate the local maximum positions identified in the backscatter profiles. The blue stars indicate the PBLH derived from radiosonde measurements. This figure is adapted from Su et al<sup>14</sup>.

Furthermore, we used the lidar-based PBLH method to identify cloud-surface coupling at a high temporal resolution. Figure 2 shows the relative positions between the cloud layer and the capping inversion of the PBL for coupled and decoupled cases<sup>16</sup>. Note that the virtual potential temperature decreases in the cloud layer, but the liquid water potential temperature is a near constant within the cloud layer. Hence, we use the relative position between the cloud layer and the capping inversion of the entrainment zone to reveal the coupling state. Specifically, the cloud base is below the capping inversion for coupled cases. We can thus determine the coupling state of continental clouds based on the quantitative differences between the PBLH, the lifting condensation level (LCL), and the cloud base. Here, the PBLH is derived from the MPL<sup>16</sup>, and the LCL is derived from surface meteorology. A cloud is identified as coupled if the cloud-base height coincides with the previous PBL top and LCL. Otherwise, the cloud is considered to be decoupled from the surface. As a result, the coupled state derived from this method is highly consistent with that derived from radiosondes<sup>16</sup>. Coupled clouds are sensitive to changes in the PBL, with a strong diurnal cycle, whereas decoupled clouds and the PBL are weakly related. Because of their distinct features, our new method offers an advanced tool to investigate coupled and decoupled clouds separately.



FIGURE 2. Idealized vertical profiles of virtual potential temperature under clear-sky, coupled-cloud, and decoupled-cloud conditions. Blue dashed lines delineate the surface layer, outer layer entrainment zone, and free atmosphere. Shaded areas show the cloudy layers, and pink dots show the PBLH. Red and green zones indicate coupled and decoupled regimes, respectively. Since it is not conserved in moist adiabatic processes, virtual potential temperatures decrease in the cloud layer. This figure is adapted from Su et al<sup>16</sup>.

#### Coupling process between aerosol and PBL entrainment

As a key factor in dictating PBL and cloud development, the role of entrainment is investigated in the context of ARI. Our observational study has demonstrated the mechanism behind the interactions between aerosols and PBL entrainment<sup>17</sup>. Because entrainment is highly variable, we proposed to use our PBLH method to derive the PBLH growth rate for computing the entrainment rate. We have employed comprehensive field observations to investigate the responses of entrainment processes to aerosols. We found that high aerosol loading can significantly suppress the entrainment rate, breaking the conventional linear relationship between sensible heat fluxes and entrainment fluxes.

Following quantitative analyses of ARM data, theoretical calculations, and reanalysis data, we proposed and demonstrated the mechanism of aerosol-entrainment coupling illustrated in Figure 3. The background grey arrow marks the vertical transport of humidity, aerosols, and heat fluxes. Under polluted conditions, aerosols can suppress sensible heat and vertical heat fluxes (blue arrows). As air masses are entrained from the free atmosphere into the PBL, the entrainment heat flux ( $F_{H_{zi}}$ ) is negative. Meanwhile, the magnitude of entrainment fluxes clearly reduces under polluted conditions. By reducing the amount of solar radiation reaching the surface, aerosols cool the surface, thus suppressing sensible heat fluxes. Nonetheless, the suppressed surface sensible heat cannot fully explain the observed responses of entrainment to aerosols, which are closely related to aerosol heating effects on the atmosphere. Since absorbing aerosols can noticeably heat the PBL, the atmosphere becomes more stable, and the PBL growth rate is reduced under polluted conditions. This effect is particularly strong for absorbing aerosols with an inverse aerosol vertical structure, leading to the suppression of both PBL turbulence and entrainment. The blue shaded area shows the perturbation in heat fluxes induced by aerosols. The suppressed entrainment process hampers

the transport of clean air from the free atmosphere to the PBL, leading to the continuous accumulation of aerosols. The abundance of aerosols within the PBL can further stabilize the atmosphere and reduce PBL turbulent fluxes, leading to a more suppressed entrainment process. These mechanisms constitute a positive feedback loop that amplifies the interaction between aerosols and entrainment. Due to these interactions, entrainment and aerosols are coupled here. The strong interaction between entrainment and aerosols is referred to as aerosol-entrainment coupling.

This study has broad implications for PBL parameterization<sup>17</sup>. Through ARI, aerosol-entrainment coupling can negate the widely used linear parameterization between entrainment fluxes and surface sensible heat fluxes. Since aerosol-induced heating in the vertical has not been accounted for in the linear scheme, we found that aerosol-entrainment coupling is poorly represented in linear parameterizations and associated model simulations. This issue can further lead to major biases in the PBLH derived from models. For instance, we found that the PBLH is considerably overestimated in ERA-5 reanalysis data under severe air pollution conditions, which is caused by the inaccurate estimation of the PBL entrainment rate. We thus reveal that aerosol-entrainment coupling accurate the polluted conditions. This calls for including aerosol-entrainment coupling in PBL parameterizations to enhance model capabilities and accuracies for a polluted environment. Neglecting aerosol-entrainment coupling would also lead to underestimating air pollution near the ground, revealed in one of our early studies<sup>3</sup>.



FIGURE 3. The background grey arrow sketches the vertical transport of humidity, aerosols, and heat fluxes. Orange, curved arrows represent solar radiation. The blue dash-dotted line represents the position of the PBL top (zi). Black, curved arrows indicate sensible heat fluxes. Red, curved arrows indicate entrainment at the PBL top.  $F_{H_s}$  and  $F_{H_{zl}}$  represent surface sensible heat fluxes and entrainment heat fluxes, respectively. The blue arrows represent the suppression effects of aerosols on surface sensible heat and vertical heat fluxes. The blue shaded area indicates the perturbation in heat fluxes induced by aerosols. Red arrows represent the accumulation of aerosols when entrainment is weak. Due to these interactions, entrainment and aerosols are coupled here (marked as the pink double arrow). This figure is adapted from Su et al<sup>17</sup>.

#### CONCLUSION

To better understand aerosol climate effects, we have conducted a series of studies to reveal interactions between aerosols, the PBL, and clouds. Cloud-surface coupling is a key factor affecting their interactions due to the following processes: (1) cloud-surface coupling determines the vertical transport of aerosols from the surface to the cloud base; (2) it controls whether ARI can directly affect cloud development; and (3) it dictates the impacts of cloud radiative effects on PBL thermodynamics. Our studies aim at advancing our understanding of these processes.

As the foundation of our studies, we have developed algorithms for determining the PBLH and cloud-surface coupling using long-term datasets of lidar, radiosonde, and meteorological variables acquired at the Southern Great Plains site from 1998 to 2019. Comprehensive evaluations of our methods indicate superior performance over the traditional approaches, with higher correlations and smaller biases under various pollution conditions. It provides more reliable tracking of the diurnal evolution of the PBL. Together with previous studies on coupling for marine clouds, we can now identify cloud-surface coupling over both ocean and land at a high temporal resolution. The state of cloud-surface coupling retrieved from lidar data with our method agrees well with those derived from radiosondes. Cloud-surface coupling can also help identify the PBLH under cloudy conditions, a common problem in lidar remote sensing.

We also applied our methods to investigate the fundamental mechanisms of aerosol-PBL-cloud coupling and interactions. By analyzing comprehensive field observations, we proposed a new mechanism to demonstrate the coupling process between aerosol and PBL entrainment. Specifically, an aerosol-inhibiting effect (noticeably stronger for absorbing aerosols) is revealed in the entrainment process, breaking the conventional linear PBL parameterization. Dictated by aerosol vertical distributions, aerosol radiative effects can alter turbulence in the PBL, leading to notable responses of entrainment to aerosols. A strong interaction and coupling process between aerosols and entrainment is demonstrated, forming the mechanism of aerosol-entrainment coupling. Strong interactions between aerosols and the entrainment process are found. Aerosol-entrainment coupling has critical impacts on PBL parameterizations and. Accounting for this new mechanism can remedy a common problem in the model simulation of the PBL.

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