

reviewer 1

It is the first time to analyze the sources of aerosols in Beijing using urban sites at a height of 528 meters. The research has clear research significance and application value. However, the article needs to conduct a thorough analysis and further clarify the following issues.

Thanks for your positive review on this work.

1. In Figure 2, why do the diurnal variation characteristics of various aerosol components differ? Is it due to different sources? Further analysis is needed.

Thanks for your valuable suggestion. Besides regional transport, we also discussed the influences of boundary layer process and aerosol chemical processing on the diurnal variation of aerosol components. We added some discussion in Section 3.2 and Section 3.4.

In Section 3.2: "The diurnal variations of aerosol components are shown in Fig. 2b, which offer insight into potential emission sources, local and regional transport, and aerosol chemical processing. Compared with observations at ground level, the diurnal variations in aerosol components at the CITIC station were remarkably weak (Fig. 2). For example, the ratio of the peak (12:00) to trough (06:00) in Org was 1.15, and the ratios in SO_4^{2-} , NO_3^- , NH_4^+ , and Cl^- were 1.09, 1.13, 1.09, and 1.15 respectively. Whereas, such ratios was about 2.0 at ground level (Zhou et al., 2020). Moreover, the peaks of Org, SO_4^{2-} , and Cl^- appear in the daytime, which are also contrary to that at ground level (Zhou et al., 2020; Quan et al., 2024).

The diurnal variability in aerosol components is strongly modulated by meteorology, particularly the PBL process (Quan et al., 2013). The PBL height (PBLH) typically decreases at night and grows during the daytime (Stull, 1988). This daytime expansion in PBLH enhances the vertical mixing and dilution of pollutants, thereby reducing their near-surface concentrations (Habineza et al., 2025). However, the circumstances differ significantly at the elevated CITIC station (528 m a.g.l.), which lies above the PBL at night but within it during the daytime. Our previous observations (17 October - 12 November 2020) indicated that the nocturnal PBLH was typically below 300 m, whereas the daytime PBLH was around 1000 m (Ma et al., 2023). At night, near-ground emissions are trapped within the shallow nocturnal PBL, while vertical transport to the overlying layer is substantially suppressed (Quan et al., 2025), resulting in low concentrations of pollutants at the CITIC station. In the morning, the PBL gradually develops due to increasing solar radiation. During this process, near-ground pollutants are vertically transported up to the CITIC station, thereby enhancing their concentrations. Additionally, enhanced formation of secondary organic aerosol (Cai et al., 2023) and sulfate (Liu et al., 2025) during daytime may also contribute to their daytime peaks. These processes jointly result in weak diurnal variations in aerosol components at the CITIC station. It is noteworthy that NO_3^- at the CITIC station remained at a high level at night but decreased slightly in daytime, which may be related to active nocturnal nitrogen chemistry above the

PBL (Ma et al., 2023). More details on vertical mixing of pollutants will be presented in section 3.4. "

In Section 3.4: " To better understand the vertical mixing of pollutants, we further analyzed the diurnal variations in aerosol components from October 17 to November 12, 2020. Driven by the daytime expansion of the PBL in the morning, strong vertical transport typically occurred. This process was identified by an abrupt increase in NO. Given that NO is a short-lived gaseous pollutant, its sharp increase in the 528 m layer serves as an indicator of upward transport from the surface. Based on the temporal evolution of NO, the strong vertical transport period was determined as 11:00-13:00 (Fig. S3). Accordingly, the variations in pollutants at the CITIC station during this specific period were investigated.

As illustrated in Fig. 10, the NO concentration at the CITIC station surged by 68.1% between 11:00 and 13:00, whereas SO₂ decreased by 5.9% during the same period. These observations substantiate the distinct emission sources of NO_x and SO₂ in Beijing. This divergence is further corroborated by the variations in NO₃⁻ and SO₄²⁻; NO₃⁻ increased by 21.9%, while SO₄²⁻ declined by 2.8%. Additionally, the mass concentration of Cl⁻ decreased by 7.3% during the same period, indicating its primary origin from regional transport. Although the mass concentrations of Org and NH₄⁺ also increased, their growth rates (Org: 6.3%; NH₄⁺: 11.2%) were notably lower than that of NO₃⁻, suggesting that regionally transported aerosols are particularly enriched in Org and NH₄⁺. Consequently, the downward entrainment of these regionally transported aerosols from the upper layer via PBL processes exacerbates surface air pollution in Beijing. "

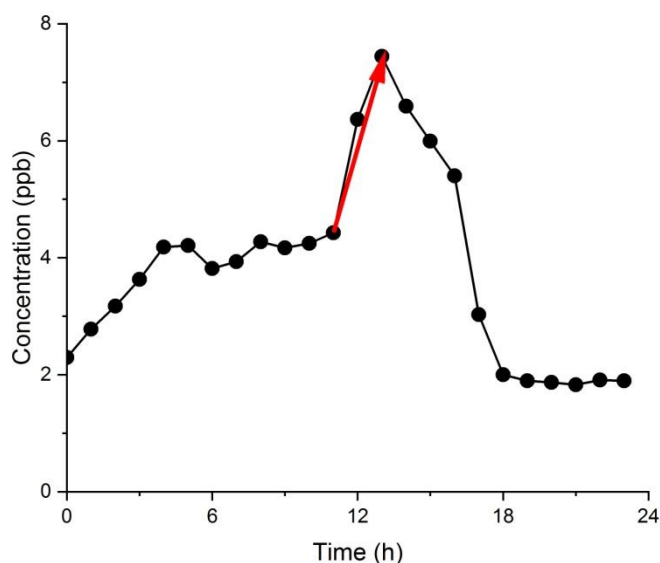


Fig. S3 The mean diurnal variation of NO at the CITIC station from October 17 to November 12, 2020. The red arrow shows the strong vertical transport period from

11:00 to 13:00.

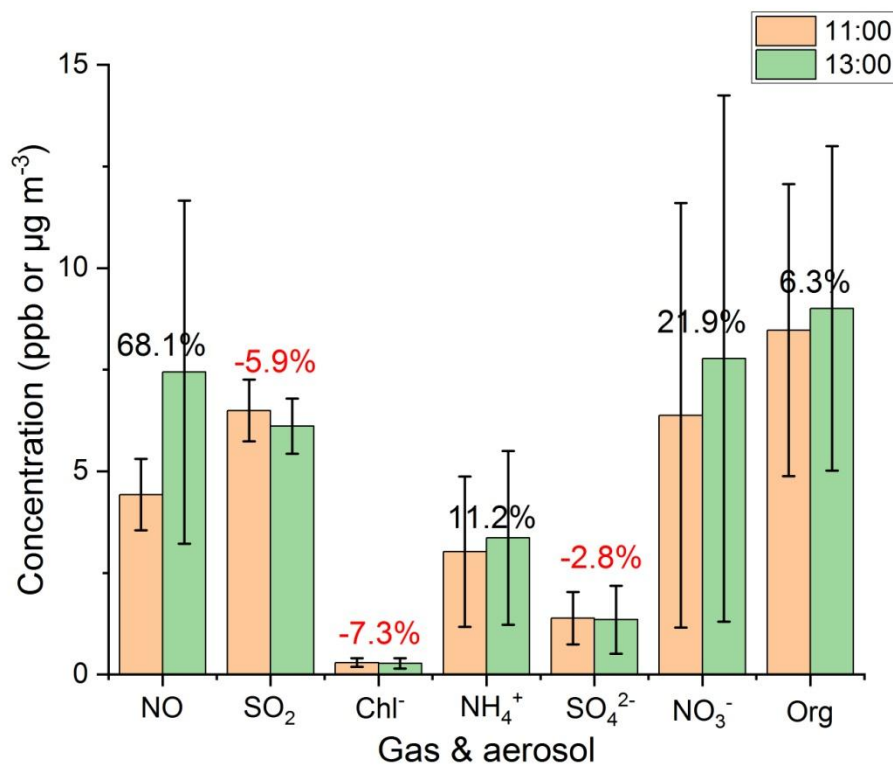


Figure 10. Mean concentrations of gases and aerosol components at the CITIC station at 11:00 and 13:00 from October 17 to November 12, 2020. Values indicate the percentage variations of pollutants between 11:00 and 13:00. Error bars represent 50% of the standard deviations.

2. What are the differences between analyzing the source of aerosol components at 500 meters and at ground observation stations? Especially, what are the differences in source statistics and potential source area analysis? A difference analysis needs to be provided.

Thanks for your valuable questions. A key distinction between the CITIC and ground stations lies in their vertical positioning: they reside in different atmospheric layers at night but within the same layer during the day. According to Ma et al. (2023), the nocturnal planetary boundary layer height (PBLH) was typically below 300 m, whereas the daytime PBLH reached approximately 1000 m. Consequently, the CITIC station was situated above the PBL at night, making it more strongly influenced by regional pollutant transport than ground-level stations. To substantiate this point, we have revised Section 3.4 to compare aerosol compositions when the CITIC station was above versus inside the PBL, as reply to Q1.

Furthermore, we examined the potential source contributions to ground-level air quality in Beijing across the six clusters (Fig. S1). The results show that potential

source contributions at the ground level originated more from local sources than those in the aloft layer.

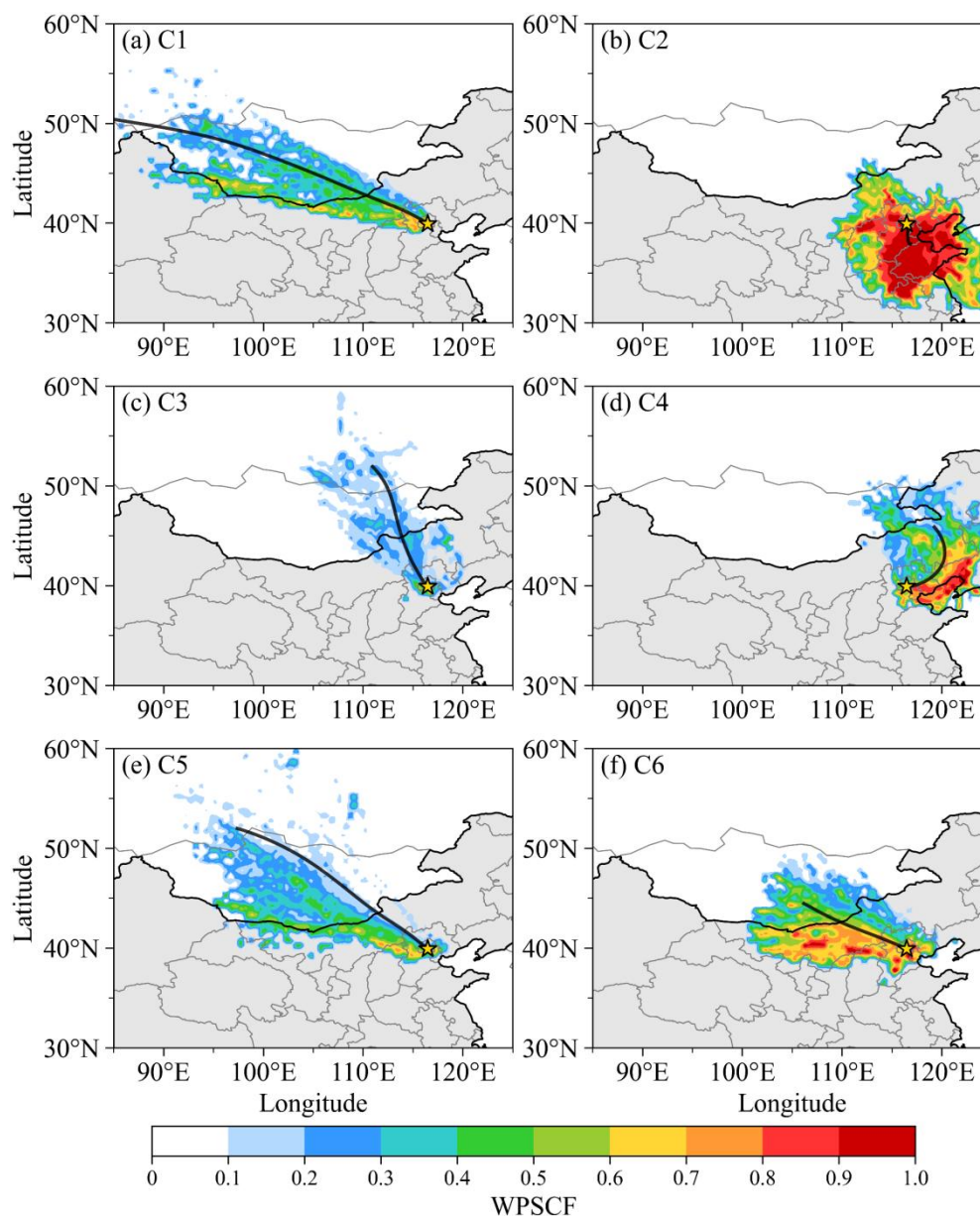


Fig. S1 Potential source contribution at ground level in Beijing in the six clusters

3. In Figure 9, why is the NO₂ at the CITIC station higher than that at the NZG station near the ground in the afternoon? Why is there still a difference in SO₂ concentration between the two stations at noon when convection is strong, instead of being close?

Thanks for your valuable suggestion. We supplied the following discussion in revision: "It is noteworthy that daytime NO₂ at the CITIC station is higher than at the NZG station, likely due to the conversion of NO to NO₂ during vertical mixing. NO accounts for about 39% of total NO_x at ground level, but decreases to 22% at the CITIC station (Ma et al., 2023). SO₂ at the CITIC station is higher than at the NZG station throughout the day, likely due to regional transport."