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Anthropogenic pollution elevates the peak height of new particle formation from planetary boundary layer to lower free troposphere

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Key points:

1. Anthropogenic pollution elevates the peak height of new particle formation from planetary boundary layer to lower free troposphere;
2. Distance of NPF peak to PBL top increases with increasing aerosol loading;
3. Findings shed new light on our understanding of NPF occurrence and NPF vertical distribution.

Abstract: New particle formation (NPF) and subsequent growth are primary sources of atmospheric aerosol particles and cloud condensation nuclei. Previous studies have been conducted in relatively clean environments; investigation of NPF events over highly polluted megacities is still lacking. Here we show, based on a recent year-long aircraft campaign conducted over Beijing, China, from April 2011 to June 2012, that NPF occurrence peaks in the lower free troposphere (LT), instead of planetary boundary layer (PBL), as most previous studies have found, and that the distance of NPF peak to PBL top increases with increasing aerosol loading. Further analysis reveals that increased aerosols suppress NPF in PBL, but enhance NPF in LT due to a complex chain of aerosol-radiation-photochemistry interactions that affect both NPF sources and sinks. These findings shed new light on our understanding of NPF occurrence, NPF vertical distribution, and thus their effects on atmospheric photochemistry, clouds, and climate.

1. Introduction

As primary sources of atmospheric aerosol particles and cloud condensation nuclei, NPF and its subsequent growth have drawn increasing attention [Kulmala *et al.*, 2004; Yu and Luo, 2009; Merikanto *et al.*, 2009; Venzac *et al.*, 2008; Zhang *et al.*, 2012]. Previous studies have identified two main regions of NPF occurrence: (1) well-mixed planetary boundary layer (PBL) where most anthropogenic pollutants and natural gas precursors are emitted and located [Väänänen *et al.*, 2016; Wehner *et al.*, 2007; Laakso *et al.*, 2007; Siebert *et al.*, 2004; Stratmann *et al.*, 2003; O'Dowd *et al.*, 2009; Pryor *et al.*, 2011; Crumeyrolle *et al.*, 2010; Dunne *et al.*, 2016], and (2) the upper free troposphere (UT) where NPF may be related to low temperature and cloud processes [Dunne *et al.*, 2016; Krejci *et al.*, 2003; Clarke *et al.*, 2013; Young *et al.*, 2007; Mirme *et al.*, 2010; Weigel *et al.*, 2011; Wang *et al.*, 2016; Lee *et al.*, 2003; Weigelt *et al.*, 2009]. Rare NPF events in lower free troposphere (LT) have been reported, and are thought to be related to the upward transport of PBL-originated air rising along valleys [Venzac *et al.*, 2008; Bianchi *et al.*, 2016] or convective movement [Rose *et al.*, 2015]. LT particles are thus thought to be either up-transported from PBL or down-transported from UT [Merikanto *et al.*, 2009; Wang *et al.*, 2016]. However, previous studies have been conducted in relatively clean environments; investigation of NPF events in LT over highly polluted megacities is limited. Also poorly understood is the quantitative relationship of NPF occurrence to aerosol loading. Furthermore, developing countries, such as China and India, are experiencing serious air pollution and high aerosol loading accompanied with rapid economic development, with the observed PM_{2.5} mass concentration reaching up to 600 $\mu\text{g m}^{-3}$ [Quan *et al.*, 2014]. In addition to harmful health effects, there is also a pressing need to understand the physical and chemical mechanisms underlying such high aerosol loading that can drastically affect Earth's energy balance, atmospheric photochemistry, clouds, and climate. To fill the gap and need, a comprehensive, year-long (Apr. 2011 to Jun. 2012) aircraft field campaign was conducted over Beijing, China, one of the most

aerosol-polluted regions in the world. Analysis of this unique dataset uncovers new NPF phenomena and provides a new understanding of NPF in highly polluted megacities like Beijing.

2. Field Campaign and Data

A total of 42 flights (118 flight hours) with an instrumented Y-12 aircraft was conducted during the field campaign. A fixed flight pattern was utilized with quasi-circular horizontal legs over the inner Beijing city, spiraling down around the rectangle of the 4th ring road from 2.1 km to 0.6 km (above sea level), with an vertical interval of 300 m, and vertical sounding flights at the Shahe airport (up to 3.6 km) after departure and before landing (Fig.S1). Aerosol properties, gas pollutants, and meteorological parameters were measured. The details about the instruments and data processing are described in the Supplementary Information (SI). A total of 28 NPF events during the campaign was identified according to the measurements of a Scanning Mobility Particle Sizer (SMPS, TSI-3936, USA), which measures particles from 14 to 661 nm in diameter with 107 size bins and a temporal resolution of three minutes. An NPF event is defined if the following two criteria are both satisfied: (1) the concentration of SMPS-measured ultrafine condensation nuclei (UCN) (diameter ≤ 25 nm) exceeds 10^3 cm^{-3} ; (2) the ratio of UCN concentration to the total SMPS aerosol (CN) concentration (hereafter UCN-CN ratio) is larger than 20%. The aerosol size distributions were bimodal for the NPF events, as opposed to the mono-modal for the non-NPF events (Fig.S2).

3. Results and discussion

To examine the effects of aerosol loading on NPF, the NPF occurrence data are partitioned into two groups of high (HA) and low (LA) aerosol loading according to the SMPS-measured aerosol surface area concentrations (SA) at ground level. The HA and LA groups represent the averages of all samples with the ground SA $\geq 500 \mu\text{m}^2\text{cm}^{-3}$ and SA $< 500 \mu\text{m}^2\text{cm}^{-3}$, respectively. A comparison of the vertical profiles of NPF events reveals striking differences between the HA and LA groups (Figure 1). Under LA, NPF events tend to occur within PBL (10 out of 13 cases, Fig.1a) whereas NPF events tend to appear near the PBL top and LT under HA (12 out of 15 cases, Fig.1b). On average, the height with peak NPF frequency elevated from 0.9 km under LA to 2.1km under HA (Fig.1c). This contrast between LA and HA can be further seen from Fig.1d, which shows the relationship between peak NPF height and PBL top height. The peak NPF height (H_{NPF}) is determined from the vertical profile of UCN-CN ratio as the height where the maximum UCN-CN ratio occurs. The PBL top height is determined at the altitude where there is an inversion or abrupt large change in dew point [Wilczak *et al.*, 1996; Quan *et al.*, 2013], and is calculated from the aircraft measurements of temperature and relative humidity.

The LA results are consistent with the previous studies [Väänänen *et al.*, 2016; Wehner *et*

al., 2007; O'Dowd et al., 2009; Laakso et al., 2007; Siebert et al., 2004; Stratmann et al., 2003; Pryor et al., 2011; Crumeyrolle et al., 2010; Dunne et al., 2016]; however, the HA results are new and worthy of further investigation. Figure 2 shows the dependence on the aerosol surface area concentration of PBL top height (a), peak NPF height (b), and the difference between the two heights (c). The results not only confirm Fig. 1, but also further suggest a clear functional dependence of NPF occurrence on aerosol loading. The position of NPF occurrence relative to PBL top results from the combined effects of decreased PBL (Fig.2a) and increased NPF height (Fig.2b) as aerosol loading increases.

The relationships of peak UCN height (H_{UCN}), determined from the vertical profile of UCN as the height where the maximum UCN occurs, with PBL top height and aerosol concentration exhibit similar behaviors (Fig.S3). The vertical distributions of UCN and CN concentrations provide more detailed information (Figure 3). Under LA, high UCN ($>2000 \text{ cm}^{-3}$) concentration generally appears at 0-1.5 km (inside PBL), with the peak UCN at 0.6 km. Under HA, UCN first increases with altitude and reaches its peak at 2.1 km (above PBL), then decrease with altitude. The maximum UCN-CN ratios appear at 1.2 km and 2.1km for LA and HA, respectively, which is consistent with the vertical NPF frequency (Fig.1c). The height of minimum median diameter (D_m) is also consistent with the peak UCN height, further supporting that NPF occurred in LT, rather than PBL under HA. Under HA, D_m in LT (60 nm) is much smaller than in PBL (84 nm) whereas D_m in LT (47nm) is slightly larger than that in PBL (42 nm) under LA. These results collectively suggest that increased aerosol loading suppresses the NPF occurrence near ground and in PBL, and delays the occurrence of NPF peak to a higher altitude.

A key process that determines NPF occurrence is the conversion of gaseous SO_2 molecules into sulfate particles [Kulmala et al., 2004; Kulmala et al., 2001]. To understand the role of aerosols on photochemical capacity and hence SO_2 -to-sulfate conversion, the concentration of OH ($[\text{OH}]$), a key atmospheric oxidant, and the sulfuric acid concentration ($[\text{H}_2\text{SO}_4]$) under two aerosol conditions (HA, LA) are calculated. The detailed methodology can be found in SI [Madronich, 1989; Nenes et al., 1998; Li et al., 2011; Petaja et al., 2009; Tie et al., 2005]. Evidently, $[\text{OH}]$ drops considerably under HA (Fig.4a). For example, the averaged $[\text{OH}]$ is $2.3 \times 10^6 \text{ molec cm}^{-3}$ in height of 0-1 km under LA, and decreased to $1.2 \times 10^6 \text{ molec cm}^{-3}$ under HA. The vertical profile of $[\text{OH}]$ also indicates that its concentration in LT (higher than 0.96 km in HA and 2.35 km in LA) is much higher than in PBL. Therefore, the decreased $[\text{OH}]$ under HA decreases the SO_2 -to-sulfate conversion, especially at surface and in PBL where aerosols are mainly located. The suppressed photochemical reaction and thus SO_2 -to-sulfate conversion in PBL permit more SO_2 molecules to be transported to LT. The calculated $[\text{H}_2\text{SO}_4]$ profile substantiates these analyses (Fig.4b): its value is suppressed under HA at surface and in PBL, and increases with altitude, reaching its peak at a height of 1.2-2.4 km. The balance may enhance NPF events in LA in view of the crucial role of SO_2 -to-sulfate conversion in

NPF [Kulmala *et al.*, 2004; Kulmala *et al.*, 2001]. The above analysis is further corroborated by the vertical distributions of SO₂ and CO under HA and LA (Fig.S4). The SO₂ concentration between 0-1km height increases from 3.5 ppb under LA to 14.1 ppb under HA, with an increase rate of 300%, much higher than the CO increase (from 1.7 to 2.2 ppm, with an increase rate of 27%). According to Parrington *et al.* [2013] and Vakkari *et al.* [2014], the difference ratio, $\Delta\text{SO}_2/\Delta\text{CO}$, can be used as an indicator of the intensity of SO₂-to-sulfate conversion; a lower $\Delta\text{SO}_2/\Delta\text{CO}$ indicates a higher photochemical activity and relatively more gaseous SO₂ is transferred to aerosol particles. The difference Δ is defined as the local measurement minus the corresponding background value represented by the averaged value in free troposphere under LA (i.e., SO₂ = 0.72 ppb and CO = 1.55 ppm). This is confirmed by our results, which indicates that the mean $\Delta\text{SO}_2/\Delta\text{CO}$ between 0-1 km heights was 14.7 ppb/ppm under LA, and increased to 20.1 ppb/ppm under HA. These results suggest that increased aerosols suppress the SO₂-to-sulfate conversion especially at surface and in PBL, and that more SO₂ molecules are transported to the free troposphere. It is noteworthy that the SO₂ concentration of 3-5 ppb above 2 km altitudes is even higher than those observed at ground in Europe and US [Stehr *et al.*, 2000; Denby *et al.*, 2010].

In addition to its effects on photochemical capacity and SO₂-to-sulfate conversion, increased aerosols can also affect NPF by altering sinks from condensation (CS) and coagulation (CoagS) [Maso *et al.*, 2002; Kulmala *et al.*, 2000]. The detailed methodology can be found in SI [Kulmala *et al.*, 2001; Fuchs and Sutugin, 1971; Fuchs, 1964; Allen and Raabe, 1985]. The profiles of calculated CS (Fig.4c) and CoagS (Fig.S5) illustrate that both increase sharply under HA, especially inside PBL. For example, the mean CS is $9.2 \times 10^{-2} \text{ s}^{-1}$ at a height of 0-1 km, and sharply decreases to $1.5 \times 10^{-2} \text{ s}^{-1}$ at a height of 2-3.6 km under HA, while under LA, the mean CS is only $1.7 \times 10^{-2} \text{ s}^{-1}$ and $0.7 \times 10^{-2} \text{ s}^{-1}$. The concurrent increases of CS and CoagS with increasing aerosol loading further reinforce the role of high aerosols in suppressing NPF in PBL through enhancing NPF sinks. McMurry and Friedlander [1979] proposed a dimensionless parameter L that describes the ratio between scavenging of newly formed particles by the pre-existing aerosol to the growth rate of newly formed particles. Kuang *et al.* [2010] revised this parameter L_{Γ} and identified that NPF is suppressed when L_{Γ} is beyond a threshold (0.7). The detailed methodology can be found in SI [McCurry *et al.*, 2005; Kuang *et al.*, 2010]. Our calculation shows that the mean L_{Γ} between 0-1 km heights was 0.15 under LA, and increased to 1.57 under HA (Fig.4d), suggesting that NPF in the low layer (0-1 km) is significantly suppressed under high aerosols. The suppressed NPF under high aerosols is caused by increased condensation sink of pre-existing aerosols and decreased atmospheric oxidation. The former appears to play a more important role. For example, the CS between 0-1 km heights increases from $1.7 \times 10^{-3} \text{ s}^{-1}$ under LA to $9.2 \times 10^{-3} \text{ s}^{-1}$ under HA, with an increase rate of 450%, about 11 times higher than the H₂SO₄ decrease (from $8.5 \times 10^6 \text{ moles cm}^{-3}$ to $5.2 \times 10^6 \text{ moles cm}^{-3}$, with a decrease rate of 40%). Dependence on

the ground L_T of PBL top height, peak NPF height, and the difference between the two heights is further analyzed (Fig.2d-2f) to understand the complex relationships of aerosol, PBL and NPF. Under HA, ground NPF is suppressed (Fig.2d), and the height NPF occurrence is elevated (Fig.2e). Hence, NPF tends to occur in FT rather than in PBL under HA, which is clearly different with LA (Fig.2f). It is noteworthy that comparative analysis of key meteorological variables (temperature, moisture, and relative humidity) further supports that the atmosphere under HA is more stable and favorable for pollutants and water vapor accumulation (Fig.S6).

In summary, our collective analyses show that high concentration of aerosols (mainly located in PBL) decreases the NPF occurrence in PBL by simultaneously reducing the source (represented by $[H_2SO_4]$) and increasing their sink (represented by CS and CoagS) of new particles. Atmospheric oxidation is lowered by high aerosols, which suppresses the SO_2 -to-sulfate conversion in PBL and enables more SO_2 transported to the free troposphere. The combined higher SO_2 and higher oxidation rate in the free atmosphere due to lower aerosol concentration enhance NPF in the free atmosphere, leading to a unique vertical distribution of NPF occurrence that peaks in LT instead of PBL, as most previous studies have found. These findings shed new light on our understanding of NPF in highly polluted environments. The dramatic differences in NPF occurrence and vertical distribution suggest potentially different NPF effects on clouds, precipitation, and thus climate in highly polluted environments. It should be noted that the freshly formed particles (a few nanometers) were not directly measured in this work. Although similar approaches have been successfully used to identify NPF events by others [Venzac *et al.*, 2008; Wang *et al.*, 2016], a direct measurement of such particles is desirable. Also noteworthy is that this study is based on the data collected in Beijing, and whether the findings can be generalized to other megacities warrants further investigation.

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343

Figures

Fig.1. Vertical distribution of the NPF events during the whole campaign for LA (a, green) and HA (b, red), respectively. The corresponding PBL top heights (black dot) are also shown for reference, the mean PBL top heights are 0.96 km and 2.35 km under HA and LA respectively; (c) vertical profile of NPF frequency under LA and HA; and (d) comparison of peak NPF height (H_{NPF}) and PBL top height (H_{PBL}) under LA and HA. All these altitudes are above sea level, the same as below.

Fig.2. Dependence on the aerosol surface area concentration (SA) at ground of (a) PBL top height, (b) peak NPF height, and (c) the difference between the two heights. Dependence on the dimensionless parameter L_T at ground of (a) PBL top height, (b) peak NPF height, and (c) the difference between the two heights. The green and red dots represent data under LA and HA, respectively.

Fig.3. Vertical profiles of (a) total condensation nuclei (CN) concentration, (b) ultrafine condensation nuclei (UCN) (diameter ≤ 25 nm) concentration, (c) UCN-CN ratio, and (d) effective diameter (D_m) under HA (red) and LA (green) in cases of NPF events.

Fig.4. Vertical profiles of (a) derived OH concentration, (b) H₂SO₄ concentration, (c) condensation sink (CS), and (d) dimensionless parameter L_T under HA (red) and LA (green).

Figure 1.

Figure 2.

Figure 3.

Figure 4.