Anthropogenic dust as a significant source of ice-nucleating particles in the urban environment

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Abstract

Anthropogenic dust is an important constituent of airborne particles in the urban environment but its ice nucleation activity remains uninvestigated. Here, we studied the sources and ice nucleating properties of size-resolved particles in the urban atmosphere under mixed-phase cloud conditions. The heat-resistant ice nucleating particles (INPs) unexpectedly contributed ~70% of the INPs in coarse mode at temperatures below -15 oC. Detailed size-resolved particle chemical composition analysis showed that these INPs were contributed by anthropogenic dust, such as traffic-influenced road dust. A parameterization based on coarse particles was developed to predict the anthropogenic dust INP concentration, due to their correlations on concentration and similarity in chemical compositions. The parameterization can be used for further evaluating the anthropogenic dust contribution to INPs on a global scale. We suggest anthropogenic dust associated with rapid urbanization will become an important factor for urban climate change by altering the cloud microphysics.

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Key Points:

- The coarse ice nucleating particles (INPs) contribute ~95.2% of the total INPs in the urban atmosphere.
- Anthropogenic dust such as traffic-influenced road dust proved to be a major source of heat-resistant INPs in the urban environment.
- Coarse particles are strongly correlated to anthropogenic dust INPs and can be used to predict their concentration.

Abstract

Anthropogenic dust is an important constituent of airborne particles in the urban environment but its ice nucleation activity remains uninvestigated. Here, we studied the sources and ice nucleating properties of size-resolved particles in the urban atmosphere under mixed-phase cloud conditions. The heat-resistant ice nucleating particles (INPs) unexpectedly contributed ~70% of the INPs in coarse mode at temperatures below −15 °C. Detailed size-resolved particle chemical composition analysis showed that these INPs were contributed by anthropogenic dust, such as traffic-influenced road dust. A parameterization based on coarse particles was developed to predict the anthropogenic dust INP concentration, due to their correlations on concentration and similarity in chemical compositions. The parameterization can be used for further evaluating the anthropogenic dust contribution to INPs on a global scale. We suggest anthropogenic dust associated with rapid urbanization will become an important factor for urban climate change by altering the cloud microphysics.

Plain Language Summary

Anthropogenic dust (dust particles generated by human activities) prevails in the urban atmosphere, but its ability to nucleate ice is not well understood. Combing the chemical composition analysis and ice-nucleating particle measurements of urban aerosols, we found anthropogenic dust, such as traffic-influenced road dust, is an important source of INPs in the urban atmosphere. On basis of the number concentration of coarse particles and INPs, a new parameterization is developed to predict the INP concentration contributed by anthropogenic dust. We propose that anthropogenic dust will become more important for the future urban climate by serving as INPs associated with the increasing anthropogenic dust loading caused by rapid urbanization.

1 Introduction

The heterogeneous ice nucleation process aided by ice nucleating particles (INPs) are important for ice crystal formation in clouds and is still an unsolved issue in aerosol-cloud interaction [*Fan et al.*, 2016]. The INP concentrations and their ice nucleation activities in the atmosphere would regulate the microphysics and radiative properties of clouds by influencing the number concentration and size of ice crystals, as a result, indirectly influencing the global climate (IPCC report 2021) [*Legg*, 2021]. A fundamental understanding of the abundance and sources of atmospheric INPs is still not achieved due to the rarity and complexity of INPs [*Kanji et al.*, 2017]. In the urban atmosphere, aerosols originate from both natural and anthropogenic emissions and undergo a number of aging and transformation processes. Such complexity adds to the difficulties to identify the major sources of INPs and quantify the respective contributions. This can be a missing piece for the cloud and climate model to predict the aerosol-ice cloud interaction in urban regions and its following impact on the urban-climate system.

The ice nucleation activities of the bulk urban aerosol and its constituents have been studied under mixed-phase cloud conditions [*Bi et al.*, 2019; *Chen et al.*, 2018a; *Chen et al.*, 2021b; *Hasenkopf et al.*, 2016; *Pereira et al.*, 2021; *Yadav et al.*, 2019; *Zhang et al.*, 2022]. The biological INPs prevail in the urban atmosphere and are considered efficient INPs at temperatures above −15 °C [*Pereira et al.*, 2021; *Yadav et al.*, 2019]. The INP concentrations can be extremely enhanced when the urban atmosphere is influenced by the long-range transport dust plume originating from desert regions [*Bi et al.*, 2019; *Chen et al.*, 2021a]. A recent study by *Tian et al.* [2022] shows the contribution of organic aerosols (heat-resistant at 350 °C) to immersion-freezing INPs at −30 °C in the urban region. However, this study was unable to exclude or evaluate the impact of local dust emissions on these refractory INPs, which is a big contributor to aerosols in urban regions [*Han et al.*, 2005]. The poor ice nucleation performance of black carbon (BC) or soot particles from fossil fuel combustion under mixed-

phase cloud conditions has been verified by both laboratory studies [*Kanji et al.*, 2020; *Vergara-Temprado et al.*, 2018] and field measurements [*Chen et al.*, 2018; *Zhang et al.*, 2022]. The INP concentrations did not show an obvious increase or decrease during heavily polluted periods in Beijing, indicating the secondary aerosols formed from the increasingly emitted anthropogenic precursors were not a major source of the observed INP concentrations [*Bi et al.*, 2019; *Chen et al.*, 2018; *Zhang et al.*, 2022]. This conclusion is also confirmed by many field studies on a global scale either conducted in highly polluted regions [*Hasenkopf et al.*, 2016; *Pereira et al.*, 2021; *Yadav et al.*, 2019] or in regions occasionally influenced by air pollution [*Creamean et al.*, 2018; *Wex et al.*, 2015], where a negligible effect on INP production from the non-dust air pollution was observed.

These studies imply that the INPs in the urban atmosphere are unlikely contributed by anthropogenic pollutants under mixed-phase cloud conditions as far as dust air pollution is not considered. Instead, the natural biological particles and the long-range transported desert dust are still considered common INP sources and there may exist unidentified INP sources in non-dust days. However, note that the atmospheric dust loading can also be changed anthropogenically due to land use changes by human activity, the so-called anthropogenic dust (as defined in *Zender et al.* [2004]). Anthropogenic dust accounts for ~30 to 70% of total dust concentrations in urban in recent decades [*Chen et al.*, 2018; *Huang et al.*, 2015]. On a global scale, anthropogenic contribution to atmospheric dust loads today is between~90 and 2000 Mt. $year^{-1}$ [*Webb and Pierre*, 2018]. It is an important constituent of airborne particles in the urban atmosphere and can be emitted by construction works, traffic-generated turbulence, and agricultural and industrial activities [*Haynes et al.*, 2020; *Philip et al.*, 2017]. The representative anthropogenic dust includes traffic-influenced road dust [*Xia et al.*, 2022] and soil particles from disturbed soil [*Wang et al.*, 2018].

Despite the high mass loading of anthropogenic dust in the urban atmosphere and its important direct and indirect effects on the urban climate system [*Philip et al.*, 2017; *Xia et al.*, 2022], limited studies have investigated the ice nucleation properties of this dust species compared to those of other pollutants. Many studies investigated the ice nucleation activities of ground-based collected soil dust. They showed that soil dust can serve as INPs in a wide temperature range (−35 to −6 ºC) [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Pereira et al.*, 2022; *Steinke et al.*, 2016; *Tobo et al.*, 2014] and its ice nucleation activity is constrained by the containing biological compounds [*Conen et al.*, 2011] and the organic matters (OM) [*Pereira et al.*, 2022; *Tobo et al.*, 2014]. The ice nucleation activity of anthropogenic soil dust from the emission of disturbed soils is less studied. Studies pointed out that the atmospheric INPs in South America can come from airborne agricultural dust [*Gong et al.*, 2022; *Testa et al.*, 2021]. *Corbin et al.* [2012] showed dust particles were riched in the ice residues activated by INPs (at a temperature of -34 ºC and relative humidity of 95% with respect to water) in Toronto. The major source of the detected dust is likely vehicular resuspension from nearby roads, implying the potential contribution of road dust to INPs [*Corbin et al.*, 2012].

In the present study, the size-resolved ambient particles were collected in the urban atmosphere when no impacts of natural desert dust were observed. The ice nucleation activities and the chemical compositions of the collected particles were investigated and evidence that anthropogenic dust makes a significant contribution to INPs in the urban environment. Regarding the continuous rapid increase of urbanization in the future, we suggest that anthropogenic dust emissions due to off-road vehicles and urban construction may play an important role in affecting the global INP budget.

2 Materials and Methods

2.1 Aerosol Sampling and Characterization

The aerosol samples were collected at Peking University Urban Atmospheric Environment Monitoring Station (PKUERS) in the summer from June 22 to July 21, 2020. PKUERS is located on the campus of Peking University and is 20 meters above the ground. This site is a representative urban site affected by multiple anthropogenic emissions, such as transport emissions and fossil fuel combustion [*Chen et al.*, 2018a; *Zhang et al.*, 2022].

Aerosols were collected onto the 47 mm diameter polycarbonate filters (Whatman, 111107) by a Micro-Orifice Uniform Deposit Impactor (MOUDI, MSP Corporation, USA) with a flow rate of 30 L min $^{-1}$. MOUDI allows the particles to be classified and collected with different aerodynamic diameters (AD). Each sample set (i.e., each set including filter samples collected particles with different cut-off sizes) was collected for 24 hours, and the detailed sampling information was listed in Table S1. Particles with the cut-off size of 0.56 μm, 1.0 μm, 1.8 μm, 3.2 μm, and 5.6 μm were collected for each set, an aerosol population that is of interest in the INP parameterizations. All reported sizes in the present study are the 50% cut-off AD (D_{50}), which corresponds to the AD of particles trapped with an efficiency of 50% at a given stage. In total, 8 sets of filter samples were collected.

An aerodynamic particle sizer (APS, model 3021, TSI) measured the number concentration of particles with AD ranging from 0.542 μm to 19.81 μm ($N_{>*}$ is defined as the number concentration of particles larger than * μm, $N_*$is the number concentration of particle with the size of * μm). Note that APS and MOUDI both detect the particle aerodynamic diameter. Elements of samples include Na, Mg, Al, K, Ca, Mn, Fe, Zn, As, Ba, Pb, and others (V, Co, Se, Sr, Mo, Tl, Bi, Th, U, Cd, Ni, Cu, Ti, Cr, and P) were measured by an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS, Bruker, aurora M90). The mass concentrations of the organic carbon (OC) and element carbon (EC) were measured by the Sunset ECOC analyzer (Sunset Lab 4) using quartz filters collected parallelly by the aerosol sampler with $PM_{2.5}$ cut-off size.

2.2 Ice Nucleation Measurement

The ice nucleation measurement was performed by the Peking University Ice Nucleation Array (PKU-INA), which is a cold-stage-based device to measure the freezing ability of droplets under mixed-phase cloud conditions [*Chen et al.*, 2018b]. In this study, each filter sample was first immersed in distilled water of 7 mL and shaken by the vortex for 40 minutes to wash particles off. The resulting suspension was then pipetted onto the cold stage to form 90 droplets with a volume of 1 μL. Droplets were separated by a spacer and then sealed by a cover glass to avoid the Wegener-Bergeron-Findeisen process. Droplets were cooled down to −32 °C with a cooling rate of 1 °C/min. Meanwhile, the status of all the droplets was recorded every 6 seconds by a high-speed camera (Q-imaging MicroPublisher 5.0 RTV, QImaging, Surrey, BC, Canada) mounted on the top of the cold stage. The obtained images were then input into the developed MATLAB program to identify the freezing temperatures of the droplets according to the brightness change of each droplet upon its phase transition.

The frozen fraction (f_{ice}) of the droplets at each temperature can be obtained using Eq. (1) by assuming a time-independent ice nucleation process of droplets:

$$f_{ice}(\mathrm{T}) = \frac{N_f}{N_t} \quad (1)$$

where N_f is the number of frozen droplets at a given temperature and N_{t} is the total number of droplets (i.e., 90 in this study).

The number concentration of INP (N_{INP}) per unit volume of sampled air collected on each sample filter (i.e., at each cut-off size) is calculated based on the f_{ice} and the total volume of the sampled air that was collected into each droplet (V_{air}) using Eq. (2):

$$N_{\mathrm{INP}}(\mathrm{T}) = \frac{-\ln\left(1 - f_{ice}(\mathrm{T})\right)}{V_{air}} \quad (2)$$

and the total number concentration of INPs each day was derived by integrating the INP number concentration with different cut-off sizes.

The cumulative number concentration of the ice active sites per unit surface area of INPs (n_s), as derived in many studies to describe the ice nucleation ability of particles [*Connolly et al.*, 2009; *Hiranuma et al.*, 2015; *Niemand et al.*, 2012], is calculated according to Eq. (3):

$$n_s(\mathrm{T}) = \frac{N_{\mathrm{INP}}(\mathrm{T})}{A} \quad (3)$$

For particles collected at each cut-off size, A ($A = \frac{\pi N_* {D_p}^2}{4}$) is the total surface area of the particles per unit volume of air sampled at each stage. A is calculated based on the number concentration of particles per unit volume of air with different aerodynamic sizes (D_p) measured by APS and assuming the particles are spherical.

3 Results and Discussion

3.1 Overview of the INP Concentration

Figure 1(a) shows the total number concentration of INPs (N_{INP}) (the sum of size-resolved N_{INP}) detected on different days as a function of temperatures. The temperature dependencies of total N_{INP} are similar and the variations of N_{INP} are less than one magnitude from −21 °C to −5 °C. This implies the INPs on different days may originate from similar aerosol sources and no specific strong sources of INPs presented on one particular day during the sampling period.

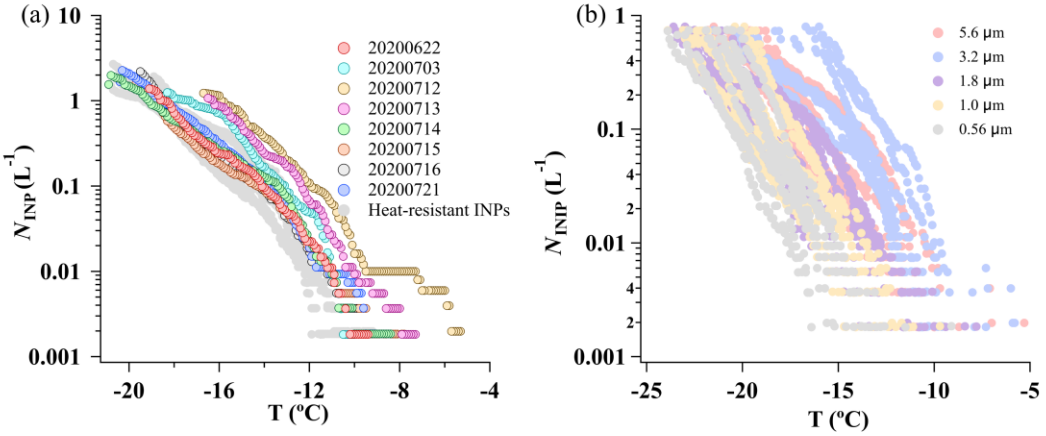

Figure 1. The total (a) and size-dependent (b) number concentration of INPs (N_{INP}) as a function of temperatures; The gray circles in (a) represent the number concentration of heat-resistant INPs after being heated at 95 °C for 20 mins. The cut-off size in (b) corresponds to the aerodynamic diameter of particles trapped with an efficiency of 50% at a given stage.

The size-dependent N_{INP} (with D_{50}=0.56 μm, 1 μm, 1.8 μm, 3.2 μm, and D_{50}=5.6 μm) is shown in Fig. 1(b). Each line represents the N_{INP} with one cut-off size collected in one day. On average, the N_{INP} in size larger than 1 μm explained 95.2%±4% of the total N_{INP} over the

temperature range from −20 ºC to −10 ºC, meaning that coarse INPs dominate the INP concentration in the urban atmosphere of Beijing. The prevalence and importance of the coarse INPs are pointed out by other studies conducted in the global atmosphere as well [*Mason et al.*, 2016; *Gong et al.*, 2020; *Mitts et al.*, 2021]. In the urban atmosphere, the coarse particles can originate from dust particles, tire debris, and biological particles (see *Wu and Boor* [2021] and the references therein). The concentration of coarse particles has a strong correlation with the INP concentrations detected in the urban environment, thus can be an important source of INPs [*Che et al.*, 2021; *Chen et al.*, 2021; *Jiang et al.*, 2023]. The sources of the detected coarse particles and INPs will be discussed in the following sections.

3.2 The Contribution of Anthropogenic Dust to INPs

The extracted sample solutions were heated to 95 ºC for 20 mins. The total N_{INP} decreased after heat treatment (Fig. 1), indicating that proteinaceous biological INPs contributed to the observed INPs (refer to [*Christner et al.*, 2008]). Fig. 2 furtherly shows the percentage of heat-resistant N_{INP} (the ratio of N_{INP} measured after and before heat treatment) at different temperatures. The contribution of heat-sensitive proteinaceous biological particles to INPs only becomes important at temperatures above −14.3 ºC (~50%). The percentage of heat-resistant N_{INP} increases with decreasing temperatures. At temperatures below −15 ºC, the heat-resistant INPs account for ~70% of the total INPs.

The heat-resistant INPs can originate from dust particles (refer to [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Perkins et al.*, 2019]) and some unidentified organic matter (OM) [*McCluskey et al.*, 2018; *Tian et al.*, 2022]. The impacts of soot particles and other inorganic components (inorganic salts) on the heat-resistant INPs are excluded due to their poor ice nucleation activities under mixed-phase cloud conditions [*Chen et al.*, 2018a; *Kanji et al.*, 2020; *Zhang et al.*, 2022].

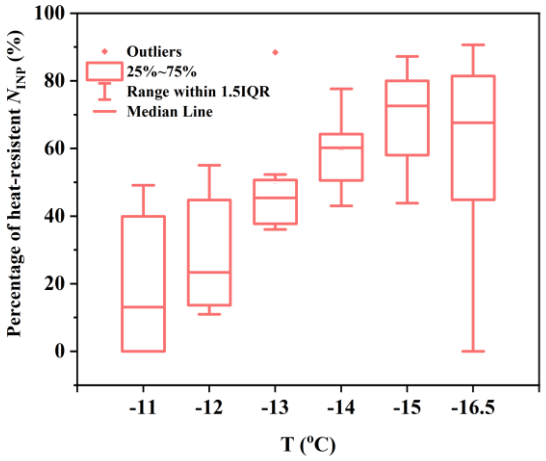

Figure 2. The percentages of heat-resistant N_{INP} vs. temperatures; only values at temperatures above −16.5 ºC were calculated where droplets in all samples did not finish frozen.

The source of aerosols and heat-resistant INPs was explored based on the chemical analysis of collected particles. The contribution of heat-resistant INPs from OM is considered to be minor under the determined conditions, as a poor correlation between the detected heat-resistant INPs and organic carbon was found (R^2=0.19, Fig. S1). The chemical elements of the coarse particles (D_{50}=3.2 μm) (Fig. 3 and Table S3) show that crustal elements including Ca (50.46%±2.27%), Mg (10.98%±0.82%), Fe (14.89%±1.48%) and Al (10.42%±1.45%) account for a major mass percentage (88.64%±2.92%) of the total element mass of particles, demonstrating the strong contribution from dust particles. Differently, the crustal elements in

fine particles (D_{50}= 0.56 μm) are much less, while a large mass percentage of non-dust related elements (Zn, Pb, K, and Na) are found (48.53%±10.61%) (Fig. 3 and Table S3). This indicates that dust particles contribute to heat-resistant INPs in the coarse particles, while fine particles may be influenced by non-dust anthropogenic sources (see detailed discussion in Fig. S2 and Text S1). This conclusion is also supported by the different ice nucleation activities (presented as n_s, calculated based on Eq.(3)) obtained for coarse and fine INPs (Fig. S3), which comes from different aerosol sources.

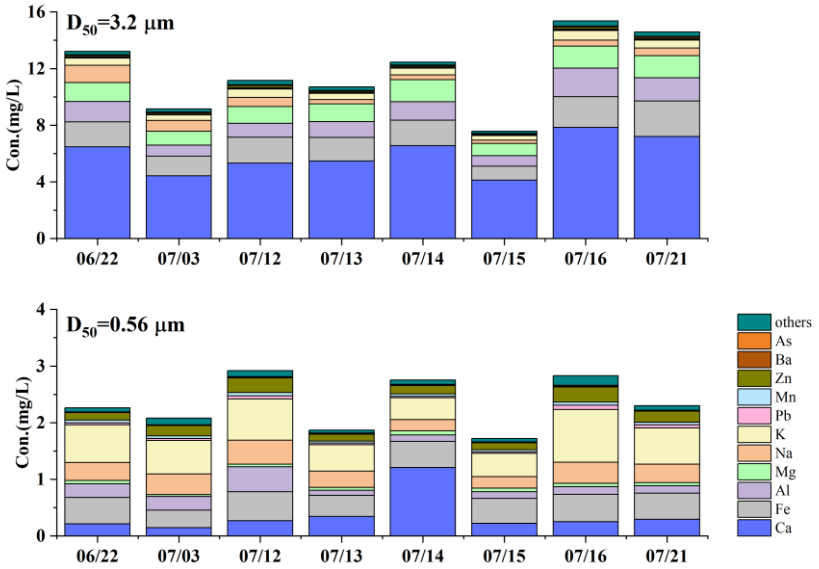

Figure 3. The element compositions of particles in different sizes.

Dust particles in the urban atmosphere can either be long-range transported desert dust or anthropogenic dust [*Han et al.*, 2005; *Zender et al.*, 2004]. Here, we confirmed natural desert dust has negligible impact on the heat-resistant INPs while anthropogenic dust is a major source, as supported by the following evidence. No obvious enhancement in the number concentration of coarse particles and in n_s has been observed during the sampling time (Fig. S4(a)), which typically occurred during Asian dust storm events (Fig. S4(b)) [*Chen et al.*, 2021a]. The n_s obtained in this study ($3*10^3$ to $2*10^5$ m^{-2} at -15 °C) were two magnitudes lower than those obtained during the spring Asian dust events (10^5 to 10^7 m^{-2} at -15 °C) at the same site [*Chen et al.*, 2021a]. The analysis of 72-hours backward trajectories (calculated every 6 hours at 1:00, 7:00, 13:00, and 19:00 UTC at the height of 500 m) (Fig. S5) shows that air masses did not pass over the desert regions. Asian dust events occur especially during spring months and not in the summer of Beijing [*Shao and Dong*, 2006], while the anthropogenic dust from local emissions can contribute ~80% of the total dust [*Han et al.*, 2005].

The impact of one representative anthropogenic dust species on the collected aerosols and INPs, the traffic-influenced road dust, is proved by the good correlation between the hour-mean concentration of the coarse particles and nitric oxide (NO) during the sampling time (R^2=0.53, Fig. S6). Coarse particles and NO are indicators of the dust particles and vehicle primary emissions, respectively. Good correlations between heat-resistant N_{INP} at −16 °C and the mass percentage of Ba and Zn were observed in coarse particles (R^2=0.39 and R^2=0.60, Fig. S7). Ba and Zn are two tracers for the road dust particles [*Gietl et al.*, 2010; *Harrison and Alghamdi*, 2023; *Peltier et al.*, 2011], which proves the contribution of road dust to heat-resistant INPs again. Other anthropogenic dust species, such as those generated from agricultural and construction activities may contribute to the collected coarse particles as well but cannot be validated in the present study due to lacking reliable tracers. Since the dust generated from construction is commonly found in urban regions [*Azarov et al.*, 2019; *Yan et al.*, 2020], we expect its contribution to the observed anthropogenic dust INPs. The impact of

soil dust from agricultural activities is hard to be evaluated here. However, the n_s values of the obtained heat-resistant INPs are 3 orders of magnitudes lower than those from soil dust (original or H_2O_2 treated soil dust) measured by *Tobo et al.* [2014] (Fig. S8) and than the n_s of inorganic INPs measured by *Testa et al.* [2021] which was presumably from land surface emission. This comparison implies that the ice nucleation activity of the airborne anthropogenic dust cannot be explained alone by the collected ground-based and near-source soil dust.

3.3 Parameterization of Anthropogenic Dust INPs

Based on the aforementioned results, the coarse heat-resistant N_{INP} can be referred to as anthropogenic dust N_{INP} in the urban atmosphere. The anthropogenic N_{INP} is highly correlated to the number concentration of coarse particles ($N_{>1\ \mu m}$, R^2=0.67, Table S2), which can be foreseen from the strong impact of dust on the coarse particles (section 3.2). Thus, $N_{>1\ \mu m}$ is used to predict the concentration of anthropogenic dust INPs. For comparison, the correlations between the anthropogenic N_{INP} and the number concentration of particles with other size ranges ($N_{>500\ nm}$ and $N_{>1.8\ \mu m}$) are listed in Table S2.

Table 1 The parameterizations to predict the anthropogenic dust INPs

$$N_{\mathrm{INP}} = (N_{>1\ \mu m})^{(\alpha(-T)+\beta)} * \exp(\gamma(-T) + \delta)$$

	α	β	γ	δ	R^2
Anthropogenic dust INPs	$-1.835*10^{-5}$	0.0294	0.652	-12.518	0.927

The parametrization to predict the anthropogenic dust INPs at a temperature range from $-21^oC \sim -7^oC$ was developed based on $N_{>1\ \mu m}$ following the form of *DeMott et al.* [2015] parameterization (hereafter D15) (listed in Table 1). The coefficient of determination (R^2) of the resulting parameterization is 0.95 with 95% confidence. The comparison between the results from observation and prediction was shown in Fig. S9. The ratios of the observed to the predicted values are within a factor of 3, indicating a good prediction from the parameterization. To be compared, the heat-resistant N_{INP} was estimated using D15 as well, which was developed based on $N_{>500\ nm}$ (Fig. S10). An overestimation of heat-resistant/total INPs about 1~2 magnitudes was observed, indicating poor prediction. This is because the particles larger than 500 nm cannot represent the exact size range of the measured INP species, the anthropogenic dust INPs here (evidenced by the different chemical compositions and ice nucleation activities of coarse and fine particles). The overestimation of N_{INP} in the urban environment by D15 was confirmed by *Chen et al.* [2018a] and *Bi et al.* [2019] as well.

Overall, the parameterization developed based on $N_{>1\ \mu m}$ showed a good performance on the prediction of the anthropogenic dust INPs. This parameterization can then be used in regional or climate models to predict the INPs contributed by anthropogenic dust and evaluate its further impact on cloud formation and the urban climate. Note that using heat-resistant N_{INP} as the proxy of anthropogenic dust N_{INP} in the urban atmosphere can cause a bias. This assumption cannot exclude that some of the measured heat-resistant INPs are from the heat-resistant OM [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Tobo et al.*, 2014], although a poor correlation between OM and heat-resistant INPs and significant dust influence has been found here. On the other hand, some dust species can significantly lose their ice nucleation activity when undergoing the heating process [*Daily et al.*, 2020], indicating the possibility of underestimating the anthropogenic dust N_{INP} based on the heat-lability of particles. These uncertainties might influence the quantification of anthropogenic dust INPs and need more investigation on measuring the contribution of anthropogenic dust to aerosols and atmospheric INPs in the urban environment.

4 Conclusions

The abundance and sources of INPs remain unclear in the urban atmosphere. In the present study, the ice nucleation activities of size-resolved particles were investigated in a typical urban environment. INPs in coarse-mode account for 95.2%±4% of the detected N_{INP} and are significantly contributed by heat-resistant INPs (~70%) at temperatures below −15 ºC. The further size-resolved chemical composition analysis indicates that anthropogenic dust such as traffic-influenced road dust is a major source of these heat-resistant INPs. The number concentration of coarse particles is strongly correlated with those of the anthropogenic dust INPs, thus was used to develop the parameterization for predicting anthropogenic dust INP concentrations. The new parameterization predicts the observed N_{INP} within a factor of 3, showing a good performance. This study highlighted the importance of anthropogenic dust particles as a significant source of INPs in urban environments, which has not been extensively studied before.

Projections of past and future atmospheric dust loads suggest that the contribution of anthropogenic emissions is increased in the last decades and it will continue to have a profound role in the Earth's future climate [*Mahowald and Luo*, 2003; *Stanelle et al.*, 2014]. Due to the good ice nucleation activity of the anthropogenic dust and its large contributions to the atmospheric INPs reported by the present study, we suggest that anthropogenic dust will become an important factor to connect with urban climate change by modifying INP concentrations and cloud microphysics.

Acknowledgments

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Conflict of interest

The authors declare no conflicts of interest relevant to this study.

Open Research

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Anthropogenic dust as a significant source of ice-nucleating particles in the urban environment

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Key Points:

- The coarse ice nucleating particles (INPs) contribute ~95.2% of the total INPs in the urban atmosphere.
- Anthropogenic dust such as traffic-influenced road dust proved to be a major source of heat-resistant INPs in the urban environment.
- Coarse particles are strongly correlated to anthropogenic dust INPs and can be used to predict their concentration.

Abstract

Anthropogenic dust is an important constituent of airborne particles in the urban environment but its ice nucleation activity remains uninvestigated. Here, we studied the sources and ice nucleating properties of size-resolved particles in the urban atmosphere under mixed-phase cloud conditions. The heat-resistant ice nucleating particles (INPs) unexpectedly contributed ~70% of the INPs in coarse mode at temperatures below −15 °C. Detailed size-resolved particle chemical composition analysis showed that these INPs were contributed by anthropogenic dust, such as traffic-influenced road dust. A parameterization based on coarse particles was developed to predict the anthropogenic dust INP concentration, due to their correlations on concentration and similarity in chemical compositions. The parameterization can be used for further evaluating the anthropogenic dust contribution to INPs on a global scale. We suggest anthropogenic dust associated with rapid urbanization will become an important factor for urban climate change by altering the cloud microphysics.

Plain Language Summary

Anthropogenic dust (dust particles generated by human activities) prevails in the urban atmosphere, but its ability to nucleate ice is not well understood. Combing the chemical composition analysis and ice-nucleating particle measurements of urban aerosols, we found anthropogenic dust, such as traffic-influenced road dust, is an important source of INPs in the urban atmosphere. On basis of the number concentration of coarse particles and INPs, a new parameterization is developed to predict the INP concentration contributed by anthropogenic dust. We propose that anthropogenic dust will become more important for the future urban climate by serving as INPs associated with the increasing anthropogenic dust loading caused by rapid urbanization.

1 Introduction

The heterogeneous ice nucleation process aided by ice nucleating particles (INPs) are important for ice crystal formation in clouds and is still an unsolved issue in aerosol-cloud interaction [*Fan et al.*, 2016]. The INP concentrations and their ice nucleation activities in the atmosphere would regulate the microphysics and radiative properties of clouds by influencing the number concentration and size of ice crystals, as a result, indirectly influencing the global climate (IPCC report 2021) [*Legg*, 2021]. A fundamental understanding of the abundance and sources of atmospheric INPs is still not achieved due to the rarity and complexity of INPs [*Kanji et al.*, 2017]. In the urban atmosphere, aerosols originate from both natural and anthropogenic emissions and undergo a number of aging and transformation processes. Such complexity adds to the difficulties to identify the major sources of INPs and quantify the respective contributions. This can be a missing piece for the cloud and climate model to predict the aerosol-ice cloud interaction in urban regions and its following impact on the urban-climate system.

The ice nucleation activities of the bulk urban aerosol and its constituents have been studied under mixed-phase cloud conditions [*Bi et al.*, 2019; *Chen et al.*, 2018a; *Chen et al.*, 2021b; *Hasenkopf et al.*, 2016; *Pereira et al.*, 2021; *Yadav et al.*, 2019; *Zhang et al.*, 2022]. The biological INPs prevail in the urban atmosphere and are considered efficient INPs at temperatures above −15 °C [*Pereira et al.*, 2021; *Yadav et al.*, 2019]. The INP concentrations can be extremely enhanced when the urban atmosphere is influenced by the long-range transport dust plume originating from desert regions [*Bi et al.*, 2019; *Chen et al.*, 2021a]. A recent study by *Tian et al.* [2022] shows the contribution of organic aerosols (heat-resistant at 350 °C) to immersion-freezing INPs at −30 °C in the urban region. However, this study was unable to exclude or evaluate the impact of local dust emissions on these refractory INPs, which is a big contributor to aerosols in urban regions [*Han et al.*, 2005]. The poor ice nucleation performance of black carbon (BC) or soot particles from fossil fuel combustion under mixed-

phase cloud conditions has been verified by both laboratory studies [*Kanji et al.*, 2020; *Vergara-Temprado et al.*, 2018] and field measurements [*Chen et al.*, 2018; *Zhang et al.*, 2022]. The INP concentrations did not show an obvious increase or decrease during heavily polluted periods in Beijing, indicating the secondary aerosols formed from the increasingly emitted anthropogenic precursors were not a major source of the observed INP concentrations [*Bi et al.*, 2019; *Chen et al.*, 2018; *Zhang et al.*, 2022]. This conclusion is also confirmed by many field studies on a global scale either conducted in highly polluted regions [*Hasenkopf et al.*, 2016; *Pereira et al.*, 2021; *Yadav et al.*, 2019] or in regions occasionally influenced by air pollution [*Creamean et al.*, 2018; *Wex et al.*, 2015], where a negligible effect on INP production from the non-dust air pollution was observed.

These studies imply that the INPs in the urban atmosphere are unlikely contributed by anthropogenic pollutants under mixed-phase cloud conditions as far as dust air pollution is not considered. Instead, the natural biological particles and the long-range transported desert dust are still considered common INP sources and there may exist unidentified INP sources in non-dust days. However, note that the atmospheric dust loading can also be changed anthropogenically due to land use changes by human activity, the so-called anthropogenic dust (as defined in *Zender et al.* [2004]). Anthropogenic dust accounts for ~30 to 70% of total dust concentrations in urban in recent decades [*Chen et al.*, 2018; *Huang et al.*, 2015]. On a global scale, anthropogenic contribution to atmospheric dust loads today is between~90 and 2000 Mt. $year^{-1}$ [*Webb and Pierre*, 2018]. It is an important constituent of airborne particles in the urban atmosphere and can be emitted by construction works, traffic-generated turbulence, and agricultural and industrial activities [*Haynes et al.*, 2020; *Philip et al.*, 2017]. The representative anthropogenic dust includes traffic-influenced road dust [*Xia et al.*, 2022] and soil particles from disturbed soil [*Wang et al.*, 2018].

Despite the high mass loading of anthropogenic dust in the urban atmosphere and its important direct and indirect effects on the urban climate system [*Philip et al.*, 2017; *Xia et al.*, 2022], limited studies have investigated the ice nucleation properties of this dust species compared to those of other pollutants. Many studies investigated the ice nucleation activities of ground-based collected soil dust. They showed that soil dust can serve as INPs in a wide temperature range (−35 to −6 ºC) [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Pereira et al.*, 2022; *Steinke et al.*, 2016; *Tobo et al.*, 2014] and its ice nucleation activity is constrained by the containing biological compounds [*Conen et al.*, 2011] and the organic matters (OM) [*Pereira et al.*, 2022; *Tobo et al.*, 2014]. The ice nucleation activity of anthropogenic soil dust from the emission of disturbed soils is less studied. Studies pointed out that the atmospheric INPs in South America can come from airborne agricultural dust [*Gong et al.*, 2022; *Testa et al.*, 2021]. *Corbin et al.* [2012] showed dust particles were riched in the ice residues activated by INPs (at a temperature of -34 ºC and relative humidity of 95% with respect to water) in Toronto. The major source of the detected dust is likely vehicular resuspension from nearby roads, implying the potential contribution of road dust to INPs [*Corbin et al.*, 2012].

In the present study, the size-resolved ambient particles were collected in the urban atmosphere when no impacts of natural desert dust were observed. The ice nucleation activities and the chemical compositions of the collected particles were investigated and evidence that anthropogenic dust makes a significant contribution to INPs in the urban environment. Regarding the continuous rapid increase of urbanization in the future, we suggest that anthropogenic dust emissions due to off-road vehicles and urban construction may play an important role in affecting the global INP budget.

2 Materials and Methods

2.1 Aerosol Sampling and Characterization

The aerosol samples were collected at Peking University Urban Atmospheric Environment Monitoring Station (PKUERS) in the summer from June 22 to July 21, 2020. PKUERS is located on the campus of Peking University and is 20 meters above the ground. This site is a representative urban site affected by multiple anthropogenic emissions, such as transport emissions and fossil fuel combustion [*Chen et al.*, 2018a; *Zhang et al.*, 2022].

Aerosols were collected onto the 47 mm diameter polycarbonate filters (Whatman, 111107) by a Micro-Orifice Uniform Deposit Impactor (MOUDI, MSP Corporation, USA) with a flow rate of 30 L min $^{-1}$. MOUDI allows the particles to be classified and collected with different aerodynamic diameters (AD). Each sample set (i.e., each set including filter samples collected particles with different cut-off sizes) was collected for 24 hours, and the detailed sampling information was listed in Table S1. Particles with the cut-off size of 0.56 μm, 1.0 μm, 1.8 μm, 3.2 μm, and 5.6 μm were collected for each set, an aerosol population that is of interest in the INP parameterizations. All reported sizes in the present study are the 50% cut-off AD (D_{50}), which corresponds to the AD of particles trapped with an efficiency of 50% at a given stage. In total, 8 sets of filter samples were collected.

An aerodynamic particle sizer (APS, model 3021, TSI) measured the number concentration of particles with AD ranging from 0.542 μm to 19.81 μm ($N_{>*}$ is defined as the number concentration of particles larger than * μm, $N_*$is the number concentration of particle with the size of * μm). Note that APS and MOUDI both detect the particle aerodynamic diameter. Elements of samples include Na, Mg, Al, K, Ca, Mn, Fe, Zn, As, Ba, Pb, and others (V, Co, Se, Sr, Mo, Tl, Bi, Th, U, Cd, Ni, Cu, Ti, Cr, and P) were measured by an Inductively Coupled Plasma-Mass Spectrometry (ICP-MS, Bruker, aurora M90). The mass concentrations of the organic carbon (OC) and element carbon (EC) were measured by the Sunset ECOC analyzer (Sunset Lab 4) using quartz filters collected parallelly by the aerosol sampler with $PM_{2.5}$ cut-off size.

2.2 Ice Nucleation Measurement

The ice nucleation measurement was performed by the Peking University Ice Nucleation Array (PKU-INA), which is a cold-stage-based device to measure the freezing ability of droplets under mixed-phase cloud conditions [*Chen et al.*, 2018b]. In this study, each filter sample was first immersed in distilled water of 7 mL and shaken by the vortex for 40 minutes to wash particles off. The resulting suspension was then pipetted onto the cold stage to form 90 droplets with a volume of 1 μL. Droplets were separated by a spacer and then sealed by a cover glass to avoid the Wegener-Bergeron-Findeisen process. Droplets were cooled down to −32 °C with a cooling rate of 1 °C/min. Meanwhile, the status of all the droplets was recorded every 6 seconds by a high-speed camera (Q-imaging MicroPublisher 5.0 RTV, QImaging, Surrey, BC, Canada) mounted on the top of the cold stage. The obtained images were then input into the developed MATLAB program to identify the freezing temperatures of the droplets according to the brightness change of each droplet upon its phase transition.

The frozen fraction (f_{ice}) of the droplets at each temperature can be obtained using Eq. (1) by assuming a time-independent ice nucleation process of droplets:

$$f_{ice}(\mathrm{T}) = \frac{N_f}{N_t} \quad (1)$$

where N_f is the number of frozen droplets at a given temperature and N_{t} is the total number of droplets (i.e., 90 in this study).

The number concentration of INP (N_{INP}) per unit volume of sampled air collected on each sample filter (i.e., at each cut-off size) is calculated based on the f_{ice} and the total volume of the sampled air that was collected into each droplet (V_{air}) using Eq. (2):

$$N_{\mathrm{INP}}(\mathrm{T}) = \frac{-\ln\left(1 - f_{ice}(\mathrm{T})\right)}{V_{air}} \quad (2)$$

and the total number concentration of INPs each day was derived by integrating the INP number concentration with different cut-off sizes.

The cumulative number concentration of the ice active sites per unit surface area of INPs (n_s), as derived in many studies to describe the ice nucleation ability of particles [*Connolly et al.*, 2009; *Hiranuma et al.*, 2015; *Niemand et al.*, 2012], is calculated according to Eq. (3):

$$n_s(\mathrm{T}) = \frac{N_{\mathrm{INP}}(\mathrm{T})}{A} \quad (3)$$

For particles collected at each cut-off size, A ($A = \frac{\pi N_* {D_p}^2}{4}$) is the total surface area of the particles per unit volume of air sampled at each stage. A is calculated based on the number concentration of particles per unit volume of air with different aerodynamic sizes (D_p) measured by APS and assuming the particles are spherical.

3 Results and Discussion

3.1 Overview of the INP Concentration

Figure 1(a) shows the total number concentration of INPs (N_{INP}) (the sum of size-resolved N_{INP}) detected on different days as a function of temperatures. The temperature dependencies of total N_{INP} are similar and the variations of N_{INP} are less than one magnitude from −21 °C to −5 °C. This implies the INPs on different days may originate from similar aerosol sources and no specific strong sources of INPs presented on one particular day during the sampling period.

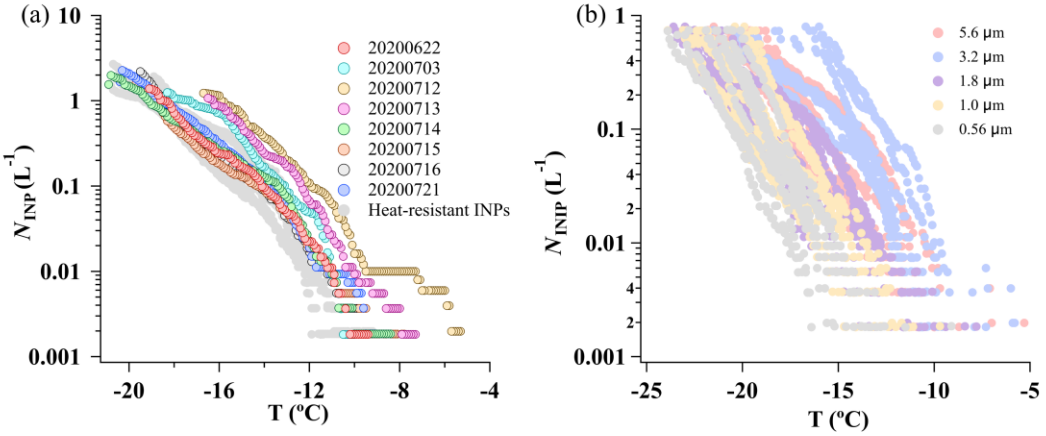

Figure 1. The total (a) and size-dependent (b) number concentration of INPs (N_{INP}) as a function of temperatures; The gray circles in (a) represent the number concentration of heat-resistant INPs after being heated at 95 °C for 20 mins. The cut-off size in (b) corresponds to the aerodynamic diameter of particles trapped with an efficiency of 50% at a given stage.

The size-dependent N_{INP} (with D_{50}=0.56 μm, 1 μm, 1.8 μm, 3.2 μm, and D_{50}=5.6 μm) is shown in Fig. 1(b). Each line represents the N_{INP} with one cut-off size collected in one day. On average, the N_{INP} in size larger than 1 μm explained 95.2%±4% of the total N_{INP} over the

temperature range from −20 ºC to −10 ºC, meaning that coarse INPs dominate the INP concentration in the urban atmosphere of Beijing. The prevalence and importance of the coarse INPs are pointed out by other studies conducted in the global atmosphere as well [*Mason et al.*, 2016; *Gong et al.*, 2020; *Mitts et al.*, 2021]. In the urban atmosphere, the coarse particles can originate from dust particles, tire debris, and biological particles (see *Wu and Boor* [2021] and the references therein). The concentration of coarse particles has a strong correlation with the INP concentrations detected in the urban environment, thus can be an important source of INPs [*Che et al.*, 2021; *Chen et al.*, 2021; *Jiang et al.*, 2023]. The sources of the detected coarse particles and INPs will be discussed in the following sections.

3.2 The Contribution of Anthropogenic Dust to INPs

The extracted sample solutions were heated to 95 ºC for 20 mins. The total N_{INP} decreased after heat treatment (Fig. 1), indicating that proteinaceous biological INPs contributed to the observed INPs (refer to [*Christner et al.*, 2008]). Fig. 2 furtherly shows the percentage of heat-resistant N_{INP} (the ratio of N_{INP} measured after and before heat treatment) at different temperatures. The contribution of heat-sensitive proteinaceous biological particles to INPs only becomes important at temperatures above −14.3 ºC (~50%). The percentage of heat-resistant N_{INP} increases with decreasing temperatures. At temperatures below −15 ºC, the heat-resistant INPs account for ~70% of the total INPs.

The heat-resistant INPs can originate from dust particles (refer to [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Perkins et al.*, 2019]) and some unidentified organic matter (OM) [*McCluskey et al.*, 2018; *Tian et al.*, 2022]. The impacts of soot particles and other inorganic components (inorganic salts) on the heat-resistant INPs are excluded due to their poor ice nucleation activities under mixed-phase cloud conditions [*Chen et al.*, 2018a; *Kanji et al.*, 2020; *Zhang et al.*, 2022].

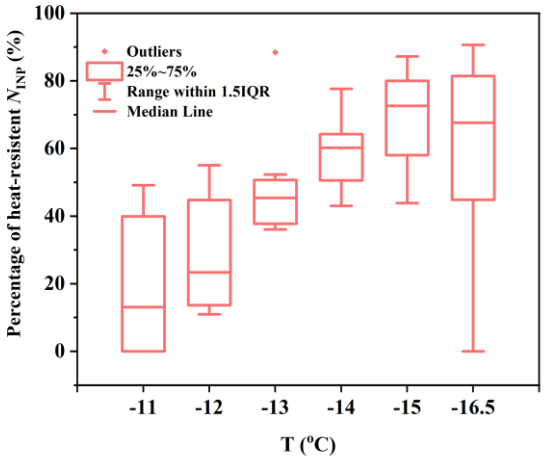

Figure 2. The percentages of heat-resistant N_{INP} vs. temperatures; only values at temperatures above −16.5 ºC were calculated where droplets in all samples did not finish frozen.

The source of aerosols and heat-resistant INPs was explored based on the chemical analysis of collected particles. The contribution of heat-resistant INPs from OM is considered to be minor under the determined conditions, as a poor correlation between the detected heat-resistant INPs and organic carbon was found (R^2=0.19, Fig. S1). The chemical elements of the coarse particles (D_{50}=3.2 μm) (Fig. 3 and Table S3) show that crustal elements including Ca (50.46%±2.27%), Mg (10.98%±0.82%), Fe (14.89%±1.48%) and Al (10.42%±1.45%) account for a major mass percentage (88.64%±2.92%) of the total element mass of particles, demonstrating the strong contribution from dust particles. Differently, the crustal elements in

fine particles (D_{50}= 0.56 μm) are much less, while a large mass percentage of non-dust related elements (Zn, Pb, K, and Na) are found (48.53%±10.61%) (Fig. 3 and Table S3). This indicates that dust particles contribute to heat-resistant INPs in the coarse particles, while fine particles may be influenced by non-dust anthropogenic sources (see detailed discussion in Fig. S2 and Text S1). This conclusion is also supported by the different ice nucleation activities (presented as n_s, calculated based on Eq.(3)) obtained for coarse and fine INPs (Fig. S3), which comes from different aerosol sources.

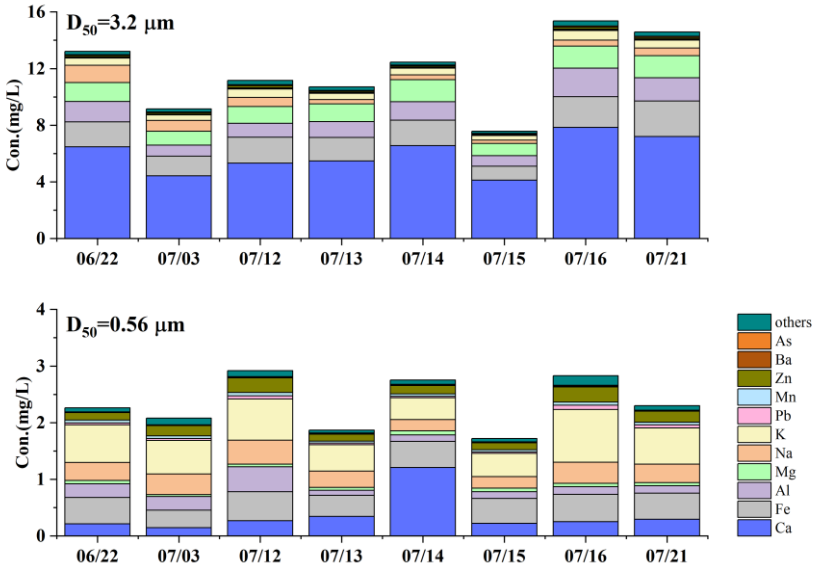

Figure 3. The element compositions of particles in different sizes.

Dust particles in the urban atmosphere can either be long-range transported desert dust or anthropogenic dust [*Han et al.*, 2005; *Zender et al.*, 2004]. Here, we confirmed natural desert dust has negligible impact on the heat-resistant INPs while anthropogenic dust is a major source, as supported by the following evidence. No obvious enhancement in the number concentration of coarse particles and in n_s has been observed during the sampling time (Fig. S4(a)), which typically occurred during Asian dust storm events (Fig. S4(b)) [*Chen et al.*, 2021a]. The n_s obtained in this study ($3*10^3$ to $2*10^5$ m^{-2} at -15 °C) were two magnitudes lower than those obtained during the spring Asian dust events (10^5 to 10^7 m^{-2} at -15 °C) at the same site [*Chen et al.*, 2021a]. The analysis of 72-hours backward trajectories (calculated every 6 hours at 1:00, 7:00, 13:00, and 19:00 UTC at the height of 500 m) (Fig. S5) shows that air masses did not pass over the desert regions. Asian dust events occur especially during spring months and not in the summer of Beijing [*Shao and Dong*, 2006], while the anthropogenic dust from local emissions can contribute ~80% of the total dust [*Han et al.*, 2005].

The impact of one representative anthropogenic dust species on the collected aerosols and INPs, the traffic-influenced road dust, is proved by the good correlation between the hour-mean concentration of the coarse particles and nitric oxide (NO) during the sampling time (R^2=0.53, Fig. S6). Coarse particles and NO are indicators of the dust particles and vehicle primary emissions, respectively. Good correlations between heat-resistant N_{INP} at −16 °C and the mass percentage of Ba and Zn were observed in coarse particles (R^2=0.39 and R^2=0.60, Fig. S7). Ba and Zn are two tracers for the road dust particles [*Gietl et al.*, 2010; *Harrison and Alghamdi*, 2023; *Peltier et al.*, 2011], which proves the contribution of road dust to heat-resistant INPs again. Other anthropogenic dust species, such as those generated from agricultural and construction activities may contribute to the collected coarse particles as well but cannot be validated in the present study due to lacking reliable tracers. Since the dust generated from construction is commonly found in urban regions [*Azarov et al.*, 2019; *Yan et al.*, 2020], we expect its contribution to the observed anthropogenic dust INPs. The impact of

soil dust from agricultural activities is hard to be evaluated here. However, the n_s values of the obtained heat-resistant INPs are 3 orders of magnitudes lower than those from soil dust (original or H_2O_2 treated soil dust) measured by *Tobo et al.* [2014] (Fig. S8) and than the n_s of inorganic INPs measured by *Testa et al.* [2021] which was presumably from land surface emission. This comparison implies that the ice nucleation activity of the airborne anthropogenic dust cannot be explained alone by the collected ground-based and near-source soil dust.

3.3 Parameterization of Anthropogenic Dust INPs

Based on the aforementioned results, the coarse heat-resistant N_{INP} can be referred to as anthropogenic dust N_{INP} in the urban atmosphere. The anthropogenic N_{INP} is highly correlated to the number concentration of coarse particles ($N_{>1\ \mu m}$, R^2=0.67, Table S2), which can be foreseen from the strong impact of dust on the coarse particles (section 3.2). Thus, $N_{>1\ \mu m}$ is used to predict the concentration of anthropogenic dust INPs. For comparison, the correlations between the anthropogenic N_{INP} and the number concentration of particles with other size ranges ($N_{>500\ nm}$ and $N_{>1.8\ \mu m}$) are listed in Table S2.

Table 1 The parameterizations to predict the anthropogenic dust INPs

$$N_{\mathrm{INP}} = (N_{>1\ \mu m})^{(\alpha(-T)+\beta)} * \exp(\gamma(-T) + \delta)$$

	α	β	γ	δ	R^2
Anthropogenic dust INPs	$-1.835*10^{-5}$	0.0294	0.652	-12.518	0.927

The parametrization to predict the anthropogenic dust INPs at a temperature range from $-21^oC \sim -7^oC$ was developed based on $N_{>1\ \mu m}$ following the form of *DeMott et al.* [2015] parameterization (hereafter D15) (listed in Table 1). The coefficient of determination (R^2) of the resulting parameterization is 0.95 with 95% confidence. The comparison between the results from observation and prediction was shown in Fig. S9. The ratios of the observed to the predicted values are within a factor of 3, indicating a good prediction from the parameterization. To be compared, the heat-resistant N_{INP} was estimated using D15 as well, which was developed based on $N_{>500\ nm}$ (Fig. S10). An overestimation of heat-resistant/total INPs about 1~2 magnitudes was observed, indicating poor prediction. This is because the particles larger than 500 nm cannot represent the exact size range of the measured INP species, the anthropogenic dust INPs here (evidenced by the different chemical compositions and ice nucleation activities of coarse and fine particles). The overestimation of N_{INP} in the urban environment by D15 was confirmed by *Chen et al.* [2018a] and *Bi et al.* [2019] as well.

Overall, the parameterization developed based on $N_{>1\ \mu m}$ showed a good performance on the prediction of the anthropogenic dust INPs. This parameterization can then be used in regional or climate models to predict the INPs contributed by anthropogenic dust and evaluate its further impact on cloud formation and the urban climate. Note that using heat-resistant N_{INP} as the proxy of anthropogenic dust N_{INP} in the urban atmosphere can cause a bias. This assumption cannot exclude that some of the measured heat-resistant INPs are from the heat-resistant OM [*Hill et al.*, 2016; *O'Sullivan et al.*, 2014; *Tobo et al.*, 2014], although a poor correlation between OM and heat-resistant INPs and significant dust influence has been found here. On the other hand, some dust species can significantly lose their ice nucleation activity when undergoing the heating process [*Daily et al.*, 2020], indicating the possibility of underestimating the anthropogenic dust N_{INP} based on the heat-lability of particles. These uncertainties might influence the quantification of anthropogenic dust INPs and need more investigation on measuring the contribution of anthropogenic dust to aerosols and atmospheric INPs in the urban environment.

4 Conclusions

The abundance and sources of INPs remain unclear in the urban atmosphere. In the present study, the ice nucleation activities of size-resolved particles were investigated in a typical urban environment. INPs in coarse-mode account for 95.2%±4% of the detected N_{INP} and are significantly contributed by heat-resistant INPs (~70%) at temperatures below −15 ºC. The further size-resolved chemical composition analysis indicates that anthropogenic dust such as traffic-influenced road dust is a major source of these heat-resistant INPs. The number concentration of coarse particles is strongly correlated with those of the anthropogenic dust INPs, thus was used to develop the parameterization for predicting anthropogenic dust INP concentrations. The new parameterization predicts the observed N_{INP} within a factor of 3, showing a good performance. This study highlighted the importance of anthropogenic dust particles as a significant source of INPs in urban environments, which has not been extensively studied before.

Projections of past and future atmospheric dust loads suggest that the contribution of anthropogenic emissions is increased in the last decades and it will continue to have a profound role in the Earth's future climate [*Mahowald and Luo*, 2003; *Stanelle et al.*, 2014]. Due to the good ice nucleation activity of the anthropogenic dust and its large contributions to the atmospheric INPs reported by the present study, we suggest that anthropogenic dust will become an important factor to connect with urban climate change by modifying INP concentrations and cloud microphysics.

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Conflict of interest

The authors declare no conflicts of interest relevant to this study.

Open Research

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Supporting Information for

Anthropogenic dust as a significant source of ice-nucleating particles in the urban environment

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Introduction

The Supporting Information shows additional figures (S1~S10) and tables (S1~S3) to provide detailed sampling information and other physicochemical characterizations of aerosols and ice nucleating particles in the present study.

Table S1. The sampling information of the filter samples

Sampling Date	Starting time*	Ending time	Air volume (L)	$PM_{2.5}$ (μg m^{-3})	PM_{10} (μg m^{-3})
2020/06/22	9:32	9:21	42870	66.3±22.9	87.7±28.6
2020/07/03	9:47	9:43	43080	58.8±24.6	68.5±30.5
2020/07/12	11:23	9:21	39540	108.0±31.4	136.7±35.7
2020/07/13	9:36	9:25	42870	53.2±22.0	79.2±25.1
2020/07/14	9:57	9:29	42360	38.4±9.8	61.2±12.9
2020/07/15	9:46	9:24	42540	35.6±9.1	57.8±12.8
2020/07/16	9:41	9:27	42780	60.6±22.1	86.8±22.2

2020/07/21	9:45	9:13	42240	44.6±12.3	73.1±18.0

*The sampling time is Beijing time (UTC +8 hours)

Table S2. The correlation between the number concentration of INP (N_{INP}) and particles with different sizes ($N_{>*}$)

The number concentration of particles larger than * μm ($N_{>*}$)	R^2: correlation between $N_{>*}$ and N_{INP}	
	Total N_{INP}	Heat-resistant N_{INP}
$N_{>500\ nm}$	0.37	0.51
$N_{>1\ \mu m}$	0.50	0.67
$N_{>1.8\ \mu m}$	0.20	0.28

Table S3. Element concentrations of particles in different size

Date	Na	Mg	Al	K	Ca	Mn	Fe	Zn	As	Ba	Pb	others
D_{50}=3.2 μm (μg/L)												
6/22	1217.4	1325.3	1443.0	507.3	6473.0	61.1	1762.8	95.6	2.4	61.8	16.6	244.1
7/03	766.2	979.1	782.7	383.6	4422.0	38.7	1392.0	98.2	1.5	54.1	13.2	213.1
7/12	638.9	1194.8	972.9	601.3	5317.4	51.9	1833.3	149.6	3.4	75.1	22.2	293.9
7/13	300.3	1260.1	1115.4	452.9	5478.9	49.1	1648.0	80.6	2.6	56.1	13.6	245.8
7/14	336.8	1546.5	1309.2	480.6	6550.4	48.8	1803.5	80.7	1.6	74.0	13.0	203.1
7/15	237.0	861.5	747.2	320.1	4118.6	30.8	977.7	70.2	1.6	44.6	8.1	159.8
7/16	435.5	1554.1	2009.6	671.4	7841.1	63.7	2171.4	136.1	2.4	82.7	43.0	344.8
7/21	535.4	1556.3	1640.4	556.9	7201.9	68.3	2510.6	109.6	2.7	81.5	18.9	296.4
D_{50}=0.56 μm (μg/L)												
6/22	317.5	59.9	238.5	665.2	213.2	49.1	466.6	132.8	8.2	6.6	33.8	68.1
7/03	366.6	31.4	244.3	590.2	144.8	46.0	307.3	179.4	8.6	7.4	36.8	118.2
7/12	426.3	46.8	439.0	724.6	268.5	61.1	510.9	255.5	15.9	8.0	55.6	105.7
7/13	289.3	51.6	86.0	464.4	343.4	36.4	373.8	120.5	13.8	6.9	29.1	53.5
7/14	198.4	73.4	115.5	383.1	1206.3	43.9	461.2	147.1	13.4	7.4	26.1	77.2
7/15	200.4	62.4	122.4	412.8	221.0	39.2	438.5	120.2	8.4	8.6	25.3	61.4
7/16	372.1	60.0	134.3	929.3	251.9	54.0	482.7	268.6	16.4	11.4	77.6	170.6
7/21	325.3	54.1	131.0	642.0	293.7	48.2	460.7	193.5	8.9	7.5	53.6	82.0

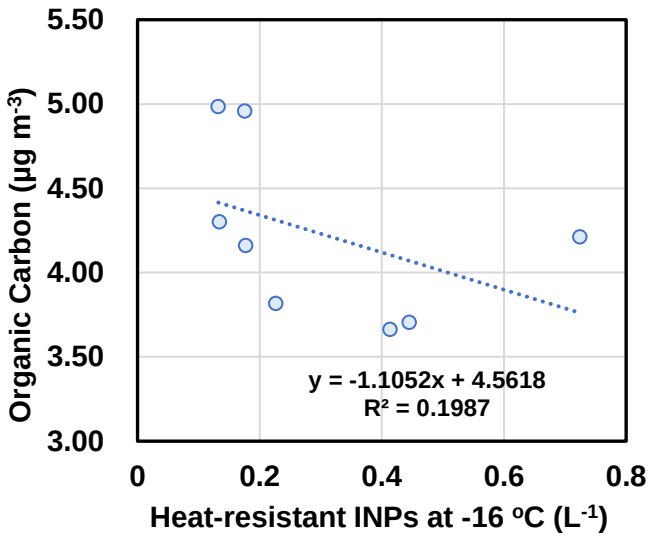

Figure S1. The correlation between heat-resistant N_{INP} at -16 °C and organic carbon (OC) measured by the Sunset ECOC analyzer.

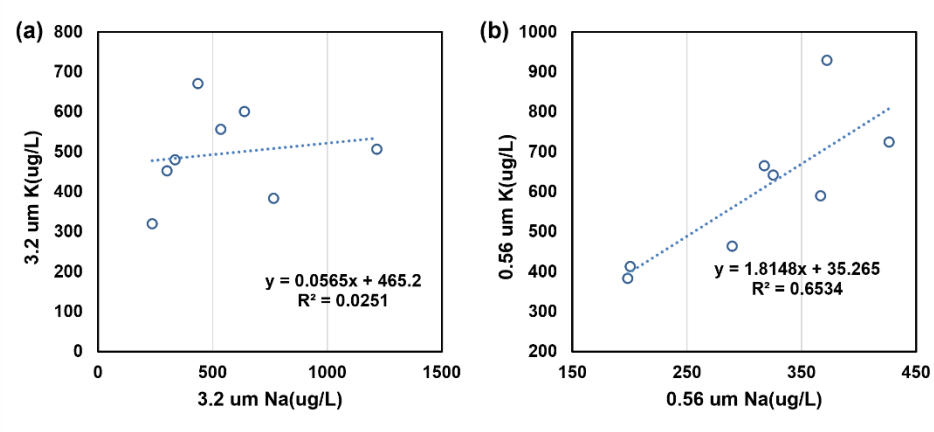

Figure S2. The correlations between the Na and K mass concentrations in 3.2 µm (a) and 0.56 µm (b) particles.

Text S1. The strong correlation between Na and K in fine particles (R^2=0.65) proved that they come from the same anthropogenic source such as biomass burning and industrial emissions, as supported by previous studies [*Lian et al.*, 2022; *Ooki et al.*, 2002]. This correlation became very weak in coarse particles (R^2=0.02) where Na was commonly found in natural sources (such as the sea-salt NaCl in marine aerosol). This result indicates that compared to coarse particles, fine particles are influenced by anthropogenic sources.

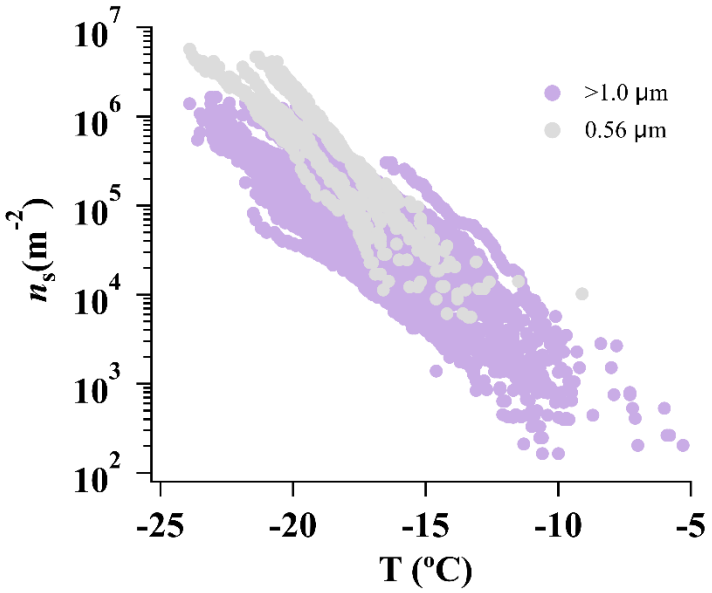

Figure S3. The cumulative number concentration of the ice active sites per unit surface area (n_s) of particles in different sizes. Particles larger than 1 µm include particles with D_{50} sizes of 1 µm, 1.8 µm, 3.2 µm and 5.6 µm.

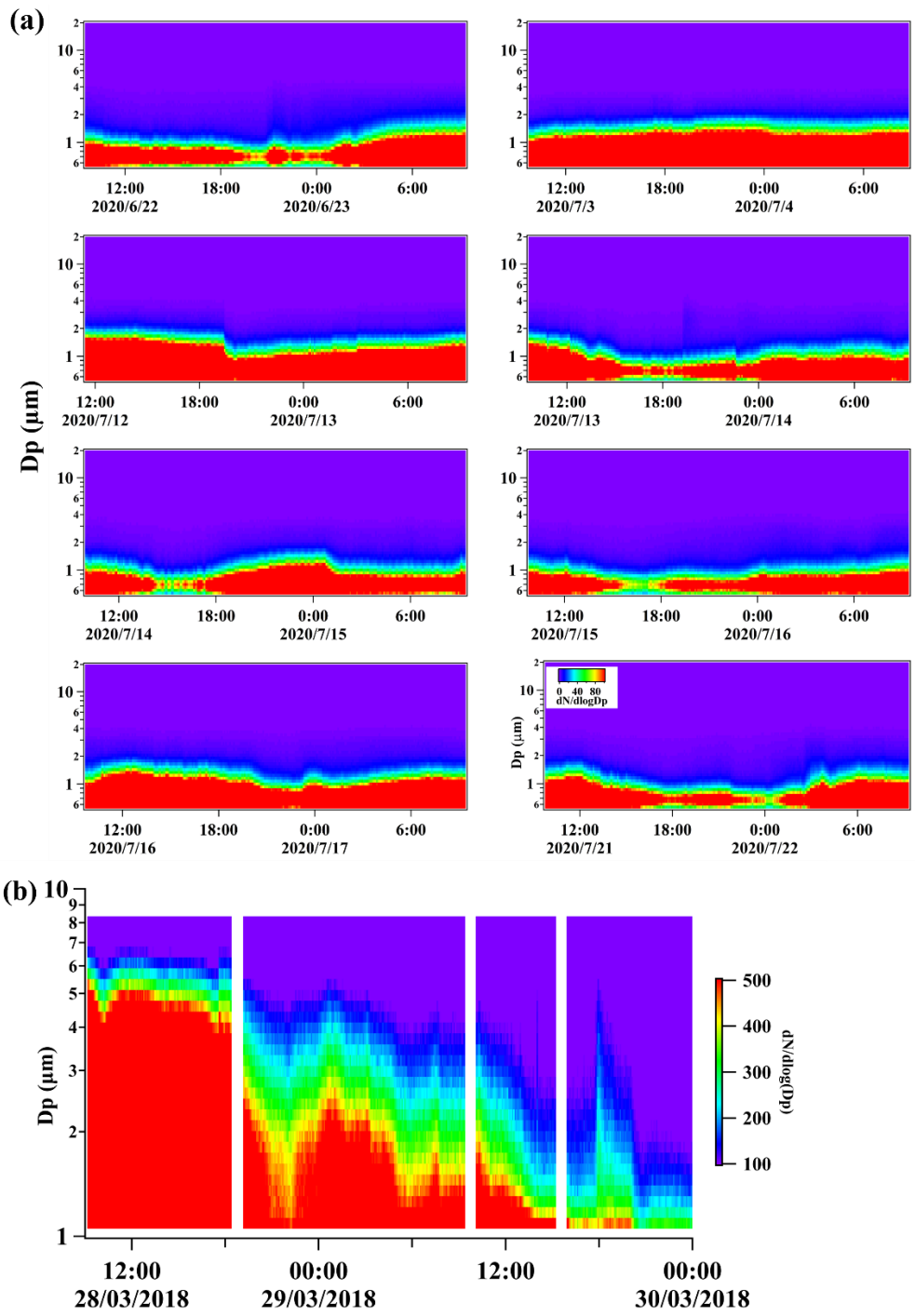

Figure S4. The time series of the aerodynamic size (Dp) distribution of aerosols from 0.542 μm to 19.81 μm measured during each sampling day (a) and compared with those detected during the dust event (b) by *Chen et al.* [2021]. The unit of dN/dlog(Dp) is cm^{-3}.

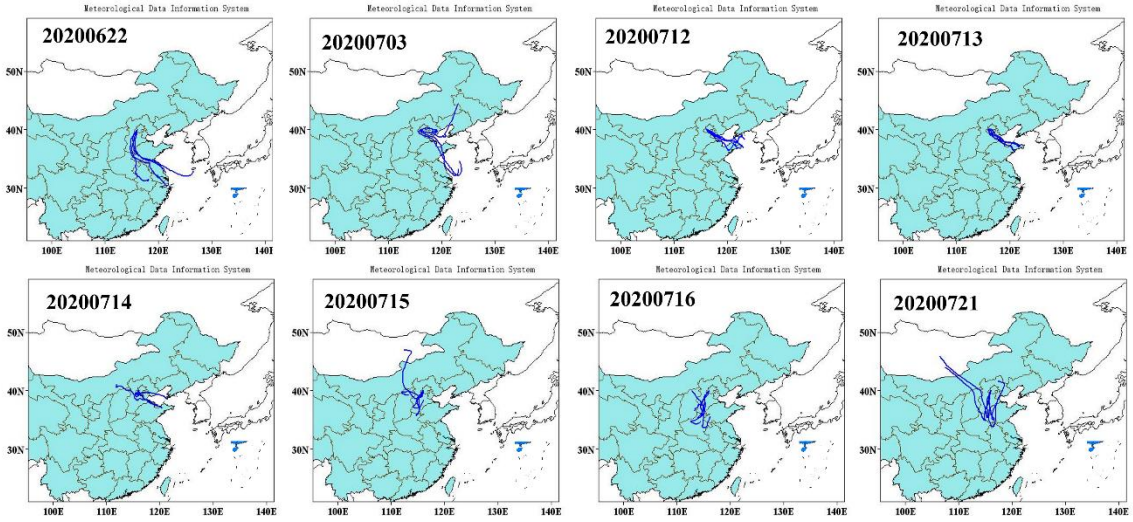

Figure S5. The 72 hours backward trajectories of air masses during the sampling time. Each trajectory was calculated every 6 hours, resulting in 4 trajectories for each sample (sampling for 24 hours) calculated at 1:00, 7:00, 13:00, and 19:00 UTC.

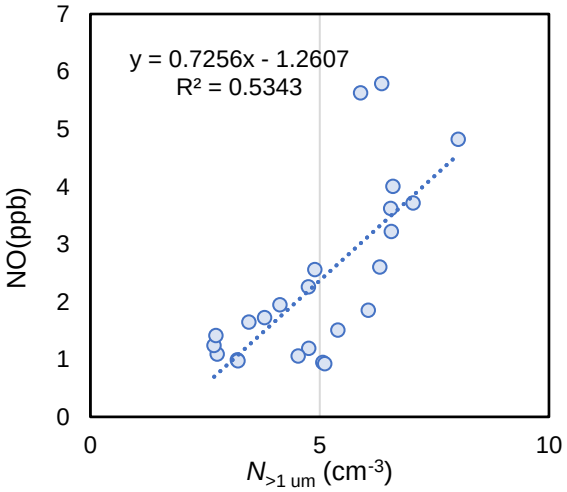

Figure S6. The hourly mean concentration of nitric oxide (NO) and the hourly mean number concentration of coarse particles during the sampling time. One data point represents the mean value of NO detected at the same time on different days (24 data points indicate 24 hours of the day).

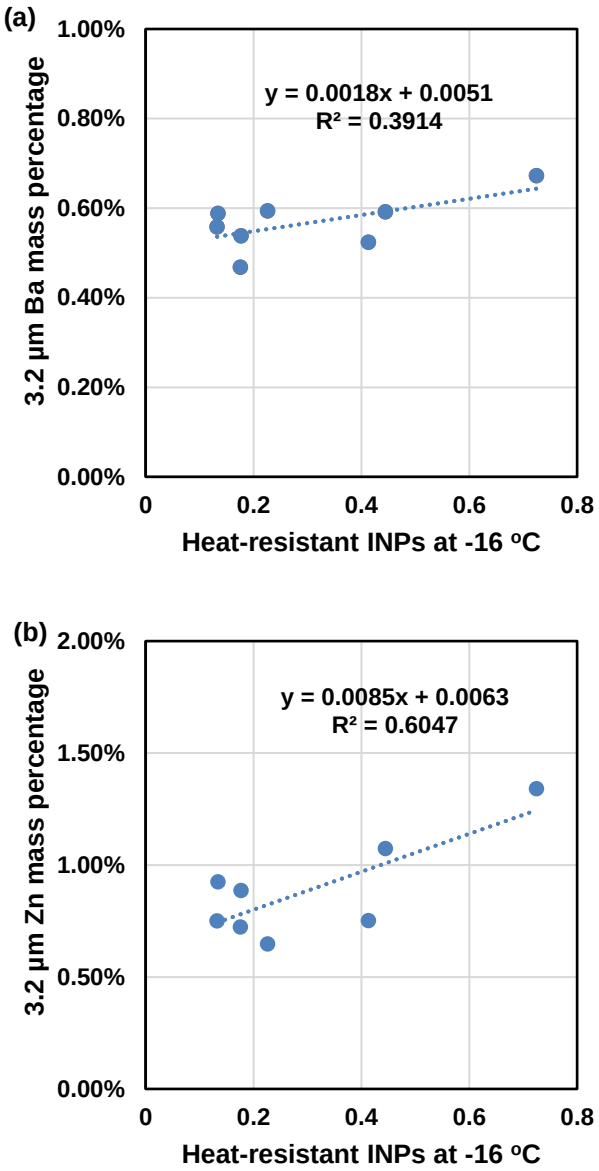

Figure S7. The correlations between the heat-resistant N_{INP} and the mass percentages of Ba (a) and Zn (b) in D_{50}=3.2 μm particles.

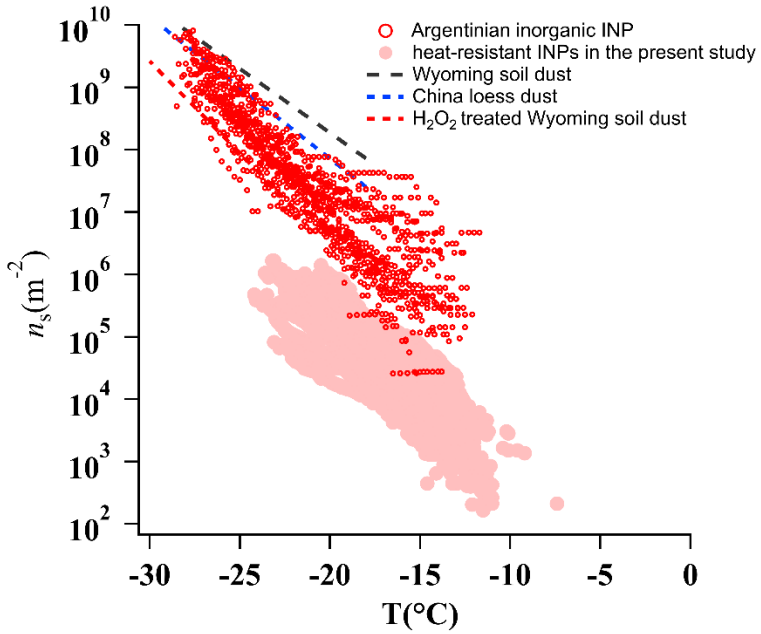

Figure S8. The comparison of n_s from the present study (purple circles) and those from the soil dust measured by *Tobo et al.* [2014] and inorganic INPs influenced by Argentinian land surface emission in *Testa et al.* [2021]. Note that the unit of n_s in *Tobo et al.* [2014] was cm^{-2} while here we converted the data to m^{-2}.

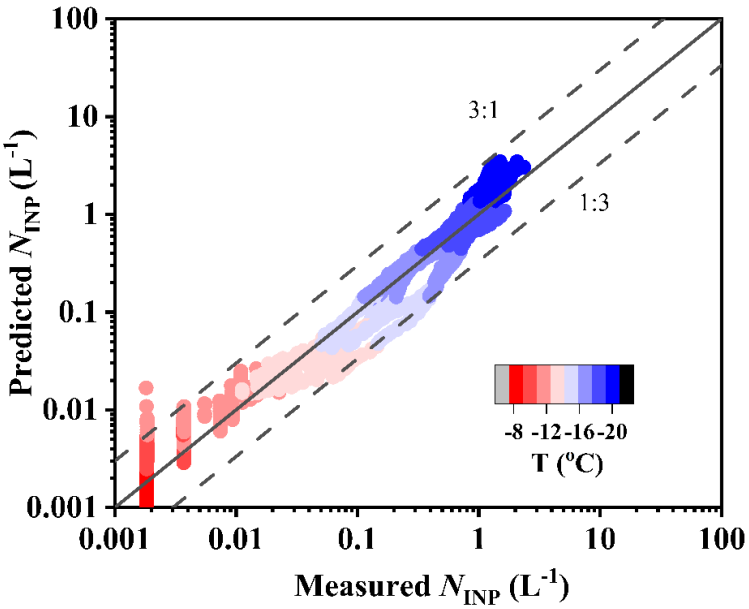

Figure S9. The comparison of N_{INP} from the observation and predicted by the newly developed parameterization in the present study.

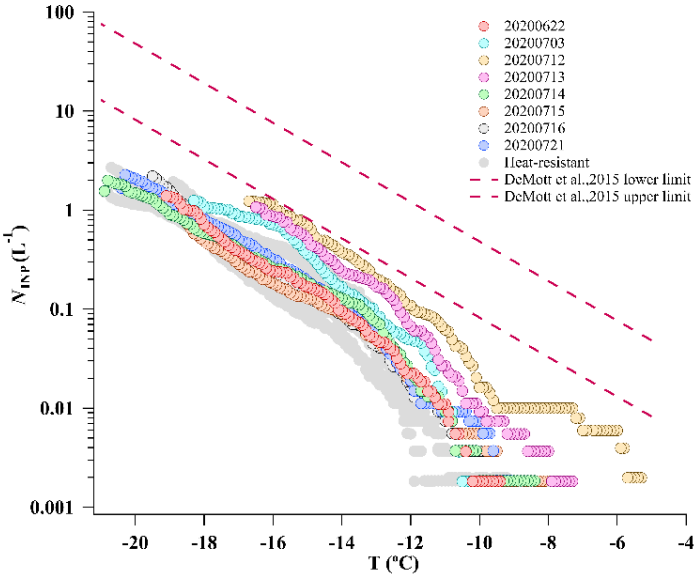

Figure S10. The number concentration of INPs (N_{INP}) as a function of temperatures measured during the sampling time; The gray circles represent the number concentration of heat-resistant INPs after being heated at 95 °C for 20 mins. The pink dashed lines represent the upper and lower limits of INPs estimated according to the parameterization proposed by *DeMott et al.* [2015] based on the highest and the lowest number concentrations of particles larger than 500 nm detected in the present study.

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