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Polluted dust promotes new particle formation and growth

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Understanding new particle formation and their subsequent growth in the troposphere has a critical impact on our ability to predict atmospheric composition and global climate change. High pre-existing particle loadings have been thought to suppress the formation of new atmospheric aerosol particles due to high condensation and coagulation sinks. Here, based on field measurements at a mountain site in South China, we report, for the first time, in situ observational evidence on new particle formation and growth in remote ambient atmosphere during heavy dust episodes mixed with anthropogenic pollution. Both the formation and growth rates of particles in the diameter range 15–50 nm were enhanced during the dust episodes, indicating the influence of photo-induced, dust surface-mediated reactions and resulting condensable vapor production. This study provides unique in situ observations of heterogeneous photochemical processes inducing new particle formation and growth in the real atmosphere, and suggests an unexpected impact of mineral dust on climate and atmospheric chemistry.

A erosol particles, being important to both global climate[1,2] and atmospheric chemistry[3], can be directly emitted from natural or anthropogenic sources and secondarily formed in the atmosphere[3]. Mineral dust, with an emission rate of 1000–3000 Tg per year from deserts or semiarid areas[4], is the most important primary aerosol particle source and dominates the aerosol mass in the global atmosphere. New particle formation (NPF) and subsequent particle growth form together a secondary aerosol particle source that controls the total particle number concentration[5–7]. The climatic effects of mineral dust and atmospheric NPF have been thought to be disconnected from each other. Mineral dust affects climate via direct light scattering and ice nucleation[1,2,8–10], whereas NPF contributes significantly to the global cloud condensation nuclei (CCN) budget[5–7] and thereby to uncertainties in the indirect radiative forcing[11,12].

Mineral dusts injected into the atmosphere can affect atmospheric photochemistry in very complicated ways. On one hand, dust tends to suppress photochemical processes via light scattering[1], scavenging of reactive gases[13] and photochemical oxidant uptake[14,15]. For example, both field and model studies have reported significant reductions of acidic gases and O_3 in dust plumes[14,16–19]. On the other hand, recent studies indicate that mineral dust can induce chemical reactions in the presence of sunlight, defined as heterogeneous photochemistry[20,21]. Such observations suggest that dust has the potential to participate in and promote the atmospheric photochemical processes directly as a reactant or catalyst[22].

NPF and subsequent particle growth are tied to photochemistry as well. Particle nucleation generally originates from the production of H_2SO_4 via the oxidation of SO_2 by the OH radical[5], while particle growth depends crucially on low-volatile vapors produced from the photo-oxidation of volatile organic compounds (VOCs)[23,24]. High pre-existing particle loadings are considered to suppress the atmospheric NPF because of high condensation and coagulation sinks[5]. Therefore, it has been thought that dust and NPF do not usually co-exist, since high dust concentrations tend to scavenge both low-volatile vapors (e.g. sulfuric acid) and small molecular clusters efficiently[6,7]. In this study, we analyzed a two-month period of comprehensive observations during the Asian dust storm season in 2009 at a mountain top site in southern China[25]. The results show, for the first time, that high dust concentrations and NPF can really co-exist, in addition to

which we provide in situ evidence of dust-related heterogeneous photochemical processes that promote atmospheric new particle formation and growth.

Results

As a part of a field campaign on aerosol-cloud interaction at Mount Heng in Hunan Province, southern China[25], we conducted intensive measurements of a wide variety of trace gases and aerosols during spring 2009 (March to May), a season with frequent dust events (see Supplementary Information (SI) for details). Fig. 1 shows the temporal variation of several quantities (including PM_{10}, $PM_{2.5}$, calcium in $PM_{2.5}$, and particle number size distribution in the diameter range of 10–500 nm) measured during 20 April to 9 May, 2009. Two episodes of very high aerosol loadings, with maximum PM_{10} mass concentrations of about 248 μg m^{-3} and 911 μg m^{-3}, appeared during 20–22 April and 25–26 April, respectively. The low mass fractions of fine particles and high calcium concentrations during the two events suggest the primary origin of these particles was a dust storm. The satellite observations on board the CALIPSO satellite on 25 April, showed clearly the presence of multi-layered dust aerosols in the lower troposphere over the southern China (Fig. S2).

Unexpectedly, intense new particle formation and growth was observed during the two major dust episodes (Fig. 1). The formation rates of 15 nm particles, J_{15}, were in general slightly higher during dust days than during non-dust days (0.27 ± 0.12 cm^{-3} s^{-1} vs. 0.23 ± 0.06 cm^{-3} s^{-1}), and the highest rate (0.45 cm^{-3} s^{-1}) was observed on the strongest dust day (April 25th, Table 1). It should be noted that J_{15} represents the combined effects of atmospheric nucleation, growth of nucleated particles to the diameter of 15 nm, and simultaneous scavenging of the growing nuclei by the pre-existing population of larger particles. As a result, J_{15} can be considered as a good metric for estimating the source rate of new particles that might eventually lead to the production of new CCN. The particle growth rates in the diameter range 15 to 30 nm were comparable to each other between the dust and non-dust days (Table 1), indicating larger production rates of extremely low-volatile vapors on dust days, when the condensation sink for such vapors was very high. In the diameter range 30 to 50 nm, the mean particle growth rate on dust days was 14.3 ± 6.5 nm h^{-1}, more than twice the value on non-dust days (6.6

± 3.4 nm h^{-1}). The highest growth rates were observed on April 22nd and 26th during the two main dust episodes (Table 1). Such growth rates are quite high compared to other regions around the world (typically below 10 nm h^{-1} in remote atmosphere and below 20 nm h^{-1} in some moderate polluted regions[6]). However, considering the high condensation and coagulation sinks at particle loadings in excess of 650 μg m^{-3} (mean value during the NPF period at 25th April), the occurrence of NPF and particle growth observed here appears to contradict our current understanding of aerosol dynamics[5–7,11].

Since sulfuric acid is the key driver of atmospheric nucleation[5,26], we utilized the sulfuric acid proxy approach, described simply as the ratio of UV × SO_2 to condensation sink (CS)[27], to investigate the role of sulfuric acid in NPF events at Mt. Heng. Fig. 2A shows the scatter plot of UV × SO_2 versus CS during typical (defined as continuous growth occurring after the particle nucleation) and non-typical NPF events (defined as no evident growth occurring after the particle nucleation, in which case particle nucleation rates and growth rates are hardly to be calculated) as well as non-NPF events. Since high sulfuric acid concentration and low CS promote NPF, it is not strange at all that the NPF event days (both typical and non-typical events) and non-NPF event days were separated into two different groups laying to the left and right of the diagonal line, respectively. However, the NPF events observed during 25–26 April behaved in the opposite way, especially on 25 April when the PM_{10} approached 650 μg m^{-3} at the period of NPF (12:00–14:59 LT). In addition, the formation of fine sulfate was observed during the dust event of 25–26 April (See strong diurnal cycle of sulfate in Fig. S7). Considering their alkaline nature, mineral dust particles should take up gaseous acids more easily than most other types of particles[13,28], which is expected to suppress NPF even further. Therefore, our observations strongly suggest an additional dust involved pathway enhancing the production of sulfuric acid. It should be noted that several recent studies in polluted urban regions also observed NPF with high CS[29], which can be attributed to even stronger source strength of low-volatile vapors resulting from very high SO_2 concentrations.

Particle growth plays a critical role in determining whether new-formed particles can reach sizes to be able to act as cloud condensation nuclei (diameter >50–100 nm). Different from particle nucleation,
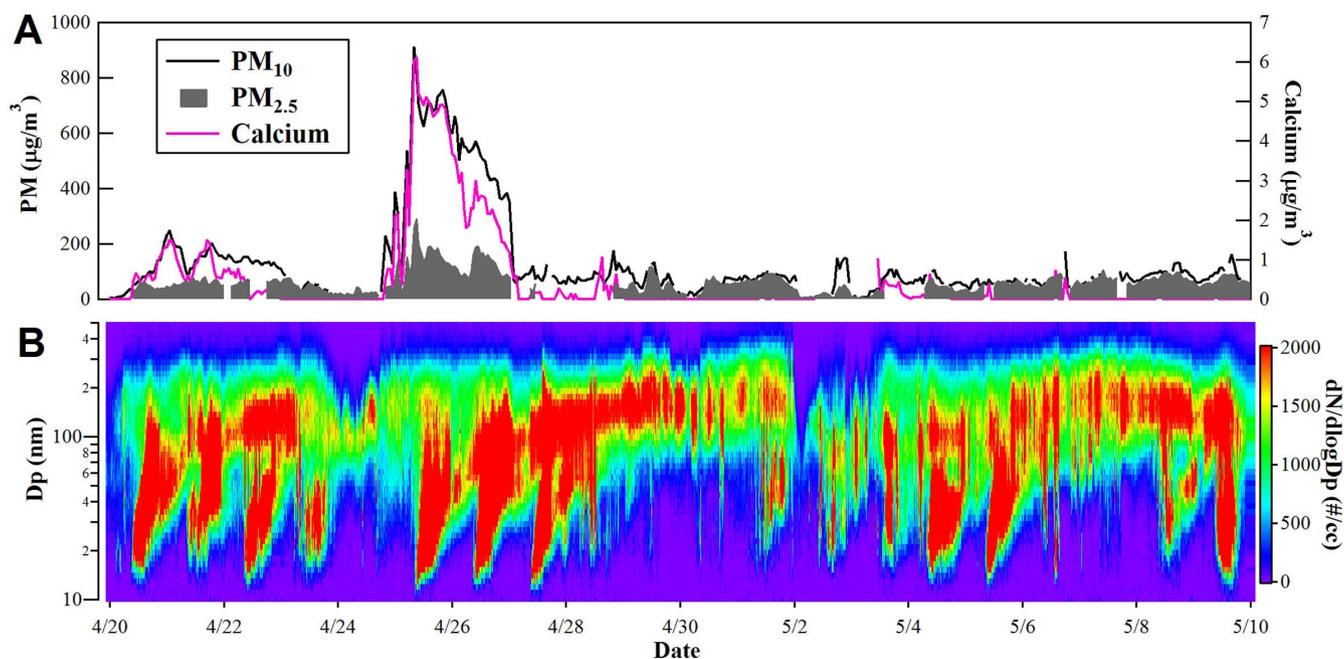

Figure 1 | Temporal variations of aerosol related parameters during 20 April to 9 May, 2009. (A) Time series of PM_{10}, $PM_{2.5}$, calcium in $PM_{2.5}$, and (B) particle number size distribution at Mt. Heng during April–May 2009.

Table 1 | Formation rates, growth rates, source rates of vapor and PM$_{10}$ concentrations during typical new particle formation events at Mt. Heng during March to May of 2009

Date	J$_{15}$* (cm^{-3} s^{-1})	GR(15–30)* (nm h^{-1})	GR(30–50)* (nm h^{-1})	Q (cm^{-3} s^{-1})	PM$_{10}$*** (µg m^{-3})
15-Mar	0.26	4.3	NA**	1.0e + 06	****
25-Mar	0.17	5.6	5.6	1.1e + 06	56.3
7-Apr	0.27	7.7	11.4	1.9e + 06	75.7
20-Apr	0.15	6.1	6.1	1.0e + 06	82.0
22-Apr	0.20	4.9	19.3	1.5e + 06	135.1
25-Apr	0.45	7.2	12	2.8e + 06	668.6
26-Apr	0.26	7.4	22.4	2.8e + 06	506.9
27-Apr	0.31	6.1	11.9	1.7e + 06	75.3
4-May	0.17	4.6	3.5	1.3e + 06	69.8
5-May	0.29	6.4	5.8	1.8e + 06	54.6

*: The time periods of particle nucleation and growth varied largely from day to day. Generally, particle nucleation started at 10:00–12:00 LT, ended at 12:00–13:00 LT; particle growth from 15 nm to 30 nm started at 11:00–12:00 LT, ended at 13:00–14:00 LT; particle growth from 30 nm to 50 nm started at 13:30–14:30 LT, ended at 15:00–17:00 LT.
**: No growth rate can be detected.
***: Hourly average values of 12:00–14:59 LT.
****: PM$_{10}$ mass was not measured.

the contribution of sulfuric acid to particle growth is usually minor, especially at sizes larger than a few nanometers[30,31]. Instead, low-volatile organic vapors (oxidized biogenic VOCs and nitrogen containing organics) have demonstrated to play a key role[23,24]. To investigate the possible roles of VOCs during the dust events, we estimated the concentration levels of directly-emitted VOCs during the campaign by carrying out Lagrangian dispersion modeling[32–34] based on the emission inventories for total anthropogenic VOCs[35] and biogenic monoterperenes[36] (See Fig. S4). The simulations showed that, during the dust events, both anthropogenic and biogenic VOCs concentrations were much lower than the corresponding mean values during the whole campaign. In addition, the ratio of ozone concentration to CS, indicating the potential of producing high concentrations of low-volatile organic vapors, clearly showed much lower values during 25–26 April. This observation violates our current understanding of particle growth, unless there were some non-traditional mechanisms promoting the oxidation of VOCs.

So, what are the most plausible additional sources of gaseous sulfuric acid and low-volatile organic vapors? In case of sulfuric acid, one potential pathway is the dust-induced photo-catalytic reaction which can produce additional OH radicals to accelerate the oxidation of SO$_2$[37]. This is a plausible explanation for the observed episode on 25–26 April, as the conditions with increased photo-sensitive components, sufficient solar radiation, water and reactive species were favorable to this pathway. As shown in Fig. S5, the strongest new particle formation that occurred on 25 April was connected to heavy loadings of Fe (mostly exist as Fe$_2$O$_3$) and the highest relative humidity among the dust event days (detailed information is described in the SI, Fig. S5 and S6). Both of these features act in favor of heterogeneous photocatalytic reactions in the water films around the dust particles. In addition, the photolysis of enhanced HONO is another possible pathway to form OH radical, which has been demonstrated to be induced by the TiO$_2$ photocatalysis of NO$_2$ in our previous study[25,38] (detailed information is also described in the SI).

In case of low-volatile organic vapors enhancing the particle growth, photo-promoted oxidation of VOCs by the illuminated dust, demonstrated by several laboratory studies[39–41], is considered as the most plausible pathway. Secondary vapors with a wide range of volatility can be produced by the oxidation of complex VOCs, such as biogenic organics. A small fraction of these vapors are extremely low volatile and can grow efficiently all-sized particles, including the very small ones[42]. Most of the oxidation products have higher volatilities and can contribute to the growth of larger particles only. Therefore, our observation that 30–50 nm particle growth rates were higher than those of 15–30 nm particles in the dust plumes (Table 1)
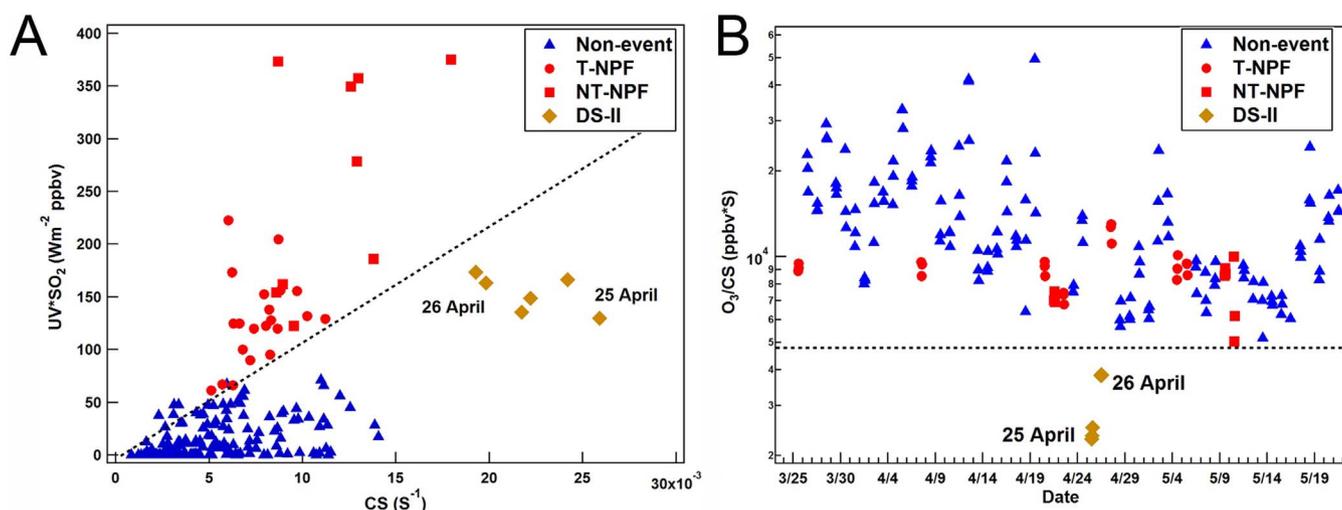

Figure 2 | Analysis of relationship between different proxies. (A) Scatter plot of hourly averaged UV × SO$_2$ versus condensation sink at noontime (12:00–14:59 LT), (B) Temporal variations of the ratios of daily noontime (12:00–14:59 LT) averaged ozone to condensation sink during March–May 2009.
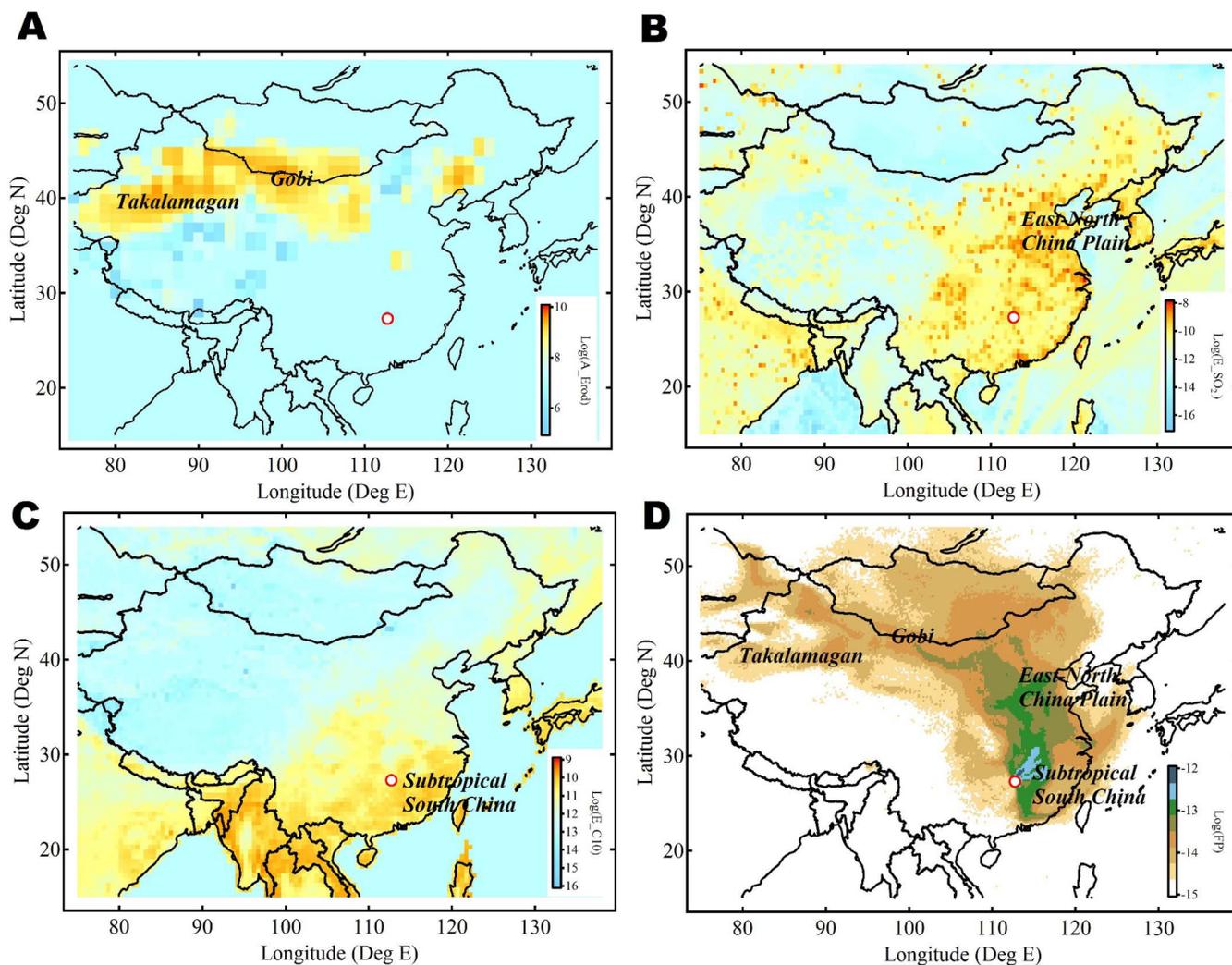
Figure 3 | Map shows eroded area, emission rates and transport pathways for the dust cases. (A) Area of eroded in Asia, (B) Emission rate of SO$_2$, (C) Emission rate of monoterperene from biogenic emission in April, and (D) 100 m footprint retroplume calculated by 7-day backward Lagrangian dispersion simulation for 20–22 April and 25–26 April, 2009. The maps were drawn by the software of Igor Pro, http://www.wavemetrics.com/.

is consistent with our hypothesis of heterogeneous production of oxidants in dust plumes. To quantify the contribution of dust-related processes to the condensable vapors, we calculated the source rates of the vapors (Q) during the typical NPF days (see Table 1). During 25–26 April, the values of Q were 2.8×10^6 cm^{-3} s^{-1}, twice those on non-dust days (about 1.4×10^6 cm^{-3} s^{-1} on average), indicating that at least half of the vapor precursors for observed particle growth on 25–26 April might have been produced from heterogeneous photochemical processes associated with the presence of dust.

In summary, our study provides observational evidence on photo-induced particle nucleation and growth, as well as HONO production[25], in the presence of mineral dust particles in the real atmosphere. These results demonstrate that mineral dust can participate in atmospheric photochemistry directly as reactants or catalysts, and that dust can enhance the atmospheric oxidation capacity by surface redox reactions[20] and gaseous radical production. Given the increasing evidence of anthropogenic industrial dust[43] and worldwide distribution of chromophores (i.e. nitrate and organics)[22], photo-sensitive aerosols might be important to atmospheric photochemistry on a global scale. In laboratory experiments, dust-related heterogeneous photochemical reactions have been demonstrated to be stronger when dust particles are mixed or coated with other reactive species such as nitrate[44]. In this study, such a mixture between mineral dust and anthropogenic pollutants actually occurred. In Fig. 3A–3D, we

show the calculated transport pathways using a Lagrangian transport and dispersion model with a "footprint" retroplume for the two events together with land-use data, maps of SO$_2$ emission and natural emission of monoterpenes. The analysis clearly shows that the observed air masses during the two dust events originated from the Taklimakan and Gobi Deserts and carried with the anthropogenic pollutants from the North China plain and eastern China to southern China. The time series in Fig. S5 show relatively high concentrations of anthropogenic pollutants (e.g. sulfate and BC in PM$_{2.5}$) during the dust events, suggesting a strong mix of dust and polluted plumes. This kind of mixed plumes provided abundant reactive species and water, which favored aging of the dust particles to form secondary coating (see Fig. S6), and induced heterogeneous photochemical reactions during the long range transport[25]. For example, the photodecomposition of coated nitrate may release NO$_3$ radical to oxidize some specific VOCs[44] (Detailed descriptions are in the SI).

Discussion

Our investigation provides "direct" observational evidence on new particle formation and growth in heavy dust plumes mixed with anthropogenic pollution, and suggests an unexpected source of nucleating and condensable vapors via dust-induced heterogeneous photochemical processes. In Asia, huge amounts of dust is being produced during the dry season from both Gobi and Taklimakan
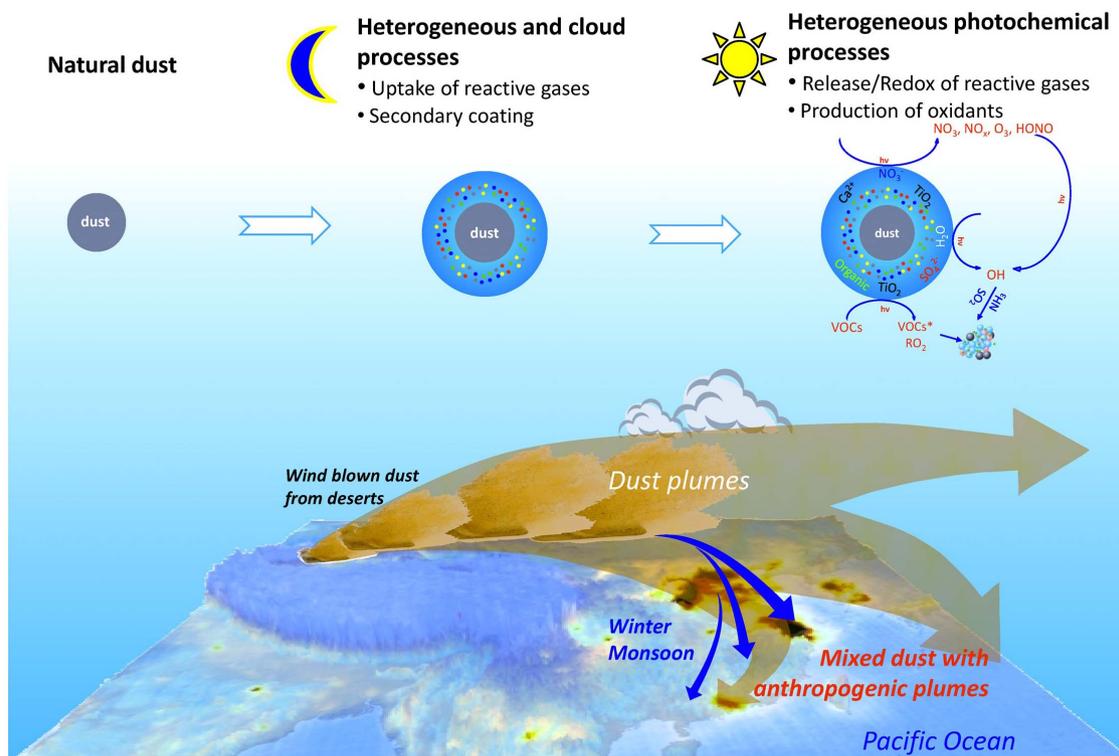
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Figure 4 | Schematic description of main stages of Asian dusts during the long range transport in the atmosphere and their main transport pathways. Stage I-Mineral dusts are injected into the atmosphere in the remote area. Stage II - dust particles uptake anthropogenic reactive gases and form secondary coatings after them transported over area with high anthropogenic emissions. Heterogeneous photochemical processes should play a role in this stage, but easily be covered up by strong gas phase photochemistry. Stage III - aged dusts transport to remote Asia-Pacific region, where the plumes experience heterogeneous photochemical reactions favoring the new particle formation and growth. The map in the figure was drawn by Global Mapper.

deserts[45,46], and the dust is being transported eastward across the polluted northern and eastern parts of China (Fig. 4). The mixture of dust and anthropogenic pollution plumes can be further transported easterly in the mid-troposphere and southerly in the lower-troposphere, causing significant impacts on the remote atmosphere in downwind areas like the Pacific Ocean even other continents in the world[47,48] (Fig. 4). Given that the polluted dust promotes new particle formation and growth, this study suggests a more crucial role of dust in both regional and global climate change than the current knowledge suggests. In addition, the observation of dust-promoted particle nucleation and growth, along with HONO production, provide strong evidence on the role of aerosol-related photochemistry, which would enhance the atmospheric oxidation capacity via photo-induced surface redox reactions and gaseous radical production (Fig. 4). These findings challenge the traditional point of view that aerosols are net sinks for the atmospheric oxidation capacity, and indicate a possible pathway for the large missing source of OH radical in heavy polluted regions[49].

Methods

Field experiment. Mount Heng located in the middle of Hunan Province, southern China, is a remote mountain area with few local sources around but generally downwind both source regions of Asia dusts and anthropogenic pollutions in the spring (Fig. S5). The field campaign was conducted March to May in 2009 at a meteorological station on the summit of Mt. Heng. Comprehensive parameters of trace gases and aerosol components, including SO_2, O_3, $PM_{2.5}$, PM_{10}, sulfate, calcium, BC in $PM_{2.5}$, were continuously measured during the campaign. Particle number size distributions in the range of 10–10000 nm were employed to detect the particle nucleation and growth. Several filter based samples were also collected and analyzed to assist interpreting the observed phenomena. Detailed information is provided in the SI.

Numerical calculation and modeling. Methods to calculate the particle formation rates, growth rates, and condensation sink (CS) are followed the suggestion of Kulmala et al.[50]. Lagrangian particle dispersion modeling (LPDM) was carried out based on a method developed and evaluated by Ding et al.[32] using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. For each hour, 3000 particles were released at the site and were traced backward for a 7-day period. The residence time at 100 m altitude, i.e. footprint retroplumes, were used to understand the contribution from potential source regions.

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Author contributions

A.D., T.W., W.W. and M.K. lead the overall scientific questions. W.N. participated in the field measurement and contributed to data analysis. V.K., C.G., T.P., X.Y., C.F. and M.K. involved in initial discussions, and commented and edited the manuscript. X.Q. contributed to the data analysis. L.X., Q.Z., X.G. and X.W. participated the campaign and operated some trace gases and PM instruments. Correspondence and requests for materials should be addressed to A.D. (dingaj@nju.edu.cn) or T.W. (tao.wang@polyu.edu.hk)

Additional information

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