Possible warming effect of fine particulate matter in the atmosphere

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Particulate matter emitted through human activities not only pollutes the air, but also cools the Earth by scattering shortwave solar radiation. However, coarser dust particles have been found to exert a warming effect that could, to some extent compensate for the cooling effect of fine dust. Here we investigate the radiative effects of sulfate containing aerosols of various sizes and core/shell structures using Mie scattering and three-dimensional finite difference time domain simulations of the electromagnetic fields inside and around particulate matter particles. We find that not only coarse dust, but also fine non-light-absorbing inorganic aerosols such as sulfate can have a warming effect. Specifically, although the opacity of fine particles decreases at longer wavelengths, they can strongly absorb and re-emit thermal radiation under resonance conditions at long wavelength. We suggest that these effects need to be taken into account when assessing the contribution of aerosols to climate change.
During the last decade, global warming has led to climate change and has triggered more extreme weather events, hotter heat waves, drier droughts, colder winters, and larger storm surges, endangering the survival of flora and fauna on Earth, including human beings. To achieve the Paris Agreement, it will be necessary to strictly limit the total amount of cumulative carbon emissions\textsuperscript{5–8}. On the other hand, particulate matter (PM) generated by human activities not only pollute the air, but also affect Earth’s radiation budget\textsuperscript{2}. The radiative effect of PM is the opposite of that of greenhouse gases. The direct scattering of short wavelength solar radiation by anthropogenic PM particles increases planetary albedo, thereby exerting a cooling influence on the planet\textsuperscript{1,11,13}. The global mean radiative forcing due to aerosol–radiation interactions is $\sim0.35$ W m$^{-2}$; aerosol–cloud interactions is $\sim0.45$ W m$^{-2}$; greenhouse gases is $\sim+2.83$ W m$^{-2}$\textsuperscript{12}.

Anthropogenic PM includes secondary organic aerosols and secondary inorganic aerosols (SIA), the main components of SIA are sulfate and nitrate\textsuperscript{13–15}. SIA particles are taken as the portion of Aitken and Accumulation mode particles\textsuperscript{16,17}. In previous studies, it was concluded that sulfate also absorbs longwave radiation, but the aerosol radiative effect is generally small because the opacity of aerosols decreases in longer wavelengths of infrared (IR) regime\textsuperscript{11}. The major natural sources of ambient PM, such as marine aerosol, mineral dust, Saharan dust, smoke plumes, and their mixtures have been found to enhance the greenhouse effect\textsuperscript{18–24}. Recently literature found that the dust in the atmosphere is substantially coarser than represented in current global climate models. They also found the contribution of very coarse dust (diameter $\geq5$ µm) was underestimated, and the coarse particles could cause warming of the atmosphere\textsuperscript{24}. More recently, they stated that climate models consistently underestimate the amount of coarse dust in the atmosphere, and the climate model must be revised\textsuperscript{25}. The improved description of the dust size distribution and its vertical distribution, as well as the IR optical properties, are important for a reliable estimate of its radiation effects\textsuperscript{23}.

The atmosphere often contains a mixture of different types of PM particles, and the mixing process leads to change in sulfate and mineral dust loads, thus altering their impact on the atmospheric radiation. Transmission electron microscopy (TEM) has been used to observe the size, structure and composition of individual fine particles, which is essential for assessing the radiation properties of PM particles and understanding their climatic effects\textsuperscript{26,27}. It has been observed that mineral dust particles often get coated with sulfates and other soluble materials\textsuperscript{28}. The fine (PM$_{2.5}$) and coarse (PM$_{10}$) particles were intended to apply to the two major atmospheric particle distributions which overlap in the intermodal size range between 1 and 3 µm\textsuperscript{17,29}. The interaction of the composite PM particles with atmospheric radiation may be different from that of homogeneous PM particles. Theoretical analysis of the relationship between various types of PM mixtures, particles sizes, absorption coefficients, and wavelength are important for the investigation on radiative properties and climate modeling. The atmospheric window offers a direct and strong driving force to dissipate heat into space in the form of thermal radiation under appropriate atmospheric conditions\textsuperscript{30–34}. The application of radiative cooling as a passive cooling option for buildings has a great potential to compensate the problems raised by global warming\textsuperscript{35,36}. The PM in the atmospheric window would affect the absorption and scattering effects of thermal radiation\textsuperscript{37}.

In this paper, we discuss the thermal radiation properties of sulfate and SO$_4^2$-sulfate core/shell mixture particles in atmospheric window, including ED and MD resonance behavior. We also used the three-dimensional finite difference time domain (3D-FDTD) method to simulate the electromagnetic fields inside and around PM particles with various sizes and core/shell structures. In this study, we used the World Health Organization air quality guidelines values to simplify the calculation of thermal radiation power density changes in a clear atmosphere with and without PM particles. These calculations of power density changes preclude any differences in geographical and seasonal distributions. From the conceptual experimental results, it was found that different optical properties related to particle size have a significant effect on the thermal emissivity, potentially affecting the temperature of the surrounding environment.

### Results and discussion

The atmosphere has a highly transparent window for IR wavelengths in the range from 8 to 13 µm (the so-called first infrared atmospheric window (IRW1)) and a relatively transparent window for IR wavelengths in the range from 16 to 24 µm (the so-called second infrared atmospheric window (IRW2)); they are highlighted by the light-cyan and yellow areas, respectively, in Fig. 1a. According to Planck’s law and Wien’s displacement law, when the surface temperature is 250, 300, and 350 K, the corresponding maximum wavelengths $\lambda_{\text{max}}$ of the thermal radiation are 11.51, 9.50, and 8.25 µm, respectively. These thermal radiation peaks from the Earth’s surface coincidentally fall within the IRW1. For these same temperatures, ~31, 38, and 41%, respectively, of the thermal radiation energy distribution is concentrated in IRW1, and ~29, 24, and 20%, respectively, in IRW2. The higher the surface temperature, the greater the effect of IRW1 on thermal radiation; furthermore, the effect of IRW1 will also be greater than that of IRW2. This feature of IRW1 permits heat to dissipate through thermal radiation emission, with the potential to provide a passive cooling mechanism for the Earth.

A recent report displays that the main constituents of both PM$_{10}$ and PM$_{2.5}$ particles are generally organic matter, sulfate, and nitrate\textsuperscript{38}. Figure 1b–e present the IR absorbance spectra of the major components of SIA—(NH$_4$)$_2$SO$_4$, NH$_2$NO$_3$, Na$_2$SO$_4$, and NaNO$_3$, respectively—in the wavelength range from 4 to 20 µm\textsuperscript{19}. Table S1 of Supplementary Note 1 lists the IR absorption bands of (NH$_4$)$_2$SO$_4$ and Na$_2$SO$_4$ in IRW1\textsuperscript{40–42}. The $\nu_3$(SO$_4^2$-) modes of (NH$_4$)$_2$SO$_4$ and Na$_2$SO$_4$ are strong absorption bands in IRW1 and would block heat transfer of thermal radiation to space. The radiative forcing caused by sulfate from fossil fuel and biofuel emissions has been estimated to be ~0.4 W m$^{-2}$ (range: ~0.6 to ~0.2 W m$^{-2}$); for nitrate, it is estimated to be ~0.11 W m$^{-2}$ (range: ~0.3 to ~0.03 W m$^{-2}$); the value for ammonium is included within the sulfate and nitrate estimates\textsuperscript{2}. The effect of PM on global climate is uncertain because the effect of PM can be to cool or to warm, depending on its optical properties. In this study, we chose (NH$_4$)$_2$SO$_4$ for analysis because it is one of the major species of SIA, and because it features a strong absorption band and induces scattering in IRW1.

### Comparison of absorption efficiency ($Q_a$) for different sizes of sulfate particles

Figure 2a, b presents the absorption cross-section ($C_a$) and absorption efficiency ($Q_a$) [where $Q_a$ is the normalized $C_a$ to the cross-section area of the particles] contours of (NH$_4$)$_2$SO$_4$ spheres, calculated by Mie theory, for wavelengths in the range from 4 to 20 µm, and for particle diameters ($D_p$) ranging from 0.1 to 10 µm (see Supplementary Note 2 for definitions of $Q_a$ and $C_a$). The value of $C_a$ becomes larger as the particle size increased, and is more obvious than 5 µm. In the normalized $Q_a$ contour, all of the particle sizes resulted in three bands, corresponding to the three IR active vibration modes, at wavelengths of ~7, 9, and 16 µm. Two strong absorption bands...
for the particles having diameters of ~2 and 4 µm appeared at wavelengths of 9 and 16 µm, respectively, corresponding to the ED resonance of the (NH₄)₂SO₄ particles. The two strong absorption bands are included within IRW₁ and IRW₂, suggesting that these particles would greatly decrease the transparency of these IR windows—and intensify the greenhouse effect.

Figure 2c, d present the \( Q_a \) of (NH₄)₂SO₄ particles having values of \( D_p \) of 0.5, 2, and 2.5 µm (PM₂.₅) and 5, 8, and 10 µm (PM₁₀₋₂.₅), respectively. In Fig. 2c, the particle having a value of \( D_p \) of 2 µm gave the largest value of \( Q_a \) because of ED resonance at a wavelength of ~9 µm. In Fig. 2d, the values of \( Q_a \) of the particles having diameters of 5, 8, and 10 µm were not significantly different, but the absorption band became broader as the size increased. When the size of a particle increases, higher-order modes of resonance would dominate the \( Q_a \) spectrum. Further information about the values of \( Q_a \) for particles having different sizes is provided in the Supporting Information (see Supplementary Note 3 and Fig. S1 for detailed contribution of 2n-poles analysis).

To verify the influence of the PM with different particle sizes in the IRW, we calculated (Fig. 2e) the maximum value of \( Q_a \) in IRW₂, and the average value of \( Q_a \) in IRW₂. In the range of IRW₁, the maximum value of \( Q_a \) for particle size \( D_p \) of 2 µm is 2.46. This \( Q_a \) peak arose from the ED resonance of the 2-µm particle. The average value of \( Q_a \) in IRW₁ and IRW₂ can represent a particle’s influence on thermal radiation for the overall IRW₁ and IRW₂ absorption, respectively. Because resonance of high-order modes broadens the \( Q_a \) peak, the average values of \( Q_a \) in both IRW₁ and IRW₂ gradually increased upon increasing the value of \( D_p \).

In general, atmospheric particles are normally classified into different modes according to their size, formation processes, and atmospheric age. Particles with a size between 0.01 and 0.1 µm (Aitken mode) account for the majority of the number of atmospheric particles, but they rarely account for more than a few percent of the total mass. Particles with diameters between 0.1 and 1.0 µm (accumulation mode) usually account for a large portion of the total aerosol mass and have the largest surface area. This makes these particles performing significant importance to atmospheric chemistry. Particles <1.0 µm contribute to the optical properties of atmospheric aerosols. Although the particles with larger values of \( D_p \) provided higher average value of \( Q_a \), the number of particles would decrease upon increasing the value of \( D_p \). Both the

Fig. 1 Atmospheric IR absorption spectra and infrared absorption spectra of sulfate and nitrate. a Black-body emission curves of the Earth at various temperatures (black curves), with atmospheric IR absorption spectra at an air mass of 1.0 (gray area) as a reference. b-e IR absorption spectra of ammonium sulfate (NH₄)₂SO₄, sodium sulfate (Na₂SO₄), ammonium nitrate (NH₃NO₃), and sodium nitrate (NaNO₃) in the wavelength range from 4 to 20 µm (Recalculated from Sigma Library of FT-IR Spectra).
number of particles and the value of $Q_a$ should be considered when calculating the absorptance of these particles. We consider the number of particles and the value of $C_a$ to calculate the absorptance of these particles with different sizes. At the same mass concentration, the number of particles is proportional to $\frac{1}{D_p^2}$, and the surface area is proportional to $D_p^2$. We define $\frac{1}{D_p^2}$ as the particle number effect ($E_p$). The values of $Q_a \times E_p$ can indicate the absorption capacity of particles with different sizes under the same mass concentration. Figure 2f displays the tendencies of the average value of $Q_a$, the particle number effect ($E_p$), and the values of $Q_a \times E_p$ in IRW1 for various values of $D_p$.

3D-FDTD simulations of the optical properties of SiO$_2$/sulfate core/shell mixtures. In the same way, we also analyzed the most
abundant compound in the Earth’s crust, silicon dioxide (SiO$_2$).

The maximum value of $Q_a$ appears around $D_p$ of 2.5 µm at the
wavelength of ca. 9 µm. The coating of atmospheric particles is
often resulting from enhanced hygroscopicity due to aging
mechanisms such as condensation, condensation, and hetero-
geneous chemical reactions$^{28,43}$. To further investigate the optical
behavior of PM, we used the 3D-FDTD method to simulate the
electromagnetic field inside and around particles of various values
of $D_p$. Aged PM may be fractionally or entirely encapsulated
within a nearly spherical shell. We studied the optical properties
of homogeneous sulfate and SiO$_2$/sulfate core/shell mixtures of
PM (see Supplementary Note 4 and Fig. S2 for detailed homo-
geneous sulfate PM analysis). According to transmission electron
microscope images$^{43}$, we assumed that silica or SiO$_2$ formed the
core, coated by a concentric spherical shell of (NH$_4$)$_2$SO$_4$. Fig. 3a
provides a schematic representation. The radius of the particle
was 0.02, 0.25, 1, or 4 µm; the core/shell ratio was set at 0.5 and
the core radius was 0.01, 0.125, 0.5, or 2 µm. Figure 3b presents
the values of $Q_a$ of core/shell particles having values of $D_p$ of 0.04, 0.5, 2, and 8 µm. Fig. 3c–j present 3D-FDTD simulations results of the normalized electric field intensities ($|E/E_0|^2$) and normalized absorption rates per unit volume ($P_{abs} = 0.5\omega\varepsilon_0^2|E/E_0|^2$ with the unit of W/m$^3$ (V/m)$^2$) of the
core/shell particle having sizes of 0.04, 0.5, 2, and 8 µm. All of the
simulation results are normalized by the electric field intensity
(with the unit (V/m)$^2$) of the incident light source. For the electric
field distribution of the particle having a value of $D_p$ of 0.04 and
0.5 µm (Fig. 3c, d), the shell thickness (0.01 µm and 0.125 µm) was
smaller than the penetration depth of (NH$_4$)$_2$SO$_4$
($d_p = \frac{\lambda}{4\pi} = 0.43$µm), such that the light field could penetrate
partially into the SiO$_2$ core. The extinction coefficient of SiO$_2$ at a
wavelength of 9 µm ($k = 0.759$) is, however, smaller than that of
(NH$_4$)$_2$SO$_4$ ($k = 1.665$). The penetrated light field and the lower
extinction coefficient of SiO$_2$ result in a small absorption rate
inside the SiO$_2$ core (Fig. 3g, h). The electric field distribution in
(Fig. 3c, d) clearly reveal that although the electric field distributed
on the outer edge of the particle shell, the area of spatial
distribution of the high electric field regime for the particle of
0.5 µm is much larger than that of 0.04 µm. In Fig. 3e, i, the electric field distribution of the particle having a value of $D_p$ of 2 µm reveals a dipole resonant pattern similar to that in the
homogeneous case in Fig. S2. Because of the high absorption rate
and small penetration depth of (NH$_4$)$_2$SO$_4$ particle having a value

![Fig. 3 The $Q_a$ spectra and simulated E-field intensities and absorption rates of 0.04, 0.5, 2, and 8 µm core/shell structure particles. a Schematic representation of the core/shell structure, with SiO$_2$ as the core surrounded by a concentric spherical shell of (NH$_4$)$_2$SO$_4$. Here, the core/shell ratio is set at 0.5. b $Q_a$ spectra of core/shell structures having values of $D_p$ of 0.04, 0.5, 2, and 8 µm. c-f Simulated E-field intensities and g-j simulated absorption rates of c, g 0.04-, d, h 0.5-, e, i 2-, and f, j 8-µm core/shell structure particles.](https://doi.org/10.1038/s43247-021-00278-5)
Comparison of atmospheric transmittance under clear sky and presence of PM. Figure 4 shows the atmospheric transmittance in the absence (clear sky) and presence of PM, including homogeneous sulfate and sulfate/SiO$_2$ core/shell mixtures at different concentrations. It can be clearly observed from Fig. 4a, b that the concentration of PM increases to 20, 50, and 1,180 µg m$^{-3}$, the atmospheric transmittance decreased, especially in IRW1 (light-cyan areas). Sulfate has a strong absorption band around the wavelength of 9 µm, and the decrease in transmittance can be clearly observed. As far as we known, the injection of sulfate particle into the stratosphere is the most discussed proposal for solar geoeengineering. However, in addition to the risk of ozone loss and heating of the lower tropical stratosphere$^{44}$, it is also likely to absorb the earth’s radiation and warm the earth’s surface.

Conceptual experimental results. To confirm the thermal radiation behavior of (NH$_4$)$_2$SO$_4$ experimentally, we set up an apparatus to conduct a conceptual experiment in the laboratory. We prepared several samples containing 0.1 wt% (NH$_4$)$_2$SO$_4$ in KBr, and measured the cooling rate of different samples shielding (see Supplementary Note 10 and 11 for detailed experimental concept analysis). We found that the larger the shielding area, the slower the cooling rate. From this experiment, we suggest that the thermal radiation absorption of PM in the atmosphere would increase the temperature of the surroundings. The thermal emissivity of the samples composed of different (NH$_4$)$_2$SO$_4$ particle sizes was also measured. Different optical properties related to particle size have a significant effect on the thermal emissivity.

Conclusions

According to our findings in this study, sulfate displays strong IR absorption, especially in the atmospheric window. The IR absorption properties of (NH$_4$)$_2$SO$_4$ make it behave like “greenhouse PM” producing positive radiative forcing that may increase the temperature of its surroundings. An increase in the concentration of sulfate would reflect more of the incoming solar radiation back into space, but it would also capture more of the outgoing thermal radiation back to the Earth. (NH$_4$)$_2$SO$_4$ and sulfate/SiO$_2$ core/shell particles having diameter of ~2 µm display the maximum absorption efficiency factor ($Q_s$) of 2.46 and 2.54, respectively, at a wavelength of ca. 9 µm. At the same concentration, considering the factors of particle size and number together, the size of (NH$_4$)$_2$SO$_4$ particles with the strongest absorption capacity appears at 1.4 µm. The particle size also has an important influence on the scattering efficiency factor ($Q_s$), as the particle size decreases, the scattering phenomenon of

Table 1 The changes in the thermal radiation power density by different cases.

| Case | Type | Concentration (µg/m$^3$) | $P_{atm}$ (W m$^{-2}$) | $\Delta P_{atm}$ (W m$^{-2}$) |
|------|------|-------------------------|------------------------|-----------------------------|
| 1    | Homogeneous | 20                      | 324.61                 | 1.19                        |
| 2    | Homogeneous | 50                      | 326.24                 | 2.83                        |
| 3    | Homogeneous | 1180                    | 350.43                 | 27.01                       |
| 4    | Core/shell | 20                      | 324.68                 | 1.26                        |
| 5    | Core/shell | 50                      | 326.43                 | 3.02                        |
| 6    | Core/shell | 1180                    | 355.87                 | 32.46                       |

*Presented the changes in the thermal radiation power density in atmosphere containing PM of various sizes and concentrations, including homogeneous sulfate and sulfate/SiO$_2$ core/shell mixtures.*
(NH₄)₂SO₄ gradually becomes dominated by BWQs. The BWQs would drive the thermal radiation scattered back from the particles of the PM, thereby decreasing the thermal emission going out to the space beyond the atmosphere. The absorption of particles at resonant conditions would be increased, even if the sizes of particles are much smaller than the wavelength. The relationship of particles size, absorption coefficients, and wavelength effect is very important for the investigation of resonance phenomena on particles. The simplified k distribution would lose the dependence of calculation results on wavelength. In this study, we used the LBL method to observe the resonance effect of PM particles with different sizes in broadband of spectral regime from 4 to 20 µm.

It is extremely difficult to describe the mixing state of an PM particle. In this study, we use simplified assumptions such as homogeneous (NH₄)₂SO₄ and sulfate/SiO₂ core/shell particles to discuss the effect of sulfate on thermal radiation absorption in IRW1 under resonance conditions. Once the optical properties of various materials are known individually, the optical properties of mixed PM can be calculated and correctly averaged. To evaluate how the longwave aerosol radiative effect varies as a function of size distribution and aerosol burden, we also briefly analyze the dependence of the longwave radiative effect as a function of median radius, distribution width, and sulfate/SiO₂ weight fraction to discuss the use of statistics data and treatment of uncertainties (see Supplementary Note 12 for a detailed discussion of...
this study, and we will analyze more PM species in future work and also investigate the LWPMRE of the aging process of the PM particles.

Methods

Sample fabrication. (NH₄)₂SO₄, KBr and KCl were purchased from Sigma–Aldrich. The (NH₄)₂SO₄ and KBr particles were crushed to sizes of several hundreds of micrometers, using a bead crushing machine (Beads Crusher GT-12), and then ground in an agate mortar to sizes of a few micrometers. The (NH₄)₂SO₄ (particle sizes: ca. 2–20 μm) were mixed into KBr at a content of 0.1 wt%; the mixed powder was pressed into several tablets, using a tablet press. The (NH₄)₂SO₄ particles were observed through scanning electron microscopy (SEM, NOVA 450). Fourier transform infrared spectroscopy (FTIR) was performed using a Fourier transform infrared spectrometer (Bruker VERTEX 70). The emissivity spectra of the samples in the mid- and far-infrared wavelengths (2.5–25.0 μm) were measured using the emissivity measurement kit (Emission Adapter, A540) and MCT detector of FTIR (Vertex 70, Bruker), and the blackbody furnace (IR 563/ 301. Blackbody system) was used as a correction source for the emissivity spectra. Temperature measurement data were recorded using K-type thermal couples and a paperless high-speed recorder (KR2000 Series Graphic Recorder).

Simulation method. High-resolution FTIR transmittance spectral data of (NH₄)₂SO₄, NaN₃, NH₄NO₃, and NaNO₃ were obtained from the Sigma Libra of FT-IR Spectra. The optical constant of (NH₄)₂SO₄ used in the simulations at a wavelength (λ) of 9 μm was 0.99 ± 1.7i. Mie theory was used to simulate the thermal radiation properties of the (NH₄)₂SO₄ particles. The absorption cross-section (α₀) and the scattering cross-section (σ₀) were computed numerically using the Matlab code. The electric field distribution of the (NH₄)₂SO₄ particles was simulated using the 3D-FDTD method (RSoft FullWAVE).

Data availability

We did not use dataset. The raw data can be obtained from the following link (https://github.com/Gibbs9487/data_of_PM) without restrictions. There are no restrictions on data availability. These data can be supplied as Supplementary Information, for reader convenience. Please contact us if you have any questions.

Code availability

The FDTD data is collected from commercial software (RSoft Fullwave). The data of Mie solution is performed and collected by home made MATLAB code. The analysis of particle size is performed by ImageJ. The Code can be obtained from the corresponding author upon request.

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Author contributions
S.-L.C. and H.-L.C. conceived the study and analyzed main results. S.-L.C. wrote the paper. S.-W.C. contributed data and analysis. Y.-J.C. assists in the measurement of thermal emissivity in supplementary note 11. All authors contributed to data analysis and scientific discussion.

Competing interests
The authors declare no competing interests.

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