

A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle

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Mineral dust aerosols impact Earth's radiation budget through interactions with clouds, ecosystems, and radiation, which constitutes a substantial uncertainty in understanding past and predicting future climate changes. One of the causes of this large uncertainty is that the size distribution of emitted dust aerosols is poorly understood. The present study shows that regional and global circulation models (GCMs) overestimate the emitted fraction of clay aerosols (<2 μm diameter) by a factor of $\sim 2\text{--}8$ relative to measurements. This discrepancy is resolved by deriving a simple theoretical expression of the emitted dust size distribution that is in excellent agreement with measurements. This expression is based on the physics of the scale-invariant fragmentation of brittle materials, which is shown to be applicable to dust emission. Because clay aerosols produce a strong radiative cooling, the overestimation of the clay fraction causes GCMs to also overestimate the radiative cooling of a given quantity of emitted dust. On local and regional scales, this affects the magnitude and possibly the sign of the dust radiative forcing, with implications for numerical weather forecasting and regional climate predictions in dusty regions. On a global scale, the dust cycle in most GCMs is tuned to match radiative measurements, such that the overestimation of the radiative cooling of a given quantity of emitted dust has likely caused GCMs to underestimate the global dust emission rate. This implies that the deposition flux of dust and its fertilizing effects on ecosystems may be substantially larger than thought.

direct radiative forcing | scale invariance | aeolian saltation | dust storms | wind erosion

Mineral dust aerosols eroded from arid soils impact weather and climate by scattering and absorbing radiation (1–4) and by modifying cloud properties (1, 5). Deposition of dust aerosols also partially controls the productivity and carbon sequestration of ocean ecosystems by providing limiting micronutrients such as iron, which affects atmospheric concentrations of greenhouse gases (6). The total impact of dust aerosols on Earth's radiative budget constitutes an important uncertainty in understanding past and predicting future climate changes (1, 6–8). In addition, dust aerosols adversely affect human health (9) and could suppress hurricane activity (10).

All these processes depend on the size of the atmospheric dust aerosols (2–5), which also determines their lifetime (3). But current treatments of the particle size distribution (PSD) of emitted dust aerosols in global circulation models (GCMs) are based on empirical relations with limited or no physical basis (3, 11–13). This use of empirical relations is necessary both because of the scarcity of measurements (14, 15) and because the understanding of the physical processes that determine the emitted dust PSD is very limited (16, 17). As a consequence, the fraction of emitted dust aerosols in the clay size range (<2 μm diameter), which both interact most efficiently with shortwave (solar) radiation and have the longest lifetime, differs by up to a factor of 4 between GCMs (3, 11–13, 18–20).

The availability of an accurate expression for the emitted dust PSD could thus reduce the uncertainty on GCM estimates of dust climate forcing. The present study derives such an expression from the analogy between the fragmentation of soil dust aggregates and the much better understood fragmentation of brittle materials such as glass (21). The resulting theoretical expression for the emitted dust PSD is in excellent agreement with measurements. In contrast, GCMs overestimate the emitted fraction of clay aerosols by a factor of $\sim 2\text{--}8$, with implications for simulations of the spatial distribution, radiative forcing, and global emission rate of dust aerosols.

The Physics of Dust Emission

Dust aerosols that undergo long-range transport predominantly have diameters smaller than 20 μm (22) and are denoted here as PM₂₀ dust. The cohesive forces on such small particles in soils are generally much larger than aerodynamic forces (22), thereby preventing PM₂₀ dust from being lifted directly by wind (15, 22). Moreover, these strong cohesive forces cause PM₂₀ dust to rarely occur loosely in soils because they easily attach to other particles, thereby forming dust aggregates of larger sizes (17, 23).

Instead of being lifted directly by wind, PM₂₀ dust is generally emitted by an intermediary process called saltation (22, 24). In saltation, larger sand-sized particles [$\sim 70\text{--}500 \mu\text{m}$ (22)], which are more easily lifted by wind because their cohesive forces are small compared to aerodynamic forces (22), move in ballistic trajectories (24). Upon impact on the soil bed, these saltating particles can eject dust particles from dust aggregates in the soil (22), a process known as sandblasting (22, 23). In addition, some saltating particles are sand-sized aggregates of dust particles, which can fragment and emit dust aerosols upon striking the surface (17).

Although the processes leading to dust emission are qualitatively understood, a detailed quantitative understanding is hindered by the large, highly variable, and poorly understood cohesive forces on PM₂₀ dust in soils (16, 17, 22). As a consequence, previous physically based theories of dust emission that account for cohesive forces (16, 17) have large uncertainties in their input parameters and can differ greatly from measurements (17). I therefore take a different approach here and utilize the closest analog problem to dust emission that is quantitatively understood: the fragmentation of brittle materials (21, 25).

The Fragmentation of Brittle Materials

When a brittle material such as glass or gypsum receives a large input of energy, for example by being dropped on a hard surface

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is further justified by the occurrence of scale invariance in the emission of dust aerosols (see below), which is unique to the fragmentation regime (27).

Fragmentation of Dust Aggregates. Although the physics of dust aggregate fragmentation is expected to be similar to the fragmentation of conventional brittle materials such as glass, there is one important difference: the size of the indivisible constituents. In conventional brittle materials, these indivisible constituents are usually individual molecules or crystal units with sizes on the order of 10^{-10} – 10^{-9} m, such that the small-size deviation from the power law of **1** due to the presence of indivisible constituents is only rarely observed (29). However, the indivisible constituents of dust aggregates are the discrete dust particles with much larger typical sizes of 10^{-7} – 10^{-5} m (16, 17, 23). As a consequence, we expect the small-size deviation from the power law to occur at a typical fragment size of ~ 1 μm .

Many dust aerosols are aggregates themselves (17, 23, 37), such that an emitted dust aerosol consists of one or more soil particles of equal or smaller size. The production of dust aerosols with size D_d is thus proportional to the volume fraction of soil particles with size $D_s \leq D_d$ that contribute to the formation of these aerosols. That is, if the fraction of soil particles with size $D_s \leq D_d$ is doubled, then the production of aerosols with size D_d will be doubled as well, provided that the shape of the soil PSD remains constant. I therefore neglect any influence of changes in the shape of the soil PSD and assume that the production of aerosols of size D_d is proportional to the volume fraction of soil particles with size $D_s \leq D_d$,

$$\frac{dN_d}{d \ln D_d} \propto \int_0^{D_d} P_s(D_s) dD_s, \quad [3]$$

where N_d is the normalized number of emitted dust aerosols with size D_d , and P_s is the fully dispersed PSD of PM20 soil particles. The distribution of fully disaggregated soil particles is usually described as a log-normal distribution (25):

$$P_s(D_s) = \frac{1}{\sqrt{2\pi} \ln(\sigma_s)} \exp\left\{-\frac{\ln^2(D_s/\bar{D}_s)}{2 \ln^2(\sigma_s)}\right\}, \quad [4]$$

where σ_s is the geometric standard deviation, and \bar{D}_s is the median diameter by volume (2). The number and volume size distributions of emitted dust aerosols are then obtained by combining 2–4,

$$\frac{dN_d}{d \ln D_d} = \frac{1}{c_N D_d^2} \left[1 + \operatorname{erf}\left(\frac{\ln(D_d/\bar{D}_s)}{\sqrt{2} \ln \sigma_s}\right) \right] \exp\left[-\left(\frac{D_d}{\lambda}\right)^3\right], \quad [5]$$

$$\frac{dV_d}{d \ln D_d} = \frac{D_d}{c_V} \left[1 + \operatorname{erf}\left(\frac{\ln(D_d/\bar{D}_s)}{\sqrt{2} \ln \sigma_s}\right) \right] \exp\left[-\left(\frac{D_d}{\lambda}\right)^3\right], \quad [6]$$

where V_d is the normalized volume of dust aerosols with size D_d , c_N and c_V are normalization constants, and erf is the error function (see p. 423 of ref. 2).

Note that Eqs. 5 and 6 are applicable only when dust emission is predominantly due to the fragmentation of soil aggregates. Eqs. 5 and 6 are, for example, not valid for (i) aerodynamically lifted dust (22), (ii) dust emitted mainly by impacts in the damage regime, which could occur for very cohesive soils, and (iii) dust with diameters larger than ~ 20 μm , which are more likely to occur as loose particles in the soil (17, 23), such that their emission is not always due to fragmenting impacts.

The Fully Dispersed PSD of Arid Soils. Eqs. 5 and 6 depend on the log-normal distribution parameters of fully dispersed soil particles with sizes extending from the submicron range up to 20 μm . Although measurements of soil PSDs extending into the submicron range have been published (38, 39), there are not nearly enough measurements to parameterize a spatially varying soil PSD in GCMs. To circumvent this problem, we must define a “typical” PM20 arid soil PSD.

Parameterizing a typical PM20 arid soil PSD is difficult, however, because there are few published arid soil PSDs that extend into the submicron range. A notable exception is the study of d’Almeida and Schütz (40), who reported such PSDs for six arid soils from across the Sahara (Table 1). These authors used wet sieving and optical and electron microscopy to determine the soil PSD from ~ 0.01 – $1,000$ μm . Although the water in which d’Almeida and Schütz suspended their samples dispersed the soil to some degree, they did not fully disperse their samples, for example, by ultrasonic shaking or using a dispersing agent (38, 39). However, measurements suggest that the difference between the fully dispersed soil PSD and that obtained from suspension in water could be limited, especially in the clay size range (41).

I thus parameterize a typical PM20 arid soil PSD from the measurements of d’Almeida and Schütz (40), supplemented by laser diffraction measurements of actively eroding soils in Utah (42). A least-squares fitting technique was used to determine the most likely values of the log-normal parameters \bar{D}_s and σ_s for each individual soil (Table 1). The values of these parameters are surprisingly uniform, despite the wide range of soil textures and geographical locations represented. The average and standard deviation of the log-normal parameters of the eight soil PSDs are $\bar{D}_s = 3.4 \pm 1.9$ μm and $\sigma_s = 3.0 \pm 0.4$, and the log-normal PSD with these average parameters appears to be a reasonable approximation to the eight available arid soil PSDs (see Fig. S1). However, the accuracy of the average values of \bar{D}_s and σ_s estimated in Table 1 remains uncertain because six of the eight soil PSDs are not fully dispersed (40) and because of the small number of soils represented.

Measurements of the Vertical Dust Flux. Testing the validity of Eqs. 5 and 6 requires measurements of the emitted dust PSD. Although measurements of the PSD of atmospheric dust aerosols are relatively abundant in the literature (37, 40, 43, 44), these measure-

Table 1. Values of the log-normal soil PSD parameters \bar{D}_s and σ_s

Study	Soil number	Soil texture	Geographical location	Best fit \bar{D}_s , μm	Best fit σ_s
d’Almeida and Schütz (1983, ref. 40)	1	Loam	Mali	2.6	2.9
d’Almeida and Schütz (1983, ref. 40)	2	Silt	Senegal	1.6	3.4
d’Almeida and Schütz (1983, ref. 40)	3	Sand	Mali	1.7	2.8
d’Almeida and Schütz (1983, ref. 40)	4	Loamy sand	Algeria	7.2	3.7
d’Almeida and Schütz (1983, ref. 40)	5	Sand	Niger	2.1	2.9
d’Almeida and Schütz (1983, ref. 40)	6	Sandy loam	Sudan	4.9	2.7
Goldstein et al. (2005, ref. 42)	00-U36	Sand	Utah	3.0	2.8
Goldstein et al. (2005, ref. 42)	00-U37	Loam	Utah	3.8	2.8
Average and standard deviation				3.4 ± 1.9	3.0 ± 0.4

Values of \bar{D}_s and σ_s were obtained using least-squares fitting of Eq. 4 to the PSDs of eight arid soils with a range of textures and geographical origins (40, 42).

ments are not representative of the emitted dust PSD because they inherently include aerosol removal due to deposition. We instead require measurements of the size-resolved vertical dust flux produced by arid soils, which is a direct measure of the PSD of the emitted dust aerosols (45). A list of published size-resolved vertical dust flux measurements is given in Table S1.

To allow size-resolved vertical dust flux measurements from the six distinct soils investigated in the literature (14, 15, 45, 46) to be collectively compared against Eq. 5, the measurements were processed as follows: Because measurements follow the power law of 1 in the range of 2–10 μm (see Fig. 2, *Inset*, and refs. 15 and 45), each set of measurements in that size range for a given soil and a given wind speed were fit to this power law. Measurements at all aerosol sizes for this given soil and wind speed were then normalized by the proportionality constant in the fitted power law to eliminate the strong dependence of the dust flux on the wind speed. For a given soil, this procedure put measurements at different wind speeds on an equal footing, except for the dependence of the shape of the dust PSD on the wind speed, which measurements suggest to be small (15, 44, 46). The normalized measurements at the various wind speeds for a given soil were then averaged for each particle size to reduce measurement noise and obtain the standard error. Because ref. 45 obtained only one reliable measurement per particle size, the standard error on these single measurements was estimated from the similar measurements of refs. 15 and 46.

Results

The theoretical emitted dust PSD (Eqs. 5 and 6) depends on the parameters \bar{D}_s , σ_s , and λ . The soil parameters $\bar{D}_s = 3.4 \mu\text{m}$ and $\sigma_s = 3.0$ are taken from Table 1, and the side crack propagation length $\lambda = 12 \pm 1 \mu\text{m}$ is obtained from a least-squares fit to the measurements in Fig. 2. This latter result is in agreement both with the expected value of $\sim 10\%$ (26, 33) of the typical dust aggregate size of $\sim 20\text{--}300 \mu\text{m}$ (17) and with the occurrence of scale invariance in dust emission up to a particle diameter of $\sim 10 \mu\text{m}$ (see Fig. 2, *Inset*, and discussion below). These values of \bar{D}_s , σ_s , and λ yield $c_N = 0.9539 \mu\text{m}$ and $c_V = 12.62 \mu\text{m}$ for the normalization constants in Eqs. 5 and 6.

Comparison of Theory with Measurements. Fig. 2 shows that the PSD of emitted dust aerosols is indeed scale invariant in the range of 2–10 μm , because it closely follows the predicted power law of 1. Moreover, the PSD is reduced relative to the power

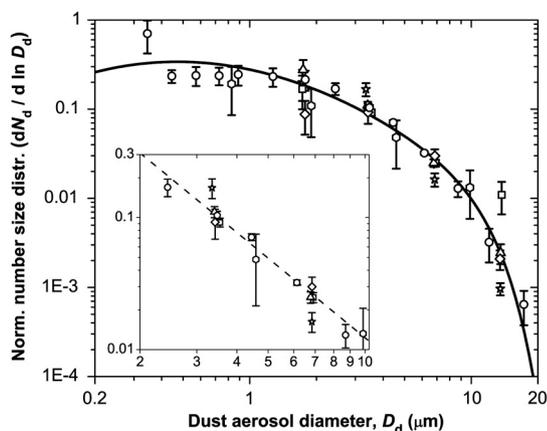


Fig. 2. Measurements with standard error of the normalized number size distribution of emitted dust aerosols [circles (14), squares (15), hexagons (45), triangles (soil 1 in ref. 46), stars (soil 2 in ref. 46), and diamonds (soil 3 in ref. 46)]. (*Inset*) Measurements in the size range of 2–10 μm only. The solid line denotes the theoretical prediction of Eq. 5, and the dashed line denotes the power law of 1 observed for the fragmentation of brittle materials (see Fig. 1).

law for small particle sizes ($< 2 \mu\text{m}$), as predicted from the dust aggregate indivisible constituent size of $\sim 1 \mu\text{m}$. The emitted dust PSD is also reduced relative to the power law for larger particle sizes ($> 10 \mu\text{m}$), as predicted from the limited propagation length of side cracks. Measurements of the emitted dust PSD thus provide strong qualitative support for the dust emission theory presented above.

In addition to this qualitative agreement, Fig. 2 shows excellent quantitative agreement between theory and measurements. Do note that the theory is poorly constrained in the submicron range because of sparse measurements (14).

The final important result evident from Fig. 2 is the small amount of scatter between the dust flux datasets, even though these data were obtained for widely varying wind and soil conditions (Table S1). This similarity suggests that changes in the wind and soil conditions have only a limited effect on the emitted dust PSD, as also suggested by the insensitivity of dust aerosol PSDs to changes in wind speed and source region (37, 43, 44). Although more research is needed to fully verify this hypothesis, the apparent insensitivity of the emitted dust PSD to specific soil and wind conditions is highly fortuitous for regional and global dust modeling (44).

Comparison of Theory with Empirical Model Relations. Fig. 3 compares the theoretical expression of Eq. 6 to empirical relations used in regional models and GCMs. The theory is clearly an improvement over empirical model relations, because it provides much better agreement with measurements. In fact, both theory and measurements show that empirical model relations substantially overestimate the mass fraction of emitted clay aerosols ($D_d < 2 \mu\text{m}$), whereas they underestimate the fraction of emitted large silt aerosols ($D_d > \sim 5 \mu\text{m}$). The greater fraction of large silt aerosols found by measurements and predicted by theory is consistent with the underestimation of the long-range transport of large silt aerosols by GCMs (11, 12, 47).

With the exception of ref. 48, GCMs assume emitted clay mass fractions ranging from $\sim 10\%$ (11, 12, 20, 49) to $\sim 35\%$ (13, 18), whereas measurements and theory indicate an emitted clay fraction of $4.4 \pm 1.0\%$ for the average values and standard errors of \bar{D}_s , σ_s , and λ listed above and in Table 1. Likewise, measured PSDs of dust aerosols indicate an atmospheric clay fraction of $\sim 10\text{--}20\%$, whereas GCMs predict an atmospheric clay fraction of $\sim 20\text{--}60\%$ (Table S2). Further evidence that GCMs overestimate the emitted clay fraction was reported by Cakmur et al. (49), who found that optimal agreement of a GCM with measurements (e.g., dust aerosol optical depth, deposition, and PSD) requires a smaller clay fraction than normally used.

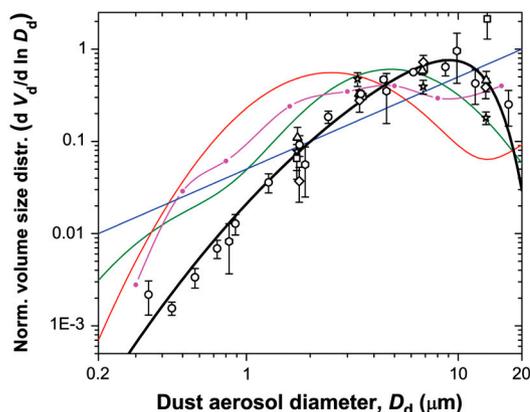


Fig. 3. The normalized volume size distribution of emitted dust aerosols used in 4 GCM studies [magenta line and circles (3), and blue (20), green (12), and red (13) lines]. The thick black line denotes the theoretical PSD of Eq. 6, and symbols and error bars denote measurements as defined in Fig. 2.

house gas concentrations through the fertilization of marine biota (6), may be substantially larger than previously thought, especially close to dust source regions.

The theoretical model presented here could be applied to fragmentation in analogous physical systems where the creation of small fragments is limited by the presence of indivisible particles. This includes dust emission on Mars and the fragmentation of small asteroids (53), granular rocks (29), and other brittle materials with a granular or crystal structure.

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