Improving simulations of fine dust surface concentrations over the western United States by optimizing the particle size distribution

Li Zhang,[1,2] Jasper F. Kok,[3] Daven K. Henze,[1] Qinbin Li,[2] and Chun Zhao[4]

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[1] To improve estimates of remote contributions of dust to fine particulate matter ($PM_{2.5}$) in the western United States, new dust particle size distributions (PSDs) based upon scale-invariant fragmentation theory (Kok_PSD) with constraints from in situ measurements (IMP_PSD) are implemented in a chemical transport model (GEOS-Chem). Compared to initial simulations, this leads to reductions in the mass of emitted dust particles with radii $<1.8\,\mu m$ by 40%–60%. Consequently, the root-mean-square error in simulated fine dust concentrations compared to springtime surface observations in the western United States is reduced by 67%–81%. The ratio of simulated fine to coarse PM mass is also improved, which is not achievable by reductions in total dust emissions. The IMP_PSD best represents the PSD of dust transported from remote sources and reduces modeled $PM_{2.5}$ concentrations up to $5\,\mu g/m^3$ over the western United States, which is important when considering sources contributing to nonattainment of air quality standards. **Citation:** Zhang, L., J. F. Kok, D. K. Henze, Q. Li, and C. Zhao (2013), Improving simulations of fine dust surface concentrations over the western United States by optimizing the particle size distribution, *Geophys. Res. Lett.*, *40*, 3270–3275, doi:10.1002/grl.50591.

1. Introduction

[2] The annual imported mass of dust aerosol from transpacific transport over North America is comparable in magnitude to all domestic aerosol sources [*Yu et al*., 2012]. Accordingly, surface dust concentrations in the western United States in spring are influenced not only by local sources [*Hwang and Hopke*, 2007; *Wells et al*., 2007] but also by transpacific transport of Asian (49%–77%, depending on season) and African (15%–34%) dust [*Husar et al*., 2001; *Chin et al*., 2007; *Zhao et al*., 2008; *Yu et al*., 2012], with the Asian dust contributing between 0.2 and $1.0\,\mu g/m^3$ in the spring [*VanCuren and Cahill*, 2002]. This transpacific dust transport contributes to concentrations of fine particulate matter ($PM_{2.5}$) above attainment thresholds set by the U.S. Environmental Protection Agency (EPA) [*Husar et al*., 2001; *Jaffe et al*., 2003; *Szykman et al*., 2003], which leads to degradation of public health and visibility [*Mahowald et al*., 2007; *Kang et al*., 2012].

[3] Chemical transport models, such as GEOS-Chem [*Bey et al*., 2001], are often used to investigate impacts of dust on surface air quality, visibility, and long-range transport [*Chung et al*., 2003; *Fairlie et al*., 2007; *Fairlie et al*., 2010]. Significant positive biases have been identified in GEOS-Chem simulations of surface dust concentration and dust aerosol optical depth (AOD) compared to observations [*Generoso et al*., 2008; *Fairlie et al*., 2010; *Ku and Park*, 2011; *Johnson et al*., 2012; *Ridley et al*., 2012; *Wang et al*., 2012]. Large uncertainties exist in the particle size distribution (PSD) of emitted dust in such models since the PSD from different soil types is not well known [*Cakmur et al*., 2006; *Kok*, 2011a]. As the lifetime and transport of dust particles are highly size dependent [*Kok*, 2011a], understanding the size distribution of dust particles is therefore of great importance for estimating the impacts of domestic versus distant sources of dust on local air quality and visibility.

[4] Recently, a simple theoretical expression for the emitted dust PSD based on the physics of scale-invariant fragmentation of brittle materials was developed by *Kok* [2011a]. This scheme leads to simulated dust PSD in good agreement with field measurements and indicates that most models overestimate the relative contribution of clay dust particles ($<2\,\mu m$ diameter) by a factor of ~2–8. Revisions to dust PSDs in atmospheric circulation models support these conclusions [*Nabat et al*., 2012; *Johnson et al*., 2012]. Based on the latter work, *Ridley et al*. [2012] showed that redistributing the PSD of submicron dust in GEOS-Chem's optical calculations (i.e., not affecting dust mass concentrations) leads to improved agreement of modeled AOD compared to the Aerosol Robotic Network. *Johnson et al*. [2012] applied the PSD of *Kok* [2011a] in GEOS-Chem across the entire size range of emitted particles, showing improved agreement with space-born measurements of AOD and extinction.

[5] While these previous works underscore the importance of reassessing the dust PSD for improving estimates of global dust AOD distributions, the impacts of the PSD of emitted dust on surface dust concentration, especially the contribution of dust to $PM_{2.5}$, have yet to be investigated. In this study, we focus on surface-level fine dust concentration in the western United States, considering both the theoretical PSD of *Kok* [2011a] (Kok_PSD) and a new parameterization,

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[1]Department of Mechanical Engineering, University of Colorado Boulder, Boulder, Colorado, USA.
[2]Department of Atmospheric and Oceanic Sciences, University of California, Los Angeles, California, USA.
[3]Department of Earth and Atmospheric Science, Cornell University, Ithaca, New York, USA.
[4]Atmospheric Science and Global Change Division, Pacific Northwest National Laboratory, Richland, Washington, USA.

Corresponding author: L. Zhang, Department of Mechanical Engineering, University of Colorado, 1111 Engineering Drive, ECME 114, Boulder, CO 80309, USA. (li.zhang@colorado.edu)

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Table 1. Size-Resolved Mass Fraction (%) of Each Bin in Different Particle Size Distribution Schemes

Radii (μm)	Bin1 (0.1–1.0)				Bin2 (1.0–1.8)	Bin3 (1.8–3.0)	Bin4 (3.0–6.0)
	Submicron Bins						
Scheme	0.1–0.18	0.18–0.3	0.3–0.6	0.6–1.0			
GC_PSD	12.2%				25.3%	32.2%	30.2%
	25%	25%	25%	25%			
Kok_PSD	4.8%				12.5%	25.7%	57.0%
	0.5%	2.6%	19.2%	77.7%			
IMP_PSD	7.7%				19.2%	34.9%	38.2%
	0.7%	3.3%	24.9%	71.1%			

developed here based on Kok_PSD, that is consistent with measurements in the size range of 0.1–1.0 μm from the Dust and Biomass-burning Experiment (DABEX) [*Osborne et al*., 2008; *Zhao et al*., 2010]. The impact of changing PSD and reducing dust mass emission on the ratio of fine dust versus coarse dust (2.5 μm < diameter < 10 μm) is also investigated.

2. GEOS-Chem Model and Observations

[6] GEOS-Chem is a global three-dimensional chemical transport model driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) [*Bey et al*., 2001]. We use GEOS-Chem version 9-01-01 driven by GEOS-5 meteorological fields, 2° (latitude) × 2.5° (longitude) horizontal resolution, and 47 vertical layers between the surface and 0.01 hPa. A nested simulation over North America with 1/2° (latitude) × 2/3° (longitude) horizontal resolution is performed for comparison. The standard dust scheme in GEOS-Chem is the dust entrainment and deposition (DEAD) mobilization scheme of *Zender et al*. [2003], combined with the source function used in the Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model [*Ginoux et al*., 2001; *Chin et al*., 2004] as described by *Fairlie et al*. [2007].

[7] Dust in GEOS-Chem is distributed across four size bins (radii 0.1–1.0, 1.0–1.8, 1.8–3.0, and 3.0–6.0 μm) following *Ginoux et al*. [2004]. The smallest size bin is further divided equally into four submicron size bins (with effective radii centered at 0.15, 0.25, 0.4, and 0.8 μm) for calculation of optical properties and heterogeneous chemistry [*Fairlie et al*., 2010; *Ridley et al*., 2012]; the mass distribution within the submicron bins does not affect dust mass concentrations. The size-resolved mass fractions of the standard dust scheme in GEOS-Chem (GC_PSD) are shown in Table 1. Fine dust concentration is estimated by summing the first bin and 38% of the second bin [*Fairlie et al*., 2007, 2010].

[8] Long-term measurements of fine dust and coarse PM (2.5 μm < diameter < 10 μm) are available in the United States since 1987 from the Interagency Monitoring of Protected Visual Environment (IMPROVE) network for the protection of visibility in class I remote areas [*Malm et al*., 1994] (data available at http://vista.cira.colostate.edu/improve/). Figure S1 in the supporting information shows the 70 IMPROVE sites in the western United States; these are remote sites situated at various elevations. Surface soil dust concentrations are calculated as the sum of the soil-derived elements (Al, Si, K, Ca, Ti, and Fe) and their normal oxides [*Malm et al*., 1994]. Fine dust mass is estimated using the formula of *Malm et al*. [1994]. GEOS-Chem model results are sampled according to the IMPROVE observations (24 h averages every 3 days) at the locations of the IMPROVE sites in California (CA, 19 sites), Washington (WA, 9 sites), Oregon (OR, 6 sites), Nevada (NV, 3 sites), Idaho (ID, 3 sites), Utah (UT, 5 sites), Arizona (AZ, 17 sites), and Colorado (CO, 8 sites). Nevertheless, the comparison between localized observations with model results that are representative of a much larger area is inherently problematic. In addition, the comparison is further complicated by the fact that many of the IMPROVE sites are located in mountainous areas, and the associated upslope flow is difficult to represent in a coarse-resolution model. Therefore, dust concentrations are averaged for IMPROVE sites within each state when comparing modeled to observed values.

3. Modified Dust Size Distributions

[9] *Kok* [2011a] derived the following theoretical expression for the emitted dust size distribution based on the physics of scale-invariant fragmentation of brittle material,

$$\frac{\mathrm{d}V_\mathrm{d}}{\mathrm{d}\ln D_\mathrm{d}} = \frac{D_\mathrm{d}}{c_\mathrm{V}}\left[1+\mathrm{erf}\left(\frac{\ln\left(D_\mathrm{d}/\overline{D}_\mathrm{s}\right)}{\sqrt{2}\ln\sigma_\mathrm{s}}\right)\right]\exp\left[-\left(\frac{D_\mathrm{d}}{\lambda}\right)^3\right] \quad (1)$$

where V_d is the normalized volume of dust aerosols with diameter D_d, the normalization constant $c_\mathrm{V} = 12.62$, the soil parameters of median diameter by volume $\overline{D}_\mathrm{s} = 3.4$ μm, the geometric standard deviation $\sigma_\mathrm{s} = 3.0$, and $\lambda = 12 \pm 1$ μm denotes the propagation distance of side branches of cracks created during a fragmentation event. We integrate the dust mass fluxes following equation (1) into each size bin. Thus, the new size-resolved mass fractions based on *Kok* [2011a], here referred to as the Kok_PSD, are redistributed as shown in Table 1. Compared to GC_PSD, the mass fractions of the first two size bins are reduced significantly, by more than 50%, when applying the Kok_PSD, while the total dust mass is held the same. A slight reduction occurs in the third size bin, while the fraction in the fourth size bin nearly doubles. While the mass fractions in the submicron size bins are divided equally (25% each) in the GC_PSD, these fractions become 0.5%, 2.6%, 19.2%, and 77.7% using the Kok_PSD. However, we note that the mass factions of the submicron size bins do not change the simulation of dust mass concentrations in GEOS-Chem; they only impact the dust optical properties.

[10] *Zhao et al*. [2010] used WRF-Chem to simulate dust, considering two different size distributions of emitted dust. One is suggested by *Osborne et al*. [2008] based on aircraft measurements [*Zhao et al*., 2010, "Modal2"], and the other one is obtained by comparing the simulated dust PSD with

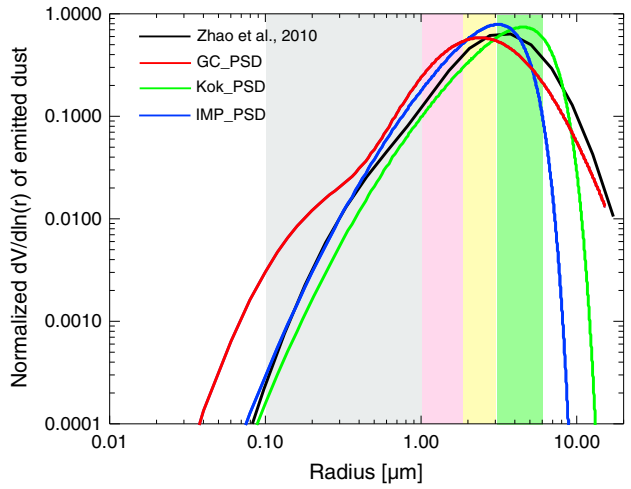

Figure 1. Comparison of normalized volume size distributions of emitted dust particles between GC_PSD (red line), Kok_PSD (green line), IMP_PSD (blue line), and the results of *Zhao et al*. [2010] (black line). The radii ranges of the four size bins in GEOS-Chem are shaded with different colors.

the observations from the DABEX field campaign in North Africa [*Zhao et al*., 2010, "Modal1"]. The dust simulation better captured the cross-sectional area size distribution of the aircraft measurements with the "Modal1" PSD [see *Zhao et al*., 2010, Figure 1]. In Figure 1, we overlap the GC_PSD (red line) and Kok_PSD (green line) with the result ("Modal1") of *Zhao et al*. [2010] (black line). Both the Kok_PSD and the *Zhao et al*. [2010] PSD are quite different from the GC_PSD, especially for particle radii less than 0.4 μm. The Kok_PSD is more consistent with that of *Zhao et al*. [2010] for particle radii less than 4 μm. However, more than 30% differences are indicated in the radius range of 0.1–0.4 μm, which is the range of the first size bin in GEOS-Chem, meaning that fewer dust particles in the first size bin would be emitted with the Kok_PSD. In equation (1) [*Kok*, 2011a, equation (6)], the side crack propagation length λ is obtained from a least squares fit to measurements. Fundamentally, λ is rather uncertain as it depends on poorly constrained factors like soil cohesion. For instance, changes in soil moisture, and thus cohesion, can produce changes in the value of λ [*Kok*, 2011b]. Therefore, the value of λ is very difficult to describe theoretically and can be expected to depend on soil parameters such as mineralogy, soil moisture, and size distribution [*Kok*, 2011a, 2011b]. For instance, the data set of emitted dust PSDs measured in the Japan-Australia Dust Experiment (JADE) [*Shao et al*., 2011] is in good agreement with the theory of *Kok* [2011a] but has a lower value of λ than that of Kok_PSD [see *Kok*, 2011b, Figure 1].

[11] Considering the uncertainties inherent in the parameter λ, we tune this parameter to maximize agreement with the results of *Zhao et al*. [2010], yielding a value of $\lambda = 8$ μm, with a corresponding change of the normalization constant ($c_V = 6.83$). This value of λ is also consistent with the spread shown in observational data sets, such as *Shao et al*. [2011]. The new PSD (blue line) nearly overlaps the results of *Zhao et al*. [2010] (see black line) in the clay size bin (<2 μm diameter, see Figure 1). We refer to this as the IMP_PSD (blue line), for which the size-resolved mass fractions are provided in Table 1. The IMP_PSD will emit more particles in the first size bin (clay size) but less in the fourth size bin compared to the Kok_PSD. Compared to the Kok_PSD, the IMP_PSD shows positive bias relative to the result of *Zhao et al*. [2010] for coarse particle with radii between 0.7 and 4 μm. The PSD for relatively large dust sizes range (>~5 μm radii) shows substantial variation between data set with different soil properties [*Kok*, 2011a, 2011b] and is thus uncertain. These variations in the dust PSD impact model estimates of local dust sources on coarse (>5 μm radii) aerosol. While the performance of the above PSDs for large particles (>5 μm radii) warrants further study, we focus in this work on the PSD for particles limited to the size range in GEOS-Chem (<6.0 μm radii), which is of most importance for assessing the influence of long-range aerosol on $PM_{2.5}$ concentrations over the western United States.

4. Results and Discussion

[12] Figure 2 shows the simulated and observed time series of fine dust averaged by state over the western United States during the season of strong transpacific dust transport for March–May 2006. The root-mean-square error (RMSE) of fine dust concentrations between IMPROVE observations and model simulations is shown with colored text corresponding to the different dust PSDs. With the GC_PSD, the model consistently overestimates surface dust concentrations by more than 50%, and the RMSEs are above 0.7 μg/m^3 in coastal states but above 1.0 μg/m^3 inland. Even performing a nested simulation with finer horizontal resolution, the fine dust concentrations are still largely overpredicted and do not improve, and the RMSEs are still above 0.7 μg/m^3 in coastal states (Figure S2). While the temporal variability is consistent regardless of the PSD, simulations using the Kok_PSD show dramatic reductions in absolute mass concentrations compared to GC_PSD. This actually leads to dust concentrations being underestimated with the Kok_PSD in most states from mid-April to the end of May. In contrast, the IMP_PSD well captures the time series of fine dust concentrations in CA, WA, and OR with the RMSEs reduced to 0.16, 0.16, and 0.13 μg/m^3, respectively, which are 40%–47% lower than that using the Kok_PSD and 79%–82% lower than that using the GC_PSD.

[13] While the IMP_PSD is an overall improvement compared to the GC_PSD and the Kok_PSD in AZ, NV, ID, and CO, some discrepancies persist after mid-April in these states, in which sites are in or downwind of local dust source regions [*Fairlie et al*., 2007]. To investigate this, we performed sensitivity simulations (SW_contrib) in which we zeroed out dust emission over the western U.S. source region (30°N–42°N, 120°W–108°W, see Figure 1). These simulations indicate that for the IMP_PSD, the peak dust concentrations in NV, AZ, and UT are dominated by local contributions after mid-April (see Figure 2, the blue dash line), especially in AZ. In contrast, coastal states (CA, WA, and OR) are not highly influenced by local emissions but are instead dominated by long-range transport, in agreement with previous studies [*Husar et al*., 2001; *VanCuren and Cahill*, 2002]. Model PSDs significantly impact the fine dust concentrations from both local and remote sources. Further, the peak dust concentrations (see the black arrows in Figure 2) in late March observed in most states, and most intensely in AZ, NV, ID, and CO, are well captured by the

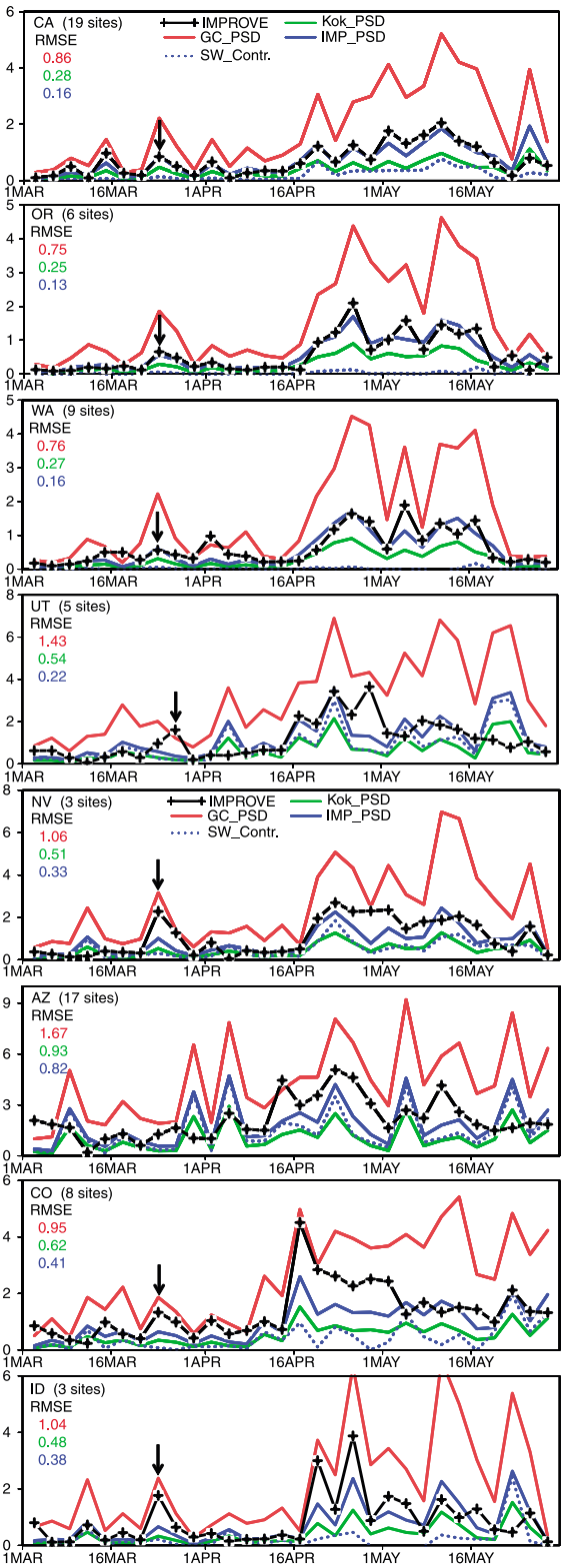

Figure 2. Simulated and observed (black dot line) surface dust concentration (μg/m^3) at IMPROVE sites by state from March to May 2006. The simulated results are based on GEOS-Chem with GC_PSD (red line), Kok_PSD (green line), IMP_PSD (blue line), and the dust contributions from the western United States using IMP_PSD (blue dash line). The colored texts are the RMSEs of dust concentrations between observation and model simulations with corresponding dust PSDs.

IMP_PSD for the West Coast states but underestimated in the four inland states. However, the magnitude of this peak is reproduced by the GC_PSD in some inland states (e.g., NV, CO, and ID). Further, nearly every modeled peak with the IMP_PSD in AZ that approaches 3 μg/m^3 has two noticeable features: more than 90% contribution from local sources (SW_contrib) and overestimation of the measurements. Therefore, the IMP_PSD appears consistent with the size distribution of transpacific dust particles emitted from Asia and Africa, in particular the African dust, since the IMP_PSD represents the DABEX measurements over Africa. It also suggests that the IMP_PSD may not represent the PSD well in the North American source regions, especially for smaller size bins.

[14] Reductions in concentrations of transpacific dust from Asian sources would be consistent with recent inverse modeling studies based on surface and remote-sensing constraints of dust in Asia [*Ku and Park*, 2011; *Wang et al.*, 2012]. Mechanistically, these changes to fine dust could also be achieved by reducing the total mass of dust emission over the source regions. To separate the impacts of PSD versus total emissions, we consider the ratio of fine to coarse dust. With the GC_PSD, this ratio is a factor of 3 larger than the observed ratio of fine dust to coarse PM over CA (Figure S3). While acknowledging that the observed ratio contains contributions from nondust species in the coarse PM and, hence, some overestimation by the model is expected, these would unlikely be substantial enough to explain the factor of 3 differences noted here. We therefore consider a sensitivity experiment based on the GC_PSD with dust emissions exterior to the western United States reduced by 50% (GC_0.5xEmi). While this does reduce the bias in the simulation of fine dust concentration over CA and AZ by 74% and 25%, respectively (Figure S4), the ratio of fine dust versus coarse dust does not change (Figure S3). In contrast, changing the PSD not only improves the fine dust simulation (Figure 2) but also improves the ratio of fine dust versus coarse dust by 65%–75% (Figure S3). While the comparisons of the Kok_PSD and IMP_PSD to the measurements of *Zhao et al.* [2010] for dust with radii between 0.7 and 4 μm imply a low bias for the former and a high bias for the latter, both schemes lead to significant reductions in the ratio of fine to coarse dust compared to GC_PSD. Thus, the overall comparisons of these schemes to the GC_PSD are not governed by the fitting (or lack thereof) of the new PSDs for particles in this size range.

5. Conclusions

[15] This study applied two new PSDs for emitted dust, the Kok_PSD and a newly developed IMP_PSD, to the GEOS-Chem model to reduce large discrepancies between the simulated surface-level fine dust concentration and measurements from the IMPROVE network in the western United States during March–May 2006. The new PSDs also improved the ratio of fine to coarse dust; simply adjusting the total dust emissions did not. Analysis of the timing of dust events indicates that the IMP_PSD is most appropriate for representing dust emitted and transported from Asia and Africa, but not the local dust sources in the western United States. Globally, it is likely that a combination of adjustments to both PSDs and dust emissions is necessary

or plausible for improving the simulations of fine dust concentrations and the ratio of fine to coarse dust. Accounting for the evolution of particle size during transport owing to aerosol mixing [*Zhang and Iwasaka*, 2004, 2006] would also likely improve the model simulations.

[16] Here the IMP_PSD has been applied globally to all dust source regions. Clearly, this is a significant simplification, as the PSD of emitted dust may vary over different source regions since soil parameters, such as the soil particle size distribution's median diameter and geometric standard deviation and the side crack propagation length, are dependent on soil properties, including cohesion [*Kok*, 2011a]. More measurements of the size distributions of emitted dust spanning a broad range of soil properties could allow the development of soil-specific source parameters. Constraints on the fundamental parameters governing dust mobilization schemes would be of value for improving our understanding of the factors governing dust emissions. Such fundamental source parameters may also be constrained using inverse modeling, which has thus far focused largely on adjusting the total mass of dust emissions [e.g., *Ku and Park*, 2011; *Wang et al*., 2012].

[17] Overall, adjustment of the dust PSD not only improves the dust simulation in GEOS-Chem but also revises our estimate of the role of long-range transport on $PM_{2.5}$ in the western United States. The U.S. EPA recently strengthened the annual National Ambient Air Quality Standard (NAAQS) for $PM_{2.5}$ from 15.0 to 12.0 $\mu g/m^3$ (http://www.epa.gov/airquality/particlepollution/actions.html#dec12). While this alone increases the fractional contribution of background sources to nonattainment, given that background $PM_{2.5}$ concentrations may have been overestimated in the spring by as much as 5 $\mu g/m^3$, controlling local sources of $PM_{2.5}$ may yet be nearly as effective in approaching attainment of the revised NAAQS as previously thought.

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