PNAS
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Supplementary Information for

Revisiting particle dry deposition and its role in radiative effect estimates

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This PDF file includes:

Supplementary text
Figures S1 to S2
Tables S1 to S3
SI References

www.pnas.org/cgi/doi/10.1073/pnas.2014761117

S1. Revised Parameterization Details

Gravitational settling, Brownian motion, impaction, and interception are three processes by which particles are lost to terrestrial surfaces. Losses due to Brownian motion in dry deposition models stems from Brownian diffusivity that is based on molecular diffusion (1) and represents a well understood physical process. The loss efficiency associated with Brownian motion includes the Schmidt number, which is simply the ratio of the kinematic viscosity of air to the diffusion term. The empirical coefficients are the exponent and coefficient of the Schmidt number and dictate the slope of the loss efficiency. We do not change the underlying physical basis for Brownian motion in our parameterization, but do modify the loss rate to terrestrial surfaces due to that motion. The loss of particles by Brownian motion is a result of the enhanced diffusivity present near the surface of a collector; particles are lost via a collision that we associate and describe by Brownian motion despite the process being a collision. Impaction is the process by which a particle is unable to follow the streamlines of a flow path and collides with some object and is collected. This physical basis of this process is well known (1) and hinges on the Stokes number of a particle. However, under both the turbulent flow conditions and the varying length scales of collectors in the ambient environment, the Stokes number cutoff for particle impaction is particularly poorly defined. As a result, the collection efficiency coefficient and exponent used to describe observed particle deposition rates to uneven surfaces (2) by impaction are approximations. Interception is the loss process by which a particle comes within a particle radius of a surface and is captured, and is perhaps the least constrained process of these three collection efficiencies. Interception is generally formulated as capture occurring along the tangent of a collector (1). Given a distribution of collectors, turbulent conditions, and length scales, this collection efficiency is parameterized based on the ratio of particle diameter to characteristic length of the collectors. There is no underlying physical basis for this term, and the ratio approach creates an inherent dependence on particle diameter that is strongly influenced by purely empirical coefficients.

To revise the existing parameterizations, we first looked to the interception terms and iteratively increased the C_{In} term and decreased the exponential term (υ) to minimize the model-measurement discrepancies in both size-dependence and magnitude of deposition velocities for the forest site. This change was inadequate for shifting the minimia in size dependence, and we thus simultaneously, iteratively suppressed the Brownian diffusion term and enhanced the impaction terms following Petroff et al. (3) to minimize the model-measurement discrepancies in the SPiFFY data. We used the same gamma term of Brownian diffusion as Petroff et al. (3), but decreased the collection term (C_B) to 0.2 in order to capture the small observed minimum – consistent with the concept that Brownian diffusion enhances dry deposition less than initially thought. We simultaneously suppressed the role of impaction to reduce the curvature that is introduced by a large impaction component at large particle diameters (>10 um). The revision of these coefficients in the parameterization was done in an iterative and methodical process in an effort to represent the data while remaining as consistent as possible with previously published data.

The revised dry deposition parameterization describes variability of literature measurements across multiple land use types. Revisions presented here are done in a manner that affects all land use types, but does not change the existing tables that describe the land use parameters in GEOS-Chem and many other chemical transport models. To be clear, this framework is both observationally constrained and based on the existing physical understanding of dry deposition. However, these principles do not mean that this parameterization is mechanistically accurate. In other words, this is a theoretical construct applied to represent observations, but there are likely complex surface level mechanisms that control particle deposition. However, we intend these revisions to be one way to more accurately represent the state of science – and current measurements in particular.

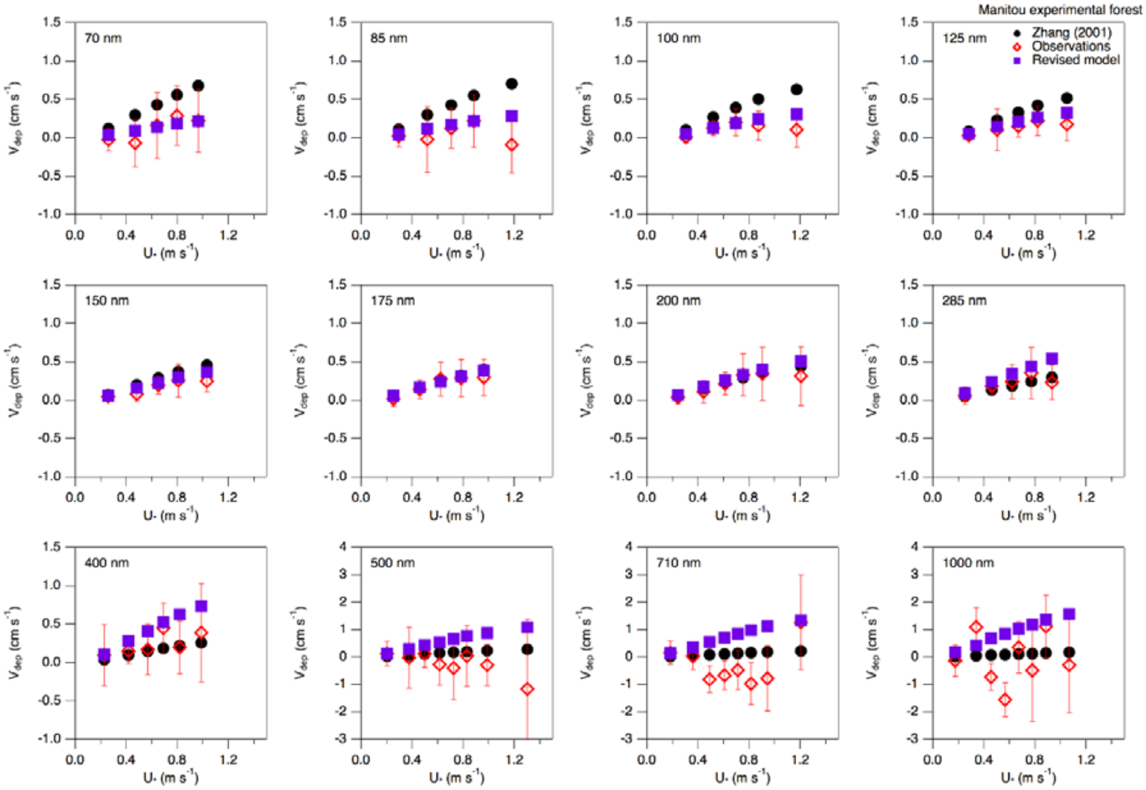

Fig. S1. Direct deposition velocity measurements from SPiFFY binned by friction velocity (N=200 per bin). Each plot is for a specific size range ending in the number listed on the plot. Also shown is the Zhang, Gong, Padro and Barrie (4) used in GEOS-Chem and a revised parameterization.

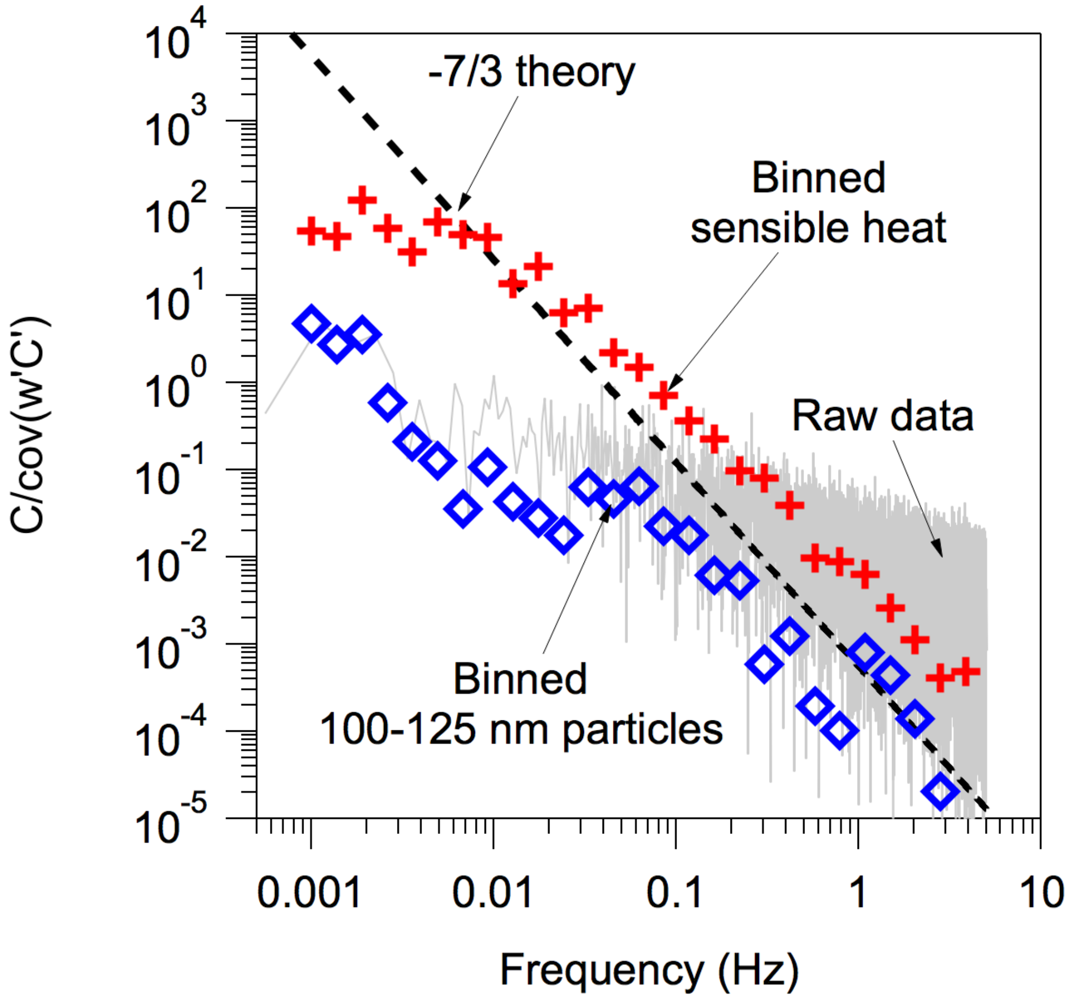

Fig. S2. Example cospectrum for 100-125 nm particles and sensible heat from 13-July 2015 at 8 AM during the SPIFFY field campaign. The shown raw data represent only the positive values from C/cov(w'C'). The binned data points represent the average of all the points over that frequency bin and thus include the negative values and tends to suppress the slope some relative to the raw data.

Table S1. Collection efficiency parameters that govern the surface resistance term from two previously published model algorithms and a revised scheme driven by observations.

	Zhang 2001	**Petroff 2010**	**Revised**
Brownian	$C_B Sc^{-\gamma}$	$C_B Sc^{-\gamma} * Re^{-1/2}$	$C_B Sc^{-\gamma}$
γ	0.5-0.58	2/3	2/3
C_B	1	0.7-1.262	0.2
Interception	$\mathrm{C_{In}}\left(\frac{d_p}{A}\right)^{\upsilon}$	$C_{In}\left(\frac{d_p}{L}\right)^{\upsilon}$ **or** $C_{In}\left(\frac{d_p}{L}\right)\left[2+\ln\left(\frac{4L}{d_p}\right)\right]$	$C_{In}\left(\frac{d_p}{A}\right)^{\upsilon}$
υ	2	1	0.8
C_{In}	0.5	0.14-0.81	2.5
Impaction	$C_{Im}\left(\frac{St}{0.8+St}\right)^{2}$	$C_{Im}\left(\frac{St}{\alpha+St}\right)^{2}$	$C_{Im}\left(\frac{St}{\alpha+St}\right)^{1.7}$
β	2	2	1.7
C_{Im}	1	0.47-0.6	0.4
Notes		For vegetated surfaces, smooth surfaces are parameterized in a different way. 26 land use surfaces are parameterized in this model	
Notes		Our measurements indicate the term $Re^{-1/2}$ is typically around 0.02±0.01 with a max of 0.18,	
Notes		L is the same parameter as A in Zhang and the revised parameterization. Slightly different values are used	

Table S2. GEOS-Chem-TOMAS sensitivity cases used.

Case	**Description**
base	The default GEOS-Chem-TOMAS model set-up with the Zhang parameterization
All_LUC	Applying the dry deposition parameterization developed in this work for all land use types
LUC1	Applying the parameterization to land use type 1 only (needle leaf forests)
10x base	Increasing the surface resistance parameter in the base case by 10 times
1/10x base	Decreasing the surface resistance parameter in the base case by 10 times
5x All_LUC	Increasing the surface resistance parameter in the All_LUC case by 5 times
1/5x All_LUC	Decreasing the surface resistance parameter in the All_LUC case by 5 times

Table S3. Changes in AIE, DRE, CDNC, $PM_{2.5}$ mass, and $PM_{2.5}$ number between the default Zhang parameterization in GEOS-Chem and the sensitivity cases (described in Table S2).

Case	AIE global (land) Δ [W m^{-2}]	DRE global (land) change Δ [W m^{-2}]	CDNC (land) % change	$PM_{2.5}$ mass global (land) % change	$PM_{2.5}$ number global (land) % change
All_LUC	-0.63 (-0.13)	-0.09 (0.02)	11.96 (11.08)	10.97 (6.53)	43.8 (26.3)
LUC1	-0.01 (-0.01)	0.02 (0.01)	0.32 (0.36)	-1.73 (-1.31)	6.99 (7.94)
10x base	-0.56 (-0.13)	-0.17 (-0.16)	10.38 (10.95)	14.83 (14.91)	16.15 (-13.72)
1/10x base	0.3 (0.08)	0.52 (0.63)	-2.44 (-10.18)	-33.17 (-35.4)	120.12 (99.62)
5x All_LUC	-0.68 (-0.16)	-0.16 (-0.13)	13.58 (13.44)	15.19 (14.41)	48.47 (3.79)
1/5x All_LUC	-0.46 (-0.07)	0.09 (0.36)	7.22 (4.11)	-0.54 (-12.47)	35.56 (51.04)

SI References

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