Revisiting dry deposition of particles in the atmosphere

U.S. DEPARTMENT OF ENERGY | Office of Science

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Deposition: An under-appreciated aspect of the aerosol life cycle?

Chemistry

Chemistry

$$\frac{d[X]}{dt} = Production - Loss - \nabla_v$$

Direct Emissions

Deposition

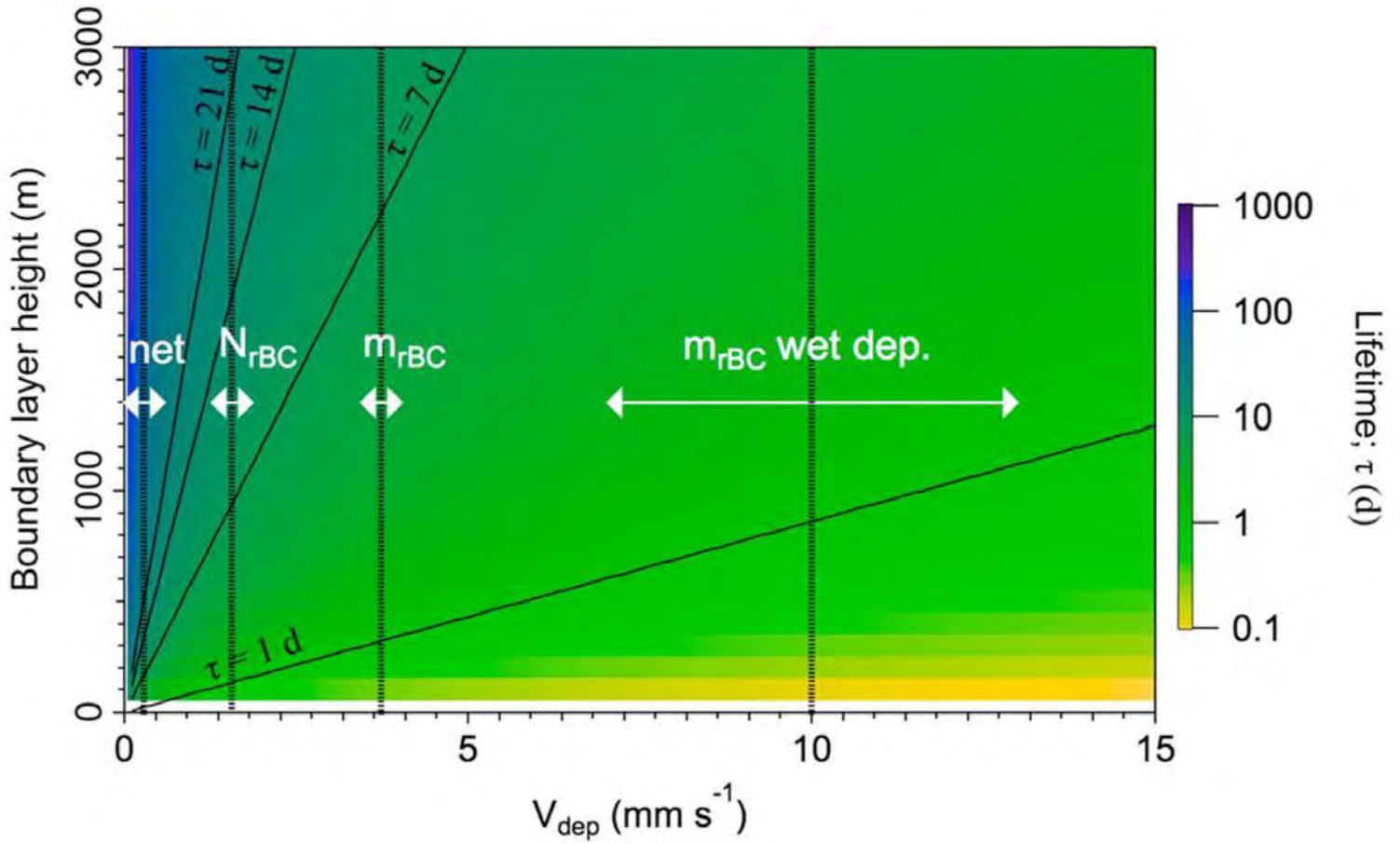





Black Carbon is a tracer for studying wet vs dry deposition → eddy covariance flux measurements at the Southern Great Plains site

Dry deposition accounted for 6 ± 4% of total BC deposition *during BCADS**

Net BC lifetime: 7-11 days, dominated by wet deposition

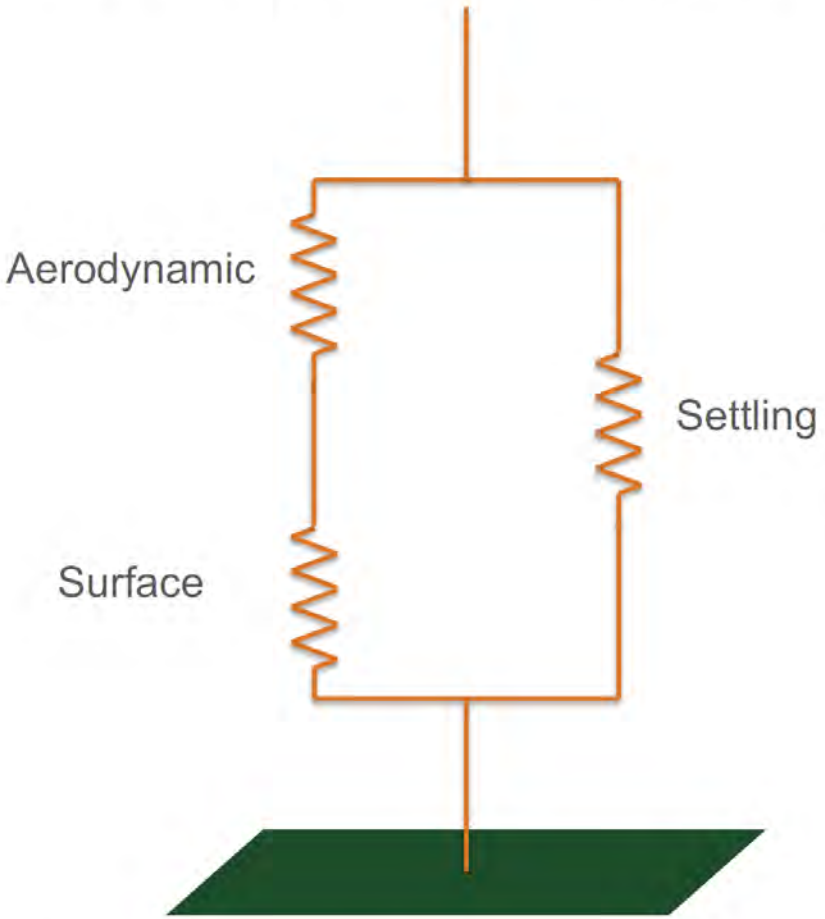


This analysis considers bulk BC, but particle deposition varies by size…

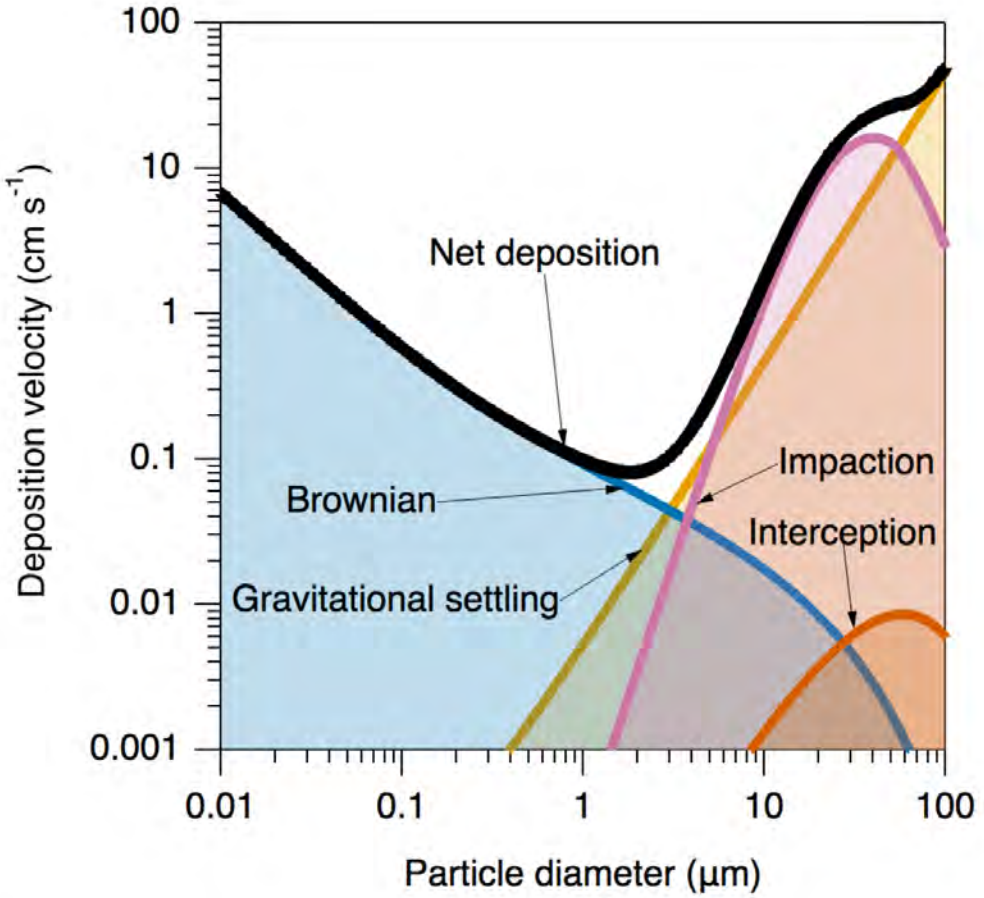
*14 cm rainfall over ~6 weeks

Emerson et al. 2019. J. Geophys. Res.

Traditional view of particle dry deposition



$$V_{dep} = V_{settling} + \frac{1}{R_{aerodynamic} + R_{surface}}$$



$$E_{Brownian} = C_b Sc^{-2/3}$$

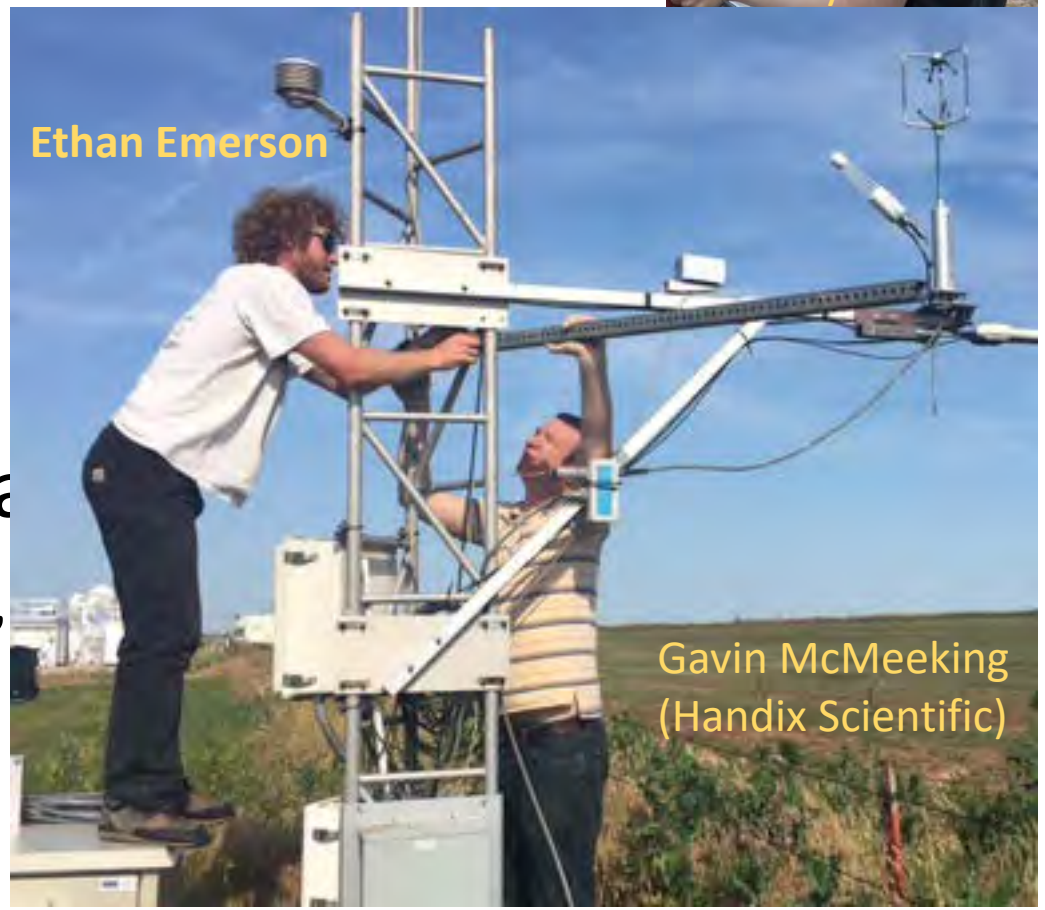
$$E_{Impaction} = C_{Im}\left(\frac{St}{\alpha + St}\right)^{\beta}$$

$$E_{Interception} = C_{In}\left(\frac{d_p}{A}\right)^{v}$$

[Zhang et al. 2001; Slinn, 1982]

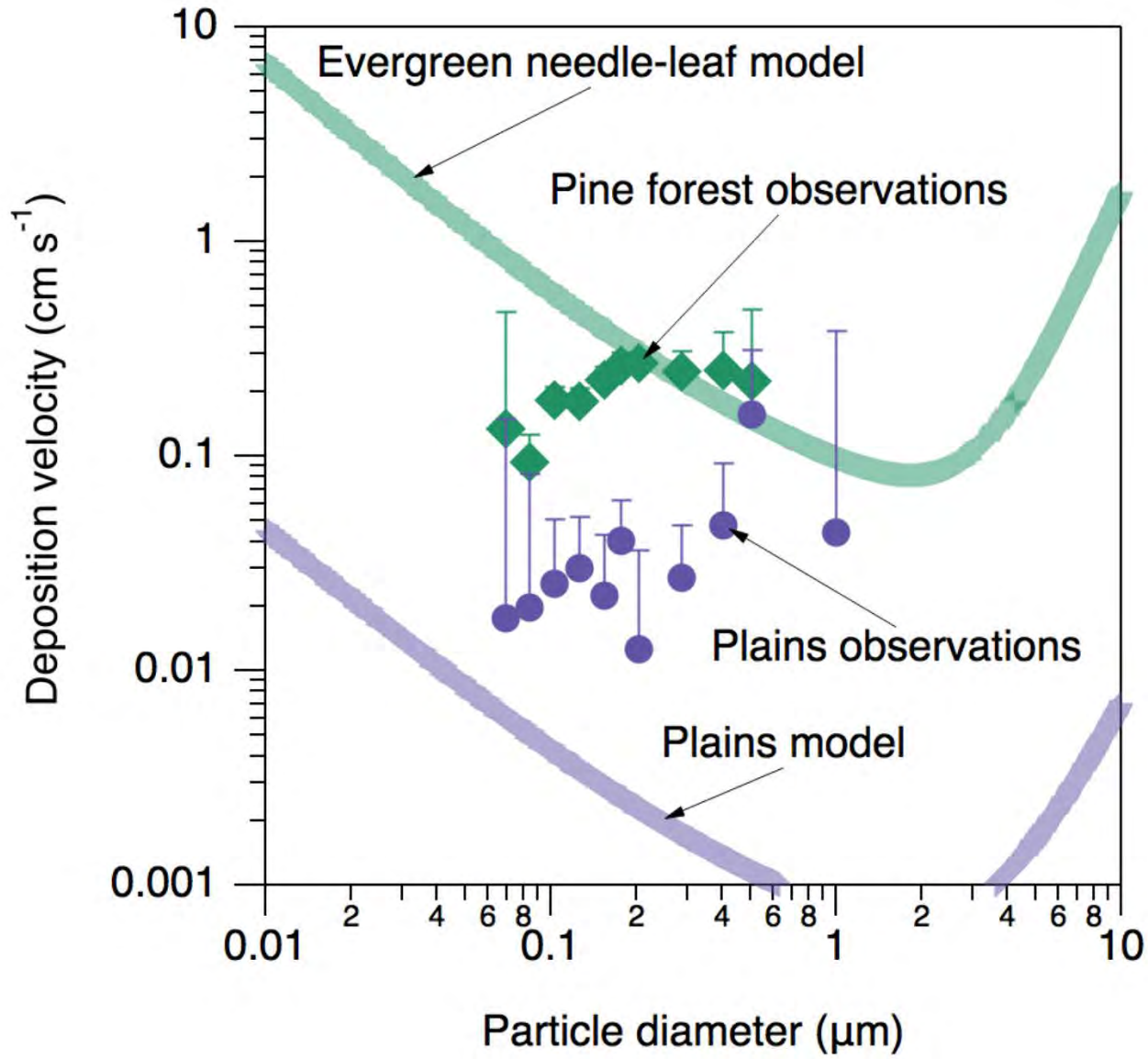
We use two sets of size-resolved particle flux measurements

Manitou Forest, CO
4 seasons,



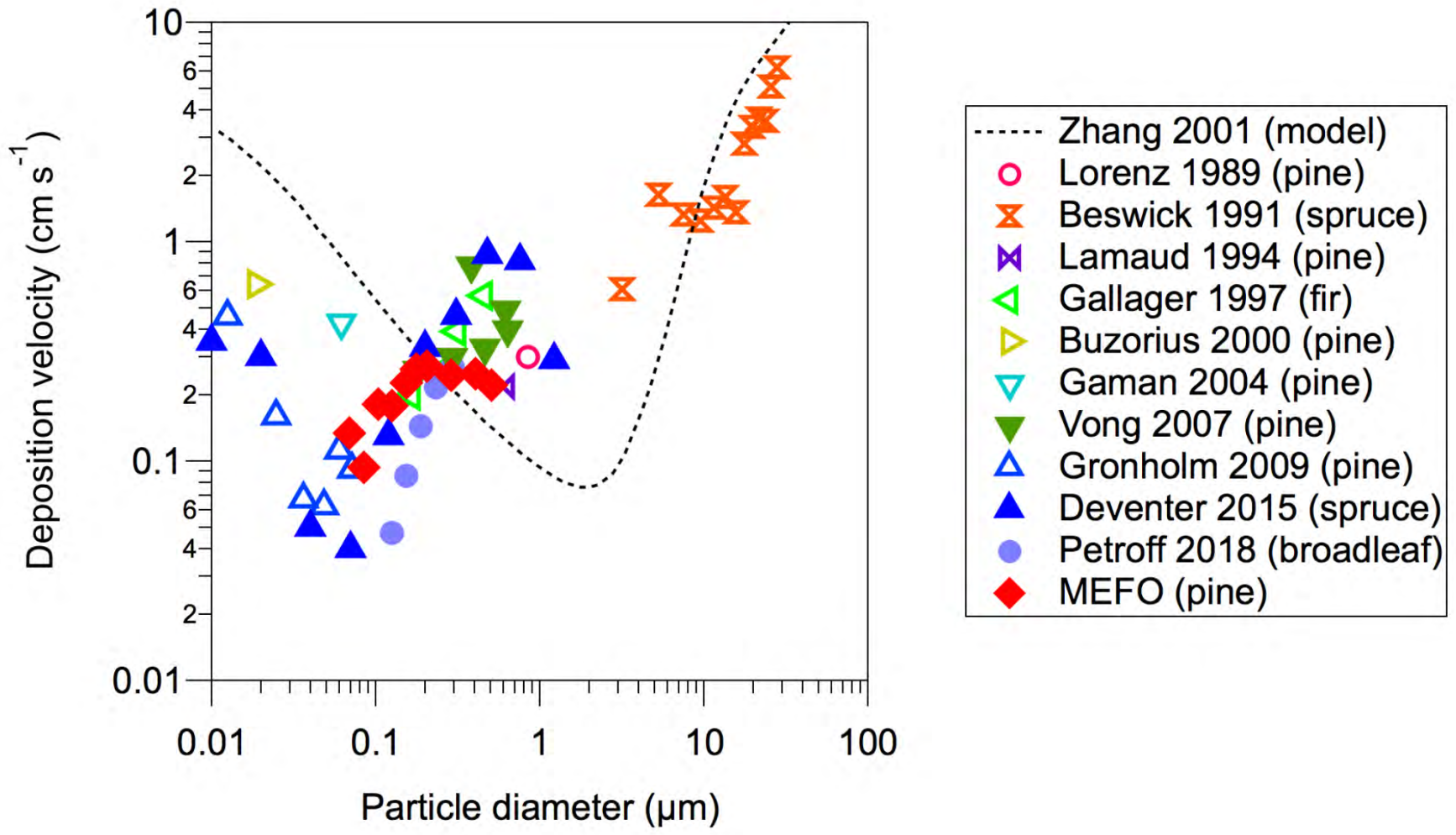
Southern Grea
6 weeks,

Widely-used models fail to capture the observational data



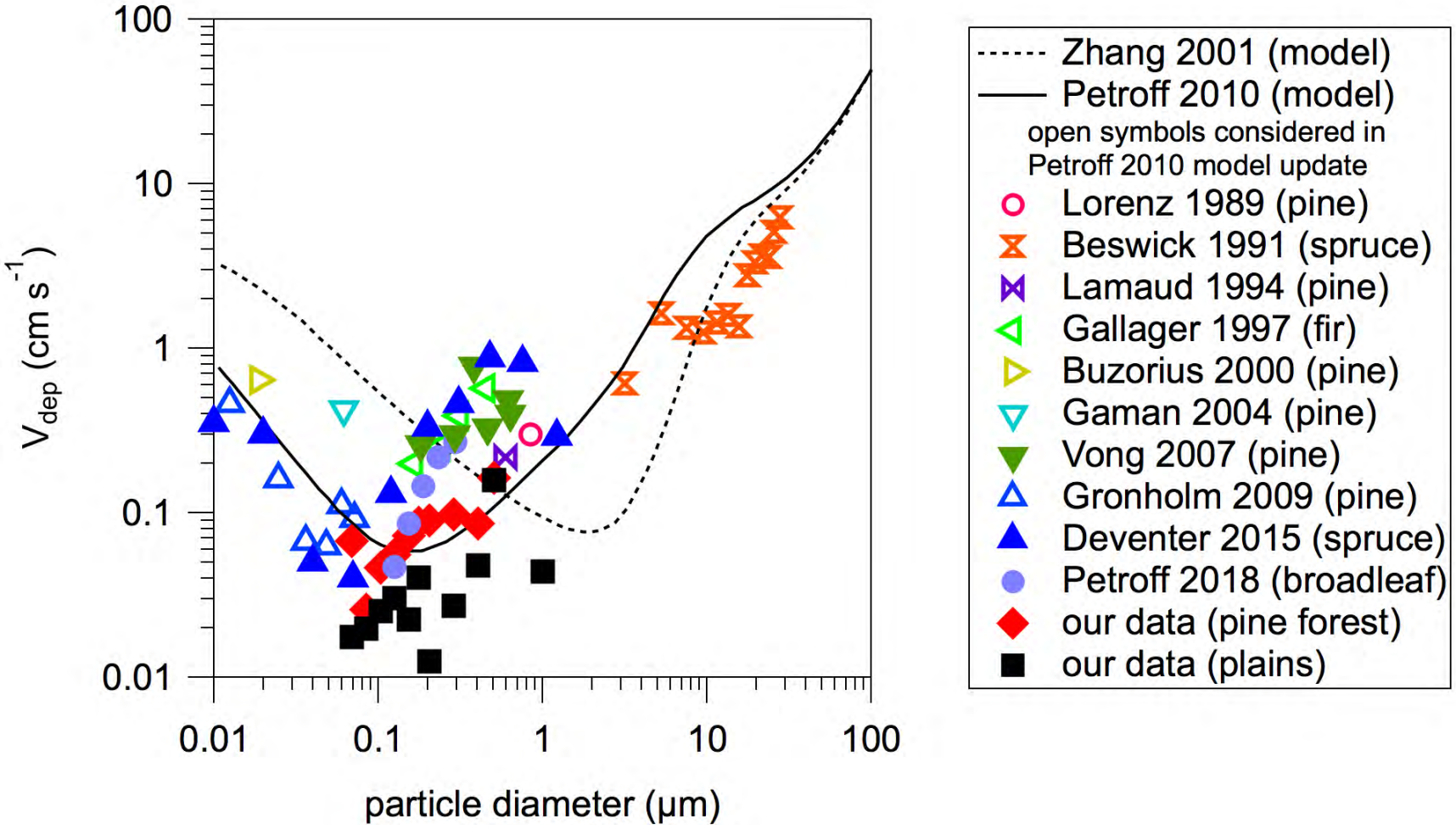
Model: Zhang et al. [2001]

Widely-used models fail to capture the observational data



Sophisticated deposition models capture the observations (but widely used simpler ones generally do not)

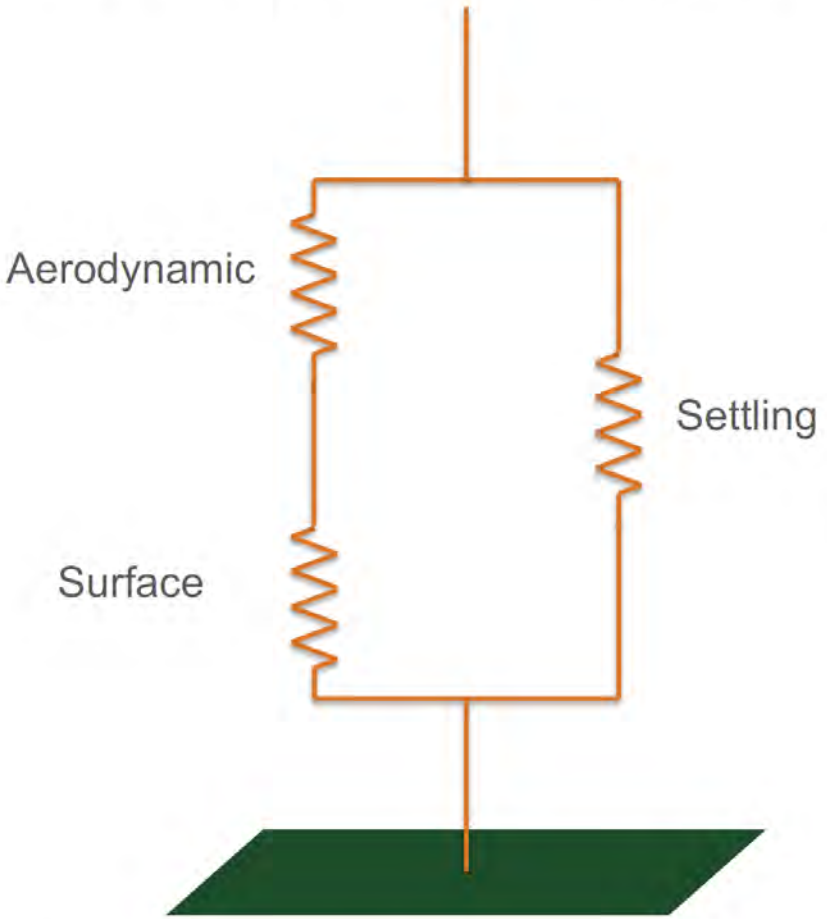
- Can we develop a simple (yet accurate) model for global models?
- What underlying processes control particle dry deposition?



* Model shown is for forested terrain

Zhang et al., (2001); Petroff & Zhang, (2010)

We use the extensive observations to modify simple parameterization terms, using the sophisticated models as a framework



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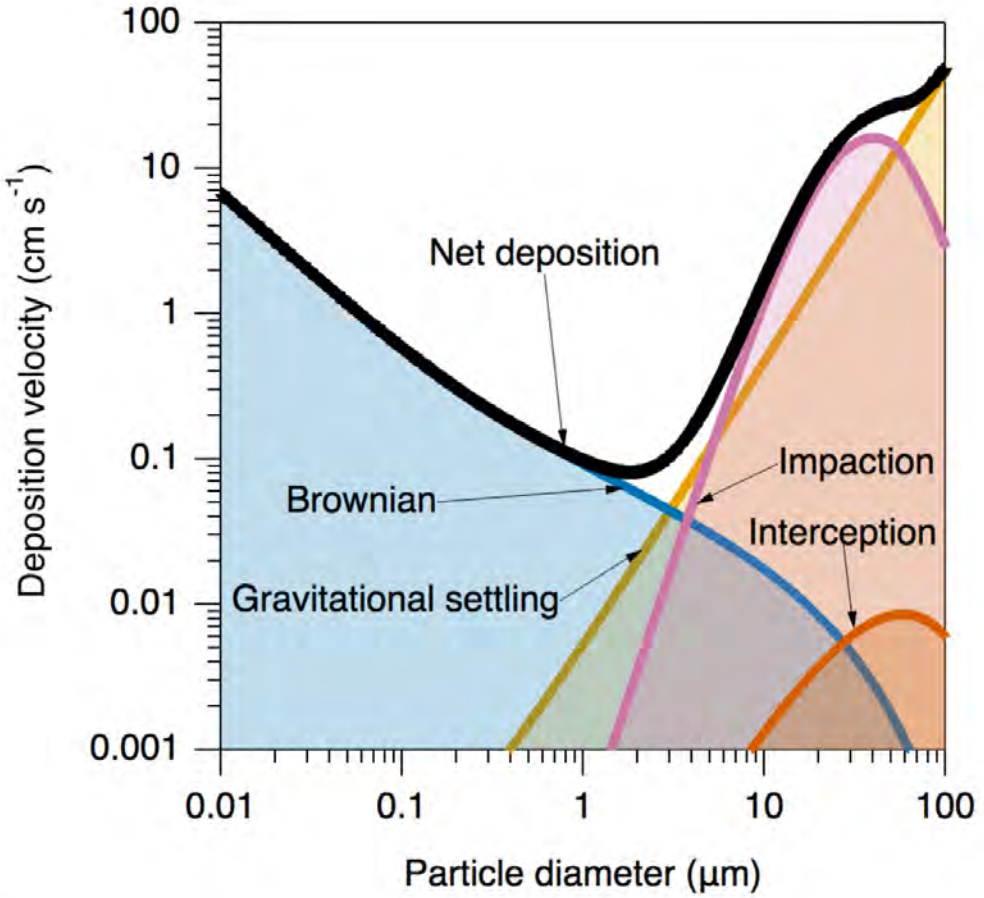

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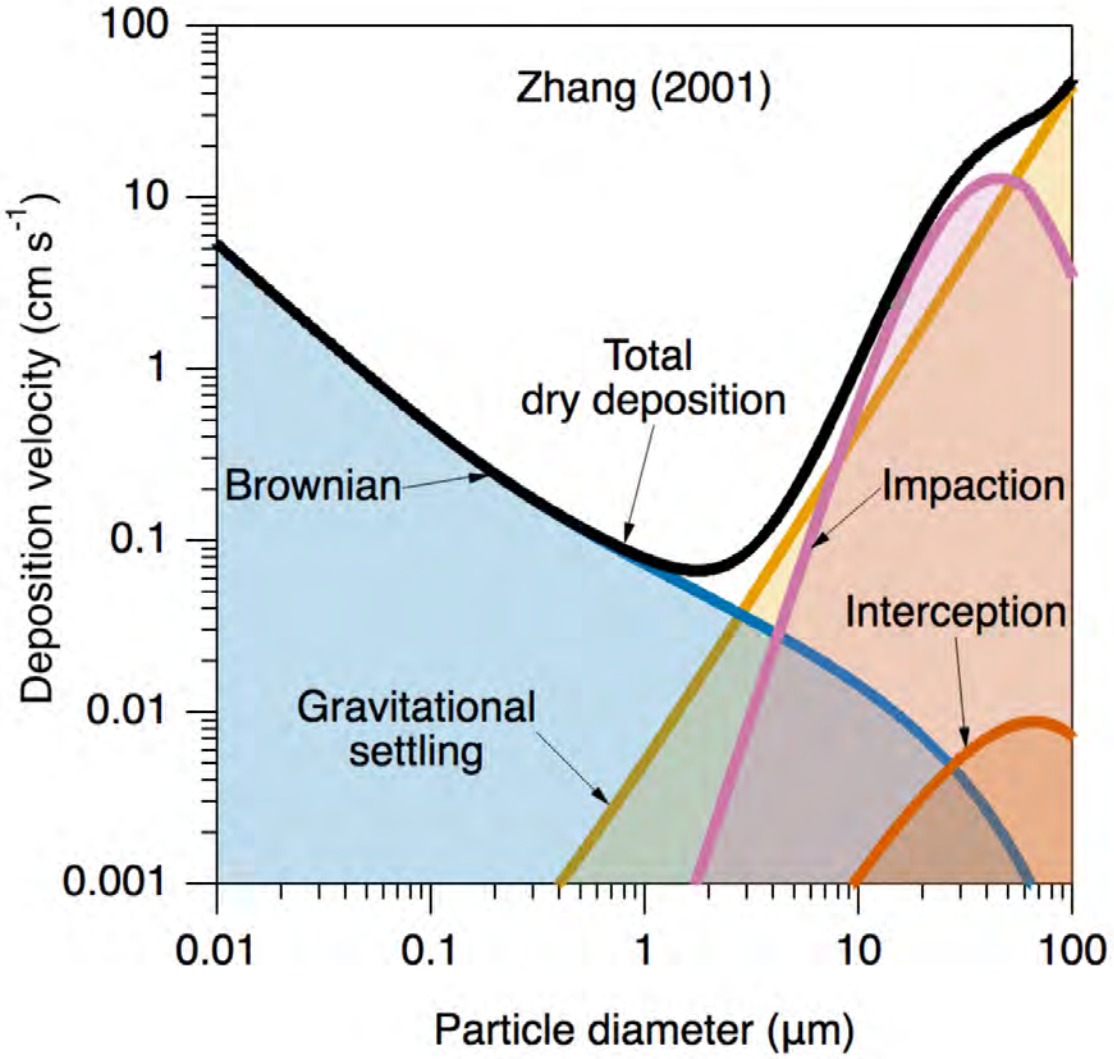
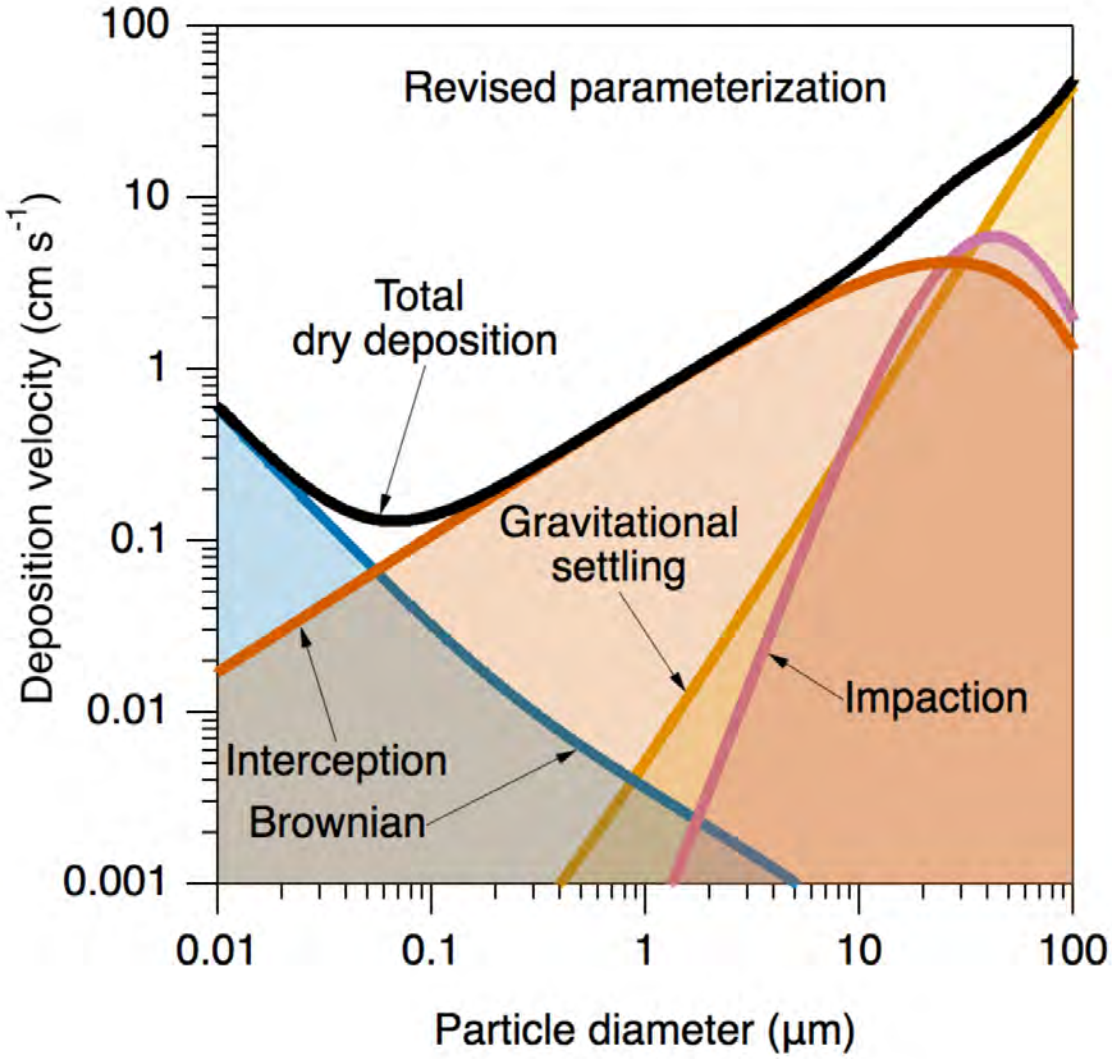
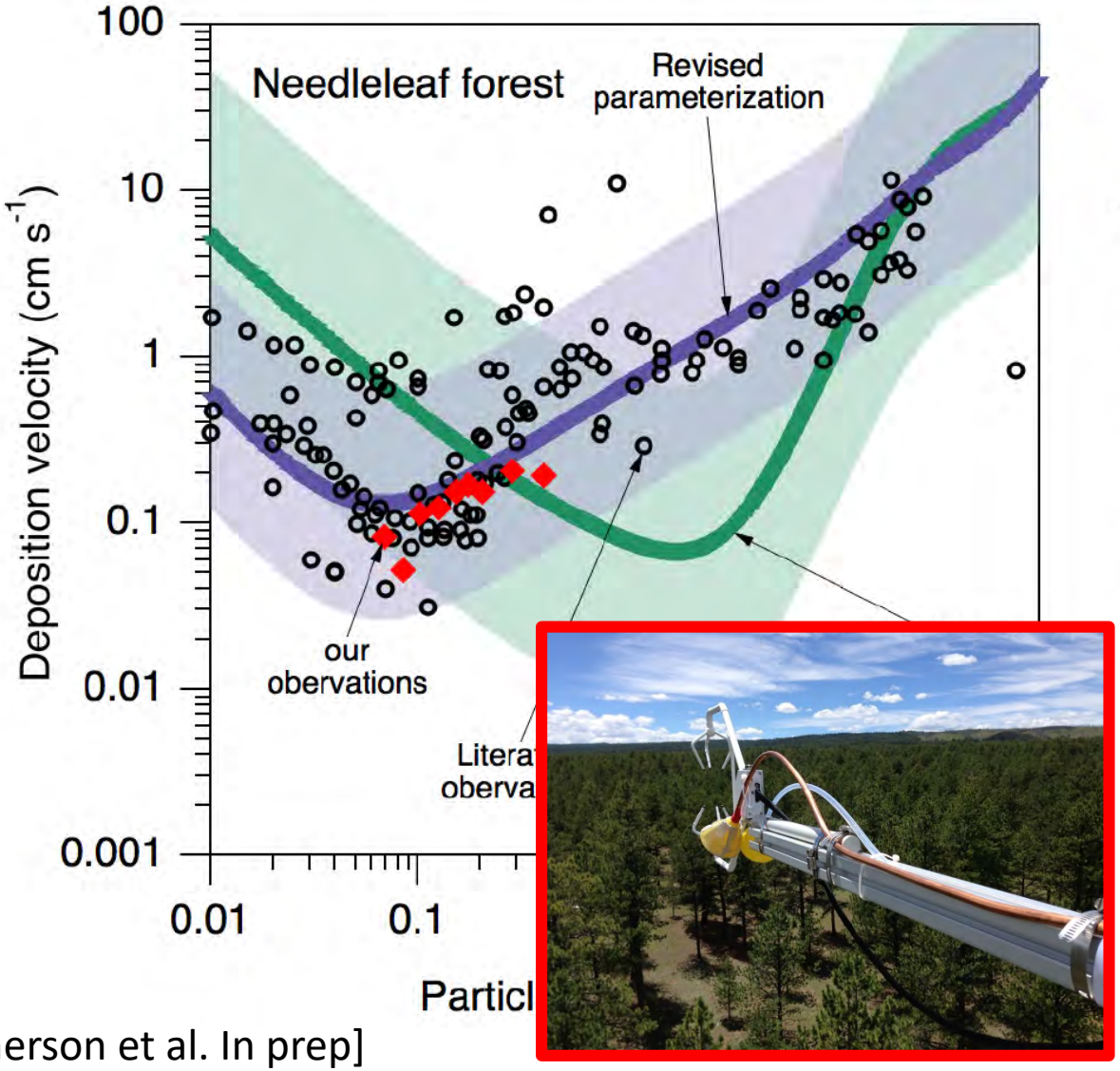
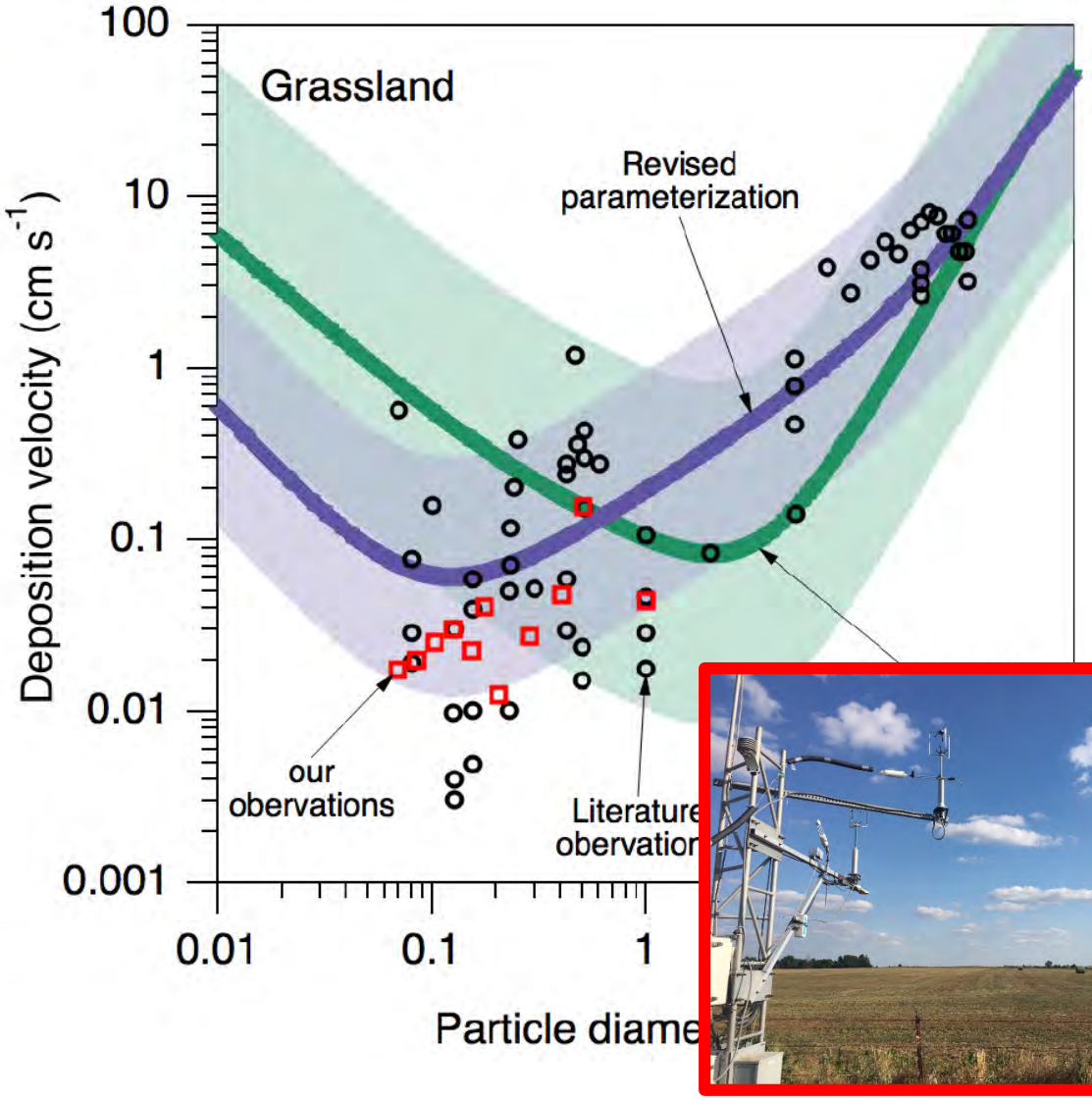
Our observations suggest current model overestimates of Brownian diffusion and underestimates of interception





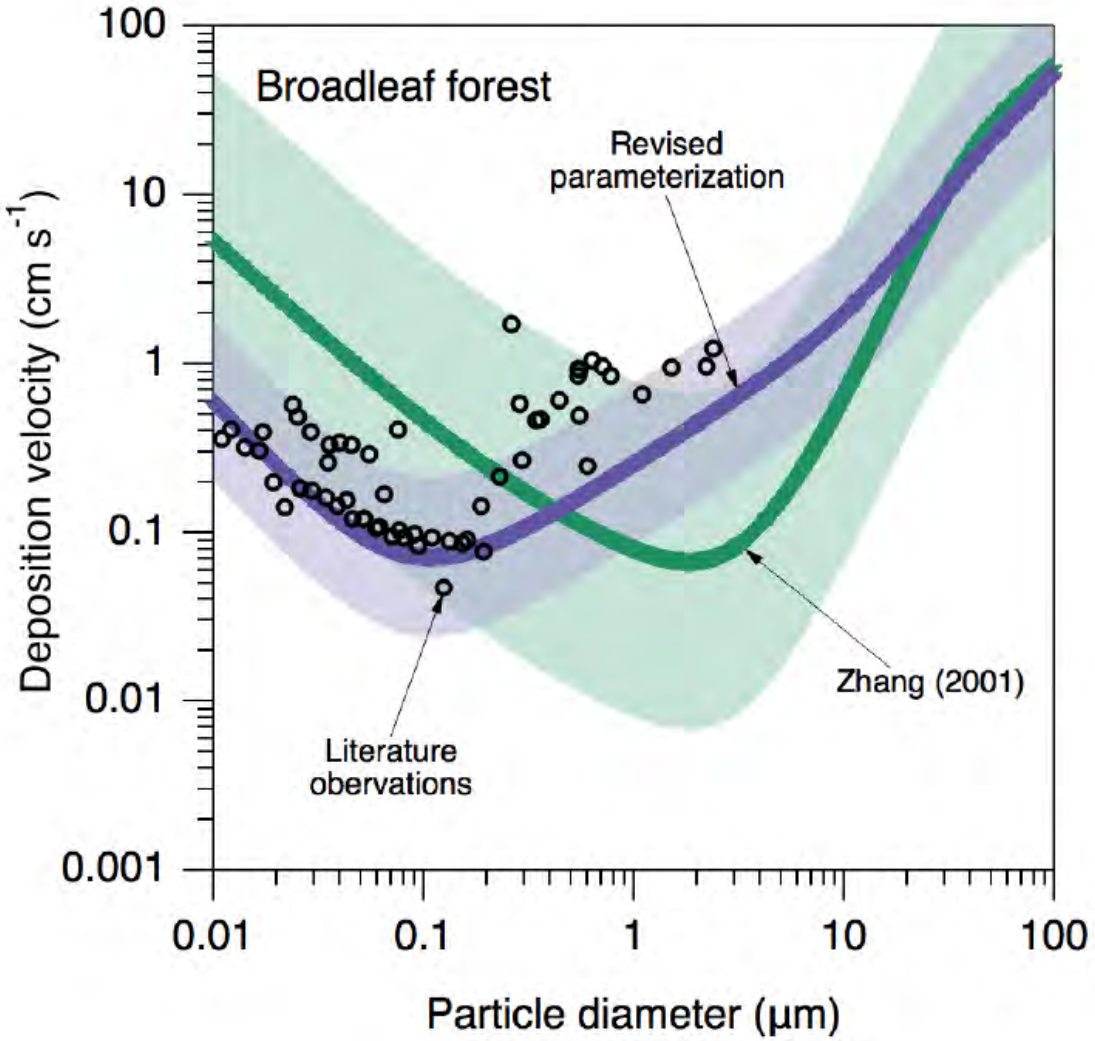
Revised vs standard parameterization for dry deposition captures data and has lower uncertainty

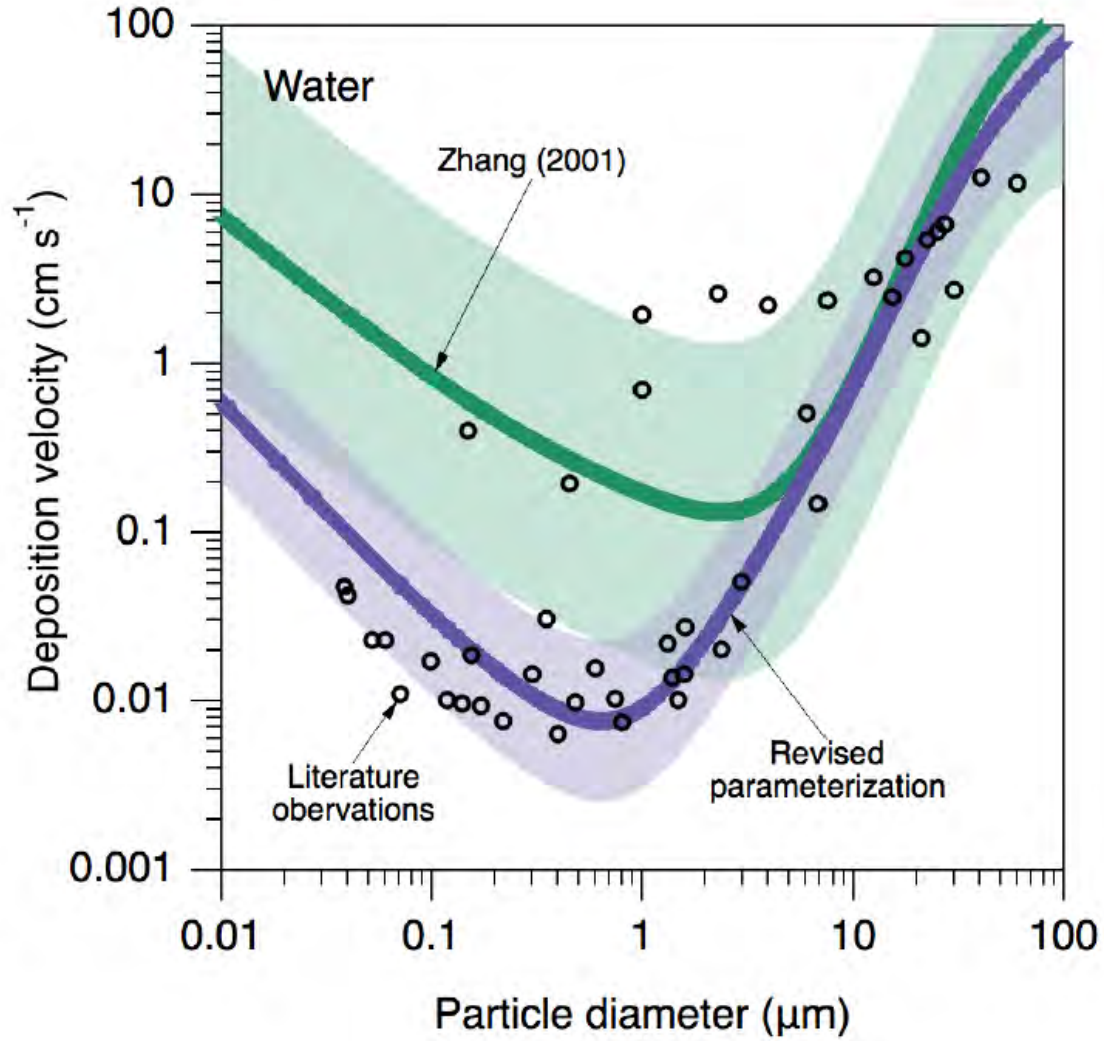




[Emerson et al. In prep]

Parameterization holds for different land cover types, but data over oceans are limited





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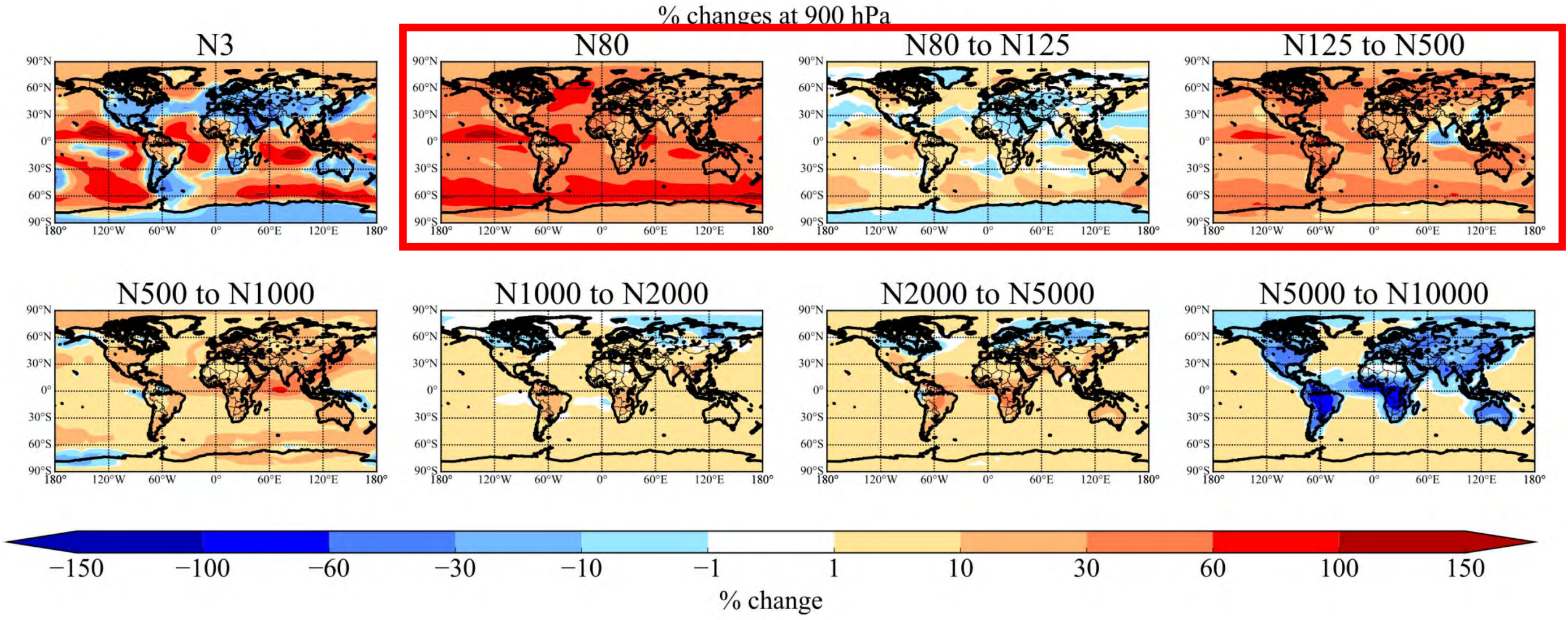
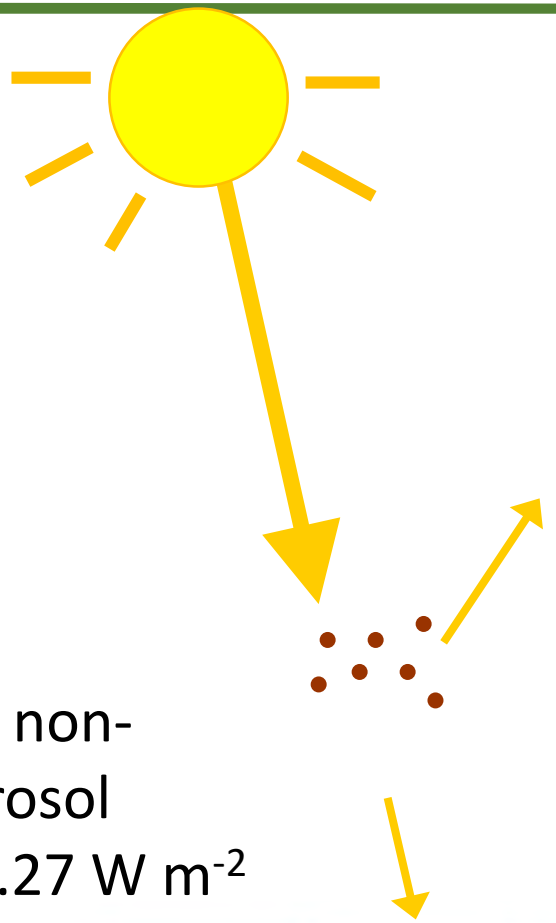
Revised particle dry deposition parameterizations have a substantial effect on modeled aerosols…



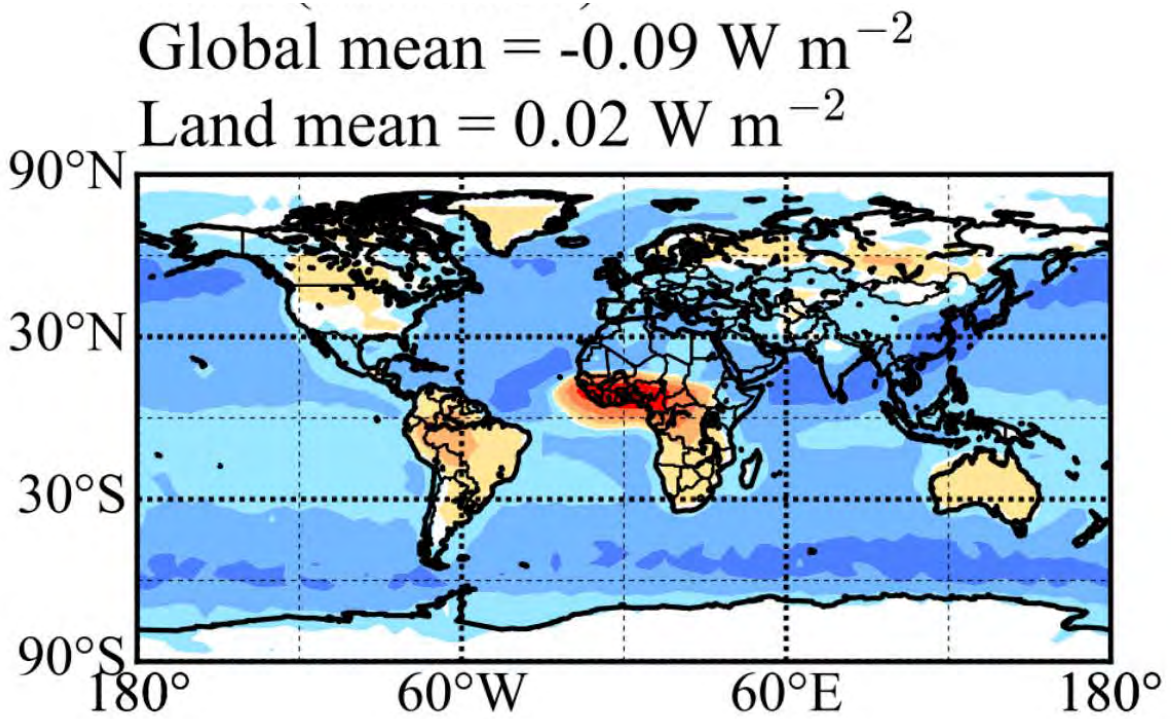
*Collaboration with Anna Hodshire + Jeff Pierce (CSU); [Emerson et al. In prep]

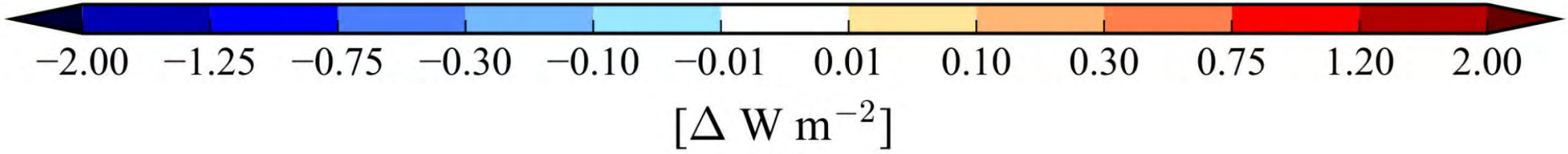
Dry deposition impacts size-resolved [particle], and thus the direct radiative effect – current models overestimate cooling effect over land



IPCC AR5 non-cloud aerosol effect: -0.27 W m^{-2}

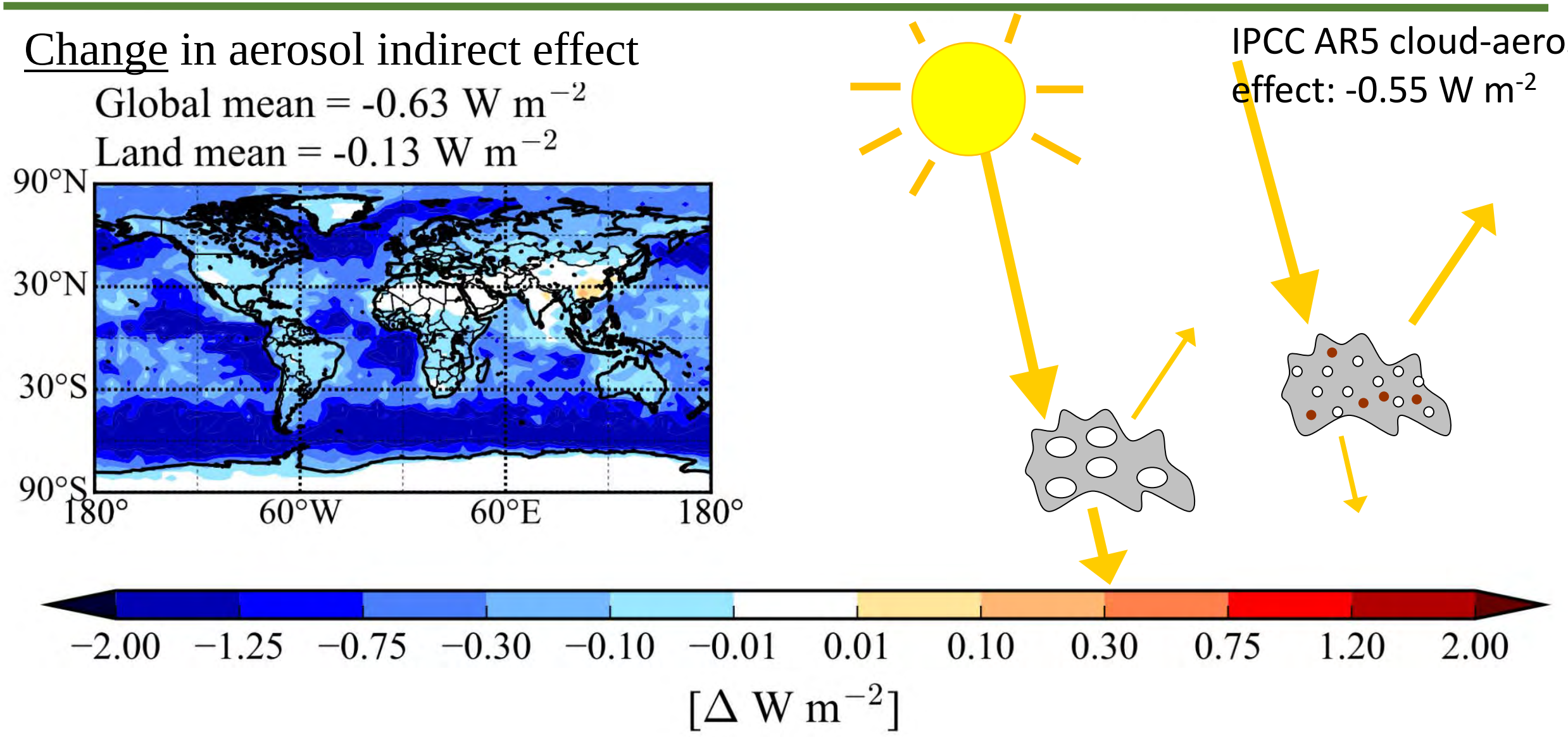
Change in aerosol direct effect (core-shell)





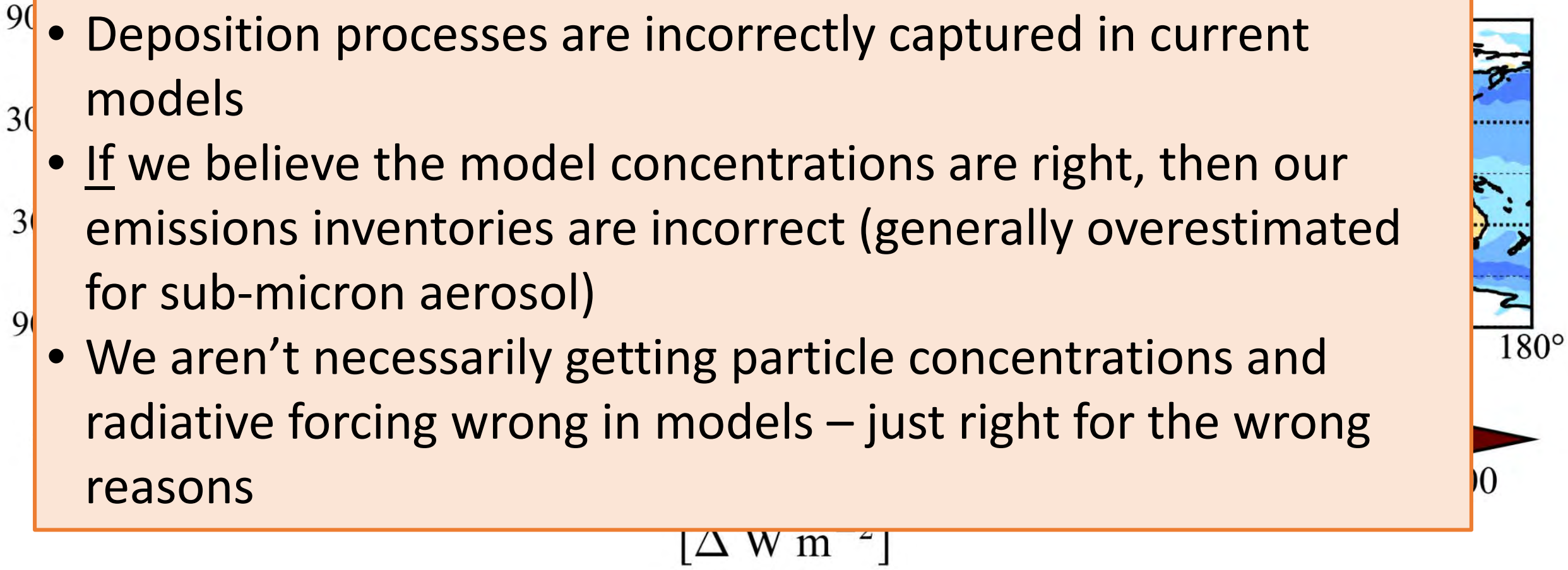
Global cooling, driven by deposition over ocean, which is highly uncertain due to lack of measurements

Changing the dry deposition parameterization changes the aerosol indirect effect – current models underestimate cooling



Changing the dry deposition parameterization changes radiative forcings substantially – but is this real??

- Particle concentrations are the result of deposition AND emission AND chemistry
- Deposition processes are incorrectly captured in current models
- If we believe the model concentrations are right, then our emissions inventories are incorrect (generally overestimated for sub-micron aerosol)
- We aren't necessarily getting particle concentrations and radiative forcing wrong in models – just right for the wrong reasons



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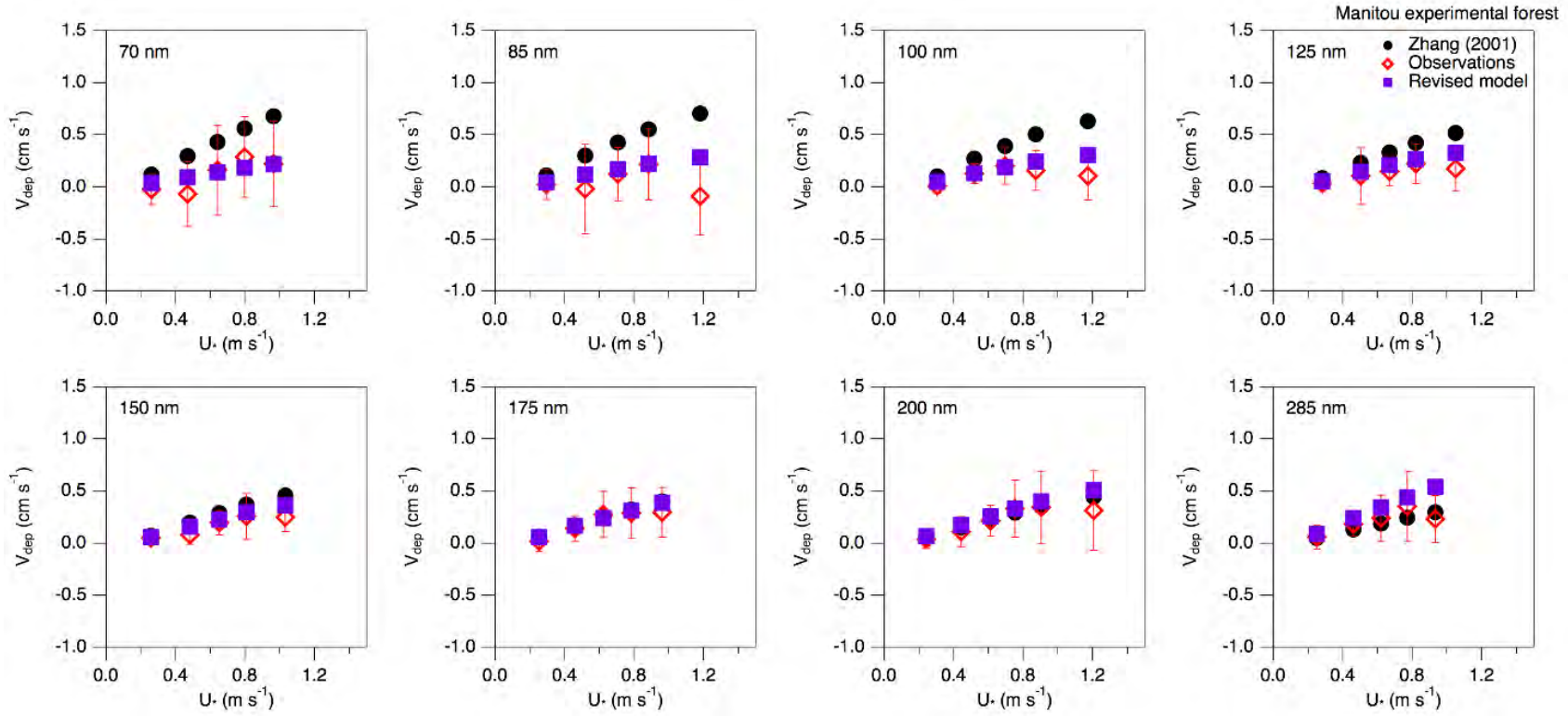
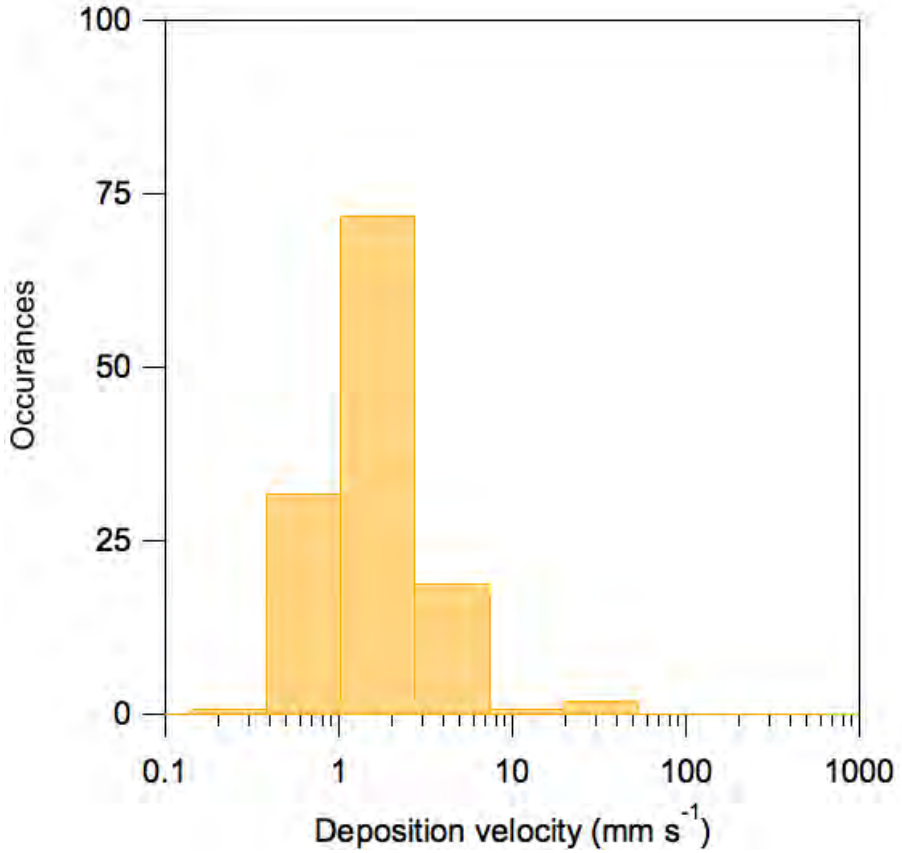


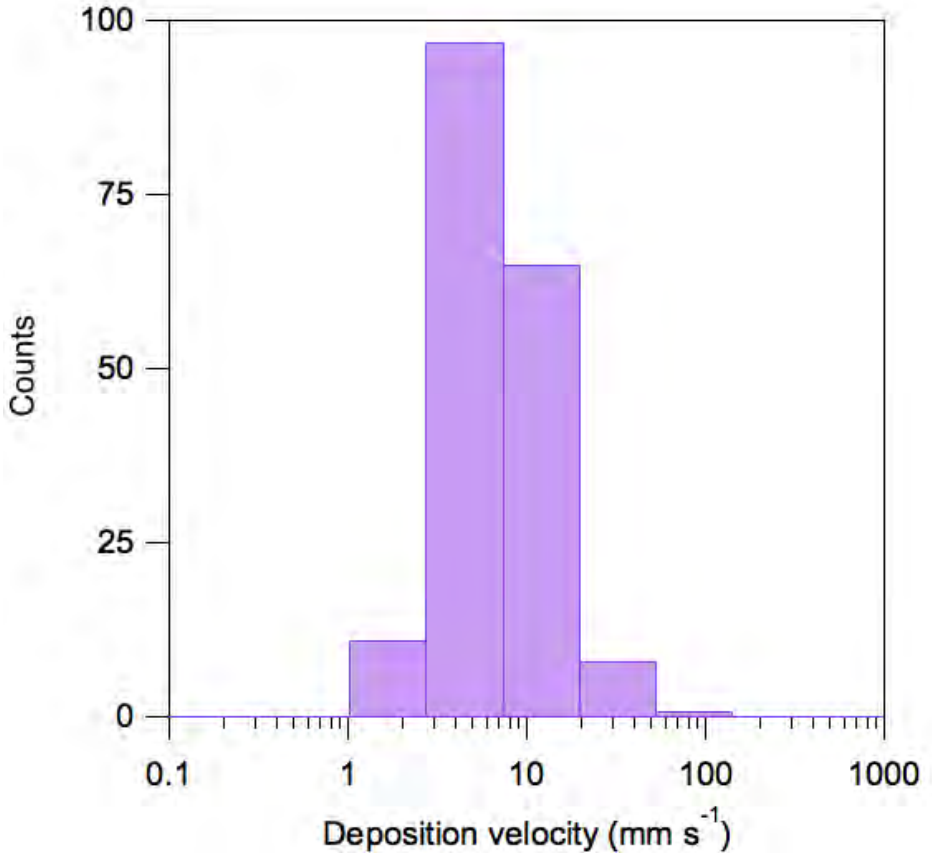
For example, turbulence (and friction velocity u* in particular) plays a strong role in the size-dependent dry deposition

Deposition velocity measures efficiency of removal

V_{dep} (rBC Particle #)
Average: 0.3 ± 0.2 mm/s
Exclude upward fluxes: 1.6 ± 0.3 mm/s



V_{dep} (rBC Particle Mass)
Average: 0.3 ± 0.2 mm/s
Exclude upward fluxes: 3.5 ± 0.3 mm/s



Comparable to model BC $V_{dep,dry}$
1 mm/s† ; 0.1-0.7 mm/s‡

† Reddy and Boucher (2004); Huang et al. (2010) ‡ Liu et al (2011)

Compare to removal by wet deposition:
$V_{dep,\ wet}$ = 6-10 mm/s