

## Comments Received from Reviewer #2

We thank Bruce Hicks for carefully reviewing our manuscript and providing valuable suggestions. We have revised the manuscript following his comments. Our responses are compiled in the supplement, where we have addressed each comment individually and detailed the respective changes to the manuscript (**RC: reviewer's comment**, **AR: author's response**).

**RC:** This is a well written paper, with conclusions that are important and with which I agree. The introduction gives one of the best overviews of the development of dry deposition formulations that I have come across. The text introduces a new (to me) method for ordering the sensitivity of a model output to the various input properties, and concludes (no surprise) that  $u^*$  is a key property. Since  $u^*$  is a measure of the level of turbulence affecting the eddy transport of all atmospheric properties, this is an expected result. However, it leads us further down a path that seems (to me) to have no foreseeable end, since  $u^*$  is among the most difficult of all atmospheric properties to quantify, both instrumentally and even more so by using available models or even tabulations. The following observations arose during my reading of the mss.

**AR:** We understand and agree with your concern regarding difficulties in measuring friction velocity ( $u^*$ ). We revised the opening paragraph of the conclusion section to reflect on your comment as follows (lines 878-883):

“One could argue that, if the different models exhibited greatest sensitivities to different parameters, and those parameters were more uncertain, the models exhibiting greatest sensitivity to the least certain parameters would be the most uncertain. In this way, sensitivity plays a potential role in determining which model is better. However, because our results showed that all models were most sensitive to  $u^*$ , or, at large size,  $RH$ , sensitivity does not end up playing a role in assigning which model is best. We also note that accurate measurement of  $u^*$  is extremely challenging (Andreas, 1992; Weber, 1999), and there exists ambiguity in its definition in boundary-layer meteorology (Weber, 1999).”

**RC:** The first sentence of the introduction could be omitted.

**AR:** We have omitted the first sentence.

**RC:** Line 33. Suggest omit “accurate

**AR:** We omitted the word as suggested (line 31) in the revised manuscript.

**RC:** Line 36. “particles” – plural.

**AR:** We have corrected the grammatical error (line 34) in the revised manuscript.

**RC:** Line 41. The time scale as stated is that which relates to gravitational settling. It is not appropriate for small particles, for which the relevant time scale needs to take into account the

depth of the layer in which the particles are mixed and the various resistances associated with their deposition. In any case, I do not see the need to refer to the time scale here.

**AR:** We agree with this comment. The equation (previously denoted as Eq. 1) stating the time scale for dry deposition is now omitted. The subsequent equations are re-numbered.

**RC:** Line 49. Terrain complexity is a central factor that none of the existing formulations Address.

**AR:** We agree that the issue of terrain complexity is not addressed in the five parameterizations we evaluated. To address this limitation, we have included the following sentences (lines 58-62):

“In all these models, the conventional resistance-based approach does not consider surface inhomogeneity or terrain complexity (i.e., deposition over flat terrain is assumed). However, Hicks (2008) argued about the importance of considering terrain complexity in dry deposition models because the assumption of surface homogeneity in existing deposition models limits the accuracy of pollutant load estimation in sensitive ecosystems that are located in complex terrain (e.g., on mountaintops or hills).”

**RC:** Line 57. “. . . there remain . . . “ singular

**AR:** We have corrected the grammatical error (line 63) in the revised manuscript.

**RC:** Line 66. There is a recent paper in JGR (lead author – Hicks) that might shed some light on the way in which data have or have not agreed with model predictions.

**AR:** Thanks for pointing out to this reference. We have included the following sentences (lines 70-72):

“In a recent study, Hicks et al. (2016) compared five deposition models with measurements conducted over forests. They found that for particle sizes less than ca. 0.2  $\mu\text{m}$ , the modeled deposition velocities agreed fairly well with measured velocities. The largest discrepancy was observed for particle sizes of 0.3 to ca. 5.0  $\mu\text{m}$ .”

**RC:** Lines 83-85. My immediate reaction is to wonder why the benefits of multi-variable partial correlation are not mentioned here. This attributes the variance among the contributing factors so that an ordering becomes obvious. The implication of the text is that a single correlation analysis is not appropriate because of the various covariances that could contribute. To my mind, this is precisely what a multiple regression is intended to resolve.

**AR:** For linear models, multiple variable partial correlation coefficients (PCC) provides the strength of linear relationship between the output and the parameter being tested for its sensitivity while removing the effects from all other input parameters (Brevault et al., 2013). Because the dry deposition models are non-linear, sensitivity analysis using PCC cannot provide accurate ranking of model input parameters in terms of their sensitivity to model outputs.

Rather, in non-linear models, a variance-based global sensitivity test approach such as Sobol' method for sensitivity analysis is more appropriate to achieve accurate parameter ranking and quantify the relative contributions of the input parameters to model outputs.

**RC:** Line 96 and onwards. This is the best description of what the models actually assume that I have come across. Congratulations to whoever it was who did the grunt work. Well done! The detail elevates my own fear that the modelers are constructing simulations that reflect articles of faith rather than of evidence. I appreciate that it is comparative examinations like that of the present text that might well shed some informed light on the benefits of model complexity. However, I have yet to see a reason not to start with something simple and add complexities as observations then warranted.

**AR:** Thanks for this comment.

**RC:** I recommend using readily apparent subheadings for each of the model descriptions.

**AR:** We have re-labeled the subheadings (sections 2.1 to 2.4) for each of the models in the revised manuscript.

**RC:** Section 3 – Methods. I note that the selection of data requires that each dataset contains measurements of all of the variables whose relevance is IMAGINED by the modelers. I consider this to be rather limiting. Surely, the selection of variables measured in field studies is a measure of what the experimentalists thought to be important factors. Examination of this might constitute a Delphic approach to determining what is important. I note, also, that requiring measurements of such things as leaf area index (LAI) assumes an application to situations in which a vegetated canopy of some specific kind dominates. For example, it has long been known that the hairs on leaves can play a role. How are these considered in the context of a LAI?

**AR:** Thank you for this comment. The presence of leaf hairs or trichomes is not considered in the context of LAI in any of the dry deposition models we evaluated in this study. We came across several measurement studies (Rauret et al. 1994; Howsam et al. 2000; Meyers et al. 2006; Burkhardt, 2010) that suggested enhanced particle deposition in the presence of trichomes. In fact, trichomes along with other microroughness elements such as epicuticular wax could increase fine particle deposition rates. We believe that the complexity associated with interactions between particle deposition and leaf microroughness elements poses a major challenge in dry deposition models to account for such interactions in a mechanistic fashion. That said, in our work, we stated that we did not consider the structural uncertainty (that which may arise from inadequate formulation of deposition processes) of the dry deposition models while evaluating their performance (lines 807-808).

**RC:** The definitions and roles of such properties as the zero plane displacement and the roughness length remain subjects of sometime heated discussion in meteorological circles. Using these concepts rather cavalierly in dry deposition models seems a poorly justified

extrapolation of a very poor basic understanding. I would welcome a more kindly-phrased reflection of this theme somewhere in the present text.

**AR:** To address your comment, we have included the following discussion (lines 796-807) in the revised manuscript:

“Additional uncertainties in the modeled deposition velocities could arise from inadequate model formulation and/or inappropriate use of certain micrometeorological parameters. For example, in dry deposition models (such as *PZ10*),  $d$  and  $z_0$  are often calculated as a fraction of canopy height ( $h$ ), and are often taken as  $d \approx 2h/3$  and  $z_0 \approx 0.1h$ . These expressions are valid for dense canopies (Katul et al., 2010). If the leaf area density is highly skewed or shows a bimodal distribution, such approximations cannot be used (Katul et al., 2010). In addition, the parameter values of  $d$  and  $z_0$  are subject to large uncertainty and are often very difficult to measure in urban areas (Cherin et al., 2015). Therefore, caution must be taken while using constant  $d$  and  $z_0$  values from lookup tables. Also, current deposition models do not explicitly treat terrain complexity in their formulations. Hicks (2008) argued that conventional use of  $d$  and  $z_0$  for non-flat terrain such as mountains is not appropriate in that context. In addition, experimentally derived values of  $d$  and  $z_0$  often represent local characteristics. Thus, it poses a challenge to scale those up in a model grid cell (Schaudt and Dickinson, 2000) of atmospheric transport models. Using remote sensing, robust scaling of these parameter values is achieved, which could be used to acquire representative values in a model grid cell (Tian et al., 2011).”

**RC:** Line 345. The wind tunnel studies CANNOT be considered in the same context as open water deposition. One cannot get 2-m white-capped waves in a wind tunnel.

**AR:** We agree that the conditions are likely to be different between a wind tunnel and the field in the context of particle deposition over water surfaces. However, in dry deposition models (e.g., Petroff and Zhang, 2010, Zhang et al., 2001), the roughness length ( $z_0$ ) for a water surface is calculated as a function of friction velocity and/or wind speed. The empirical expressions commonly used to estimate  $z_0$  for water surfaces were developed based on laboratory or wind-wave tunnel experiments (Uneo and Deushi, 2003). For example, in the Petroff and Zhang, 2010 (*PZ10*) model, the  $z_0$  was parameterized using an expression based on a wind-profile experiment conducted on a laboratory reservoir by Charnock (1955). In Charnock’s expression, the roughness length does not depend on the wave state parameters such spectral peak frequency, wave steepness, and wave age (Uneo and Deushi, 2003). In our work, we used the formulations given by Keller and Keller (1992) to calculate roughness length for water surfaces, which were developed based on experiments conducted on a wave-tank. In their formulation, Keller and Keller (1992) also did not relate  $z_0$  to wave state parameters (e.g., spectral peak frequency). They compared the wave-tank derived  $z_0$  values with those obtained through open ocean momentum flux measurements conducted by Large and Pond (1981) and found good agreement. Given these reasons, we have included the wind-tunnel measurements in our study.

We changed the following sentence as “We note that the studies by Moller and Schumann, and Sehmel et al. were conducted in the wind tunnels, and thus the observed deposition does not (replaced *may not* with *does not*) necessarily reflect deposition under natural conditions” in the revised manuscript (line 361).

**RC:** Line 359. “. . . were used . . .”

**AR:** We have made the suggested correction (line 366) in the revised manuscript.

**RC:** Equations (63) and (64) are no more than expressions that quantify how much the averages differ. Why go into all of the mathematics when the description is so simple. And why not simply call it the amount by which the ratio of the averages differs from unity. (Defining BNMBF seems rather extreme. In practice, the distributions involved are likely to be log-normal or close to it, and hence the arithmetic average is an incorrect concept.)

**AR:** We have expressed these equations (now re-numbered as Eqs. 64-65) in their basic (simplified) forms. We have included a sentence to clarify the meaning of  $B_{NMFB}$  (lines 370-371) as suggested.

We agree that the distribution of deposition velocity ( $V_d$ ) for a wide range of particle sizes (e.g., 0.01 -100  $\mu\text{m}$ ) should exhibit a log-normal distribution (e.g., Figure 7 in Hicks et al., 2016). However, in most of the measurement studies used to evaluate model performance in our study, particle sizes ( $d_p$ ) were confined to from 0.01 to 0.80  $\mu\text{m}$ , while the number of data points for  $d_p > 1.0 \mu\text{m}$  (column 10 in Table 1) were minimal. Therefore, we think using  $B_{NMFB}$  is a reasonable approach to quantify the accuracy of the models.

**RC:** At this point, I encountered Tables 1 and 2. Some comments . . .

**RC:** What time of day are the numbers meant to represent?

**AR:** We are aware that the base values of certain parameters (e.g., relative humidity ( $RH$ ), and Monin-Obukhov length ( $L_O$ )) could exhibit more diel variability than some other parameters listed in Table 2. To provide a basis for relative comparison, the base parameter values listed in Table 2 were used to perform uncertainty analysis, with the objective of demonstrating the overall uncertainties in modeled deposition velocities in the five parameterizations across five LUCs. These base values used for uncertainty analysis should be viewed as reasonable, arbitrary choices within the range of the reported values of those parameters in the measurement studies listed in Table 1. Here, we did not explicitly differentiate between daytime vs. nighttime conditions.

**RC:** What do the uncertainty ranges mean? If they are meant to refer to long-term ensemble averages base on some other variable, then I would agree with some of them.

**AR:** Each uncertainty range denotes the degree of imprecision (in percentage) associated with each of corresponding parameters listed in Table 2. To avoid confusion, we have referred to it

as ‘uncertainty’ in Table 2 in the revised manuscript. The uncertainty values (in percentage) were obtained from a literature search and the references are listed in column 3 of Table 2.

**RC:** The humidity assumption of 80% seems very high. Unless nighttime conditions are assumed.

**AR:** In several measurement studies (e.g., Neumann and den Hartog, 1985; Ruijgrok et al., 1997; Vong et al., 2004; Grönholm et al., 2009), particle deposition was measured at both daytime and nighttime. The threshold for very high humidity (*RH*) was reported as >95% (e.g., Ruijgrok et al., 1997) in the context of their measurement study. In addition, the ranges of *RH* were reported as 35-100% and 49-90% by Grönholm et al. (2009) and Vong et al. (2004), respectively. The choice of *RH* = 80% was arbitrary, but was within the reported range of *RH* by the measurement studies listed in Table 1. In addition, at high *RH* (e.g., *RH* close to 90%), atmospheric particles are likely to grow (the phenomena commonly known as hygroscopic growth) to twice their initial dry diameter (Vong et al., 2004). The hygroscopic growth was accounted for in the models according to Eq. (15).

**RC:** However, I note that nighttime is indeed assumed: the value of *L* is positive. A value of 50 m is commonly VERY stable.

**AR:** We understand your concern regarding the choice of Monin-Obukhov length (*L<sub>o</sub>*) value as 50 m for the uncertainty test. In this study, we denoted uncertainties in the modeled *V<sub>d</sub>* in normalized terms (i.e., the difference between the 95<sup>th</sup> and 5<sup>th</sup> percentiles *V<sub>d</sub>* was divided by the 50<sup>th</sup> percentile *V<sub>d</sub>*) rather than in absolute terms (i.e., the difference between the 95<sup>th</sup> and 5<sup>th</sup> percentiles *V<sub>d</sub>* or the central 90% values of *V<sub>d</sub>*). Therefore, the choice of *L<sub>o</sub>* has minimal effect on the normalized uncertainties reported in Table 10.

**RC:** I could argue with all of the other “base values,” all of which represent the environment that some investigator saw out his window. For example, grass 50 cm tall seems very unlikely. At that stage, it should have been harvested, grazed or mowed.

**AR:** The measurement studies used for model evaluation on grass reported typical values of grass height as 30-60 cm (e.g., Neumann and den Hartog, 1985; Vong et al., 2004). We agree that grass height of 50 cm could be higher than average height, but it was not uncommon in particle deposition measurement studies. For example, an article by Wesely et al. (1985) (not used in study) reported sulfur deposition on grass with height ranging from 40-50 cm.

**RC:** The use of a one-sided LAI worries me a little. Particles will deposit to both sides of leaves. Why use the one-sided value?

**AR:** We used one-sided LAI because atmospheric transport models (e.g., GEOS-Chem) regularly use MODIS derived LAI values as inputs to the dry deposition models. According to MODIS LAI database (<https://modis.gsfc.nasa.gov/data/dataproduct/mod15.php>), “LAI is defined as one-sided green leaf area per unit ground area in broadleaf canopies and as half the total needle surface

area per unit ground area in coniferous canopies.” In addition, reported values of LAI in the measurement studies (listed in Table 1) over vegetative surfaces often do not differentiate between two vs. one-sided LAI. Thus, to avoid ambiguity, we used one-sided LAI.

**RC:** In Table 2, I hope that the separate listing of ranges for  $u$  and  $u^*$  does not mean that these are allowed to vary independently. The friction velocity is usually from 4% of wind speed (over water) to 20% of the wind speed (over a forest), with the variation of this proportionality being well known. It changes from day to night of course, but it seems that the analysis presented here is only for nighttime.

**AR:** The separate uncertainty ranges (now, for clarity, denoted as ‘uncertainty’ in Table 2, as compared to ‘uncertainty range’ in the previous manuscript) for wind speed ( $U$ ) and friction velocity ( $u^*$ ) denote respective imprecisions associated with those two parameters. In the uncertainty analysis presented in this paper, we used these two parameters independent of each other by allowing them to vary only within their specified imprecision/uncertainty ranges. The objective of the uncertainty analysis was to quantify the effects of imprecision in measured values of the input parameters (e.g.,  $U$ ,  $u^*$ ,  $RH$ ,  $L_0$ , etc.) to the modeled  $V_d$ . Thus, we did not explicitly distinguish between daytime and nighttime periods.

**RC:** The variation of  $L$  from 10 to 100 m is very constraining, since once again the analysis represents conditions that would often be considered uncommonly stable.

**AR:** The analysis presented here assumed near-neutral conditions. The range (10 to 100 m) for  $L_0$  (from stable side) was consistent with reported range for neutral conditions ( $-200 < L_0 < 200$ ) by Pryor et al. (2007) (study #13 in Table 1). In addition, we did not find any change in parameter rankings reported in Table 11 by increasing the upper bound value of  $L_0$  (in the assumed range of 10-100 m) by factors of 10 to 20.

**RC:** The roughness length quoted as the lower limit for snow/ice is 0.02 mm. As far as I know, this is less than aerodynamically smooth, and is therefore unlikely (some would say impossible). Please be careful.

**AR:** The lower limit of the roughness length ( $z_0$ ) is consistent with the reported  $z_0$  values in the measurement studies of particle deposition on ice/snow (e.g., Fig. 2a in Held et al., 2001a (study#24 in Table 1)). In addition, the article by Tjernström (2005) reported the lower bound  $z_0$  value as 0.01 mm from Arctic Ocean Experiment of 2001.

**RC:** In Table 3, I note that all of the field programs are for daytime conditions, yet the Base Values tabulated indicate an assumption of stable (nighttime) conditions. Something must be wrong. A suitable explanation would be that the sign for  $L$  is incorrect.

**AR:** In several measurement studies listed in Table 1 (e.g., Neumann and den Hartog, 1985; Ruijgrok et al., 1997; Vong et al., 2004; Grönholm et al., 2009), particle deposition was

measured under both daytime and nighttime conditions. The analysis presented here assumed stable conditions, thus the sign for  $L_0$  is correct.

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