Evaluation of five dry particle deposition parameterizations for incorporation into atmospheric transport models

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Abstract. Despite considerable effort to develop mechanistic dry particle deposition parameterizations for atmospheric transport models, current knowledge has been inadequate to propose quantitative measures of the relative performance of available parameterizations. In this study, we evaluated the performance of five dry particle deposition parameterizations developed by Zhang et al. (2001) (Z01), Petroff and Zhang (2010) (PZ10), Kouznetsov and Sofiev (2012) (KS12), Zhang and He (2014) (ZH14), and Zhang and Sao (2014) (ZS14), respectively. The evaluation was performed in three dimensions: model ability to reproduce observed deposition velocities, \( V_d \) (accuracy), the influence of imprecision in input parameter values on the modeled \( V_d \) (uncertainty), and identification of the most influential parameter(s) (sensitivity). The accuracy of the modeled \( V_d \) was evaluated using observations obtained from five land use categories (LUCs): grass, coniferous and deciduous forests, natural water, and ice/snow. To ascertain the uncertainty in modeled \( V_d \), and quantify the influence of imprecision in key model input parameters, a Monte Carlo uncertainty analysis was performed. The Sobol’ sensitivity analysis was conducted with the objective to determine the parameter ranking, from the most to the least influential. Comparing the normalized mean bias factors (indicator of accuracy), we find that the ZH14 parameterization is the most accurate for all LUCs except for coniferous forest, for which it is second most accurate (BSMBF = -2.31). From Monte Carlo simulations, the estimated mean normalized uncertainties in the modeled \( V_d \) obtained for seven particle sizes (ranging from 0.005 to 2.5 μm) for the five LUCs are 17%, 12%, 13%, 16%, and 27% for the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations, respectively. From the Sobol’ sensitivity results, we suggest that the parameter rankings vary by particle size and LUC for a given parameterization. Overall, for \( d_p = 0.001 \) to 1.0 μm, friction velocity was one of the three most influential parameters in all parameterizations. For giant particles \( (d_p = 10 \mu m) \), relative humidity was the most influential parameter. Because it is the least complex of the five parameterizations, and it has the greatest accuracy and least uncertainty, we propose that the ZH14 parameterization is currently superior for incorporation into atmospheric transport models.

1. Introduction

Of the processes involved in the atmosphere-surface exchange of gaseous species and particles, dry deposition refers to the process by which aerosols are removed from the atmosphere in the absence of precipitation through vertical turbulent transport and interaction with the earth’s surface. Dry deposition is a complex process that is influenced by the chemical properties of aerosols and their sources, meteorological conditions, and surface characteristic features. The transference of particles from the atmosphere to the earth’s surface is controlled by forcings such as frictional drag and terrain induced flow modification (Giorgi, 1986; Stull, 1988). Understanding the processes and factors controlling dry deposition is necessary to accurately estimate the residence time of atmospheric particles, which governs their atmospheric transport distance, trans-boundary fluxes, and potential climate effects (IPCC, 2001; Nemitz et al., 2002; Pryor et al., 2008). An accurate estimation of dry deposition is also needed to quantify the atmospheric loads of particle containing sulfate, nitrate, and ammonium that contribute to acidification and eutrophication of ecosystems, toxic elements such as Pb, Zn, and Cd, and base cations such as Na⁺, K⁺, Ca²⁺, and Mg²⁺ that alter the nutrient cycling in soil (Ruijgrok et al., 1995; Petroff et al., 2008a).
Over the last three decades, several indirect and direct methods were developed to measure dry particle deposition flux to ecosystems (McMahon and Denisot, 1979; Sehmel, 1980; Gallagher et al., 1997; Zhang and Vet, 2006; Pryor et al., 2008). In particle flux measurements, the timescale for dry deposition ($\tau_{dep}$) for a given measurement height $z$ is estimated as:

$$\tau_{dep}(z) = \frac{z}{V_d}$$

(1)

where the dry deposition velocity $V_d$ at height $z$ is defined as the ratio of the total flux $F(z)$ divided by the particle concentration at the same height $C(z)$ (Pryor et al., 2013; Rannik et al., 2016) and is mathematically expressed as:

$$V_d = \frac{F(z)}{C(z)},$$

(2)

One of the major limitations of direct flux measurement is limited spatial coverage because the measurement stations are confined to only a limited number of sites (Nemitz et al., 2002). The application of spatially and temporally resolved 3-D atmospheric transport models, from regional to global scale, can produce estimates of the dry deposition fluxes for a suite of atmospheric species over various natural surfaces such as bare soil, grass, forest canopies, water, and ice/snow. To predict the dry deposition fluxes using atmospheric transport models, a parameterization/scheme that can adequately account for the major physical processes of particle deposition (e.g., turbulent diffusion, gravitational settling, interception, impaction, and Brownian diffusion) must be embedded in the host model.

Many dry particle deposition models have been developed for scientific research and operational purposes (see model review by Petroff et al., 2008a). In mechanistic or process-based dry deposition models, an electrical resistance based approach is widely used to parameterize the dry particle deposition velocity (Venkatram and Pleim, 1999). In this approach, dry particle deposition occurs via two parallel pathways: turbulent diffusion (expressed as aerodynamic resistance) and gravitational settling (expressed as resistance due to gravitation). In addition, particle collection by surfaces via Brownian diffusion, interception, and impaction are represented using separate surface resistance terms (Slinn, 1982; Hicks et al., 1987; Wesely and Hicks, 2000; Zhang et al., 2001; Seinfeld and Pandis, 2006; Petroff and Zhang, 2010; Zhang and He, 2014).

Despite considerable efforts in developing dry particle deposition parameterizations of varying complexity, there remains considerable gaps in systematic performance evaluation of existing schemes with reliable field measurements. We note that the evaluation of dry deposition parameterizations with field measurements is very limited and not up to date. Van Aalst (1986) evaluated the performance of six dry deposition parameterizations against field measurements, and reported large discrepancies in terms of the modeled deposition velocities. He reported that over water surfaces the modeled deposition velocities for 1.0 µm particles by the Williams (1982) scheme were factors of 10 to 50 higher than those predicted by the Sehmel and Hodgson (1978) scheme. For forest canopy, the Wiman and Agren (1985) model over-predicted the deposition velocities of the Slinn (1982) model by a factor of five. Studies also suggest that in many dry deposition parameterizations, the largest uncertainty exists for 0.1-1.0 µm particles because of the differing treatments of some key particle deposition processes such as Brownian diffusion (Van Aalst, 1986; Petroff and Zhang, 2010; Zhang and Sao, 2014).

Uncertainty in modeled dry deposition velocities is an area that requires a thorough investigation. Only a few studies have been conducted in quantifying the uncertainties in dry particle deposition parameterizations. Ruijgrok et al. (1992) performed an uncertainty evaluation of the Slinn (1982) model by assessing the variabilities in nine input parameters to the model outputs. Using Slinn's model, Gould and Davidson (1992) determined the influence of uncertainties in the size of the collection elements, roughness length, canopy wind profile and wind speed on the modeled deposition velocities. As far as we know, a detailed uncertainty analysis to address the influence of varying particle size, meteorological conditions, and surface features has not been performed on existing dry deposition parameterizations. The results from an uncertainty analysis could be used as one of the
model’s performance indicators, and help guide the modeling community to adequately account for uncertainties in the modeled deposition fluxes of pollutants to ecosystems.

Sensitivity analysis is often performed to determine the most influential parameters to the model outputs. Typically, a dry particle deposition model incorporates a large number of input parameters, which are subject to variability. In addition to identifying the most sensitive parameter(s), a sensitivity analysis can provide important insight as to the processes that control the overall deposition process, and identify those that may require further improvement. However, a detailed sensitivity test that encompasses exploring the entire parameter spaces of the input parameters of a dry deposition parameterization has not yet been performed. Some researchers conducted one-at-a-time (OAT) sensitivity analysis test (SA) (Ruijgrok et al., 1997; Zhang et al., 2001) of dry deposition models. In OAT-SA, the effect of varying one model input parameter is tested at a time while keeping all others fixed (Saltelli and Annoni, 2010). Because in reality the variabilities in a set of model input parameters are expected to occur simultaneously, a OAT-SA is not a useful tool to determine the most influential parameter(s) in the deposition models. Rather, a global sensitivity test approach is needed. In global sensitivity analysis, the potential effects from simultaneous variabilities of model input parameters over their plausible range is considered (Lilburne and Tarantola, 2008).

In the present study, five dry particle deposition parameterizations, developed by Zhang et al. (2001), Petroff and Zhang (2010), Kouznetsov and Sofiev (2012), Zhang and He (2014), and Zhang and Sao (2014), are selected for an intercomparison of performance in terms of accuracy, uncertainty, and sensitivity. Throughout this paper, these models are referred to as Z01, PZ10, KS12, ZH14, and ZS14, respectively. The objectives of this study are threefold. The first objective is to evaluate the accuracy of five dry particle deposition parameterizations using measured dry deposition velocities obtained from field observations. Data of measured deposition velocities were collected from the literature, which comprised of measurements conducted over land use categories (LUCs) including grass, coniferous and deciduous forests, natural waters, and ice/snow. The second objective is to perform an uncertainty analysis of the modeled dry deposition velocities related to imprecision in model input parameter values. The third objective is to quantify the most influential parameters in the modeled dry deposition velocities by applying a global variance-based sensitivity analysis.

2. Background

The five dry particle deposition schemes used in this paper are described briefly below. For each scheme, the major expressions/equations used to compute the dry particle deposition velocities are provided.

2.1. The Z01 scheme

The Z01 scheme estimates dry particle deposition velocity as a function of particle size and density, meteorological variables, and surface properties. In the Z01 scheme, the dry particle deposition velocity \( V_d \) is expressed as:

\[
V_d = V_g + \frac{1}{R_a + R_s} \tag{3}
\]

where \( V_g \) is the gravitational settling velocity, \( R_a \) is the aerodynamic resistance above the canopy, and \( R_s \) is the surface resistance. The expression for gravitational settling velocity \( V_g \) is given as:

\[
V_g = \frac{n_d \rho g \eta_v}{18 \nu} \tag{4}
\]

where \( \rho \) is the dry density of the particle, \( d_p \) is the particle aerodynamic diameter, \( g \) is the gravitational acceleration, \( C \) is the Cunningham correction factor, and \( \eta_v \) is the temperature dependent viscosity coefficient of air. The correction factor \( C \) is applied to account for the molecular structure of the air and is expressed as:

\[
C = 1 + \frac{2a}{d_p} \left( 1.257 + 0.4e^{-0.559d_p} \right) \tag{5}
\]
where $\lambda$ is the mean free path of air molecules.

The aerodynamic resistance ($R_a$) is calculated as:

$$R_a = \frac{\ln(z_R^2) - \psi_H}{u_*}$$

where $z_R$ is the reference height where $V_d$ is typically computed, $z_R$ is the roughness height, $\kappa$ is the von Kármán constant, $u_*$ is the friction velocity, and $\psi_H$ is the stability function for heat. The expression for $\psi_H$ is: $\psi_H = 2\ln[0.5(1 + (1 - 16x)^{0.5})]$ when $x \in [-2; 0]$, and $\psi_H = -5x$ when $x \in [0; 1]$. Here, $x = z/L_O$, where $z$ is the measurement height and $L_O$ is the Monin-Obukhov length.

The surface resistance term, $R_s$ in Eq. 3, is a function of particle collection efficiencies due to Brownian diffusion ($E_B$), impaction ($E_{IM}$), and interception ($E_{IN}$). Accordingly, $R_s$ is parameterized as:

$$R_s = \frac{1}{\epsilon_0 u_* (E_B + E_{IM} + E_{IN}) R_1}$$

where $\epsilon_0$ is an empirical constant and its value is taken as 3 for all LUCs, and $R_1$ is the correction factor for particle rebound, which is included to modify the collection efficiencies at the surface. $R_1$ is parameterized as a function of Stokes number ($St$) as:

$$R_1 = \exp(-St^{-0.5})$$

The parameterizations for $E_B$, $E_{IM}$, and $E_{IN}$ are expressed by Eqs. (9-11). The particle collection efficiency ($E_B$) is parameterized as a function of Schmidt number ($Sc$) as:

$$E_B = Sc^{-\gamma}$$

where $Sc$ is the ratio of kinematic viscosity of air ($\nu$) to the particle Brownian diffusivity ($D$). $\gamma$ is a LUC dependent variable, and the typical values of $\gamma$ range from 0.54 to 0.56 for rough surfaces and from 0.50 to 0.56 for smooth surfaces. Brownian diffusivity ($D$) is calculated as:

$$D = \frac{ck_B T}{3n \mu}$$(10)

where $C$ is the Cunningham correction factor as expressed by Eq. (5), $k_B$ is the Boltzmann’s constant ($1.38 \times 10^{23}$ J K$^{-1}$), and $\mu$ is the dynamic viscosity of air at temperature $T$.

For smooth surfaces, particle collection efficiency by impaction ($E_{IM}$) is parameterized as:

$$E_{IM} = 10^\frac{7}{2\pi}$$

And, for rough surfaces,

$$E_{IM} = (\frac{Se^A}{3 + 5St})^\beta$$

where $\alpha$ and $\beta$ are constants; values of $\alpha$ are LUC dependent, and $\beta$ is taken as 2. In Eqs. (11-12), the Stokes number ($St$) is expressed as:

$$St = \frac{V_d u_*}{g A}$$ (for vegetative surfaces),

$$St = \frac{V_d u_*^2}{v}$$ (for smooth surfaces),

where $A$ is the characteristic radius of the surface collector elements. The values of $A$ are given for different LUCs for various seasons by Zhang et al. (2001).

Collection efficiency by interception ($E_{IN}$) is calculated as:

$$E_{IN} = \frac{1}{2} (\frac{dp}{x})^2$$

Growth of particles under humid conditions is considered in the Z01 scheme by replacing the $dp$ with a wet particle diameter ($d_{wp}$), which is calculated as:
\[
d_w = \left[ \frac{C_1(d_w)}{2} \frac{C_2}{2} \right]^{1/3} + \frac{d_w}{2} \log \frac{C_3}{2},
\]

where \( C_1, C_2, C_3, \) and \( C_4 \) are the empirical constants (values given in Table 1 of Zhang et al., 2001), and \( RH \) is the relative humidity.

### 2.2 The PZ10 scheme

Petroff and Zhang (2010) parameterized dry particle deposition velocity using an expression similar to Eq. (3) with some improvements of the surface resistance and collection efficiency terms. In the PZ10 scheme, the effect of gravity and drift forces (e.g., phoretic effects) were taken into account by introducing the term drift velocity \( (V_{drift}) \). Thus, dry deposition velocity \( (V_d) \) at a reference height \( (z_R) \) is given as:

\[
V_d = V_{drift} + \frac{1}{\tau_a + \tau_v} \tag{17}
\]

Here, the drift velocity \( V_{drift} \) is equal to the sum of gravitational settling velocity and phoretic velocity, and the expression of \( V_{drift} \) is:

\[
V_{drift} = V_g + V_{phar}. \tag{18}
\]

\( V_g \) is calculated using Eq. (4). The LUC dependent values of \( V_{phar} \) were given by Petroff and Zhang (2010).

Surface resistance \( (R_s) \) is commonly expressed as an inverse of the surface deposition velocity, \( V_{ds} \) (i.e., \( R_s = 1/V_{ds} \)). In the PZ10 scheme, \( V_{ds} \) is parameterized as:

\[
\frac{V_{ds}}{u_*} = E_g \frac{\theta}{\theta_g} \frac{\tanh(n)}{\tanh(n)} \tag{19}
\]

The parameters (e.g., \( Q, \theta_g, \alpha, \) and \( \eta \)) used in Eq. (19) are dependent on the aerodynamic and surface characteristic features. The parameterization of the total particle collection efficiency on the ground below the vegetation \( (E_g) \) has two components: (i) collection by Brownian diffusion \( (E_{gb}) \) and (ii) collection by turbulent impaction \( (E_{gt}) \). In the PZ10 scheme, formulation of \( E_{gb} \) is expressed as:

\[
E_{gb} = \frac{Sc^{-2/3}}{14.5} \left[ \frac{1}{6} \ln \left( \frac{(1+F)^2}{1-F} \right) + \frac{1}{3} \text{Arctan} \left( \frac{2F-1}{\sqrt{3}} \right) + \frac{\pi}{6\sqrt{3}} \right]^{-1}, \tag{20}
\]

where \( F \) is a function of the Schmidt number \( (Sc) \) and is expressed as \( F = Sc^{-1}/2.9 \).

Collection efficiency by turbulent impaction, \( E_{gt} \), is a function of dimensionless particle relaxation time \( (\tau_{ph}^+) \) and a coefficient \( C_{ir} \) (taken as 0.14). In the PZ10 scheme, \( E_{gt} \) is parameterized as:

\[
E_{gt} = 2.5 \times 10^{-3} C_{ir} \Phi_{m}^{2/3} \tau_{ph}^{2/2}, \tag{21}
\]

\( \tau_{ph}^+ \) is calculated as \( \tau_{ph} = \tau_{p} u_f^2 / v \). The local friction velocity \( (u_f) \) is expressed as:

\[
u_f = u_* e^{-\alpha}, \tag{22}
\]

where \( \alpha \) is the aerodynamic extinction coefficient and is expressed as:

\[
\alpha = \frac{k_s \text{LAI}}{12 \pi^2 \left( 1 - \frac{1}{2} \right)^2} \Phi_m^{2/3} \left( \frac{h-h_0}{L_o} \right). \tag{23}
\]

In Eq. (21), \( k_s \) is the inclination coefficient of canopy elements, \( \text{LAI} \) is the leaf area index, \( d \) is the zero-plane displacement height, \( h \) is the height of the canopy, \( L_o \) is the Monin-Obukhov length, and \( \Phi_m \) is the non-dimensional stability function for momentum.

The expressions for \( \Phi_m \) is, \( \Phi_m(x) = (1 - 16x)^{-1/4} \) when \( x \in [-2; 0] \) and \( \Phi_m(x) = (1 + 5x)^{-1/4} \) when \( x \in [0; 1] \).

In Eq. (19), the non-dimensional time-scale parameter, \( Q \), is defined as the ratio the turbulent transport time scale to the vegetation collection time scale. The magnitude of \( Q \) can be used to characterize the dominant mechanism of the vertical transport.
of particles to the surface. For particle deposition over a canopy, \( Q \ll 1 \) describes a condition when homogeneous concentration of Aitken and accumulation mode particles prevails throughout the canopy. This condition occurs when turbulent mixing is very efficient and transfer of particles is limited by the collection efficiency on leaves. In contrast, \( Q \gg 1 \) characterizes a situation in which an inhomogeneous particle concentration within the canopy prevails, which is typical for coarse mode particles. Under such conditions, efficient collection of particles by leaves takes place and transfer to the surface is usually limited by the turbulent transport.

In the PZ10 scheme, \( Q \) and \( Q_B \) are parameterized using Eqs. (24) and (25), respectively:

\[
Q = \frac{L_{AI} E_T h}{l_{mp}(h)}, \quad (24)
\]

\[
Q_B = \frac{E_T h}{l_{mp}(h)}, \quad (25)
\]

where \( E_T \) is the total particle collection efficiency by various physical processes and \( l_{mp}(h) \) is the mixing height for the particles. The mixing height for particles, \( l_{mp}(h) \), is calculated as:

\[
l_{mp}(h) = \frac{\kappa(h-d)}{\Phi_h \left( \frac{h}{L_e} \right)} \quad (26)
\]

where \( \Phi_h \) is the stability function for heat and expressed as: \( \Phi_h(x) = (1 - 16x)^{-1/2} \) when \( x \in [-2; 0] \) and \( \Phi_h(x) = 1 + 5x \) when \( x \in [0; 1] \).

The total collection efficiency \( (E_T) \) is expressed as:

\[
E_T = \frac{U_h}{u_s} (E_B + E_{IN} + E_{IM}) + E_{IT}, \quad (27)
\]

where \( U_h \) is the horizontal wind speed at canopy height \( h \), and \( E_B, E_{IN}, E_{IM} \), and \( E_{IT} \) are the collection efficiencies by Brownian diffusion, interception, impaction, and turbulent impaction, respectively. Note that the physical meaning of the first three efficiency terms are similar to those of the Z01 scheme. However, the parameterizations of these terms differ from the Z01 scheme. The term describing turbulent impaction efficiency \( (E_{IT}) \) is absent in the Z01 scheme.

Parameterization of deposition efficiencies (i.e., \( E_B, E_{IN}, E_{IM} \), and \( E_{IT} \)) are given below according to the PZ10 scheme:

Particle collection efficiency by Brownian diffusion \( (E_B) \):

\[
E_B = C_B S c^{-2/3} R e_h^{-1/2}. \quad (28)
\]

In Eq. (28), \( C_B \) is the LUC dependent coefficient, \( R e_h \) is the Reynolds number of the horizontal air flow calculated at top of the canopy height \( h \) as \( R e_h = \frac{U_h L}{\nu} \). Here, \( L \) is the LUC dependent characteristic length of the canopy obstacle elements.

Particle collection efficiency by interception \( (E_{IN}) \):

\[
E_{IN} = C_B \frac{d_p}{L} \ (\text{for needle-like obstacle}), \quad (29)
\]

\[
E_{IN} = C_B \frac{d_p}{L} \left[ 2 + \ln \frac{d_p}{4d_L} \right] \ (\text{for leaf of plane obstacle}). \quad (30)
\]

In Eqs. 29-30, \( C_B \) is the LUC dependent coefficient.

Particle collection efficiency by impaction \( (E_{IM}) \):

\[
E_{IM} = C_{IM} \left( \frac{S t_h}{S t_h + \beta_{IM}} \right) \ . \quad (31)
\]

In Eq. (31), \( S t_h \) is the Stokes number on top of the canopy, which is calculated as \( S t_h = \frac{r_p U_h}{L \tau_p} \). \( r_p \) is the particle relaxation time calculated as \( r_p = V_p / g \cdot C_{IM} \) and \( \beta_{IM} \) are LUC dependent coefficients.

Particle collection efficiency by turbulent impaction \( (E_{IT}) \) is parameterized as:

\[
E_{IT} = 2.5 \times 10^{-3} C_{IT} \tau_p^2 \quad \text{if } \tau_p > 20, \quad (32)
\]
\[ E_{IR} = E_{IT} \quad \text{if } \tau_{ph}^+ \geq 20, \quad (33) \]

In Eqs. (32-33), the dimensionless particle relaxation time, \( \tau_{ph}^+ = \frac{\tau_p u^*_h}{v} \).

220 The term \( \eta \) in Eq. (19) is taken as:

\[ \eta = \frac{\sqrt{a_z^2} + Q}{4} \quad (34) \]

For non-vegetative surfaces, such as bare soil, natural water and ice/snow, a modified form of Eq. (17) is used in the form of Eq. (35), which is expressed as:

\[ V_d = V_{drift} + \frac{1}{\pi a^{+1/12} R g h v} \quad (35) \]

2.3. The KS12 scheme

Kouznetsov and Sofiev (2012) developed a dry particle deposition parameterization by extending the conventional resistance-based analogy using the exact solution of the steady-state equation for aerosol flux. According to the KS12 scheme, for rough surfaces, dry deposition velocity \( (V_d) \) is computed as:

\[ V_d = V_{diff} + V_{int} + V_{imp} + V_g, \quad (36) \]

where \( V_{diff}, V_{int}, V_{imp} \), and \( V_g \) are the velocities for the depositing particles due to Brownian diffusion, interception, impaction, and gravitational settling, respectively. The parameterizations for these terms are provided below.

\[ V_{diff} \] was parameterized as:

\[ V_{diff} = 2u^* R e_*^{-1/2} S c^{-2/3}, \quad (37) \]

where \( R e_* \) is the canopy Reynolds number given by

\[ R e_* = \frac{u^* a}{v}, \quad (38) \]

where \( a \) is the length scale for different LUCs.

\[ V_{int} \] is parameterized as:

\[ V_{int} = u^* R e_*^{1/2} \left( \frac{\xi}{a} \right)^2, \quad (39) \]

\[ V_{imp} \] is parameterized as:

\[ V_{imp} = \frac{2u^2}{U_{top}} \eta_{imp} \left( St - \frac{u^*}{U_{top}} R e_*^{-1/2} \right), \quad (40) \]

where \( U_{top} \) is the mean horizontal wind speed on top of the canopy, \( \eta_{imp} \) is the particle collection efficiency due to impaction, and \( St \) is the Stokes number. Kouznetsov and Sofiev (2012) used Eq. (41) to parameterize \( \frac{u^*}{U_{top}} \) as:

\[ \frac{u^*}{U_{top}} = \min \left[ (C_e + C_R L A I / 2)^2 \left( \frac{u^*}{U_{top}} \right)_{max} \right] \quad (41) \]

where \( C_e = 0.003, C_R = 0.3, \) and \( \left( \frac{u^*}{U_{top}} \right)_{max} = 0.3 \) are constants.

245 The Stokes number \( St \) is expressed as:

\[ St = \frac{\tau_p u^*_h}{a}, \quad (42) \]

where \( \tau_p \) is the particle relaxation time calculated as \( \tau_p = V_g / g \).

The expression for \( \eta_{imp} \) is given as:

\[ \eta_{imp} = \exp \left( \frac{-0.1}{St_e - 0.15} - \frac{1}{\sqrt{St_e - 0.15}} \right) \quad \text{if } St_e > 0.15, \quad (43) \]

\[ \eta_{imp} = 0 \quad \text{if } St_e \leq 0.15, \quad (44) \]
where $St_e$ is the effective Stokes number calculated as:

$$St_e = St - Re_e^{-\frac{1}{2}},$$  \hspace{1cm} (45)

where $Re_e$ is the critical Reynolds number calculated as:

$$Re_e = \left(\frac{u_{top}}{u_*}\right)^2 Re_c.$$  \hspace{1cm} (46)

255 The term $V_g$ in Eq. (36) is parameterized using Eq. (4).

Note that in the ZH14 scheme, the parameterization of $V_d$ over smooth surfaces requires solving the universal velocity profiles (either numerically or analytically) described by Kouznetsov and Sofiev (2012). We exclude the details of the solution procedure in this paper. We used the analytical solutions of the velocity profile obtained from the author of the ZH12 scheme through personal communication.

260 2.4. The ZH14 scheme

Zhang and He (2014) developed an empirical resistance-based parameterization for dry particle deposition by modifying the Z01 scheme. The overall structure of the ZH14 scheme is similar to that of the Z01 scheme (i.e., $V_d$ is calculated using Eq. (3)). In the ZH14 scheme, the parameterizations of $R_d$ and $R_g$ are similar to those of the Z01 scheme. However, in ZH14 scheme, parameterizations for the surface resistance term $R_s$ were modified for three bulk particle sizes (i.e., PM$_{2.5}$, PM$_{2.5-10}$, and PM$_{10+}$).

265 Recalling, $R_e = 1 / V_d$, the parameterizations of $V_{ds}$ are given below.

For particle sizes less than or equal to 2.5 µm (PM$_{2.5}$), $V_{ds}$ is expressed as:

$$V_{ds}(PM_{2.5}) = a_1 u_*,$$  \hspace{1cm} (47)

where $a_1$ is an empirical constant derived by regression analysis. Values of $a_1$ are given by Zhang and He (2014) for five groups of 26 LUCs.

270 For particle sizes between 2.5 and 10 µm (PM$_{2.5-10}$), $V_{ds}$ is expressed as:

$$V_{ds}(PM_{2.5-10}) = (b_1 u_* + b_2 u_*^2 + b_3 u_*^3) e^{k1 (\frac{LAI}{LAI_{max}} - 1)},$$  \hspace{1cm} (48)

where $b_1$, $b_2$, and $b_3$ are LUC dependent constants, $LAI_{max}$ is the maximum leaf area index for a given LUC, and $k1$ is a constant, which is a function of $u_*$, and expressed as:

$$k1 = c_1 u_* + c_2 u_*^2 + c_3 u_*^3,$$  \hspace{1cm} (49)

275 where $c_1$, $c_2$, and $c_3$ are the LUC dependent constants.

For particle sizes larger than 10 µm (PM$_{10+}$), $V_{ds}$ is expressed as:

$$V_{ds(10+)} = (d_1 u_* + d_2 u_*^2 + d_3 u_*^3) e^{k2 (\frac{LAI}{LAI_{max}} - 1)},$$  \hspace{1cm} (50)

where $d_1$, $d_2$, and $d_3$ are the LUC dependent constants, and $LAI_{max}$ is the maximum leaf area index for a given LUC. The parameter $k2$ is a constant, which is a function of $u_*$, and is expressed as:

$$k2 = f_1 u_* + f_2 u_*^2 + f_3 u_*^3,$$  \hspace{1cm} (51)

280 where $f_1$, $f_2$, and $f_3$ are the LUC dependent constants.

2.5. The ZS14 scheme

Zhang and Sao (2014) used an analytical solution of the steady-state flux equation to derive an expression to compute dry deposition velocity $V_d$ as:

$$V_d = \left( R_g + \frac{R_e - R_g}{\exp\left(\frac{LAI}{LAI_{max}}\right)} \right)^{-1},$$  \hspace{1cm} (52)
For neutral atmospheric stability conditions, the parameterizations of \( R_a \) for rough and smooth surfaces are given in Eqs. (53), and (54), respectively:

\[
R_a = \frac{Sc \ln \left( \frac{z}{h_c - d} \right)}{\kappa u_r}, \quad \text{(rough surfaces)}
\]

\[
R_a = \frac{\beta Sc \ln \left( \frac{z}{c_0} \right)}{\kappa u_r}, \quad \text{(smooth surfaces)}
\]

where \( B_1 \) is an empirical constant (0.45), and \( Sc_T \) is the turbulent Schmidt number expressed as:

\[
Sc_T = \left( 1 + \frac{\alpha^2 u^2}{u_*^2} \right),
\]

where \( \alpha \) is a dimensionless coefficient taken as 1.

The gravitational resistance term \( R_g \) is calculated as \( R_g = 1/V_g \). The parameterization of the surface resistance term \( R_s \) is given by Zhang and Sao (2014) as follows:

\[
R_s = \left\{ R_{Vdm} \left[ \frac{E}{C_d} \frac{\tau_c}{\tau} + \left( 1 + \frac{\tau_c}{\tau} \right) Sc^{-1} + 10^{-\frac{1}{3}} \right] + V_{g,w} \right\}^{-1},
\]

where \( R = \exp(-b \sqrt{St}) \) with \( b \) is an empirical constant, \( E \) is the total collection efficiency, \( C_d \) is the drag partition coefficient, \( Sc \) is the Schmidt number, \( T_{p,\delta} \) is the dimensionless particle relaxation time near the surface, and \( V_{g,w} \) is the gravitational settling velocity of particle after humidity correction. \( \tau_c \) is the ratio of the drag on the roof of the roughness element \( (\tau_c) \) to the total shear stress \( (\tau) \) and is calculated as:

\[
\frac{\tau_c}{\tau} = \frac{\beta \lambda_e}{1 + \beta \lambda_e}.
\]

where \( \beta \) is the ratio of the pressure-drag coefficient to friction-drag coefficient, and \( \lambda_e \) is the effective frontal area index. The parameter \( \lambda_e \) is a function of frontal area index or roughness density \( (\lambda) \), and plane area index \( (\eta) \). The expression of \( \lambda_e \) is

\[
\lambda_e = \frac{\lambda}{(1-\eta)^c_2} \exp \left( -c_1 \lambda \right).
\]

where \( c_1 = 6 \) and \( c_2 = 0.1 \).

Eq. (57) is used to compute \( T_{p,\delta} \) as:

\[
T_{p,\delta} = \frac{T_{p,\delta} u_*^2}{V}.
\]

where \( T_{p,\delta} \) is the particle relaxation time near the surface \( (T_{p,\delta} = V_g/g) \).

\( V_{dm} \) is calculated using two separate expressions for rough and smooth surfaces, as expressed in Eqs. (60) and (61), respectively:

\[
V_{dm} = \frac{u_*}{u_a h_c} \quad \text{(for rough surfaces)},
\]

\[
V_{dm} = B_2 u_* \quad \text{(for smooth surfaces)},
\]

where \( u_a \) is the horizontal air speed and \( h_c \) is the height of the roughness element.

In Eq. (56), the total collection efficiency \( (E) \) comprised of collection efficiencies by Brownian diffusion \( (E_B) \), impaction \( (E_{IN}) \), and interception \( (E_{IH}) \). The parameterizations for each of these three terms are given below:

\[
E_B = C_B Sc^{-2/3} Re^{n_B} \!
\]

where \( C_B \) and \( n_B \) are empirical parameters function of flow regimes, and are given by Zhang and Sao (2014).

\[
E_{IH} = \left( \frac{St}{5.6 + St} \right)^2,
\]

where \( St \) is the Stokes number and is expressed as \( St = \tau_c u_*/d_c \). Here, \( d_c \) is the diameter of the surface collection element. Values of \( d_c \) are given by Zhang and Sao (2014) for various surfaces.
where $A_{in}$ is a surface dependent micro-roughness characteristic element, and $d_{p,w}$ is the wet diameter of the particle.

3. Methods

3.1. An evaluation of the dry deposition parameterizations

To assess the accuracy of the five parameterizations, the modeled dry deposition velocities were compared with field measurements from both rough and smooth surfaces. The measurement studies conducted on various natural surfaces were collected from the literature. More specifically, the studies cited in the review article on particle flux measurements by Pryor et al. (2008) were collected to acquire the meta-data on particle deposition. The availability of the measured and/or reported parameters (e.g., particle size and density, air temperature, relative humidity, horizontal wind speed, friction velocity, atmospheric stability parameter, canopy height, roughness height, zero-plane displacement height, and leaf area index) from these measurement studies was thoroughly investigated and compiled. It was found that the many (ca. 50%) of the studies cited by Pryor et al. (2008) did not report most of the aforementioned parameters necessary to run the parameterizations to perform a valid comparison between the model output and measurements. To reduce uncertainty, those studies were excluded from the parametrization accuracy evaluation. In addition, a literature search was performed in Web of Science® to find measurement studies published after 2008, and those studies were thoroughly assessed to determine the availability of required input parameters to run the dry deposition models. Finally, 29 measurement studies covering five land use categories (LUCs) were selected to evaluate the accuracy of the five parameterizations. The five LUCs include grass, deciduous, and coniferous forests (rough surfaces), and natural water and ice/snow (smooth surfaces). Table 1 summarizes information related to sampling location, latitude, longitude, elevation above mean sea level (AMSL), sampling periods, and particle sizes reported in the measurement studies. The global spatial distribution of these measurement studies is shown in Figure 1 according to the five LUCs.

Figure 1. Global distribution of dry particle deposition measurement locations (listed in Table 1) used to evaluate the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations. Note that for multiple measurement campaigns conducted in one location, only one data point is shown. Two wind tunnel studies on water surfaces are not shown.

Measurements conducted over grass by Wesely et al. (1977), Neumann and den Hartog (1985), Allen et al. (1991), Nemitz et al. (2002), and Vong et al. (2004) were used to evaluate the performance of the five parameterizations. For coniferous forest,
modeled deposition velocities were compared with measurements from Lamaud et al. (1994), Wyzers and Duyzer (1996), Gallagher et al. (1997), Ruijgrok et al. (1997), Buzorius et al. (2000), Rannik et al. (2000), Gaman et al. (2004), Pryor et al. (2007), and Grönholm et al. (2009). Experiments conducted over deciduous forest are limited, and only three studies (Wesely et al., 1983; Pryor, 2006; Matsuda et al., 2010) were used in the present paper.

To evaluate the performance of the parameterizations over water surfaces, studies by Möller and Schumann (1970), Sehmel et al. (1974), Zufall et al. (1998) and Caffrey et al. (1998) were used. We note that the studies by Moller and Schumann, and Sehmel et al. were conducted in the wind tunnels, and thus the observed deposition may not necessarily reflect deposition under natural conditions. Particle deposition measurements on ice/snow pack were collected from eight studies: Ibrahim (1983); Duan et al. (1987); Nilsson and Rannik (2001); Gronlund et al. (2002); Contini et al. (2010); Held et al. (2011a); Held et al. (2011b); and Donateo and Contini (2014). The parametrizations were fed using reported values of particle properties (diameter and density), meteorological conditions (stability parameter, temperature, wind speed, etc.), and surface properties (canopy height, roughness length, leaf area index, etc.). However, reasonable values of the missing parameters were assumed when needed.

In the present study, the accuracy of the dry deposition parameterizations was evaluated using the normalized mean bias factor (BNMBF). The BNMBF provides a statistically robust and unbiased symmetric measure of the factor by which the modeled dry deposition velocities differ from the measured ones, and the sense of that factor (i.e., the positive and negative values imply the overprediction and underprediction by models, respectively). The interpretation of the BNMBF is simple, and it avoids any inflation that may be caused by low values of measured quantities (Yu et al., 2006).

To quantify the disagreement between the modeled and observed quantities, the normalized mean bias factors were calculated for the pairs of modeled \( V_{d(\text{modeled})i} \) and measured dry deposition velocities \( V_{d(\text{measured})i} \), respectively. In this study, the expressions for computing the BNMBF used in two different forms, which are:

For the \( V_{d(\text{modeled})i} > V_{d(\text{measured})i} \) case (i.e., overestimation):

\[
BNMBF = \frac{\sum V_{d(\text{modeled})i} - \sum V_{d(\text{measured})i}}{\sum V_{d(\text{measured})i}} = \frac{\sum \left( V_{d(\text{measured})i} - \frac{V_{d(\text{modeled})i}}{V_{d(\text{measured})i}} \right)}{\sum V_{d(\text{measured})i}}
\]

(65)

For the \( V_{d(\text{modeled})i} < V_{d(\text{measured})i} \) case (i.e., underestimation):

\[
BNMBF = \frac{\sum V_{d(\text{measured})i} - \sum V_{d(\text{modeled})i}}{\sum V_{d(\text{modeled})i}} = \frac{\sum \left( V_{d(\text{measured})i} - \frac{V_{d(\text{modeled})i}}{V_{d(\text{measured})i}} \right)}{\sum V_{d(\text{modeled})i}}
\]

(66)

The step-wise derivation of the Eqs. (65-66) and their application on training air quality datasets are illustrated by Yu et al. (2006).

### 3.2. Uncertainty analysis of the dry deposition parameterizations

To quantify the influence of imprecision in the model input parameter values on the modeled velocities, a classical Monte Carlo uncertainty analysis was applied. The Monte Carlo techniques have been widely used to evaluate the propagated uncertainty in the modeled outputs in many geophysical models (e.g., Alcamo and Bartnickj, 1987; Derwent and Hov, 1988; Chen et al., 1997;
Tatang et al., 1997; Hanna et al., 1998, 2001; Bergin et al., 1999; Bergin and Milford, 2000; Beekman et al., 2003; Mallet and Sportisse, 2006). Monte Carlo uncertainty evaluation techniques are relatively straightforward and flexible means for incorporating probabilistic values in the modeled dry deposition velocities. Indeed, the techniques are less reliant on assumptions about distributions of the input parameters (Hanna et al., 2002).

In this study, we define uncertainty in the parameterizations as the inability to confidently specify single-valued quantities because of the imprecision in the model input parameters. A classical Monte Carlo uncertainty method was applied to assess the overall uncertainty of a dry particle deposition parameterization with regard to the uncertainties in the following input parameters: RH, h, zo, d, LAI, U, u*, and Lo. The uncertainty estimates for those input parameters were obtained from the literature and are presented in Table 2. Using the uncertainty ranges for each of these parameters, uniform probability distribution functions were assigned since information on their actual distributions are lacking.

The Monte Carlo simulations were performed using R statistical software (version 3.2.4). Each simulation was run by randomly drawing 100 samples from the assigned uniform probability density function (PDF). The simulations were repeated 10,000 times. Frequency distributions or the PDFs of the modeled dry deposition velocity are the basic results of the Monte Carlo simulations. These PDFs were approximated assuming normal distributions, and then the 5th, 50th, and 95th percentile dry deposition velocities were computed. We use the range of the central 90% (the difference between 95th and 5th percentiles) of the PDFs as a convenient measure of uncertainty in the modeled deposition velocity. These steps were repeated for all five parameterizations using seven different particle sizes: 0.005, 0.05, 0.5, 1.0, 1.5, 2.0, and 2.5 µm on the five selected five LUCs (i.e., grass, deciduous and coniferous forests, water, and ice/snow). These particle sizes were selected to represent four distinct particle modes: nucleation (<0.01 µm), Aitken (0.01-0.1 µm), accumulation (0.1-1.0 µm), and coarse (>1.0 µm), respectively.

3.3. Sensitivity analysis

In this study, the Sobol’ sensitivity method (Sobol’ 1990) was applied to identify the most influential input parameter or the set of parameters of a dry deposition parameterization, and to characterize the relative contribution of the parameters to the overall variability in the modeled $V_d$. As opposed to the local sensitivity analysis (e.g., OAT approach), the Sobol’ method is a global sensitivity approach, in which a set of input parameters of a model can be varied simultaneously over their entire parameter value space to identify their relative contributions to the overall model output variance. The Sobol’ method has been applied in environmental modeling applications (Tang et al., 2007; Pappenberger et al., 2008; van Werkhoven et al., 2008; Yang, 2011), but has not yet been applied in dry particle deposition modeling research. Given that in most of the dry deposition parameterizations, model inputs can span a wide range within their physical realms, the application of a global sensitivity analysis used in this study should be viewed as a critical step toward the understanding of different sub-physical processes of particle deposition.

In the Sobol’ method, the variance contributions to the total output variance of individual parameters and parameter interactions can be determined. These contributions are characterized by the ratio of the partial variance ($V_i'$) to the total variance ($V$) as expressed in Eq. 67. This ratio is commonly termed as Sobol’ first order index ($S_i$) (Saltelli et al., 2010; Nossent et al., 2011). The first order indices represent the fractions of the unconditional model output variance. In this study, Sobol’ first order sensitivity indices were calculated as:

$$S_i = \frac{V_i}{V} = \frac{V_x(E_{x_{-i}}(V_d|X_i))}{V(V_d)} \tag{67}$$

where $X_i$ is the $i$-th input parameter and $X_{-i}$ denotes the matrix of all input parameters but $X_i$. The meaning of the inner expectation operator is that the mean of $V_d$ is taken over all possible values of $X_{-i}$ while keeping $X_i$ fixed. The outer variance is taken over all possible values of $X_i$. The variance $V(V_d)$ in the denominator is the total (unconditional) variance.
The numerator in Eq. (67) can be interpreted as follows: \( V_{X_i}(E_{X_i}(V_d|X_i)) \) is the expected reduction in variance that would be obtained if \( X_i \) could be fixed. In regard to the variability of the model input parameters in dry deposition schemes, \( S_i \) provides a means to quantify the effect of parameter \( X_i \) by itself. A higher order \( (S_{ij}) \) or total order \( (S_{ij}) \) can be computed when the total effect of a parameter, inclusive of all its interaction with other model input parameters, are of interest. In this paper, we confine the sensitivity analysis to Sobol’ first order indices only.

For each of the five parameterizations evaluated here, four to nine input parameters were selected for determining the first order Sobol’ sensitivity indices. An exception to applying the Sobol’ method was made for the KS12 parameterization while evaluating the parameter sensitivity for smooth surfaces. Due to the complex nature of KS12 smooth surface parameterization, it was not computationally feasible to apply the Sobol’ method. Instead, the OAT approach was applied for water and ice/snow surfaces. Note that the total number of input parameters that go into each model varies between parameterizations, and LUC types. For each parameterization, five particle sizes \((d_p = 0.001, 0.01, 0.1, 1.0, \text{ and } 10 \mu m)\) were assessed for Sobol’ analysis. The sensitivity of each parameterization was tested for the following three sets of input parameters for five LUCs: (i) particle properties, (ii) aerodynamic parameters, and (iii) surface characteristics of particle deposition. First, the sensitivity of particle deposition to particle properties (aerodynamic diameter and density) was tested. Sensitivity indices were calculated for the particle size range of 0.001 \( \mu m \) to 10 \( \mu m \). Second, the sensitivity of the schemes was tested for aerodynamic parameters (friction velocity, wind speed, and stability condition) for different particle sizes one-at-a-time. Third, the sensitivity of the schemes to surface characteristics was tested. Surface characteristics include \( h, z_0, d, \text{ and } LAI \). The sensitivity ranges for the parameter values used for Sobol’ analysis are reported in Table 3.

The Sobol 2007 package in R statistical software package (version 3.2.4) was used to perform the Sobol’ sensitivity analysis. In the Sobol’ method, the Monte Carlo simulations were performed by drawing samples from the assigned parameter value distribution. In this study, all the selected parameters were approximated using uniform PDFs. To assert uncertainty in the simulations, bootstrapping (Efron and Tibshirani, 1993) with re-sampling was used to achieve 95% confidence intervals on the Sobol’ first order indices. For a fixed particle size, the simulations were run 100,000 times and samples were bootstrapped 1,000 times. To identify the most important parameters in each of the five dry deposition models with respect to particle size and LUC, a parameter ranking (e.g., from most to least influential) was conducted.

The results section is organized in the following manner. First, the accuracy of five dry particle deposition parameterizations (i.e., Z01, PZ10, KS12, ZH14, and ZS14) are compared with measured dry deposition velocities obtained from five LUCs. Second, the uncertainties in modeled dry deposition velocities due to the imprecision in the model input parameter values quantified using Monte Carlo simulation techniques are presented. Third, the sensitivity analysis results for modeled dry deposition velocities by the five parameterizations are presented.

4. Results

4.1. Evaluation of the dry particle deposition parameterizations

Field measurements conducted on five LUCs: grass, coniferous forest, deciduous forest, water surfaces, and ice/snow were used to evaluate the agreement between measured and modeled dry deposition velocity \( (V_d) \). The parameterizations were run using reported values of the meteorological (e.g., \( U, u’, T, RH, \text{ and } L_0 \)) and canopy (e.g., \( h, z_0, d, \text{ and } LAI \)) parameters, and particle properties (e.g., \( d_p \) and \( \rho \)) from the measurement studies. Reasonable parameter values were assumed for any missing or unreported parameters. Normalized mean bias factors (BMRF) were used as an indicator of the agreement between measured and modeled \( V_d \). BMRF is a signed quantity—its magnitude indicates the factor by which the modeled and observed \( V_d \) differ from each other, and its sign provides an indicator as to whether the modeled \( V_d \) is greater or less than the measured \( V_d \). It is to be noted that uncertainties
in the measured dry deposition velocities were not considered while evaluating the performance of the five parameterizations in terms of accuracy.

4.1.1. Evaluation of dry particle deposition to grass

Five measurement studies conducted on grass (Wesely et al., 1977; Allen et al., 1991; Neumann and den Hartog, 1985; Nemitz et al., 2002; and Vong et al., 2004) were used to evaluate the accuracy of the parameterizations. In those studies, reported values of meteorological parameters, canopy properties, and particle size vary widely. For example, the wind speed (U) varies from 0.67 to 6.20 m/s, particle size (dp) varies from 0.05 to 2.28 µm, and LAI varies from 2 to 4 m²/m². The parameterizations were fed with reported values from each of the studies to reduce any uncertainty in the accuracy comparison, however, for any missing parameter value(s), the assumed input parameter values typically fell within the aforementioned ranges.

Table 4 summarizes the BNMBF for modeled Vd computed against five measurement studies on grass. The BNMBF is interpreted as follows: for example, if BNMBF is positive, the parameterization overestimates the measured Vd by a factor of BNMBF+1. If BNMBF is negative, the model underestimates the measured Vd by a factor of 1- BNMBF. For the case using the observations from Allen et al. (1991), the BNMBF values of -17.61, -18.12, -0.55, and -5.13 indicate that the Z01, KS12, ZH14, and ZS14 parameterizations underestimated the measured Vd by factors of 18.61, 19.12, 1.55, and 6.13, respectively, whereas, the BNMBF value of +15.96 indicates that the PZ10 parameterization overestimated the observations by a factor of 16.96.

These results provide means for a relative comparison of the parameterizations’ accuracy. For instance, the BNMBF values corresponding to the Allen et al. study suggest that the ZH14 parameterization is the most accurate and the KS12 parameterization is least accurate. Similar comparison between the modeled and observed Vd can be made using the BNMBF values for the remaining four studies in Table 4. Nonetheless, it is evident that none of the parameterizations performed best in terms of accuracy for all of the five studies since the BNMBF values show high variability both in terms of the magnitude and direction of the bias (i.e., positive or negative) when assessed against all the five studies listed in Table 4.

The characteristics of a parameterization (e.g., Z01) to simultaneously over-predict (i.e., the positive BNMBF for Neumann and den Hartog, and Nemitz et al.) and under-predict (i.e., the negative BNMBF for Allen et al. 1991, Wesely et al. 1977, and Vong et al. 2004) the measurements could be misleading, resulting in erroneous judgement of the performance of the parameterizations.

To address this limitation, an ensemble approach was taken, in which BNMBF was calculated for each of the parameterizations using all the observations reported in the five studies. The results from this ensemble analysis indicate that, except for the Z01 parameterization, the other four parameterizations underestimated the measured Vd by factors ranging from 1.54 to 10.37. In contrast, the Z01 parameterization overestimated the observation by a factor of 6.45 (Table 4). Overall, these results indicate that the ZH14 parameterization provided the best agreement between the measured and modeled Vd of the five parameterizations.

4.1.2. Evaluation of dry particle deposition to coniferous forest

Nine studies conducted on coniferous forest (Lamaud et al., 1994; Wyer and Duyzers, 1996; Gallagher et al., 1997; Ruijgrok et al., 1997; Rannik et al., 2000; Buzorious et al., 2000; Gaman et al., 2004; Pryor et al., 2007; and Grönholm et al., 2009) were used to evaluate the accuracy of the parameterizations. In these studies, the largest variations (ranges are given in the parentheses) were associated with wind speed (U (0.06-1.30 m/s), U (0.60-6.19 m/s), LAI (6-10 m²/m²), and dp (0.01-0.60 µm). For any missing parameter value(s), the assumed input parameter values typically fell within the aforementioned ranges.

Comparison of the computed BNMBF values for coniferous forest (Table 5) shows that the majority of the simulations performed using the five parameterizations underestimated the measured Vd. For example, the PZ10 parameterization
underestimated observed $V_d$ by factors ranged from 1.51 to 27.98 (B_{NMBF} values varied from -0.51 to -26.98) for eight of the nine studies on coniferous forest. Table 5 also illustrates that both the magnitude and sign of the B_{NMBF} values varied widely when the accuracy of the five parameterizations was evaluated against only one study (e.g., Pryor et al., 2007). Of the B_{NMBF} values associated with the Ramik et al. (2000) study, the Z01 and KS12 parameterizations overestimated the measured $V_d$ by factors of 4.16 and 1.51, respectively, whereas the PZ10, ZH14, and ZS14 parameterizations underestimated the measured $V_d$ by factors of 3.54, 2.13, and 19.75, respectively. The bias factors for the Z01 parameterization for the following studies: Laman et al. (1994), Gallagher et al. (1997), Buzorious et al. (2000), and Gaman et al. (2004), were +0.77, -1.74, +0.75, and -0.90, respectively. Comparing these values with the corresponding B_{NMBF} values of the other four parameterizations, it can be deduced that the Z01 parameterization is the most accurate against those observations reported in these four studies. However, the accuracy of the Z01 parameterization is not the best for the other five studies, as can be seen from Table 5.

An ensemble approach similar to the one described in the previous section was used to determine the most and the least accurate parameterizations. From this analysis, the bias factors for the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations are -2.35, -3.93, -1.75, -2.31, and -3.67, respectively, suggesting that the KS12 is the most accurate parameterization (i.e., under-predicted the observations by a factor of 2.75), and the PZ10 is the least accurate parameterization (i.e., under-predicted the observations by a factor of 4.93) for coniferous forest. It can be noted that the performance of the Z01 and ZH14 parameterizations are nearly identical, while the ZH14 is the second most accurate (i.e., under-predicted the observations by a factor of 3.31).

4.1.3. Evaluation of dry particle deposition to deciduous forest

A similar comparison between measured and modeled $V_d$ was performed using three studies (Wesely et al., 1983; Pryor, 2006; and Matsuda et al., 2012) for deciduous forest. In these studies, the largest variations (ranges are given in the parentheses) were associated with $u^*$ (0.12-1.13 m/s), $U$ (1.20-6.00 m/s), LAI (0.20-10 m^2/m^2), and $d_p$ (0.05-2.50 µm). For any missing parameter value(s), the assumed input parameter values typically fell within the aforementioned ranges.

Computed B_{NMBF} values for deciduous forest are presented in Table 6. For the Wesely et al. (1983) study, comparison of the B_{NMBF} values between the parameterizations show that the performance of the ZS14 parameterization was the most accurate (i.e., B_{NMBF} = -2.28; under-predicted the observations by a factor of 3.28). The B_{NMBF} values associated with the PZ10 parameterization showed strong variation between the studies (e.g., two orders of magnitude discrepancy between the Wesely et al. (1983) and Pryor (2006) or Matsuda et al. (2012) studies).

Evidently, none of the parameterizations performed consistently better for all the three studies. Overall, the results from the ensemble approach show that all the parameterizations overestimated the observations reported in three studies. Considering the B_{NMBF} values obtained by this approach, it is apparent that the ZH14 is the most accurate parameterization (i.e., B_{NMBF} = -3.75, underestimated the observed $V_d$ by a factor of 4.75), and the ZS14 is the least accurate of the five parameterizations (i.e., B_{NMBF} = -10.93, underestimated the observed $V_d$ by a factor of 11.93) for deciduous forest.

4.1.4. Evaluation of dry particle deposition to water surfaces

Only a limited number of measurement studies on size-segregated dry particle deposition over natural water surfaces are available in the literature. In this research, four studies (Möller and Schumann, 1970; Schmel et al., 1974; Zuffal et al., 1998; and Caffery et al., 1998) conducted over water surfaces were used to evaluate the parameterizations’ accuracy. From these studies, the reported values of the parameters that show the largest variations (ranges are given in the parentheses) are $u^*$ (0.11-0.40 m/s) and $d_p$ (0.03 to 48 µm).
Table 7 shows that the PZ10 parameterization performed best for two studies (i.e., Möller and Schumann, 1970; and Caffery et al., 1998), in which BA values were -1.65 and +0.35, respectively. Comparison of the BA values between the Z01 and ZH14 parameterizations reveal that the accuracy of the two parameterizations varied widely among the studies (e.g., BA ranged from -0.144 to +18.87 and -0.33 to +10.28, respectively). Nevertheless, none of the five parameterizations was able to reproduce the measured Vd satisfactorily for all the four studies. Comparison of the BA values obtained by the ensemble approach showed that the ZH14 parameterization is the most accurate, which underestimated the measured Vd by a factor of 1.25 (i.e., BA = -0.25), and the PZ10 is the least accurate parameterization (i.e., BA = -0.89).

4.1.5. Evaluation of dry particle deposition to snow and ice surfaces

Two studies over snow (Ibrahim, 1983; and Duan et al., 1987), and six studies over ice surfaces (Nilsson and Rannik, 2001; Gronlund et al., 2002; Contini et al., 2010; Held et al., 2011a; Held et al., 2011b; and Donateo and Contini, 2014) were used to evaluate the accuracy of the four parameterizations (Z01, PZ10, KS12, and ZH14) for smooth surfaces. The ZS14 parameterization was not included here because it does not allow prediction of deposition over ice/snow surfaces. The BA values for the parameterizations are presented in Table 8.

Of the four parameterizations, agreement between the modeled and measured Vd is not satisfactory for the PZ10 and KS12 parameterizations because they significantly underestimated the measured Vd (e.g., the bias factors from ensemble approach are -53.03 and -21.80, respectively). In contrast, the Z01 and ZH14 parameterizations predicted the measured Vd with reasonable accuracy (e.g., the bias factors from ensemble approach were +1.98 and +0.26, respectively). Table 8 also shows that the ZH14 parameterization performed best for six of the eight measurements in which the BA varied between -0.74 to 3.98. Overall, for the nine studies combined (i.e., ensemble measurements), the ZH14 parameterization is the most accurate (overestimated the measured Vd by a factor of 1.26), and the PZ10 is the least accurate parameterization (underestimated the measured Vd by a factor of 54.03).

To summarize, the results from the ensemble evaluation of the parameterizations is graphically shown in Figs. 2(A-B) for the five LUCs. The horizontal dotted-dashed line in the plots indicates 100% agreement between modeled and measured Vd, whereas any dispersion from this line either above (i.e., over-estimation) or below (i.e., under-estimation) indicates the degree of the model’s accuracy.

Figure 2. Ensemble averaged, normalized mean bias factors for the five parameterizations: a) three rough surfaces and water, b) Ice/snow.

4.2. Uncertainty analysis results from the Monte Carlo simulations

The overall uncertainty in the modeled Vd due to imprecision in the model inputs was assessed by performing a set of Monte Carlo simulations on the five dry particle deposition parameterizations. Uncertainties (in terms of imprecision) in the...
following model input parameters: RH, U, u*, L0, h, z0, d, and LAI were approximated using uniform distributions. Note that not all of the five parameterizations require an identical number of input parameters. For example, Monte Carlo simulations performed on rough surfaces (i.e., grass, coniferous, and deciduous forests) for the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations, imprecision in four (RH, L, u*, and z0), eight (RH, L, u*, U, z0, h, d, and LAI), four (RH, u*, U, and LAI), four (RH, L, u*, and z0), and two (RH, u*) input parameters, respectively, were assessed to evaluate the overall uncertainty in modeled dry deposition velocities.

The results from the Monte Carlo simulations are summarized in Table 9 and are presented and discussed in two steps. First, the uncertainty estimates that are shown in Table 9 for five parameterizations on five LUCs are used to elucidate the models’ precision, which is one of the indicators of overall performance of the parameterization (Sections 3.2.1-3.2.5). Second, the size-dependent uncertainty ranges (i.e., the difference between the 95th and 5th percentiles) was divided by the 50th percentile $V_d$, which can be treated as a normalized measure of uncertainty. This approach was taken to make reasonable comparison between different particle sizes for different parameterizations (Section 3.2.6). Note that the ZS14 parameterization does not treat different vegetative covers separately; therefore, inter-comparison of the Monte Carlo simulation results is confined to the first four parameterizations listed in Table 9.

4.2.1. Uncertainties in the modeled $V_d$ for grass

The uncertainties in simulated $V_d$ (i.e., differences between 95th and 5th percentiles of distribution) for the given range of $d_p$ (i.e., 0.005-2.5 µm) on grass varied widely (Table 9). In the Z01 parameterization, the estimated uncertainty for nucleation mode particles (0.0038 m s$^{-1}$ for $d_p = 0.005$ µm) was larger than that of coarse mode particles (0.0001 m s$^{-1}$ for $d_p > 1.0$ µm). Overall, in the Z01 parameterization, the trend was that as the particle size increased from 0.005 to 2.5 µm, uncertainties in modeled $V_d$ decreased considerably. In the PZ10 parameterization, the range of uncertainty for the simulated particle sizes is narrower as compared to those of the Z01 parameterization. Although not consistent, a decreasing trend in uncertainties can be seen for all the particle sizes in the PZ10 parameterization. Of the simulated particle sizes, the uncertainty for $d_p = 0.005$ µm is the largest (0.0016 m s$^{-1}$) in the KS12 parameterization. As particle size increased from 0.005 to 2.5 µm, significant decrease in uncertainties is observed. For $d_p = 0.05$ to 1.5 µm, the 5th and 95th percentile $V_d$ were nearly identical (Table 9), suggesting that the KS12 parameterization is the most precise of five parameterizations specifically for those particle sizes. From Table 9, it can be deduced that the uncertainties associated with the ZH14 parameterization, which is an improved and simplified version of the Z01 parameterization, were fairly constant (ca. 0.0003 m s$^{-1}$) for the seven particle sizes simulated here for grass.

4.2.2. Uncertainties in the modeled $V_d$ for coniferous forest

For nucleation mode particles (i.e., $d_p = 0.005$ µm), the largest uncertainty (0.0036 m s$^{-1}$, median $V_d = 0.0180$ m s$^{-1}$) was associated with the Z01 parameterization (Table 9). Overall, the uncertainties in the Z01 parameterization showed a decreasing trend as the particle size increased from 0.005 to 2.5 µm. We note that in the PZ10 parameterization, the relative magnitude of the uncertainties associated with 0.005, 1.0, 1.5, 2.0, and 2.5 µm particles were of the same order (i.e., varied between 0.0010 to 0.0031 m s$^{-1}$). In comparison, uncertainties in modeled $V_d$ for 0.05 and 0.5 µm particles were smaller by factors of ca. 10. In the KS12 parameterization, the largest uncertainty was found for the nucleation mode particles (i.e., 0.0027 m s$^{-1}$; median $V_d = 0.0299$ m s$^{-1}$), and the uncertainties in modeled $V_d$ decreased substantially as $d_p$ increased. The uncertainties in modeled $V_d$ in the ZH14 parameterization were constant (0.0002 m s$^{-1}$) for all seven particle sizes indicating the model’s ability to reproduce dry deposition velocities with high precision.

4.2.3. Uncertainties in the modeled $V_d$ for deciduous forest

A similar comparison of the uncertainties in modeled $V_d$ can be made for deciduous forest. It is seen from Table 9 that, for all the parameterizations except for ZH14, the largest uncertainties were associated with nucleation mode particles. That is, Z01
and KS12 parameterizations showed substantially greater uncertainties for \(dp = 0.005 \ \mu m\) (0.0030 and 0.0027 m s\(^{-1}\), respectively) as compared to the Aitken or coarse mode particles, for which the relative magnitude of the uncertainties were smaller by factors of ca. 13-30. In the KS12 parameterization, the identical values of the 5\(^{\text{th}}\) and 95\(^{\text{th}}\) percentile \(V_d\) resulted in uncertainty values of zero for each simulated particle size of 0.5 to 2.0 \(\mu m\), which indicates that it is the most precise of all four parameterizations. In addition, the uncertainties in the modeled \(V_d\) in the ZH14 parameterization were constant (0.0004 m s\(^{-1}\)) for all seven particle sizes.

4.2.4. **Uncertainties in the modeled \(V_d\) for water surfaces**

For water surfaces, the uncertainties in modeled \(V_d\) varied largely for the Z01 parameterization (Table 9). That is, the largest uncertainty (0.0021 m s\(^{-1}\)) was associated with \(dp = 0.005 \ \mu m\) (median \(V_d = 0.0099 \ \text{m s}^{-1}\)), and as \(dp\) increased to 2.5 \(\mu m\), the uncertainty decreased to 0.0001 m s\(^{-1}\) (for 2.5 \(\mu m\) particles, median \(V_d = 0.0009 \ \text{m s}^{-1}\)). Relatively narrower ranges in the uncertainties in modeled \(V_d\) for the PZ10 and KS12, and constant uncertainties in the ZH14 parameterizations with regard to changes in particle size suggest their higher precision as compared to the Z01 parameterization under similar model input parameter uncertainties. Overall, as compared to the simulated uncertainties in the modeled \(V_d\) by the Z01, PZ10, KS12, and ZH14 parameterizations, uncertainties in the ZS14 parameterization are larger for \(dp = 0.05\) to 2.5 \(\mu m\).

4.2.5. **Uncertainties in the modeled \(V_d\) for ice/snow surfaces**

Comparison between the simulated uncertainties in modeled \(V_d\) revealed that the uncertainties vary significantly for the Z01 and KS12 parameterizations as \(dp\) changes. For example, uncertainties estimated from Table 9 for these two parameterizations decreased from 0.0023 to 0.0003 m s\(^{-1}\) and 0.0027 to 0.0008 m s\(^{-1}\), respectively, as particle size increased from 0.005 to 2.5 \(\mu m\). Note that the median \(V_d\) by the PZ10 parameterization is an order of magnitude lower than that of other three parameterizations, which results in close to zero uncertainties for all seven particle sizes. Also revealed in Table 9, the uncertainties in the ZH14 parameterization are constant (0.0002 m s\(^{-1}\)) with regard to changes in the particle size.

4.2.6. **Normalized uncertainties in the modeled \(V_d\)**

An extended analysis of the results presented in the previous sections are summarized here. The normalized uncertainties presented in the Table 10 can be interpreted as follows: any value that is closer to zero indicates higher model precision (i.e., less uncertainty). As shown in Table 10, the normalized uncertainties for grass and \(dp = 0.005 \ \mu m\) associated with the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations are 0.20, 0.11, 0.09, 0.20, and 0.20, respectively. These results suggest that KS12 is the least uncertain (i.e., most precise) parameterization for nucleation mode particles, whereas, the Z01, ZH14, and ZS14 are the most uncertain (i.e., least precise) parameterizations. Similar comparisons can be made for other particle sizes, as well as between the different LUCs. For example, the uncertainties associated with \(dp = 0.05 \ \mu m\) is greater for the PZ10 parameterization for deciduous forest as compared to grass (0.20 > 0.13).

Comparison of the normalized uncertainties in modeled \(V_d\) over smooth surfaces (i.e., water and ice/snow) also reveals interesting findings. For example, for \(dp = 0.5 \ \mu m\), the normalized uncertainties over water surfaces for the Z01, PZ10, KS12, and ZH14 parameterizations are 0.20, 0.00, 0.50, and 0.17, respectively. These results suggest that the PZ10 parameterization is the least uncertain (i.e., most precise), whereas, the KS12 is the most uncertain (i.e., least precise) parameterization for accumulation mode particles. Over ice/snow surfaces, with \(dp = 0.005 \ \mu m\), both the Z01 and ZH14 parameterizations have large uncertainties (normalized uncertainties are 0.18 and 0.17). In contrast, PZ10 is the most precise parameterization with close to zero normalized uncertainty value.

The normalized uncertainties presented in Table 10 also reveal interesting findings about the relative magnitude of imprecision for a given particle size on various LUCs by one parameterization. For example, with \(dp = 0.005 \ \mu m\), the range in normalized uncertainties varies from 0.18-0.20 and 0.09-0.20 for all the five LUCs for the Z01 and KS12 parameterizations, respectively.
Figs. 3(A-E) show the relative comparison between uncertainties in modeled $V_d$ by five parameterization for seven particle sizes across five LUCs. For LUC grass, Fig. 3A shows that in the uncertainties in the $Z01$ and $ZH14$ parameterizations show nearly identical trends, which are relatively narrow. That is, the uncertainties for particle sizes from 0.005 to 2.5 µm varied from 12-22% and 11-20% in the $Z01$ and $ZH14$ parameterizations, respectively. In contrast, uncertainties in the $PZ10$ and $KS12$ parameterizations exhibit large dispersion (i.e., uncertainty ranges from ~0-13% in the $PZ10$, and ~0-14% in the $KS12$ parameterizations). The largest uncertainties in the simulated $V_d$ are associated with the $ZS14$ parameterization, in which the range of uncertainty varied from ~0-34% for the seven particle sizes. We note that the minimum $V_d$ produced by the $KS12$ parameterization is at $d_p = 0.5$ µm for grass, coniferous and deciduous forest, and ice/snow surfaces, which can be confirmed from the Fig. 3(A-C and E). In addition, Fig. 3(D-E) show that the position of this minimum $V_d$ in the $PZ10$ parameterization ranged from $d_p = 0.5-1.0$ µm for water and ice/snow surfaces.

![Graphs showing uncertainty comparison](image-url)

Figure 3. Comparison of the simulated uncertainties in the modeled dry deposition velocities as a function of particle size in five parameterizations for five LUCs.

Similar comparison can be made to evaluate the relative magnitude of the uncertainties in the modeled $V_d$ predicted by the parameterizations over other LUCs from Figs. 3(B-E). In Fig. 3B, uncertainties in modeled $V_d$ for coniferous forest ranged
from 11-20%, 8-15%, ~0-21%, and 13-17%, in the Z01, PZ10, KS12, and ZH14 parameterizations, respectively. In Fig. 3C, uncertainties in modeled $V_d$ for deciduous forest ranged from 10-21%, 20-29%, ~0-21%, and 15-18% in the Z01, PZ10, KS12, and ZH14 parameterizations, respectively. In Fig. 3D, uncertainties in modeled $V_d$ for water surfaces ranged from 11-21%, ~0-25%, 18%, 14-18%, and 18-36% in the Z01, PZ10, KS12, ZH14, and ZS14 parameterizations, respectively. In Fig. 3E, uncertainties in modeled $V_d$ for ice/snow surfaces ranged from 14-30%, ~0-8%, ~0-28%, and 13-17% in the Z01, PZ10, KS12, and ZH14 parameterizations, respectively.

4.3. Sensitivity analysis results: Sobol’ first order sensitivity index

For Sobol’ first order sensitivity analysis, five particle sizes (i.e., $d_p = 0.001, 0.01, 0.1, 1.0, \text{and} 10 \text{ µm}$) were selected. A sample size ($n$) of 100,000 was used for model evaluations for each of the five particle sizes. To assess the confidence intervals for the first order Sobol’ sensitivity index, bootstrapping resampling was used. In the bootstrapping method, the $n$ samples used for the sensitivity simulations were sampled 1,000 times with replacement. In the following sections, the results from the Sobol’ sensitivity analysis, and evolution of the parameter rankings are presented.

The Sobol’ sensitivity analysis performed here is used to achieve a ranking of the model input parameters. The ranking of the parameters from most to least sensitive of the five particle sizes for the five parameterizations is shown in Table 11. Tables S1-S5 show the first order Sobol’ indices of the various input parameters used in five dry particle deposition parameterizations for five LUCs. In these tables, particle size-dependent first order Sobol’ index ($S_i$) for different model input parameters are presented with 95% confidence intervals (CI) obtained by bootstrap sampling. For example, the results of the first order Sobol’ indices for the Z01 parameterization on five LUCs are presented in Table S1. It is important to note that the number of parameters tested for Sobol’ analysis varied between different LUCs, mainly because the number of parameters required for modeling $V_d$ for one LUC may be more or less as compared to another LUC.

As shown in Table S1, for the Z01 parameterization on grass, the importance of the most influential parameters on the modeled dry deposition velocities for five particles sizes can be compared using the corresponding $S_i$ values of the model input parameters (e.g., $i = RH, \rho, L_0, u_*, \text{etc.}$). For example, with $d_p = 0.001 \text{ µm}$, it can be clearly seen that the $u_*$ is by far the single most sensitive parameter with an $S_i$ value of 0.918, which indicates that 91.8% of the variation in the modeled $V_d$ can be attributed to variations in $u_*$. The other parameters that have significant effect on the modeled $V_d$ are $z_0$ and $L_0$. These two parameters have $S_i$ values of 0.044 and 0.009, respectively. As compared to the first order Sobol’ value of $u_*$, these values are significantly smaller; however, the lower limits of the corresponding 95% C.I. intervals for $z_0$ and $L_0$ are greater than zero, indicating that they have a significant effect on the modeled velocities. The $S_i$ values for the other two parameters, $RH$ and particle $\rho$, were approximately zero for $d_p = 0.001 \text{ µm}$ (Table S1), and indicate that these variables have no influence on the modeled $V_d$.

Comparison between the first order Sobol’ indices for different particle sizes for grass shows strong variations for certain input parameters, which reveals interesting findings about the relative importance (from the most to the least) of the model input parameters to the modeled dry $V_d$. For example, as seen from Table S1, as $d_p$ increases from 0.001 to 10 µm, $S_i$ values of $u_*$ decrease from 0.918 to 0.245, which indicates that deposition of coarse particles is not strongly influenced by variations in friction velocity.

From Table S1 it is also seen that parameters that influence particle properties (i.e., $RH$ and $\rho$) have higher $S_i$ values for the coarse particles as compared to the fine or accumulation mode particles. Similar comparisons between size-dependent behavior of parameter sensitivity for other rough surfaces (i.e., coniferous and deciduous forests) can be made using the $S_i$ values reported in Table S1.
The results of the first order Sobol’ indices for the Z01 parameterization on two smooth surfaces: water and ice/snow are also presented in Table S1. Over liquid water surfaces, variation in $u*$ values has the largest influence modeled $V_d$ for $d_p=0.001$ to 10.0 $\mu$m. As is seen from Table S1, the $S_i$ values of $u*$ can alone explain 98.3-99.5% of the variations in modeled $V_d$ for particle sizes up to fine mode (i.e., 0.001-0.01 $\mu$m). For coarse mode particles (e.g., $d_p = 10 \mu$m), $u*$ is also the most influential parameter, contributing ca. 56% of the total variation in modeled $V_d$, while relative humidity is the second most influential/sensitive parameter with an $S_i$ value of 0.393. The influence of $u*$ also tends to dominate the modeled $V_d$ over ice/snow surfaces. This theory can be confirmed by comparing the size-dependent $S_i$ values of $u*$ shown in Table S1, which suggest that $u*$ is the single most sensitive parameter ($S_i = 0.978$) for $d_p = 1.0 \mu$m. As the particle size increased to 10 $\mu$m, RH and $u*$ can explain 92.7% of the total variation in modeled $V_d$ in the Z01 parameterization.

The results of the first order Sobol’ indices for the PZ10 parameterization on five LUCs are presented in Table S2. The size-dependent $S_i$ values on coniferous forest can be compared here to elucidate the contribution of different input parameters on the modeled $V_d$. It can be noted that, on rough surfaces, the PZ10 parameterization was tested for the most number of input parameters (i.e., nine) among the five parameterizations. Some canopy properties such as $h$, $d$, LAI, and meteorological properties such as $U$ were tested for their influence on modeled $V_d$ in addition to those parameters that were tested for the rough surfaces in the Z01 parameterization. As seen from Table S2, for coniferous forest, for $d_p = 0.001 \mu$m, $u*$ and $L_d$ are the two most influential parameters ($S_i$ values of 0.492 and 0.462, respectively). Although LAI is not the most influential parameter for the range of $d_p$ tested here, its influence on the overall variability in the modeled $V_d$ increase from 0.5 to 31.3% as particle size increases from 0.001 to 0.1 $\mu$m. Similarly, wind speed tends to show an increasing influence as $d_p$ increases from 0.001 to 1.0 $\mu$m (overall contribution of $U$ in the variability in $V_d$ shows an increase from 0.1% to 27.7%). For coarse particles (i.e., $d_p=10 \mu$m), $u*$ and $L_d$ are the two most influential parameters with $S_i$ values of 0.372 and 0.350, respectively. Together with RH, the three parameters can explain 92% of the variation in the modeled $V_d$. Results from the first order Sobol’ indices for the other LUCs for the PZ10 parameterization presented in Table S2 can be explained in a manner similar to that used to explain the contribution of the most sensitive parameters to the modeled dry deposition velocities. For the water surface, $u*$ is the most influential parameter for $d_p = 0.001 \mu$m as 99.4% of the total variance on the modeled $V_d$ is attributed to its variability. Indeed, for particle sizes up to 0.1 $\mu$m, the $u*$ itself is most sensitive parameter. As seen from Table S1, RH becomes the most influential model parameter for $d_p = 1.0$ and 10.0 $\mu$m, which alone contribute to 69.5% and 95.6% of the total variabilities in the modeled $V_d$, respectively.

Table S3 shows the first order Sobol’ indices for the KS12 parameterization on five LUCs. For brevity, the results of the first order sensitivity indices for deciduous forest are discussed herein. It is seen that $u*$ is the single most influential parameter for $d_p = 0.001$ to 0.1 $\mu$m (e.g., total contribution on the modeled $V_d$ attributable to $u*$ ranges from 94.4 to 96.7%). For $d_p = 1.0$ and 10 $\mu$m, RH is the most influential parameter with $S_i$ values of 0.629 and 0.934, respectively.

Table S4 shows the first order Sobol’ indices for the ZH14 parameterization on five LUCs. The results show a strong influence of $u*$ on the modeled $V_d$. As shown in Table S4, the $S_i$ values alone can explain nearly 100% of the variation in the modeled $V_d$ for $d_p = 0.001$ to 1.0 $\mu$m. For large particles (e.g., $d_p = 10 \mu$m), RH is the most influential parameter, however, the contributions of other parameters as listed in Table S4 vary with regard to changes in LUCs.

5. Discussion

The accuracy of the parameterizations should be interpreted within the context of the field measurements used in this study assuming that they were accurate. In addition, the inter-comparison of the parameterizations’ accuracy is subject to uncertainties with regard to the assumed values of missing meteorological parameters, particle properties, or surface features.
Evidently, the normalized mean bias factors obtained using the ensemble approach is a useful measure to inter-compare the parameterizations’ performance against a sub-set of field measurements for a given LUC. Extending the comparison of the normalized mean bias factors across the five LUCs for the five parameterizations investigated in this study provides a relative assessment of their accuracy. However, the ZH14 parameterization is most accurate for all parameterizations except coniferous forest, where it is a close second to the KS12 parameterization.

For rough surfaces, our results suggest that ZH14 is the most accurate parameterization for grass and deciduous forest, and it is the second most accurate parameterization for coniferous forest. In contrast, KS12, PZ10, and ZS14 are the least accurate parameterizations for grass, coniferous, and deciduous forests, respectively. It is interesting that in most cases the models underpredicted the measured dry deposition velocities (negative bias factors in Tables 4-8). Indeed, for grass, except for the Z01 parameterization, the other four parameterizations under-predicted the measured $V_d$ by factors of 1.54 to 10.37 ($B_{NMBF}$ varied from -0.54 to -9.37). With regard to deciduous and coniferous forests, all of the five models (from the most to the least accurate: ZH14, PZ10, KS12, Z01, and ZS14; KS12, ZH14, Z01, ZS14, and PZ10) under-predicted the measured $V_d$ by factors of 4.75 to 11.93, and 2.75 to 4.93, respectively.

A direct quantitative comparison of the accuracy of the five parameterizations with those reported in other studies is impossible because the metric used in the present study ($B_{NMBF}$) is not commonly used to evaluate the accuracy of the dry deposition models. However, qualitatively, our findings regarding the PZ10 performance for coniferous forests are in accordance with those reported by Petroff and Zhang (2010). They reported that the PZ10 parameterization under-predicted the measured deposition velocities for the following subset of observations that we also used for coniferous forest: Lammal et al. (1994), Gallagher et al. (1997), Buzorious et al. (2000), Gaman et al. (2004), and over-predicted for Grönholm et al. (2009).

The accuracy results over smooth surfaces suggest that, for the water surface, the best agreement between the measured and modeled $V_d$ was found for the ZH14 parameterization. Overall, the accuracy ranking from best to worst is as follows: ZH14, Z01, KS12, PZ10, and ZS14. Over ice/snow surface, the results suggest that the ZH14 is the most accurate parameterization, and PZ10 is the least accurate. Qualitatively, this finding is consistent with Petroff and Zhang (2010), who reported that their model significantly underestimated the measured deposition velocities over ice/snow surface for the following studies: Ibrahim (1983), Duan et al. (1987), Nilsson and Rannik (2001), and Contini et al. (2010), which were also used in the present study. We also note that the Z01 parameterization overestimated the measured $V_d$ from the aforementioned studies. This finding is consistent with Petroff and Zhang (2010), as they compared their model with Z01 over the ice/snow surface. One possible explanation for a large discrepancy between modeled and measured $V_d$ by PZ10 is an incorrect magnitude of the drift velocity applied, corresponding to phoretic effects on ice and snow.

Collectively for both rough and smooth surfaces, it is found that the ZH14 scheme is the most accurate for these LUCs: grass, deciduous forest, water, and ice/snow surfaces. KS12 performed slightly better for coniferous forest only. The performance of the PZ10 scheme could be viewed as moderate. This finding is interesting considering that the ZH14 is the simplest resistance-based scheme of the five parameterizations. We emphasize that Z01 and ZH14 parameterizations share similar structural features, but simplifications of the particle collection processes by constant values by ZH14 (see Eqs. (47-51)) could produce better agreement. In addition, we note that the KS12 parametrization is exclusively based on wind tunnel measurements and its performance over forest canopies is not satisfactory, as reported by the model developers Kouznetsov and Sofiev (2012). However, we find that KS12 performed the best over coniferous forests with the nine studies used in this research. However, Kouznetsov and Sofiev (2012) did not use the same subset of studies to evaluate the model performance as we used.

Given the complex nature and incomplete knowledge of the dry particle deposition process, it is of importance to account for the uncertainties in the modeled deposition velocities in atmospheric transport models (Petroff and Zhang, 2010; Zhang et al., 2017).
2012). Although there have been many dry deposition models developed over the years, the information on the model output uncertainties is meager and not up-to-date. To assert uncertainty on the modeled dry deposition velocities, Gould and Davidson (1992) adopted a step-wise uncertainty test of Slinn’s (1982) model. However, in reality, the model parameters are subject to simultaneous variability, and a OAT test cannot adequately propagate the error to the overall model outputs. This limitation was partially overcome by Ruijgrok (1992), who performed a probabilistic uncertainty test of Slinn’s model.

The Monte Carlo uncertainty analysis performed in this study assumes that in the five parameterizations all the major physical processes (e.g., turbulent diffusion, Brownian diffusion, impaction, interception, and gravitational settling) of dry particle deposition are accounted for satisfactorily. Thus, the uncertainty analysis conforms to the uncertainties in the model input variables and their overall contribution to the propagated uncertainties in the modeled dry deposition velocities. Additional uncertainties in the modeled deposition velocities could arise from inadequate model formulation. However, addressing this issue of a model’s structural uncertainty was outside the scope of this paper.

The values of the eight model parameters, covering four meteorological (\(U, u^*, L_o, \text{ and } RH\)) and four canopy morphological (\(z_o, d, h, \text{ and } LAI\)) properties, used in the Monte Carlo simulations were assumed to be uniformly distributed because their true distributions were unknown. It is emphasized that these parameters are not all necessarily independent; \(z_o\) and \(d\) are functions of the surface characteristics (Zhang and Sao, 2014; Shao and Yang, 2005, 2008). Considering these underlying assumptions, the uncertainties in modeled \(V_d\) reported in this paper should be viewed as the effect of the chosen parameter PDFs on the output uncertainty. The uncertainty bounds (i.e., the central 90\% values) reported in the Table 9 could be treated as a metric of the quality of the modeled outputs. The normalized uncertainties reported in this study are a useful indicator to assess the overall performance of a model for four particle modes (seven particle sizes) across five LUCs.

We applied Sobol’ sensitivity analysis to identify the most influential parameter(s) of the five parameterizations. Parameter rankings achieved using the Sobol’ first order indices for different models provide a robust evaluation of the models’ sensitivity by varying a set of input parameters within their plausible ranges. It is emphasized that a local sensitivity analysis such as OAT could lead to incomplete or misleading inference of the parameter sensitivity on the model’s output because assumptions of model linearity are not always justified for dry deposition parameterizations due to their complex formulations.

The Sobol’ sensitivity rankings presented in Table 11 can be used for inter-comparison between models’ parameter sensitivity. Over rough surfaces, for nucleation size particles (e.g., \(d_p = 0.001 \mu m\)), \(u^*\) is the most sensitive parameter for Z01, PZ10, KS12, and ZH14 parameterizations. As particle size increases from 0.001 \(\mu m\) to 1.0 \(\mu m\), except for the PZ10 scheme and for 1.0 \(\mu m\) for grass in KS12 scheme, \(u^*\) remains the most influential parameter. This finding is in accordance with previous studies (Zhang et al., 2001; Zhang and He, 2014) that show that dry particle deposition velocities for atmospheric particles are greatly influenced by friction velocity. We note that in the PZ10 scheme, \(LAI\) and \(L_o\) are the two most commonly-found sensitive parameters for \(d_p = 0.001\) to 1.0 \(\mu m\) for rough surfaces. As seen from the parameter rankings (Table 11), for \(d_p = 10 \mu m\) in the Z01, PZ10, KS10, ZH14 schemes, RH is the most influential factor. We postulate that with particle growth, high humidity may have a significant effect on coarse mode particles, and as a result, other model input parameters become less sensitive. The parameter ranking of the PZ10 scheme for deciduous forest shows that \(L_o\) is the most influential parameter. Similarly, for coniferous forest, \(L_o\) is found to be one of the most sensitive parameters for most particle sizes. One possible reason for this finding could be the interdependency of the particle mixing length parameter and \(L_o\) in the PZ10 scheme. Indeed, the mixing length indirectly relates to particle collection efficiencies in the PZ10 parameterization (see Eqs. 19, 26-27)). The rankings of the Z01 and ZH14 parameters are nearly identical for rough and smooth surfaces. This finding is not surprising given that these two parameterizations were developed using similar assumptions.
In general, dry particle deposition parameterizations developed for different particle size ranges and surfaces vary widely in terms of their complexity in model structure. The complexity in their numerical formulations often depends on the purpose (e.g., operational or research) of the model development (Petroff et al., 2008a). Comparing two previously developed one-dimensional aerosol deposition models for broadleaf and coniferous canopies (see details in Petroff et al., 2008b; Petroff et al., 2009) with the ZH14 parameterization, Petroff and Zhang (2010) argued that the mathematical formulations in those models are too complex and require numerous input parameters for implementation in aerosol transport models. Following this hypothesis, we attempt to qualitatively evaluate the relative complexity of the five dry particle deposition parameterizations tested in this study for incorporation into atmospheric transport models.

Of the five parameterizations, we note that the model structure of the PZ10 is relatively more complex than those of the Z01, ZH14, and ZS14 parameterizations. The complexity of the KS12 parameterization tends to be different by a large degree between rough (i.e., vegetative canopies and snow) and smooth (i.e., water) surfaces. The ZS14 formulation (Eqs. (52-65)) is of comparable complexity to the rough surface formulation in the KS12 parameterization (Eqs. (36-46)), and these parameterizations can be viewed as moderately complex. The formulation of the Z01 parameterization can be viewed as moderately complex as well. In this parameterization, three processes (Brownian diffusion, interception, and impaction) were parameterized using Eqs. (9-15) to describe the particle deposition at the collection surface. We claim that the KS12 parameterization for smooth surfaces is the most complex of the five models. This is mainly because it requires solving the dimensionless dry deposition velocity profiles over smooth surfaces using an analytical approach, which can be complex and computationally-expensive.

A direct qualitative comparison of the relative complexities of the major process terms in the PZ10 and Z01 parameterizations is possible because both of these parameterizations are resistance-based (i.e., expressions of $V_d$ in Eqs. 3 and 7 are of similar forms). It is evident from Eqs. (20-32) that the formulations in the PZ10 parameterization to compute the three surface collection process terms are relatively complex as compared to those in the Z01 parameterization. In the ZH14 parameterization (a resistance-based scheme as well), these process terms are not explicitly parameterized. Presumably, by incorporating a large number of LUC dependent constants to compute surface deposition velocity using Eqs. (47-51), simplifications were made possible to the ZH14 parameterization. The use of fitting parameters to account for poorly understood dry deposition processes in parameterizations is not uncommon. Due to the complex nature and inadequate understanding of the particle collection processes to leaf surfaces, suggestions were made to treat particle deposition on vegetative surfaces in a simplified manner using empirically derived fitting parameters (Petroff et al., 2008a). Consequently, Petroff and Zhang (2010) also introduced a large number of artificial parameters to account for characteristic length and orientation of the canopy obstacle, and different LUCs to parameterize the particle collection efficiencies (e.g., due to Brownian diffusion, interception, turbulent and inertial impaction). Based on these considerations and those in the previous paragraph, we claim that the ZH14 is the simplest of the five parameterizations.

6. Conclusions

In terms of overall performance for incorporation in atmospheric transport models, we suggest that parameterization accuracy and uncertainty should be considered jointly, while sensitivity of the model input parameters should be treated separately for each dry particle deposition parameterization. The paper presents a comprehensive evaluation of the performance of five parameterizations in terms of their accuracy, model output uncertainty, and parameter sensitivity. Based on the results, it is evident that the ZH14 parameterization is the most accurate for four of the five LUCs (grass, deciduous forest, water, and ice/snow surfaces) and second most accurate for the fifth LUC (coniferous forest). Of the five parameterizations, the uncertainty range for the ZH14 (11-20%) has the lowest upper bound across the five LUCs for particle size ranging from 0.005-2.5 µm. In terms of the lower bound of the uncertainty range, the ZH14 is second to the Z01 (10-30%) parameterization. We demonstrated
that the Sobol’ sensitivity analysis can be successfully applied to dry deposition models to rank the input parameters by taking into account the complex interactions between them.

The large dispersion in the parametrizations’ accuracy may indicate that despite considerable efforts in developing sophisticated process-based dry deposition models, there remain major gaps in our understanding of the dry deposition process. Another possible explanation for the large dispersion may be that it is significantly caused by measurement uncertainties, which were not addressed in this paper. However, inter-variability in modeled deposition velocities is not uncommon, as pointed out by Ruigrok et al. (1995) in an inter-comparison study of several earlier dry deposition models. We emphasize that the accuracy results presented in this paper should be discussed in terms of the locations in which the parameterization accuracy has been evaluated against measurements for the five LUCs (Table 1; Figure 1).

The results from the uncertainty analysis using the Monte Carlo simulations on the size-segregated particles should be of interest to atmospheric transport modelers as well as to the scientific community interested in quantifying the uncertainty bounds in the atmospheric deposition fluxes of pollutants to ecosystems using concentration data from monitoring stations. This is because until now, uncertainties in modeled $V_d$ for size-segregated particles for a suite of currently-available dry particle deposition parameterizations has been unavailable. We stress that future work on probabilistic uncertainty analysis should focus on quantifying uncertainties for additional LUCs than those covered in this study. One of the major limitations of our uncertainty analysis approach is the assumption of uniform distribution of all imprecise model input parameters. To address this limitation, accurate information on the input parameter PDFs is needed.

With the help of field observations, and improved theoretical knowledge of dry deposition, the Sobol’ parameter rankings could be used to fine-tune dry deposition models to better account for processes that are currently lacking or poorly parameterized. Future work should focus on estimating higher order (i.e., second order and total order) Sobol’ indices. Such indices would be useful for model developers interested in understanding the joint influence of multiple input parameters on the modeled deposition velocities.

Based on the qualitative evaluation of relative complexity of the five parameterizations, we suggest that the model structure of the ZH14 parameterization is the least complex. After reviewing over 100 air quality models, Kouznetsov and Sofiev (2012) reported that resistance-based approaches are extensively implemented in most of those models. Thus, in practice, it may be preferable to use a relatively simple parameterization over a complex (and potentially computationally expensive) one, if the accuracy and uncertainty of the model justify it. Based on these criteria (i.e., accuracy, uncertainty, and complexity), we propose that, of the five parameterizations we tested, the ZH14 parameterization is currently superior for incorporation into atmospheric transport models.

7. Code availability
The codes supporting this study can be found online at: https://osf.io/a6q97/

Notes: The authors declare no competing financial interest.

8. Acknowledgements
This project was funded by the National Science Foundation’s (NSF’s) Coupled-Natural-Human System Program through Award No. 1313755.
9. References


Ibrahim, M., Barrie, L., and Fanaki, F.: An experimental and theoretical investigation of the dry deposition of particles to snow, pine trees and artificial collectors, Atmospheric Environment (1967), 17, 781-788, 1983.


Zhang, J. and Shao, Y.: A new parameterization of particle dry deposition over rough surfaces, Atmospheric Chemistry and Physics, 14, 12429-12440, 2014.


Table 1. Measurement studies used to evaluate the five parameterizations.

<table>
<thead>
<tr>
<th>Land use category</th>
<th>No.</th>
<th>Title</th>
<th>Authors</th>
<th>Location</th>
<th>Latitude</th>
<th>Longitude</th>
<th>Elevation (m)</th>
<th>Sampling dates</th>
<th>Particle size (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grass</td>
<td>1</td>
<td>Dry deposition of fine aerosol to a grass surface</td>
<td>Allen et al. (1991)</td>
<td>U.K.</td>
<td>51.88° N</td>
<td>0.94° E</td>
<td>28</td>
<td>June, 1988–June, 1989</td>
<td>0.48</td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>An eddy-correlation measurement of particulate deposition from the atmosphere</td>
<td>Westoby et al. (1997)</td>
<td>U.S.</td>
<td>38.84° N</td>
<td>90.06° W</td>
<td>225</td>
<td>February–March, 1976</td>
<td>0.075</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>Eddy correlation measurements of atmospheric fluxes of ozone, sulphur, and particulates during the Champaign intercomparison study</td>
<td>Neumann and den Hartog (1985)</td>
<td>U.S.</td>
<td>40.11° N</td>
<td>88.27° W</td>
<td>233</td>
<td>June, 1982</td>
<td>0.10–2.28</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>Micrometeorological measurements of particle deposition velocities to moorland vegetation</td>
<td>Nemitz et al. (2002)</td>
<td>U.K.</td>
<td>55.79° N</td>
<td>3.23° W</td>
<td>109</td>
<td>May–October, 1999</td>
<td>0.12–0.45</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>Eddy correlation measurements of aerosol deposition to grass</td>
<td>Vogt et al. (2004)</td>
<td>U.S.</td>
<td>44.46° N</td>
<td>123.11° W</td>
<td>81</td>
<td>May–June, 2000</td>
<td>0.52</td>
</tr>
<tr>
<td>Coniferous forest</td>
<td>6</td>
<td>The Landes experiment: Biopshere-atmosphere exchanges of ozone and aerosol particles above a pine forest</td>
<td>Lamont et al. (1994)</td>
<td>France</td>
<td>44.84° N</td>
<td>5.08° W</td>
<td>58</td>
<td>June, 1992</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>Micrometeorological measurement of the dry deposition flux of sulphate and nitrate aerosols to coniferous forest</td>
<td>Wyers and Duyers (1996)</td>
<td>The Netherlands</td>
<td>52.27° N</td>
<td>5.71° E</td>
<td>26</td>
<td>April–December, 1993</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>8</td>
<td>Atmospheric particles and their interactions with natural surfaces</td>
<td>Gallagher et al. (1997)</td>
<td>The Netherlands</td>
<td>52.27° N</td>
<td>5.71° E</td>
<td>26</td>
<td>June–July, 1993</td>
<td>0.16–0.50</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>The dry deposition of particles to a forest canopy: a comparison of model and experimental results</td>
<td>Rongrok et al. (1997)</td>
<td>The Netherlands</td>
<td>52.27° N</td>
<td>5.71° E</td>
<td>26</td>
<td>June–July, 1993</td>
<td>0.35–0.60</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>Deposition velocities of nucleation mode particles into a Scots pine forest</td>
<td>Rannik et al. (2000)</td>
<td>Finland</td>
<td>61.85° N</td>
<td>24.28° E</td>
<td>181</td>
<td>September, 2000</td>
<td>0.015–0.35</td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>Vertical aerosol fluxes measured by the eddy covariance method and deposition of nucleation mode particles above a Scots pine forest in southern Finland</td>
<td>Buzorius et al. (2000)</td>
<td>Finland</td>
<td>61.85° N</td>
<td>24.28° E</td>
<td>181</td>
<td>March–May, 1997</td>
<td>0.015</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>Relaxed eddy accumulation system for size-resolved aerosol particle flux measurements</td>
<td>Geman et al. (2004)</td>
<td>Finland</td>
<td>61.85° N</td>
<td>24.28° E</td>
<td>181</td>
<td>September–October, 2001</td>
<td>0.05</td>
</tr>
<tr>
<td>Deciduous forest</td>
<td>13</td>
<td>Analyses of flux methods and functional dependencies</td>
<td>Pryor et al. (2007)</td>
<td>Finland</td>
<td>61.85° N</td>
<td>24.28° E</td>
<td>181</td>
<td>January–December, 2004</td>
<td>0.016–0.07</td>
</tr>
<tr>
<td></td>
<td>14</td>
<td>Aerosol particle dry deposition to canopy and forest floor measured by two-layer eddy covariance system</td>
<td>Grottoli et al. (2009)</td>
<td>Finland</td>
<td>61.85° N</td>
<td>24.28° E</td>
<td>181</td>
<td>March, 2003</td>
<td>0.015–0.05</td>
</tr>
<tr>
<td></td>
<td>15</td>
<td>Fluxes of gosses and particles above a deciduous forest in wintertime</td>
<td>Westoby et al. (1983)</td>
<td>U.S.</td>
<td>35.98° N</td>
<td>78.91° W</td>
<td>77</td>
<td>January–February, 1981</td>
<td>0.48</td>
</tr>
<tr>
<td></td>
<td>16</td>
<td>Size-resolved particle deposition velocities of sub-100nm diameter particles over a forest</td>
<td>Pyor (2006)</td>
<td>Denmark</td>
<td>55.48° N</td>
<td>11.64° E</td>
<td>40</td>
<td>May–June, 2004</td>
<td>0.025–0.065</td>
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<tr>
<td></td>
<td>17</td>
<td>Deposition velocity of PM2.5 sulfate in the summer above a deciduous forest in central Japan</td>
<td>Matsuda et al. (2012)</td>
<td>Japan</td>
<td>36.40° N</td>
<td>138.58° E</td>
<td>1380</td>
<td>July, 2009</td>
<td>2.5</td>
</tr>
<tr>
<td>Water</td>
<td>18</td>
<td>Mechanisms of transport from the atmosphere to the Earth’s surface</td>
<td>Möller and Schumann (1970)</td>
<td>(Wind tunnel)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.03–1.13</td>
</tr>
<tr>
<td></td>
<td>19</td>
<td>Particle deposition rates on a water surface as a function of particle diameter and air velocity</td>
<td>Schimel et al. (1994)</td>
<td>(Wind tunnel)</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.25–29</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>Airborne concentrations and dry deposition fluxes of particulate species to surrogate surfaces deployed in southern Lake Michigan</td>
<td>Zulfali et al. (1998)</td>
<td>U.S.</td>
<td>44.00° N</td>
<td>87.00° W</td>
<td>-</td>
<td>July, 1994</td>
<td>0.4–24.0</td>
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<tr>
<td></td>
<td>21</td>
<td>Determination of size-dependent dry particle deposition velocities with multiple intrasurface elemental tracers</td>
<td>Caffrey et al. (1998)</td>
<td>U.S.</td>
<td>44.00° N</td>
<td>87.00° W</td>
<td>-</td>
<td>July, 1994</td>
<td>0.05–48.0</td>
</tr>
<tr>
<td>Ice/snow</td>
<td>22</td>
<td>Aerosol dry deposition measured with eddy-covariance technique at Waza and Aboa, Dronning Maud Land, Antarctica</td>
<td>Grønland et al. (2002)</td>
<td>Antarctica</td>
<td>73.00° S</td>
<td>13.41° W</td>
<td>584</td>
<td>January, 2000</td>
<td>0.02–0.2</td>
</tr>
<tr>
<td></td>
<td>23</td>
<td>Deposition velocity of ultrafine particles measured with the Eddy-Correlation Method over the Nansen Ice Sheet (Antarctica)</td>
<td>Constan et al. (2010)</td>
<td>Antarctica</td>
<td>74.88° S</td>
<td>163.17° W</td>
<td>84.7</td>
<td>December, 2006</td>
<td>0.06–0.070</td>
</tr>
<tr>
<td></td>
<td>24</td>
<td>On the potential contribution of open lead particle emissions to the central Arctic aerosol concentration</td>
<td>Held et al. (2011a)</td>
<td>Arctic Ocean</td>
<td>87.00° N</td>
<td>6.00° W</td>
<td>-</td>
<td>August–September, 2008</td>
<td>0.045</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>Near-surface profiles of aerosol number concentration and temperature over the Arctic Ocean</td>
<td>Held et al. (2011b)</td>
<td>Arctic Ocean</td>
<td>87.00° N</td>
<td>6.00° W</td>
<td>-</td>
<td>August–September, 2008</td>
<td>0.045</td>
</tr>
<tr>
<td></td>
<td>26</td>
<td>Correlation of dry deposition velocity and fraction velocity over different surfaces for PM2.5 and particle number concentrations</td>
<td>Donato and Contini (2014)</td>
<td>Antarctica</td>
<td>74.88° S</td>
<td>163.17° W</td>
<td>-</td>
<td>December, 2006</td>
<td>0.015–0.13</td>
</tr>
<tr>
<td></td>
<td>27</td>
<td>An experimental and theoretical investigation of the dry deposition of particles to snow, pine trees and artificial collectors</td>
<td>Ibrahim (1983)</td>
<td>Canada</td>
<td>51.25° N</td>
<td>85.32° W</td>
<td>450</td>
<td>February, 1989</td>
<td>0.7–0.8</td>
</tr>
<tr>
<td></td>
<td>28</td>
<td>Eddy correlation measurements of the dry deposition of particles in wintertime</td>
<td>Dunn et al. (1987)</td>
<td>U.S.</td>
<td>40.70° N</td>
<td>77.96° W</td>
<td>177</td>
<td>December, 1985</td>
<td>0.22–0.73</td>
</tr>
<tr>
<td></td>
<td>29</td>
<td>Turbulent aerosol fluxes over the Arctic Ocean 1. Dry deposition over sea and pack ice</td>
<td>Nilsson and Rannik (2001)</td>
<td>Arctic Ocean</td>
<td>88.00° N</td>
<td>15.00° E</td>
<td>-</td>
<td>August, 1999</td>
<td>0.02–0.08</td>
</tr>
</tbody>
</table>
Table 2. Parameter values and associated uncertainties used in Monte Carlo simulation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Base value (assumed)</th>
<th>Uncertainty range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative humidity, RH (%)</td>
<td>80 (all LUCs)</td>
<td>±5%</td>
<td>Heinonen (2002)</td>
</tr>
<tr>
<td>Wind speed, U (m/s)</td>
<td>4 (all LUCs)</td>
<td>±3%</td>
<td>Högström and Smedman (2004)</td>
</tr>
<tr>
<td>Friction velocity, u* (m/s)</td>
<td>0.3 (all LUCs)</td>
<td>±10%</td>
<td>Andreas (1992)</td>
</tr>
<tr>
<td>Monin-Obukhov length, L0 (m)</td>
<td>50 (all LUCs)</td>
<td>±10%</td>
<td>Weidinger et al. (2000)</td>
</tr>
<tr>
<td>Roughness length, z0 (m)</td>
<td>0.04 (Grass)</td>
<td>±25%</td>
<td>Su et al. (2001)</td>
</tr>
<tr>
<td>Canopy height, h (m)</td>
<td>15 (Coniferous forest)</td>
<td>±5%</td>
<td>Larjavaara and Muller-Landau (2013)</td>
</tr>
<tr>
<td>Zero-plane displacement height, d (m)</td>
<td>7 (Coniferous forest)</td>
<td>±25%</td>
<td>Su et al. (2001)</td>
</tr>
<tr>
<td>Leaf area index (one-sided), LAI (m²/m²)</td>
<td>10 (Deciduous forest)</td>
<td>±5%</td>
<td>Richardson et al. (2011)</td>
</tr>
</tbody>
</table>

Table 3. Input parameter ranges for the Sobol' sensitivity analysis.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative humidity, RH (%)</td>
<td>10-100 (all LUCs)</td>
<td>Assumed</td>
</tr>
<tr>
<td>Wind speed, U (m/s)</td>
<td>1-5 (all LUCs)</td>
<td>Studies # 1-29</td>
</tr>
<tr>
<td>Friction velocity, u* (m/s)</td>
<td>0.1-0.5 (all LUCs)</td>
<td>Studies # 1-29</td>
</tr>
<tr>
<td>Monin-Obukhov length, L0 (m)</td>
<td>10-100 (all LUCs)</td>
<td>Studies # 1-29</td>
</tr>
<tr>
<td>Roughness length, z0 (m)</td>
<td>0.02-0.10 (Grass)</td>
<td>Studies # 1-5</td>
</tr>
<tr>
<td>Canopy height, h (m)</td>
<td>14-20 (Coniferous forest)</td>
<td>Studies # 6-14</td>
</tr>
<tr>
<td>Zero-plane displacement height, d (m)</td>
<td>7-12 (Coniferous forest)</td>
<td>Studies # 6-14</td>
</tr>
<tr>
<td>Leaf area index (one-sided), LAI (m²/m²)</td>
<td>0.2-10 (Deciduous forest)</td>
<td>Studies # 6-14</td>
</tr>
</tbody>
</table>

1Studies are listed in Table 1.

Table 4. Results of the normalized mean bias factors for grass (bold-faced value indicates the most accurate parameterization).

<table>
<thead>
<tr>
<th>Study</th>
<th>Z0I</th>
<th>PZ10</th>
<th>KS12</th>
<th>ZH14</th>
<th>ZS14</th>
</tr>
</thead>
<tbody>
<tr>
<td>Allen et al. (1991)</td>
<td>-17.61</td>
<td>15.96</td>
<td>-18.12</td>
<td>-0.55</td>
<td>-5.13</td>
</tr>
<tr>
<td>Wesely et al. (1977)</td>
<td>-2.78</td>
<td>-28.78</td>
<td>-7.56</td>
<td>-10.62</td>
<td>-102.92</td>
</tr>
<tr>
<td>Neumann and den Hartog (1985)</td>
<td>0.96</td>
<td>-0.12</td>
<td>-0.50</td>
<td>4.79</td>
<td>0.56</td>
</tr>
<tr>
<td>Nemitz et al. (2002)</td>
<td>5.15</td>
<td>1.12</td>
<td>-3.82</td>
<td>2.17</td>
<td>-0.10</td>
</tr>
<tr>
<td>Vong et al. (2004)</td>
<td>-4.55</td>
<td>-4.55</td>
<td>-25.71</td>
<td>-2.12</td>
<td>-4.03</td>
</tr>
<tr>
<td>Five studies</td>
<td>5.45</td>
<td>-1.80</td>
<td>-9.37</td>
<td>-0.54</td>
<td>-4.30</td>
</tr>
</tbody>
</table>
Table 5. Results of the normalized mean bias factors for coniferous forest (bold-faced value indicates the most accurate parameterization).

<table>
<thead>
<tr>
<th>Study</th>
<th>Dry particle deposition parameterization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Z01</td>
</tr>
<tr>
<td>Lamaud et al. (1994)</td>
<td>0.77</td>
</tr>
<tr>
<td>Ruigrok et al. (1997)</td>
<td>-5.70</td>
</tr>
<tr>
<td>Rannik et al. (2000)</td>
<td>3.16</td>
</tr>
<tr>
<td>Buzorious et al. (2000)</td>
<td>0.75</td>
</tr>
<tr>
<td>Gaman et al. (2004)</td>
<td>-0.90</td>
</tr>
<tr>
<td>Pryor et al. (2007)</td>
<td>0.69</td>
</tr>
<tr>
<td>Grönholm et al. (2009)</td>
<td>0.95</td>
</tr>
<tr>
<td>Nine studies</td>
<td>-2.35</td>
</tr>
</tbody>
</table>

Table 6. Results of the normalized mean bias factors for deciduous forest (bold-faced values indicate the most accurate parameterization).

<table>
<thead>
<tr>
<th>Study</th>
<th>Dry particle deposition parameterization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Z01</td>
</tr>
<tr>
<td>Wesely et al. (1983)</td>
<td>-9.25</td>
</tr>
<tr>
<td>Matsuda et al. (2012)</td>
<td>-1.34</td>
</tr>
<tr>
<td>Three studies</td>
<td>-8.11</td>
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</tbody>
</table>

Table 7. Results of the normalized mean bias factors for water surfaces (bold-faced value indicates the most accurate parameterization).

<table>
<thead>
<tr>
<th>Study</th>
<th>Dry particle deposition parameterization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Z01</td>
</tr>
<tr>
<td>Möller and Schumann (1970)</td>
<td>18.87</td>
</tr>
<tr>
<td>Sehmel et al. (1974)</td>
<td>0.44</td>
</tr>
<tr>
<td>Zufall et al. (1998)</td>
<td>-0.47</td>
</tr>
<tr>
<td>Caffrey et al. (1998)</td>
<td>0.75</td>
</tr>
<tr>
<td>Four studies</td>
<td>0.52</td>
</tr>
</tbody>
</table>

Table 8. Results of the normalized mean bias factors for ice/snow surfaces (bold-faced value indicates the most accurate parameterization).

<table>
<thead>
<tr>
<th>Study</th>
<th>Dry particle deposition parameterization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Z01</td>
</tr>
<tr>
<td>Gronlund et al. (2002)</td>
<td>-1.22</td>
</tr>
<tr>
<td>Contini et al. (2010)</td>
<td>5.68</td>
</tr>
<tr>
<td>Held et al. (2011a)</td>
<td>2.96</td>
</tr>
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<td>Held et al. (2011b)</td>
<td>2.78</td>
</tr>
<tr>
<td>Donatoe and Contini (2014)</td>
<td>1.62</td>
</tr>
<tr>
<td>Duan et al. (1987)</td>
<td>0.22</td>
</tr>
<tr>
<td>Nilsson and Rannik (2001)</td>
<td>1.69</td>
</tr>
<tr>
<td>Eight studies</td>
<td>1.98</td>
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</table>
Table 9. Median (50th percentile) values of the simulated dry deposition velocities (m s\(^{-1}\)) with 5th and 95th percentiles of distribution indicating the range of uncertainty.

<table>
<thead>
<tr>
<th>Land use category</th>
<th>Particle size (μm)</th>
<th>5th</th>
<th>95th</th>
<th>5th</th>
<th>95th</th>
<th>5th</th>
<th>95th</th>
<th>5th</th>
<th>95th</th>
<th>5th</th>
<th>95th</th>
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</thead>
<tbody>
<tr>
<td>Grass</td>
<td>0.005</td>
<td>1.72×10(^{-3})</td>
<td>1.99×10(^{-3})</td>
<td>2.10×10(^{-3})</td>
<td>2.21×10(^{-3})</td>
<td>9.00×10(^{-4})</td>
<td>9.58×10(^{-4})</td>
<td>3.06×10(^{-4})</td>
<td>1.17×10(^{-4})</td>
<td>1.77×10(^{-4})</td>
<td>1.68×10(^{-4})</td>
</tr>
<tr>
<td>Coniferous Forest</td>
<td>0.005</td>
<td>4.42×10(^{-3})</td>
<td>4.92×10(^{-3})</td>
<td>5.32×10(^{-3})</td>
<td>1.05×10(^{-2})</td>
<td>0.00×10(^{-2})</td>
<td>0.00×10(^{-2})</td>
<td>2.63×10(^{-2})</td>
<td>2.78×10(^{-2})</td>
<td>2.92×10(^{-2})</td>
<td>1.44×10(^{-2})</td>
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<tr>
<td>Deciduous Forest</td>
<td>0.005</td>
<td>3.09×10(^{-3})</td>
<td>3.09×10(^{-3})</td>
<td>3.26×10(^{-3})</td>
<td>1.16×10(^{-2})</td>
<td>1.20×10(^{-2})</td>
<td>1.34×10(^{-2})</td>
<td>1.45×10(^{-2})</td>
<td>1.56×10(^{-2})</td>
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<tr>
<td>Water</td>
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<td>4.00×10(^{-4})</td>
<td>4.43×10(^{-4})</td>
<td>4.83×10(^{-4})</td>
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<td>1.00×10(^{-5})</td>
<td>1.00×10(^{-5})</td>
<td>5.00×10(^{-6})</td>
<td>6.00×10(^{-6})</td>
<td>6.00×10(^{-6})</td>
<td>5.00×10(^{-6})</td>
</tr>
<tr>
<td>Ice/snow</td>
<td>0.005</td>
<td>1.15×10(^{-4})</td>
<td>1.26×10(^{-4})</td>
<td>1.38×10(^{-4})</td>
<td>4.00×10(^{-5})</td>
<td>4.00×10(^{-5})</td>
<td>4.00×10(^{-5})</td>
<td>4.00×10(^{-5})</td>
<td>4.00×10(^{-5})</td>
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<td>4.00×10(^{-5})</td>
</tr>
</tbody>
</table>

*The parameterization does not categorize rough or vegetative surfaces into different LUCs. In this analysis, grass is used to represent a rough surface.
Table 10. Normalized uncertainties in modeled dry deposition velocities.

<table>
<thead>
<tr>
<th>Land use category</th>
<th>Particle size, $d_p$ (µm)</th>
<th>Dry particle deposition parameterization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Z01</td>
</tr>
<tr>
<td>Grass</td>
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</tr>
<tr>
<td></td>
<td>0.05</td>
<td>0.18</td>
</tr>
<tr>
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<td>0.22</td>
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<tr>
<td></td>
<td>1.0</td>
<td>0.16</td>
</tr>
<tr>
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<td>1.5</td>
<td>0.14</td>
</tr>
<tr>
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<td>2.0</td>
<td>0.12</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>0.14</td>
</tr>
<tr>
<td>Coniferous forest</td>
<td>0.005</td>
<td>0.20</td>
</tr>
<tr>
<td></td>
<td>0.05</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>0.13</td>
</tr>
<tr>
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<td>1.0</td>
<td>0.16</td>
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<tr>
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<td>1.5</td>
<td>0.13</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>0.11</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>0.14</td>
</tr>
<tr>
<td>Deciduous forest</td>
<td>0.005</td>
<td>0.21</td>
</tr>
<tr>
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<td>0.13</td>
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<td>0.10</td>
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<tr>
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<td>2.5</td>
<td>0.14</td>
</tr>
<tr>
<td>Water</td>
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<td>0.21</td>
</tr>
<tr>
<td></td>
<td>0.05</td>
<td>0.18</td>
</tr>
<tr>
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<tr>
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<td>2.0</td>
<td>0.15</td>
</tr>
<tr>
<td></td>
<td>2.5</td>
<td>0.11</td>
</tr>
<tr>
<td>Ice/snow</td>
<td>0.005</td>
<td>0.18</td>
</tr>
<tr>
<td></td>
<td>0.05</td>
<td>0.16</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
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<td>2.5</td>
<td>0.30</td>
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</tbody>
</table>

Note: a normalized uncertainty value of zero indicates that the 95th and 5th percentile $V_d$ are of equal magnitude.
<table>
<thead>
<tr>
<th>Land use category</th>
<th>$d_p$ (μm)</th>
<th>Z01</th>
<th>PZ10</th>
<th>KS12</th>
<th>ZH14</th>
<th>ZS14</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grass</td>
<td>0.001</td>
<td>$u_c, z_0, L_0$ (RH, $p_o$)</td>
<td>$u_c, L_0, z_0, U, L_0, (h, d, RH, $p_o$)</td>
<td>$u_c, (RH, p_o, LAI)$</td>
<td>$u_c, (z_0, L_0, RH, p_o)$</td>
<td>$U, u_c, (RH, p_o)$</td>
</tr>
<tr>
<td></td>
<td>0.01</td>
<td>$u_c, z_0, L_0$ (RH, $p_o$)</td>
<td>$u_c, L_0, z_0, U, (L_0, h), (z_0, d, RH, $p_o$)</td>
<td>$u_c, (RH, p_o, LAI)$</td>
<td>$u_c, (z_0, L_0, RH, p_o)$</td>
<td>$U, u_c, (RH, p_o)$</td>
</tr>
<tr>
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<td>$u_c, L_0, U, d, (u_c, L_0, h, z_0, RH, $p_o$)$</td>
<td>$u_c, (RH, p_o, LAI)$</td>
<td>$u_c, (z_0, L_0, RH, p_o)$</td>
<td>$U, U_c, (RH, p_o)$</td>
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<tr>
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<td>$RH, u_c, p_o, LAI$</td>
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<td>$U, U_c, RH, p_o$</td>
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<td>$RH, u_c, U, LAI, (p_o, z_0, L_0, h, d)$</td>
<td>$RH, u_c, p_o, LAI$</td>
<td>$RH, p_o, (U_c, L_0, z_0)$</td>
<td>$U, U_c, RH, p_o$</td>
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<tr>
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<td>$u_c, (RH, p_o, LAI)$</td>
<td>$u_c, L_0, (z_0, RH, p_o)$</td>
<td>$U, u_c, (RH, p_o)$</td>
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<tr>
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<td>$u_c, L_0, (z_0, RH, p_o)$</td>
<td>$U, u_c, (RH, p_o)$</td>
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<tr>
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<td>$u_c, (L_0, RH, p_o)$</td>
<td>$u_c, (L_0, p_o, LAI)$</td>
<td>$u_c, (L_0, RH, p_o)$</td>
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<td>$u_c, (L_0, RH, p_o)$</td>
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<td>$u_c, (L_0, RH, p_o)$</td>
<td>$U, L_0, (z_0, RH, p_o)$</td>
</tr>
<tr>
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<td>$p_o, u_c, (L_0, RH)$</td>
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<td>$U, RH, (L_0, p_o)$</td>
</tr>
<tr>
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<td>$U, U_c, (L_0, z_0, RH, p_o)$</td>
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<td>$U, U_c, (L_0, z_0, RH, p_o)$</td>
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</table>

Note: the ranking of the parameters is from most sensitive to least sensitive. Parameters within the parentheses have identical Sobol’ first order indices.