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Evaluation and Development of Models for Resuspension of Aerosols at Short Times after Deposition

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Abstract

Resuspension is known to transport hazardous particles in the natural environment, moving a fraction of deposited material back into the atmosphere. This process is notoriously difficult to model, given the complexity of the turbulent boundary layer and chemistry of the three-phase interface (air, liquid, solid) typically found at the land surface. Wind tunnel studies have demonstrated the importance of resuspension within a short time after deposition, but there exists no robust model for short-term resuspension. Numerical simulations of accidental or terrorist releases of hazardous materials need such a model to accurately predict fate-and-transport of the materials within hours to days after release. Many accepted conventional models were derived from resuspension data for aged sources, such as former weapons test sites; these data sets, and the associated models, may not be appropriate for short-time resuspension. The study described here re-examined historical wind tunnel data on short-term resuspension, with the goal of developing a model appropriate for numerical simulations. Empirical models are derived from these data using a suite of parameters (friction velocity, particle diameter, surface roughness, particle density, and time). These empirical models, and the wind tunnel data, are compared quantitatively with existing conventional models from the literature. The conventional models underpredict short-time resuspension, resulting in order-of-magnitude errors in predictions of resuspended mass. Only three models perform reasonably well: the empirical models derived from the data and an adaptation of the NCRP 129 model. More data are needed to validate the empirical models and build the physical understanding of the processes involved.

Keywords

resuspension, deposition, aerosol, particle, modeling

Introduction

Resuspension is the process by which particles are moved from an outdoor surface (the ground, a tree leaf, etc.) into the atmosphere. Episodes of resuspension have been demonstrated to transport a variety of contaminants, including radionuclides (Anspaugh et al., (1975), Garland and Pomeroy, (1994), Pires Do Rio et al., (1994)), metals (Warren and Birch, (1987)), PCBs (Bremle and Larsson, (1998)), and biological spores (Chinn, (2000)). A review of literature relevant to the field of resuspension is available in Loosmore, (2000). These and other studies establish the necessity of including resuspension in modeling and predictive studies of contaminant fate and transport in the environment. In particular, in the event of an accidental or malicious release of a chemical, biological, or radiological material, emergency responders need fast and accurate predictions of air concentrations and deposition patterns in the immediate post-release period (hours to days) to make decisions about sheltering, evacuation, and decontamination. The process of resuspension depends on micro-scale features of the particle-surface interface, such as the local water content and chemistry, as well as the larger-scale fluid mechanical removal forces. There currently exists no robust mechanistic model that is applicable to small and large fluxes at short and long times after deposition. Instead, modelers rely on simple models, such as those for the resuspension factor K and resuspension rate Λ.

The resuspension factor K [1/m] is defined to be the air concentration at the breathing height divided by the initial surface deposit; this quantity is readily measured in the field and thus appears in numerous data sets. This quantity is difficult to interpret when there is the potential for upwind contamination, when the surface deposition is inhomogeneous, and when there is variability in meteorological conditions (Nicholson, (1988)). The resuspension rate Λ [1/s] represents the resuspension flux from the surface, divided by the initial surface concentration; fluxes are not measured directly in the field but may be inferred from concentration profiles (Anspaugh et al., (1975)). A variety of mathematical models exist for K and Λ, however the models show great variability and uncertainty even when applied to longer-term data sets (Garger et al., (1996),(1997)). Garger et al., (1999) pointed out in detail the problems in existing models, including sensitivity to the choice of initial resuspension factor; these authors indicate that these models work at best for predicting annual air concentrations in homogeneous conditions. The applicability of the existing models to short-term resuspension (occurring within the first hours to days of an event) has not been tested.

Most of the existing data on resuspension was obtained months to years after the initial deposition. For example, studies of plutonium resuspension at the Nevada Test Site and elsewhere occurred years after the weapons testing had ceased (Shinn et al., (1986), Johnston et al., (1993)). The Chernobyl release lasted two weeks, so the initial deposition period is poorly defined; moreover, much of the resuspension data was measured in the years following the accident (Garland and Pomeroy, (1994), Garger et al., (1996), Hollander, (1994)). These long-term data sets are poor surrogates for short-term emergency response scenarios. After deposition, particulates can become mixed in the soil profile, through natural faunal activity, rain, soil erosion, and human intervention (Brimhall et al., (1991), Rogowski and Tamura, (1970)). Immediately after deposition, particles with the smallest adhesive forces are thought to be removed by wind, leaving behind a more firmly-held contaminant reservoir (Cleaver and Yates, (1973), Ziskind et al., (1995), Zimon, (1969)). For these reasons, the resuspension flux is expected to decrease non-linearly with time after deposition.

This paper describes an effort to develop a model appropriate to short-time resuspension. Data sets for wind tunnel studies of short-time resuspension were taken from the literature and analyzed here to develop an empirical model for the resuspension rate. Five data sets were considered. Three were considered excellent surrogates for short-time resuspension in the natural environment: Nicholson, (1993), Garland, (1982), Giess et al., (1997); these were used to develop the NGG model. The remaining data sets were used for model testing. The data sets, the model development, and testing are described below.

The NGG Data Sets

The three NGG data sets (Nicholson, (1993), Garland, (1982), Giess et al., (1997)) were used for model development because the experiments were described in detail, the deposited material was unaltered before exposure to the wind, and resuspension rates could be calculated easily from measured quantities. The three experiments include 159 data points, spanning a range of parameters, as shown in Table 1. Three types of surfaces were represented: bare soil, concrete, and grass with different heights. The set includes friction velocity (u_*) values from 0.1 to 1.4 m/s. Two types of particles were used, with different densities, and with a size range from submicron to silt. These wind tunnel experiments yield evidence that quite a bit of material can be removed at very short times. For example, in several of Nicholson's trials ~15% of the deposited material was removed; moreover, in many trials, nearly 50% of the total resuspension occurred within the first 10 seconds (Nicholson, (1993)).

The resuspension rates are shown plotted as a function of time in Figure 1. For the Nicholson data sets, as indicated in Table 1, resuspension rates were computed from the time-history of the fraction removed. As discussed by Nicholson, (1993), because these fractions were measured over intervals of time, care must be exercised in assigning a time to the derived resuspension rate. One possibility is to assume that the measured resuspension rate is representative of the interval mid-point. However, it might be more appropriate to take account of the fact that resuspension rates are known to fall roughly as 1/t. Thus, find the value of t′ within T+ΔT that satisfies:

$$t' = \frac{1}{\Delta T} \int_{T}^{T+\Delta T} \frac{1}{t} dt$$

This method ensures that the product of t′ and ΔT matches the area under the 1/t curve.

Roughness heights for the experiments were estimated at 1/10th the effective obstacle height, which corresponds approximately to the average of the results of formulas given by Lettau, (1969). The effective obstacle heights used were the grass heights as given; the bare soil and concrete were assumed to have effective obstacle heights of 1 cm and 3 mm, respectively. Particle size is specified explicitly by Nicholson, (1993) and Giess et al., (1997). Garland, (1982) uses a submicron powder, but the particle distribution is not given: a value of 0.5 micron is used here.

The friction velocity was not specified by Garland, (1982). The velocities given in his paper are assumed to be the centerline velocities at 0.5 m. Using the estimated roughness of 0.001 m, the ratio of the centerline velocity to the friction velocity was computed at roughly 15, from the semilog profile:

$$\frac{U}{u*} = 2.5\ln\left(\frac{z}{z_o}\right)$$

where z_o is the aerodynamic roughness height, after Stull, (1999) p. 378. By contrast, data in Nicholson, (1993) show a factor of 30 for flow over concrete.

Model Development

The NGG data sets described above were used to develop a model for short-time resuspension. Efforts were made to develop an analytic model for the resuspension rate using non-dimensional analysis, but none were satisfactory. Because there is quite a bit of scatter in the data, as seen in Figure 1, a relatively straightforward least-squares regression was performed in Excel. Five parameters were used: the friction velocity, u_*; time since the windflow began, t; the particle diameter, d_p; the particle density, ρ_p; and the roughness, z_o. The resuspension rate was assumed to vary either in power law relationships or exponential relationships with the parameters, and a variety of models were examined. The best model fits to these data were the following, named "emp1 and "emp3" for *empirical*:

$$\Lambda = 0.42\frac{u_*^{2.13} d_p^{0.17}}{t^{0.92} z_o^{0.32} \rho_p^{0.76}} \ [\mathrm{s}^{-1}] \qquad \text{model emp1}$$

$$\Lambda = 0.01\frac{u_*^{1.43}}{t^{1.03}} \ [\mathrm{s}^{-1}] \qquad \text{model emp3}$$

where the resuspension rate Λ has dimensions of s^{-1} and all other dimensions are as in Table 1. Although empirical, this model is physically realistic: the resuspension rate increases with u_* and particle diameter (within the range of interest) and decreases with time, surface roughness, and particle density. These trends are consistent with intuition and observations: resuspension increases with velocity but falls in time; larger surface roughness provides more shielding; heavier particles are harder to move; larger particles stick up higher into the boundary layer and thus experience higher removal forces.

The Excel regression provides some measures of the goodness-of-fit. The student t statistic is available to test the contribution of the various parameters. The t statistic and R^2 are given in Table 2 below. For a 2-tailed test, the critical t values corresponding to 5%, 1%, and 0.1% confidence are 1.96, 2.58, and 3.29, respectively. As may be seen in the table, t and u_* are unequivocally important parameters, but the contributions of the other parameters are less convincing. Model performance is evaluated in the next section.

Comparison to Existing Models

The above two models were tested against four other models, which predict resuspension factors:

1. $K = 10^{-6} \ \{\mathrm{t} < 1\,\mathrm{day}\} \qquad K = \dfrac{10^{-6}}{t} \ \{1 < \mathrm{t} < 1000\,\mathrm{days}\}$

2. $K = \dfrac{10^{-6}}{t} \ \{\mathrm{t} < 1000\,\mathrm{days}\}$

3. $K = 10^{-4}\exp\left(-0.15\sqrt{t}\right) + 10^{-9}$

4. $K = \left[10^{-5}\exp(-0.07t) + 6\times10^{-9}\exp(-0.003t) + 10^{-9}\right]\times 10^{\pm 1}$

For all models, t has units of days; K has units of 1/m. These models were chosen for evaluation for the following reasons. The first model comes from NCRP 129 (NCRP, (1999)), which is used by early responders to emergency scenarios. The second is an adaptation (by this study) of the first (NCRP) model, incorporating the effect of time in the first day. The third and fourth models are expected to provide good results for aged sources. The third model is historical, from Anspaugh et al., (1975) and the fourth is a recent model from an expanded resuspension data set (Anspaugh et al., (2002)). This fourth model represents a vast review of data on aged sources and may be the best model for longer-term scenarios, but its effectiveness for very-short-time resuspension is not known. Model 4 includes a factor of 10 uncertainty.

These resuspension factors were converted to resuspension rates in the following way: air concentrations in resuspension scenarios typically follow a power law distribution with height (Anspaugh et al., (1975)):

$$\frac{\partial C}{\partial z} = -p\frac{C}{z}$$

This profile can be derived from the assumption of a steady-state balance between the upper flux of contaminant due to turbulent eddies and the downward settling flux:

$$V_d C = -K_z \frac{\partial C}{\partial z} = -\frac{u_* \kappa z}{\phi} \frac{\partial C}{\partial z}$$

where K-theory is used to represent the eddy diffusivity; κ is the von Karman constant. The parameter φ is the diabatic influence function, which may be computed from the Businger-Dyer relationships (Stull, (1999)). Generally, φ has a value of 1 for neutral conditions, falls to approximately 0.5 in unstable conditions, and increases above 1 for stable conditions. From these approximations:

$$p = \frac{V_d \varphi}{u_* \kappa}$$

Here V_d can be interpreted as a settling velocity or as a turbulent deposition velocity. Values for p were previously reported to fall typically between 0.25 and 0.35 (Anspaugh et al., (1975)). Given that a larger dataset from a variety of sites (Shinn, (2002)) shows a range from approximately 0.05 to 0.5 and a median value of 0.18, the present study used a value of 0.25. Note that the observed values of p lend evidence for the interpretation of V_d as a turbulent deposition velocity that exceeds the gravitational settling velocity. For example, 10 micron diameter particles of unit density have a settling velocity of approximately 3e-3 m/s. For a neutral surface layer, given a moderate friction velocity of 0.25 m/s, p would be only 0.03 if V_d were the settling velocity. To fit the observations, V_d must be larger than the gravitational settling velocity, such as might occur with turbulent deposition.

Making the assumption that the emission rate is equivalent to the turbulent contaminant flux, in the neutral boundary layer we have:

$$F = -u_* \kappa z \frac{\partial C}{\partial z} = u_* \kappa p C = u_* \kappa p K D_o$$

where D_o is the initial surface deposit. The resuspension rate is the normalized flux:

$$\Lambda = \frac{1}{D_o}\frac{\partial D}{\partial t} = \frac{1}{D_o} F = u_* \kappa p K$$

The model predictions from these and from the two empirical models are compared in Figure 2, below. Only model 2 and the empirical models capture the early-time behavior observed in the experiments. Models 1, 3, and 4 perform poorly.

A quantitative model comparison was performed using the uncertainty analysis code BOOT (Hanna et al., (1993)). Because of the wide range of values of interest, the models were compared logarithmically (natural logarithms of the models were compared to those of the observations and each other). Table 3 contains the result of that analysis. The metric *vg* is the geometric mean variance; *fa2* is the fraction of predictions with a factor of 2 of the observations, and *mg* is the geometric mean bias. The first row refers to the observations.

The empirical models provide the best fit to this data, which is not surprising considering that they were derived from the data. Of the other four models, model 2 has the best performance. The *fa2* from model 2 is almost as good as from emp3; its geometric mean is a little low and the bias is larger. Models 1 and 4 are so different from the observations that the geometric mean variance overloads BOOT; that from model 3 is not far behind. The factor of 10 uncertainty in model 4 is inadequate to address the difference seen between its results and the observations. These results demonstrate that the conventional models 1, 3, and 4 may be inappropriate for resuspension modeling at very short times (as noted, this data set extends to just beyond one day after deposition).

Model Testing

The models above were compared against two other data sets from the literature: Braaten et al., (1990) and Wu et al., (1992); these experiments are described in Table 4, below. Not all parameters were specified: some were estimated here, as explained below. The roughness heights were estimated at 10^{-6} m. The friction velocities for Wu et al., (1992) were estimated by assuming a semi-logarithmic profile. The ratio of the model predictions to the observed values is presented in Figure 3; only the results from the empirical models and model 2 described earlier are presented again here. All models underestimate the resuspension rate. However, in both the Braaten and Wu experiments, charge neutralizers were used on the surface before the resuspension experiments, which may have reduced the effective adhesive forces on the particles, thereby enhancing resuspension. This underestimation is quantified in the positive bias seen in Table 5, which also provides additional statistics from BOOT. As indicated by the metrics in Table 5, the empirical model emp1 performs the best.

In order to estimate the importance of the resuspension occurring within the first day, it of interest to compare the predicted fraction of material removed by these models. Models 1 and 2 are compared here, as two extremes of model performance. The fraction removed (FR) is equivalent to the time-integral of the resuspension rate:

$$FR = \frac{dD}{D_o} = \int_{t_1}^{t_2} \Lambda(t)dt = \kappa u_* p \int_{t_1}^{t_2} K(t)dt$$

The fraction removed should be dimensionless, so care must be taken in the specification of K. For model 1:

$$FR_1 = 10^{-6} u_* \kappa p (t_2 - t_1)$$

Because u_* has units of m/s, and K has units of 1/m, then t must be specified here in s. For model 2:

$$FR_2 = 0.0864 u_* \kappa p \ln\left(\frac{t_2}{t_1}\right)$$

Here, the constant 0.0864 has units of s/m, so FR is dimensionless. Because we cannot integrate 1/t from t = 0, consider t_1 = 1s. Then:

$$FR_1 = 0.0864 u_* \kappa p$$
$$FR_2 = 0.982 u_* \kappa p$$

FR_2 is more than a factor of 10 greater than FR_1. (Note that using t_1 = 60 s, instead, results in a factor of 7 difference between FR_1 and FR_2). If u_* = 0.5 m/s, then $FR_2 \approx 5\%$, which is consistent with Nicholson's observations (using a value of 0.25 for p, as above). Contrast these results with the expected fraction removed in the period from 1 day to 1 year (here the two models are equivalent in form):

$$FR = 0.51 u_* \kappa p$$

The fraction removed during the period from 1 day to 1 year, FR, is half of the material predicted to be removed in the first day, by model 2. In other words, this model predicts resuspension to fall so sharply with time that two-thirds of the total removal in the first year occurs in the first day. This result emphasizes the need for a robust model for short-time resuspension.

Discussion

The empirical model that includes all parameters, emp1, performed the best throughout the study. Although emp1 performed better than emp3 or model 2 against the Braaten et al., (1990) and Wu et al., (1992) data sets, all three models may have utility, depending on the availability of input parameter data for a system. The models emp3 and model 2 cannot take into account the effects of the surface and particle characteristics, but where these values are uncertain, emp3 and model 2 may be more appropriate.

The data sets considered here are really not adequate to test these models, as these data will not test the model performance beyond the first day. It should be noted that the observed effect of increasing resuspension rates with particle diameter, is expected to be valid only within the range of silt and smaller particles. Similarly, the effect of shielding for increasing roughness is likely to be true only on a local scale; use of a large roughness to represent, for example, a forest or urban environment would not be appropriate, as particles may quite easily resuspend from the top of leaf surfaces or buildings. Note also that it would be preferable to have a larger data set including more values of particle density.

Some elementary sensitivity testing is presented below. Table 6 includes, for each parameter, the multiplicative change in Λ that would result from a 10-fold increase in the parameter, for emp1. An order of magnitude error in u_*, for example, translates into two orders of magnitude in resuspension rate. This error in velocity is unlikely. Estimation of the roughness height is potentially difficult, given the great variability in outdoor surfaces. These results show that an order of magnitude error in z_0 translates only into a factor of two in the resuspension rate, well within the natural spread of data seen in even these careful wind tunnel studies.

Conclusion

This paper has reexamined historical data sets from wind tunnel studies applicable to resuspension at very short times (less than one day). The data demonstrate the importance of including the time dependence of resuspension in short-time modeling. Several models were evaluated. Those models from the literature that were derived from aged sources perform poorly against these data. Only three models perform reasonably well: the empirical models derived from the data and an adaptation of the NCRP 129 model. These models are recommended as the best (currently) available models for use in very-short-time predictions of resuspension, as would be appropriate for emergency responders. More data are needed to test and improve the models herein and to build a stronger physical foundation for the trends observed.

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Figures

Figure 1: NGG data: resuspension rates Λ plotted versus time

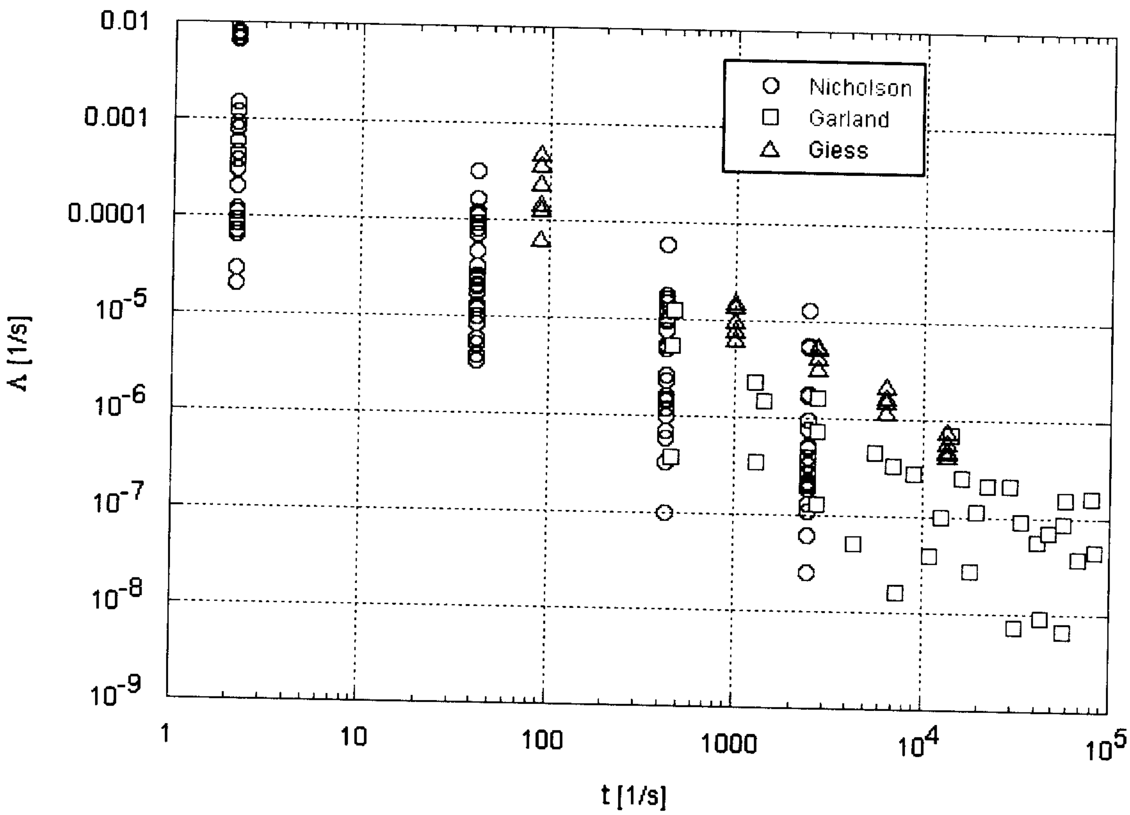

Figure 2: The ratio of predicted to observed Λ versus time for a suite of models

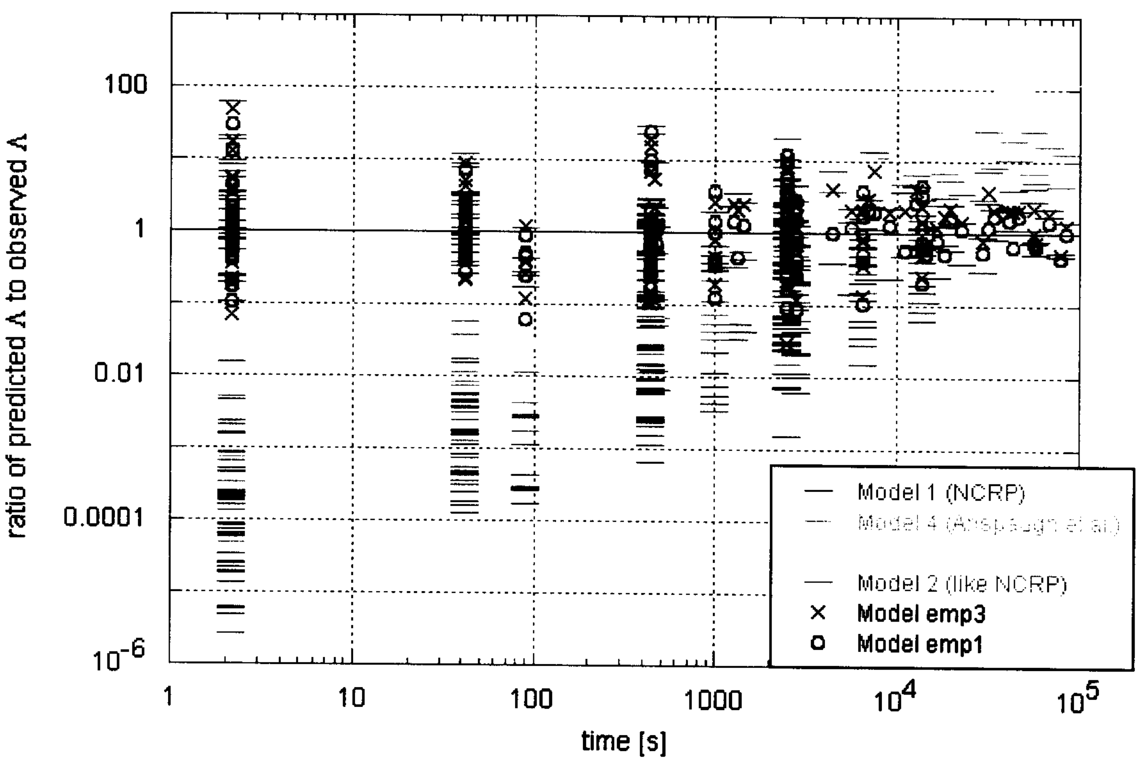

Figure 3: Performance of the models against the Wu and Braaten data

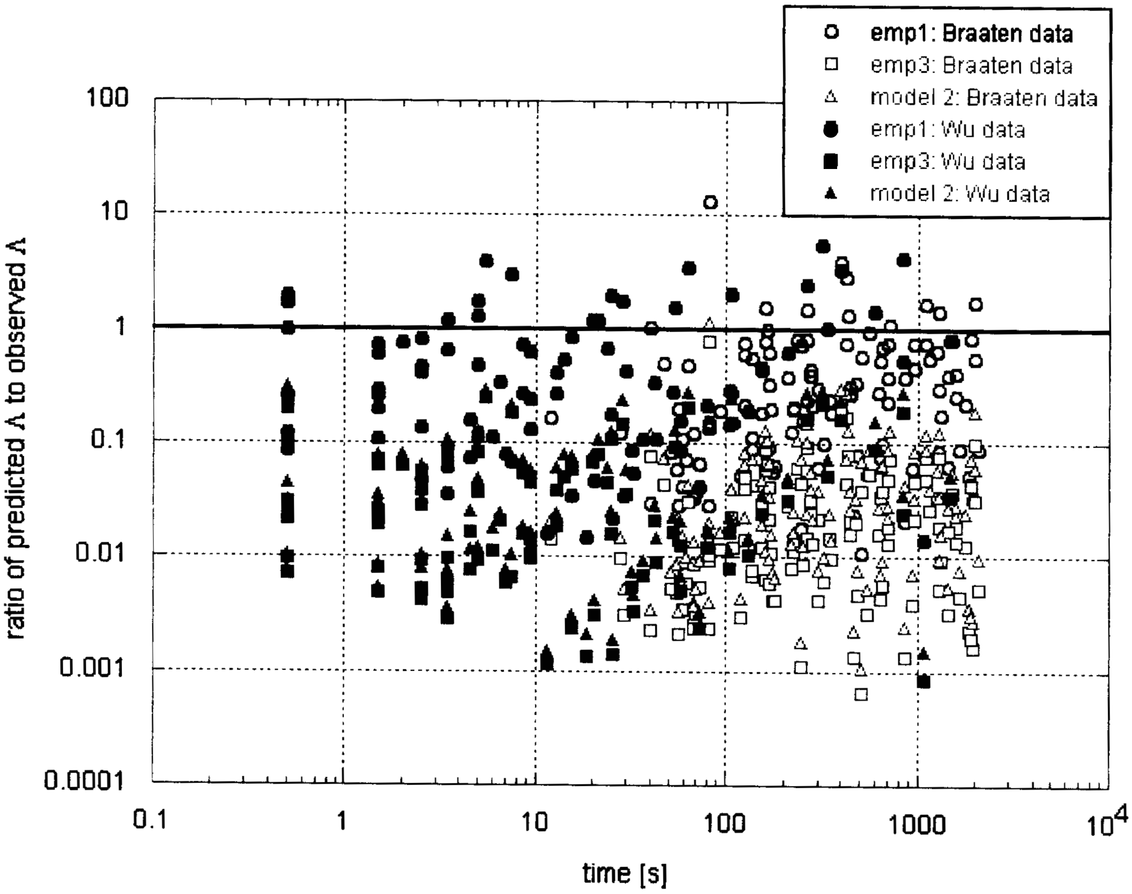

Table 1: Parameters of the NGG data sets

	Nicholson, (1993)	Giess et al., (1997)	Garland, (1982)
wind tunnel dimensions	1m × 1m	1.2m × 0.8m	1m × 1m
surface types	grass and concrete	grass	bare soil
u_*	0.1-0.4 m/s	0.3-1.4 m/s	0.2-0.8 m/s
time	2-2491 s	90-13590 s	540-108000 s
particle type	silica spheres	silica spheres	tungstic oxide
d_p	4-22 μm	1 & 10 μm	0.5 μm
ρ_p	1000 kg/m3	1000 kg/m3	7160 kg/m3
surface feature height	0.003 and 0.05 m	0.1 and 0.3 m	0.01 m
inferred roughness height	0.0003 and 0.005 m	0.01 and 0.03 m	0.001 m
resuspension rate data	computed from fraction removed	taken directly from Table 1	taken directly from Figure 3a
number of data points	96	30	33

Table 2: Goodness of fit metrics for the empirical models

		t-test t value				
	R^2	t	u_*	z_o	d_p	ρ_p
model emp1	0.89	-26.03	11.77	-3.83	1.56	-4.07
model emp3	0.85	-29.90	10.31	na	na	na

Table 3: Statistical comparison of models to the NGG data

model	mean	sigma	bias	vg	fa2	mg
obs	-1.24E+01	3.05E+00	0.00E+00	1.00	1.000	1.000
emp1	-1.24E+01	2.87E+00	1.14E-05	2.88	0.610	1.000
emp3	-1.24E+01	2.82E+00	-1.65E-04	3.95	0.428	1.000
model 1	-1.73E+01	7.44E-01	4.91E+00	**********	0.075	135.096
model 2	-1.21E+01	2.79E+00	-3.47E-01	4.88	0.409	0.706
model 3	-1.27E+01	7.32E-01	3.26E-01	26092.64	0.157	1.386
model 4	-1.50E+01	7.41E-01	2.61E+00	**********	0.138	13.572

Table 4: Parameters for data sets for testing

	Braaten et al., (1990)	Wu et al., (1992)
wind tunnel dimensions	1m × 1m	1m × 1m
surface types	glass	glass
u_*	0.17-0.26 m/s	estimated from 0.16 – 0.4
time	12-2093 s	1-1500 s
particle type	lycopodium spores	lycopodium spores
d_p	28 microns	28 microns
ρ_p	1000 kg/m3	1000 kg/m3
z_o (roughness height)	estimated at 1e-6	estimated at 1e-6
resuspension rate data	computed from fraction removed	computed from fraction removed
number of data points used	113	90
other details	used ^{210}Po charge neutralizer on surface	used ^{85}Kr charge neutralizer on surface

Table 5: BOOT output for model performance on the Braaten and Wu data sets

Braaten						
model	*mean*	*sigma*	*bias*	*vg*	*fa2*	*mg*
obs	-8.17E+00	1.88E+00	0.00E+00	1.00	1.000	1.000
emp1	-9.76E+00	1.10E+00	1.59E+00	59.48	0.289	4.927
emp3	-1.25E+01	1.21E+00	4.30E+00	**********	0.000	73.529
Model2	-1.19E+01	1.17E+00	3.77E+00	6708067.00	0.000	43.449
Wu						
model	*mean*	*sigma*	*bias*	*vg*	*fa2*	*mg*
obs	-5.83E+00	2.76E+00	0.00E+00	1.00	1.000	1.000
emp1	-7.00E+00	2.32E+00	1.17E+00	28.21	0.263	3.217
emp3	-9.69E+00	2.51E+00	3.86E+00	**********	0.009	47.506
Model2	-9.36E+00	2.42E+00	3.53E+00	1758143.00	0.009	34.285

Table 6: the sensitivity of emp 1 to a factor-of-10 change in a parameter value

parameter	10-factor
u_*	130
t	0.1
d_p	1.5
ρ_p	0.2
z_o	0.5