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# Dry deposition models for radionuclides dispersed in air: a new approach for deposition velocity evaluation schema

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**Abstract.** In the framework of a National Research Program funded by the Italian Minister of Economic Development, the Department of Energy, Information Engineering and Mathematical Models (DEIM) of Palermo University and ENEA Research Centre of Bologna, Italy are performing several research activities to study physical models and mathematical approaches aimed at investigating dry deposition mechanisms of radioactive pollutants. On the basis of such studies, a new approach to evaluate the dry deposition velocity for particles is proposed. Comparisons with some literature experimental data show that the proposed dry deposition scheme can capture the main phenomena involved in the dry deposition process successfully.

## 1. Introduction

As highlighted in ATMES (Atmospheric Transport Model Evaluation Study) report [1], the greatest number of uncertainties in numerical evaluations of pollutant transport and dispersion in air are introduced by the parameterizations both of the source term and of dry or wet deposition processes.

In the nuclear field, in case of severe accident that causes a release of radionuclides in the atmosphere, key challenges are the characterization of the specific type of release, the analysis of the dispersion and deposition phenomena and, consequently, the definition of appropriate corrective actions for the protection of the population in case of these extreme emergency conditions.

Recently, ENEA research center at Bologna, Italy, performed some research activities concerning "Studies in support to emergency preparedness" in case of accident in a nuclear power plant near the border of Italy, in collaboration with the Department of Energy, Information Engineering and Models Mathematicians (DEIM) of the University of Palermo. These activities included the study of physical-mathematical models able to properly simulate the dry deposition flux of radioactive pollutants.

Until now, due to limited understanding of some key aspects of dry deposition processes, there isn't a single accepted theoretical description of the involved phenomena because of the multiplicity and complexity of the fluid-dynamic processes which can influence the deposition rate.

Various experimental campaigns have been performed in international laboratories to evaluate the deposition velocity for different types of pollutants. Nevertheless, it is difficult to obtain a global generalization since the velocity values differ by four orders of magnitude for gases and three orders for particles [2]. These issues limit the possibility to study the dry deposition process with a single modelling



approach.

In literature there are different semi-empirical correlations characterized by well-defined criteria for application, and, in this field, parameterizations of the dry deposition velocity cover only some general conditions which may be of interest in nuclear applications. There are some models that can be applied only to one type of surface or canopy configuration, others that can work with different types of surface. The comparison between these two groups however highlights large differences between their predictions.

In this paper a new schema for parameterization of dry deposition velocity for particles is described. The work has involved comparisons with the experimental data obtained in different international laboratories. The results shows that the proposed approach allows to catch, with good agreement, some aspects of the phenomena involved in the dry deposition process for the examined environmental conditions.

## 2. Description of dry deposition processes

Dry deposition process refers to all phenomena of meteorological, chemical and biological nature that can influence a pollutant flux interacting with a ground surface, without involving the water into the atmosphere. The process is very complex and involves both gaseous pollutants and particulate of various granulometry and density, even if general phenomenological factors involving the two pollutant categories can be very different. The knowledge on particle dry deposition is far from being completely understood due to the complex dependences of deposition on particle size, density, terrain, vegetation, meteorological conditions and chemical species.

The main phenomena that are considered to influence the deposition process, are described as follows:

- transport due to atmospheric turbulence in the lower layer of the Planetary Boundary Layer (PBL) very near to the ground, called Surface Layer (SL). This process is independent of the physical and chemical nature of the pollutant and it depends only on the turbulence level;
- diffusion in the thin layer of air which overlooks the air-ground interface (named quasi-laminar sublayer resistance), where the dominant component becomes molecular diffusion for gasses, Brownian motion for particles and gravity for heavier particles;
- transfer to the ground that exhibits a pronounced dependence on surface type with which the pollutant interacts (i.e. urban context, grass, forest, etc.).

In the case of gas, containment of a pollutant by a surface depends on the surface chemical property, which absorbs, dissolves or involves pollutant in chemical reactions; in case of particles the efficiency of pollutant capture is connected to resuspension and redeposition phenomena that depend both on the surface type and wind velocity. Moreover, the deposition process changes quite a lot over the year, for example due to the seasonal variation of vegetation (with or without leaf) or over the day in connection with meteorological conditions (e.g. influence of temperature on leaf stoma).

## 3. Dry deposition models

A key concept to study the dry deposition process is the deposition velocity  $v_d$  [m/s] (i.e. deposition velocity at a given height  $z$ ) that links the pollutant vertical flux  $F_d$  [ $\text{g}/(\text{m}^2\text{s})$ ] to the concentration  $C(z)$  [ $\text{g}/\text{m}^3$ ] measured at quota  $z$  [m] above the ground reference level:

$$v_d = \frac{F_d}{C(z)} \quad (1)$$

Considering that the reciprocal of  $v_d$  is the overall resistance to mass transfer, the influence of the various phenomena on deposition velocity can be expressed in terms of an electrical analogy. Resistance to the deposition is configured as resistance in parallel and series combinations to describe transfer factor between air and surface.

For gaseous pollutant collection, it is possible to schematize the deposition process as resistances in series, so we can write the relationship:

$$v_d = (r_a + r_b + r_s)^{-1} \quad (2)$$

where  $r_a$  is the aerodynamic resistance considering the turbulence phenomenon in SL;  $r_b$  is the quasi-laminar sublayer resistance related to the diffusion phenomenon for gas and collisions due to the Brownian motion for particles; and  $r_s$  is the surface resistance, which depends on the nature of the receptor ground.

The evaluation of parameters  $r_a$  and  $r_b$  for gas pollution is reported in [3-7]. The calculation of gas surface resistance  $r_s$  is complicated and depends on the primary pathways for uptake such as diffusion through leaf stomata. A revised parameterization that includes a realistic treatment of cuticle and ground resistance in winter (low temperature and snow-covered surfaces) as well as the handling of seasonally-dependent input parameters is reported in [8].

For particle pollutant, in SL region the turbulence acts on particle motion exactly like on gas, however the process is influenced also by gravity. Into the quasi-laminar sublayer, the particle diffusion is influenced especially by Brownian motion. In fact, dusts are deposited also because of collisions with less dense but more numerous particles, such as gasses.

In this case the aerodynamic resistance  $r_a$ , the resistance to travel across the quasi-laminar surface layer  $r_b$  and the resistance to surface uptake  $r_s$  are considered in parallel to a second pathway: the gravitational settling defined as the reciprocal of settling velocity,  $v_s$  [9-11].

Seinfeld and Pandis in [11] derived a dry deposition flux relationship based on the assumption that  $r_s=0$ , and by equating the vertical fluxes in two layers over a surface to the total resistance.

The velocity  $v_d$  is obtained by using the equation reported below:

$$v_d = v_s + (r_a + r_b + r_a r_b v_s)^{-1} \quad (3)$$

where the product  $r_a r_b v_s$  represents a virtual resistance.

The settling velocity  $v_s$  grows in proportion to the square of particle diameter,  $d_p$ , according to the law of Stokes valid for particles with a diameter of up to 50  $\mu\text{m}$ :

$$v_s = \frac{d_p^2 g (\rho_p - \rho) C_c}{18\nu} \quad (4)$$

where  $g$  is the acceleration due to gravity,  $\rho_p$  the particle density,  $\rho$  the air density,  $\nu$  the air kinematic viscosity and  $C_c$  the correction factor evaluated as follows:

$$C_c = 1 + \frac{\lambda_{\text{air}}}{d_p} \left( 2,514 + 0,8 e^{-\frac{0,55 d_p}{\lambda_{\text{air}}}} \right) \quad (5)$$

with  $\lambda_{\text{air}}$  the mean free path of air.

### 3.1. Estimation of aerodynamic resistance

In [11-14] the parametrization of the aerodynamic resistance is as follows:

$$r_a = \frac{1}{k u_*} \left[ \ln \frac{z}{z_0} - \Psi_h \right] \quad (6)$$

with  $z_0$  the height above the displacement plane at which the mean wind becomes zero downward through the surface layer,  $u_*$  the shear velocity,  $k$  the von Karman constant (generally equal to 0.4).

Brandt et al. in [15] suggest the following relationship for calculating the parameter  $\Psi_h$ :

$$\Psi_h = -5 \frac{z}{L} \quad \text{with } \frac{z}{L} > 0 \quad (7)$$

$$\Psi_h = e^{\left\{ 0,598 + 0,390 \ln\left(-\frac{z}{L}\right) - 0,09 \left[ \ln\left(-\frac{z}{L}\right) \right]^2 \right\}} \quad \text{with } \frac{z}{L} < 0 \quad (8)$$

with  $L$  the Monin-Obukhov length that characterizes the stability of PBL layer in the lower part of atmosphere, computed as follows:

$$L = \frac{u_*^3 c_p \bar{T}}{kgH} \quad (9)$$

where  $c_p$  is the specific heat at constant pressure,  $\bar{T}$  is the average temperature in SL, and  $H$  is the sensible heat.

It is worth to note that Eq. (6) is a general expression that may be used for every pollutant and the parameter  $r_a$  depends only on mechanical and convective turbulence levels that are typical of the examined condition.

### 3.2. Estimation of the quasi-laminar sublayer resistance for particles

Once in the quasi-laminar sublayer, most particle dry deposition models treat surface uptake in terms of inertial impaction by inertial forces, interception, Brownian diffusion and gravitational settling.

The resistance to transfer across the quasi-laminar surface layer, that includes the effects of brownian motion (Schmidt number,  $Sc$ ) as well as the influence of inertial impaction (Stokes number,  $St$ ), can be evaluated by using the following relationship [9, 13, 16-18]

$$r_b = \frac{1}{(Sc^{-2/3} + 10^{-3}/St)u_*} \quad (10)$$

where  $Sc$  is defined as  $Sc=D/v$ , and  $St$  is evaluated as:

$$St = \frac{v_s u_*^2}{g v} \quad (11)$$

with  $v_s$  sedimentation velocity from Eq. (4)

## 4. Influence of captation processes on particle dry deposition parameterization

A bibliographical analysis of the determination of dry deposition velocities for vegetative canopy (experiment in situ) or derived by wind tunnel measurements demonstrates a substantial number of variations that are more pronounced for forest canopies or in the accumulation range. Comparisons of several models reveal that they differ from each other greatly and the largest uncertainty is for the accumulation mode particles (around 0.1–1.0 micron diameter range).

Although Brownian diffusion and gravitational settling are well-known to dominate small and large particle deposition, the dominant deposition mechanism for particles of intermediate  $d_p$  (in the range about of 0.1–1  $\mu\text{m}$ ) depends strongly on atmospheric conditions, surface characteristics and particle size.

Depending on the value of the roughness length  $z_0$ , the eddy turbulence effect is a major contribution to the total dry deposition velocity for particles in the size range from 0.01  $\mu\text{m}$  to approximately a few micrometers. Above this range, the dry deposition velocity is dominated by the inertial impaction term, and the Brownian diffusion and the eddy turbulence terms can be neglected.

Therefore the deposition of aerosol requires to describe Brownian diffusion phenomena and sedimentation process together with the following processes:

- Interception. This occurs when the particles, that follow the streamlines of the mean flow field, have small inertia. In the vicinity of an obstacle, they are held back because the distance between the particle center and the surface is smaller than half the particle diameter [19].
- Inertial impaction. If the particle inertia is too large, the particle, which is transported by the flow towards an obstacle, cannot follow the flow deviation in the vicinity of the obstacle. Thus it collides with the obstacle and remains on the surface.
- Turbulent impaction. By increasing the particle diameter, the inertial effect influences the deposition

process. However, in this mechanism, the particle has a sufficiently high velocity that turbulent eddies give some of them a transverse “free fight velocity”, so that some particles possess sufficient momentum to reach the wall [20-22]. When gravity is in the direction of the flow, the lift force enhances the particle deposition rate. For upward flow, however, the lift force moves the particle away from the wall and reduces the deposition rate [23].

- Rebound. It is thought to influence coarse particle deposition (size typically larger than 5  $\mu\text{m}$ ) [24]. From a mechanistic point of view, rebound is related to the kinetic energy of the incident particle, obtained from the particle velocity component normal to the deposit surface, and to the nature of the impact [25-27].

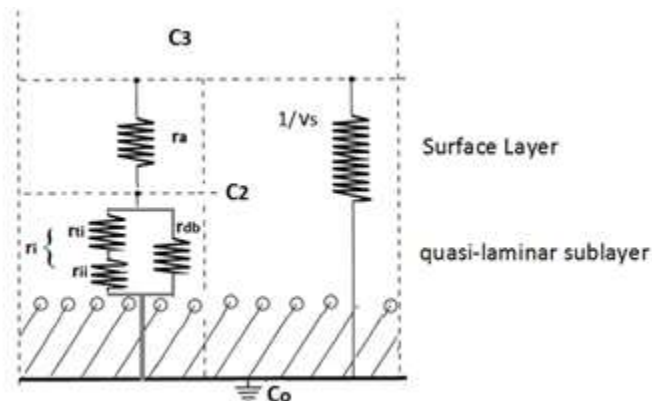
As above said, a review of the existing mechanistic models emphasizes the wide variety of ways that captation can take place, however, a comparison of two similar scenarios provides large discrepancies, generating values with different order of magnitude for the same particle diameter.

In the framework of the work performed to revise the various model as reported in literature, the comparison with the models from Slinn [16], Zhang et al. [28], and Wesely et al. [29] indicates that the Slinn model underestimates the deposition velocities by a factor of 40 to 100, while the Zhang and Wesely models better represent the experimental measurements, with the measurement/model differences generally lower than 30. In particular, the model of Zhang et al. [28] shows rather better agreement with the observations but tends to overestimate  $v_d$  for  $d_p$  under approximately 1.0  $\mu\text{m}$ .

#### 4.1. A modified approach for dry deposition as inferred from various parameterization schemes

To evaluate the effects of turbulent impaction processes on dry deposition phenomena of particles the following considerations can be performed. The new schematization for parametrization of particle deposition process, based on the electrical analogy, is depicted in Figure 1.

The resistance  $r_a$  is connected in series to two resistances in parallel, that is the resistance  $r_{bd}$  which represents the Brownian diffusion and the resistance  $r_i$  which allows to treat impaction processes. This last resistance is evaluated by considering two resistances in series: resistance  $r_{ii}$  that takes into account the inertial impact condition, and resistance  $r_{ti}$  that considers the effects resulting from turbulent impaction. This assumption allows to take into consideration effects on particle concentration coming from both the inertial and turbulent impaction (i.e. reciprocal influence of the two impact processes on dry deposition efficiency). It is worth to note that all the resistances regard processes which influence the flux over the canopy.



**Figure 1.** New schematization for parametrization of particle deposition process.

On the basis of the above described hypothesis, it is possible to write the following relationship:

$$v_d = v_s + (r_a + r_i)^{-1} \quad (12)$$

where the resistance  $r_a$  is calculated by using Eq.s (6)-(9) and the resistance  $r_i$  as follows:



$$\frac{1}{r_i} = \frac{1}{r_{bd}} + \frac{1}{r_{ii} + r_{ti}} \quad (13)$$

where  $r_{bd} = 1/(u_* Sc^{-2/3})$ ,  $r_{ii} = 1/(u_* 10^{-3/St})$ .

The transport of particles by Brownian diffusion represented as function of  $Sc^{-2/3}$  is recommended in [12, 25, 30, 31]. Moreover some Authors [9, 16, 32, 33] have parameterized the inertial impaction by using the term of  $10^{-3/St}$ . This was also confirmed by the observations conducted by Aluko and Noll [34] for large airborne particles (diameter greater than about 10  $\mu\text{m}$ ).

For the calculation of resistance  $r_{ii}$  in Eq. (13) the following considerations are taken into account.

As well known, empirical relations of turbulent deposition are typically presented in terms of the dimensionless particle relaxation time  $\tau_+$  [35] that, for a spherical particle, is defined as:

$$\tau_+ = \tau \frac{u_*^2}{\nu} \quad (14)$$

where  $\tau$  is the particle relaxation time that, for a spherical particle, is defined as:

$$\tau = \frac{d_p^2 \rho_p C_c}{18\mu} \quad (15)$$

where  $\mu$  is the dynamic viscosity of the air.

For turbulent deposition in pipe flows, the following regimes have been observed [32,33]:

- Regime with about  $\tau_+ < 0.1$  (very small particles); Brownian diffusion becomes significant and deposition is affected by a combination of Brownian and eddy diffusion. For the Brownian regime the deposition velocity is proportional to  $Sc^{-2/3}$ ;
- Regime with about  $0.1 < \tau_+ < 30$ ; particle motion is strongly dependent on turbulent fluctuation in the fluid flow and the deposition velocity increases with  $\tau_+$ . For larger particles the deposition velocity can be considered proportional to  $\tau_+$  squared.
- Regime with about  $\tau_+ > 30$ ; the particles have large inertia and the effect of turbulence on particle motion is significantly reduced; deposition velocity decreases with increasing particle size.

On the basis of these considerations, the parameter  $r_{ii}$  is assumed to be an exponential function of  $\tau_+$  as reported below:

$$r_{ti} = \frac{1}{u_*(a\tau_+^b)} \quad (16)$$

with  $a$  and  $b$  constants.

In Eq. (16) the constants  $m$  and  $n$  have been evaluated by fitting Eq.s (4), (12), (13) and (16) to the experimental data reported in [36, 37] where the Authors investigated the deposition of aerosol particles in a turbulent vertical pipe flow. The results reported in [36] show that the dimensionless deposition velocity increases linearly with the square of the dimensionless relaxation time for  $\tau_+ < 10$ . Furthermore, they found that the particle deposition velocity increases until about  $\tau_+ = 30$  and then slightly decreases beyond this value.

The fitting procedure provides for the constants  $a$  and  $b$  the values of 2 and 1, respectively.

## 5. Comparison with experimental data

The new scheme for computing the deposition velocity  $v_d$  is validated by comparison with experimental data reported in literature for several typology of soil and meteorological conditions. Some of the obtained results are shown in Figures 2-6.

In Figure 2, comparisons with the experimental data reported by Liu and Agarwal in [36], valid for grass surfaces, and Chamberlain in [37], for sticky grass surfaces, are reported. As expected, a good agreement is obtained, comparable in trend and values.

A further comparison has been carried out by using experimental measurements reported in [38], for several surface typologies. The obtained results are reported in Figures 3-6. It is worth to note that Authors in [38] reveal uncertainties on experimental data especially significant for  $d_p$  values less than  $10\ \mu\text{m}$ .

From the comparison we can say that in case of “sticky wood” surfaces (Figure 3), “sand” (Figure 4) and “plant” (Figure 5) prediction of  $v_d$  is good, both in trend and values, for  $u_*$  with high values, whereas it is underestimated for  $u_*$  with the lowest value within range, especially for “plant” surface (Figure 5).

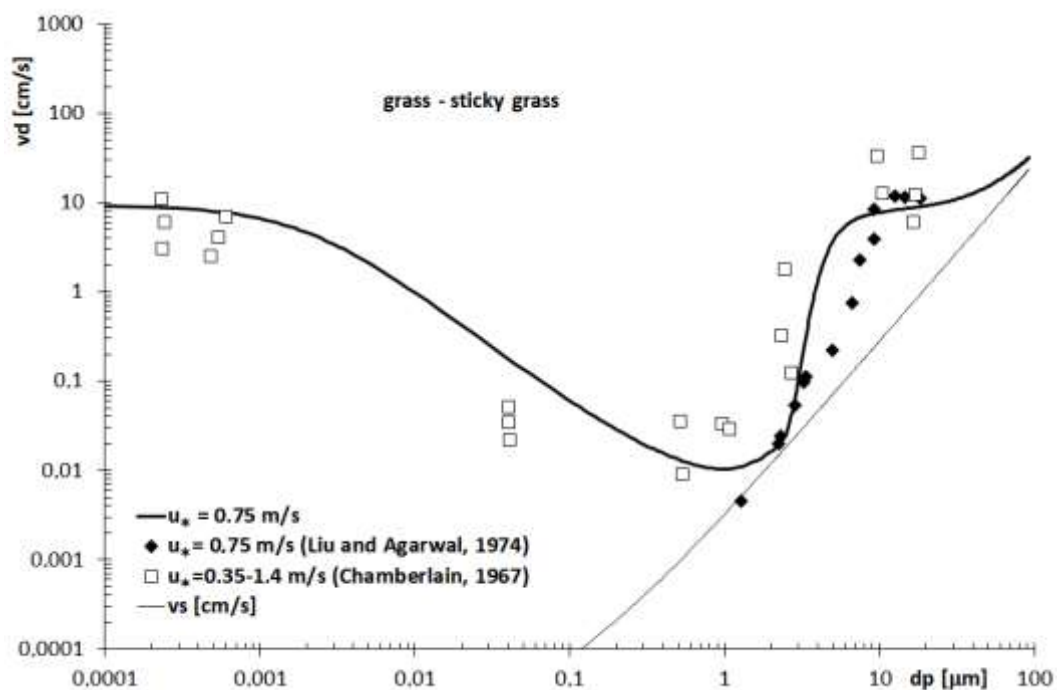
Note that the experimental data reported in [34, 39, 40] are also shown in Figures 3 and 5. These data refer to experimental activities performed in similar surface conditions and various range of velocity  $u_*$ .

The comparison between  $v_d$  prediction and the experimental data obtained by using very close values of  $u_*$  (i.e.  $u_* = 0.52\ \text{m/s}$ ) shows a good agreement.

By comparison of prediction of  $v_d$  with experimental data related to “water” surface (Figure 6), we can see that the model underestimates the experimental data for  $d_p$  below  $5\ \mu\text{m}$ . In this Figure the experimental data reported in [41] for water surface are also shown. It is worth to note that as a whole the prediction of  $v_d$  allows to capture the trend of the experimental data also in case of low values of  $d_p$ .

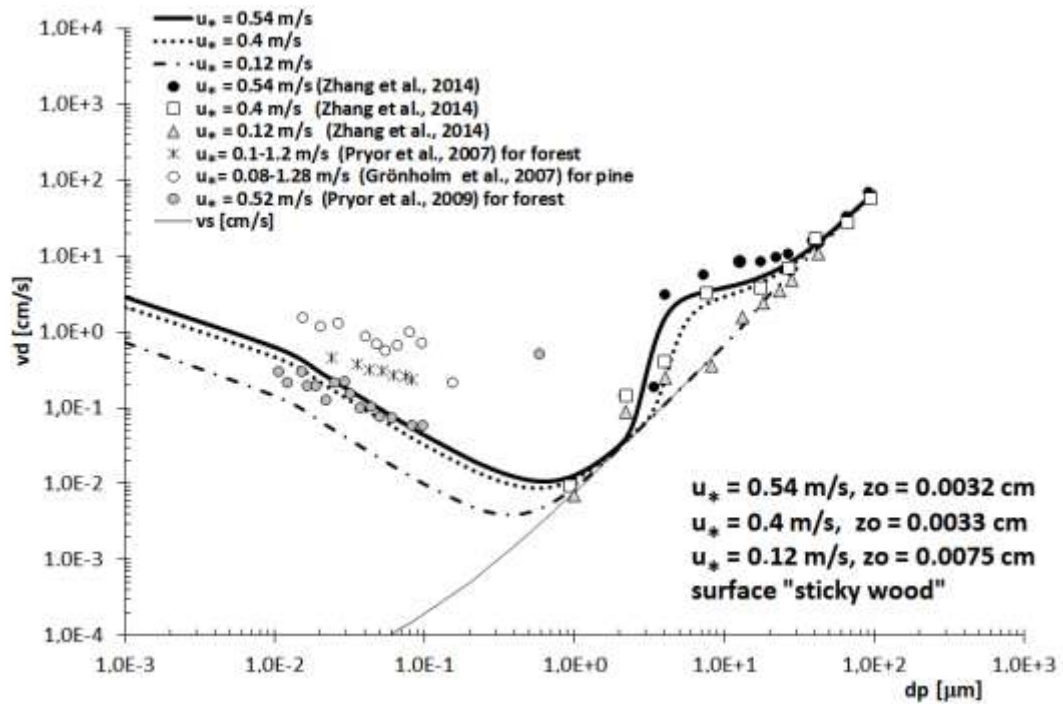
## 6. Conclusion

A scheme to evaluate particles deposition velocity is proposed. The validation work, carried out by using a significant number of experimental data collected from literature, shows that the proposed model reproduces with accuracy the data. It is worth to note that the correlation obtained with the new approach are able to catch the main phenomenology involved in the deposition process and partially to reduce those singularities that characterize the different models reported in literature that limit the applicability of the model to the entire canopy. However, further research activities are under-way to improve the model with the aim of take into account the rebound phenomenon that influences the coarse particle deposition.

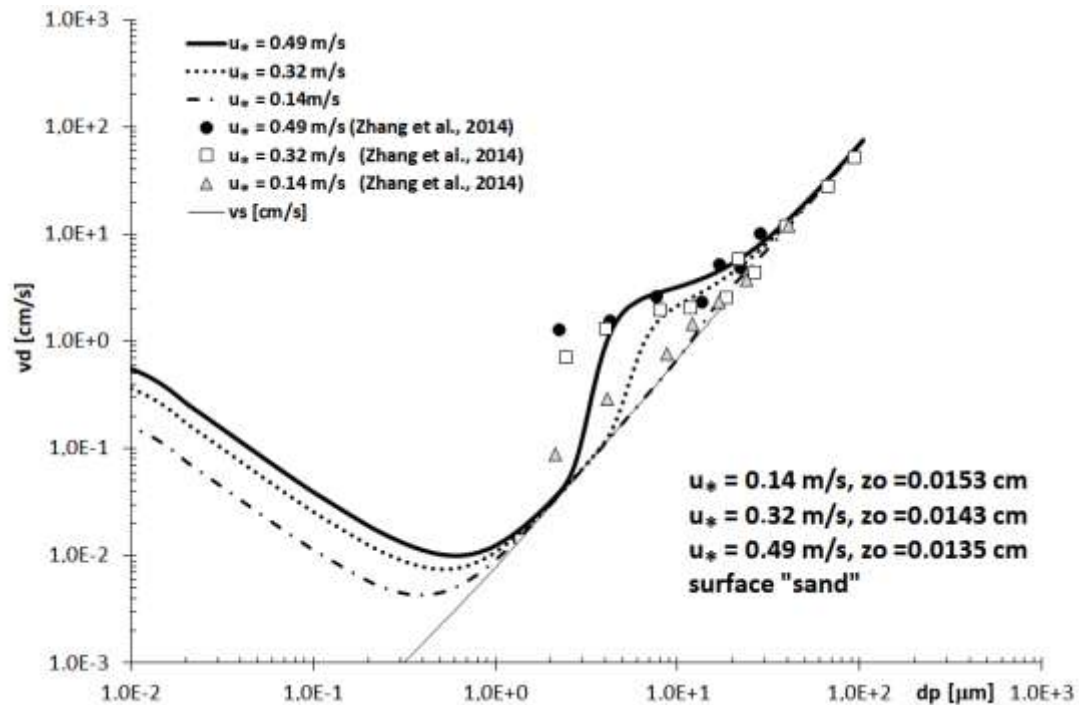


**Figure 2.** Comparison between the calculated deposition velocity and the experimental data reported in [36, 37] for grass surfaces and sticky artificial grass.

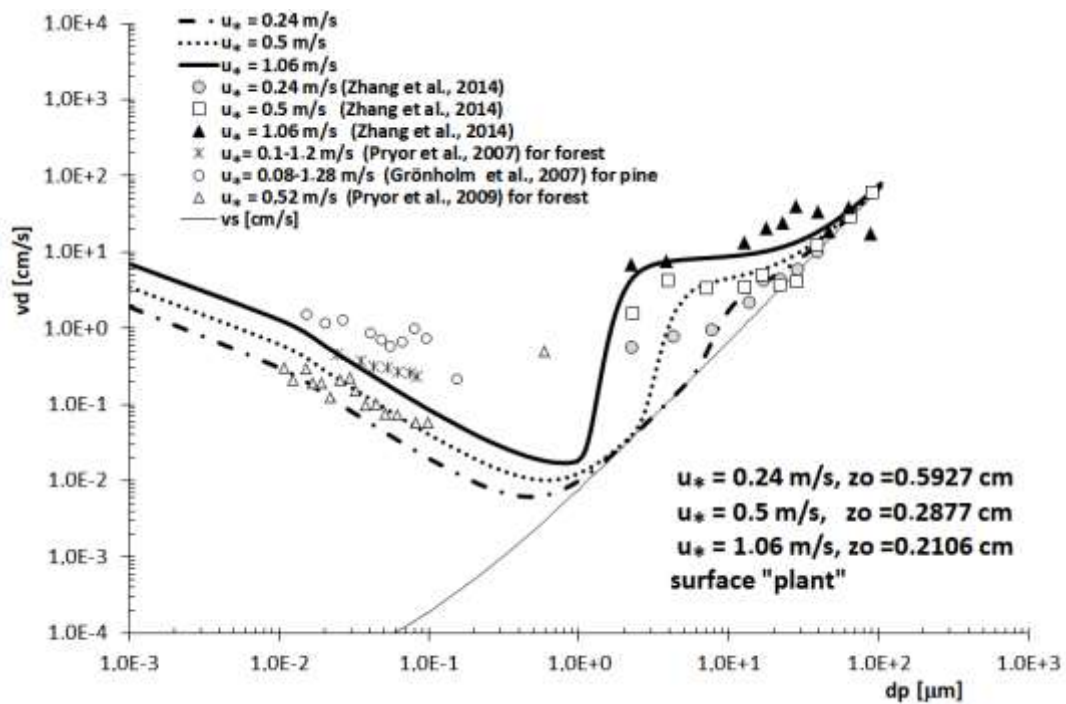




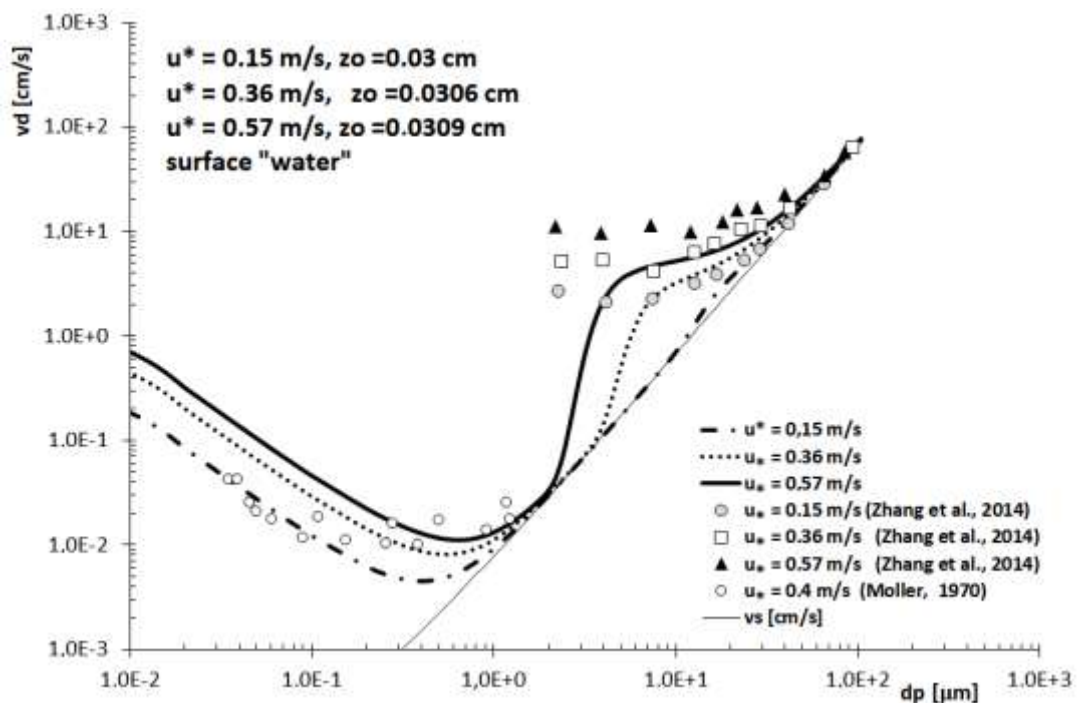
**Figure 3.** Comparison between the calculated deposition velocity and the measurements reported in [34, 38-40] for “sticky wood” surfaces, “forest” and “pine”.



**Figure 4.** Comparison between the calculated deposition velocity and the experimental data reported in [38] for “sand” surface.



**Figure 5.** Comparison between the calculated deposition velocity and the experimental data reported in [34, 38-40] for “plant” surface, “forest” and “pine”.



**Figure 6.** Comparison between the calculated deposition velocity and experimental data reported in [38, 41] for “water” surface.

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