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Report on model equations and comparison with generic models

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Summary

Within the European Horizon 2020 TRANSAT project, the WP3 entitled "Impact of tritiated products on environment and human health" deals with several topics, including one on the "Radioecology of tritiated products" (Topic 2). Among the various removal processes of pollutants in the atmosphere, the dry deposition process is recognized as an important pathway. In this field, there are several models reported in the literature useful to predict the dry deposition velocity of particles of different diameters but many of them are not capable of representing dry deposition phenomena for several categories of pollutants and deposition surfaces. Moreover, their application is valid for specific conditions and if the data in that application meet all of the assumptions required of the data used to define the model. In this paper, to overcome the above issues, the dry deposition velocity generic modelling approach proposed by Giardina and Buffa (2018) based on an electrical analogy schema is used and adapted to efficiently address the deposition of radioactive pollutant in particulate aerosol form onto grass and vegetable surfaces. The dry deposition velocity is calculated as a function of the most relevant driving factors, including the particle characteristics (size and density), the meteorological conditions and the surface features of the environmental receiving cover. Comparisons with extensive published measurements and other empirical models based on field data show that combining the mechanistic aspects of the approach of Giardina and Buffa (2018) along with existing empirical parametrizations of impaction efficiency allows to predict reasonable deposition velocities for a wide particle size range over grass and vegetable surfaces. The parametrizations suggested in this report - although not exhaustive of a given phenomenon (here impaction) - has allowed to lay the groundwork to configure a deposition rate model for tritiated particles of (sub)micron size on grass and vegetable surfaces. This work is a prerequisite to formalize in a simple model the processes of foliar uptake and internalization in plant biomass - which occur after deposition - of (sub)micronic (possibly tritiated) particles.

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Abbreviations

C_c	Cunningham slip correction factor (-)
D	Particle's Brownian diffusivity ($m^2.s^{-1}$)
d_p	Particle diameter (m)
g	Gravity acceleration ($m.s^{-2}$)
HTO	Tritiated water (in vapour/gaseous or liquid form)
K	Von Karman constant
K_B	Boltzmann constant
Kp	Particle eddy diffusivity ($m^2.s^{-1}$)
L	Obhukov length (m)
m,n	Non-dimensional numbers (-)
OBT	Organic Bound Tritium
QLS	Quasi-Laminar Sublayer (-)
r	Total resistance ($s.m^{-1}$)
r_a	Aerodynamic resistance ($s.m^{-1}$)
r_{db}	Brownian diffusion resistance ($s.m^{-1}$)
r_{ii}	Inertial impact resistance ($s.m^{-1}$)
r_{ql}	Quasi-laminar sublayer resistance ($s.m^{-1}$)
r_{ti}	Turbulent impact resistance ($s.m^{-1}$)
Sc	Schmidt number (-)
SL	Surface Layer (-)
St	Stokes number (-)
T	Temperature (K)
TFWT	Tissue-free-water-tritium
u*	Friction velocity ($m.s^{-1}$)
U	Horizontal mean flow velocity ($m.s^{-1}$)
v_d	Deposition velocity ($m.s^{-1}$)
V_s	Settling velocity ($m.s^{-1}$)
WP	Work Package
z_0	Roughness length (m)
τ+	Non-dimensional particle relaxation time (-)
τ	particle relaxation time (s)
ρ_a	Density of air ($kg.m^{-3}$)
ρ_p	Density of particle ($kg.m^{-3}$)
μ_a	air dynamic viscosity ($kg.m^{-1}.s^{-1}$)
ν_a	air kinematic viscosity ($m^2.s^{-1}$)
λ_a	Mean free path of air (m)

Summary

Within the European Horizon 2020 TRANSAT project (TRANSversal Actions for Tritium; http://transat-h2020.eu/), the WP3 entitled "Impact of tritiated products on environment and human health" deals with several topics, including one on the "Radioecology of tritiated products" (Topic 2). Among the various removal processes of pollutants in the atmosphere, the dry deposition process is recognized as an important pathway. In this field, there are several models reported in the literature useful to predict the dry deposition velocity of particles of different diameters but many of them are not capable of representing dry deposition phenomena for several categories of pollutants and deposition surfaces. In this paper, to overcome the above issues, the dry deposition velocity generic modelling approach proposed by Giardina and Buffa (2018) based on an electrical analogy schema is used and adapted to efficiently address the deposition of radioactive pollutant in particulate aerosol form onto grass and vegetable surfaces. According to this approach, the dry deposition velocity is calculated as a function of the most relevant driving factors, including the particle characteristics (size and density), the meteorological conditions and the surface features of the environmental receiving cover. Comparisons with extensive published measurements and other empirical models based on field data show that combining the mechanistic aspects of the approach of Giardina and Buffa (2018) along with existing empirical parametrizations of impaction efficiency allows to predict reasonable deposition velocities for a wide particle size range over grass and vegetable surfaces. The parametrizations suggested in this report - although not exhaustive of a given phenomenon (here impaction) - has allowed to lay the groundwork to configure a deposition rate model for tritiated particles of (sub)micron size on grass and vegetable surfaces. This work is a prerequisite to formalize in a simple model the processes of foliar uptake and internalization in plant biomass - which occur after deposition - of (sub)micronic (possibly tritiated) particles.

1 Context and objectives

Nuclear facilities are likely to discharge different types of radionuclides into the atmosphere, either as gaseous and aerosol species in situations of chronic or accidental releases. Among those forms, tritiated particles were yet locally observed in atmospheric aerosols (Maro et al., 2014a, 2014b, 2015) leading to a different behavior of tritium in ecosystems than tritiated water (HTO) or tritium bound to organic compounds (OBT). These airborne tritiated particles may also originate from other anthropic sources including past uses of tritium, e.g. in watch-making or tritium use for emergency lighting.

In the future, tritiated particles might be released within the ITER international project. Among refractory materials, tungsten has been selected as the main suitable plasma facing material in tokamaks and future nuclear fusion reactors. When the ITER project attains its operational mode, the future fusion reactor could thereby generate tritiated tungsten dust-like particles.

Anthropic releases of tritiated particles might also increase in the future due to dismantling activities. Indeed, during the decommissioning of nuclear facilities, operations are intended to remove or eliminate any tritiated material. These operations generate fine airborne dust of tritiated aerosols.

Following their release into the atmosphere and dispersion, tritiated aerosols - as other radioactive aerosols - are subjected to various processes such as radioactive decay, atmospheric deposition in wet or dry conditions on soil and vegetation, possible re-emission from any contaminated surfaces, including natural covers, if not, transfer and integration into living organisms. Internal exposure to humans occurs from inhalation of atmospheric radioactive particles – in particular those that are very small and do not dissolve easily - and the use of contaminated plants as food or as feed for domestic animals.

In recent years, an increasing interest has been directed to the application of particles to ecological terrestrial species (e.g., plants) for agronomic purposes, by investigating several processes such as foliar uptake (uptake through leaves) of atmospheric aerosols by plants, bioaccumulation, and risks of (sub)micronic or nano-particles for plants (Rauret et al., 1995; Birbaum et al., 2010; Larue, 2011; Remédios et al., 2012; Wang et al., 2013). However, the assimilation of airborne particles by plants and their transfer along the food chain have so far been poorly documented and have not been taken into account in the models. In particular, knowledge about their interaction with vegetation is still required - foremost their uptake by guard cells in the epidermis of leaves regulating the opening of stomata - to understand how nano- or microscale materials can affect food chains and, ultimately, human health. Upstream of the processes mentioned above, understanding the elementary mechanisms governing deposition of tritiated aerosols onto vegetation is first needed to propose a fit-for-purpose modelling approach for deposition velocities, taking into account, if relevant, the particle characteristics, the meteorological conditions and the surface features of the environmental receiving cover.

In the framework of WP3 entitled “Impact of tritiated products on environment and human health” of the TRANSAT Horizon 2020 project, investigations are proposed to improve knowledge in the field of radiobiology, dosimetry, radiotoxicology, genotoxicology, ecotoxicology and environmental fate in case of contamination by tritiated products. In particular, a specific task of this WP (Task 3.2: Radioecology of tritiated products) aims to study the consequences of an accidental release of such tritiated particles in terms of radioecology and ecotoxicology. The very first action of this task is the assessment of the dry deposition velocity of tritium (and subsequent incorporation processes) in particulate aerosol form within plants, with a particular focus on grass and vegetable surfaces.

In this context and on the basis of the existing modelling approaches and measurements obtained for grass and for any other plants representative of natural and/or agricultural covers, the first and main objective of this project is to use, implement, (re)parametrize and test an existing model of deposition of (sub)micronic particles onto grass and vegetable surfaces along with published measurements and other empirical models. The second objective, in a longer run, will be to formalize in a simple model subsequent transfer and incorporation processes (i.e. foliar uptake and metabolism) - that occur after deposition - of (possibly tritiated) particulate aerosols in the vegetation.

As a prerequisite to the present modelling report, a short review on the main transfer processes of radionuclides in a particulate form is given in the next section. Literature review is based, either specific to tritiated particles, if existing, or to micrometric particles.

2 Main transfer processes of radionuclides to plants

Following their release into the atmosphere, airborne radionuclides, either as gaseous or in aerosol/particle forms, may be submitted to atmospheric dispersion, dry and wet deposition (Figure 1). Wet deposition is due to the entrainment of radionuclides towards the surfaces by precipitation and specifically wind-driven rain; therefore wet deposition takes place during rain events and dry deposition all the rest of the time. A part of the radionuclides retained by leaves through foliar transfer may be decreased by washout, due to rainwater or irrigation (Rauret et al., 1995; Madoz-Escande et al., 2005). Another fraction may be absorbed through the epidermis of aerial parts and then be reallocated by translocation (and further remobilization). Finally, the fraction present in soils may be incorporated by the plant through the plant roots (Figure 1).

In this report, we solely focus on the process of dry deposition. More specifically, the existence of knowledge and/or specific experimental data for radionuclides in particulate form is reviewed for this process. Alternatively, the question of whether or not we can consider tritium-specific parameterization is raised, as well as whether existing options are sufficient.

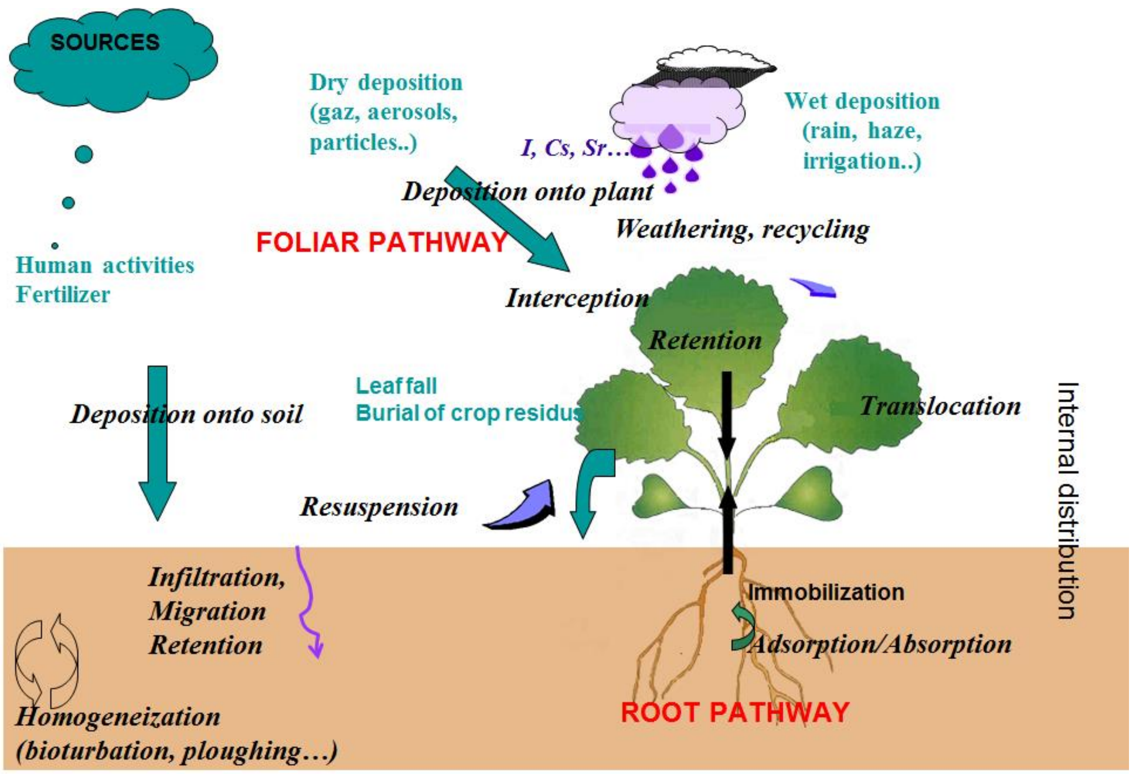

Figure 1 : Transfer processes of radionuclides to plants through foliar and root pathways

3 Dry deposition process of airborne contaminants

3.1 Description, complexity and uncertainty

The dry deposition process is recognized as an important pathway among the various removal processes of pollutants in the atmosphere. It refers to all phenomena of meteorological, chemical and biological nature that influence a flux of particles (and gas) interacting with a ground surface without involving water in the atmosphere (i.e. in the absence of precipitation). Since it controls the transfer of pollutants from the atmosphere to the surface, the study of the dry deposition process is a major issue concerning the impact of pollutants on the population and the environment. If event of a severe accident and the release of radionuclides in the atmosphere occur, it may be useful for defining effective mitigation measures and actions to protect the population.

The main phenomena that are considered to influence the dry deposition process are described as follows (see more details and definitions in Giardina and Buffa, 2018) :
• Transport due to **atmospheric turbulence** in the lower layer of the Planetary Boundary Layer (PBL) in the close vicinity of the ground, called Surface Layer (SL). This process is independent of the physical and chemical nature of the radionuclides 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, QLS), where the dominant component becomes molecular diffusion for gases, **Brownian motion** for particles and **gravity** for heavier particles (e.g. particle diameter higher than 100 µm);
• Transfer to the ground that exhibits a pronounced dependence on surface type with which the radionuclide interacts (i.e. urban context, grass, forest, etc.) though **interception** and **impaction**

Many scientific articles have reviewed the state of knowledge regarding dry deposition during the last 30-40 years. Despite of this, understanding key aspects of dry deposition process is far from complete due to (1) the multiplicity and complexity of the fluid-dynamic processes (atmospheric transport and diffusion, transfer to the ground, etc.) that influence the deposition flux, (2) the complex dependence of deposition on many variables including radionuclide size, density, landscape heterogeneity, vegetation, meteorological conditions and chemical species, (3) the lack of a complete experimental set of data covering all scenarios of interest that limits the understanding of certain key aspects occurring in the process (Giardina et al., 2017; Giardina and Buffa, 2018), (4) highly time-dynamics of dry deposition 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 light and temperature on leaf stomata opening). As a consequence, there is no single accepted theoretical description of the dry deposition phenomena.
The next sections present a brief update of dry deposition reviews with a sole focus on particles, with respect to modelling and experimental aspects.

3.2 Experimental campaigns

Various experimental campaigns, performed in different international laboratories, allowed the evaluation of the dry deposition velocities for particles and various vegetation surfaces (forest canopies, grassland,…), resulting in a large database of experimental results, obtained under different conditions (aerosol characteristics, meteorological situations) with several techniques (see the reviews of Sehmel, 1980; Zhang and Vet, 2006; Pryor et al., 2007, Tab.1; Guha, 2008). It contains data from about 40 years of theoretical work on dry deposition, field and wind-tunnel experiments, such as those carried out on forest (Pryor et al., 2007; Petroff et al., 2008a) or grassland ecosystems (Pröhl, 1990, cited by Thiessen et al. 1999; Pellerin, 2017), or other vegetables (e.g. leaf vegetables: Tschiersch et al., 2009). Nevertheless, since information describing the experimental conditions is not always documented, a comparison of different campaign techniques and results remains difficult (Petroff et al., 2008a). In addition, there is a difficulty in generalizing the particle dry deposition phenomenon because the velocity values evaluated by many particle flux studies conducted during the past four decades differ by **three orders of magnitude** (Sehmel, 1980; Thiessen et al., 1999; Pryor et al., 2007; Guha, 2008; Petroff et al., 2008a; Pellerin, 2017; Giardina et al., 2017; Giardina and Buffa, 2018). IRSN studies have also performed measurements of dry (and wet) deposition of aerosols onto different covers (maize, grass, bare soil, forest…) as a function of driving variables including particle size distribution (Petroff et al., 2008b) and micrometeorological/turbulent parameters such as wind friction velocity and sensible heat flux for dry deposition (Damay et al. 2009; Damay, 2010; Maro et al. 2006) (and rain intensity for wet deposition; Maro, 2011). A recent study (Pellerin, 2017) allowed the quantification of dry deposition velocity above grassland according to the particles diameter – particularly for particles under 10 nm - and micrometeorological features of the atmospheric stability.

4 Dry deposition models of airborne contaminants

4.1 Short review of the existing dry deposition models for particles

Numerous dry deposition models in the literature based on the estimation of gaseous dry deposition velocities have been developed worldwide in air quality and climate modelling or other specific applications (Wesely and Hicks, 2000; Zhang et al. 2003; Hirabayashi et al., 2012; Giardina and Buffa, 2019). The concept of deposition velocity v_d (i.e. the deposition velocity at a given height z), that has been used extensively in modelling the dry deposition process for gas (Pryor et al., 2007), can also be applied for particles: v_d (m s^{-1}) can link the gas/particle vertical flux *F* (g m^{-2} s^{-1}) to the atmospheric concentration *c(z)* (g.m^{-3}) of gas/particles measured at height z (m) to the ground reference level as follows:

$$v_d = \frac{F}{c(z)} \tag{1}$$

4.1.1 Influence of particle size (diameter)

The particle-size spectrum is influenced by the release type (Thiessen et al., 1999). Planned releases are usually filtered, and the cut-off point of the filter device determines the maximum particle size. Unplanned releases may be unfiltered, and the whole size spectrum may be released. However, with increasing distance from the release point, the size spectrum approaches that of **atmospheric background aerosols**, i.e. in the range of approximately 0,1 to 1 µm. Particles below 0,1 µm coagulate or attach to larger particles, whereas particles larger than about 1 µm are lost due to **sedimentation**. The deposition velocity typically decreases with increasing distance from the release point, since the ratio of small-to-large particle concentrations increases due to the effective deposition of large particles (Thiessen et al., 1999; Pröhl, 2003).

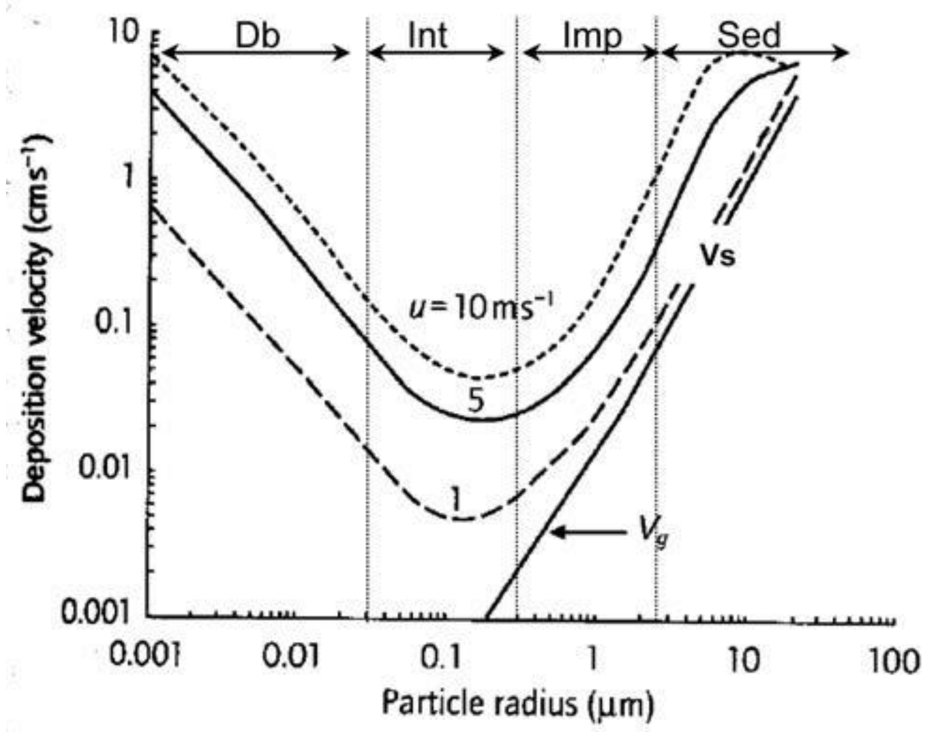

Figure 2 : Variation of the particle dry deposition velocity to vegetation as a function of particle size (radius) according to the theorical approach proposed by Slinn (1982) (Db:brownian diffusion, Int: Interception, Imp: impaction, Sed: sedimentation, vs: gravitational/settling velocity, u=wind speed).

The dry deposition of ultrafine particles - in the size range from 0,01 µm to approximately a few micrometers - is essentially caused by **Brownian diffusion** (Pröhl, 2003; Giardina et al., 2017). This process are assumed to dominate the diffusion processes in the quasi-laminar sublayer surface (QLS). With regard to particles with a diameter of less than 0.2 µm, as and when the diameters increase, the impact of the Brownian diffusion is increasingly low, but at the same time, the deposition caused by interception and impaction phenomena increase. A minimum deposition velocity is observed for particle of this diameter size (Figure 2). Beyond 0.5 µm and up to 1.2 µm, the influence of Brownian diffusion is negligible, the increase is caused by the sharp escalation in the influence of **interception** and **impaction**. At diameters >1 µm, deposition increases with increasing particle size, because the effects of **impaction** and **gravitaional settling** and become more important, and the Brownian diffusion and the eddy turbulence can be neglected (Petroff et al., 2008a; Giardina et al., 2017). All these processes are detailed by Giardina et al. (2017). As a consequence, a second pathway for particles is integrated - **gravitational settling**, which is considered to be in parallel to the resistances r_a, r_b and r_s (see Eq(2) and Figure 3 below), and can be defined as the reciprocal of the settling velocity (Giardina et al., 2017; Giardina and Buffa, 2018).

4.1.2 Influence of plant canopy structure

For particles (as well as for gases), the deposition velocity is also influenced by the **structure of plant canopy**. In general, deposition is more effective for well-developed canopies, since the area of interface between vegetation and atmosphere is increased (Thiessen et al., 1999). Although this effect is well known, it is considered in only a few assessment models. In models for routine releases, it can be avoided by the choice of appropriate long-term mean values. However, the problem is more serious for model applications to single releases, due to the pronounced seasonality of the standing biomass.

When parameterizing the deposition velocity to vegetation and other surfaces, existing approaches for **gaseous** radionuclide collection mostly utilize the **multiple resistance analogy** approach represented as three resistances in series (cf. Figure 3).

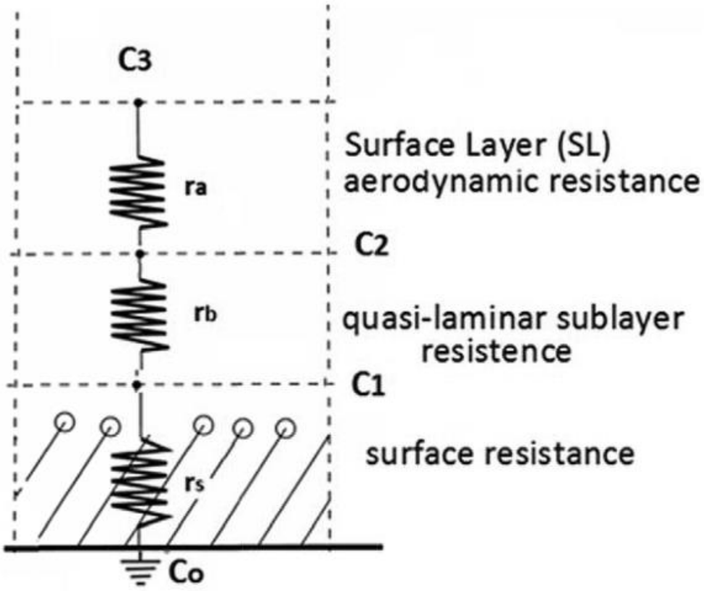

Figure 3 : Electrical analogy for the dry deposition of gaseous pollutants (from Seinfeld and Pendis, 1998; Giardina and Buffa, 2018).

Several theoretical frameworks reported in the literature aim to predict the dry deposition velocity of particles of different diameters to vegetation (as shown in Figure 2). However, many of them are not capable of representing dry deposition phenomena for several categories of pollutants and deposition surfaces. Relying on some semi-empirical correlations, their applications are valid for specific conditions and only if the data in that application meet all of the assumptions required of the data used to define the model. There are some models that can be applied only to a **unique type of surface** or canopy configuration, others that can work with **different types of surface**. In the first category, strong physical simplifications made in operational models based on past experiments often conducted in simplified configurations resulting in **significant discrepancies** between measurement results and model predictions of particles dry deposition velocity to vegetation (Slinn, 1982; Zhang et al. 2001; Pryor et al., 2007; Petroff, 2008a; Damay, 2010; Maro, 2011). In the second category, models seek to explain the deposition process to complex surfaces by taking into account variables such as particle granulometry (size distribution), surface morphology and meteorology. However, there are still substantial and systematic discrepancies between these process-based models of particle dry deposition and micro-meteorologically derived observations made over high roughness vegetated surfaces such as forests, particularly for submicron diameter particles (Thiessen et al. 1999; Pryor et al., 2007; Giardina et al. 2017). Dry deposition of this size range of particles is influenced by three main driving variables, including **particle diameter** (cf.Figure 2), **friction velocity** and/or **surface roughness**, and **stability** (Pryor et al., 2007; Maro et al. 2014a; Pellerin, 2017).

In summary, it has been emphasized the wide variety of ways for dry deposition formalization, leading to large differences between the various model predictions (Giardina et al., 2017) **particularly for particles in the submicronic range** (Petroff et al., 2008a; Damay, 2010; Pellerin, 2017). For this size range of particles, uncertainties on the dry deposition velocity values are up to several orders of magnitude discrepancies according to the model used (Petroff et al., 2008a). **These issues**, in addition to the ones mentioned in section 3.1, **limit the possibility of studying the dry deposition process of particles using a single modeling approach**.

4.2 A modified approach for dry deposition as inferred from various parameterization schemes

In this context, the generic approach that has been proposed recently for modelling dry deposition velocity by (Giardina et al., 2017; Giardina and Buffa, 2018) allows to overcome the above-mentioned issues by addressing different environmental conditions and deposition surfaces for several particles. In this approach, based also on an electrical analogy (Figure 4) as for gaseous pollutants (Figure 3), particles exchange at the air-vegetation interface is viewed as occurring as a sequence where the particle is first vertically transported towards the SL via **atmospheric turbulent** movements of air then diffuses across the quasi-laminar sublayer (QLS) of air which overlooks the air-ground interface largely by **Brownian motion** for particles (and **gravity** for heavier particles) and is finally transported to the ground, which exhibits a strong dependence on the surface type with which the pollutant interacts (i.e. urban context, grass, forest, etc.). In the case of particles, the efficiency of capture is connected to **resuspension** and **redeposition** phenomena that depend both on the surface type and wind velocity.

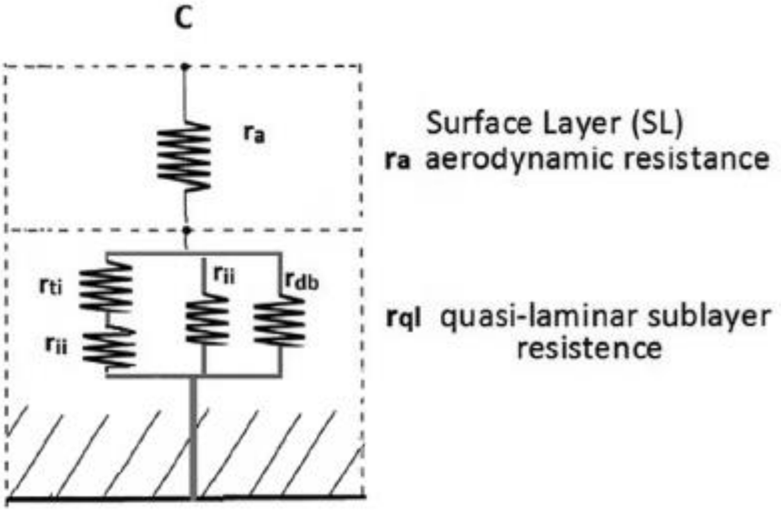

Figure 4 : Schematization for the parametrization of the deposition velocity for particles, as proposed by Giardina and Buffa (2018).

By considering that the reciprocal of v_d is the overall resistance to the mass transfer, the influence of the various phenomena on the deposition velocity of atmospheric particles can be conceptualized in terms of the electrical analogy for gas (cf. Figure 3) as three resistances in series (Pryor et al. 2007), in parallel to the gravitational settling pathway for particles (Giardina and Buffa, 2018), as follows:

$$v_d(z) = \frac{1}{r_a(z) + r_b + r_s} + v_s \qquad (2)$$

Where: r_a is the **aerodynamic resistance** considering the turbulence phenomenon in SL; r_b is the **quasi-laminar sublayer resistance** related to collisions due to the Brownian motion for particles (and diffusion phenomenon for gas); and r_s is the **surface resistance** (or canopy resistance), which depends on the nature of the receptor ground. Unlike for gases, r_s is often assumed to be equal to zero for particles, but the **settling velocity v_s** needs to be taken into account (see below).

However, as highlighted by Venkatram and Pleim (1999), the electrical analogy described above is imperfect in the context of particle dry deposition modelling because it is inconsistent with mass conservation equation (Pryor et al. 2007; Giardina and Buffa, 2018). Several authors derived alternative formulations, from simple ones for particles of a given size (Venkatram and Pleim, 1999) to process-based approaches including multiple size classes (Slinn, 1982). In this latter approach, the influence of particle size on v_d is accounted for only through the collection efficiency coefficient derived from wind tunnel deposition measurements in the late 1950s and 1960s. Petroff et al (2008b) derived a parameterization of the particle collection for each deposition mechanisms: Brownian diffusion, interception, inertial and turbulent impactions, and gravitational settling.

For larger particles, the dominant processes of interception and impaction may circumvent r_b and as particle diameter increases, v_s increasingly dominates the total flux (Petroff et al., 2008a).

More recently, Giardina and Buffa (2018) proposed a new approach for modelling dry deposition velocity, based on the assumption that the vertical transport of particles can be modeled by adding together the **turbulent transport** and **particle settling**. More specifically, the dry deposition velocity is evaluated by assuming that the resistances that affect the particle flux in the QLS can be combined to take into account local features of the mutual influence of Brownian diffusion motions and inertial impact processes in the QLS and the atmospheric turbulence in the SL. This assumption leads to a new scheme for the parametrization of the deposition velocity of particles, based on a different electrical analogy than described above (cf. Figure 4). In this approach, aerodynamic resistance r_a (i.e. contribution to the deposition due to the atmospheric turbulence in the SL) is connected in series with the resistance r_{ql} across the quasi-laminar sublayer (QLS) related to mechanisms of **diffusion by Brownian motions** and the **impaction** phenomena.

5 Model equations for dry deposition of particles

The previous scheme based on the electrical analogy is used for our modelling approach of dry deposition velocity of particles.

5.1 Estimation of total resistance r

Accordingly to Figure 4, the total resistance r can be evaluated using the equation as follows (Giardina and Buffa, 2018):

$$r(z) = r_a + r_{ql} \tag{3}$$

Where the aerodynamic resistance r_a and the resistance r_{ql} are evaluated according to Eq(4) and (8), respectively.

5.2 Estimation of aerodynamic resistance r_a

As mentioned above for particle pollutants in the SL region, the **turbulence** acts on particle motion similar to that on gas; however, the process is also influenced by **gravity** (for heavier particles). Therefore, the aerodynamic resistance r_a to transfer considering the turbulence phenomenon in SL can be determined by using the Monin-Obukhov similarity theory leading to the following relationship (Giardina et al., 2017; Giardina et al., 2018):

$$r_a = \frac{1}{ku*}(ln\frac{z}{z_0} - \varphi_h) \tag{4}$$

Where: u* is the friction velocity, which represents the intensity of the atmospheric turbulence; z_0 is the surface roughness height above the displacement plane; and k is the von Karman constant (generally equal to 0.4).

φ_h can be calculated as follows :

$$\varphi_h = -5\frac{z}{L} \quad \text{with } \frac{z}{L} > 0 \ \text{(stable atmospheric conditions)} \tag{5}$$

$$\varphi_h = e^{\left\{0.598+0.390\ln\left(-\frac{z}{L}\right)-0.09\left[\ln\left(-\frac{z}{L}\right)^2\right]\right\}} \quad \frac{z}{L}<0 \text{ (unstable atmospheric conditions)} \qquad (6)$$

Where L is the Monin-Obukhov length that characterizes the stability of the SL layer in the lower part of atmosphere; L can be computed as follows:

$$L = \frac{u*^3 C_p \rho T}{kgH} \qquad (7)$$

Where C_p is the specific heat at constant pressure, T the average temperature in SL and H the sensible heat at constant pressure.

5.3 Estimation of the quasi-laminar sublayer resistance for particle r_{ql}

Once in the quasi-laminar QLS sublayer, most particle dry deposition models treat surface uptake in terms of inertial impaction by inertial forces, interception, Brownian diffusion and gravitational settling (e.g. Slinn, 1982). However, as mentioned above, the deposition process in this layer is particularly influenced from the Brownian motion and the gravity due to heavier particles (and diffusion phenomenon for gas).

$$\frac{1}{r_{ql}} = \left(\frac{1}{r_{db}} + \frac{1}{r_{ii}} + \frac{1}{r_{ii}+r_{ti}}\right) \qquad (8)$$

- r_{db}: resistance related to **Brownian diffusion** phenomena: the Brownian's diffusivity of a particle of a given size should be computed from the slip-flow corrected Stokes–Einstein relation based on the Schmidt number (S_c) (Pryor et al 2007 ; Giardina and Buffa, 2018).

$$r_{db} = \frac{1}{u*} c S_c{}^p \qquad (9)$$

Where c and p are constants. The parameter p usually lies between 1/2 for water surfaces and 2/3 with larger values for rougher surfaces.

S_c is evaluated as:

$$S_c = \frac{v_a}{D} \qquad (10)$$

Where v_a is the air kinematic viscosity ($m^2.s^{-1}$), and D is the particle's Brownian diffusivity of air ($m^2.s^{-1}$) determined from Stokes-Einstein equation:

$$D = \frac{K_B T C_c}{3\pi \mu_a d_p} \qquad (11)$$

With K_B the Boltzmann constant (J/K); T the absolute temperature; μ_a the air dynamic viscosity; and C_c the Cunningham factor from Eq.(18).

For all surface conditions the following relationship is assumed:

$$r_{db} = \frac{1}{u * S_c{}^{-2/3}} \quad \text{for all surface conditions} \qquad (12)$$

- r_{ii}: resistance for the **inertial impact** process, based on the evaluation of the Stokes number (S_t) which is a function of settling velocity v_s (Giardina and Buffa, 2018). Two different equations for the impaction efficiency are classically used, as a function of smooth surface and surfaces with rough elements:

$$r_{ii} = \frac{1}{u^* E_{IM}} \tag{13}$$

Where E_{IM} is the **impaction efficiency** expressed as follows:

$$E_{IM} = \frac{St^2}{St^2+400} \text{ for smooth surfaces} \tag{14}$$

$$E_{IM} = \frac{St^2}{St^2+1} \text{ for rough surfaces} \tag{15}$$

Where S_t is the Stokes number defined as:

$$S_t = \frac{v_s}{g}\frac{u*^2}{v_a} \tag{16}$$

With the settling velocity v_s increasing in proportion to the square of the particle diameter, d_p, according to the law of Stokes, which is valid for particles with a diameter of up to 50 μm :

$$v_s = \frac{{d_p}^2 g\left(\rho_p - \rho_a\right) C_c}{18\mu_a} \tag{17}$$

Where g is the gravitational acceleration; ρ_p the particle density; $\rho_a$the air density; $\mu_a$the air dynamic viscosity; and $C_c$the Cunningham factor. The parameter C_c can be given as follows:

$$C_c = 1 + \frac{\lambda_a}{d_p}\ (2.514 + 0.8e^{(-\frac{0.55d_p}{\lambda_a})}) \tag{18}$$

Where λ_a is mean free path of air.

- r_{ti}: resistance related to **turbulent impact phenomena**, evaluated as a function of a dimensionless particle relaxation time τ_+ (Giardina and Buffa, 2018).

$$r_{ti} = \frac{1}{u^* m \tau_+^n} \tag{19}$$

Where τ_+ is evaluated using the following relationship:

$$\tau_+ = \tau\frac{u^{*2}}{v_a} \tag{20}$$

With τ the particle relaxation time defined for spherical particle as follows:

$$\tau = \frac{{d_p}^2 \rho_p C_c}{18\mu_a} \tag{21}$$

Based on these resistance calculations, Giardina and Buffa (2018) proposed an evaluation of the particle deposition velocity by the following equation, replacing Eq(2) :

$$v_d = \frac{v_s}{1 - e^{-r(z) \cdot v_s}} \quad (22)$$

Where: the settling velocity v_s increases in proportion to the square of the particle diameter, D_p, according to the law of Stokes, which is valid for particles with a diameter of up to 50 µm (Giardina et al., 2017; Giardina and Buffa 2018). r(z) is the total resistance to the transport, see Eq.(3), which is therefore computed as a function of D_p and height z, as described above.

This model has been successfully validated through a comparison with experimental data from literature depending on the particle diameter, D_p, for different meteorological conditions and surface typologies, such as short grass, grassland, sand, forest - as well as with other models. As these measurements have been obtained with different methods and under different aerodynamic conditions, there are still uncertainties about the size dependence of the deposition and about the influence of other parameters that describe the meteorological conditions and the canopy geometry (Giardina and Buffa, 2018).

For calculating dry deposition velocity of tritiated particles, a similar equation than proposed by Giardina and Buffa (2018) in Eq.(22) has been used and parameterized for grass and vegetables, based on the electrical analogy in which the total resistance to the transport is calculated according to Eq(3) (with the different resistances calculated in equations (4) to (19)).

6 Model-data and model-model comparison results

Parametrization of the inertial impact process for grass and vegetable surfaces

Eqs(14)-(15) proposed by Giardina et al (2018) for computing the **impaction efficiency** for smooth and rough surfaces have been used in our study (cf. Figures 5 and 6 below).

Various authors suggested similar equations or formulae for computing the impaction efficiency as a function of smooth surface and surfaces with rough elements (Slinn, 1982; Giorgi, 1986; Peters and Eiden, 1992). For example, Eq(15) has also been suggested by Slinn (1982) for vegetative canopies, and will be used in our model application for vegetables.

Giorgi (1986) suggested the following formulas for parametrization of impaction efficiency, one for smooth surfaces and surfaces elements (the same than Eq**(**14)) and the following one for vegetated surfaces, also function of the Stokes number (S_t):

$$E_{IM} = \left(\frac{St}{St + \alpha}\right)^{\beta} \quad (23)$$

This form is the same as the one used by Peters and Eiden (1992) for a spruce forest and by Zhang et al (2001) for different values of α and β varying with the land use cover. In our study, Eq(23) is used with α chosen as 1,2 and β as 2, respectively, for both grass and crops (Zhang et al., 2001).

The suggested parametrization for modelling the deposition velocity v_d is validated through a comparison with several experimental data reported in the literature depending on the particle diameter, d_p, for different meteorological conditions and surface typologies, including short grass, grassland and vegetables. Comparisons against results obtained by using dry deposition models based on other parametrization of the impaction process (e.g. Eq(23), Zhang et al. 2001) for the studied type of surfaces are also shown. Additionally, the settling velocity v_s evaluated using Eq(17) is also depicted in the figures described in the following sections.

6.1 Deposition on grass surfaces

The deposition velocity for smooth surface or surface with bluff roughness elements like grass or grassland is calculated using Eq(22), where the parameter r_{ii} is evaluated from Eq(13) with the impaction efficiency being computed according to Eq(14).

Figure 5 illustrates a comparison between predictions of dry deposition velocity (v_d, in $cm.s^{-1}$) obtained with the parametrization from the model of Giardina and Buffa (2018) for friction velocities of u* =0.75 and 0.26 ($m.s^{-1}$) and experimental measurements reported in literature.

With a focus on intermediate size particles, Figure 6 compares predictions of v_d obtained using the same model for higher friction velocity values of u* =1; 5 and 10 ($m.s^{-1}$) with experimental measurements reported in Figure 5. Indeed it has been highlighted that the friction velocity of the wind is a parameter related to the natural surface that has the strongest impact on the deposition (Pellerin et al., 2017; Pellerin, 2017). The other parameters related to the natural surface, such as Leaf Area Index (LAI) or vegetation cover properties (adherence, micro-roughness), have a second order impact.

In the simulation runs, the deposition velocity is modeled for particle density of 2650 ($kg.m^{-3}$) and roughness length z_0=0.02 (m) for grass, as suggested in (Pellerin, 2017).

Additionally, model runs using parametrization of Zhang et al. (2001) for the same values of friction velocities u* =0.75 and 0.26 ($m.s^{-1}$) (Figure 5) and u* =1 ($m.s^{-1}$) (Figure 6) are reported for the purposes of comparison with some theoretical approaches most in use at present for modelling the impaction process. In these simulations, the impaction efficiency has been computed according to Eq(23) with α and β chosen as 1.2 as 2, respectively. The other input parameters of the Zhang et al model are reported in (Zhang et al, 2001) and (Pellerin et al. 2017, Table 5).

Experimental measurements reported in (Liu and Agarwal, 1974) for grass and in (Chamberlain and Chadwick, 1953), (Chamberlain 1966, 1967) for sticky artificial grass are also depicted in both figures 5 and 6, as well as experimental data reported in (Maro et al, 2006), Damay (2010), Pellerin (2017) for grassland. For the latter, the dry deposition velocities v_d obtained during the four experimental campaigns (DEPECHEMOD1 to 4) are reported for atmospheric aerosol particles of diameter size between 2.5 nm and 1.2 µm, above grassland surfaces (Pellerin et al. 2017, Pellerin, 2017).

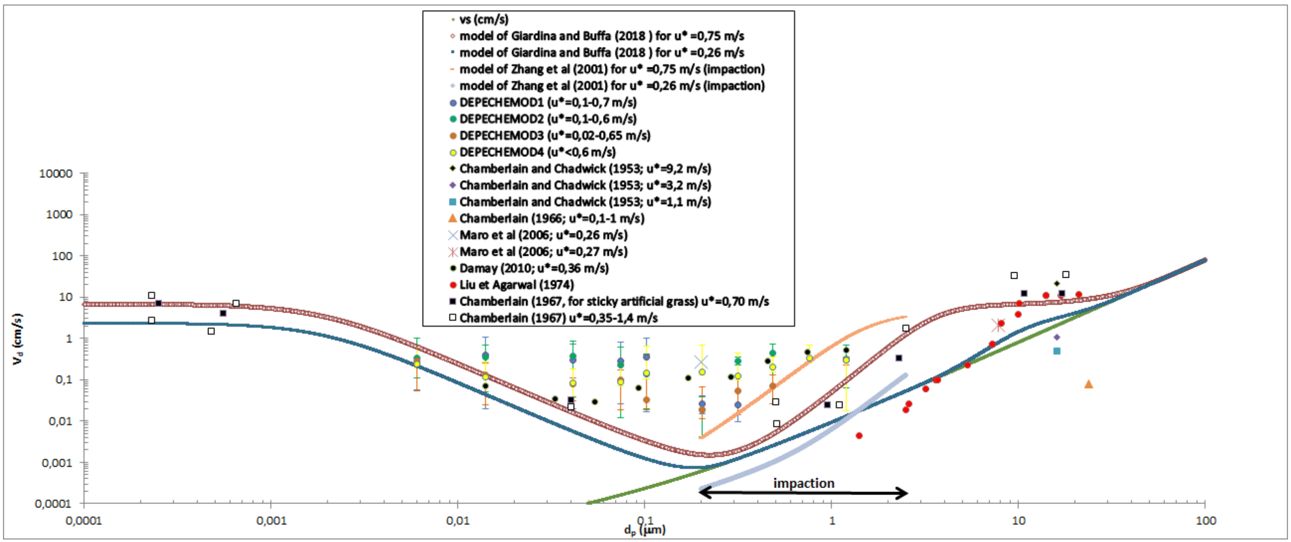

Figure 5 : A comparison between (1) dry deposition velocity predictions (V_d, in $cm.s^{-1}$) obtained using the model of Giardina and Buffa (2018) and the parametrization suggested by Zhang et al. (2001) for impaction, with friction velocities u*=0,75 and 0,26 ($m.s^{-1}$) and (3) experimental measurements reported in literature (Chamberlain and Chadwick, 1953; Chamberlain, 1966, 1967; Liu and Agarwal, 1974; Maro et al 2006; Damay, 2010; Pellerin, 2017 during the DEPECHEMOD campaigns).

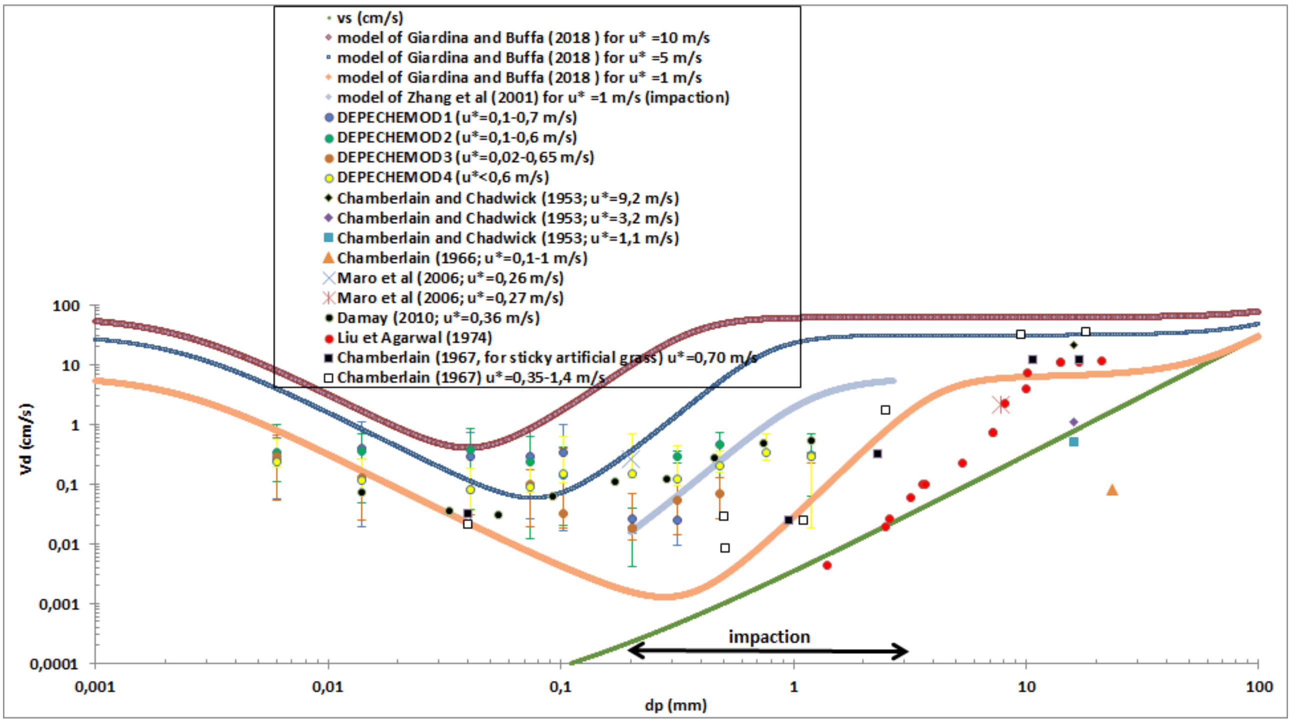

Figure 6 : A comparison between dry deposition velocity predictions obtained using the model of Giardina and Buffa (2018) for friction velocities u*=10; 5 and 1 (m.s^{-1}) and predictions using the parametrization proposed by Zhang et al. (2001) for the impaction, for u*= 1 (m.s^{-1}). The experimental measurements reported in Figure 5 are also shown.

6.2 Deposition on vegetable surfaces

The deposition velocity for vegetables is calculated using Eq(22), where the parameter r_{ii} is evaluated from Eq(13) with the impaction efficiency being computed according to Eq(15) dedicated to rough surfaces. The predictions obtained using the model of Giardina and Buffa (2018) for friction velocities of u*=0.24; 0.5 and 1.06 (m.s^{-1}) are reported in Figure 7.

Additionally, Figure 7 illustrates the predictions obtained from the parametrization of Zhang et al. (2001) for the same values of friction velocities. In these simulations, the generic Eq(23) suggested by Giorgi (1986) has also been used for parametrizing the impaction efficiency as a function of the Stokes number, with α and β chosen as 1.2 as 2, respectively, for vegetated surfaces according to Zhang et al. (2001).

A further comparison has been performed with the experimental measurements reported for plant in (Zhang et al., 2014) for the same values of friction velocity. The values of the friction velocity u* and roughness length z_0 used in Zhang's experimental tests are reported in Table 1. The same values as suggested in this table are used in the simulation runs.

For a comparison with experimental data for quite similar surfaces, the data obtained by (Hofken and Gravenhorst, 1982, cited by Giardina and Buffa, 2018); (Pryor et al., 2007) for forest, and Damay (2010) for maize are also depicted in Figure 7.

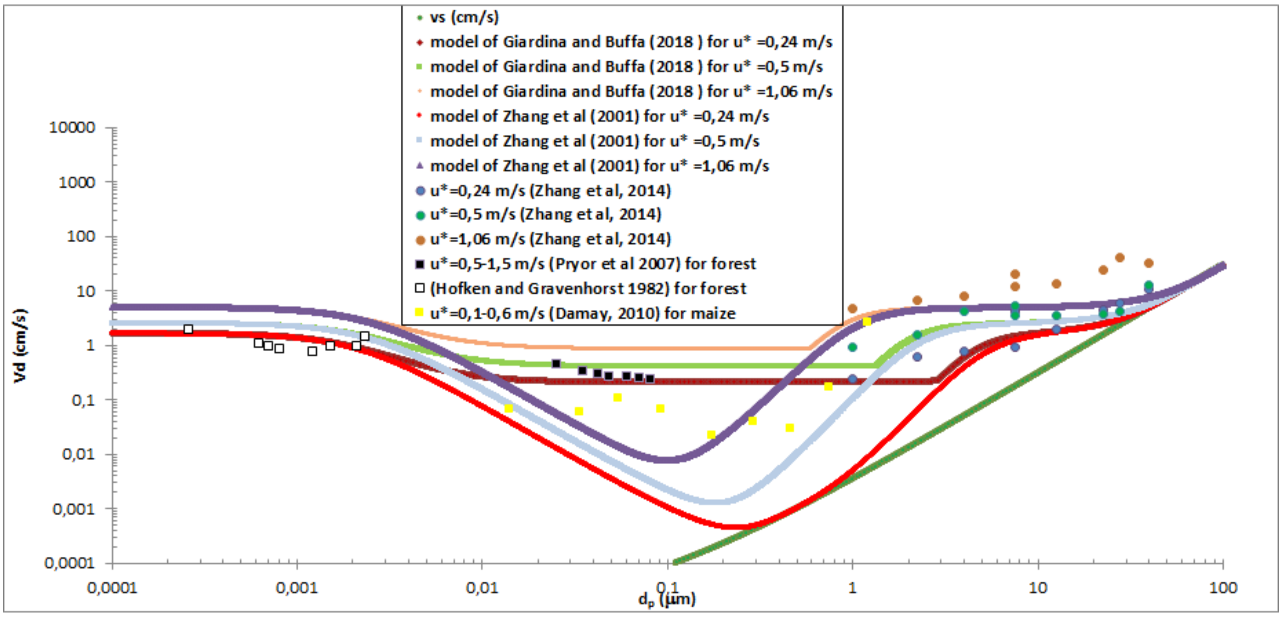

Figure 7 : A comparison between deposition velocities predicted by the model of Giardina and Buffa (2018) and using the parametrization of Giorgi (1986) for impaction as suggested by Zhang et al (2001) for vegetables, with friction velocities of u*=0.24; 0.5 and 1.06 (m.s^{-1}). The experimental data obtained by Zhang et al (2014) are also shown for the same friction velocities. For a comparison among experiments characterized by similar test conditions, the data reported in (Hofken and Gravenhorst, 1982; Pryor et al., 2007; Damay, 2010) are reported for other natural surfaces (forest and maize).

Table 1: Data used for the validation of the model with the experimental measurements reported in (Zhang et al., 2014)

	u* (m/s)	z_0 (mm)
Plant	0.24 0.50 1.06	5.927 2.877 2.106

6.3 *Application to the steel and cement particles in TRANSAT WP3*

In the framework of TRANSAT, the first step in the objectives defined in WP3 was the production and characterization of particles that represented decommissioning process in both fusion and fission within nuclear facility containing tritium. Once identified, the relevant steel and cement particles generated during decommissioning process were then produced and characterized in term of physical and chemical stabilities. The results obtained make it possible to calculate the aerosol size distribution in the case of a great number of stainless steel and cement piece cuttings, with an aerodynamic median diameter equal to 13.3 µm for steel particles (Gensdarmes et al., 2019) and equal to 4.24 µm for cement particles (Rose et al., 2019), and a geometric standard deviation of 1.35 and 2 for steel and cement particles (Gensdarmes et al., 2019; Rose et al., 2019).

The estimated dry deposition values inferred from the modelling outputs shown in Figures 6 and 7 for grass and vegetable surfaces, respectively, are reported in Table 2.

Table 2: Estimated dry deposition velocity values (V_d, cm/s) onto grass and vegetable surfaces for the stainless steel and cement particles characterized in the framework of TRANSAT WP3

V_d (cm/s)	Steel particles	Cement particles
Grass surface	[4.8 – 51.1]	[2.2 -49.8]
Vegetable surface	[3.2 – 6.1]	[0.4 – 6.0]

7 Discussion

The performance of the dry deposition velocity parametrization has been compared to theoretical observations, as well as a number of experimental measurements obtained for smooth and rough surfaces, represented in our application by grass and grassland surfaces (Figures 5 and 6) and vegetable surfaces (Figure 7), respectively.

Globally, the different curves predicted by the model follow the theoretical curve form of dry deposition velocity with the effects of the three dry deposition mechanisms: Brownian diffusion, interception and impaction (Sehmel, 1980; cf. Figure 2), depending on particle size, showing the main noticeable features (Figures 5 and 6): the decrease of deposition velocity for very small particles (< 0,1 μm) due to the **Brownian diffusion** and the eddy turbulence, down to a minimum value with regard to particles with a diameter of less than (or around) 0.2 µm. Beyond 0.5 µm and up to 1.2 µm, the increase of v_d is caused by the sharp escalation in the influence of **interception** and **impaction**. At diameters >1 µm, deposition increases with increasing particle size, because the effects of **gravitational settling** and **impaction** become predominant (Petroff et al., 2008a; Giardina et al., 2017). This model pattern is consistent with the behavior recommended according to the physical meaning of the deposition processes.

The overall change of deposition velocity as a function of particle size also strongly depends on atmospheric conditions, with smoother variations for higher friction velocity values, and in these situations, a minimum value of deposition velocity being reached for smaller particle sizes (<0,3 μm). (see Figure 6 and Figure 7). This confirms the first influence of friction velocity on dry deposition velocity, as highlighted by (Pellerin et al. 2017, Pellerin 2017). By contrast, it has been tested that any change in surface roughness z_0 has a mitigated impact on dry deposition velocity values (not shown).

Figure 5 shows experimental data valid for dry deposition processes to grass and grassland surfaces. When applied on these smooth surfaces, the model outputs from Giardina and Buffa (2018) do not agree as well with measurements and differences more than of one order of magnitude can arise for particle diameter of intermediate values (d_p between 0.1 - 1μm) and more specifically for lower friction velocities u*=0.26 (m.s^{-1}) (cf. Figure 5). Given the discrepancy between the model and the measured values for grass in these situations, the Giardina and Buffa (2018) parametrization of the impaction efficiency (cf. Eq(14)) leads to an underestimation of the phenomena of interception and impaction in our application on smooth surfaces.

The parametrization suggested by Zhang et al (2001) for the impaction process with u*=0.75 (m.s^{-1}) allows a better agreement than the model of Giardina and Buffa (2018) with the experimental data obtained in the DEPECHEMODi (i from 1 to 4) campaigns reported in Pellerin (2017), especially for particle diameter of intermediate values (d_p between 0.1 - 1μm; cf. Figure 5). On the other hand, the model of Giardina and Buffa (2018) based on the value of u*=0.26 (m.s^{-1}) allows a better agreement with the reported experimental data for d_p with very low values (below 0.01 μm) or high values (above 1 μm). For these latter values, the experimental data reported for u*=0.70 and 0.75 (m.s^{-1}) (i.e. Liu and Agarwal, 1974; Chamberlain, 1967)) show that the impaction process is predominant with respect to the gravitational effects, this can be deduced from data that are located over the v_s curve, representing the gravitational settling velocity trend (green line). This aspect is well captured by the Giardina and Buffa (2018) approach.

Figure 6 shows similar trends of dry deposition velocity predictions obtained using this approach for friction ranging from u* = 1 to 10 (m.s^{-1}). Additionally, the predictions based on the parametrization proposed by Zhang et al. (2001; for u*= 1 (m.s^{-1})) produce results that are higher in accordance with the large number of measurements for particles of intermediate diameter, allowing predictions that are sufficiently accurate and sensitive to the change of canopy. On this point, this result is achieved by accounting a different parametrization of inertial impact processes. Indeed, the Zhang's parametrization of impaction (cf. Eq(23) for both grass (Figures 5 and 6) and vegetables (Figure 7) provides similar predicted patterns of v_d but higher values to that of Giardina and Buffa (2018). The model of Zhang et al. (2001) is mainly governed by the phenomena of sedimentation and Brownian diffusion, this drawback was offset in our application by taking into account the recent formulation of the particle deposition velocity of Giardina and Buffa (2018, Eq(22)) along with the parametrization proposed by Giorgi (1986) and reformulated by (Zhang et al. 2001) for computing the impaction phenomena in grass and vegetables. When applied on rougher surfaces than grass or grassland, such as vegetables, it seems that the Zhang et al suggested parametrization for impaction results is closer to measurements. This has been confirmed by Petroff et al (2008a,b) according to which the model of Zhang et al. (2001) is particularly more suitable for predicting the fine particle deposition rates on very rough surfaces than for less rough or smoother surfaces (Petroff et al., 2008a,b).

Further parametrizations should be tested in the future by using similar or other formulas suggested by various authors (Slinn, 1982; Giorgi, 1986; Peters and Eiden, 1992…) for computing the impaction efficiency as a function of smooth and rough surfaces.

Additionally, in the turbulent-impaction regime, a sharp increase of the deposition velocity is noticeable, despite a variability of the results (Figures 5 and 6), and denotes a strong influence respect to the particle inertia. The combination of the resistances r_{ii} and r_{ti}, as described in section 5.3 allows to catch this phenomenon, improving the predictions especially for intermediate size and coarse particles.

In the framework of the TRANSAT WP3, the stainless steel and cement particles were characterized by aerodynamic diameters that are higher than the diameter range of [0.1-3 µm] were impaction has the greatest influence on dry deposition velocity. With a focus on the size of our particles of interest, i.e. characterized by an aerodynamic median diameter around 13 µm and 4 µm for steel and cement particles, respectively, modeling results as well as experimental data when available, show smoother variations than for smaller particles (i.e. < 2 µm). However, estimated dry deposition velocities inferred from the several modeling parametrization show higher variability for grass than for vegetable surfaces. The friction velocity of the wind, which is related to the natural surface, may explain this difference in a greater extent than other parameters including LAI or vegetation cover properties (roughness...).

8 Conclusions and perspectives

As highlighted in ATMES (Atmospheric Transport Model Evaluation Study) report (Klug et al., 1992), the highest uncertainties in numerical evaluations of pollutant transport and dispersion in air are introduced by the parameterization both of the source term and deposition velocities, particularly for particles in the submicronic range (Petroff et al., 2008a; Damay, 2010; Pellerin, 2017). For this size range of particles, uncertainties on the dry deposition velocity values are up to several orders of magnitude discrepancies according to the model used (Petroff et al., 2008a; Giardina et al., 2017). Additionally, experimental uncertainness precludes drawing general conclusions from the deposition rate measurements performed by various international laboratories (Sehmel, 1980). All these issues limit the possibility of studying the dry deposition process of particles using a single modeling approach and further complicate the model-measurement intercomparison analysis.

In this study, the approach proposed by Giardina and Buffa (2018) based on an electrical analogy schema has been advocated to be used along with other suggested parametrizations of the laminar sublayer resistance for the inertial impact process in order to predict the dry deposition velocity of particles. The dry deposition velocity is calculated as a function of the most relevant driving factors, including the particle characteristics (size and density), the meteorological conditions and the surface features of the environmental receiving cover. Comparisons with published measurements and other parametrizations show that by combining the semi-mechanistic resistance equations of the model of Giardina and Buffa (2018) along with existing empirical parametrization of impaction efficiency can predict reasonable deposition velocities for a wide particle size range over grass and vegetable surfaces. This revised parameterization could be easily implemented within atmospheric dispersion modeling codes and be capable of efficiently addressing different deposition surfaces, from smooth to rough natural surfaces, for several radioactive pollutants.

However, knowledge, experimental data and modelling approaches reported in this review (and in the literature), when available for aerosol particles, never referred to tritiated particles, since the behavior of airborne tritiated (sub)micronic particles in the atmosphere-soil-plant continuum has been very poorly understood and documented so far (Le Dizès, 2019). Among the different processes and approaches reviewed, no existing tritiated particles-specific model (i.e. one specifically designed to account for the processes that determine the fate and behavior for tritiated particles once released in the environment) has been identified so far, with the exception of the wet deposition process that has been well formalized and parametrized for airborne tritiated particles (Le Dizès, 2019).

Despite the lack of knowledge, models and data available on the behavior of airborne tritiated particles, similar information may be borrowed, at least qualitatively, from processes related to generic (sub)micrometric particles, by making some assumptions. It is reasonable to hypothesize that specific processes related to tritiated particles must be taken into account in relation to a generic particle of the same size... if not, we are moving towards a possibly specific parameterization. The general concern is whether (or not) the size of the particles means that tritiated particles have behavior that might be predictable from knowledge of the behavior of the other radioactive particles.

The generic and well-documented approach of Giardina and Buffa (2018) presented in this report, with suggested parametrizations of a given phenomenon (here impaction)- although not exhaustive of all mechanisms governing dry deposition - has allowed to lay the groundwork to configure a dry deposition rate model for tritiated particles of (sub)micron size on prairies and vegetables. The parameterization has been derived from existing knowledge and data reported from literature for micron or nanometer sized particles, and will need, as far as possible, to be specifically devoted to tritiated particles. This configuration of a model of deposition of tritium in particulate aerosol form onto prairies and vegetables, is a first step towards the formalization in a simple model of the processes of foliar uptake and internalization in plant biomass - which occur after deposition - of (sub)micronic (possibly tritiated) particles.

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