Dry deposition of particle on urban areas

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Abstract. Dry deposition process is recognized as an important pathway among the removal processes of radioactive pollutants in atmosphere. There is not a unique and accepted theoretical description of involved dry deposition phenomena due to the complexity of the fluid-dynamic processes that influence the deposition flux, but also because there is a lack of experimental data covering all scenarios of interest. In this paper, that is the result of a National Research Program a research activity conducted by DEIM Department of the University of Palermo and ENEA and funded by the Italian Minister of Economic Development, a new schema for parameterization of particle dry deposition velocity on urban area is proposed. The work required comparisons with some experimental data reported in literature for different particle deposition scenarios. The results show that the proposed approach can catch some aspects of phenomena involved in dry deposition processes for the examined environmental conditions with good agreements.

1. Introduction

Urbanization and industrialization are the cause of the formation of pollutants that are deposited on trees, grass, crops, water bodies, and buildings with ecological and non-ecological impacts. In the nuclear field, in the event of a severe accident, which leads to a release of radionuclides in the atmosphere, key challenges are the characterization of the specific isotopes that are released as well as studying the dispersion and deposition phenomena. This is particularly important for the definition of effective mitigation measures and actions to protect the population.

In this field, dry deposition process is recognized as an important pathway among the various removal processes of radioactive pollutants in atmosphere.

There is not a unique and accepted theoretical description of the involved dry deposition phenomena because the complexity of the fluid-dynamic processes, that influence the deposition flux, and lack of a complete experimental set of data covering all scenarios of interest. Various experimental campaigns, performed in different international laboratories, allowed evaluations of deposition velocities for different types of pollutants and deposition surfaces. Nevertheless, there is a difficulty of generalization since the velocity values differ by four orders of magnitude for gases and three orders for particles.

All the above issues limit the possibility to study the dry deposition process with a single modelling approach.

In this field research activities have been focused on the identification, among the models reported in literature, of those approaches that are capable of representing dry deposition phenomena for several categories of pollutants and deposition surfaces [1-3]. On the basis of this study, a new schema for parameterization of particle dry deposition velocity onto rough surfaces, such as an urban area, is proposed. The main aim is to develop an approach that is easy to implement within established atmospheric dispersion modeling codes as well as capable of dealing efficiently with different deposition surfaces.

The work required comparisons with some experimental data reported in literature for different particle deposition scenarios. The results show that the proposed approach can catch some aspects of the phenomena involved in dry deposition processes for the examined environmental conditions with good agreements.

2. A modified approach for dry deposition

The primary phenomena that are considered to affect the process can be described as follows:

- transport due to atmospheric turbulence in the lower part of the Planetary Boundary Layer (PBL), which is called the Surface Layer (SL). It is independent of the physical and chemical nature of the pollutant and depends only on the atmospheric turbulence level (i.e. turbulent movements of air);
- diffusion in the thin layer of air, which overlooks the air-ground interface (named Quasi-Laminar Sublayer, QLS), where the molecular diffusion for gas, Brownian motion and turbulent diffusion for particles and gravity for heavier particles become dominant;
- transfer to the ground (e.g. interception, impaction, and rebound), which exhibits a pronounced dependence on the surface type with which the pollutant interacts.

A key concept to study the dry deposition process is the deposition velocity v_d [m/s] (i.e. the deposition velocity at a given height z) that links the pollutant vertical flux to the concentration measured at quota z [m] to the ground reference level:

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

with F pollutant flux removed per unit area and C(z) the pollutant concentration at quota z.

Considering that the reciprocal of v_d is the overall resistance to mass transfer, the influence of the above described phenomena on deposition velocity can be expressed in terms of an electric analogy, in which the resistances to the mass transfer are configured in parallel and series circuits to describe transfer factors between air and surface.

The vertical transport of particles can be modeled by assuming that the turbulent transport and particle settling can be added together as follows:

$$K_p \frac{dC}{dz} + v_s C = F \tag{2}$$

where K_p is the eddy diffusivity for the mass transfer of species with a concentration C and v_s is the settling velocity evaluated as follows:

$$v_s = \frac{d_p^2\, g\, (\rho_p - \rho_a)\, C_c}{18\, \nu_a} \tag{3}$$

being g the gravitational acceleration; ρ_p the particle density; ρ_a the air density; ν_a the air kinematic viscosity; and C_c the Cunningham slip correction factor.

By integrating the above equation, it is possible to obtain the expression of the deposition velocity as follows:

$$v_d = \frac{v_s}{1-e^{-[r(z)\, v_s]}} \tag{4}$$

where $r(z)$ is the total resistance to the transport, which can be computed as a function of d_p and height z (quota to the ground reference level), as explained in the following section.

Based on the electrical analogy, in this paper it is proposed to evaluate the resistance r for urban rough surfaces by using the schema shown in figure 1.

It is to be noted that in urban-scale dispersion models, the lowest portion of the boundary layer is often represented using surface layer similarity parameterizations. Boundary layer formulations of this type are only applicable in the inertial sublayer well above the building tops, but not in the immediate vicinity of the urban canopy elements where the flow locally depends on the particular building arrangement and thus it has a rather complex structure.

As above said, interception and inertial forces may transport particles across the various sublayers. Once a particle has traversed also the viscous sublayer, it will interact with the surface. Depending on the characteristics of the contaminant and surface, a particle may stick or bounce off. Particle deposition by any combination of these transport mechanisms largely depends on types of deposition surfaces and particle characteristics [4].

The scheme of deposition processes upon the canopy of urban surfaces proposed in this work has been modified to include the particle rebound or resuspension phenomena together with Brownian diffusion, impaction process, and turbulent transfer.

In figure 1 the aerodynamic resistance r_a (i.e. contribution to the deposition due to the atmospheric turbulence in SL) is connected in series with the resistance r_{ql} across the quasi-laminar sublayer to take into account mechanisms of diffusion by Brownian motion and impaction phenomena. The resistance r_{ql} is evaluated by considering two resistances in parallel, that is: resistance r_{bd}, which represents the Brownian diffusion, and resistance r_i, which allows to treat impaction processes.

The resistance r_i is evaluated by considering two resistances in series: resistance r_{ii} takes into account the inertial impact condition, and resistance r_{ti} considers the effects resulting from turbulent impaction.

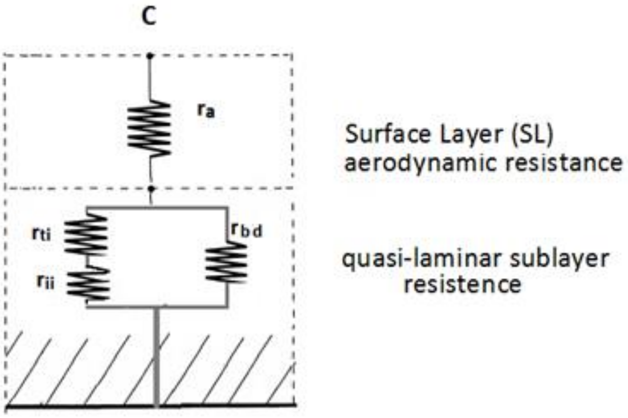

Figure 1. New schematization for parametrization of particles deposition velocity.

These last assumptions allow 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).

Accordingly, the overall resistance r can be evaluated by using the following equations:

$$r = r_a + r_{ql} \tag{5}$$

where r_{ql} and r_i are evaluated as follows:

$$\frac{1}{r_{ql}} = \frac{1}{r_{bd}} + \frac{1}{r_i} \qquad (6)$$

$$r_i = r_{ii} + r_{ti} \qquad (7)$$

Following the usual methods of micrometeorology for homogeneous terrain, the pollutant concentration flux can be expressed in terms of the local flux–gradient relationship (surface-layer similarity theory) [5]. Therefore the resistance r_a can be determined by using Monin–Obukhov similarity theory as follows [6-8]:

$$r_a = \frac{1}{ku_*}\left[\ln\frac{z}{z_o} - \Psi_h\right] \qquad (8)$$

being z_0 the surface roughness height above the displacement plane and k the von Karman constant (generally equal to 0.4).

Brandt et al. in [9] suggested the following relationship for calculating parameter Ψ_h in equation (8):

$$\Psi_h = -5\frac{z}{L} \qquad \text{with } \frac{z}{L} > 0 \text{ (stable atmospheric conditions)} \qquad (9)$$

$$\Psi_h = e^{\left\{0{,}598 + 0{,}390\ln\left(-\frac{z}{L}\right) - 0{,}09\left[\ln\left(-\frac{z}{L}\right)\right]^2\right\}} \quad \text{with } \frac{z}{L} < 0 \text{ (unstable atmospheric conditions)} \qquad (10)$$

where L is the Monin-Obukhov length computed as follows:

$$L = \frac{u_*^3 c_p \rho \overline{T}}{kgH} \qquad (11)$$

with c_p specific heat at constant pressure, $\overline{T}$ average temperature in SL, and H sensible heat.

For the resistance r_{bd} various models predict a functional dependence on Sc number such that in general:

$$r_{bd} = \frac{1}{u_*} c\, Sc^p \qquad (12)$$

where c and p are constant.

The parameter p usually lies between 1/2 and 2/3 with larger values for rougher surfaces. For example, Slinn and Slinn in [10] suggested a value of 1/2 for water surfaces. Slinn in [11] suggest a value of 2/3 for vegetated surfaces. Zhang et al. in [12] used values of parameter p varying with land use categories.

In this work it is assumed in equation (12) the following relationship:

$$r_{bd} = \frac{1}{u_* Sc^{-2/3}} \qquad (13)$$

The transport of particles by Brownian diffusion represented as function of $Sc^{2/3}$ in equation (12) is recommended in various works on the basis of theoretical and empirical results [13, 14].

It is proposed to evaluate the resistance for inertial impact process r_{ii} in equation (7) by using the following relationships valid for rough surfaces:

$$r_{ii} = \frac{1}{u_*\left(\frac{St^2}{St^2+1}\right)R} \qquad (14)$$

Some authors suggested this formula or similar for impaction efficiency as function of smooth surfaces and surfaces with roughness elements [11]. Note that particle rebound is also included via the factor R [12].

Slinn in [11] suggested the following form for R:

$$R = e^{(-b\sqrt{St})} \qquad (15)$$

where b is an empirical constant [12]. In this work it is assumed b=2 as suggested in [15].

For the calculation of resistance r_{ti}, general assumptions are reported below.

As well known, empirical relations of turbulent deposition are typically presented in terms of the dimensionless particle relaxation time τ_+:

$$\tau_+ = \tau \frac{u_*^2}{\nu} \tag{16}$$

where τ is the particle relaxation time defined, for a spherical particle, as follows:

$$\tau = \frac{d_p^2 \rho_p C_c}{18\mu} \tag{17}$$

Various models predict a functional dependence of resistance turbulent impact phenomena r_{ti} on τ_+ as follows:

$$r_{ti} = \frac{1}{u_* m \tau_+^n R} \tag{18}$$

It is to be noted that in equation (18) there is the correction factor R related to the collection efficiency for rebound evaluated by equation (15). This allows us to take into account the functional dependence of rebound phenomena on turbulent impact conditions. The constants m and n in equation (18) have been evaluated by fitting some data reported in literature for urban surfaces And they are set to 0.05 and 0.75, respectively.

3. Comparison with experimental data

The new approach for computing deposition velocity v_d is validated by comparison with experimental data reported in [16] for several meteorological conditions and deposition urban surfaces.

Figures 2 through 5 show experimental data for urban area in terms of deposition velocity as a function of frictional velocity u_*.

The datasets were taken from different experimental campaigns over a wide range of degrees of complexity: urban background, urban canopy, and industrial district or Venice lagoon surface (Italy). Summary of experimental sites in terms of measurement height (z), and roughness height z_o are reported in table 1.

In all figures the vertical bars present errors of 30%.

On the whole, a good agreement between the prediction trend obtained by using the proposed model and the examined experimental data is found.

In particular, it can be observed that there are good predictions for u_* values above about 0.3 m/s. Only limited effect of stability is observed with a slight reduction of the deposition velocities in stable conditions. This aspect is captured by the proposed approach.

Table 1. Summary of experimental sites used in aerosol sampling reported in [16].

Site	**Height z (m)**	**Roughness length z_o (m)**
Bologna	10	0.35 ± 0.02
Lecce	10	0.53 ± 0.02
Maglie	10	0.52 ± 0.02
Venice lagoon	9.6	0.11 ± 0.03 land, 0.01 ± 0.03 water

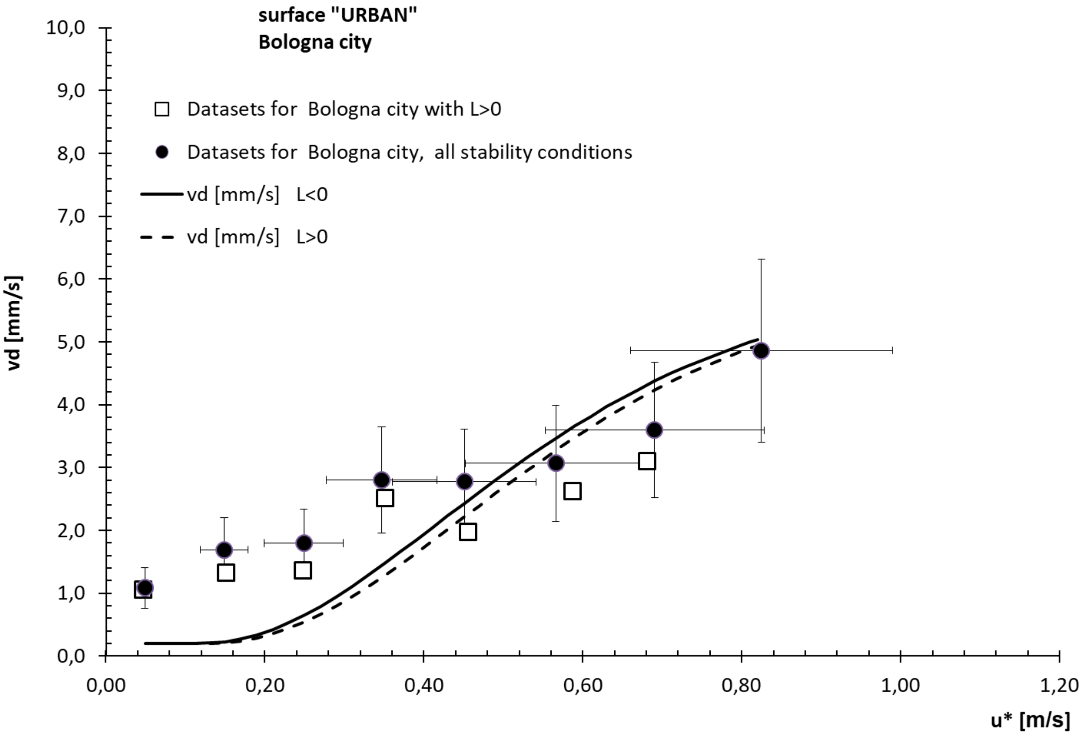

Figure 2. Deposition velocity predictions as functional dependence from friction velocity $u_*$and comparison with measurement datasets relevant to Bologna city, as reported in [16].

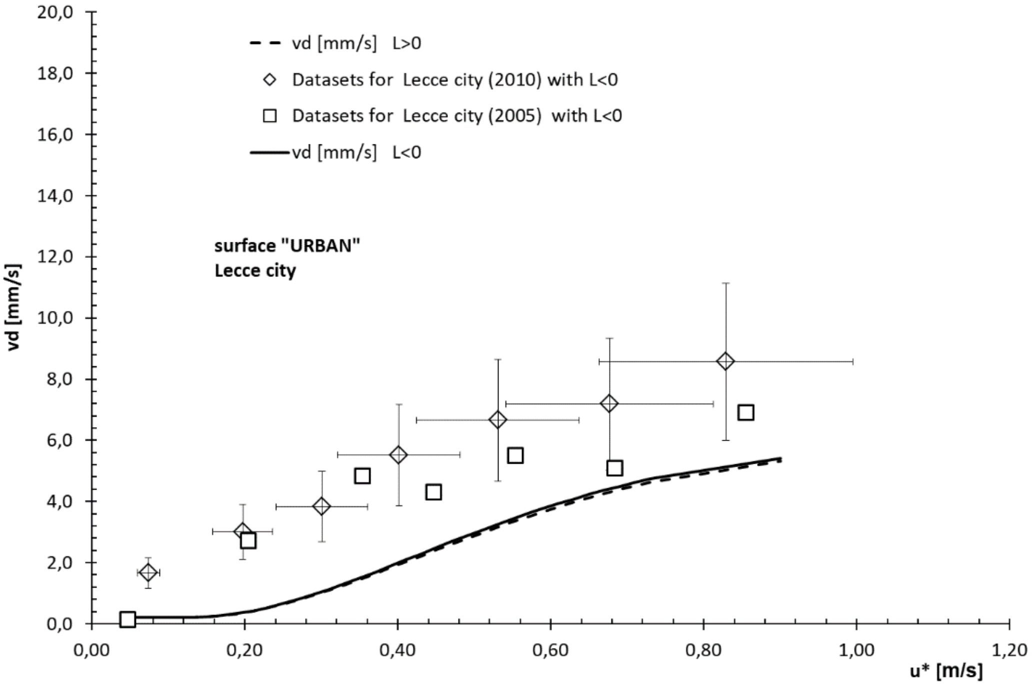

Figure 3. Deposition velocity predictions as functional dependence from friction velocity $u_*$and comparison with measurement datasets relevant to Lecce city, as reported in [16].

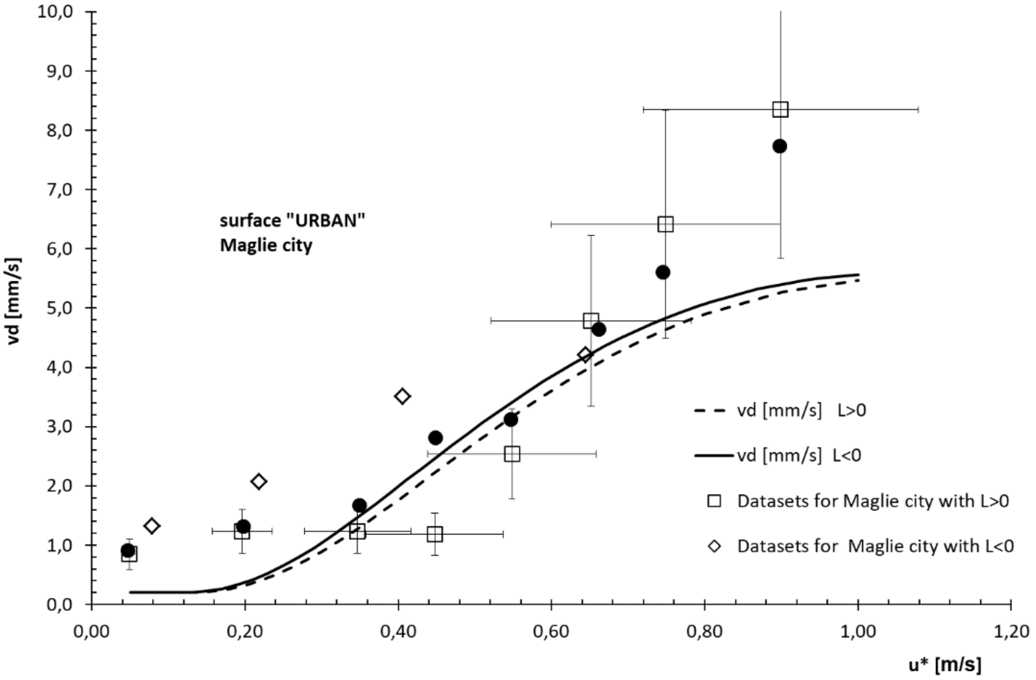

Figure 4. Deposition velocity predictions as functional dependence from friction velocity $u_*$and comparison with measurement datasets relevant to Maglie city, as reported in [16].

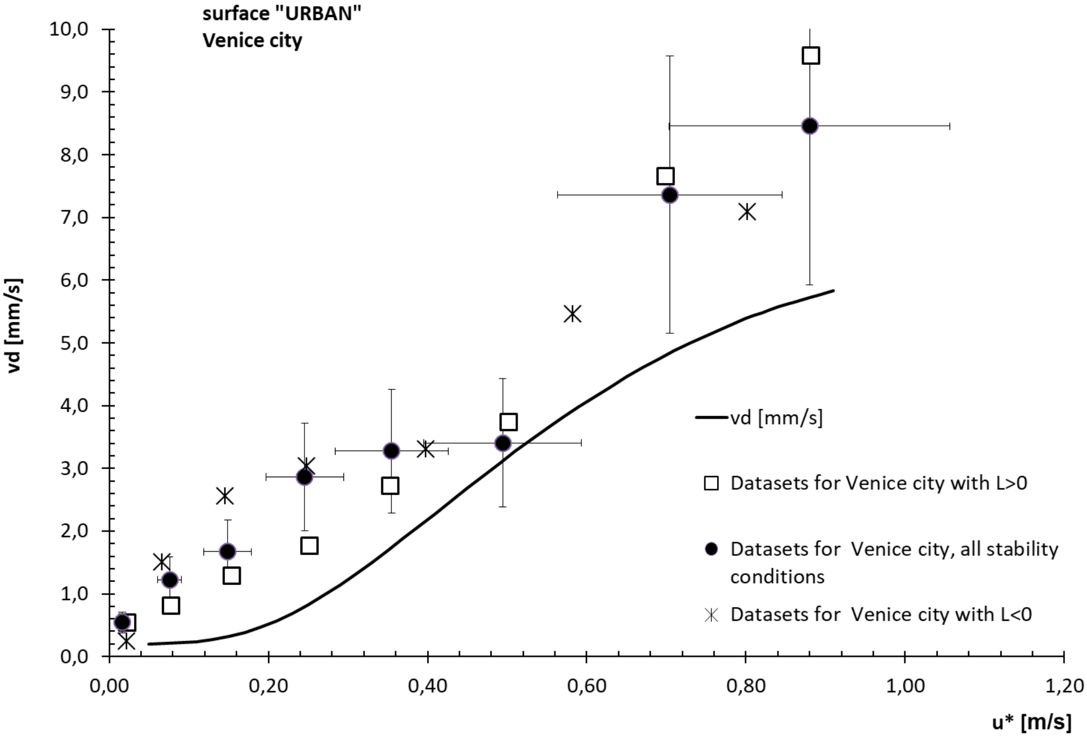

Figure 5. Deposition velocity predictions as functional dependence from friction velocity $u_*$and comparison with measurement datasets relevant to Venice city, as reported in [16].

4. Conclusion

The ATMES (Atmospheric Transport Model Evaluation Study) report, relevant to the study of models for the evaluation of radioactive pollutants disperse in the ambient atmosphere, highlighted that the highest uncertainties are in the parametrization of source terms and dry and wet deposition velocities.

In literature there are several models, but no one is able to treat exhaustively most of phenomenologies related to pollutants deposition because of the number and complexity of the involved processes. A review of the existing mechanistic models emphasizes the wide variety of ways captation can take place, however, a comparison of two similar scenarios provides large discrepancies between each other, which can differ by two orders of magnitude for the same particle diameter.

As highlighted in literature, the measurements of deposition velocity carried out by different international laboratories don’t allow to draw general conclusions due to experimental uncertainties.

In fact, for the same typology of pollutant, experimental data show that, for gasses, the values of deposition velocity differ even by four orders of magnitude and, for particles, by up to three orders of magnitude.

The main aim of our research activity was to develop an approach easy to implement within atmospheric dispersion modeling codes as well as an approach capable of dealing efficiently with different deposition surfaces for several radioactive pollutants. Some parametrizations reported in literature are based upon a number of assumptions which may be frequently violated in practice that can strongly dependent on a number of parameter combinations such as land use classification and seasonal categories.

In this paper, a new scheme for particles deposition velocity, based on an electrical analogy, is proposed to evaluate the resistances for urban rough surfaces. It is worth to note that the correlation obtained with the new schematization is based on the hypothesis that the impact phenomena in the quasi-laminar sublayer can be affected by specific local features of the mutual influence of inertial impact processes and turbulent impact phenomena. The proposed approach is further modified to take in consideration the rebound phenomena.

The validation work, carried out by using some experimental data from literature, allowed to verify the effectiveness of the proposed approach.

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