H13-10
VALIDATION OF AEROSOLS DRY DEPOSITION VELOCITY MODELS WITH NEW DATA

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Abstract: In order to evaluate the impact of aerosol pollution on ecosystems, we have to study the transfer functions of particles on vegetated canopies. One of them is the dry deposition, which is defined by the deposition velocity (Vd): the ratio between particles surface flux and the atmospheric aerosol concentration nearby the surface. This deposition velocity depends on many parameters, for example, the topography ground, the substrate, the micrometeorological conditions (turbulence) and the aerosol characteristics (size, electric charge).
Nowadays, there are several models of aerosol dry deposition which consider the effects of the turbulence, the particles size for a large range of diameter (some nm to 100 µm). In the case of nanoparticles, there is not enough reliable experimental data to allow a comparison with the dry deposition models. For operative models, the scattering of Vd experimental data of nanoparticles in a rural environment creates uncertainties larger than one order of magnitude. The study of the aerosols dry deposition velocity has remained an international challenge since the sixties and involves an in situ experimental approach, in order to consider the local particularities (substrate, turbulence, vegetated canopies, etc…)
The main aim of this study is to obtain experimental data on aerosol dry deposition velocities onto rural areas. Therefore we have developed a direct eddy covariance method with a spectral analysis. The use of an Electrical Low Pressure Impactor (Outdoor ELPI, Dekati Inc.) for this method enables to calculate dry deposition velocities for atmospheric aerosols sizing from 7 nm to 2 µm (Damay *et al*., 2009). In this work, we present the impact of micrometeorological parameters and particle size on the dry deposition velocity and make comparison with the Slinn model and the Zhang *et al.* model.

Key words: *Aerosols, deposition velocity, eddy correlation, models.*

INTRODUCTION
In order to evaluate the impact of the aerosol pollution on ecosystems, we have to study the transfer functions of particles on vegetated canopies. One of them is the dry deposition, which is defined by the deposition velocity (Vd): the ratio between particles surface flux and the atmospheric aerosol concentration nearby the surface. This deposition velocity depends on many parameters, for example, the topography ground, the substrate, the micrometeorological conditions (turbulence) and the aerosol characteristics (size, electric charge).

There are several models for the dry deposition of aerosols onto vegetative canopies, which consider the effects of turbulence and the different mechanical processes (Brownian diffusion, impaction, interception, and sedimentation which drive deposition according to particle size for a wide range of diameters). As an example, the models of Slinn (1982) and Zhang *et al*. (2001) are often used for air quality prediction. There is more than one order of magnitude between the predictions of these two models for fine particles (< 2.5 µm). In this context, data are needed to validate the models.

Most of the dry deposition measurements come from air quality studies (McMahon and Denison, 1979; Sehmel, 1980). In another study (Petroff *et al*., 2008), the relationship between dry deposition velocity over grassland and aerosol size is highlighted. These studies underline that the data are highly scattered for particle sizes below 1 µm. Furthermore, no values are available as a function of micrometeorological parameters for diameters under 0.05 µm.

The aim of this study is to obtain experimental data on aerosol dry deposition velocities onto rural areas. Therefore we have developed a direct Eddy Covariance (EC) method: the dry deposition flux is calculated from the covariance between the fluctuations of the vertical wind speed and of the atmospheric particle concentration. The use of an Electrical Low Pressure Impactor (outdoor ELPI) for this method enables to calculate dry deposition velocities for atmospheric aerosols sizing 7 nm to 2 µm. This method has been developed and presented in detail by Damay *et al*. (2009). So the method will be presented rapidly, to focus on the results, especially the impact of micrometeorological parameters and of particle size on the dry deposition velocity and comparison with the models of Slinn (1982) and Zhang *et al*. (2001).

MEASUREMENT OF THE AEROSOL DRY DEPOSITION VELOCITY BY A DIRECT COVARIANCE METHOD
The experiment was carried out in June 2007 and June 2008, on a rural area covered by maize. The site is situated 70 km of the city of Bordeaux (44°25'N, 0°40'W) in France. The site was well-suited for using the Eddy Correlation method with more than 600m of similar vegetation observed in all the wind directions and with few strong emission sources of aerosols which could convect to the measurement system. These conditions promote a steady concentration of aerosols, but nevertheless, rigorous quality tests were applied to each calculated flux to check for the steadiness of the aerosol concentration, and those measurements were in a constant layer flux. This will not be presented here because it has been described in the work of Damay *et al.* (2009). During the campaign of measurements, different micrometeorological conditions have been observed: stable, neutral and unstable conditions.
The wind speed is measured by a sonic anemometer fixed at a height of z=6m, with a sampling rate of 10Hz. The aerosol concentration is measured by an Electric Low Pressure Impactor (ELPI) which measures concentration for 9 classes of aerodynamic size between 7 nm and 2µm.

For each 30 min period, and for each range of aerosol size, the aerosol vertical flux F is calculated as the covariance between the vertical wind velocity, w ($m.s^{-1}$), and the aerosol concentration, c ($particles.m^{-3}$). The dry deposition velocity is deduced from the ratio between this aerosol vertical flux and the mean aerosol concentration. Due to the low response time of the experimental setup compared to the turbulent time scales of the particle concentrations, a flux correction has been applied. It is based on the spectral analysis because a similarity has been observed between the cospectra of the vertical wind speed and temperature and the cospectra of vertical wind speed and particle concentration in the surface layer.

Therefore, an important data processing with quality tests has been developed and a method to correct the spectral loss, based on spectral similarity, has been performed and validated. All this work has been presented in detail in the work of Damay *et al*. (2009) and will not be presented here.

IMPACT OF MICROMETEOROLOGICAL PARAMETERS AND PARTICLE SIZE

The dry deposition velocity has been determined for different sizes of aerosol corresponding to each stage of the ELPI, at different periods in a day and for different days. An example (June 16th 2007) of diurnal pattern of dry deposition, V_d, for two aerosol aerodynamic diameters (70 nm and 120 nm for respectively the stage 3 and 4 of the ELPI) is shown on the figure 1. Similar evolutions have been observed for the other aerosol sizes and for the other days of the measurements.

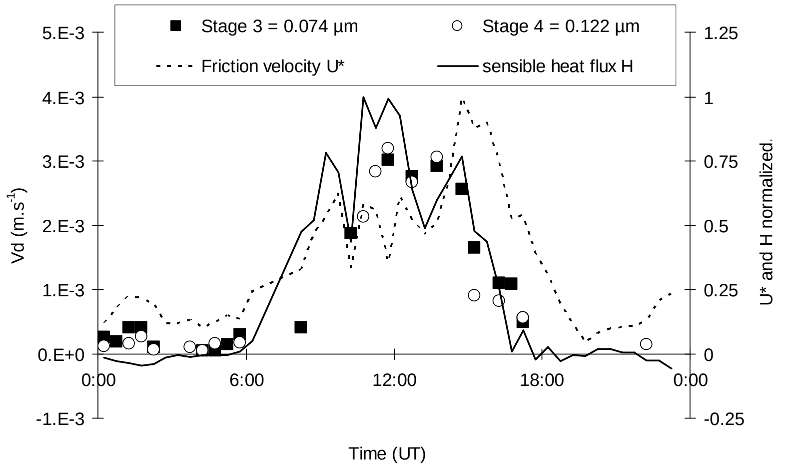

Figure 1: Example of diurnal pattern of the dry deposition velocity for 2 aerosol size: 70 and 120 nm
Comparison with the diurnal evolution of the sensible heat flux H and friction velocity U*, normalized by their respective maximum.

The results are similar to those of Lamaud *et al* (1994 a and 1994 b) and those of Held and Klemm (2006), although it was over a different canopy (forest). The evolution of Vd is similar and the maximum value of 3.10^{-3} ms^{-1} is the same order of magnitude for the studies cited above. A large variation for Vd was observed between nocturnal and diurnal results.
During the campaign of measurement, different meteorological conditions have been observed. The atmospheric turbulence is principally due to thermal and mechanical effects. The sensible heat flux (H, $W.m^{-2}$) and the friction velocity (U*, $m.s^{-1}$), are two important parameters to characterize the atmospheric turbulence because H characterizes the thermal properties of the turbulence and U* the mechanical properties. A large variation of these two parameters has been observed between diurnal and nocturnal episodes. The evolutions of the sensible heat flux and the friction velocity normalized by their respective maximum (H_{max}=144 $W.m^{-2}$ and U^*_{max}=0.6 $m.s^{-1}$) are plotted in fig.1 and show a large variation between diurnal and nocturnal episodes as well.

Impact of micrometeorological parameters

The figure 1 shows there are some correlations between the evolutions of the dry deposition velocity Vd, the friction velocity U*and the sensible heat flux H. An increase of H or U* seems to increase the dry deposition velocity. So, the evolution of Vd has been plotted in function of U* (A, on the right) and H (B, on the left) on the figure 2. This figure represents the results obtained for the second stage of the ELPI (which corresponds to an aerodynamic diameter of 33nm). Similar evolutions have been observed for the other aerosol sizes.

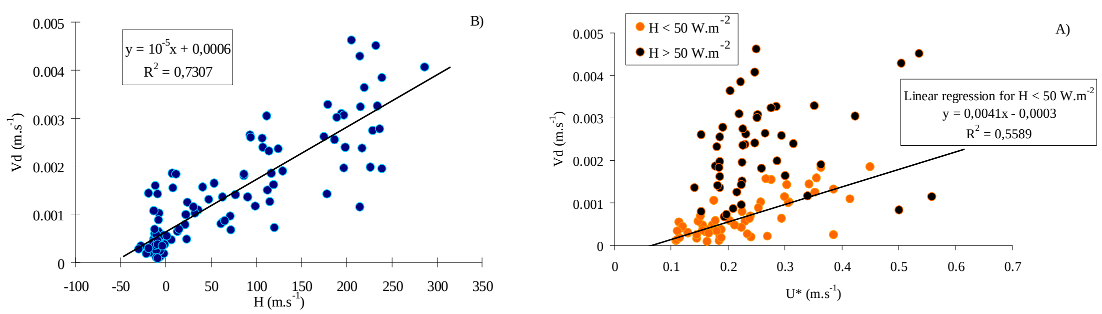

Figure 2: Evolution of Vd in function of the friction velocity (A, on the right) and of the sensible heat flux (B, on the left).
This was obtained for the second stage of the ELPI, i.e. for an aerosol size of 33nm.

The figure on the left (B) shows clearly the effect of the sensible heat flux on the dry deposition velocity values: Vd increases with H and we could suggest a linear evolution of Vd with H but we can't forget that the thermal turbulence (represented by H) and the mechanical turbulence (represented by U*) can influence Vd simultaneously. So, the linear regression plotted on the figure 2 is only an indication of the evolution. The dispersion of the points around the curve is due to experimental uncertainties and to the mechanical effect of U*. The figure on the right (A) shows the effect of the friction velocity U*. The dispersion of the points is more important than for the figure B. In order to dissociate the thermal and the mechanical effect, Vd has been plotted in function of U* for two different scales of values for H: low sensible heat flux (H<50 Wm^{-2}) and high sensible heat flux (>50 Wm^{-2}). This value has been chosen because during the night, the values of H stays smaller than 50 Wm^{-2} and in the day, the values are greater than 50 Wm^{-2}. When the thermal turbulence is low (I.E. H small), Vd is linear with U* and the ratio Vd/U* can be considered as constant. Similar shapes have been observed for the other aerosol sizes but only for the days where the values of H are contrasted between day and night. Another step would be to introduce the Monin-Obukhov length, which characterizes the atmospheric stability, as it has been realized by Wesely *et al.* (1985) and Lamaud *et al.* (1994b).

Impact of aerosol size

For neutral and stable atmospheric conditions, the thermal turbulence is generally low, and in that case, Vd/U* can be considered as constant. So, an average of Vd/U* can be calculated for each range of aerosol size for neutral and stable conditions. On the figure 3, each average of Vd/U* (with its standard deviation) has been plotted in function of the aerodynamic diameter of the aerosol. The values are constant for little sizes (less than 0.1 µm), then, decrease slowly, to increase rapidly for the biggest sizes.

COMPARISON WITH MODELS OF DEPOSITION

Two analytic models, often used for air quality prediction, have been compared with experimental data: Zhang *et al.* (2001) and Slinn (1982). The figure 3 also shows the results obtained with these two models. For these models, a mean friction velocity value of 0.26 $m.s^{-1}$ was taken and the main parameters chosen are d=0.94 m (the displacement height), z_0=0.095 m, h=1.25 m (the height of the canopy), z=6 m and 1/L= -0.006 m^{-1} (L being the Monin-Obukov length).

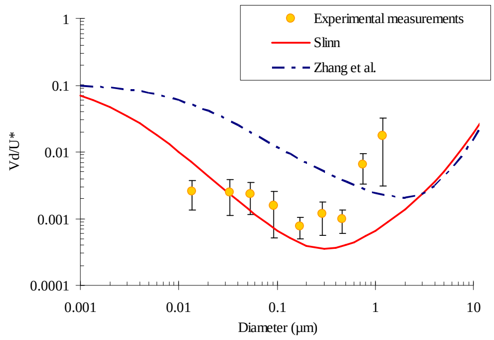

Figure 3: Evolution of the ratio Vd/U* in function of the aerodynamic size of the aerosol for neutral and stable conditions. Comparison between experimental results and the analytic models of Zhang *et al.* and Slinn.

The global shape of the evolution of Vd/U* in function of the aerosol size is similar for the two models: the values decrease and then increase. Nevertheless, there are important differences between the two models: the minimum is not located at similar sizes: 0.2 µm for Slinn and 2 µm for Zhang *et al.* and the values of the minimum for each model are different: practically one order of magnitude.

For sizes smaller than 0.2µm, Slinn is not so far from experimental measurements. But generally there is a difference between experiments and models, especially for Zhang *et al.*: on the one hand, for small sizes, the experimental values stay almost constant (compared to models shape) between 7 nm and 0.1 µm whereas the models values decrease strongly. On the other hand, for the biggest sizes (after 0.5 µm), there is an important difference between experimental and models values (one order of magnitude) and the experimental values increase more rapidly than the models values.

To determine the dry deposition velocity, the models take into account the different mechanical processes that drive deposition: Brownian diffusion, interception, impaction and sedimentation. The figure 4 shows the part of each mechanical process for the two models. The figure 4.A (on the left) represents the evolution of Brownian diffusion and the figure 4.B (on the right) represents the evolution of interception and impaction.

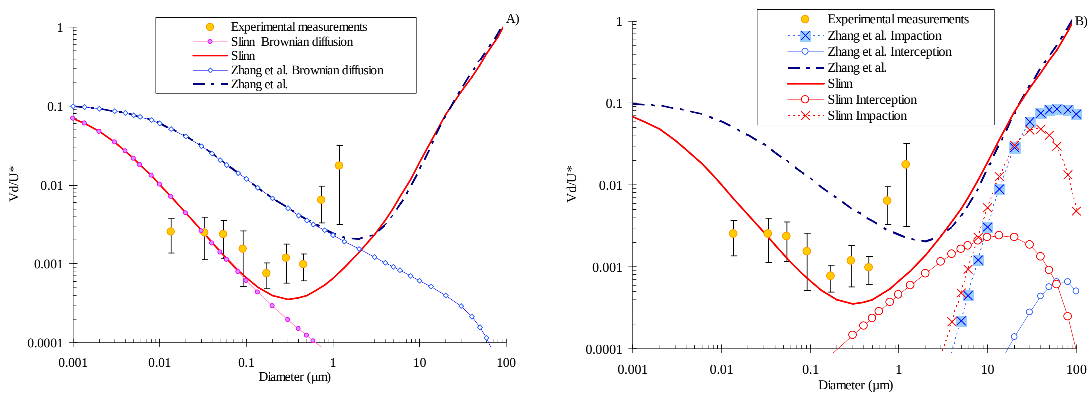

Figure 4: Comparison between experiment and models – Neutral and stable cases
At left (A): zoom on the Brownian diffusion – At right (B): zoom on impaction and interception

The figure 4.A shows that the Zhang *et al*. model overestimates the Brownian diffusion. This is due to the lack of empirical weightings in the efficiency expression of the Zhang *et al*. model. The Slinn model predicts also important values of the dry deposition velocity but only for very small particles (less than 20nm).

Between 0.5 μm and 2μm, the figure 3 shows that there is an important difference between experimental and model values (one order of magnitude) and the experimental values increase more rapidly than the model values. This can mean that the interception and the impaction are underestimated by the models whereas they are the preponderant mechanical processes of deposition in this range of size. The figure 4.B represents each mechanical process for the two models: Slinn and Zhang *et al.* give the same results for the impaction but not for the interception: the Zhang *et al.* interception is a lot smaller than the Slinn interception. Furthermore, the Slinn interception is a little underestimated and avoids the total dry deposition velocity of Slinn to reach the experimental values. For the impaction, even if the two models give the same results, the deposition velocity values due to impaction are not sufficient to reach the experimental tendency for the biggest aerosol sizes.

CONCLUSION

A method based on direct Eddy Covariance with a spectral analysis has been developed (Damay *et al*., 2009) and used to obtain experimental data on aerosol dry deposition velocities onto rural areas covered with maize. The use of an ELPI enables to calculate dry deposition velocities for atmospheric aerosol sizing from 7 nm to 2 μm. The influence of 2 micrometeorological parameters, the sensible heat flux H and the friction velocity U*, on the dry deposition velocity has been shown. In neutral and stable meteorological conditions, the experimental results have been compared with the Slinn model and the Zhang *et al*. model which take into account the principal mechanical processes that drive deposition (Brownian diffusion, interception, impaction and sedimentation). Experimental results are nearer to Slinn results than to Zhang *et al.* results. Nevertheless, compared to the new data obtained in this work, the two models seem to overestimate the Brownian diffusion (especially the Zhang *et al.* model) and to underestimate interception and impaction mechanisms. A sensibility study of the models could be interesting to see the effect of each parameter used in the models because these parameters are linked to measurements realized in wind tunnels (Chamberlain, 1972) and not on real rural areas.

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