

## ***Interactive comment on “Development and validation of a size-resolved particle dry deposition scheme for applications in aerosol transport models” by A. Petroff and L. Zhang***

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Please find below the responses to the comments of the three referees. A revised manuscript is prepared to account for these minor modifications.

Alexandre Petroff

Referee 1.

The present paper aims at improving the representation of dry deposition in aerosol transport model. More specifically, authors propose to improve the modelling of dry deposition over vegetated surface by deriving an improved scheme from a previous

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work from Petroff et al (2008a; 2009) with an updated representation of surface resistance and collection efficiencies. The dry deposition module is also applied to other surface types mainly water surface, deserts-like and snow/ice covered surfaces. This new scheme is based on the resistive approach following the work of Zhang et al (2001; 2003). This latter model that is currently used in aerosol models is compared to the new one and with a large set of available observations. The results show that this new scheme allows a better representation of dry deposition velocities over 2 types of vegetated surfaces. Especially the representation of the amplitude and position of the minimum of these velocities as a function of the particle diameter and surface type seems to be better reproduced. The subject of the paper is interesting. Authors have made huge efforts to present the tools and the underlying theory. The methodology used to derive the new scheme from the detailed scheme of Petroff et al (2008a; 2009) and to evaluate its skills is sound nevertheless the analysis of the relative performances of both models remain maybe too much qualitative. Even if the number of available observations for each surface type is weak a better quantification of the results would greatly improve the paper especially in the case of the vegetated surfaces. Maybe a more clear synthesis of the results is also needed. I think that these aspects would highlight the results of the paper and this way could convince aerosol modellers to use it for their applications. For these reasons, i agree with the publication of this article in the GMD journal with some minor revisions concerning the previous remark. I propose few corrections and /or clarifications to the authors that i hope could improve the paper.

Abstract: I think that the abstract as well as the conclusion needs to be completed with a more explicit quantification of the results.

We agree. A more precise and explicit quantification of the results is included in the revised manuscript, particularly regarding the comparison of the two models.

Section 1 - Introduction: P1318 – line 22-23: It is missing more details about the impact of the dry deposition process (as a sink of aerosols) comparatively to other sinks (especially wet deposition) and their related life time.

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We have added a short discussion on the relative importance in the revised paper (first paragraph of the Introduction)

P1319 – line 09-10: Please give the Vd values obtained for rougher canopies.

A rough estimate of a few tenths of cm.s-1 is given in the revised manuscript for this size range(section 1 paragraph 2)).

P1320 – line 10-11: Could you precise the kind of canopies?

The paragraph dealing with the minimum deposition velocity was confusing and has been rewritten in the revised manuscript. References have been added to studies, where this minimum was identified, ie. on water or single fiber (section 1 paragraph 5).

P1320 – line 12-13: To which kind of surfaces the range of Vd values in the accumulation mode you are mentioning is corresponding?

We are not sure we identified the part of the text you are referring to. Whenever we mentioned value range for the deposition velocity, we tried to make the kind of surface more explicit.

Section 2 - Theoretical considerations

P1321 – line 17: “. . .Paulson (1970); Dyer (1974) . . .”, replace the “;” by “..and..” Corrected in the revised paper.

P1321 – line 19: correction needed for “. . .formulations. . .” Corrected in the revised paper.

P1323 – line 11: correction needed for “. . .example. . .” Corrected in the revised paper

P1324 – line 16-17: you made the assumption that the aerosol was a homogeneous phase with interactions of any kind between particles. Can you argue more about that? Are this processes negligible in your case? If not can we estimate the associated uncertainties?

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We agree that these are strong assumptions that need to be further discussed. Reference to studies that questioned these assumptions have been added to the manuscript (section 2.2 paragraph 2). We are not aware of a way to quantify the associated uncertainties, as these effects strongly depends on the meteorological conditions and the chemical composition of both the gas and the particle phases. Mention of these uncertainties has also been added to the perspectives of the paper.

Section 3 – Results In general, concerning this section, it would have been interesting for each evaluation of the scheme (i.e for different surface type) to clearly explain (recall) what it is expected concerning the discrepancies between both model in light of the different settings that are used. Especially, are we supposed to wait the same results for both for non vegetated surfaces? Why?

A brief section has been added to the first part of the "results", where the main differences between the two models are given (section 3, paragraph 2).

P1333 – line 20: you do not justify the choice of the aerosol density. Moreover, you are using other values in the following. It is maybe details but it would be nice to clarify it.

For consistency, a common value of 1500kg.m-3 is chosen in the applications, water included. It is a value "typical" of the tropospheric aerosol (for example Pöschl 2005). This choice is mentioned in the first paragraph of the "results" section.

P1334 – line 7: my question here is related to the general remark concerning this section. Why you do not present the results obtained with Zhang et al (2001)? Is it too much similar?

It is a negligence. The results of zhang et al's model are added to the figure and the text is modified accordingly. We also noticed there were two inconsistencies between the text and the figure of the original manuscript, related to the roughness length and the reference height: zr taken as 1e-2m while z0 was taken as 4e-5m for the model runs. In the revised manuscript, z0 is taken as 4cm (as in LUC 24-table 2) and zR is taken

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as 1m, so  $z_R \gg z_O$ . This change does not have an impact on the models ( $z_r/z_O$  stays the same), but the measured deposition velocity is recalculated at a higher altitude and decreases for the coarse particles. This explains why there is a better agreement between the model and the measurements.

P1334 – line 12: Explain clearly what is driving the choice of  $z_R$ . The reference height is the altitude within the inertial sub-layer, where the flux is evaluated based on the concentration. It is chosen as a few times of the height of the canopy or the lowest height resolved by the chemical transport model (if the model layer is higher than the canopy height). Details about this choice is added in the revised manuscript (section 2.2 paragraph 6).

P1336 – line 5-11: this paragraph should be clarified maybe just rephrase it. We rephrased this paragraph in the revised manuscript to highlight how the configuration of (Beswick et al., 1991) was different from the model configuration and explains the gap between the model and these measurements (section 3.4 paragraph 2).

Conclusion Cf remarks for the abstract and in general the need of the closer analysis of the results.

The conclusion has been rewritten and the results are more closely analysed.

Referee\_2. Üllar Rannik

The authors are presenting a size-resolved aerosol particle dry deposition model simple enough for application in large-scale numerical models. Clearly, as indicated also in the paper, such a model with sufficient simplicity and capability of capturing most significant features such as dependencies on surface type as well as particle size is currently missing. Additional source of uncertainty in dry deposition modelling to vegetative canopies is dependence on atmospheric stability. The presented model takes into account the influence of stability indirectly through dependence of aerodynamic properties above the canopy. Considering current (limited) knowledge and experimen-

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tal evidence on aerosol dry deposition into vegetative canopies and canopy flows during different stratification conditions the constructed model combines up to date theoretical knowledge with relative simplicity. The ability of the model to represent deposition to ground surface in the form of its asymptotic limit is an additional nice feature of the model. Thus the article deserves certainly publication in GMD but could be improved by considering following suggestions.

The presented model is based on simplification of more complex deposition model into vegetative canopies with the aim to achieve the same descriptive power by introducing coefficient values, that aim to fit the results. Such coefficients and therefore model “versions” are derived for 26 land use classes. A reader being not familiar with global scale modelling deserves short explanation why exactly 26 LUC's were used and/or are there other standards of land use classification in global modelling communities.

Details about land-use categories have been added to the revised manuscript (section 1 last paragraph).

In addition, treatment of urban environment as canopy may look weird (again for those not being familiar with global modelling). Although the authors recognize that such treatment is open to criticism (P1333 line 7-10), it deserves a short explanation of what is meant by “urban trees with LAI 2” and how the concept of GEM model combines emissions (that usually dominate over deposition except at larger vegetative areas such as parks) with modeled deposition.

We agree. We modified the text accordingly (section 2 last paragraph).

P1323 lines 25-26 and eq. (7) P1324. What boundary conditions have resulted in such functional form of the extinction coefficient as a function of stability length above canopy? Presumably different assumptions can lead to different functional form. Either reference or explanation needed.

This model is an extension of what was used in our previous studies and is based on

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Inoue, 63. Details on the derivation of equation (7) and references are added to this section of the revised manuscript (section 2.1 last paragraph).

Figure 1 is little informative in my opinion i.e. different lines do not carry information separately and the same information would be given also by presenting only variation boundaries for each color. Instead, consider presenting deposition velocity size dependence separately for each deposition mechanism and deviation (relative error) between current model and 1D-model also separately for different mechanisms (for some chosen configuration(s) of particular interest). Such presentation would be probably more illustrative and helpful in understanding. Fig. 1 in present form could be summarized in text by a few sentences.

We agree this figure is confusing and not very informative. We followed the referee's suggestion and study the contribution of processes on long grass for different friction velocity. This section highlights the evolution of the process balance.

Referee\_3

The paper is for a size-resolved particle dry deposition scheme for application in large-scale models. Authors propose to improve dry deposition scheme for several land use used in model from Petroff et al. (2008a; 2009). Mostly dry deposition scheme is developed based on vegetated surface. However, this paper show dry deposition velocities of other land types as water surface, desert, and snow/ice covered surface are considered. This approach is important especially in global model. Also, Authors compared with observations limited to evaluate present model. They show that results in this study is more representative than previous model. It is interesting to concern phoretic effect as well as gravity to drift velocity and to assign a constant small value to phoretic effect. For these reasons, I agree with the publication of this paper in the GMD journal with some minor revisions.

Abstract I think that it is need taht you describe explicitly your results in abstract.

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We modified the abstract in order to make our results more explicit.

1. Introduction You compare your results with the result of Zhang et al (2001) in several graph. What do you think the reason why dry deposition developed by Zhang et al (2001) is higher than most earlier models? I do not see what difference between Zhang et al (2001) and your module has. I think you need to clarify this.

Specific details about the differences between zhang formulation and the present model are given in the first paragraph of the "results" section. Briefly, the major reason why Zhang et al. (2001) predicted higher  $V_d$  for fine particles is the choice of Brownian diffusion collection efficiency. This term is a power function of  $Sc$ , a very small change in the power for  $Sc$  makes a large difference in the calculated  $V_d$  for very small particles.

2. Theoretical considerations aerodynamics P1326 line: explain reason using constant as  $5 \times 10^{-5} \text{ m s}^{-1}$  to  $V_{phor}$  to water, ice, and snow surface. Also, if water and ice/snow have different  $V_{phor}$ , how much this affects the change of dry deposition velocity?

The constant assigned to the deposition by phoretic effects is chosen by comparison with experimental results. On water, this value is chosen as  $5 \times 10^{-5} \text{ m/s}$ . This choice is an arbitrary fit and does not correspond to a calculation. Similarly, this value is chosen as  $1.5 \times 10^{-4} \text{ m/s}$  on ice and snow. Following the referee's comment, it appear to the authors that these values are not supposed to be the same. The text of the revised paper is modified accordingly (section 2.2.1 first paragraph).

P1330 eq(23): In general, collection efficiency for Brownian diffusion is used the equation of Wesely. Why this equation is chosen?

Classically, an empirical formulation, initially proposed by Wesely and Hicks (1977), is used for describing the "quasi-laminar" sublayer resistance to transfer by molecular or Brownian diffusion of gas and particles. This "quasi-laminar" resistance is assumed to account for the transfer across all obstacle layers within the canopy (ground, leaves,

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trunks etc...). Thus, it describes the integration of deposition over multiple surfaces. Its form is  $E_b = Sc^{(-2/3)/\alpha}$ , where  $\alpha$  is equal to 1 for Wesely and Hicks (1977) and Padro et al. (1991), while Seinfeld and Pandis (2006, p.908) proposes to use  $\alpha=5$ . Meanwhile, Zhang et al. (2001) proposes to use  $\alpha=1/3$  and a variable exponent of  $Sc$  (close to  $1/2$ ). In the present paper, the equation (24) is used to describe the transport efficiency by Brownian diffusion through the turbulent layer developed on the ground. This expression is preferred to the equation of Wesely, because it is based on theoretical developments and is not adjusted like (Wesely, 1977) on deposition measurements at the canopy scale.

P1331 line 18-19: Do you calculate or approximate  $C_b$ ,  $C_{in}$ ,  $C_{im}$ , and  $C_{it}$ ? justify how to obtain these constants.

These constants are found by fit between the present model and the 1D-model over a very large number of canopy configurations. Typically, different values are tried until a minimum of the relative error is found on the entire size range 1 nm-1000 micron. The paragraph is partially rewritten in the revised manuscript to be more explicit (section 2.2.5 3rd paragraph).

P1333 line 1-5: Are  $z_0/h$  and  $d/h$  sensitive to dry deposition velocity? If these values are sensitive, how can these be applied in 3-d air quality models?

We guess that the reviewer meant that if the dry deposition velocity is sensitive to the choices of  $z_0/h$  and  $d/h$ . Dry deposition velocity is, to some extent, sensitive to these aerodynamic parameters.  $z_0/h$  and  $d/h$  depend on the vegetation amount and vertical profile. It is thus normal that they change with the land cover.

fig 1: I do not know what information it shows.

This figure has been modified following the comment of the second referee, Mr. Rannik.

fig 2: In graph, there is not explanation box for lines. It is better you change shape of line to distinguish between lines.

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We add the legend to the first of these graph to make the reading easier.

3. Results In comparing present results to the results of Zhang et al, it seems that your results generally represent observation data rather than Zhang et al (2001). Why?

There are three reasons for that. - The first is that the present model explicitly account for LAI, while Zhang et al.'s model doesn't, and is thus more sensitive to vegetation change. - The second is that Zhang et al.'s model has been developed in 2001, and there were at the time hardly no measurements of deposition in the Aitken mode. As a result, the fitting of constants in Zhang et al. empirical parameterisation only relies mostly on measurements in the accumulation mode. Now that Aitken mode measurements are more common, the model of Zhang et al. appears to over-estimate them. - The third is that the present model is developed based on a 1D model, where there is hardly no empirical fittings and is thus more trustful.

P1334 line 12, P1335 line 6, and P1336 line 4: justify why  $z_r$  is chosen.

The justification of  $z_r$  is given in the revised manuscript and some information about the sensitivity of the model to this parameter is given (section 2.2 paragraph 6).

You used each  $z_r$  according to land surface. When this dry deposition module is applied to air quality models, do you think  $z_r$  is used as a constant or different values according to land use? values?

To complete the previous answer, it is suggested to use a reference height of 2 times of the canopy heights for vegetation canopy, or  $z_r=10m$  for non-vegetative surfaces.

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Interactive comment on Geosci. Model Dev. Discuss., 3, 1317, 2010.

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