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Interactive comment on "A simple model to estimate exchange rates of nitrogen dioxide between the atmosphere and forests" *by* J. Duyzer et al.

J. Duyzer et al.

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Our paper "A simple model to estimate exchange rates of nitrogen dioxide between the atmosphere and forests" was not well received. In the sequel we react to the comments of each of the four referees that have commented on our manuscript. In this paragraph we would like to make a few general comments in response to all referees and other interested readers.

The goal of our study was to make a simple model and test it on real data. It is our opinion that there is a need for a simple model that: - needs relatively simple input - can be solved simply in such a way that it can be implemented in a large scale model such as the EMEP model - is consistent with the common resistance layer model

In our opinion some of these goals were met. But the referees obviously claim that the

result is not good enough. In response we have some general comments:

Quality of field data Two authors were involved in field experiments specifically aimed at obtaining the data needed to validate such a model. So we are fully aware of the limitations of the field data. It is clear that they are too noisy and generally of limited quality, but such problems are expected when making measurements over forests in relatively clean (low-NOx) conditions. This is made clear in the paper. Having this in mind we did not intent to validate the model but rather to illustrate the model by using real data as input and compare the results with realistic data.

Lack of detail We could easily have expanded the paper with a full description of our experiments but we wonder whether it would really have changed the message or the appreciation of the paper. The experimental setup used in Soroe was similar to that used in Speulderbos. There were of course some minor differences such as heights of inlets etc., but generally the same instruments were used. Some gaps occur in the data and the reasons have not been provided. Of course we have not selected data that fit our purpose. That would have made the comparison with measurements much better and our work much easier. The reason (which we agree should have been given in the paper) was that the model needs all input data to be present, and when one parameter is lacking the model cannot run. In such cases all data from this specific period are omitted.

Better parameterisation We are aware of newer and more sophisticated descriptions of turbulence in forests and they may give better results for some components (e.g. H2O). However, these models are typically far too complex to fit our purpose and most do not allow the incorporation of the effect of chemical reactions on the transport-process. Even where models have been developed for the incoporation of NOx-ozone reactions, they have required numerical solutions which make their implemenation in regional or global scale models rather inpractical(Ganzeveld et al. 2001, J. Geophys. Res., 107, D16, 4297; Ganzeveld+Leileveld, 2004, Geophys. Res. Letters, 31, doi:10.129/2003GL019205).

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We have attempted to present ideas which are more suitable for inclusion in regional and global-scale models. To compensate for the limitations inherent in starting from the big-leaf approach, we carried out a sensitivity analysis on the most important parameters. For example the parameter describing the exchange between the crown level and the trunk space was based on the so-called in canopy resistance described by van Pul and Jacobs (1994). This parameterisation is quite common, and has indeed been found to work well for ozone deposition modelling over forests (e.g. Tuovinen et al., 2001, Water Air and Soil Pollution, Focus: 1, pp 263-274; Tuovinen et al., 2004, Atmos. Environ. 38, 2373-2385), but all such in-canopy resistance formulations are very uncertain (Massman, 2004, Atmos. Env., 38, 2323). Therefore we varied its value by a factor of ten and also varied the dependency of this parameter on atmospheric stability. The differences between the outcome of calculations with the highest and lowest values of this parameter are significant but not dramatic. This leads us to believe that even a more sophisticated description of the exchange coefficients would not have altered the outcome of our calculations.

Raupach and Finnigan's nice paper (Single-layer models of evapotranspiration from plant canopies are incorrect but useful, whereas multilayer models are correct but useless: Dicsuss, Aust. J. Plant Physiol., 1988, 15, 705-716) makes some nice and relevant points about the difficulties of applying complex models within the 'relatively simple' field of latent heat prediction. For ozone the applicability of such models is further hampered by our lack of knowledge of many of the loss processes, such as to the ground vegetation, external vegetation surfaces, moisture films or chemical reactions with other components (e.g. VOCs released by the canopy), as well as the NOx reactions we would like to include. Massman (Atmos. Env., 2004, Vol. 38, pp 2323) has reviewed some of the problems associated with straightforward ozone deposition models, and illustrates the range of resistances found in the literature.

Conclusions We still believe that the model could serve a purpose. It provides a general framework which allows the incorporation of chemical reactions in the common

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resistance layer model. The exchange rates can be parameterised in the way scientists like best and the result can be used as a sub-model in large scale dispersion models. In fact it could even be used as a simple correction factor. That would be vary attractive and an improvement. Even where the influence on atmospheric NOx chemistry may be small (as correctly pointed out by one of the referees) the influence of ozone deposition rates can be significant. To really improve the situation and carry out a true validation would require better quality of field measurements of NO and NO2 fluxes above tall vegetation, and especially for temperate forests. It seems that up to date these measurements do not exist.

Referee #1 Top page S614 OK with respect to Gao et al.

In Duyzer et al. it is shown that the methods given by Kramm lead to unrealistic results outside the area where the simulations are carried out. The reason for this is rather complex but is related to the photo-stationary state which is most likely to occur at larger heights. This is not handled very well in the methods such as those described by Kramm. We are aware of the effect that errors may be associated with the use of gradient transport to describe turbulent transport. But we are dealing here with a 'first concepts' for a correction of the classical description of the deposition process according to the resistance layer model. So a simple, working method may sometimes be preferred over a complex method that is difficult to apply. S616 Unfortunately there is a misunderstanding here. For Rinc we use the value of u* above the canopy. So u* would rather have values between 0.5 and 1.5 m/s in the daytime. It is true that b is not a factor but has a dimension. Van Pul and Jacobs give no explanation for this and we chose to leave it there.

We are not so sure about large stability effects inside the canopy. It seems that the large driving factors such as sunlight are absent in this area. As was stated, u* (at least the square root of at that level) values of less than 0.1 m/s were observed at these levels. So exchange is very small and stability effects may be rather unimportant. In fact we parameterised Rx as a function of stability but this did not improve the results

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nor did it have a large effect. The referee complaints that the reader is left with a factor of 10 uncertainty in Rx. Even this large variation does not lead to a dramatic effect on the results. Therefore we conclude that our results are rather insensitive and robust to the choice of a parameterisation.

Page S616 QUOTE from the referees comments: e) If a model is tested for implementation it should be (1) compared with data have a given absolute accuracy. (2) The data should cover all possible situations which can be observed Yes we would also like to use these data. And we showed only a fraction of our effort to obtain them. To our knowledge there are no such data available.

The statement on the EMEP model is of course correct. Please note that we are far away from using this correction in the EMEP model.

Page S617 top Of course Rx varies with time in accordance with variations of u*. The flux direction changes because of the balance between chemical reactions and exchange rates etc. The model calculates the flux based upon parameters measured in that particular period and nothing else. So in each measurement point a new calculation is made. This leads to the noisy looking course of the NO2 fluxes with time. But this is also what is observed. So that could very easily be realistic.

S617 lower on the page I find this line of arguments rather difficult to follow. Please note that horizontal gradients of O3 are rather small across tens of kilometres. So it is difficult to imagine large advection errors

Referee #2

Page S496: - We disagree with the statement that no concepts are provided. The incorporation of chemical reactions in a resistance layer model is rather new to our belief. - We are aware of more sophisticated models. Unfortunately it is has not been published how these complex models could incorporate second order chemical reaction.

Page S497 - As far as we know all large scale models use a resistance layer approach

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to describe surface exchange. To run the model for each of the two canopies all relevant parameters have been changed of course to describe the exchange in each correctly. This of course within the limits of the resistance layer approach - The use of an in-canopy resistance derived from a maize canopy may sound a bit strange but in the absence of other parameterisation we feel it is justified to use it. Especially because it seems have the right parameters in the equation. Pag S498 - Leaf area indexes for Speuld have been derived by several techniques including destructive ones. Yes, it is a very dark forest. - We did measure NO fluxes using eddy correlation methods. But the uncertainty was rather high because of uncertainty in efficiency of converters etc. and drift. We did measure gradients of NO but did not use them in the absence of fluxes measured by eddy correlation methods. S499 - We chose to use the measured Ozone fluxes because the influence of chemistry on ozone deposition is only small. And we wanted to focus on NOX exchange S500 - Biological uptake is parameterised in the resistance layer model. We fail to see why (or even how) this would need to be improved. - We agree that the parameterisation of in canopy UV light intensity might be weak. But a sensitivity analysis showed that the light intensity below the canopy is not really relevant.

Referee #3 Page S622 We agree with the referee that the agreement with the results of measurements is not really obvious and convincing. We think that this is partly due to instrumental noise. With respect to the interpretation of figure 4 we like to note that there is no discussion whether there is emission of NO from the soil. The emission was observed directly during the experiment. We are sorry to say that in the black and white presentation shown here it is not obvious that the agreement is better. Better graphs would have been more helpful.

Lower on page S622 We agree that Rx is crucial and the scientific basis for the used parameterisation is poor. The literature references given by the referee may indeed give clues for different and better parameterisations. But, and this was not treated in the manuscript, we have changed parameterisations of Rx based on all available

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parameters (u*, stability, LAI etc). This did not lead to significant improvement of the comparison. So it is our belief that we could have improved the looks of the equations but not the comparison with the results of measurements. Therefore we chose the Rinc version which is attractive because it is simple, uses terms which are physical in origin, and is used in many European models. We agree with the analysis on page S623. The effect on atmospheric NO, NO2, O3 chemistry could be small. But, and that was to our surprise,: the effect on O3 deposition was significant. This is especially true during the night. That is a rather new finding that is based upon well-known chemical and physical processes and does not need new processes such as variable cuticular resistances etc. to explain higher than expected nocturnal uptake. This does not mean that we want to state that the latter processes may not occur. Page S624 Yes the description of the equations could have been done better. And that is true for the choice of parameter names as well. We thank the referee for helpful suggestions.

Referee #4

Page S631 We have not used constant flux layer gradient equations as such. We have used the resistance layer model. If the referees claim that this is a violation of rules they are of course correct, but as discussed in the Raupach and Finnigan paper mentioned above there are good reasons for making use of the simple resistance model. Such models are in fact in widespread use in air pollution models dealing with reactive compounds like O3, NO, NO2, NH3, NH4, NO3, HNO3. So yes, it is wrong but we are in good (at least large) company. What is added to the classical resistance layer approach is transport directly from the crown level to the trunk space. That is a very large simplification indeed. But it is attractive to put it in a resistance layer model because that is the framework that is currently used.

Interactive comment on Biogeosciences Discussions, 2, 1033, 2005.