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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.

Anonymous Referee #3

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The paper presents a simple analytical two-layer resistance model to describe the soil-vegetation-atmosphere exchange of NO, NO2, and ozone for forest ecosystems. In addition to commonly applied two-layer resistance schemes for deposition modeling (Wesely, 1989, Atmos. Environ., 23, 1293-1304), the authors introduced NO-NO2-O3-chemistry in the lower canopy or trunk layer. The strongly reduced photolysis of NO2 within the canopy compared to above favors the net production of NO2 from NO and ozone. As simulated earlier by multi-layer chemistry models (e.g. Gao et al., 1993, JGR, 98, 18'339-18'353) this effect can lead to an upward flux of NO2 and a downward flux of NO at the canopy top despite a NO emission at the soil surface and a NO2 uptake of the leaves. The aim of the study is to show that the simple analytical approach is able to reproduce this effect to a reasonable degree, so that it could

be used to improve the surface exchange parameterization of atmospheric chemistry-transport models like EMEP. For this purpose model simulations are compared to field measurements (several days) of two forest sites.

GENERAL COMMENTS

In my opinion, the type and quality of the presented figures are not sufficient even for a rough verification of the model approach. In Figs. 5, 6, and 8, I can see no systematic agreement between observations and model calculations of the NO2 flux (although both show positive and negative values). If the agreement with field data is not really obvious like in the present case, it has to be supported by quantitative statistical evaluations (e.g. regression, correlation analysis). If this is not possible, e.g. due to a large uncertainty of the field data, a comparison/verification is not meaningful. The variability of the field measurements (including the difference between the two measurement levels above the canopy) is interpreted by the authors as "noise" (Section 4.1). It is argued that the flux difference of the two heights cannot be explained by chemical flux divergence. However, since the difference partly looks more systematic than random-like, it should also be discussed whether or not it could have been caused by measurement/instrumental problems or by advection effects. I can also not agree with the interpretation of Fig. 4 in p.1045,line 12-17. It is argued that the simulation with reduced R_x agrees better with the observations than the simulation with zero soil emission. However, based on the displayed data in Fig. 4, I more tend to the opposite conclusion.

As stated by the authors, the most uncertain parameter of the simple model is R_x describing the turbulent exchange within the canopy. Unfortunately it is probably the most crucial parameter concerning the bi-directional NO2 exchange, because the chemistry effect mainly depends on the residence time of the air in the trunk-layer. Considering the importance of R_x, the scientific basis and the discussion of its influence and uncertainty is too poor in the manuscript and needs to be improved significantly. In addition to the cited references there exist a number of other studies about the topic e.g.

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Van den Hurk and McNaughton (1995, J. Hydrology, 166, 293-311) or Nemitz et al. (2001, QJRMS, 127, 815-833). The former included the near-field dispersion concept after Raupach (1989, QJRMS, 115, 609-632) in the parameterization of the in-canopy transfer. For unstable conditions, studies of the canopy exchange time scale (roughly equal to h_{can}*[R_a+R_x]) derived from the analysis of coherent structures (e.g. Paw U et al., 1992, Agric. For. Met., 61, 55-68; Zelger et al., 1997, Atmos. Environ., 31,217-227) may also provide useful data for a parameterization.

In the motivation and discussion of the paper, it should be clearly distinguished between true deposition of ozone (destruction on/in leafs and soil) and the apparent deposition by reaction with NO to NO2. The same holds for the true emission of NO from the forest soil and deposition of NO2 to leaf and soil surfaces and the apparent emission and deposition fluxes at the canopy top. Obviously there is a cycling process of NO rich air entering the canopy where there is a net conversion to NO2 which again leaves the canopy with a high probability (because surface deposition of NO2 is slow). Once the increased NO2 concentrations is exposed again to the high radiation levels above the canopy, it will be rapidly reduced to the old equilibrium that existed before the air parcel entered the canopy. This NO-NO2 cycling also implies a cycling of ozone of the same amount (ozone entering the canopy where it is converted to NO2, leaving the canopy as NO2 and being photolysed back to ozone. Since the NO-NO2-O3 chemistry is relatively fast, large scale models like EMEP might not be very sensitive to the actual NO/NO2-partitioning but only to the net NOx exchange with the canopy and thus to the “true” surface emission and deposition processes that are already included as common bulk parameterizations.

The requirement of a strongly simplified model approach is usually that it does not have to be very accurate for a specific case but that it has to cover the main variability effects of most plausible cases. I admit that this is difficult due to the limited number and uncertainty of available field measurements. However, I would expect that at least the sensitivity of the modeled NO2 flux to variable environmental conditions (ratios of

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NO, NO2, and O3 ambient concentration, LAI, canopy height, leaf and soil surface resistances) within plausible ranges for European forests is analyzed and discussed.

Due to the described major shortcomings of the manuscript I cannot recommend it for publication in Biogeosciences.

SPECIFIC AND TECHNICAL COMMENTS

The equations are not numbered which unnecessarily complicates the presentation of the model calculations and parameter variations. In addition the choice of the displayed equations in the main text (chapter 2) and in the appendix seems somewhat arbitrary. Some equations are displayed twice (main text and appendix) and some parameters are not clearly defined. For example, the value of the parameter b in p.1040,line 10 is only defined by the respective equation in p.1050,line 17 (without any reference between the two equations). A more systematic presentation of the model equations and the parameter definitions is necessary.

There are some inconsistencies in the resistance network as presented in Fig. 1, described in the main text and in Appendix A: 1) The leaf surface resistance of the upper part of the canopy should not be denoted as "R_c" as it is done in the manuscript. It only represents a sub-component of the bulk resistance of the entire canopy that is commonly denoted as R_c in the literature (e.g. Wesely, 1989, Atmos. Environ., 23, 1293-1304). Therefore a different symbol like R_{lcrown} (in analogy to R_{lus}) should be used. 2) The symbol C_{leaf} in Fig.1 is misleading because it does not represent a leaf surface concentration (the latter would be below R_b) but rather a crown-layer concentration. Thus a different notation like C_{crown} in analogy to C_{trunk} would be more appropriate. 3) In Fig. 1 and in the main text, only an uptake by leaves in the understorey (R_{lus}) is mentioned whereas in the Appendix also a soil resistance (R_{soil}) is introduced in the equations. The parameterization and importance of both resistances should be discussed, because they are very important for the canopy reduction effect for soil emitted NO.

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p.1036, line 1: "The structure of the model is strongly related ..."

p.1036, line 4: "...and compare the results of model calculations with ..."

p.1036, line 9f.: reformulate sentence

p.1036, line 23-25 (incl. footnote 1): I agree that chemical reactions of VOC and ozone may be neglected in this context. However, more important for the NO2 flux is the reaction of NO with peroxy radicals RO2. According to Gao et al. (1993, JGR, 98, 18'339-18'353) it can be as effective as the reaction with ozone (at least in sunlight conditions). This should be discussed in more detail.

p.1043, line 12: the general downward direction of the NO2 flux is not a result of a simple resistance model (without in-canopy chemistry) but a pre-assumption!

p.1043, line 15: discuss this statement in more detail (see also general comments)

p.1045, line 24: "In other words: R_x = ..."

p.1049, line 19: "...+ k_1*h_{trunk}..."

p.1050, line 17: The definition of R_{inc} does not have the right units for a resistance (the same applies to p.1045, line 24)

p.1051, line 4: "...the uptake of O3 and NO2 by the under-storey ..."

All Figures: the figure captions are too short. Figures with captions should be self-explaining to a reasonable extent.

Figs. 2,5-9: do not plot the axis labels inside the diagram frame where they overlap with the plotted data!

Fig. 2: The second y-axis for the soil NO flux is not helpful. Better use the same y-scale for NO and NO2 fluxes (allowing a direct quantitative comparison) and, if necessary, a different scale or panel for the ozone fluxes.

Figs. 3 and 4: The figure captions are wrongly assigned.

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Interactive Comment

Figs. 3 and 4: The measurement height given in the figure caption and displayed in the symbol legend do not agree.

Figs. 3,4,10,11: remove diagram titles

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