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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 #1

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

The paper describes a two layer mechanistic model approach to calculate the exchange of reactive trace gases (NO, NO_2, O_3) between the soil, the forest and the atmosphere. Chemical reactions in this three component system are considered for the purpose to estimate the observed direction and magnitude of the NO_2 - flux above two forest canopies. Two short series of measurements are compared to model results. It is discussed which one of the model parameter has to be selected in a certain way to approach the measured fluxes and mixing ratios. This is done to justify that this simple model may be implemented in the EMEP modul.

Technical comments:
There are some printing errors in the manuscript:

- page 1035: line 13, the author is Lenschow
- page 1040: line 27, a point behind "temperature" is missing
- page 1046: line 24, a point behind "Dorsey et al. (2004)" is missing
- page 1056: there is no information available which period is shown. Is it 22-06-03 to 04-07-93?
- page 1057 and 1058: The notation on x-axis is for "June and July 1993", the title gives "May - June 1993" and the cited literature (e.g. Dorsey et al., 2004) gives 19-06-93 to 02-07-93 and 20-06-93 to 02-07-93 (Duyzer et al., 2004) for the duration of the experiment as also earlier reported during a BIATEX - workshop.
- Fig. 2 to Fig. 9 have captions which should be renewed because they are not simply readable.
- There may be false printings in the data if Fig. 3 and Fig. 4 are compared to Fig. 1 and Fig. 3 from Duyzer et al. (2004) or did you find differences of some ppb over about 10m above the canopy?
- Comparing with Dorsey et al. (2004), one can conclude that Fig.2, Fig.7, Fig.5 and Fig.6 describe measurements from 24-06-93 to 29-06-93. If this is true the reader should be informed in detail in the text on the reasons for significant flux divergence for the NO_2-flux above the canopy.

To summarize these findings, the authors should carefully renew the graphical presentation of their results.

BGD
2, S612–S619, 2005

General comments:

a) The authors mention (page 1035, line 20) that “detailed models have been constructed, which are able to explain some of the phenomena observed above forests (e.g. Gao et al., 1991; Duyzer et al., 1995).” Giving Gao et al. (1991) as reference is misleading because this paper describes exchange processes above low vegetation. The authors potentially have the paper of Gao et al. (1993), J. Geophys. Res. 98, 18339 - 18353 in mind, which discusses turbulent diffusion and chemical reactions in and above forests.

b) The authors mention (page 1035, line 14) that the properties of the canopy flow are even more complex for a forest than for low vegetation where procedures to correct for the influences of chemical reactions on fluxes are described in literature (they give: Kramm et al., 1991; Gao et al., 1991). G. Kramm and R. Dlugi, (1994); J. Atmos. Chem. 18, 319 - 157 showed that over low vegetation different turbulence parameterisations did not change significantly their results and could approach results of Gao et al. (1991). They applied a first order closure model, and, therefore, could relate their fluxes directly to resistances and local gradients of mixing ratios as also published by Kramm et al. (1995); Atmos. Environ 29, 3209 - 3231. The application of a simplified version of such models to dry deposition of these chemical compounds is decribed, for example, by Spindler et al., (1996), Meteorol. Zeitschrift (NF)5, 205 - 220 (article in English language), and one may get some idea on the accuracy with which fluxes of these compounds can be calculated for grassland.

c) In and above tall vegetation the application of resistance models (which are equivalent to a first order closure formulation) need significant local modifications especially for the determination of fluxes of sensible heat, water vapour or trace compounds like CO_2 (see for example: A.S. Thom et al., (1975): Quart. J. Roy. Meteorol. Soc. 101, 93 - 105 or M.R. Raupach et al., (1996), Boundary Layer Meteorol. 78, 351 - 382; J.

BGD

2, S612–S619, 2005

Graefe (2004):, Agr. For. Met. 124, 237 - 251; J.C. Kaimal and J.J. Finnigan (1994): Atmospheric Boundary Layer Flows: Their Structure and Measurement, Oxford University Press, N.Y., 189pp; G.G. Katul, J.D. Albertson (1999): J. Geophys.Res. 104, 6081 - 6091; R. Leuning (2000): Boundary Layer Meteorol 96, 293 - 314).

Therefore, a considerable number of studies refer to the application of inverse Lagrangian dispersion theory (ILDT) for the estimation of the source/sink strength of sensible heat, water vapour, carbon dioxide or methane and their fluxes at canopy top. This is performed in a way that characteristics of a forest (e.g.: canopy height; projected leaf area index; plant density; vertical leaf area index profile; plant type) are used to estimate turbulent parameter to determine the near and far field diffusivity inside and above the canopy. The authors of this paper replace this concept by an estimation of the first order turbulent exchange coefficient K_M for momentum. They apply K_M (in the vertical integrated form as inverse turbulent resistance inside and above the canopy to the transport of trace gases. Compared to ILDT this method considers only the trace of the diffusion matrix and is proven to be an insufficient description of the turbulent transport inside canopies. Therefore, the flux of heat, water vapour or CO_2 at canopy top may have an unknown systematic error of order of a factor of about $\pm(2\ to\ 4)$ if calculated this way. In general trace substances are considered to be transported like heat or water vapour. It seems that Duyzer et al. chose another concept. How to justify this?

d) The determination of fluxes of reactive compounds from the chemical triad NO-NO_2-O_3, requires the knowledge of various quantities. The authors state that they had only to adapt a resistance $R_x = R_{inc}/\beta$ (page 1040, line 15 - 18, page 1045, line 12 - 17 and line 18 - 20) by varying β between $\beta = 1$ to $\beta = 10$ to force an approximate agreement for the two forests they had data to compare with model results. R_x describes the turbulent transport in the trunk space. Taking characteristics from Speulderbos and the equation from page 1045 (line 24) this results in

BGD
2, S612–S619, 2005

$R_{inc} = 1.4 * 10m^2/m^2 * 22.2m/u* \approx 310,8m/u_*$. For u_* in the canopy the authors estimate “only a few percent of its value above the canopy” (page 1040, line 4 - 5) but give no data. Therefore taking simply $u_* = 0.01ms^{-1}$ to $u_* = 0.1ms^{-1}$ gives $R_{inc} \approx 31080s$ and $R_{inc} \approx 3108s$. At first, R_{inc} and R_x have the dimension of a time (s) but not a resistance (sm^{-1}). Therefore factor b (page 1040, line 10) has a dimension of m^{-1}, which is not given but mentioned in W.A.J van Pul and A.F.G. Jacobs (1994). So b is no “factor”, but an “inverse length”. This is one example of many where the authors stay with inaccurate formulations. In addition, the reader has to read all cited paper in detail to try to understand what is really presented in this paper. At second, dividing R_{inc} by 10 leads to $3108s$ (or $3108sm^{-1}$) and $310.8s$ (or $310.8sm^{-1}$) leaving the reader with a factor of 10 (or more) variability. For example, it is known that R_{inc} should also vary significantly with stability chances especially if vertical mixing is surpressed, as one can assume in a forrest with $LAI > 8$. So what is the meaning of b and β for a certain canopy? The authors should be aware that they plan to implement this diagnostic tool (model) into the EMEP model all over Europe, but actually stay with fitting b and β to approach fluxes from local experiments! How are b and β and other parameters predicted for each grid point ?

e) If a model is tested for implementation it should be
(1) compared to data which have a given absolute accuracy.
(2) The data should cover all possible situations which can be observed.
Such a procedure is sometimes called validation and is the necessary requirement to apply a model especially for environmental regulation purposes, as done with the EMEP model. The facts mentioned in d) leave the readers with a parameter variation to approach flux divergences obtain from a study of only some days at Speulderbos. These data, if carefully considered, show as well upward as downward NO_2 fluxes above the canopy with and without a flux divergence. In addition data gaps are obvious for the $25m$ height. During daytime a strong variability in the magnitude and direction of the NO_2-fluxes is visible. Therefore, from the model design R_x is supposed to

BGD
2, S612–S619, 2005

EGU

vary with time. This variability cannot be only inversely proportional to u_* because also the flux direction changes. There are data where at $25m$ height an emission is observed with deposition at $35m$ height and at the next time step a reverse situation. The authors should describe in detail how this behaviour is handled within their model.

f) Considering (1) there is no reference which describes the absolute accuracy of the fluxes being determined. In addition, the assumption is made, that advection has no influence on the fluxes in such a heteroge- neous landscape (the tree height varies at least between $12m - 20m$ in a distance of about $200m - 300m$ to the West and South sector with some clearings and a small road). The authors should show that advection can be neglected. Otherwise the observed flux divergences could be simply estimated by the influence of horizontal and vertical advective transport or/and instationarity inside and above the canopy. The following example may give examples for inconsistent descriptions in the text. For $R_{inc} \simeq 310sm^{-1}$, the vertical transport velocity in the trunk space is about $0.003ms^{-1}$ if $u_* = 0.01ms^{-1}$. Thus it needs about $1h$ to transport a compound release from the ground vertically through the trunk space. In the same time the horizontal transport is over a distance of about $180m$ or more below $10m$ height. The authors mention in their cited references mixing ratios of about $1 - 6ppb$ for NO and about $40 - 45ppb$ for O_3 in the trunk space. A simple calculation (see below) shows that no NO should be present after 1h for transport to the canopy top.

The cited data also show a vertical flux divergence up to about $0.001ppbs^{-1}$ for the ozone flux between $h = 25m$ and $h = 35m$. This is proportional to the storage term of $36ppb/h$ before noon, a rate which cannot be achieved by chemical production but only advective (horizontal and vertical) transport. If ozone is advected, why not NO_x ? As mentioned, the NO_2 - flux (see: Dorsey et al., 2004) near the canopy ($h = 25m$) is sometimes negative while at $h = 35m$ can be positive (emission of NO_2). Therefore a negative flux (deposition) turns into a positive flux (emission) with a maximum divergence of about $0.007ppbs^{-1}$ over a distance of $10m$ during daytime (noon) conditions.

BGD
2, S612–S619, 2005

A mean divergence might be about $0.001 ppbs^{-1}$. Let us assume $u_H = 2ms^{-1}$ and a low value of $u_* = 0.3ms^{-1}$. Then the exchange coefficient is at least $0.5ms^{-1}$ and the vertical travel time is less than about $20s$ over the distance of $10m$. The chemical reaction $O_3 + NO$ yields about
$0.00042ppb^{-1}\ s^{-1} * 70ppb(O_3) * 1ppb(NO) = 0.0294ppb\ s^{-1} NO_2$ for $1ppb\ NO$ (in $20s$ one obtains about $0.59ppb\ NO_2$).

Therefore, we add about 10% to 20% to the mean level of NO_2-mixing ratio. The maximum divergence observed is only about 25% of the divergence being possible by the source reaction. The sink reaction (photolysis) constant is below about $5 * 10^{-3} s^{-1}$ at maximum and destroys less than about $0.1ppb\ NO_2$ during $20s$. Unfortunately, no data are given in this paper and the cited references for the actual photolysis rates. Also there are no data or given references for u_* or the single terms of the energy balance or other parameter being necessary to understand the dynamic and energetic conditions which control exchange and chemistry at both field sites. For the mean divergence about $0.08ppb\ NO_2$ are added during $20s$ to the mean mixing ratio of about $2 - 5ppb\ NO_2$ above the canopy. It is reported that the NO - mixing ratio in the trunk space is between $1 - 6ppbv$ (Dorsey et al., 2004). What happens to the remaining NO if only $1ppb$ can react?

In summary, the presentation of the model and, especially any comparison to measurements is performed in a way that common requirements to a validation procedure are not fulfilled. Neither the complete micrometeoro- logical and meteorological conditions during the measurements nor the detailed data and the concept to parameterize and calculate fluxes are given. Every argument seems to be plausible at a first glance, but rather incomplete proofs are given and essential details are missing. Even the presentation of results (figures) is performed in a way which do not fulfil the requirements of Biogeosciences (BG). Therefore, the paper needs essential additions and major revisions.

BGD
2, S612–S619, 2005

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