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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Technical Note: Anthropogenic and natural offline emissions and the online EMissions and dry DEPosition submodel EMDEP of the Modular Earth Submodel system (MESSy)" *by* L. N. Ganzeveld et al.

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Dear reviewer,

Thank you for the feedback on our manuscript submitted as a technical note to ACPD in 2006. Based on your and the other reviewers comment's as well as discussions with the co-authors and editor, we have changed the set-up of the manuscript such that it is now resubmitted to ACP as a regular manuscript instead of a technical note. This, and having repeated some of the model simulations, also explains the rather long time period in between the publication in ACPD and the resubmission of this revised version of the paper.

„The authors introduce a MESSy subroutine EMDEP, which calculates/organises emissions and deposition of chemical species and present results from 1-year integration. In general I do appreciate a detailed documentation of model systems. However in this case I have the feeling that a duplication of work is performed. The two papers by Kerkweg et al. (same special issue) describe in more detail the routines OFFLEM, ONLEM, and DRYDEP and are basically the same as in EMDEP. And unless I missed important differences between these routines, I would recommend not to publish the methodology (= Sec. 3) twice.“

To adress this issue we have substantially revised the manuscript and indicated the differences of EMDEP/DRYDEP and ONLEM. ONLEM is a comprehensive submodel that combines the former two and includes additional process details and options to be used in the development of process parameterizations. The publications by Kerkweg et al. (2006a,b) provide technical descriptions only, and the present publication rather describes some of the scientific issues involved, including important options relevant for the development of parameterizations.

We have furthermore introduced paragraphs to clearly indicate the link between the various emission and deposition submodels, e.g., in the beginning of Section 3:

EMDEP signifies the representation of online simulations of EMissions and dry DEPosition of gases and aerosols and the coupling of these two processes relevant to atmosphere-biosphere exchanges (including in-canopy chemical transformations and turbulent exchange), based on earlier work by Ganzeveld et al, 1995, 1998, 2002. Because of the large number of various model components and the complicated structure of EMDEP, two alternative submodels have been developed to calculate online emission and dry deposition fluxes based on the concepts applied in EMDEP (more details about these submodels, ONLEM and DRYDEP, can be found in Kerkweg et al., 2006a,b). Here we focus on describing specific modifications with respect to the representation of online emission processes in EMDEP compared to the earlier work as well as the sensitivity of these online emissions to the spatial resolution of the driver

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model ECHAM5. Note that EMDEP is also a prime tool for the further development of process descriptions and parameterizations, for example by applying a single column version of the GCM (Ganzeveld et al., 2002a).

Hopefully this will remove the confusion concerning the availability of various submodels for similar processes in MESSy. To further prevent duplication of the description of model components we have also removed some more technical issues, e.g., the description of the calculation gaseous and aerosol dry deposition, described in section 3.2, where from the included text above it can be inferred that more technical details, i.e., the actual code implementation, can be found in the Kerkweg et al. 2006 manuscripts.

„On the other hand the description of the implemented emission data set is not given in Kerkweg et al. so that I would recommend that Ganzeveld et al. concentrate on this point in combination with an analysis of the impact of emission heights and an analysis of calculated budgets. However, the analysis performed so far is quite hand-waving and lacks a detailed investigation. Therefore, I think the paper can be a valuable contribution only after major revisions, which include a change in the focus.“

A more detailed analysis of the sensitivity of the simulated atmospheric chemistry, in particular the boundary layer concentrations associated with changes in the assumptions on emission heights has been included. This is also expressed by the new title: *Sensitivity of atmospheric chemistry and surface deposition to emissions in the ECHAM5/MESSy1 model.* We realize that the presented sensitivity analysis could be further extended introducing more parameters (e.g., wet deposition, boundary layer entrainment, etc.). However, considering that there is still so little information available on how to realistically introduce effective emissions heights in these large scale models limits a more quantitive assessment of the impact of these assumptions on emission heights on atmospheric chemistry. The presented sensitivity analysis intends to stress that this specific feature of the respresentation of the emission processes deserves a

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large priority in further model development and evaluation.

Major comments:

1. Difference EMDEP $<->$ (EMDEP and OFFLEM, ONLEM) unclear. As far as I understand from the text EMDEP is a routine including various processes. In a further step processes were separated/extracted from EMDEP and OFFLEM, ONLEM DRYDEP routines developed. The following question should be answered to better understand the relations:

(a) Is EMDEP obsolete, because it can be substituted by the updated subroutines OFFLEM, ONLEM and DRYDEP? In that case I would not recommend the publication!

See also the reply above and modifications introduced in Section 3; EMDEP is not obsolete. The submodels ONLEM and DRYDEP will not change in future, whereas EMDEP will be the choice for new developments and additional features. Note for example that EMDEP has been extended by the introduction of a coupled approach of emissions and dry deposition using a multi-layer exchanges model (Ganzeveld et al., 2002a,b), which will become available to the MESSy users in new releases. Furthermore, EMDEP will be updated including the results of specific analyses focusing on surface exchanges such as the recently published work on snow-ice ozone exchanges (Helmig et al., ACP, 2006).

(b) Is the methodology different in the approaches EMDEP vs. (OFFLEM, ONLEM, DRYDEP)? If not, the methodology should only be published once either in a paper on EMDEP or in OFFLEM, ONLEM DRYDEP papers.

The submodel OFFLEM only processes (reading and assigning boundary conditions, i.e, emission fluxes) prescribed input fields, such as the offline emission inventory being described in our technical note, to constrain MESSy simulations. It does not include a specific emission inventory, such as the one described in our manuscript. We have

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modified some of the text to explain this more clearly:

An important feature of the offline emission inventory is the application of source type dependent emission height profiles. These are also used in the MESSy submodel OFFLEM (submodel for process prescribed boundary conditions, see Kerkweg et al., 2006a) to assign the read-in emission fluxes, e.g., of the inventory presented in Section 2, to the model layers closest to the emission heights.

ONLEM and DRYDEP are benchmark submodels to consider online emissions and dry deposition without considering their potential direct interactions. There are many similarities between the three submodules, with one essential similarity being the concepts based to describe the emission (NOx and VOC's) and dry deposition processes but there are especially differences concerning the interfaces. Having modified the explanation of these similarities and differences as well the removal of the detailed description of the gaseous and aerosol dry deposition, we hope that this issue is resolved.

2. OFFLEM: Emissions: Original work in emissions unclear. As far as I understand from the text from 5460/10 to 5461/8 is a reference of existing work. 5461/9-5461/17 describes how the speciation is done, based on previous work. The rest of this section is a summary, which datasets were put together.

(a) It should be made clearer what is new, i.e. what part is not 'simply' copied from other emission datasets.

We have described in our ms the compilation of an emission dataset that we currently apply in a default setup of MESSy to conduct present-day atmospheric chemistry studies. It's various components have been described elsewhere, also indicated by the various references to the original work, Olivier et al, Van Aardenne et al and Van der Werf et al. However, the consistent coupling of these various emission sources including the EDGAR-FT2000 dataset for anthropogenic emissions, including the biomass burning, complemented with the biogenic emissions from MATCH provides a crucial component of MESSy and consequently needs to be properly documented. There-

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fore we have included this description of the inventory especially stressing some of the key features of the inventory also relevant to presented sensitivity analysis, e.g., the emission height table and the main differences between the v3.2 and v3.2-FT-2000 inventory.

(b) It should be emphasised how the authors decide which data set they take. Is there some sort of philosophy behind? EDGAR covers only anthropogenic emissions? That should be made clear to avoid the impression that emission data were picked arbitrarily.

The motivation to take the various sources for the anthropogenic, biogenic and natural emissions that are considered in the offline emission dataset is based on a selection of state-of-the-art emission representations available within our group and extensively tested, such as the biogenic emissions from the MATCH model, which were included in an extensive evaluation of this model, as well as our participation to the further development of the EDGAR dataset (in a joint effort of the JRC(I), MNP (NL) and MPI-C). We admit that this selection includes arbitrary elements, but we consider it of utmost importance that the different components of the emission inventory have been checked for consistency within our own group. By being directly involved in all aspects of the development of emission inventories and their implementation in the model we have removed many inconsistencies associated with the use of model parameters and the assignment of emission categories. We have developed a number of budgeting routines that are applied after each change in the emission dataset to ensure consistency and avoid double counting.

Note that the EDGARv3.2-FT2000 includes biomass burning emissions based on the Van der Werf et al., 2003 inventory.

(c) A lot of detailed information is given on the datasets. A table would be helpful which includes source types, species, methods applied (if), and references.

See also our reply to comment (a); we have included various references that provide

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such details to arrive at the global annual emission fluxes of all species and decided to limit the presentation of the offline emission inventory to global annual emission fluxes as they have been applied in the extensive evaluation of the ECHAM5/MESSy and the key features relevant to the sensitivity analysis presented in this ms.

(d) Kerkweg et al. gave information on how the input data in OFFLEM are processed in more detail. Is EMDEP working the same way?

EMDEP only deals with online emissions and dry deposition whereas OFFLEM is a submodel only to process preprocessed boundary conditions to the model.

3. ONLEM: The discussion of the treating of online emissions is identical to Kerkweg et al., except that Ganzeveld et al. present it more verbally, without describing the algorithms but simply referring to previous papers, and Kerkweg actually present the formulas, an approach which I support and which I think is the right way for a Technical Note. I suggest to remove this part and refer to Kerkweg et al. With clear indications whenever the method deviates.

See also previous replies; we limit the discussion on the online emissions to what is especially different compared to the implementation of the online emissions in the ECHAM4 model (Ganzeveld et al., 2002b), and have removed the more technical description of the dry deposition process and the ms is resubmitted as an „normal“ paper.

4. DRYDEP: Same as above. Here we have a verbal description of what Kerkweg et al. (p 6856) describes in a more technical way. Both refer to the same basic papers by Ganzeveld and Lelieveld (1995) and Ganzeveld et al., (1998). So why duplicating it?

Duplication; see previous replies and the removed description of the dry deposition discussion.

5. The design of the numerical experimental lacks information. Is ECHAM run in a nudged mode or not? If nudged how is it done, which impact does it have on

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the boundary layer?

We have modified the description of the how the experiments have been done (see Section 4), including a statement that these simulations reflect runs with ECHAM5/MESSy without nudging the model, and that the meteorology of the two sensitivity runs is exactly the same since we have ignored the coupling between the atmospheric chemistry and radiative forcing. See als Section 4.2;

Note that this sensitivity analysis reflects solely the impact of different emission heights on chemistry since the simulated meteorological conditions in these simulations are identical.

6. The discussion of the results, especially concerning the resolution leaves a lot of question open. Especially the analysis of the impact of the resolution on the emissions is not accurately performed.

This comment was initially not very clear; however, also based on the other comments (especially on the VOC emissions, see below), indicates that the reviewer requires a more detailed analysis of the role of the differences in emissions due to resolution versus temporal variability. This point is therefore addressed in more detail below.

Minor comments:

5459 / 10: I do not understand the phrasing 'testing model' what exactly will be tested? And how can one decide, which subroutine is giving the correct answer to allow testing?

Testing model has been applied here to indicate that EMDEP is a submodel which is continuously further developed, including new processes but also sometimes recoding components of the model and which require extensive tests to secure the application of these subroutines in MESSy. It is used to stress the difference with ONLEM and DRYDEP that are developed to provide „benchmark" model representations of online emissions and dry deposition. We have removed the term testing model.

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5467/18: Is 1 year integration and climatologically integration not a antagonism?

You are right; we wanted to indicate that it is a free, non-nudged simulation that has been presented. With the substantial change in the structure of the paper, the explanation of the model integrations has been modified and hope it now properly explains how the integrations have be done.

5467/27: Why is it a consequence?

The consequence of having established that apparently the use of a maximum LAI of 10 does reflect observed maximum LAI's. However, he sentence has been changed.

5468/3: use reduced instead of corrected.

Has been modified

5468/4: Where is the increase in the 'simulated fraction Eą ' coming from? How do the authors know that there is compensation? Did they run extra simulations? Does it directly follow from some equations? This has to be explained in more detail.

This and following point really require a quite detailed analysis of the representation of the VOC emissions in ECHAM5/MESSy. Since the aim of our paper was to indicate some of the main features of the general representation of the emissions and deposition as well as their in role in atmospheric chemistry in ECHAM5/MESSy and not so much quite specifically the VOC emissions we did not include such specific information. However, we do provide some more details here and have introduced some additional modifications in the paper.

The point about the compensating effect is well taken. To address this we have performed additional tests with a column version of the model for tropical forests (see Kuhn et al., ACPD, 2007) including simulations for an LAI of 10 and 6 with the same canopy radiation algorithm implemented in EMDEP. These simulations show an up to $\sim$8% increase in the simulated fraction of sunlit leaves. We have underlined the term

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simulated here since it should be carefully checked if this is a real feature or more a numerical feature where we are aware that with the large radiation gradients in the top of the canopy, these simulations are sensitive to the vertical discretization used for the calculation of the canopy radiation regime.

We have added a extra sentence: *Sensitivity analysis with the canopy-radiation algorithm of EMDEP (Ganzeveld et al., 2002a) shows for tropical forest an increase in the fraction of sunlit radiation of ∼8% reducing the LAI from 10 to 6.*

5468/8: Is the agreement also good in terms of seasonal cycle / horizontal patterns?

We did not check the temporal and spatial distribution of the VOC emissions in this study and have actually never conducted such a detailed comparison of the global simulated fluxes but focused on the evaluation for specific sites using a column version of the model, which contains the same implementation of the VOC emission algorithm (e.g., tropical rainforest, Kuhn et al., ACPD, 2007; coniferous forest, Ganzeveld et al., Atmos. Environ., 2006). The fact that the implementation of the VOC emission inventory, based on a very similar methodology compared to that presented by Guenther et al. (1995) including the use of 1992 Olson ecosystem database (to determine the global distribution of the VOC emission factors), complemented with the remote sensing biomass products and using ECHAM's radiation and surface temperature, resulted in global annual fluxes similar to that reported by Guenther et al. (1995) gave us confidence in the correct implementation of the algorithm in ECHAM4 and which has consistently being implemented in ECHAM5/MESSy's submodels EMDEP (and ONLEM).

5468/13 Why? Is the T dependence that linear?

No, actually the temperature curves show clearly non-linear behaviour. However, the fact that the monoterpene emissions in the algorithm depend only on temperature (although some measurements also a light dependence), in contrast to the isoprene emis-

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sions that depend on radiation and temperature, the fact that both emissions reflect the same amount biomass and a global distribution of the emissions factors consistent with that of Guenther et al. (1995). This implies that the differences between the global isoprene emission fluxes of EMDEP and Guenther et al. (1995) should be due to differences in radiation (unfortunately this cannot be directly validated since the old ECHAM4 results are not available anymore).

5468/17-19 + 26 So what is the result in ECHAM5? Applying the larger isoprene emissions of 607 TgC/yr, are the concentrations also exceeding the measurements?

It is clear that with the larger emission flux estimate of ECHAM5/EMDEP (and ONLEM) compared to ECHAM4 simulations, the isoprene concentrations are even more overestimated compared to already too large maximum monthly mean concentration over tropical forests $>$ 20 ppbv in ECHAM4. A more extensive evaluation of isoprene concentrations of the ECHAM5/MESSy1 evaluation run simulations by Pozzer et al. (2007, MESSy special issue) shows that also for a significantly smaller global emission flux (0.6 the simulated flux), surface layer isoprene concentrations are still larger compared to observations. A solution of this problem is currently investigated and is, as we state in the manuscript not only due to a possible misrepresentation of the emissions but also potential problems with the chemical destruction, turbulent and convective exchanges and surface removal (oxidation products).

5469/1 The caption of table 3 states that the budget is only an estimate. Then the whole discussion is somehow artificial. What is the uncertainty range? In the case of a strong diurnal cycle, which I assume, the monthly mean value may be dominated by only a couple of instantaneous values. Why hasn't the emitted mass not been accumulated, which would give the exact value?

The emission budgets presented in Table 3 are actually inferred by integrating the mass between the 25-hour frequency output instanteneous values. Using a 25 hr frequency

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assures that the timing, i.e. the diel cycle including the peak emissions, is included in the global annual budget (we use this frequency to construct monthly mean diurnal cycles). Indeed accumulating the total emitted mass as an extra diagnostic parameter would avoid some confusion about total budgets. These extra diagnostics are already or will be included in further model development.

On top of that there is an interannual variability in the emission strength, which has to be included in the discussion, since results from different years are compared.

We do not compare different years. Simulations for all three resolutions use the same reference year. To avoid the issue about comparing different years, we have redone the emission budget calculations extending it to three years. Possibly some confusion has been introduced by stating in the original ms that the runs reflects the year 1987-1988. However, with a set-up using an SST climatology, the runs reflect mostly the present climate. The model setup description has been modified accordingly.

Concerning the role of interannual variability in explaining differences between models (ECHAM4-5) or resolutions, we have also included an indication about using the results of the evaluation run with ECHAM5/MESSy, setup in different configuration compared to our model integrations with as most particular feature the nudging of these simulations. This interannual variability is on the order of $<$ 6% (a range from 510-540 TgC yr-1), so smaller then the difference between the 3-year flux estimate of EMDEP and the Guenther et al. (1995) and ECHAM4 simulations.

5470/7-17(28) As far as I understood an explicit year has been simulated. Does this imply that the meteorology is nudged? Is it done in the same way as in Jöckel et al.? I.e. are the near surface layer not nudged? Since this information is missing it is hard to assess the significance of the results concerning the different resolution. How different are the near surface temperature and wind fields? If those would be the same, will the emission be the same even in a

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finer resolution, I guess not? Are the chosen resolutions, some sort of standard resolutions, recommended for the use of ECHAM5.MESSy? There are a number of open questions concerning the impact of resolution on the emission strength which should be clarified.

We hope that with the introduced modifications in the manuscript with respect to the setup of the various runs presented in ms, indicating the differences with the evaluation run, and the extended simulations to address the role of interannual variability on the calculated emission budgets, the discussion of the role of the resolution on the emissions is indeed clarified.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5457, 2006.