



Interactive comment on “Source apportionment using LOTOS-EUROS: module description and evaluation” by R. Kranenburg et al.

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First of all, thanks for your comments. The issues raised have been addressed, and are discussed below point by point.

- Formulas should be written more precisely and include the units in the text. (see below)

We have unified the notations in the formulas and also added unit descriptions to them

- Some processes presented in Section 3 are not explained in detail. (see below)

In the paper we distributed the processes into four types (emissions, transport processes, removal processes and chemistry). The emissions are used to label the

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sources. From the transport processes, we took only the advection for a detailed explanation. All other processes that govern one-dimensional transport, i.e. diffusion, sedimentation and adjust are implemented following the same approach. We have added some more explanatory sentences in section 3.3 and in section 3.4.

- Figures: Labels are far too small, hard to read

We agree with that, and recreated the figures with larger font sizes.

- Section 4.3: Since the brute force method is not valid as a benchmark, which has been shown nicely by Emmons et al., 2012 (GMDD), Grewe et al. (2010,2012),(GMD and Atmos. Environm.), it is not clear whether this part is of relevance. Please consider a revision of the objective

In principle, it is impossible to give a technical validation of the functioning of the labelling routine for a full chemistry simulation as the chemistry scheme is non-linear. Comparing a labeling to a scenario simulation will show the impact of indirect effects through changes in oxidant levels and the change in pollutant levels. Hence scenario calculations can not provide a real benchmark for the chemistry (Emmons et al., 2012), Grewe et al., 2012). And this point has been explicitly mentioned in section 4.3. Still, to illustrate the functioning of the module, we discuss experiments for CO, MGLY and NOx. The objective to illustrate the functioning rather than a real benchmark is written down in the introduction of chapter 4.

- Section 5 includes a major draw back of this method. This has to be addressed in more detail. It is not clear, why this source is not labelled additionally. Taking into account the agricultural emissions change the results totally.

We would not consider that a drawback of the method, but rather a result of the model implementation. The system presently tracks the source contributions to oxidized (e.g. nitrogen oxides) and reduced (e.g. ammonia) nitrogen species separately. Hence, the origin of nitrate is not connected to that of ammonia, which is needed to form

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ammonium nitrate. Hence, ammonium nitrate is one of the few examples where the interaction between the different labeled classes is important. The present approach is chosen as the implementation is clear and results are easily explainable. An alternative could be to perform a post processing by attributing the mass of ammonium nitrate equally to the origin of ammonia and fine mode nitrate.

In a post processing we can create an extra tracer ammoniumnitrate, with a combined contribution of the NH₄ and NO₃ contribution. (in terms of mass we have to into account the different molecule masses). In the manuscript, Figure 8 is changed such that the impact of this postprocessing is quantified. In this figure the source apportionment for nitrate for the default and the alternative approach are compared. The new figure supports the text in section 5.3 better than the original figure.

- How does this technique compare to Emmons et al., 2012 (GMD), Grewe et al., 2010, (GMD) and Wang et al., 2009; J. Geophys. Res., 114, D21206.

Tagging methods have also been developed for ozone by Emmons et al., (2012), Butler et al., (2011), Grewe et al., (2010) and Wang et al., (2009). In these methods the model description is extended with duplicate tracers and chemical reactions, also substantially increasing computation time. These studies are mentioned in the introduction. In our approach, the chemical transformations are tracked by storing the reacted mass per reaction and combining that with source attribution of the reactant. Hence, we do not duplicate tracers or reactions.

The tagging methods for ozone (Grewe, 2004), (Butler et al., 2011), (Dahlmann et al., 2011), (Grew et al., 2012). required to assume a NO_x of VOC limited chemical regime. With the same assumptions our methodology could also be used for ozone. As the chemical limitation varies across Europe, a sensible application is not straightforward and therefore not pursued here. This is discussed in section 3.5

- For tracking reasons it would be good to have a module name and version number (also in the title).

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The method is a generic method which fits in the LOTOS-EUROS model version 1.8. Later versions probably needs specific adaptations for the labeling routine if new processes are implemented. We included this information in section 3.1

- I propose to include a supplementary material, which describes the input, etc. some sort of a handbook

The input and settings of the model are described in section 2. Although this could be extended (or added as supplementary material), we wonder what kind of detail is necessary for which purpose. A report with more detailed information is available (Schaap et al., 2012). Users who would like to know more detail on a certain processes could also contact one of the authors directly, in order to obtain the latest settings and input data.

From the specific comments We have revises the typing errors and other unclear points. Other specific comments

- 3962/17 "shows comparable performance to other European models": Please give a more quantitative summary.

The LOTOS-EUROS model shows a systematic underestimation of PM of about 40 %. This is mainly due to the missing process descriptions for secondary organic aerosols in the model. For the secondary inorganic aerosols the model shows a much smaller underestimation, while the temporal correlation is about 0.6 to 0.8. This is added in section 2.

- Section 3.4 Can you explain how re-evaporation of rain is treated. Since this is a complex non-linear process this seems to be difficult for labelling. I.e. if rain is form in a column but not reaching the ground water from different levels with different concentrations and fractions might be released at one level.

This process is not taken into account in LOTOS-EUROS. Because the model has a dynamic mixing layer, the clouds are generally in the same model layer. So it is not

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incorporated in the model. If we do have this process, than for the labelling we should track the fluxes resulting from this process and label the rain water concentrations.

- 3968/19 "PAN is taken from both precursor species (C2O3 and NO2)." Please explain in detail how this is done. Is it the same mechanism as in Grewe et al., (2010) GMD? What is about reaction like $\text{NO}_2 + \text{NO}_3 \rightarrow \text{N}_2\text{O}_5$. Either species might have a different fractions?

The problem in this reaction is that we track both the Carbon and the Nitrogen components. The module implementation needs only one source contribution per tracer. So the assumption is that PAN gets half of the NO2 and half of the C2O3 contribution. With the disassociation of PAN, the original contributions of NO2 and C2O3 are not conserved, so there will be some artificial mix up of Carbon and Nitrogen sources. For N2O5, the nitrogen atoms in NO2 and NO3 after disassociation may not be the same as the original N-atoms in NO2 and NO3 which formed N2O5. But NO3 is formed from NO2 at night and has a short life time. So NO3 radical has the same source attribution as NO2 during one time step, thus we do not feel there is a problem with N2O5.

- Section 4.4 : Please explain the sharp decrease for a few labels

The sharp decrease in run time for only a few labels can be explained by the overhead cost. Within one run, the computational cost of reading and processing the input data is relatively large. This is added to section 4.4

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