

Interactive comment on “An advanced method of contributing emissions to short-lived chemical species (OH and HO₂): The TAGGING 1.1 submodel based on the Modular Earth Submodel System (MESSy 2.53)” by Vanessa S. Rieger et al.

Anonymous Referee #1

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General comments

The present study by Rieger et al. presents an updated tagging algorithm for OH and HO₂ which allows for an attribution of HO_x to various emission categories. In contrast to the previous version V1.0 the new version takes into account tropospheric as well as stratospheric chemistry. Furthermore, the new scheme considers rest terms to take into account HO_x production and loss reactions which are not explicitly tagged.

Since the tagging mechanism does not only consider primary contributions, but also

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secondary effects via the long-lived species, the paper is rather difficult to read. I am sure for the authors, who are familiar with the tagging approach, it is clear what's going on, but for an unacquainted reader it is difficult to understand why an updated tagging method for HO_x results in different contributions of, e.g., biomass burning emissions to ozone. Maybe a schematic would be helpful. Furthermore, the HO_x tagging includes several assumptions and special cases. There are several open questions (details see below). My major concern is related to the steady-state assumption and the rest terms. Overall, I think the paper needs a clearer description of the method, a better justification of the assumptions and a more thoroughly explanation of the presented results.

In the introduction the authors argue that mitigation of climate change requires attribution of certain chemical trace gases to specific emission categories, and therefore propose the tagging approach. I wonder how robust the presented results are. For example, the presented model simulations do not consider direct CH₄ emissions. I assume that tagging CH₄ from the applied emission categories would have a large impact on the attribution of HO_x to certain emissions. In my opinion it is inconsistent to consider CH₄ decomposition as one category, while NO_x, NMHCs etc. are split into road traffic, non-traffic etc. For an evaluation of the overall climate impact all traffic related emissions have to be tagged in the same manner. Therefore, I have doubts that the method is already applicable.

In the first sentence of Sect. 3.3 the authors state it is crucial that HO_x production and loss of the reduced mechanism (almost) equal the complete HO_x production and loss. Furthermore, steady state means to me that HO_x production equals HO_x loss. This is valid for the complete HO_x chemistry derived from MECCA, but not or only partly for the reduced system. For example, stratospheric OH production of V1.1 deviates by 9% from the total stratospheric OH production and, maybe even more important, from the stratospheric OH loss of V1.1. Therefore, I do not agree with the conclusion at the end of section 3.3 that the steady-state assumption for the reduced reaction system V1.1 is justified. If the steady-state assumption was fulfilled, the rest terms to close

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the budget would be needless. Furthermore, it is not clear from the paper that the steady-state assumption for H is valid, as claimed in Sect. 3.4. Table 3 presents the “main reactions of H” for the reduced system, but is this reaction system identical to the complete chemical mechanism in MECCA or only a subset?

Section 4 is mainly a description of the presented figures, but lacks explanations, for example for the differences between V1.0 and V1.1. The argumentation is often rather vague, namely that V1.1 considers now more reactions which contributes to the differences. That leaves the impression that the authors themselves do not fully understand the changed patterns. From what is written in the paper it is hard to understand the presented results and differences, but I think this is important to judge the performance and shortcomings of the tagging method. For example, what is the reason for the changes in the contribution of stratospheric O₃ production to tropospheric OH? Furthermore, I am concerned about the HO₂ shipping contribution discussed in Fig. 6. In this case the authors provide a clear explanation, but frankly speaking this example seems to show that the tagging method does not work. Two reactions (production and loss) with the same reaction rate, but only half of the loss is considered???

Specific comments

- P1, L11-14: As mentioned above, at first glance it is hard to understand why the tagging of HO_x affects other tagged species. If you do not want to lose your readers right at the beginning, you should consider rewriting the last part of the abstract.
- P2, L8: For which specific environments? Please clarify.
- P4, L2/3: Why do you use a different time period for the global and the regional simulations (2007/2008 vs. 2009/2010)?
- P5, L7/8: I am not sure if I understand this statement correctly: In V1.0 the OH loss by the reaction with NMHCs was obtained from the total chemical CO production, no matter if OH was involved or not?

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- P5, L17/18: Reaction rates depend on the concentrations of the reactants and via the rate coefficient often on temperature, and therefore vary in time and space. So how is the threshold reaction rate of 1e-15 mol/mol/s to interpret? Is that an annual and global mean value?
- P5, L21-31: What's the point of those two paragraphs? As long as you do not show any numbers, this part is rather vague.
- P5, L33: Why are reactions 19, 28 and 30 listed in Table 1, when they are not part of V1.1? That's inconsistent with the table caption.
- Table 1: How are NMHCs treated in the HO_x tagging? Is there only one lumped NMHC or are the individual NMHCs treated separately? If it is a lumped NMHC, how often can it react with OH?
- Table 2: How exactly are the tropospheric and stratospheric production and loss rates calculated? I assume there is a kind of air mass weighting applied? mol/mol/s is a somehow weird unit, I would usually expect something like Tg(OH)/yr. . .
- P8, L2: It is true that the OH production for “all” differs only by 2% from the total production, but for “tag” the difference is about the same as for V1.0, namely around 11%. Which quantity is used for the tagging – “all” or “tag”?
- P9, L25: Why are the mentioned species not explicitly tagged? Please explain.
- P10, L8: I do not understand this sentence. Ratio of what to what?
- P10, L19: Why are H₂O₂ production and loss not balanced? Please explain. And HO_x production and loss of the V1.1 reaction system are also not balanced, but nevertheless steady-state is assumed. This is inconsistent to me.
- P11, L14: In my view this is in contradiction to the statement at the end of Sect. 3.3 (-> steady-state assumption justified).
- P11, L28/29: Why are the rest terms equally distributed amongst the source cat-

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egories (division by n) and not according to the contribution of the individual source categories to the total, e.g. OH_i/OH ? Couldn't it be that the linear distribution of the rest terms leads to an artificial exaggeration of a minor source category? I would be interested to see the contribution of the rest terms to the individual source categories.

- P14, Fig. 2: What is the reason for the different patterns in OH from anthrop. non-traffic, traffic and shipping? All three categories represent surface emissions.

- P16, L5/6: Please explain how the inclusion of more NMHC reactions leads to the changed pattern. In general, the treatment of NMHCs is not clear to me. Is there one lumped NMHC tracer?

- P17, L2/3: How do NO_y emissions contribute to the category "N₂O decomposition"?

- P19, L16/17: How does the HO_x tagging affect stratospheric O₃ production? Is it that HO_x produced from ozone formed in the stratosphere leads to stratospheric ozone formation/destruction via the catalytic HO_x cycles? But how would that fit with the family concept?

- Conclusions: The first paragraph has a lot of redundancy and should be shortened. This holds for several parts of the manuscript. And the second paragraph is more a repetition of the abstract than real conclusions.

- P21, L24-26: I assume that the local maximum around 5 hPa is a secondary effect via ozone?

- There is no single reference to the supplement in the manuscript. So why are those additional figures shown at all in the supplement? Honestly, I would prefer to see Fig. 1 and 2 from the supplement in the main paper instead of Fig. 2 – 4. That would make the comparison of V1.0 and V1.1 easier.

Technical comments

- Avoid overusing the definite article, e.g. P1, L21: "HO_x impacts global warming and

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local air quality..." or P2, L4: "...human impact on climate and air quality..."

- Caption Table 1, 3: "In the column "tropos." ("stratos.") reactions which are ..."

- P7, L2: Do you mean Sect. 3.3 instead of 3.4?

- Eqn. 5: There is a mistake. For a unimolecular reaction, there is no LossBi, rather a ProdBi. And the reaction rate is reduced to $R=kA$, right? Should be mentioned.

- Eqns. 8 and 9: Is it possible that ProdHi and LossHi are swapped?

- Eqn. 18: I assume the third term on the right side should read -LHO₂ instead of -PHO₂

- Eqns. 23 and 24: Why are the rest terms included in A_i and B_i? Doesn't that lead to a double-counting of the rest terms in eqn. 17 and 18?

- P14, L26: "This overall shift..."

- P21, L22: "large negative minimum" is a rather sloppy expression, please rephrase.

- Fig. 1: The reddish and pinkish lines are hard to distinguish, at least when printed. And I suggest to change the y-axis to 1.e-14 or 1.e-13 mol/mol/s to be consistent with the numbers given in the text.

- Caption Fig. 1: "(a) and (b) show the rates for the troposphere, ..."

- Caption Fig. 2, 3, 6, A1, A2: "Zonal means ... are shown." Simulation -> Simulation

- Caption Fig. 4, 5: consisted -> consistent

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