

Interactive comment on “The atmospheric chemistry box model CAABA/MECCA-4.0gmd” by Rolf Sander et al.

Anonymous Referee #1

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This paper accompanies the preliminary community release of version 4.0 of the CAABA/MECCA atmospheric chemistry box model into the GMD discussions forum, with the model code included as an electronic supplement. The paper summarises updates to the model, implemented since those described previously by Sander et al. (2005, 2011a). It first gives an overview of updates to the default chemical mechanism (MOM), and the provision of other optional mechanisms (CB05BASCOE, MOZART, JAM002 and MCM subsets) in compatible format. It goes on to outline other new features, which include skeletal mechanism reduction based on the DRGEP method, updated isotope tagging and improvements to the photolysis code, including the availability of new modules (JVAL, RADJIMT, DISSOC). Finally, a summary of recent developments of MECCA are given, that are related to its implementation into the MESSy

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modelling system.

This paper acts as a reference document for both users and developers of this community model, and the publication of overview papers of this type is therefore important and necessary. In addition to providing a suitable citation that can acknowledge and reflect the considerable time and effort that goes into developing and maintaining a state-of-the-science code of this type, it acts as a record of how the representation of scientific knowledge evolves.

However, it therefore encounters the main difficulties and limitations commonly associated with such papers in that (i) they often provide only overview statements of the (many) methods, with limited justification in some areas, to ensure that the paper does not become too long and unwieldy; (ii) process representations are inevitably a snapshot of understanding at a given time, and therefore can lag behind the latest developments in fast moving subject areas; and (iii) they generally present few or no results to illustrate the performance of the methods and tools, these being deferred to future publications where they can be presented and discussed in greater detail. These inevitable limitations therefore provide some difficulties for reviewers when judging a paper of this type against some of the GMD review criteria.

Although a lot of useful information is presented, this paper suffers from some limitations in all the identified areas, as highlighted in the comments below. The authors should therefore consider whether they can provide more information and justification on some topics. Similarly, some illustration of the impacts of the updates (where relevant) might be useful. In practice, the simultaneous (or at least imminent) publication of an application paper might have been helpful. Several (presumably) ongoing and proposed activities are listed in Sect. 6, but there are no references to papers by the developers that are in press or in preparation.

Specific comments

Page 2, line 3: MESSy (Modular Earth Submodel System) should be defined.

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Page 2, line 6: Non-methane hydrocarbon (NMHC) chemistry is listed as treated, but the term VOC is used everywhere else.

Page 2, line 25: "The rate of 1,4-H-shift for the MACRO2 radical is treated as predicted by Taraborrelli et al. (2012), which is about an order of magnitude lower than proposed by Crouse et al. (2012)."

However, the Crouse et al. (2012) study includes an experimental determination and is not simply "proposed" based on theory. Do the uncertainties in the theoretical value of Taraborrelli et al. (2012) encompass the experimental value of Crouse et al. (2012)? If so, surely the Crouse value should be applied. If not, further justification for the use of the Taraborrelli value is required.

In addition to this, I could not find any information on this specific reaction in Taraborrelli et al. (2012). Is that reference correct? It seems that Taraborrelli et al. (2012) considers 1,5 H-shifts involving transfer of a hydroxyl H atom and 1,6 H shifts involving transfer from CH₂OH groups (focused on OH-isoprene-O₂ radicals), whereas Crouse et al. (2012) considers the 1,4 H-shift involving transfer of the formyl H atom in MACRO2. Are the rates of two different types of H-shift reaction therefore being compared?

Page 4, section 2.1.1. This section presents a description of how the treatment of reactions of OH with VOCs (and their degradation products) has been updated, e.g. the use of the Peeters et al. (2007) approach for the reactions with alkenes. Some additional clarification of the methods would be helpful, including the following:

i) Although a very useful reference data set, the Atkinson et al. (2006) IUPAC evaluation is now quite old. Updates, refinements and expansions to the IUPAC evaluation are available at <http://iupac.pole-ether.fr/>. Although some of the preferred values may be unchanged from Atkinson et al. (2006), it seems strange not to take advantage of the more recent information.

ii) For the updates to the Kwok and Atkinson SAR method, what set of preferred data is

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being used? Kwok and Atkinson used a much larger dataset than covered by Atkinson et al. (2006), which (with some exceptions) has a cut-off at C3.

iii) On page 4, line 9, it is stated that the substituent factors are "...updated or calculated ex novo by computing the relative rate coefficient of OH with the simplest VOC bearing the substituent relative to the one of its parent compound." First of all, it is not clear why the substituent factors are based on such a restricted dataset. Secondly, it is not at all clear what this means. The immediate suggestion is the substituent factor for an -OH group (for example) is determined from a comparison of the relative rate coefficient for CH₃OH and CH₄ (which is about 140 at 298 K). However, this is not compatible with the description of the method further down, which suggests the factor is probably actually determined from the rate coefficient for CH₃OH, in conjunction with the Kwok and Atkinson value of k_p (and $k_{abst}(-OH)$). This latter procedure would give an $F(-OH)$ value of 5.6 at 298 K, which is substantially greater than the Kwok and Atkinson value of 3.5, based on optimization to the full dataset of OH-containing compounds. If this is the revised method, I cannot see that this is any improvement on Kwok and Atkinson, and is almost certainly a retrograde step. Given that the current MCM uses the Kwok and Atkinson method, this would also not support the statement on page 4, line 11, "No rigorous evaluation of the SAR has been conducted and the estimation uncertainty is expected to be in the same range as for the SAR used by the MCM", which surely needs some further justification.

iv) There are a few other mentions of how the method adopts, or differs from, that used with the MCM. On page 4, lines 6 and 7 it states that "For the C6 to C11 species, the MCM rate coefficients are retained." and that they "...have no temperature-dependence and are only given at 298 K." However, inspection of the MCM website (<http://mcm.leeds.ac.uk/MCM/>) reveals that many OH rate coefficients for C6-C11 species are temperature dependent (e.g. those for C6-C10 n-alkanes). Does the given statement therefore mean that MOM is using temperature independent rate coefficients derived from the temperature-dependent expressions used in the MCM? If so, this

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should be made clearer. It should also be noted that the MCM and GECKO-A teams recently published a comprehensive update to the way OH rate coefficients are to be calculated in the future (Jenkin et al., ACP, 2018), although it is recognised that this might be too recent for uptake into MOM, and is not yet used in MCM.

Page 5, line 1: should CH₃CH₂O₂ be CH₃CH₂ONO₂?

Page 8, line 8: Figure 3 is introduced here. Although this illustrates the comparative performance of the three mechanisms for the given scenario, no further discussion of the differences is given. Is the trace for CB05BASCOE obscured in the terpene panel, or are terpenes not represented?

Page 10, line 25: The term "Targets" is defined within the description of the skeletal reduction method. However, elsewhere they seem to be referred to as either "targets" or "target species".

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