

Authors' Response to Reviews of

GENerator of reduced Organic Aerosol mechanism (GENOA v1.0): An automatic generation tool of semi-explicit mechanisms

Zhizhao Wang, Florian Couvidat, Karine Sartelet

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RC: *Reviewers' Comment*, AR: Authors' Response, Manuscript Text

Reviewer # 2

RC: *The paper discusses the development of a semi-explicit reduced organic aerosol mechanism for sesquiterpenes (Bcary), with the aim for employing it in air quality and large scale models. GENOA mechanism is based on the widely used near-explicit Master chemical mechanism (MCM). The mechanism used different strategies namely, lumping, replacing, jumping and removing to reduce the MCM scheme. The reduction procedure is tested under various environmental conditions (RH, temp etc.), resulting in a final reduced mechanism (Rdc.) suitable to simulate SOA. The simulated SOA using Rdc has low average error when compared to the near-explicit MCM scheme. This is a well thought out work, with suitable implications to better reproduce SOA in large scale and air quality models. I would therefore, recommend the publication of this work after the authors have answered the following questions:*

AR: We would like to thank reviewer # 2 for the positive comments and constructive suggestions, which are much useful to improve the manuscript. We have carefully considered all of these comments and revised the manuscript accordingly. Please note that the line and section numbers mentioned in the response correspond to the version of the manuscript before revision.

1. General comments

RC: *The main question is why did the authors chose sesquiterpenes? Why not isoprene or monoterpenes? The motivation to use sesquiterpenes should be highlighted.*

AR: Thank you for the questions. Sesquiterpene was selected because it is a well-known source of SOA, and its formation mechanism is well documented. Sesquiterpene is also an ideal candidate for model development and demonstration of the reduction methodology. The oxidation products of sesquiterpene are less volatile and tend to condense more readily than those of lighter molecules such as monoterpene and isoprene.

We have added the explanation to the main paper:

Section 1, line 72:

The application of GENOA to the MCM degradation scheme of β -caryophyllene (BCARY) (Jenkin et al., 2012) is described in Sect. 3. ~~The β -caryophyllene species is selected because it widely serves as a benchmark for modeling the tropospheric chemistry and SOA formation relevant to is selected~~

for investigation and demonstration of the GENOA algorithm, because it is one of the most abundant and representative sesquiterpene (SQT) (e.g., Li et al., 2015; Xavier et al., 2019), and its degradation has been evaluated in chamber simulations (Jenkin et al., 2012). The BCARY scheme in the STOCHEM-CRI mechanism (Khan et al., 2017) and . Sesquiterpenes are well-known source of SOA (Hellén et al., 2020; Tasoglou and Pandis, 2015 and the formation mechanism of BCARY is well documented in the experiment data of Tasoglou and Pandis (2015) are also used for evaluation of near-explicit MCM mechanism (Jenkin et al., 2012). Studies have also compared BCARY SOA yields simulated using the MCM mechanism to chamber data (e.g., Xavier et al., 2019). BCARY is therefore an ideal candidate for model development and demonstration of the reduction methodology. In this paper, the near-explicit MCM BCARY degradation scheme serve as a reliable benchmark for GENOA. The experiment data from Tasoglou and Pandis (2015); Chen et al. (2012) are also compared to the newly developed reduced mechanism in Appendix A.

RC: *Since GENOA is a semi-explicit mechanism, can it be used with any box or air quality model?*

AR: Yes. The semi-explicit SOA mechanisms generated by GENOA can be added to the implicit gas-phase mechanism to model SOA in any box or air quality model. In the case of the sesquiterpene SOA, the "Rdc." mechanism adds 14 organic species including six condensables to SOA models, which is computationally feasible even for global 3-D modeling.

Generally, implicit SOA mechanisms applied to 3-D models are often based on the surrogate approach (e.g., Odum's two-product) or the Volatility Basis Set. They are added to implicit gas-phase mechanisms without altering the pathways of oxidants. The same approach is adopted for the semi-explicit SOA mechanisms. The reduced SOA mechanism includes gas-phase chemical mechanism and aerosol properties for condensable species, which can be added to the implicit gas-phase mechanism in air quality models, without altering the pathways of ozone and major radicals.

For example, the 3-D CHIMERE model can simulate SOA using the implicit gas-phase mechanism MELCHIOR2 and SOA mechanism H₂O [Couvidat et al., 2018]. With the reduced SOA mechanisms, SOAs in CHIMERE are simulated by MELCHIOR2, the reduced SOA mechanisms generated by GENOA, and H₂O for other SOA precursors not covered by our mechanisms.

In the paper, We have added the explanation of how the coupling is generally resolved between VOC species and oxidants in 3-D models:

Section 1, line 36:

~~Along with~~ To complete implicit gas-phase mechanisms, implicit SOA mechanisms have been developed to model specifically the SOA formation without modifying the concentrations of ozone and major radicals (Kim et al., 2011). In 3-D modeling, implicit SOA mechanisms or parameterizations are usually added to implicit gas-phase mechanisms, conserving the oxidant chemistry of the implicit gas-phase mechanism.

Implicit SOA mechanisms are often established based on experimental data from smog chamber experiments to represent the formation and evolution of SOA, such as the two-product empirical SOA model (Odum et al., 1996) and the volatility basis set (VBS) that splits VOC oxidation products into a uniform set of volatility "bins" Donahue et al., 2006).

We have also added the following statements about our semi-explicit mechanisms:

Section 2, line 80:

The generated semi-explicit mechanisms are designed to preserve the accuracy of explicit mechanisms for SOA formation, while keeping the number of reactions/species low enough to be suitable for large-scale modeling, particularly ~~3D~~ in 3-D AQMs. The focus of the semi-explicit mechanism is solely on the accurate modelling of SOA. Because ozone, major radicals, and other inorganics are also affected by inorganic and other VOC chemistry, their concentrations are not tracked with the semi-explicit mechanism. Instead, they are simulated using existing implicit gas-phase chemical mechanisms.

RC: *Although comparison has been made against MCM, the performance of a model can be made by comparing it against existing experimental SOA yields. There has been quite a lot of published experimental Bcary SOA yield experiments. I would suggest the authors to discuss GENOA derived SOA yields in comparison to these experiments.*

AR: As GENOA is used to reduce the MCM mechanism, we compared the mechanisms before and after the reduction to ensure that the generated SOA mechanism preserves the performance of the explicit mechanism on SOA formation.

We have also compared SOA yields simulated by MCM and "Rdc." mechanisms to the experimental data from [Tasoglou and Pandis, 2015] and [Chen et al., 2012] in Appendix A. As shown in Fig. A1 of Appendix A, the results of the "Rdc." mechanism (noted as "Rdc.") are in good agreement with the experimental data (noted as "Tasoglou." and "chen.") and the results of the MCM mechanism ("v1b2"). Moreover, as now mentioned in the paper, [Xavier et al., 2019] has already performed some evaluations of BCARY SOA from the MCM mechanism against to chamber data.

RC: *Is GENOA a carbon number conserving mechanism. Its is not clear from the manuscript if the mechanism is carbon conserving or not? If it is not then how do the authors justify it?*

AR: Strictly speaking, the carbon number is not conserved explicitly during the reduction process. However, GENOA is designed to provide a good estimation of the contribution of the different functional groups. Carbon number is constrained in reduction by lumping (the difference in the carbon number between lumped species cannot exceed 2), jumping and replacing (the difference cannot exceed 3). For lumping and replacing, there is also a restriction on the total mass ($< 100 \mu\text{g}/\text{m}^3$). For removing, there are no restrictions on either mass or carbon number.

Consequently, as we always constrain the total SOA concentration with strict error criteria (i.e., ϵ_{ref} and ϵ_{pre}), the OM/OC, H/C, and O/C ratios are well reproduced (see Fig. 9 in the paper), so the carbon number should be also be well reproduced. If necessary, a specific restriction on conserving the carbon number can be added by the user in the reduction.

2. Specific questions

RC: *L13-15: Motivation -> Although the health and climate effects of aerosols are introduced in every paper, the authors should maybe consider to explain these in a few words or a sentence. Also, it would be nice to explain why there is a need to improve the SOA representation in AQMs.*

AR: Thank you for the suggestion. We have rephrased the motivation as follows:

Section 1, line 1:

Atmospheric aerosols ~~garnered considerable attract~~ attentions due to their ~~undesirable effects on human health (Breysse et al., 2013) and climate (Seinfeld et al., 2016; McNeill, 2017), and as such, they must be well-represented~~ effects on climate and human health: they change the earth's radiation balance and cloud formation (Ramanathan et al., 2001; McNeill, 2017); they trigger a wide variety of acute and chronic diseases (Breysse et al., 2013). Because the effects of aerosols on health depend on their size and composition (Schwarze et al., 2006), adequate representations of the aerosol composition, mass and number concentrations are required in air quality models (AQMs).

Besides being directly emitted, aerosols can be secondary, i.e., formed in the atmosphere through chemical reactions and gas-particle mass transfer as secondary aerosols. ~~Secondary~~ Based on the chemical composition, they can be further divided into secondary inorganic aerosol (SIA) and secondary organic aerosol (SOA). SOA, which represents a significant fraction of aerosols (e.g., Gelencsér et al., 2007) Hallquist et al., 2009), and, is largely formed by the condensation of the oxidation products from the degradation of volatile organic compounds (VOC) (Kanakidou et al., 2005). As SOA formation involves multiple processes such as the emission of SOA precursor gases, VOC gas-phase chemistry, gas-to-particle partitioning (Kanakidou et al., 2005; Hallquist et al., 2009), there are great complexity and uncertainty to accurately predict SOA formation with the simplified representations currently used in air quality models (Porter et al., 2021). .

RC: L18-22: This sentence seems to contain quite a lot of information. I would suggest the authors to rephrase it into smaller sentences.

AR: Rephrased.

Section 1, line 18:

~~The chemistry of VOC degradation is comprehensively described in explicit atmospheric~~ The state of knowledge on VOC chemistry can be reflected by explicit chemical mechanisms, ~~such as which contain all known important reaction pathways in VOC degradation. For instance, Jenkin et al. (1997); Saunders et al. (2003) developed the near-explicit Master Chemical Mechanism (MCM) (Jenkin et al., 1997; Saunders et al., 2003), which details, which describes detailed~~ gas-phase chemical processes related to VOC oxidation, ~~or~~. Another example is the Generator for Explicit Chemistry and Kinetics of Organics in the Atmosphere (GECKO-A) (Aumont et al., 2005), which uses a prescribed protocol to assign complete reactions pathways and kinetic data to the degradation of VOCs. Explicit mechanisms represent the state of knowledge current understanding of atmospheric chemistry, including information about reaction pathways, kinetics data, and chemical structures (which may be used to deduce thermodynamic properties based on structure-activity relationships).

RC: L24: "box models". Although, it is true that explicit schemes are used in box models due to relaxed computational burdens, they have been also been used in 1-D column models or 2-D Lagrangian models (these are still not as computationally expensive compared to AQMs or Global climate models).

AR: Thank you for pointing this out. The paragraph has been revised to emphasize the limitations of using explicit mechanisms in modelling, and to include the use of explicit mechanisms in other models besides box models:

Section 1, line 24:

~~Explicit mechanisms are mainly used in the box model for solving the multigeneration oxidation of multitudinous VOCs (e.g., Yuan et al., 2018). While the near-explicit MCM mechanism has been applied to three-dimensional models (e.g., Ying and Li, 2011; Li et al., 2015), explicit mechanisms are generally~~ The MCM mechanism has been used by 2-D Lagrangian models to simulate the chemical evolution of major air pollutants and some SOAs in plumes (e.g., Evtugina et al., 2007; Sommariva et al., 2008; Zhang et al., 2021). Moreover, it has been used for simulating the formation of more complex SOAs at a regional level in 3-D models over a few weeks (e.g., modified MCM with 4642 species and 13,566 reactions in the simulations of Ying and Li (2011), and with 5727 species and 16,930 reactions in the simulations of Li et al. (2015)). Even so, explicit mechanisms of that size are too computationally intensive to be ~~used in 3D air quality model because of the large number of species involved~~ widely employed in 3-D AQMs for SOA formation.

RC: L32: “carbon-bond” instead of “carbon-bound”.

AR: Changed.

RC: L34-35: *Are all the above mechanisms (lumped, CB05, MCM, GECKO-A) developed primarily for ozone simulation? Also it would be good to give examples of a few model species.*

AR: Because grand-level ozone is one of the most important air pollutants, most gas-phase chemical mechanisms used in air quality models focus primarily on predicting accurate ozone concentration. As this may not be the case for all models, we have revised this phrase to be more precise:

Section 1, line 34:

~~These mechanisms were primarily developed for ozone simulation and reflect only the most significant chemical phenomena occurring in the atmosphere, where VOCs are represented by~~ Implicit gas-phase mechanisms were developed and validated to simulate the concentrations of oxidants and other conventional air pollutants such as ozone and NO₂. In these mechanisms, VOCs have been grouped into a limited number of model species because of computational considerations, and the SOA formation is usually not considered.

RC: L55: “suitable to”

AR: Revised.

RC: L112-113: *Why is this order used in the reduction strategy for BCARY? How would any other order influence the reduction strategy?*

AR: This reduction order was the most effective among all the reduction orders we tested. As each validated reduction can affect the subsequent reductions, reductions with small changes or in favor of other reductions are preferred to be run first. Hence, we adopted this reduction order:

First, the strategies of removing reactions (deleting trivial reactions) and jumping (jumping over negligible species) are tested, which trim the scheme for further reduction. In the following step, lumping and replacing (extension of lumping) are applied, which results in a significant merge of both reaction pathways and species. Finally, the strategy of removing species is adopted, following removing gas-particle partitioning for condensable species that cannot be removed with removing species.

We have added the explanation to the paper:

Section 2.1, line 112:

For the BCARY reduction, the reduction strategies are employed in the following order: removing reactions, jumping, lumping, replacing, removing species, and finally removing gas-particle partitioning.

The reduction strategies are ordered based on their potential influences on the mechanism. The first applied strategies, removing reactions and jumping, trim trivial reactions and species without altering the properties of the species. They are followed by lumping and replacing (as an extension to lumping), which refine the mechanisms considerably by merging the species and reactions involved. Afterwards, the "removing species" strategy attempts to delete all merged and unmerged species. Finally, the strategy of removing gas-particle partitioning is applied in order to remove the partitioning of condensable species, which cannot be removed by removing species. This current order has been tested and found to be efficient for the BCARY mechanism, but it can be changed by the user along with other user-chosen parameters shown in Table 5.

RC: *Table1: Typo in reaction 8. I think it is supposed to say 0.2 BCBOH instead of 0.2 BCAOH.*

AR: Actually, both BCAO2 and BCBO2 (Fig. 1) form the same compound BCAOH (Fig. 2) through the self- and cross-reactions of peroxy radicals (RO2-RO2 reaction). As a consequence, BCBOH does not exist in MCM mechanism.

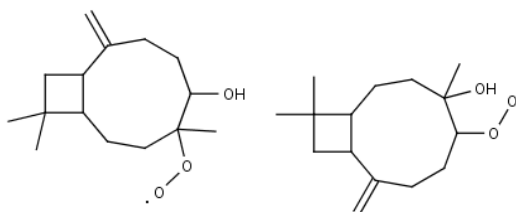


Figure 1: The molecular structures of the MCM species BCAO2 (left) and BCBO2 (right).

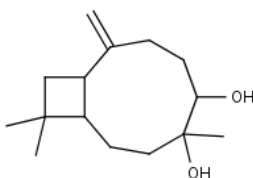


Figure 2: The molecular structure of the MCM species BCAOH.

RC: *Table2: Typo in lumped reaction R2: $0.753 * (f_{w,a} BCANO3 + f_{w,b} BCBNO3 + f_{w,c} BCCNO3)$. I would guess the factor is 0.247 instead of 0.753.*

AR: Corrected.

RC: *Table2: $C_{r,b}$ and $C_{r,c}$ are not defined. It should be defined similar to $C_{r,a}$ for better clarity.*

AR: Thanks for the suggestion. The definitions of $C_{r,b}$ and $C_{r,c}$ have been added to Table 2.

RC: L119: It would be much clear to write it as “In this example, a total of 12 chemical reactions involving three organic compounds are reduced to five reactions (4 lumped (R1-4) + 1 surrogate (R0))”.

AR: Thanks for the suggestion. The sentence has be rephrased:

Section 2.1.1, line 119:

In this example, a total of 12 chemical reactions involving three organic compounds are reduced to ~~four reactions with one new surrogate~~ five reactions (new surrogate production R0 and four lumped reactions R1 - R4).

RC: L122-123: How are the BCARY isomers undergoing similar reactions with HO₂, NO and NO₃? Are the authors referring to the R1-4 in the lumped scheme. Please make this clear to the readers.

AR: Here we wanted to point out that MCM species BCAA₂, BCBO₂, and BCCO₂ share similar structures and properties. The fact that they are isomers (which is not necessary for lumping) may explain why all of them reacted with the same species (HO₂, NO, NO₃, and other peroxy radicals (RO₂)). As long as species meets the lumping criteria, they can be merged together via lumping, and their reactions with different oxidants are also be lumped accordingly.

In order to avoid ambiguity, we have rephrased the sentence as follows:

Section 2.1.1, line 122:

As demonstrated in the tables, the organic compounds BCAA₂, BCBO₂, and BCCO₂ from the original MCM scheme are the peroxy radicals formed from the OH-initiated oxidation of β -caryophyllene (Table 2). ~~Because these isomers undergo similar reactions with HO₂, NO, NO₃, they may conform to the lumping standard~~ It is evident from their structures (shown in fig. C1) that they are isomers and may share similar chemical properties. When applying the lumping strategy, BCAA₂, BCBO₂, and BCCO₂ are merged into a new surrogate named "mBCAA₂" (Table 3).

RC: Why is Cr,a,b,c an arithmetic mean of 5 day simulations? I.e was this 5 day period selected?

AR: A five-day simulation period is chosen for calculating C_{r,a,b,c} and all other reduction parameters, as a compromise between a shorter period that may not reflect the aging of SOA, and a longer period that may less adequately address the SOA formation and be computational expensive. As the concentrations of ozone, radicals, inorganics and environmental parameters (e.g., temperature, relative humidity) are extracted from the 3-D CHIMERE simulations (24-hour monthly averages). Their diurnal profiles are repeated for five days in 0-D simulations.

To clarify, the following explanation has been added for the settings of 0-D simulation:

Section 2.3, line 268:

Unless stated otherwise, ~~a 5-day simulation is performed~~ two simulations are performed for each condition starting at midnight (0 h) and noon (12 h) ~~for each condition, focusing on,~~ taking into account both the daytime and nighttime chemistry. All 0-D simulations are run for five days in order to consider adequately SOA formation and aging processes.

RC: L129: kinetic -> kinetics

AR: Corrected.

RC: *L130: weighting -> weighing*

AR: Corrected.

RC: *L135: What is this specific behavior?*

AR: Here we wanted to point out that different types of compounds may be involved in different types of reactions. A radical, for instance, may be better lumped with another radical than a condensable compound. A PAN compound has a decomposition reaction that does not have other compounds. Therefore, a restriction on certain structural groups was applied to the BCARY reduction.

The term "specific behavior" has been rephrased for clarity:

Section 2.1.1, line 135:

~~Due to specific chemical behavior, compounds with~~ Compounds with specific structural groups sharing common chemical behavior may be more appropriately merged together. Thus, compounds containing the following functional groups can only ~~participate in lumping with the compounds sharing the same group~~ be lumped with compounds containing the same groups: peroxyacetyl nitrates (PAN), organic nitrates (RONO₂), organic radicals (R), oxy radicals (RO), peroxy radicals (RO₂), carboxylic acids (RC(O)OH), percarboxylic acids (RC(O)OOH).

RC: *L162: Aren't alkoxy radicals are RO.*

AR: Corrected.

RC: *L179: Can the authors explain what the maximum hourly branching ratio is?*

AR: We have added the definition of the maximum hourly branching ratio to the main paper:

Section 2.1.4, line 178:

There is no particular restriction to exclude species from the reduction attempt via the strategy of removing compounds or removing gas-particle partitioning. However, for removing reaction, a threshold ~~for its maximum hourly branching ratio under training conditions on the branching ratio of the reaction is applied to the reduction.~~ The branching ratio is defined as the ratio of the destruction rate of one reaction to the sum of the destruction rates of all reactions of the targeted species. In the BCARY reduction, a maximum branching ratio (B_{rm}) is applied to the reduction defined as a restriction criterion. All reactions with hourly branching ratio (averaged over the training conditions) under this value (reactions that are likely to a minimal effect on SOA formation) are considered as candidate for removal.

RC: *L185: Do the authors mean that that after one loop of reduction (as shown in Figure 1), the subsequent reduction is carried out in the reverse order? And why is saturation vapor pressure used only for the lumping strategy. This part needs more clarity.*

AR: Sorry for the ambiguity. For each strategy, the search for potential reduction is conducted following the reverse lists of reaction/species. For example, with removing reactions, GENOA attempts to remove the reaction from the end to the beginning of the reaction list. When applied to the jumping strategy, GENOA

tries to jump the species that has the highest generation and then move down to the species that has the lowest generation.

In lumping, we consider that a condensable species should be first grouped with another compound that has a similar volatility. Thus, the saturation vapor pressure is used to determine the most appropriate lumpable species. The saturation vapor pressure of species is not affected by other strategies and is therefore not used as a criterion for them.

We have added the explanation to the paper:

Section 2.1.4, line 185:

Moreover, the searches for viable reductions via removing are conducted in reverse order of the reaction/species list, ~~beginning with the species/reactions of the highest generation~~ which means that GENOA attempts to remove reactions from the bottom of the list and moves to the previous reactions. The same reverse sequence is followed for other strategies, ~~where the~~. When applied to the jumping strategy, for instance, GENOA tries to jump the species that has the highest generation and then move down to the species that has the lowest generation. Among all reduction strategies, only lumping alters the saturation vapor pressure of condensable species. Therefore, a rank of saturation vapor pressure for condensable compounds is used exclusively for lumping in lumping to determine the most appropriate lumpable species.

RC: *Table 5: Are the two conditions ADD1 and ADD2 high or low NO_x regimes?*

AR: ADD1 is under low NO_x regime and ADD2 is under high NO_x regimes. We have added columns in Table 1 (Table 5 in the paper), i.e., the average SOA concentration and NO reactive ratio with RO₂ (R_{RO_2-NO}), which more clearly indicates the chemical regimes of conditions. If R_{RO_2-NO} is high, the conditions are in the high NO_x regime. Otherwise, the conditions are in the low NO_x regime. For clarity, the digits in Table 5 of the paper have also been changed for temperature and relative humidity.

RC: *Figure 2: How do I interpret this? Is the top bar representing 0h and the bottom one representing 12h? Please add this to the figure caption, since 0 h and 12 h are not represented by empty and hashed lines.*

AR: Thank you for pointing out the error in Figure 2. The hash line for the 12-h condition was not shown. The figure (Figure 2 in the paper) has been revised to Fig. 3.

RC: *L233: I wonder what are the conditions which cause such low O₃ and NO₃ concentrations?*

AR: This condition is located in the northern part of Italy, within the Alpine arch, close to the metropolitan city of Milan. The concentrations of NO transported from polluted areas consume O₃ and NO₃ and produce NO₂, which explains the low concentrations of O₃ and NO₃ in this area.

We have added the more information about this "ADD2" training condition to the paper:

Table 1: Geographic and meteorological conditions of the training dataset

Condition Name ^a	Lat	Lon	Time	TEMP	RH	R_{NO} ^b	SOA ^c
	°N	°E	month	K	%	%	$\mu\text{g}/\text{m}^3$
OH NO	36.0	15.4	Jul.	299.4	78.6	60	4.1
OH HO ₂	32.0	-9.4	Jul.	295.9	76.7	20	6.1
NO ₃ NO	40.25	-3.4	Jul.	302.4	27.9	69	4.4
NO ₃ HO ₂	32.0	36.6	Aug.	302.2	38.7	29	5.7
O ₃ NO	69.0	33.8	Jan.	260.7	84.2	99	5.2
O ₃ HO ₂	68.0	18.2	Dec.	265.5	88.7	25	4.6
ADD1	41.5	-14.2	Dec.	288.6	75.8	20	5.5
ADD2	45.75	9.0	Dec.	279.1	84.5	100.	4.4

^a from left to right: name, latitude, longitude, time period, average temperature, average relative humidity, daily average NO reacting ratio, simulated total SOA concentration of the training conditions.

^b the daily average NO reacting ratio is calculated out of the RO₂ reactivity of NO, HO₂, NO₃, and RO₂. Conditions with high R_{NO} ratio are considered as in high NO_x regime. ^c the initial concentration of BCARY is 5 $\mu\text{g}/\text{m}^3$.

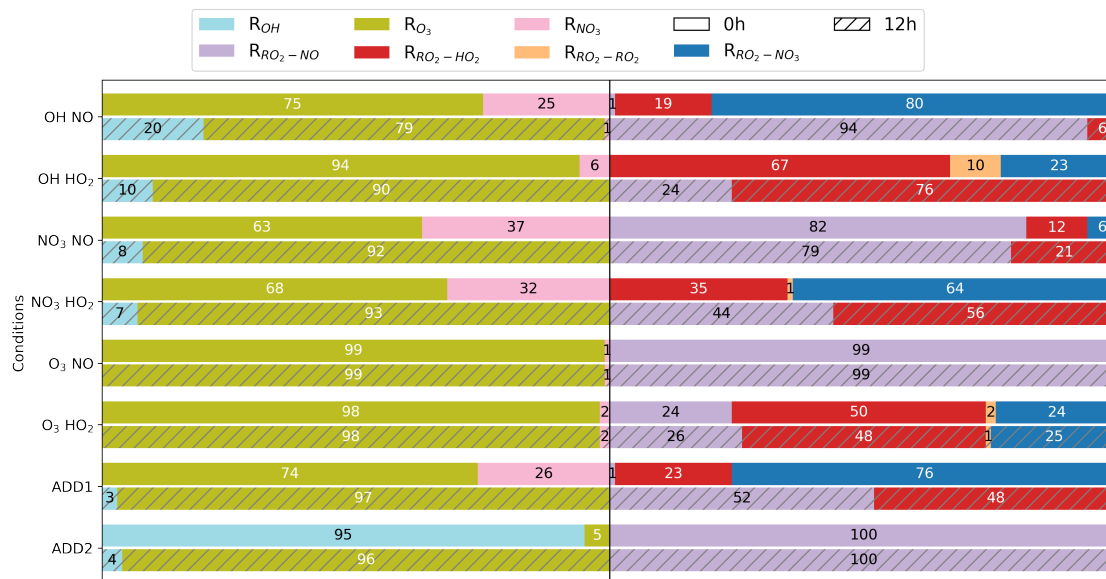


Figure 3: A bar plot showing the occupancy of seven reacting ratios in BCARY initiation reactions and RO₂ reactions, under the training conditions at midnight (0 h, top bar) and noon (12 h, bottom bar).

Section 2.2, line 232:

One specific exception is the additional condition ADD2, ~~in which~~ which is located in the northern part of Italy, within the Alpine arch, close to the metropolitan city of Milan. This condition is in extremely high-NO_x regime, as high concentrations of NO are transported from polluted areas. These high NO concentrations consume O₃ and NO₃, causing low concentrations of O₃ and NO₃. At night, ADD2 has a high R_{OH} of 95 % at midnight is not due to an abundance of OH, but rather to extremely low concentrations of O₃ (2.9×10^{-4} ppb) and NO₃ (1.1×10^{-9} ppb) that leads to an absence of nighttime reactivity.

RC: *L258: Is Kelvin effect not taken into account for gas-to-particle partitioning?*

AR: The Kelvin effect is not taken into account in the simulations, as thermodynamic equilibrium is assumed between gas and particle phases.

RC: *L262: "vapor pressure is computed using Myrdal and Talkowsky". This phrase is repeated already in the earlier part of the sentence.*

AR: Rephrased.

RC: *Appendix A: It should be "v1:Nannolal (2008)" not Nannolal 2004.*

AR: Corrected.

RC: *Appendix A, L485: There seems to be a contradiction here. The authors say that v1b2 (Nannolal and Jacob and Reid) show the best estimate in comparison with the experiments "As shown in Fig.A1, the SOA distribution simulated with "v1b2" agrees best with the experimental data. Therefore, this method with the vapor pressure computed by Myrdal and Yalkowsky (1997) and the boiling point computed by Joback and Reid (1987) is used in the BCARY reduction". But in the manuscript why has Myrdal and Talkowsky been selected in place of Nannolal 2008. Why?*

AR: Thank you very much for pointing out this error. "v1" is the method of Nannolal et al. (2008), but in the paper, it was incorrectly referred to as the method of Myrdal and Yalkowsky (1997).

We have corrected the typo:

Appendix A, line 481:

Eight methods are provided in UManSysProp, including SIMPOL.1 of Pankow and Asher (2008) ("sim"), EVAPORATION of Compernelle et al. (2011) ("evp"), and six methods out of the combination of two methods to compute the vapor pressure ("v0": Myrdal and Yalkowsky (1997) and "v1": ~~Nannoolal et al. (2004)~~Nannoolal et al. (2008)) and three methods to compute the boiling point ("b0": ~~Nannoolal et al. (2008)~~Nannoolal et al. (2004), "b1": Stein and Brown (1994), and "b2": Joback and Reid (1987)). As shown in Fig. A1, the SOA distribution simulated with "v1b2" agrees best with the experimental data. Therefore, this method with the vapor pressure computed by ~~Myrdal and Yalkowsky (1997)~~Nannoolal et al. (2008) and the boiling point computed by Joback and Reid (1987) is used in the BCARY reduction. The results simulated with the final reduced mechanism "Rdc." is also presented in Fig. A1, which has a great resemblance to the experimental data.

RC: *L275: I think it would be better to rephrase the sentence defining the FME. Is it so that the simulation error is the larger FME of the two errors I.e the FME of day 1 and the FME of rest of the simulation days? This has*

be more clear in the text.

AR: The following sentences have been rephrased to better explain how we calculate error:

Section 2.4, line 274:

~~To address the difference of~~ The error of one simulation is defined as the larger of the FME on day one and the FME on days two to five, in order to address the difference in performance of the reduced mechanisms at the early stage of the simulations (first day SOA formation dominates) and at the later stage, the error of one simulation is defined as the larger FME between the FME of the first simulation day 1 and the FME from day 2 to 5, and it is compared (SOA aging dominates). This error is used to evaluate reduction by comparing it to the error tolerance specified in training (Eq. 1). For the evaluation on the training dataset, two errors are estimated compared to the previously verified reduced mechanism with a tolerance denoted ϵ_{pre} , and the MCM mechanism with a tolerance denoted ϵ_{ref} . The error tolerances are used to restrict both the maximum and the average (half of the tolerance) errors of the training conditions. As for the evaluation on the pre-testing dataset, ~~one only the error compared to the MCM mechanism is calculated, with an error tolerance denoted $\epsilon_{pre-testing}$.~~ The error tolerances $\epsilon_{pre-testing}^{ace}$ and $\epsilon_{pre-testing}^{max}$ are set to the average and maximum errors, respectively.

RC: Eq 1: What does $i1$ and $i2$ represent?

AR: $i1$ and $i2$ are meaningless and have been removed from Eq.1.

RC: L283: Why does the ϵ_{pre} vary in such a way with respect to ϵ_{ref} . More explanation is need here.

AR: ϵ_{ref} and ϵ_{pre} are the criteria we set for evaluating the reduction. The difference between the two criteria is that ϵ_{ref} is compared to the reference mechanism and ϵ_{pre} is compared to the previous validated mechanism. ϵ_{ref} is used to track the performance of the reduction, while ϵ_{pre} is used to avoid large errors introduced by one reduction attempt. So logically, ϵ_{pre} should be less or equal than ϵ_{ref} .

In practice, when ϵ_{ref} increases by 1 %, the value of ϵ_{pre} is set from 1 % (minimum value) to the value of ϵ_{ref} . By doing this, GENOA first accepts reductions that introduce small errors compared to the previous validated mechanism, and then accepts reductions that introduce larger errors up to ϵ_{ref} .

The explanation has been added to the paper:

Section 2.4, line 282:

In order to begin with a conservative BCARY reduction, the initial values of ϵ_{pre} and ϵ_{ref} are both set to 1 %. The values of these error tolerances are then increased to larger values, reflecting the looser criteria used throughout the reducing. ϵ_{ref} is used to track the performance of the reduction, while ϵ_{pre} is used to avoid large errors introduced by one reduction attempt. Therefore, ϵ_{pre} is lower or equal than ϵ_{ref} . For every 1 % increase in ϵ_{ref} , ϵ_{pre} is stepped up by 1 % from 1 % to the value of ϵ_{ref} . By doing this, GENOA first accepts reductions that introduce small errors compared to the previous validated mechanism, and then accepts reductions that introduce larger errors up to ϵ_{ref} .

RC: L288: Is this true? As mentioned the <3% avg error is for pre-testing + training dataset. Will the average error still be < 3% for test dataset?

AR: For the BCARY reduction, the pre-testing dataset selected can provide an accurate representation of the average conditions in the testing dataset. Thus, when the average error of the pre-testing condition is less than

3 %, the error of the testing dataset is less than 3 % as well.

Generally, it depends on whether the pre-testing dataset is representative of the testing dataset. As the pre-testing dataset is selected randomly from the testing dataset, it may not initially be a reliable representation of the testing dataset (e.g., the error of pre-testing is much smaller or much larger than the error of testing). It is necessary to modify the pre-testing dataset in such a case. For example, a few conditions with large errors from the testing dataset can be added to the pre-testing dataset to improve the performance of pre-testing if the error of pre-testing is smaller than the error of testing.

We have added the explanation to the paper:

Section 2.2.2, line 240:

Meanwhile, the size of the mechanism has already been significantly reduced, which makes the evaluation of each reduction attempt on the pre-testing dataset less computationally expensive.

In principle, the pre-testing dataset should be able to provide a fairly accurate representation of the testing dataset. However, this may not always be the case, since the pre-testing dataset is selected almost randomly from the testing dataset. Therefore, an adjustment may be required to increase the representativeness of the pre-testing dataset by adding or removing a few conditions.

RC: *L303: How does GENOA decide what condensable species to remove? Is it based on species super saturation values?*

AR: In the reduction via removing gas-particle partitioning, GENOA tries to remove the partitioning of each condensable regardless of the saturation vapor pressure. The reduction is accepted only if the errors of training/pre-testing conditions are small enough.

In the late-stage reduction, the aerosol-oriented treatments are applied. Since there is strong competition among reduction strategies at the late stage of the reduction process, these treatments are used to reduce species rather than reactions, thereby reducing condensable species.

To avoid confusion, we have rephrased the statements about the aerosol-oriented treatments:

Section 2.5, line 301:

These treatments, which reduce species rather than reactions, are done when the size of the mechanism is below a certain threshold (20 for BCARY reduction). ~~They are aerosol-oriented, aiming to~~ Consequently, the late-stage treatments encourage the reduction via the removing of condensable species, and are referred to as the aerosol-oriented treatments.

RC: *L326: Effective partitioning coeff. is temperature dependent. What is the assumed temperature for the classification of SVOCs, LVOCs and ELVOCs? And since the authors use a range of temperatures in their training conditions (268-302 K) does it really make any sense in classifying the SVOCs, LVOCs and ELVOCs at an arbitrary temperature?*

AR: The effective partitioning coefficient (K_p) is computed at 298 K. We have added the assumed temperature for K_p in the paper.

RC: *L332: Are the species in the reduced Rdc. and Khan 2017 mechanism overlapping or identical?*

AR: The mechanism of [Khan et al., 2017] preserves information regarding the reaction/species of the first and

second generations of the MCM mechanism. In contrast, the "Rdc." mechanism may preserve information concerning up to the tenth generation of MCM. Thus, the "Rdc." mechanism may provide a more detailed description of SOA formation and aging than the mechanism of [Khan et al., 2017].

RC: *L341: Condensable species drop to less than 20 ? It seems that the condensable species is 10 % of its original values (10% of 493 48/49 species or thereabout) as shown in Figure 3. Also ϵ_{pre} is not shown in Figure 3. Also on L 346 it is mentioned as 41 species on the 75th time. So what is the correct number?*

AR: The number of condensable species indeed dropped to 20 at the 74th step, with a fraction of 5.61 % and the initial number of condensable species in MCM BCARY mechanism is 356. As of the beginning of the 75th step (by the end of the 74th step), the number of species is 41, including both condensable and non-condensable species.

Though the numbers in the text have been verified to be accurate, the number of reduction steps might not be clear. According to figure 3 in the paper, the size at the nth step refers to the size at the end of the nth reduction step. However, in the text, we confused it with the size at the beginning of the nth reduction step. Accordingly, the following statements have been revised:

Section 3.1, line 341:

- Early stage, from the first to the 74th reduction step, ~~where the number of condensable species drops to 20.~~ By the end of the 74th reduction step, the mechanism is reduced to 68 reactions and 41 species (including 20 condensable species). The early-stage reduction is trained only on the training dataset with the seven pre-described reduction strategies. After ϵ_{ref} reaching 3 %, the list of B_{rm} is changed from [0.05, 0.10, 0.50] to [0.10, 0.50, 1.0].
- Late stage I, from the 75th to the 107th reduction step, ~~where the number of condensables decrease to.~~ By the end of the 107th reduction step, the reduced mechanism consists of 38 reactions and 19 species (including 7 and no more condensable species), and no further reduction can be found within $\epsilon_{ref} \leq 10$ % and $\epsilon_{pre} \leq 10$ %. ~~On the 75th reduction step, the size of the mechanism drops to 68 reactions and 41 species. From there, the~~ In this stage, the reduction is trained on the pre-testing dataset if the condensable species are removed with lumping, replacing, or jumping. For reduction with other types of reduction strategies, it is first trained on the training dataset and then on the pre-testing datasets. From all reduced mechanisms with seven condensable species, GENOA selected the one with the minimum average errors on pre-testing dataset (2.44 %) to start the next stage.
- Late stage II, from the 108th to the 113rd reduction step. At this stage, the reduction strategy of removing elementary-like reactions is applied to the training. All reductions that reduce the condensables are ~~evacuated~~ evaluated exclusively on the pre-testing dataset. ~~From the 108th reduction step to the end of the training, the~~ The size of the reduced mechanism was reduced ~~from 37 reactions and 19 species~~ to 23 reactions and 15 species, among which the number of condensable species is reduced ~~from 7~~ to 6. The average (maximum) error of the final reduced mechanism "Rdc." is 2.65 % (17.00 %) under the pre-testing dataset compared to MCM.

As ϵ_{pre} is only compared to the previously validated mechanism, it does not reflect the performance of the mechanism as does ϵ_{ref} . Therefore, we keep ϵ_{ref} only as an estimation of the performance of the reduction at different stages of the reduction in Figure 3 of the paper.

RC: *L346-348: Aren't all the reduction strategies trained first with training data sent and then with pretesting data*

set? So what is the difference here between, lumping, jumping and replacing compared to other strategies?

AR: In the late stage of reduction, the reduction via removing is evaluated first on the training dataset, and then on the pre-testing dataset, whereas the reduction via lumping, replacing, and jumping is evaluated only on the pre-testing dataset. The reason is that lumping, replacing, and jumping may be more effective in terms of altering the scheme. Compared to removing, they offer more possibilities for reducing species. Therefore, reductions via lumping, replacing, and jumping are evaluated only on the pre-testing dataset. This treatment was tested and adopted for the BCARY reduction. It can be turned off by the user when applied to other reductions.

RC: *L351: “evacuated”-> “evaluated”*

AR: Corrected.

RC: *L358: It should be specified that lumping reduces the condensable species by 35 %.*

AR: We have removed the misleading statement:

Section 3.1, line 357:

As expected, the reduction strategy of removing reaction contributes the most to the decrease in the number of reactions (48 %), followed by the strategy of removing species with a contribution of 37 % ~~and lumping with a contribution of 31 %~~. Meanwhile, both lumping and removing species are significant in the reduction of species, by 35 % and 31 %, respectively.

RC: *L366: There is not mC1330 in Figure 4.*

AR: C1330 in the "Rdc." mechanism has the same properties as the one with the same name in the MCM mechanism. Therefore, the name C1330 remains unchanged after reduction.

RC: *L392: Why is more uncertainty found in regions with low RH and high temperatures? Is it because the training data set does not have enough data to work with in these conditions? Looking at figure 6, it shows that even Russian data points have high uncertainty between 3-6%. This cannot be only due to low RH and high temperature. What could be the other possible reasons for that?*

AR: The relatively high error in the regions with low RH and high temperatures indicates that such conditions are not sufficiently represented in the pre-testing dataset. As these conditions are rare to encounter over Europe, they are not included in the pre-testing dataset, such as not increasing the size of the mechanism.

Considering that the average error for pre-testing is 3 %, there are, of course, conditions with errors greater than 3 %, which are placed in the error category between 3 % and 6 %. As we be seen in Fig. 5 of the main paper, the testing errors in July and August (corresponding to the results in Fig. 6 of the paper) are actually very close to 3 %. These conditions are scattered throughout Europe, such as in the Russian area, in northern Europe, as well as in the Mediterranean.

RC: *Figure 8: Didn't the authors claim that PAN is under-represented in GENOA on L 418? But Figure 8 shows higher PAN concentrations for GENOA and lower for MCM.*

AR: We apologize for the mix-up in group names in the paper, and there is no doubt that the PAN concentrations are overestimated by the reduced mechanism shown in Figure 8 of the paper.

We have corrected the typo:

Section 3.2.2, line 415:

In comparison to MCM, only two condensable species containing nitrogen are retained in the "Rdc." mechanism: NBCOOH and C131PAN, leading to an ~~overestimation of the nitrate mass of the~~ underestimation of the organic nitrate group (~~0.10~~ $\mu\text{g m}^{-3}$ ~~0.31~~ in MCM and ~~0.30~~ ~~0.04~~ in "Rdc.") and an ~~underestimation of the overestimation of the nitrate mass of the~~ peroxyacetyl nitrate group (~~0.31~~ $0.10 \mu\text{g m}^{-3}$ in MCM and ~~0.04~~ ~~0.30~~ in "Rdc.").

RC: *L438: It should be explained why due to different volatility species Rdc delays SOA production. Is it due to low LVOC concentrations or high ELVOC concentrations?*

AR: Sorry for the misleading. The statement "Rdc delays SOA production" may not accurately describe the general situation in simulations that involve extreme SOA loading and large errors.

Thus, we have removed it from the discussion:

Section 3.2.3, line 336:

The result indicates that the "Rdc." mechanism may introduce relatively large uncertainty with extreme SOA loading (~~conditions that were outside the range of conditions used for the construction of the "Rdc." mechanism~~). ~~Results indicate that the "Rdc." mechanism may delay the SOA formation under large mass loading (larger than 500 $\mu\text{g m}^{-3}$), because of differences of the volatility of the oxidation products which was outside the range of conditions used for the construction of the "Rdc." mechanism and MCM.~~

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