Li et al. present a box model with a new photochemical solver, ROMAC v1.0, that aims to be flexible and computationally efficient through an unique adaptive dynamic optimization module. It also improves over traditional box models with better quantification of physical effects. A new chemical solver and box model development is exciting news for the field and well fit for the scope of GMD. I'm happy to recommend this manuscript for publication in GMD.

Major comments:

1. An important development for a highly efficient chemical solver is to move beyond the box model and improve the efficiency of 3-D chemical transport models. Could the authors elaborate on ROMAC's extensibility to be used, eventually, as a chemical solver component in 3-D models?

Response:

As described in Section 2.1, the VSVOR solver in ROMAC is a versatile chemical solver. Unlike the EBI solver in CMAQ, which is often customized for specific chemical mechanisms, the VSVOR solver is designed to be universal. This means that it possesses the capability to handle various chemical mechanisms in different models.

In the current version of ROMAC, the chemical solver is an integral part of the software and is not separately callable by the user. However, we are actively working on the development of the ROMAC-plug-in modeling tool. Please see our response to the general comment of the first anonymous referee for more details. This forthcoming tool will enable users to utilize the chemical solver program within ROMAC in their own models. Its design and development are currently underway, and we have added this development plan to the future development section in the revised manuscript.

Line412-416: In future development roadmap, we have plans to introduce a modeling framework version of ROMAC known as “ROMAC-plug-in”. This ROMAC-plug-in will support calls from Python or Fortran, ensuring compatibility and flexibility for

users. Importantly, the efficient design of ROMAC will be maintained, allowing for optimized performance. The kernel of ROMAC-plug-in will be specifically engineered to provide users with flexibility to effortlessly construct their own models or integrate ROMAC with existing frameworks, such as CTMs.

In addition, the description and explanation of the extensibility of the chemical solver in ROMAC is also added.

Line111-114: This solver is engineered to enhance computational efficiency while accommodating the universal attributes of atmospheric chemical mechanism. It approaches all differential equations uniformly, eliminating the need for customized solution schemes tailored to specific chemical mechanisms. Therefore, the VSVOR solver offers a universal and versatile method for chemical solving.

Line152-154: The aforementioned characteristics are inherently present within the Jacobian matrix of chemical mechanisms and are impervious to variations in specific chemical mechanisms. As a result, this scheme proves to be universally applicable across different chemical mechanisms.

2. In the intro around L51, authors discuss simplified chemical mechanisms. These approaches have a long history. The authors mainly talk about "fixed" reductions of the chemical mechanism, where a larger mechanism (e.g., MCM) is processed down to fewer species beforehand and used. But there are general methods for reduction (e.g., Young & Boris, 1977; Djouad & Sportisse, 2002) and their on-line implementations (e.g., Sander et al., 2019; Shen et al. 2022; Lin et al. 2023) that provide stability and efficiency at some cost of error, and would be useful to include in the introduction.

Response:

Thanks for your comments, we have refined the parts of simplified chemical mechanisms and solution methods in the introduction section.

Line56-58: The simplified chemical mechanism can effectively improve the solution

efficiency of chemical processes, such as SAPRC07 (Carter, 2012), CB6 (Yarwood, 2010), MOZART(Emmons et al., 2010) and the Mainz Organic Mechanism (MOM) (Sander et al., 2019).

Line59-60: General methods for reduction (Young & Boris, 1977; Djouad & Sportisse, 2002) and their on-line implementations (Sander et al., 2019; Shen et al. 2022; Lin et al. 2023) had been developed.

3. More generally, I think that saying simplified mechanisms lead to bias to simulation results is not a fully fair statement to make. There is no one true chemical mechanism that gives 100% accurate answers - so is the bias defined against the MCM, or against the observations? Reduced mechanisms generally have a focus on getting particular parts of chemistry that are of interest correct, and thus introduce some degree of bias in species that are not fully represented (and it is unavoidable to have some bias in fast-cycling radicals).

Response:

Thanks for your comments, we wish to clarify that our intention is not to undermine the value of the simplification mechanism. Rather, we aim to convey that while the simplification mechanism can significantly enhance program efficiency, it cannot entirely substitute for the role of the near-explicit mechanism. Therefore, it remains imperative to enhance the efficiency of the near-explicit mechanism.

Furthermore, we recognize that the term '*bias*' may not be the most suitable choice. To address this concern, we wish to emphasize that our discussion revolves around distinguishing between a simplified mechanism and a near-explicit mechanism. According to your suggestion, we have revised the manuscript.

Line60-65: These simplified mechanisms are typically tailored to emphasize specific aspects of chemistry. As a result, the simulation results for certain species may diverge from those obtained using near-explicit chemical mechanism, particularly concerning

radicals (e.g., OH, HO_2, RO_2) and the concentrations of secondary pollutants (Ying and Li, 2011; Jimenez, 2003). The adoption of near-explicit chemical mechanisms enables a more detailed representation of the intricate process of photochemical reactions. Consequently, the simplified mechanism cannot adequately replace the role of the near-explicit mechanism.

4. The software is currently limited-access to reviewers. Will the software be open in the future / upon publication to GMD? That is very important to the community and I believe in line with GMD policy.

Response:

The current version of ROMAC coupled MCM v3.3.1 is archived on Zenodo: https://doi.org/10.5281/zenodo.7900781. This is mentioned in the code availability section. The software will be open access when the final manuscript is accepted and published in GMD. Future versions will continue to be published and open access on Zenodo, and updates will be noted in the current link.

Minor comments:

1. L71: The authors say that "Since the ROMAC model is computationally efficient, accurate and stable, users can dynamically optimize the influence of physical processes on pollutant concentration, and overcome the shortcomings of the lack of physical processes in the traditional box models." I'm not sure I follow here. What is the main difficulty in incorporating physical processes in other box models, is it a deliberate choice to focus on chemistry, or limited by efficiency, or (as the authors imply) affected by the stability of the solver? Incorporation of physical processes in a box model is a large part of the paper and I think the introduction would be better if more could be elaborated here, e.g., on why these processes weren't fully implemented and their effects.

Response:

We apologize for any confusion that may have arisen. One of the primary challenges is the limitations of 0-D box model, which lacks a three-dimensional structure. This limitation restricts the model's capability to directly simulate physical processes. This issue could be addressed by minimizing uncertainty in the chemical reactions while iteratively approximating the values associated with physical processes based on observations and simulations. However, this approach necessitates a model with high computational efficiency. We are confident that the ROMAC framework is well-suited for this task. In response to your suggestion, we have included these points into the introduction of the revised manuscript.

Line38-42: However, it is important to consider the impact of physical transport on long-lived species, such as its effect on O_3 concentration (Li et al., 2021; Liu et al., 2022). The 0-D model, which lacks a 3-D structure, is unable to directly estimate the impact of physical processes (e.g., vertical and horizontal transport) on pollutant concentrations. Therefore, it is necessary to find a proper scheme to estimate the physical process for these models.

2. L109-110: "...explicit methods ... are difficult to solve these problems." can be worded more clearly, "...explicit methods ... cannot achieve a stable solution without using a timestep shorter than all lifetimes in the system, which is computationally infeasible."

Response:

Thank you very much for your suggestions. We have revised the sentences in the manuscript.

Line130-132: Therefore, the ODEs system of atmospheric chemical kinetics simulation is extremely stiff, and explicit methods (*e.g.,* explicit Euler method, explicit Runge-

Kutta method) cannot achieve a stable solution without using a timestep shorter than all lifetimes in the system, which is computationally infeasible.

3. L130: The ROMAC model uses a Diagonal-Simplified-Newton (DSN) method which approximates the inverse of the Jacobian. Is there a quantified estimate of how much error this will introduce and effects on stability?

Response:

Similar to the Simplified-Newton method, the DSN method eventually converge to the result. The ROMAC model actively manages its computational precision. We add a supplementary note to this part. However, due to variations in initial conditions among different simulations, providing an exact quantification of its error is challenging. In Section 2.4, we assessed ROMAC's error control capabilities by comparing its results with those obtained using high-precision solvers.

Under the error control scheme, ROMAC can run stably. The occurrence of instability (*e.g.,* non-convergence or excessively large error) is typically attributed to a too large integration time step, and ROMAC will adaptively shorten the integration time step.

Line158-160: The solution process was iterated until the difference between the results of two iterations was less than one-tenth of the preset truncation error tolerance (*etc.,* $0.1 \times atol$ or $0.1 \times rtol$) for ODEs solution.

4. L255: Specify the OS version, compilers & versions used.

Response:

Thanks for your suggestion. The details information of OS version and compilers have been added to the manuscript.

Line282-283: The operating system was 64-bit Ubuntu (version 20.04.1) and the software was compiled using Intel Fortran (ifort version 2021.2.0).

5. In Section 2.4 authors show the accuracy of the model as expressed in maximum relative error %. Does the % grow over time throughout the integration? It would be useful to show a time series plot.

Response:

Thank you for your insightful suggestion. Ensuring stable error control is indeed crucial to the integrity of our work. To address this, we have updated Appendix A by including a time-series plot of the error in Figure A2. Additionally, the rate of change of the error over time is also added in Figure A2. As illustrated in Figure A2(a), the relative error gradually stabilizes rather than diverging over time. Further, Figure A2(b) demonstrates that after 225,000 seconds, the growth rate of the relative error is extremely minimal, falling within a range of -1.0×10^{-6} %/s to 1.0×10^{-6} %/s.

Line300-303: The time series of relative error and its growth rates are depicted in Figure A2. The relative difference between the solution results of ROMAC and that of AtChem gradually stabilizes, and the rate of change of the relative error after 225,000 seconds is extremely minimal, falling within a range of -1.0×10^{-6} %/s to 1.0×10^{-6} %/s.

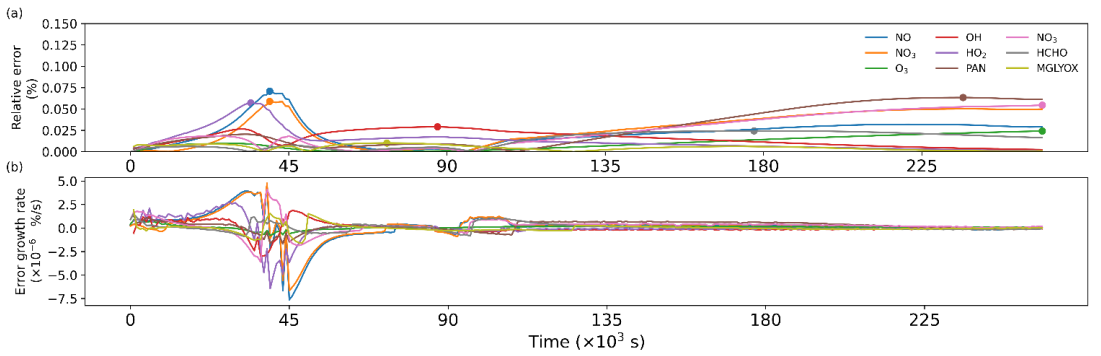

Figure A2. (a) Time series of relative errors, with dots marking the maximum values. (b) Growth rate of relative errors.

6. In the abstract authors claim a 96% improvement in computational efficiency in ROMAC compared to "other box models". It may be useful to say which, and at what expense in error (which is small but worth mentioning).

Response:

Thanks for your suggestion, we made the following changes to the abstract to show that we did not sacrifice too much computational accuracy while improving efficiency.

Line16-20: Since the development of a variable-step and variable-order numerical solver that eliminates the need for Jacobian matrix processing, ROMAC's computational efficiency has seen a marked improvement with only a marginal increase in error. Specifically, ROMAC's computational efficiency has improved by 96% when compared to several established box models, such as F0AM and AtChem. Moreover, the solver maintains a discrepancy of less than 0.1% when its results are compared to those obtained from a high-precision solver in AtChem.

Specific corrections:

1. L132 "specie" -> "species"

Response:

Sorry for the syntax error, the following changes have been made.

Line156: According to the equations associated with the implicit Euler method in Eq. (1) to Eq. (13), the iteration formula for species *i* is shown in Eq. (14).

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