Response to Reviewer 1

Dear Dr. Gianluca Armeli,

We greatly appreciate the efforts made by you to improve the quality of our manuscript (MS ID: egusphere-2025-1241). We have carefully reviewed and implemented all the comments provided by you and made significant revisions to the manuscript to address the concerns raised. In this response letter, your comments copied verbatim beneath are in black italic font, the author responses are in normal font, revised text is in blue, and line numbers refer to those in the Track Change manuscript.

General comments

The study in question presents a new model for the prediction of reaction rate constants of volatile organic compounds (VOCs). The authors used the reaction rate constant dataset by McGillen et al. to train a Siamese message passing neural network (MPNN) to predict these rate constants. The outcoming model was given the name "Vreact" and it was shown to outperform existing models for reaction rate constant prediction.

The dataset used in this study comprises 2802 gas-phase reaction rate constants for 1586 VOCs and 4 oxidants (• OH, • Cl, • NO3 and O3). The authors underline this diversity of oxidants as one of their advantages compared to previous models which only use a single oxidant per model. Because of the wide value range of reaction constants, the values were log-transformed. Vreact takes the SMILES string of the VOC and the oxidant as inputs, which is an established and modern approach in chem-informatics. Graph representations are generated from these inputs and fed to the neural network that creates the molecular feature tensors A and B. Further mathematical operations are executed to account for the effects of molecular interactions. Finally, the prediction value for the reaction rate constant is made.

Moreover, the authors evaluate how Vreact can contribute to the understanding of aerosol formation mechanisms. They showcase the oxidation of 2-methyl-4-penten-2-ol, discussing different reaction pathways and how the interaction layer of Vreact can be used for comprehension. Furthermore, the authors gathered more data from 2020 and onwards, which they called the 'post-2020 test set' to analyze the extrapolation ability of Vreact, leading to satisfactory results. Besides, more insights on the reaction rates of specific chemical classes are provided.

All in all, the article presents a modern and sustainable study. The Vreact model that is the key component of this work was built on well-established methods and principles and could overall convince with its performance. Vreact's advantages and improvements towards other models were clearly outlined in a comprehensible way. The study was conducted scientifically correct with no obvious shortcomings. Despite it being a rather data scientific topic, its atmospheric relevance became evident. The illustrations used are helpful and supporting. The supplementary material contains further details on the model architecture and is useful for a deeper understanding.

Another valuable resource is the web tool version of Vreact, reinforcing reproducibility and open data.

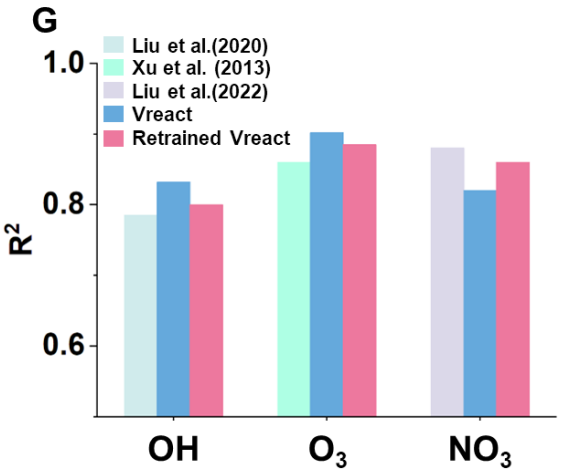
Response: Thank you for your thoughtful and constructive comments. We appreciate your recognition of the significance and quality of our work. We have provided detailed responses and made revisions addressing your comments.

Specific comments

After the results of the test set were presented, the authors provided more extensive evaluations and showcases of the model's abilities. First, they draw a more detailed comparison between Vreact and the existing single-oxidant models. Therefore, they use two independent approaches: 1) using the pre-trained Vreact to predict the test sets from the literature and 2) retraining Vreact on the original train/test splits of the literature. Approach 2) is a bullet-proof method that really isolates the model's predictive capability and delivers a nice comparison. Approach 1) has the potential problem, that the literature test sets contain data points that are part of Vreact's training set. This would be problematic, because generally, machine learning models perform significantly better on seen data, resulting in an unfair comparison. It would be appreciated, if the authors could address this issue briefly, since it was unmentioned in the text so far.

Response:

We thank the reviewer for highlighting the issue in Approach 1 regarding the partial overlap between the literature test sets and the Vreact training set, which could lead to an unfair performance comparison. We identified and removed the duplicate molecules (2 of 38 for NO_3, 13 of 35 for O_3, and 6 of 36 for OH) from the literature test sets. The revised R^2 values were recalculated and are now presented in the updated Figure 3G. While the R^2 values have decreased slightly (OH:0.024/O_3:0.016/NO_3:0.027), the overall comparative trends remain unchanged. To enhance clarity, we now refer to the modified test sets as "cleaned literature test sets" and the original ones as "original literature test sets" throughout the revised manuscript.



(G) R^2 comparison among previously published single-oxidant models, the original Vreact (evaluated on cleaned literature test sets), and Retrained Vreact (trained and tested using the

same original splits as the literature models) highlighting adaptability.

The revised main text lines 232-241:
The original text "Liu et al. (2020) for •OH (180 data points), Xu et al. (2013) for O_3 (95 data points), and Liu et al. (2022) for NO_3 radicals (189 data points). " has been revised to "Liu et al. (2020) for OH (training/test = 144/36), Xu et al. (2013) for O_3 (60/35), and Liu et al. (2022) for NO_3 radicals (151/38)."
"To ensure a fair comparison, overlapping data points between the literature test sets and the McGillen training set were removed (2 of 38 for NO_3, 13 of 35 for O_3, and 6 of 36 for OH). Second, Vreact was retrained on each literature dataset using their original train/test splits (Retrained Vreact), allowing a direct comparison with published models on original literature test sets." has been added.

Technical corrections
No typing errors or other technical problems were found.
Response: Thank you for checking for typing errors or other technical problems.

Finally, we would like to thank you again for your great efforts on improving the quality of this manuscript.

Thank you very much,
Yours sincerely,
Xian Liu