

Manuscript ID egusphere-2025-1364, entitled "Criegee + HONO reaction: the dominant sink of Criegee, and the missing non-photolytic source of OH". The work is interesting to understand the atmospheric oxidation capacity of Criegee in the atmosphere. Although the investigation claimed a new source of OH and the sink of HO₂ and suggested that HONO may be the dominant sink of Criegee, the claims could not be based on scientific foundation. Therefore, the importance of Criegee + HONO should be turned down.

- (1) The atmospheric lifetimes of CH₂OO with water dimer had been detailedly investigations from theoretical and experimental methods. Please read the reference (J. Am. Chem. Soc. 2021, 143, 8402-8413). The atmospheric lifetime is 2.12×10^{-4} s at 0 km in Table 7 in J. Am. Chem. Soc. 2021, 143, 8402-8413. Although the atmospheric lifetime of CH₂OO with water dimer is very long in the stratosphere, the concentrations of Criegee intermediates are very low at altitude above 15 km. Therefore, the importance of Criegee intermediates occurs in the troposphere. According to Table 1 in the present work, I assume that the concentration of HONO is about 10^{10} molecules cm⁻³ in the troposphere, which leads to the atmospheric lifetime of CH₂OO with HONO is about 10² s. This shows that HONO does not make any contribution to the sink of CH₂OO. In addition, Criegee intermediates are mainly produced from the ozonolysis of BVOCs, while HONO is mainly produced at urban regions.
- (2) The second issue is computational methods. In fact, there are dozens of papers that have shown that post-CCSD(T) calculations are required to obtain quantitative barrier heights for the reactions including Criegee intermediates. Although I have to admit the introduction of the calculations will extremely increase the computational costs, it should be clearly explained and reviewed in the present progress. This is very helpful for potential readers to know the progress. Please read these articles (J. Am. Chem. Soc. 2025, 147 (14), 12263-12272.; Atmos. Environ. 2025, 341, 120928.; Research **2024**, 7, 0525.; Fundam. Res. **2024**, 4 (5), 1216-1224.; Proc. Natl. Acad. Sci. USA 2018, 115, 6135-6140. And so on).
- (3) In kinetics calculations, there are still lots of factors that do not consider such as recrossing effects, torsional anharmonicity, and anharmonicity. In addition, the low energy barrier, what is the rate-determining step. I guess that the formed pre-reactive complex is the rate-determining step like Criegee reaction with HCOOH, Therefore, VRC-TST is necessary for the barrierless process.
- (4) Lines "Our study also suggests that HONO has the potential to become the most dominant sink of Criegee intermediate, surpassing SO₂ and water dimer, even in high humid condition", it is not validated.
- (5) Lines "the bimolecular reaction paths can be the main sink of sCI (Osborn and Taatjes, 2015; Lin et al., 2015; Sheps et al., 2014; Vereecken and Francisco, 2012)." Some important key references have been missed such as . J. Am. Chem. Soc. 2016, 138, 14409-14422. and J. Am. Chem. Soc. 2021, 143, 8402-8413.
- (6) Kinetics methods should be moved into computational section.
- (7) It needs to add some tables for showing the atmospheric lifetimes of Criegee intermediates with H₂O, (H₂O)₂, SO₂, HCOOH, and HONO as the function of altitude.