Reviewer 2

General comment

Xu et al. systematically studied the reaction mechanisms and kinetics of the ·Cl + imines reactions using computational methods. The authors found that imine-derived N-centered radicals exhibits various fates under tropospheric conditions, which significantly differs from the behavior of previously reported amines. This is an interesting study and the paper is a nice addition to the literature on the oxidation of organic nitrogen compounds. I believe this paper is fitting well in ACP. I can recommend publication after the following comments have been addressed.

Response: We highly appreciate your insightful and helpful comments on our manuscript. We have revised the manuscript to enhance its quality.

Special Suggestions and Comments

(1) *I'm confused why two different kinetic simulation software programs are used for the initial and subsequent reactions*

Response: Thanks for the comment. In fact, all the reaction rate constants were calculated using MultiWell 2014.1 software. In the revised manuscript, the sentences have change to:

MultiWell 2014.1 software was employed to simulate the reaction kinetics (Barker et al., 2014; Barker, 2001).

(Please see Lines 125-126)

(2) *Not clear to this reviewer which reaction steps were simulated using MultiWell program, and which were simulated using the MESMER program.*

Response: We appreciated the comment and agree that the statement is not clear here. As discussed in the response on the comment (1), we have rewritten the sentence. Please see Lines 125-126.

(3) *I am missing labels (A)~(E) in the Table 1.*

Response: Thanks. We have corrected it. (Please see Table 1)

(4) *The branch ratio in Table 1 does not appear to be 1.*

Response: Thanks. We have corrected it. (Please see Table 1)

(5) *The overall reaction rate constants in the abstract are reported with a unit factor of* $\times 10^{-11}$ cm^3 $molecule^{-1}$ s^{-1}, *while the same constants in Table 1 use a factor of* $\times 10^{-10}$ cm^3 $molecule^{-1}$ s^{-1}. *Please ensure consistency throughout the manuscript.*

Response: Thanks. We have corrected it. (Please see Table 1)

(6) *Missing reference for Molclus program.*

Response: Thanks. We have corrected it. (Please see Line 98 and reference Lu, 2025)

(7) *The parameters used in the long-range transition-state theory were not shown in the manuscript.*

Response: As discussed in the response on the Reviewer 1 comment (5), we added the parameters used in the LRTST to Table S1 in the revised SI.

(8) *I suggest some important results can be moved from SI to the main text, for example, Figure S3 and S4.*

Response: Thanks for the comment and suggestion! We have combined Figures S3 and S4 into Figure 1 and included it in the revised manuscript.

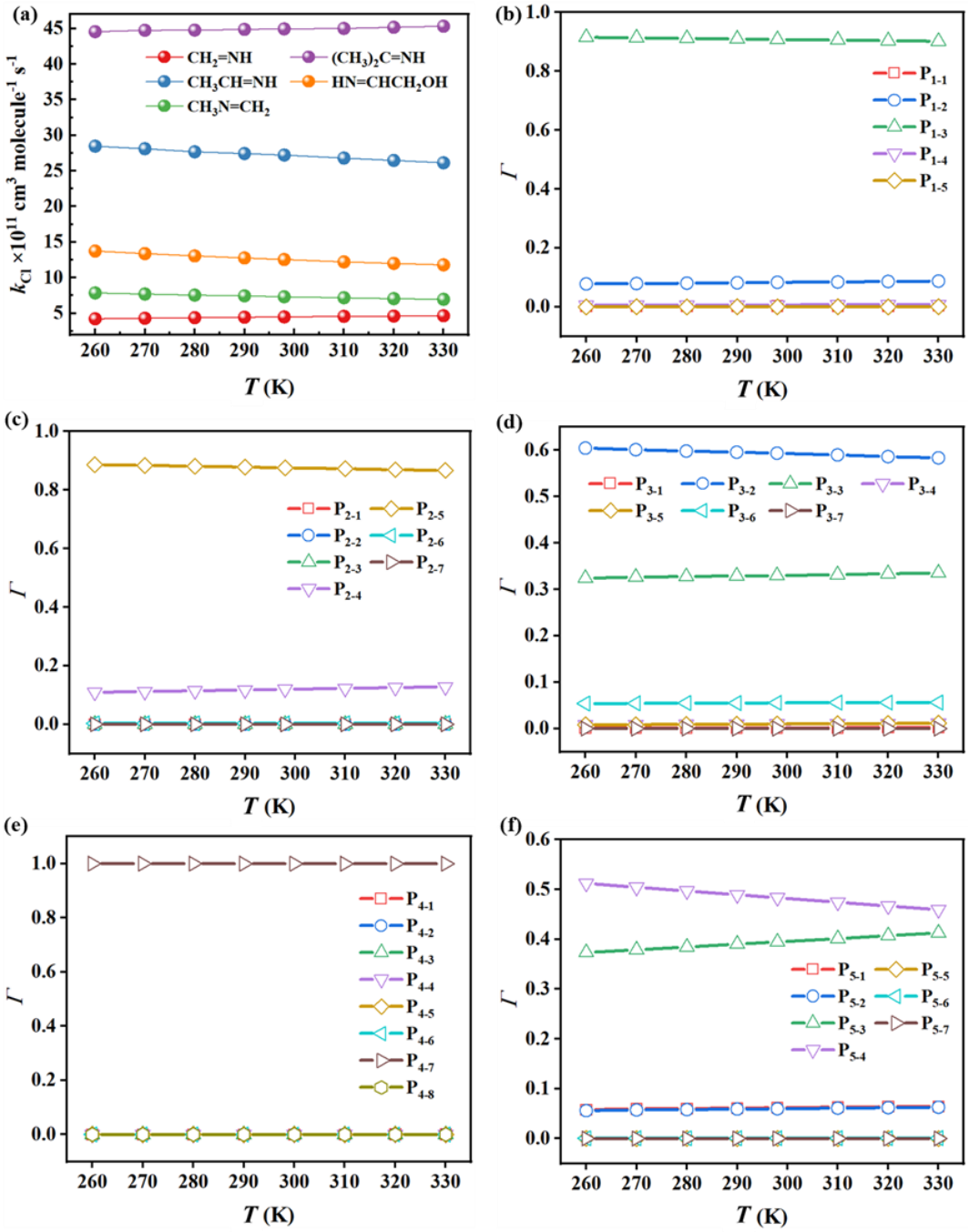

Figure 1. Reaction rate constants (k_{Cl}) for the reactions of five imines (a) and branching ratios (Γ) for the species involved in the reactions of (b) CH_2=NH, (c) CH_3CH=NH, (d)

$CH_3N{=}CH_2$, (e) $(CH_3)_2C{=}NH$, and (f) $HN{=}CHCH_2OH$ initiated by ·Cl in the temperature range of 260 - 300 K and 1 atm.