Reviewer 1 2 The authors are presenting an innovative approach to address two well-known recycling 3 issues. This starts from improving the reactor, particularly, to prevent UV photon leaks 4 and introduce excess NO. 5 The manuscript is easy to follow and has merits to the atmospheric chemistry 6 7 community particularly who intends applying the CRM method for their field observations or using the dataset for the research. I would recommend the publication 8 of this manuscript after addressing following points. 9 Reply: We would like to thank the reviewer for the insightful comments, which 10 helped us tremendously in improving the quality of our work. Please find the response 11 to individual comments below. 12 13 1. The reaction of CO with OH will produce HO₂, which ends up recycling OH in the 14 excess NO environment. Please include the discussion in the manuscript. 15 16 Reply: Many thanks, we have included the discussion in the manuscript. We added a sentence in the revised manuscript (Line 338-340): Similarly, the 17 produced HO₂ from the reactions of CO and SO₂ with OH will end up recycling 18 OH in the excess NO environments and thus reduce the fitting slopes. 19 20 21 2. It is certainly a good start to test the ICRM reactor with three VOCs but likely VOCs with various function groups required to evaluate. If the authors could suggest the list 22 of compounds to be tested and provide justification, that would trigger follow up studies 23 24 from other research groups. 25 Reply: We appreciate the reviewer for the comment. In addition to the four individual VOC species, we also calibrate using a mixed gas standard with 16 VOC 26

trimethylbenzene, phenol, m-cresol, naphthalene. The calibration slope is close to those 29

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Responses to Reviewers

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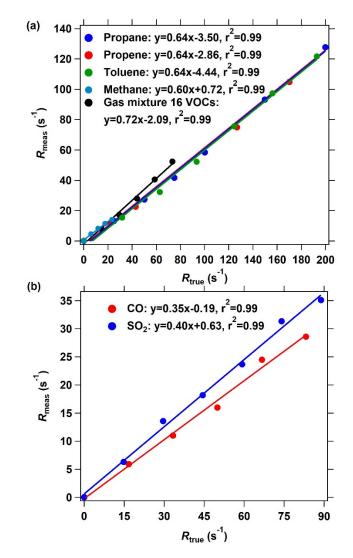
species, namely acetaldehyde, methanol, ethanol, isoprene, acetone, acetonitrile,

methyl vinyl ketone, methyl ethyl ketone, benzene, toluene, o-xylene, α-pinene, 1,2,4-

of the three individual VOC species, indicating that the $RO_2 + NO$ reactions for these 30 investigated VOCs should be similar. In addition, we also calibrated methane in the 31 revised manuscript and the results were added in Figure 4. Nevertheless, we agree with 32 the reviewer that it is necessary to calibrate more VOC species in the future, especially 33 considering that different VOCs species dominate in different environment, such as 34 forest areas and various emission sources. For example, isoprene and terpenes have 35 high reactivity contribution in forests and rural sites. Therefore, isoprene, α -Pinene and 36 β-Pinene is suggested to be calibrated in the following study. Typical branched olefin, 37 other aromatics (such as ethylbenzene) and important oxygenated VOCs (such as 38 formaldehyde and glyoxal) should also be calibrated in the future. 39

We added the following sentences in the revised manuscript (Line 404-414): In 40 this study, we calibrated four individual representative VOC species (methane, 41 propane, propene, toluene). In addition, we also calibrated the mixed standard 42 gases with 16 VOC species including representative oxygenated VOCs 43 (acetaldehyde, methanol, ethanol, acetone, acetonitrile, methyl vinyl ketone, 44 45 methyl ethyl ketone), biogenic VOCs (isoprene, α -pinene), typical aromatics (benzene, toluene, o-xylene, 1,2,4-trimethylbenzene, naphthalene, phenol, m-46 cresol). The calibration slope is close to those of the four individual VOC species, 47 indicating that the RO₂ + NO reactions for these investigated VOCs should be 48 similar. Nevertheless, given that there are different VOCs compositions in 49 different environment such as forest, urban area and emission sources, 50 calibrations for more individual VOCs species might be also needed. 51

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Figure 4. The OH reactivity calibration of the improved CRM system using different standard gases. (a) The calibrating results of organic species including methane, propane, propene, toluene, and a mixture of 16 VOC species through arm C. (b) The calibrating results of inorganic species including CO and SO₂. The measured OH reactivity was calculated based on the C2 mode shown in Fig. 2 in the ICRM system.