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The improved comparative reactivity method (ICRM): measurements of OH reactivity under high-Title:

NOx conditions in ambient air

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Abstract:

The comparative reactivity method (CRM) was developed more than a decade to measure OH reactivity (i.e., OH loss frequency) in both laboratory and field studies. However, accurate OH reactivity quantification remains challenging under real ambient conditions, especially for OH reactivity measurements in high-NOx (e.g., > 10 ppbv) environments, as ambient NO enhances the regeneration of OH radicals in the CRM reactor. To solve this problem, we design a new and improved CRM reactor (ICRM) and add NO into the system continuously so that the HO2 radical concentration is suppressed. We confirmed the appropriate level of NO by determining the maximum decrease in the pyrrole level caused by regenerated OH radicals from NO + HO2. RO2 radicals induced by volatile organic compounds (VOCs) in the ICRM reactor were also found to react with NO, which led to the regeneration of OH radicals and thus the underestimation of OH reactivity. This effect was quantified by the calibration of representative VOC species at different NO levels, and the correction coefficients obtained were used to correct the measured OH reactivity. All these efforts resulted in reducing the uncertainty of the NO-artifact correction by at least an order of magnitude compared to the original CRM system. Additionally, these technological improvements also considerably reduced the systematic errors from pyrrole photolysis in the original system. A new operation mode was proposed for the ICRM, which is able to avoid the interference resulting from OH radicals produced by photolysis of residual humidity and save time for ambient measurement. The ICRM system was employed in a field campaign to measure OH reactivity and performed well with ambient NO levels ranging from 0 to 50 ppbv, which are typically observed in the urban and suburban atmosphere.

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