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The Comparative Reactivity Method – a new tool to measure total OH Reactivity in ambient air

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Abstract. Hydroxyl (OH) radicals play a vital role in maintaining the oxidizing capacity of the atmosphere. To understand variations in OH radicals both source and sink terms must be understood. Currently the overall sink term, or the total atmospheric reactivity to OH, is poorly constrained. Here, we present a new on-line method to directly measure the total OH reactivity (i.e.~total loss rate of OH radicals) in a sampled air mass. In this method, a reactive molecule (X), not normally present in air, is passed through a glass reactor and its concentration is monitored with a suitable detector. OH radicals are then introduced in the glass reactor at a constant rate to react with X, first in the presence of zero air and then in the presence of ambient air containing VOCs and other OH reactive species. Comparing the amount of X exiting the reactor with and without the ambient air allows the air reactivity to be determined. In our existing set up, X is pyrrole and the detector used is a proton transfer reaction mass spectrometer. The present dynamic range for ambient air reactivity is about 6 to 300 s^{-1}, with an overall maximum uncertainty of 25% above 8 s^{-1} and up to 50% between 6–8 s^{-1}. The system has been tested and calibrated with different single and mixed hydrocarbon standards showing excellent linearity and accountability with the reactivity of the standards. Potential interferences such as high NO in ambient air, varying relative humidity and photolysis of pyrrole within the setup have also been investigated. While interferences due changing humidity and photolysis of pyrrole are easily overcome by ensuring that humidity in the set up does not change drastically and the photolytic loss of pyrrole is measured and taken into account, respectively, NO>10 ppb in ambient air remains a significant interference for the current configuration of the instrument. Field tests in the tropical rainforest of Suriname (~53 s) and the urban atmosphere of Mainz (~10 s^{-1}) Germany, show the promise of the new method and indicate that a significant fraction of OH reactive species in the tropical forests is likely missed by current measurements. Suggestions for improvements to the technique and future applications are discussed.

Final Revised Paper (PDF, 1576 KB)   Discussion Paper (ACPD)

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