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Title: Intercomparison of the comparative reactivity method (CRM) and pump-probe technique for

measuring total OH reactivity in an urban environment

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Abstract:

The investigation of hydroxyl radical (OH) chemistry during intensive field campaigns has led to the development of several techniques dedicated to ambient measurements of total OH reactivity, which is the inverse of the OH lifetime. Three techniques are currently used during field campaigns, including the total OH loss rate method, the pump-probe method, and the comparative reactivity method. However, no formal intercomparison of these techniques has been published so far, and there is a need to ensure that measurements of total OH reactivity are consistent among the different techniques. An intercomparison of two OH reactivity instruments, one based on the comparative reactivity method (CRM) and the other based on the pump-probe method, was performed in October 2012 in a NOx-rich environment, which is known to be challenging for the CRM technique. This study presents an extensive description of the two instruments, the CRM instrument from Mines Douai (MD-CRM) and the pump-probe instrument from the University of Lille (UL-FAGE), and highlights instrumental issues associated with the two techniques. It was found that the CRM instrument used in this study underestimates ambient OH reactivity by approximately 20 % due to the photolysis of volatile organic compounds (VOCs) inside the sampling reactor; this value is dependent on the position of the lamp within the reactor. However, this issue can easily be fixed, and the photolysis of VOCs was successfully reduced to a negligible level after this intercomparison campaign. The UL-FAGE instrument may also underestimate ambient OH reactivity due to the difficulty to accurately measure the instrumental zero. It was found that the measurements are likely biased by approximately 2 s-1, due to impurities in humid zero air. Two weeks of ambient sampling indicate that the measurements performed by the two OH reactivity instruments are in agreement, within the measurement uncertainties for each instrument, for NOx mixing ratios up to 100 ppbv. The CRM technique has hitherto mainly been used in low-NOx environments, i.e. environments with ambient NOx mixing ratios lower than a few poby, due to a measurement artifact generated by ambient NO inside the sampling reactor. However, this study shows that this technique can also be used under NOx-rich conditions if a NOx-dependent correction is carefully applied on the OH reactivity measurements. A full suite of 52 VOCs, NOx, and other inorganic species were monitored during this intercomparison. An investigation of the OH reactivity budget for this urban site suggests that this suite of trace gases can account for the measured total OH reactivity.

Description: Only IISERM authors are available in the record.

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