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Title: Intercomparison of two comparative reactivity method instruments inf the Mediterranean basin

during summer 2013

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Abstract:

The hydroxyl radical (OH) plays a key role in the atmosphere, as it initiates most of the oxidation processes of volatile organic compounds (VOCs), and can ultimately lead to the formation of ozone and secondary organic aerosols (SOAs). There are still uncertainties associated with the OH budget assessed using current models of atmospheric chemistry and direct measurements of OH sources and sinks have proved to be valuable tools to improve our understanding of the OH chemistry. The total first order loss rate of OH, or total OH reactivity, can be directly measured using three different methods, such as the following: total OH loss rate measurement, laserinduced pump and probe technique and comparative reactivity method. Observations of total OH reactivity are usually coupled to individual measurements of reactive compounds in the gas phase, which are used to calculate the OH reactivity. Studies using the three methods have highlighted that a significant fraction of OH reactivity is often not explained by individually measured reactive compounds and could be associated to unmeasured or unknown chemical species. Therefore accurate and reproducible measurements of OH reactivity are required. The comparative reactivity method (CRM) has demonstrated to be an advantageous technique with an extensive range of applications, and for this reason it has been adopted by several research groups since its development. However, this method also requires careful corrections to derive ambient OH reactivity. Herein we present an intercomparison exercise of two CRM instruments. CRM-LSCE (Laboratoire des Sciences du Climat et de l'Environnement) and CRM-MD (Mines Douai), conducted during July 2013 at the Mediterranean site of Ersa, Cape Corsica, France. The intercomparison exercise included tests to assess the corrections needed by the two instruments to process the raw data sets as well as OH reactivity observations. The observation was divided in three parts: 2 days of plant emissions (8-9 July), 2 days of ambient measurements (10-11 July) and 2 days (12-13 July) of plant emissions. We discuss in detail the experimental approach adopted and how the data sets were processed for both instruments. Corrections required for the two instruments lead to higher values of reactivity in ambient air; overall 20% increase for CRM-MD and 49% for CRM-LSCE compared to the raw data. We show that ambient OH reactivity measured by the two instruments agrees very well (correlation described by a linear least squares fit with a slope of 1 and R2 of 0.75). This study highlights that ambient measurements of OH reactivity with differently configured CRM instruments yield consistent results in a low NOx (NO + NO2), terpene rich environment, despite differential corrections relevant to each instrument. Conducting more intercomparison exercises, involving more CRM instruments operated under different ambient and instrumental settings will help in assessing the variability induced due to instrument-specific corrections further.

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