

Collaborative PhD project between Universities of Lille and Kyoto

Understanding the Atmospheric Chemistry of Peroxy Radicals through Laboratory and Field Studies

Gas-phase chemical reactions are important in a wide variety of environments such as combustion, Earth's atmosphere, and dense interstellar clouds. Modelling such environments requires a detailed understanding of the mechanisms and rate coefficients of key reactions that define the system. In the atmosphere, the oxidation mechanism of volatile organic compounds (VOC) influences on a regional scale the air quality and hence human health. On a global scale it affects the oxidizing capacity of the atmosphere and with this the global climate: on one hand through effects on the lifetime of greenhouse gases such as methane, and on the other hand via tropospheric ozone chemistry and formation of secondary organic aerosols (SOA). In combustion systems, the fuel oxidation mechanism influences the efficiency of the process and the formation of pollutants, and so is intimately linked to the chemistry of the atmosphere.

In general, these gas-phase chemical systems are driven by reactions of short-lived intermediates such as OH, HO₂ and RO₂ (peroxy) radicals. Different approaches are combined to improve the understanding of this chemistry. Field campaigns gathering different research groups together at a well-selected place will collect data such as radical and trace gas concentration and meteorological data for defined periods (often several weeks). Subsequent modelling of these data using an appropriate model will try reproducing the experimental time traces of the field measurements. Disagreement between model and measurements can then indicate chemistry that is not well described in the model. At this point, laboratory measurements jump in to improve the knowledge of these processes.

The current PhD project is involved in all three aspects: (a) the major task will consist in laboratory experiments in Lille designed to elucidate the chemistry of peroxy radicals using a sophisticated set-up consisting of laser photolysis coupled to two different detection techniques: cavity ring down spectroscopy (cw-CRDS) for the quantification of peroxy radicals and laser induced fluorescence (LIF) for the time-resolved, relative detection of OH radicals. Setting-up a third technique, broad-band UV absorption spectroscopy for a complementary detection of peroxy radicals, halogen species or Criegee intermediates, is planned in the frame of this PhD. Through a secondment of 3-6 months to Kyoto University during the 2nd or 3rd year, the student will get inside into the two other aspects: (b) participating in a field campaign in Japan (if timing permits) and (c) modelling of field campaign data under the supervision of Prof. Yoshizumi Kajii.

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