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The sensitivity of natural cloud water to photochemical processes

A. Marinoni (1,4), M. Parazols (1,2), P. Amato (1,2,3), A.M. Delort (3), S. Zappoli (4), P. Laj (2), **G. Mailhot (1)**

(1) LPMM, CNRS-University of Clermont-Ferrand, France, (2) OPGC-LaMP, CNRS-University of Clermont-Ferrand, France, (3) SEESIB, CNRS-University of Clermont-Ferrand, France, (4) Department of physical and inorganic chemistry, university of Bologna, Italy, (gilles.mailhot@univ-bpclermont.fr / Fax: +33 473 405136 / Phone: +33 473 407173)

Formaldehyde and hydrogen peroxide are key constituents of atmospheric photochemical processes, in particular those involved in the HO_x and NO_x cycles. Direct photolysis of hydrogen peroxide is a source of radicals in both gas and aqueous phases and could therefore be used as an indicator of photochemical activity. Formaldehyde is both a primary pollutant and is produced by oxidation of organic compounds by reactions that can take place both in gas and liquid phases. As such, it can be considered an indicator of organic matter state of oxidation. Quantifying the behaviour of these 2 species in clouds is therefore of importance to better constraining the relative importance of gaseous versus liquid phase processes and to check the validity of current cloud chemistry models.

In this study, we have used four years of cloud samples collected at the puy de Dôme atmospheric station to investigate the seasonal and diurnal dependence of both H_2O_2 and HCHO concentrations. A significant seasonal and diurnal dependence of H_2O_2 concentration in cloud droplets is found with maxima during summer time and around noon time. This shows that solar radiation is one of the most important factors that drive the cloud aqueous hydrogen peroxide concentration. On the contrary, the aqueous concentrations of HCHO do not appear to be linked to solar intensity.

In addition, cloud samples collected at puy de Dôme have been used in laboratory experiments to better understand chemical phase processes. Both natural cloud samples and artificial cloud solutions were irradiated in a photo-reactor and the variations

of pH, redox potential and key atmospheric substances (HCHO, H_2O_2, dissolved organic carbon (DOC), and carboxylic acids) were continuously monitored during the experiments. The behaviour of cloud samples differs significantly from that of artificial solutions. H_2O_2 concentration during the irradiations shows a decrease of about 15% h^{-1} while it usually increases for artificial solutions. DOC is not varying, while the behaviour of carboxylic acids and formaldehyde appears to depend on the cloud pH.

These results lead to conclusion that H_2O_2 in cloud is mainly produced by gas-to-liquid transfer and subsequently photo-chemically consumed in the cloud water. Instead, the processes controlling the concentration of HCHO are more complex and clearly change with cloud pH, and hence the degree of anthropogenic influence of the air mass.