Ecole Doctorale des Sciences Fondamentales

Title of the thesis: Fate of trace gases and aerosol in the cloudy atmosphere.

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Summary : Clouds, but also fogs, cover 70% of the Earth's surface. They can modify the budget of trace gas compounds and aerosols via various physicochemical processes including: the trapping of soluble compounds in the liquid phase by mass transfer, (2) the activation of some aerosols into droplets, the soluble fraction of these dissolving in the droplet, (3) chemical and biological transformations of these compounds, (4) the redistribution of compounds between the different phases when the cloud evaporates, (5) the wet deposition, a net sink for gases and aerosols (Ervens, 2015). Their oxidation leads to less volatile compounds including potentially oligomers which are components of the secondary organic aerosol (SOA). In addition, 90% of the clouds dissipate, leading to the evaporation of the most volatile species and the condensation of the least volatile ones. However the results from the field are contrasted. While some studies show the significant formation of SOA via aqueous phase processes (aqSOA according to Ervens et al., 2011), others do not show a significant change between the amounts of residual and interstitial organic aerosol (Shrivastava et al., 2011). Moreover while some studies have shown in the past that clouds could have an impact on ozone in the gas phase through the effective dissolution of the Hox radical (Lelieveld and Crutzen, 1991; Monod and Carlier , 1999), the significance of this effect remains controversial and uncertain (Liang and Jacob, 1997). Ozone depletion of around 30% has already been observed at the altitude of tropical cirrus clouds activated by the presence of ice particles (Roumeau et al., 2000). Finally, these results reflect the lack of knowledge of the processes in the cloudy atmosphere. As a consequence, the international community working on this subject has recently highlighted the need of implementing model intercomparison exercises and dedicated campaigns to fill these gaps (Lance et al., 2017). These processes are expected to affect aerosol-cloud interactions, notably by modifying the CCN properties of aerosols. This is one of the major uncertainties in the estimation of radiative forcing according to the latest IPCC report (IPPC, 2013).

The objective of the thesis will be to better understand the role of clouds on the atmospheric chemistry of trace gas compounds and aerosols from observations collected at the Puy de Dôme observatory (center of France), a real natural laboratory. The work will consist of the implementation of dedicated observation campaigns thanks to the new high resolution mass spectrometry set MOCCA (Mass spectrOmetry for the multiphasic Composition of the Cloudy Atmosphere) for measuring organic matter and a climatological study from existing observations. The project if part of the European project ATMO-ACCESS for which a field campaigns hosting international researchers will be implemented at the PUY station.