Geophysical Research Abstracts
Vol. 16, EGU2014-12061, 2014
EGU General Assembly 2014

Ozone adsorption on carbon nanoparticles

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Carbonaceous particles produced by incomplete combustion or thermal decomposition of hydrocarbons are ubiquitous in the atmosphere. On these particles are adsorbed hundreds of chemical species. Those of great concern to health are polycyclic aromatic hydrocarbons (PAHs). During atmospheric transport, particulate PAHs react with gaseous oxidants. The induced chemical transformations may change toxicity and hygroscopicity of these potentially inhalable particles. The interaction between ozone and carbon particles has been extensively investigated in literature. However ozone adsorption and surface reaction mechanisms are still ambiguous. Some studies described a fast catalytic decomposition of ozone initiated by an atomic oxygen chemisorption followed by a molecular oxygen release [1-3]. Others suggested a reversible ozone adsorption according to Langmuir-type behaviour [4,5]. The aim of this present study is a better understanding of ozone interaction with carbon surfaces. An aerosol of carbon nanoparticles was generated by flowing synthetic air in a glass tube containing pure carbon (primary particles $<$ 50 nm), under magnetic stirring. The aerosol was then mixed with ozone in an aerosol flow tube. Ozone uptake experiments were performed with different particles concentrations with a fixed ozone concentration. The influence of several factors on kinetics was examined: initial ozone concentration, particle size (50 nm $\leq$ D_p $\leq$ 200 nm) and competitive adsorption (with probe molecule and water).

The effect of initial ozone concentration was first studied. Accordingly to literature, it has been observed that the number of gas-phase ozone molecules lost per unit particle surface area tends towards a plateau for high ozone concentration suggesting a reversible ozone adsorption according to a Langmuir mechanism. We calculated the initial reaction probability between O_3 and carbon particles.An initial uptake coefficient of 1.10^{-4} was obtained. Similar experiments were realized by selecting the particles size with a differential mobility analyser. We observed a strong size-dependent increase in reactivity with the decrease of particles size. This result is relevant for the health issues. Indeed the smallest particles are most likely to penetrate deep into the lungs.

Competitive reactions between ozone and other species like H_2O or atomic oxygen were also considered. Oxygen atoms were generated by photolysis of O_3 (or O_2) and were chosen because it is believed to form the same reactive oxygen intermediates than ozone. A weak water physisorption on soot was observed revealing hydrophobic properties of particles. Oxygen atoms were found to be strongly reactive. A Langmuir behavior was observed for oxygen atoms adsorption on carbon particles and we were able to determine an initial uptake coefficient of approximately 2.10^{-2}.

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