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Changes of fatty acid aerosol hygroscopicity induced by ozonolysis under humid conditions

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Abstract. Unsaturated fatty acids are important constituents of the organic fraction of atmospheric aerosols originating from biogenic or combustion sources. Oxidative processing of these may change their interaction with water and thus affect their effect on climate. The ozonolysis of oleic and arachidonic acid aerosol particles was studied under humid conditions in a flow reactor at ozone exposures close to atmospheric levels, at concentrations between 0.5 and 2 ppm. While oleic acid is a widely used proxy for such studies, arachidonic acid represents polyunsaturated fatty acids, which may decompose into hygroscopic products. The hygroscopic (diameter) growth factor at 93% relative humidity (RH) of the oxidized arachidonic particles increased up to 1.09 with increasing RH during the ozonolysis. In contrast, the growth factor of oleic acid was very low (1.03 at 93% RH) and was almost invariant to the ozonolysis conditions, so that oleic acid is not a good model to observe oxidation induced changes of hygroscopicity under atmospheric conditions. We show for arachidonic acid particles that the hygroscopic changes induced by humidity during ozonolysis are accompanied by about a doubling of the ratio of carboxylic acid protons to aliphatic protons. We suggest that, under humid conditions, the reaction of water with the Criegee intermediates might open a pathway for the formation of smaller acids that lead to more significant changes in hygroscopicity. Thus the effect of water to provide a competing pathway during ozonolysis observed in this study should be motivation to include water, which is ubiquitously present in and around atmospheric particles, in future studies related to aerosol particle aging.

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