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Relating physical state and reactivity: humidity dependent ozone uptake on tannic and shikimic acid

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Atmospheric aerosols are an important focus of environmental research due to their effect on climate and human health. Recent findings show that organic aerosol particles are capable of forming amorphous solids and semi-solids under atmospheric conditions [1]. Such particles should be highly viscous, leading to low diffusivity within the bulk. This would then slow down eventual chemical reactions in the bulk, thereby increasing the lifetime of the organic compounds involved. First indications of such behavior were recently shown for the reaction of thin protein films with ozone [2].

To investigate the influence of the physical state on the reactivity of atmospherically relevant compounds, the uptake of ozone on two different organics was measured using a coated wall flow tube system. The investigated organic compounds are tannic acid, which is a proxy for atmospheric polyphenolic materials, and shikimic acid, a constituent of biomass burning aerosols. The viscosity of the organic film was adjusted by varying the humidity of the system, assuming a correlation between the two parameters due to water acting as a plasticizer. The investigated humidity range was 0% - 95% RH for tannic and 0% - 92% RH for shikimic acid. It was found that both of the compounds show a long term uptake of ozone which lasts for more than 20 h. The uptake coefficient is clearly humidity dependent and increases by close to two orders of magnitude between driest and wettest conditions. At a given humidity, shikimic acid is the more reactive compound.
The measured humidity dependence supports the hypothesis that the uptake coefficient is a strong function of the diffusion coefficient of ozone in the organic film.

1. Virtanen, A., et al., An amorphous solid state of biogenic secondary organic aerosol particles. Nature, 2010. 467(7317): p. 824-827.
2. Shiraiwa, M., et al., Gas uptake and chemical aging of semisolid organic aerosol particles. Proceedings of the National Academy of Sciences of the United States of America, 2011. 108(27): p. 11003-11008.