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Protein cross-linking and oligomerization through dityrosine formation upon exposure to ozone

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Air pollution is a potential factor for the increasing prevalence of allergic diseases. Airborne allergenic proteins can be directly exposed to air pollution promoting post-translational modifications, which can enhance the allergenic potential of proteins. The formation of dimers or oligomers of allergenic proteins has been reported to result in an enhanced allergenicity. However, the oligomerization process for proteins at atmospherically relevant concentration of O_3 is still largely unknown.

In this study, the kinetics and reaction mechanism of protein oligomerization upon ozone exposure were studied at atmospherically relevant ozone concentrations and relative humidity (RH) in coated-wall flow tube experiments. Bovine Serum Albumin (BSA) was used as a model protein. Protein ozone exposure was studied for different protein phase-states, i.e. amorphous solid (45% RH experiments), semi-solid (96% RH experiments) and liquid (bulk solution experiments) to account for the differences of phase in atmospheric particulates, e.g., aerosol particles and cloud droplets. Product analysis was performed using a size exclusion chromatography-high performance liquid chromatography-diode array detector (SEC-HPLC-DAD).

We demonstrate that protein cross-linking upon ozone exposure can be attributed to the formation of covalent intermolecular dityrosine species by gel electrophoretic and fluorescence spectroscopic methods. The exposure experiments indicate that in addition to ozone concentration, the oligomerization process was depending on the phase-state of protein. In liquid-phase experiments, dimer formation was significantly enhanced, thus indicating a potential relevance of in-cloud processes for protein oligomerization. The reactive turnover is higher at 96% RH compared to 45% RH, indicating a higher bulk diffusion coefficient at high RH, which is explicitly resolved by kinetic modeling. Further, the reactive turnover showed a strong correlation to particle surface-to-volume ratio, confirming the bulk diffusion limitation.