

Investigation of the SOA formation potential of particulate photosensitized oxidation processes

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Processes leading to the formation and growth of secondary organic aerosol (SOA) are an intensively researched topic in atmospheric science. Despite major progress, atmospheric chemistry models based on available parametrizations of laboratory studies are still underestimating SOA masses compared to the observed SOA masses from field studies (Hallquist et al., 2009). This discrepancy between ambient and modeled SOA mass concentrations indicates that unaccounted SOA sources and formation processes are missing.

A potential SOA source that has often been neglected so far, is photosensitized particulate-phase chemistry, which was recently shown to contribute to SOA formation (Aregahegn et al., 2013; Monge et al., 2012; Rossignol et al., 2014; Wang et al., 2020). In such processes, a variety of reaction channels is opened through light-absorbing compounds, so-called photosensitizers (PS), which absorb solar radiation and transfer their excess energy to other compounds such as oxygen, halogenides (Tinel et al., 2014) or organic compounds (i.e., volatile organic compounds, VOCs). The photosensitized particulate-phase reactions can occur either within or at the interface of the aerosol particle resulting in SOA formation, growth, and processing (Figure 1).

However, data on these photochemical mechanisms, their kinetics as well as their potential contribution to SOA formation, growth, and aging are sparsely available for the particle/aqueous phase while their presence on atmospheric particles was already reported (Teich et al., 2016).

In the present study, chamber experiments were performed to investigate the impact of photosensitized oxidation reactions on SOA formation and growth. The TROPOS Atmospheric Chemistry Department Chamber (ACD-C) was used to study in detail gas-particle interactions as well as aqueous- / particulate-phase chemistry. Additionally, offline filter analysis using LC-MS techniques was performed to determine the composition of SOA particles after potential photosensitized reactions took place.

As potential photosensitizers imidazole-2-carboxaldehyde (2-IC), 3'-methoxyacetophenone (3'-MAP), and 3,4-dimethoxybenzaldehyde (DMB) were used. The present study provides insights into the reactivity of the photosensitizers regarding to SOA

formation and growth for different experimental conditions, such as variations of the pH and the chemical composition of seed particles (i.e., addition of carboxylic acids). Furthermore, to simulate more realistic atmospheric conditions and to increase the chemical complexity of the particles, Suwannee river fulvic acid (SRFA) was added to the seed particle matrix.

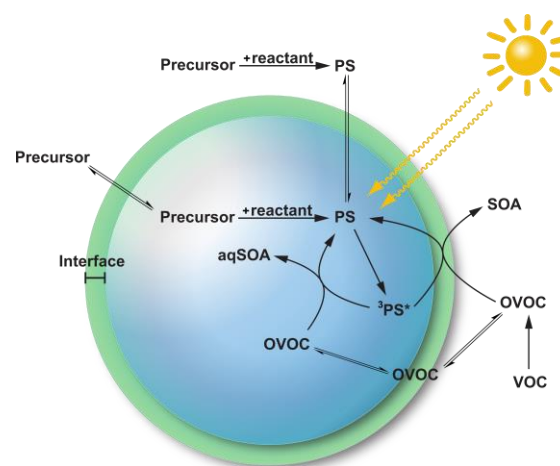


Figure 1. Scheme of photosensitized oxidation processes of oxygenated VOCs (OVOCs) within and/or at the interface of an aerosol particle.

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