Final Report Summary - AIRSEA (Air-Sea Exchanges driven by Light)

Due to recent advances in our knowledge, increasing attention has been given to the physical and chemical processes occurring at the uppermost layer of the oceans i.e. the sea-surface microlayer (SML). This region, at the uppermost tens to hundreds of micrometers of the water surface, has physical, chemical and biological properties that differ from those of the underlying subsurface water. SML chemical composition is controlled by biogeochemical and physical processes in the ocean. On one hand, it influences the organic fraction of marine aerosol produced by sea spray processes, on the other hand it controls trace gas deposition to the ocean and may be involved in secondary organic aerosol formation in the marine boundary layer. These aspects are central to the atmospheric composition changes, air quality and associated climate change.

Therefore, the project AIRSEA aimed therefore at a better chemical characterization and understanding of the photochemistry taking at the oceanic microlayer. Indeed, the presence of complex and photoactive compounds at the air/sea interface and thus also in the primary marine aerosol give rise to new processes affecting chemistry in the marine boundary layer. Upon light absorption, these compounds will go into an excited triplet state, and new chemical pathways such as the efficient degradation of organic water constituents but will also induce efficient charge exchange reactions, almost at the diffusion limit, thereby activating halogenated radicals in the marine environment, enhancing the local oxidation capacity.

Also, specific chemistry has been identified at the air/water interface. We demonstrated that, while fatty acids are believed to be photochemically inert in the actinic region, complex volatile organic compounds (VOCs) are produced during illumination of an air-water interface coated solely with a monolayer of carboxylic acid. When aqueous solutions containing fatty acids at bulk concentrations that give rise to just over monolayer coverage are illuminated with actinic radiation, saturated and unsaturated aldehydes are seen in the gas phase and more highly oxygenated products appear in the aqueous phase. This chemistry is probably initiated by triplet state NA molecules excited by direct absorption of actinic light at the water surface. As fatty acids covered interfaces are ubiquitous in the environment, such photochemical processing will have a significant impact on local ozone and particle formation.

Also, photochemical processes at the air/water interface involving authentic biogenic surfactants leads to the abiotic production of VOC. Interestingly, VOC fluxes increased with the decay of microbial cells in the samples, indicating that cell lysis due to cell death was the main source for surfactants, and VOC production. Up to now such VOC emissions were directly accounted to high biological activity in surface waters. However, we suggest that abiotic photochemistry can lead to similar emissions into the atmosphere, especially in less biologically-active regions. This can be exemplified in the case of isoprene thought to be solely produced in the sea water by various biological processes. This chemical interfacial processing may represent a significant abiotic source of isoprene in the marine boundary layer.

In summary, at the air / sea interface, biogenic surfactants are critical in providing a concentrated medium allowing radical-radical reactions to occur in parallel with molecular oxygen additions. This chemical competition at the interface, leads to the production of a large variety functionalized and unsaturated compounds, including isoprene. We suggest now that biological
activities are of course emitting VOCs, but cell lysis is a significant surface of biogenic surfactant that are inducing specific photochemistry at the air/sea interface leading to the emissions of reactive VOCs. This starts a new paradigm on why, when and where VOC emissions can take place above oceans.

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