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Sum-Frequency Generation Spectroscopy for Studying Organic Layers at Water-Air Interfaces: Microlayer Monitoring and Surface Reactivity

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The sea surface microlayer, according to commonly accepted terminology, comprises the topmost millimetre of the oceanic water column. It is often enriched with organic matter and is directly influenced by sunlight exposure and gas exchange with the atmosphere, hence making it a place for active biochemistry and photochemistry as well as for heterogeneous reactions. In addition, surface active material either is formed or accumulates directly at the air-water interface and gives rise to very thin layers, sometimes down to monomolecular thickness. This “sea surface nanolayer” determines the viscoelastic properties of the seawater surface and thus may impact the turbulent air-sea gas exchange rates. To this effect, this small scale layer presumably plays an important role for large scale changes of atmospheric trace gas concentrations (e.g., by modulating the ocean carbon sink characteristics) with possible implications for coupled climate models.

To date, detailed knowledge about the composition, structure, and reactivity of the sea surface nanolayer is still scarce. Due to its small vertical dimension and the small amount of material, this surfactant layer is very difficult to separate and analyse. A way out is the application of second-order nonlinear optical methods, which make a direct surface-specific and background-free detection of this interfacial layer possible. In recent years, we have introduced the use of vibrational sum frequency generation (VSFG) spectroscopy to gain insight into natural and artificial organic monolayers at the air-water interface.

In this contribution, the application of VSFG spectroscopy for the analysis of the sea surface nanolayer will be illustrated. Resulting spectra are interpreted in terms of layer composition and surfactant classes, in particular with respect to carbohydrate-containing molecules such as glycolipids. The partitioning of the detected surfactants into soluble and non-soluble (“wet” and “dry”) surfactants will be discussed. Furthermore, the application of a combined VSFG/Langmuir trough experiment to investigate the reaction kinetics of heterogeneous oxidation processes will be highlighted. The ozonolysis of monolayers of unsaturated fatty acids serves as model system for natural aging processes of surfactant layers at the sea surface. Finally, a VSFG time series study of the sea surface nanolayer at a western Baltic Sea near-shore sampling station will be presented. The observed seasonality reveals a significant temporal shift with respect to the spring algal bloom showing that high organic material content in the microlayer does not necessarily correlate with high nanolayer abundance. This interesting finding and implications for the formation of surfactant material by degradation of biological material will be discussed briefly.