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Air-sea exchange of acetone, acetaldehyde, DMS and isoprene at a UK coastal site.

Daniel Phillips^{1,2}, Frances Hopkins¹, Thomas Bell¹, Charel Wohl^{1,2}, Claire Reeves², Philip Nightingale^{1,2,3}, Peter Liss², and Mingxi Yang¹

¹Plymouth Marine Laboratory, Plymouth, UK

²Centre for Ocean and Atmospheric Sciences, University of East Anglia, Norwich, UK

³Sustainable Agriculture Systems, Rothamsted Research, Devon, UK

Volatile organic compounds (VOCs) are ubiquitous in the atmosphere and are important for atmospheric chemistry. Large uncertainties remain in the role of the ocean in the atmospheric VOC budget because of poorly constrained marine sources and sinks. There are very few direct measurements of air-sea VOC fluxes near the coast, where natural marine emissions could influence coastal air quality (i.e. ozone (O₃), aerosols) and terrestrial gaseous emissions could be taken up by the coastal seas.

To address this, we present air-sea fluxes of acetone, acetaldehyde and dimethylsulfide (DMS) at the coastal Penlee Point Atmospheric Observatory (PPAO) in the South-West UK during the spring (Apr-May 2018). Fluxes are quantified simultaneously by eddy covariance (EC) using a proton transfer reaction quadrupole mass spectrometer. Comparisons are made between two wind sectors representative of different air-water exchange regimes: the open water sector facing the North Atlantic Ocean and the fetch-limited Plymouth Sound fed by two estuaries.

Mean EC (± 1 standard error) fluxes of acetone, acetaldehyde and DMS from the open-water wind sector were 8.01 ± 0.77 , 1.55 ± 1.44 and $4.67 \pm 0.56 \mu\text{mol m}^{-2} \text{d}^{-1}$ respectively (- sign indicates air-to-sea deposition). These measurements are generally comparable (same order of magnitude) to previous measurements in the Eastern North Atlantic Ocean at the same latitude. In comparison, the terrestrially influenced Plymouth Sound wind sector showed respective fluxes of -12.93 ± 1.37 , -4.45 ± 1.73 and $1.75 \pm 0.80 \mu\text{mol m}^{-2} \text{d}^{-1}$. The greater deposition fluxes of acetaldehyde and acetone within the Plymouth Sound were largely driven by higher atmospheric concentrations from the terrestrial wind sector. The reduced DMS emission from the Plymouth Sound was caused by a combination of lower wind speed and likely lower dissolved concentrations as a result of the freshwater estuarine influence (i.e. dilution).

In addition, we measured the near surface seawater concentrations of acetone, acetaldehyde, DMS and isoprene from a marine station 6 km offshore. Comparisons are made between EC fluxes from the open water and diffusive VOC fluxes calculated with a two-layer (TL) model of gas transfer using air/water concentrations. The calculated TL fluxes are largely consistent with our direct measurements in the directions and magnitudes of fluxes. Generally, the TL model

predicted acetone and acetaldehyde fluxes that were ~12–33 % higher (greater deposition) than the EC measurements. This could be due to sea surface processes that produce these carbonyl compounds that were not accounted for by the TL technique.