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Partitioning of methanesulfonate and non-sea-salt sulfate in individual sea salt particles collected at the Pt. Reyes national seashore.

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Sea salt particles are chemically and physically modified by condensation of various oxidation products of dimethylsulfide (DMS), the major source of sulfur over oceans. For many years, effective conversion of DMS to particulate sulfate was assumed to be the dominant reaction pathway for DMS in the marine boundary layer. However, recent modeling studies (von Glasow and Crutzen, Atm.Chem.Phys. 2004) indicated that under certain conditions DMS does not predominantly convert to sulfate (nss-SO₄²⁻), but rather ends up in sea salt particles in a form of methanesulfonate $(CH_3SO_3^-)$, which previously has been considered only of minor importance. Specifically, high $CH_3SO_3^-/nss-SO_4^{2-}$ values have been reported in model runs for cloudy MBL at winter conditions (surface temperature of 3-8°C over the ocean). These findings have been confirmed in our field study presented here. We report that CH_3SO_3Na is a dominant form of the nss-sulfur found in the sea salt particles that traveled over the areas of cold ocean stream prior their sampling in the vicinity of Pt. Reyes, CA, north of San Francisco. In this presentation we demonstrate the effective and complementary coupling of a multitude of microprobe analytical techniques (CCSEM/EDX, TOF-SIMS and STXM/NEXAFS) to unravel the chemical composition of individual sea salt particles with the focus on the quantitative assessment of the $CH_3SO_3^-/nss-SO_4^{2-}$ partitioning of the nss-sulfur in these particles. We report the particle size specific data on the nss-S/Na and the $CH_3SO_3^-/nss-SO_4^{2-}$ ratios measured in dry residues of marine cloud droplets and particles collected during the Marine Stratus Experiment (MASE) in July 2005. Characteristic ratios of nss-S/Na > 0.10 are reported for sea salt particles, with higher values for small particles indicating extensive formation of sulfur containing salts in small particles. Characteristic ratios of $CH_3SO_3^-/nss-SO_4^{2-} > 0.70$ are reported with higher values for large particles, indicating the higher capacity for $CH_3SO_3^-$ (lower conversion to SO_4^{2-}) for large particles. To the best of our knowledge, this is the first time that $CH_3SO_3^-/nss-SO_4^{2-}$ have been quantitatively reported based on the individual particle measurements