Text S1: Details of Sampling Fog Water at Sea, Seawater Incubation, and Microlayer Sample Collection Experiments

This section describes the mounting configurations, operational methodologies and tactical sampling considerations for the at-sea fog water sampling during this program. Also described are the acid incubation and microlayer measurements of MMHg.

R/V *Point Sur*

 The R/V *Point Sur* is a 135 foot, 500 ton Regional Class Research Vessel with the wheel house towards the midpoint of the vessel. On the R/V *Point Sur*, sufficient bow space allowed for the placement of a sampling tower 3 meters aft of the bow of the vessel, yet forward of the wheel house (Figure S-3). The tower was constructed of two Rhone 25G sections. The tower was hinged at the base and supported a stainless steel plate adapter at the top. The active strand collector was mounted to the stainless steel adapter and the galvanized tower was supported by three stainless steel guy cables, secured to the bulwarks and tightened using turnbuckles. The freeboard at this location was about 5 meters and the tower extended another 6 meters above this resulting in a sampler height of about 11 meters above the water. This placed the sampler about 20 meters forward of the ship’s stacks.

R/V *Robert Gordon Sproul*

 The R/V *Sproul* is a 125 foot, 700 ton, converted drilling re-supply vessel, now Coastal Class research vessel (mud-boat). The wheel house is far forward which accommodates a large and low fantail. There is insufficient space on the bow to mount a tower. On the R/V *Sproul*, the active strand sampler was mounted to the ship’s mast above the wheel house (Figure S-4). A stand was fitted to the stainless steel base and supported by Unistrut affixed to the ship’s mast about 2 meters above, and just forward of, the wheel house. In this location, the sampler was approximately 7 meters above the water and about 9 meters forward of the ship’s stacks. At this time, the R/V *Sproul* was burning biodiesel as a primary fuel.

R/V *Oceanus*

 The R/V *Oceanus* is a 177 foot, 960 ton Ocean Class research vessel with the wheel house in the front 1/3 of the vessel and a reconfigured stack just aft of the wheel house. The bow was obstructed by an existing, bow-mounted MET tower constructed of a single pole which precluded at-sea access but also collected water droplets that could be blown into a sampler mounted immediately aft of the pole. The active strand sampler was affixed to the top of the wheel house and protruded slightly forward of the lifeline rails. Here, the sampler was located about 8 meters forward and 5 meters below the ship’s stacks.

 *Sea water incubation experiments*

 To test for potential demethylation of DMHg under acidic conditions, seawater was taken from the Moss Landing Marine Laboratory pumphouse and put into two carboys. One of these carboys was acidified to pH = 5.2 and the other was unacidified. Six samples were taken from each carboy. Three were incubated under natural light conditions in an outdoor incubator for 6 hours and three were incubated in the dark for 6 hours. At the end of this time the samples were analyzed for MMHg according to methods given in Hammerschmidt et al. (2011) and Lamborg et al. (2012).

 *Microlayer measurements of MMHg*

 Surface microlayer samples were collected using the glass plate method in its original form (Harvey and Bruzell, 1972) and as modified by Ebling and Landing (2015).  In addition a dipping device consisting of 7.3 cm diameter by 50 cm in length borosilicate glass tubes was also tested.  All three methods were used and yielded similar concentrations of MMHg.  Acid cleaned glass plates (or tubes) were dipped into seawater from the bow of an inflatable boat moving slowing into the wind. Blanks were determined from MQ water rinses of the sampling devices.   Samples were collected into cleaned borosilicate bottles (200 mL) and stored acidified for methylmercury analysis ashore. Methods for mercury sampling and analysis follow Hammerschmidt et al. (2011) and Lamborg et al. (2012).

References

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