MERCURY SPECIATION IN OPEN OCEAN WATERS

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ABSTRACT. Vital to our understanding of the biogeochemical cycling of Hg and the origin of the enhanced monomethylmercury (MMHg) concentrations in biota is knowledge of the sources, behavior and fate of methylated Hg species in natural waters. Methylated Hg species, dissolved gaseous and reactive mercury were measured in the equatorial Pacific Ocean in early 1990. Both dimethylmercury (DMHg) and MMHg were found in the sub-thermocline waters of the equatorial Pacific Ocean. Maxima in alkylmercury species in the O_2 minimum region coincided with a decrease in reactive mercury. A significant inverse correlation between DMHg, and MMHg, concentration and O_2 content was observed. A maximum in reactive mercury was observed in the region of the thermocline, with similar concentrations in the surface and deeper waters. Atmospheric deposition is not a significant source of MMHg to this region. The data suggest formation of alkylmercury species in the low oxygen zone, with Hg(II) being the most likely substrate. A model for the cycling of Hg species in the equatorial Pacific Ocean is presented. These results are the first direct evidence of a significant open ocean source of methylated Hg species and suggest a pathway for mercury accumulation in pelagic fish.

1. INTRODUCTION

There is well-documented evidence for the bioaccumulation of mercury in biota (e.g. Huckabee et al, 1979, Windom and Kendall, 1979, Craig, 1986) and increased levels of Hg in fish have resulted in health advisories regulating fish consumption in the United States, Canada and Scandinavia (Wiener and Stokes, 1990). While information on the ability of organisms to methylate Hg is available (Robinson and Tuovinen, 1984; Beijer and Jernelov, 1979), there is still a paucity of data on the sources, behavior and sinks of Hg in natural waters. Recent studies in northcentral Wisconsin seepage lakes have identified some important conditions conducive to monomethylmercury (MMHg) production in fresh-water lakes (Bloom and Watras, 1989a, Hurley, Watras and Bloom (this volume)). Studies in freshwater and estuarine regions have indicated that anoxic waters and sediments are important environments for MMHg production (Olson and Cooper, 1976; Beijer and Jernelov, 1979; Gilmour and Henry (in press)); however, little is known about the source of Hg found in ocean fish (Windom and Kendall, 1979).

Improvements in analytical techniques have made possible the determination of low levels of alkylmercury species, and elemental Hg (Hg^0) in natural waters (Bloom, 1989; Kim, 1987; Fitzgerald et al., 1989). Monomethylmercury has now been measured in lakewater in

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both the dissolved and particulate fractions (Bloom and Watras, 1989a). There is also evidence of MMHg in marine particulate matter and plankton (Topping and Davies, 1981; Knauer and Martin, 1972). This investigation was undertaken as previous studies of the equatorial Pacific Ocean (Kim and Fitzgerald, 1986; Kim, 1987) have demonstrated the presence of and significant sea-air exchange of Hg^0, and have provided evidence of a volatile organic Hg species, most probably DMHg, at depth (Kim and Fitzgerald, 1988). It was hypothesized that the low oxygen waters of the equatorial Pacific Ocean should be conducive to the production of alkylmercury species and Hg^0. This hypothesis was investigated during a 1990 NOAA cruise of the Malcolm Baldridge.

We recently reported unequivocal observations of both MMHg and dimethylmercury (DMHg) in open ocean waters (Mason and Fitzgerald, 1990). These alkylmercury species were associated with the low O_2 sub-thermocline waters of the equatorial Pacific Ocean. The evidence suggests the existence of significant regions of alkylmercury production in the open ocean. The presence of both DMHg, a volatile uncharged species, and MMHg, an ionic complex, has important implications for the understanding of the biogeochemistry and cycling of Hg in the marine environment, and for the accumulation of methylated Hg species in fish. On the basis of these results we put forward a preliminary view of the interactions and cycling of alkylmercurials and other Hg species in the equatorial Pacific Ocean surface waters (Mason and Fitzgerald, op. cit.).

This paper will outline the methodologies employed in the determination of DMHg, MMHg, total dissolved gaseous Hg (DGM), and reactive Hg (Hg_R) in ocean water. In addition, results from two stations that highlight important characteristics of the distributions of the Hg species in the equatorial region will be presented. Details of the distributions of methylated mercury species were discussed in Mason and Fitzgerald (1990).

2. EXPERIMENTAL METHODS

As illustrated in Figure 1, the research cruise in the equatorial Pacific Ocean of the Malcolm Baldridge from 3 January to 18 February, 1990 consisted of a transect between the Panama Canal and Tahiti (Leg 1), and a track from Tahiti to 5°N, 180°W, then to 5°S, 180°W (Leg 2). Two stations were occupied on the equator (stations 2 and 8); two were north of the equator (stations 1 and 7); and five south. All stations, except station 5 (12°S), were between 5°N and 5°S. The stations covered the longitudes between 87°W and 180°W.

The ship was equipped on the bow with a 7.5 m aluminum tower which was used to collect atmospheric gaseous and particulate samples, and to collect rain samples. A clean room was available on deck. All sample manipulations and analyses were performed under clean room conditions. Quantification of Hg species, trapped either on gold or graphitized carbon columns (see details below) was by atomic fluorescence spectroscopy (AFS). The AFS system was housed in the main chemical laboratory.

Water samples were collected using trace metal free techniques (Gill and Fitzgerald 1985) in the mixed layer and below the thermocline to a maximum depth of 900 m, using 8 L and 30 L teflon-lined Go-Flo^R bottles (General Oceanics) attached to a Kevlar^R line. Samples for dissolved O_2 and nutrient determinations were either taken from a conductivity-temperature-depth (CTD) rosette sampler deployed at the same time, or directly from the Go-Flo^R bottles. Expendable bathythermograph (XBT) deployments to 450 m were conducted prior to and on station. Samples from the surface mixed layer were collected for chlorophyll determinations and for productivity studies using 10 L Go-Flo^R bottles. Shipboard determinations of DMHg, MMHg, DGM and Hg_R were performed on unfiltered seawater samples. Dimethylmercury and DGM measurements were completed within 8 hr on separate 2 L aliquots taken directly from the Go-Flo^R bottles. A further 2 L were

acidified with 20 mL of 6N HCl and then subsampled for the MMHg and Hg_R determinations. In some instances, 20 L of seawater were filtered through quartz fiber filters and the filter analyzed for total particulate Hg. Samples have been retained for total Hg determinations.

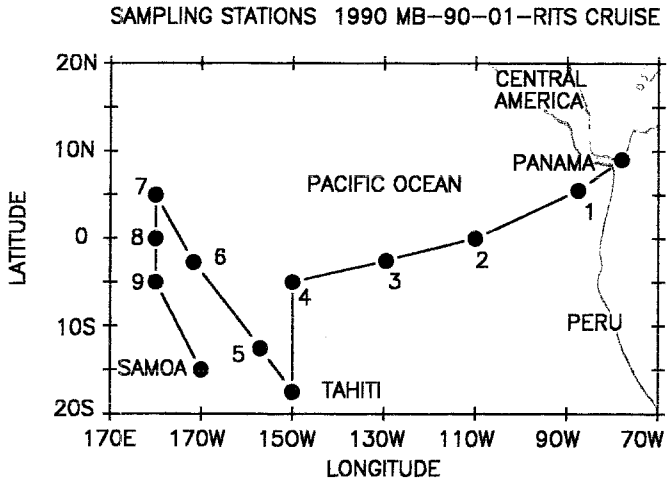

FIGURE 1: Cruise track of the Malcolm Baldridge showing stations where water samples were collected. (After Mason and Fitzgerald, 1990).

Dimethylmercury was stripped from seawater using a stream of argon and trapped on a graphitized carbon (Carbotrap^R; Supelco Inc.) column (Fitzgerald et al., 1989). The species on the column (DMHg and Hg^0) were separated and quantified by cryogenic gas chromatography with AFS detection (Bloom and Fitzgerald, 1988). The Carbotrap^R column was heated to elute the trapped compounds which were retrapped on a OV-1 chromatographic column at liquid nitrogen temperatures. The OV-1 column was then transferred to a heated oven and the species eluted. Prior to AFS quantification all Hg compounds were decomposed to Hg^0, after elution, using a heated quartz column at 900°C. Peaks corresponding to elemental Hg and DMHg were identified by their retention time. No other peaks appeared in the chromatogram. The procedural blank, determined by rebubbling the water, was <5 fM.

For the DGM determination, an additional 2 L of seawater were degassed and the volatile species trapped on a gold column (Kim, 1987). Detection was by AFS. The procedural blank was consistent throughout the cruise and averaged 145 fM $\pm$ 45 fM (n=20). Blank corrected concentrations for DGM determined by the speciation method (DMHg and Hg^0 on the Carbotrap^R column plus Hg on the gold backup column) were within 20 % of those obtained using the gold trapping method, further confirming the presence of DMHg. At station 9, two casts were made within 150 km of each other. The measured concentrations of DMHg and DGM, at three depths, differed by less than 30 %.

Extractable MMHg was determined on an acidified unfiltered 400 mL aliquot. A

methylene chloride extraction step (2X20 mL) was necessary to remove chloride ions which interfere with the ethylation (Bloom, 1989). Buffered distilled water (pH 4.9) was added and the methylene chloride was evaporated, thereby transferring the MMHg into the aqueous phase. The MMHg was derivatized using tetraethylborate. The volatile alkyl derivatives were sparged from solution and trapped on a Carbotrap^R column. The methylethylmercury derivative was separated from Hg° and diethylmercury (Hg(II) derivative) by cryogenic gas chromatography and was measured by AFS. The overall procedural blank was 40 ± 17 fM (n=9), determined by a re-extraction of the water. Rain samples of 200 mL were analyzed without extraction. The procedural blank for the analysis of rain was similar.

For the Hg_R determination, 500 mL of acidified sample was reduced using $SnCl_2$. The volatile species were sparged from solution and trapped on a gold column (Gill and Fitzgerald, 1987) before being quantified by AFS. The procedural blank over the duration of the cruise was 0.58 ± 0.16 pM. To determine total Hg, water samples were oxidized with BrCl solution (Szakacs et al., 1980; Bloom and Crecelius, 1983). Quartz fiber filters used to collect particulate Hg were also digested using BrCl solution. Excess oxidant was reduced using hydroxylamine solution prior to $SnCl_2$ reduction and AFS detection.

Air samples were collected throughout the cruise and analyzed for total gaseous Hg (TGM), alkylmercury species, and for total particulate Hg using well-known methodologies (Fitzgerald, 1989). Levels of TGM and particulate Hg were similar to values obtained on previous cruises in the region (Fitzgerald, op. cit.).

3. RESULTS AND DISCUSSION

Dimethylmercury and MMHg were found in measurable quantities in the deeper waters at all stations (Mason and Fitzgerald, 1990). The trends in DMHg, MMHg, DGM and Hg_R were found to be similar at all the stations and therefore, in this report, only two stations (4 and 9) will be presented in detail (Figure 2). In addition, the overall trends of all the stations will be discussed. Generally, the profiles of DMHg and MMHg exhibited a maximum in concentration below the thermocline (Mason and Fitzgerald, op. cit.; Figure 2). A peak in Hg_R was found in the thermocline region at most stations, while concentrations of Hg_R were similar in the mixed layer and in the sub-thermocline waters. Elemental mercury (calculated as DGM-DMHg) concentrations were typically lowest in the surface mixed layer and a maximum was generally found in the sub-thermocline region. The profiles for DMHg, MMHg and DGM suggest a decrease in concentration with depth below the O_2 minimum. The DMHg concentrations approached the detection limit in the thermocline and no DMHg (<5 fM) was found in the mixed layer. At stations 1, 3, 5, 6, 7 and 9, MMHg concentrations were below the detection limit (50 fM) in the surface waters (<150 m). Stations 2 and 4 were atypical in that measurable concentrations of MMHg were found in the thermocline region. The concentration of DMHg in the sub-thermocline waters (>200 m) was the greatest at station 2 (555 ± 154 fM, n=3); and the least at station 9 (163 ± 85 fM, n=5). For MMHg, sub-thermocline concentrations ranged from zero to 280 pM. The DMHg and MMHg concentrations in the sub-thermocline waters were generally smaller at the more westerly stations (Figure 2).

Total dissolved gaseous mercury ranged between 40 and 325 fM for the surface waters, and from 270 to 990 fM for the sub-thermocline waters. Concentrations of Hg° ranged from 15 to 610 fM below the thermocline and no general east-west trend was evident. These concentrations are similar to previously measured values (Kim and Fitzgerald, 1986; Kim and Fitzgerald, 1988). In addition, Kim and Fitzgerald (1988) found evidence of a volatile organic Hg species at depth, probably DMHg, at stations between 5°N and 5°S, 150°W.

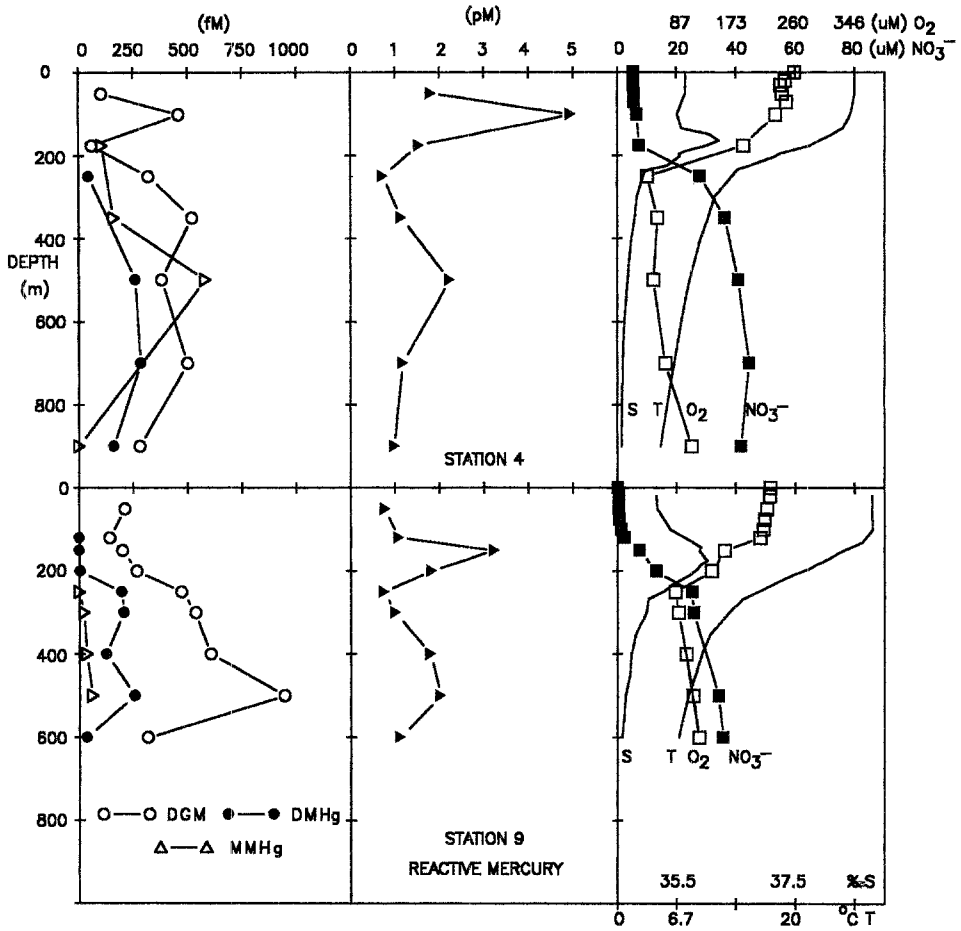

Figure 2: Concentration profiles for DMHg, MMHg and DGM (left); reactive mercury (middle); and hydrographic parameters (temperature, salinity, dissolved O_2, and nitrate) (right) at stations 4 (5°S, 150°W) and 9 (5°S, 180°W). (After Mason and Fitzgerald, 1990).

For the sub-thermocline waters (>200 m), Hg_R ranged between 0.72 and 2.85 pM. In the surface waters (<100 m), Hg_R concentrations were similar. A maximum in Hg_R at most stations was found in the thermocline region (100-200 m; Figure 2). Concentrations ranged from 1.4 to 6.9 pM in this zone. A general decrease for comparable latitudes, from east to west, was evident for Hg_R in the surface mixed layer (Figure 4).

Dimethylmercury and MMHg profiles suggest scavenging losses or decomposition in

the surface waters and production at depth. Gas evasion could account for the distribution of DMHg if the flux at the sea surface is greater than the flux across the thermocline. Particulate scavenging in the surface waters could remove MMHg. Both processes would yield undetectable concentrations in the mixed layer. Mason and Fitzgerald (1990) showed that the expected dissolved MMHg concentration in the surface waters in equilibrium with the observed particulate Hg concentrations (12 ± 8 pg L^{-1}) would be below the current detection limit of 50 fM, in agreement with the measured concentrations.

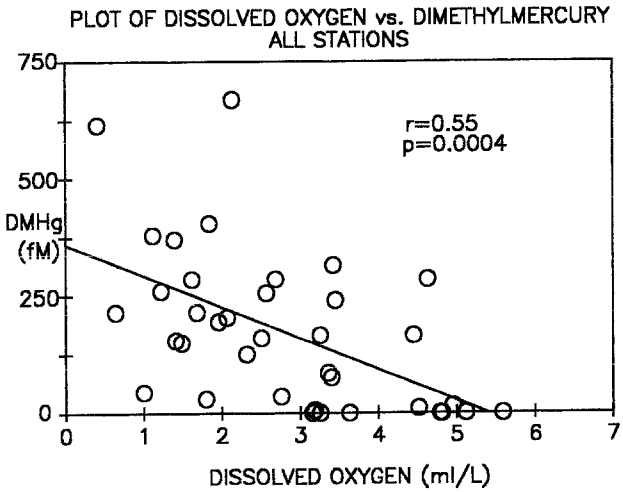

FIGURE 3: Plot of DMHg against dissolved O_2 for all stations at all depths. The regression line is shown.

The vertical and spatial distributions of the various species provide the basis for a preliminary interpretation of the biogeochemical cycling of Hg in the upper ocean. The low sub-thermocline Hg_R generally corresponds with the low O_2 regime and with high DMHg and MMHg. Based on these observations, we suggest that DMHg and MMHg are produced in the O_2 minimum zone and that production at other depths is small (Mason and Fitzgerald, 1990). A correlation between DMHg concentration and dissolved O_2 is statistically significant ($r=0.55$, $n=37$, $p=0.004$, Fig 3). A weaker correlation between MMHg and dissolved O_2 was also found ($r=0.43$, $n=36$, $p=0.012$). There was no significant relationship between Hg_R and dissolved O_2 concentration. However it was noted that the sub-thermocline Hg_R and the maximum Hg_R concentration generally increased as the surface Hg_R increased (Figure 4a). These results indicate that the system is strongly coupled, most likely through incorporation of ionic Hg into particulate matter in the surface waters and subsequent release in the deeper waters. The lower concentrations of Hg_R found at the equatorial stations (stations 2 and 8) relative to the concentrations at stations of similar longitude (Figure 4b) provide additional evidence of the importance of particulate matter in the cycling of Hg_R. Gill (1987) found a decrease in Hg_R in the surface waters at the equator at 160°W, which corresponded to a decrease in the $^{234}Th/^{238}U$ ratio, and to an increase in nitrate+nitrite. These results indicate that increased scavenging of mercury from the surface waters by particles is occurring in

regions of upwelling and associated enhanced productivity.

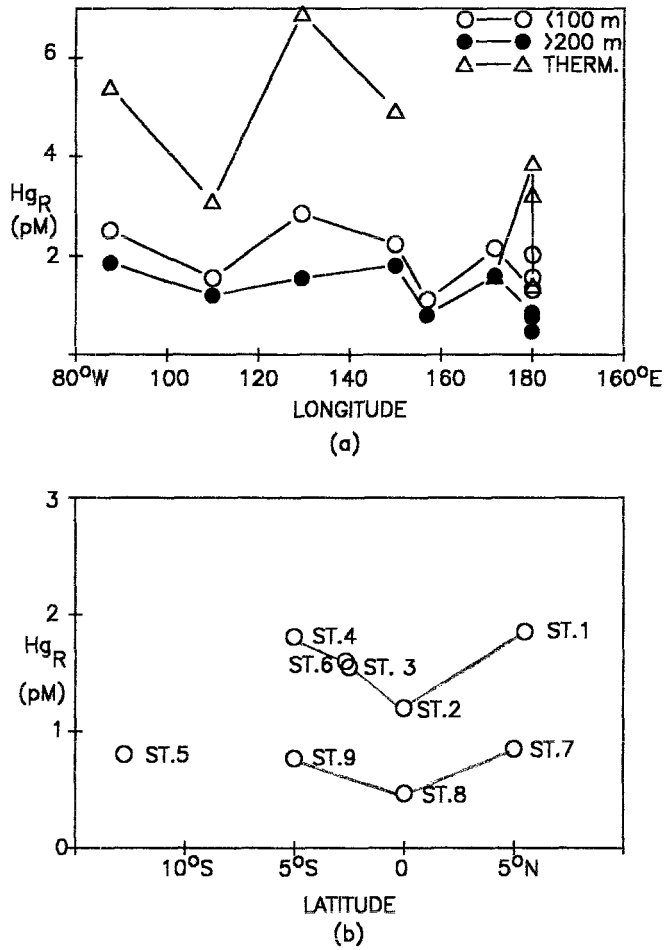

FIGURE 4: (a) Plot of Hg_R variation with longitude in the surface mixed layer (<100 m), the thermocline region (100 to 200 m), and the maximum concentration in the sub-thermocline waters (200 to 900 m) showing variability between stations. (b) Plot of Hg_R in the surface waters (<100 m) with latitude.

The maximum in Hg_R in the thermocline zone suggests that regeneration and release of dissolved Hg from particulate matter is occurring. Knauer and Martin (1981) showed that only 12% of the carbon fixed into particulate matter in the surface waters reached the bottom of the thermocline. Particulate dissolution could release both Hg_R and MMHg back

into the water column.

Biological demethylation processes which are known to occur throughout the water column in lakes (Xun et al., 1987) could account for the loss of DMHg in the more oxygenated thermocline waters, and could release additional inorganic Hg in the low O_2 region due to demethylation of MMHg. Prelimin-ary results from laboratory experiments indicate that DMHg is stable in oxygenated unfiltered coastal surface seawater at the natural pH of seawater, but not at low pH. Fagerstrom and Jernelov (1972) state that DMHg is unstable at a pH below 5.6. Thus DMHg is likely to be stable in the open ocean surface waters.

There was no evidence of MMHg in the atmospheric gaseous fraction. Rain collections were made on five occasions and analyzed for Hg_R, MMHg and total Hg using procedures described above. The reactive component was typically greater than 80% of the total Hg concentration. Total Hg and Hg_R concentrations were similar to values found previously for remote ocean regions (Fitzgerald, 1989). There was no evidence of MMHg in rain, contrary to what has been found for continental rains (Bloom and Watras, 1989b; Fitzgerald et al., 1989). Thus it appears that the atmosphere is not a significant source of MMHg to the open ocean. This observation is corroborated by studies in northcentral Wisconsin lakes where in lake processes were found responsible for a large proportion of the MMHg found in fish (Fitzgerald et al., 1989, Hurley, Watras and Bloom (this volume)).

Alkylation of Hg, both biotically and abiotically, can occur under a wide variety of environmental conditions (Beijer and Jernelov, 1979; Robinson and Tuovinen, 1984; Gilmour and Henry (in press)). However, biological methylation in anoxic environments is thought to be the predominant process (Olson and Cooper, 1976). Dimethylmercury is a major product of the reaction between Hg(II) and methylcobalamin under mildly reducing conditions (Wood et al., 1968). However, MMHg is formed if excess Hg(II) is present (Craig, 1986). Jensen and Jernelov (1969) demonstated the production of DMHg in the presence of dead fish tissue and Fagerstrom and Jernelov (1972) showed that DMHg is the major product in alkaline anoxic sediments. Monomethylmercury was formed in sediments of lower pH. These authors suggest that although pH's between 5 and 9 do not affect the methylation rate, the pH does affect the amount of DMHg formed. At higher pH values they found almost all the methylated Hg to be DMHg. Compeau and Bartha (1984) showed that DMHg is produced in anoxic estuarine sediments spiked with MMHg. In addition, these authors suggest that sulphate reducing bacteria are the predominant methylators of Hg in anoxic estuarine sediments (Compeau and Bartha, 1985). Water column methylation in coastal marine surface waters (Topping and Davies, 1981) and in freshwater (Furutani and Rudd, 1980; Xun et al., 1987) has been found. These results suggest that the low O_2 waters of the equatorial Pacific Ocean are conducive to formation of DMHg, and MMHg.

The evidence of alkylmercury species in low O_2 ocean waters, remote from continental margins, and the distributions of the various mercury species have been integrated into a preliminary conceptual model (Figure 5) of the cycling of Hg species in equatorial Pacific Ocean waters (Mason and Fitzgerald, 1990). We speculate that DMHg and MMHg are produced in the O_2 minimum region with concentrations decreasing as the minimum O_2 concentration increases. In addition, we suggest that "labile inorganic Hg" provides the substrate for methylation. This fraction is composed of labile inorganic and organic complexes with Hg(II), and of labile particulate associations. Fagerstrom and Jernelov (1972) have previously stated that "the methylation rate is dependent on the concentration of inorganic divalent Hg in the substrate". We suggest that labile inorganic Hg is supplied to the surface waters primarily by atmospheric deposition, and to the sub-thermocline waters by vertical mixing and by particulate dissolution. The correlation between O_2 concentration and alkylmercury production suggests that a low O_2 environment is conducive to the formation of alkylmercury species. Therefore production is enhanced in the low O_2 environment of the equatorial Pacific.

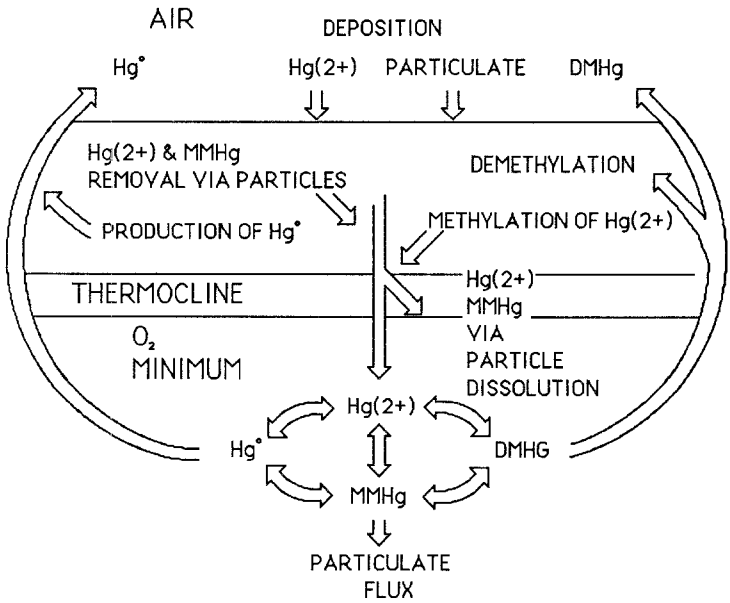

FIGURE 5: Diagram representing the overall processes likely to affect the concentrations and distributions of the various Hg species in the surface waters of the equatorial Pacific Ocean (After Mason and Fitzgerald, 1990).

The distribution of the various mercury species will be modified by horizontal advection, upwelling, and diffusion. Horizontal advection of mercury species in the sub-thermocline region could be a significant process. Upwelling is likely to enhance exchange between the surface mixed layer and the deeper waters and could supply additional Hg(II) to the sub-thermocline zone. Dimethylmercury will be rapidly decomposed in the atmosphere (Niki et al., 1983) after evasion from the surface waters. Particulate scavenging will remove any MMHg mixed across the thermocline into the surface waters. Particulate dissolution in the deeper waters could release bound MMHg and Hg(II) into solution.

While more research is needed to investigate the importance of the formation and decomposition processes for alkylmercury species and Hg^0, this model highlights areas for further investigation. This work suggests that in situ production of alkylmercury species occurs in the open ocean away from continental margins. In addition, this investigation provides the first substantial evidence of a pathway for Hg accumulation in pelagic marine fish.

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