Increase in mercury in Pacific yellowfin tuna

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Abstract

Mercury is a toxic trace metal that can accumulate to levels that threaten human and environmental health. Models and empirical data suggest that humans are responsible for a great deal of the mercury actively cycling in the environment at present. Thus, we would predict that the concentration of mercury in fish should have increased dramatically since the Industrial Revolution. Evidence in support of this hypothesis has been hard to find, however, and some studies have suggested that analyses of fish show no change in mercury concentration. By compiling and re-analyzing published reports on yellowfin tuna (*Thunnus albacares*) caught near Hawai'i over the past half century, we find that the concentration of mercury in these fish is currently increasing at a rate ≥ 3.8 % per year. This rate of increase is consistent with a model of anthropogenic forcing on the mercury cycle in the North Pacific, and suggests fish mercury concentrations are keeping pace with current loadings increases to the ocean. Future increases in mercury in yellowfin tuna and other fishes can be avoided by reductions in atmospheric mercury emissions from point sources.

Introduction

Mercury is a potent toxin that can accumulate to high concentrations in fish, posing a health risk to humans who eat fish. Methylmercury, the predominant form of mercury in fish, is formed from mercuric ions (Hg[II]) by microbes and perhaps abiotically in waters and sediments, then enters the base of the food web and increases in concentration with each successive trophic level. Consumption of mercury-contaminated fish from gross pollution events, e.g., in Minamata, Japan [1] where fish with concentrations as high as 36 ppm could "easily be captured by hand", has resulted in severe neurologic damage in humans, most acutely in children exposed prenatally via maternal fish consumption. In waters not directly affected by local pollution, mercury concentrations in fish are typically < 1 ppm. However, even at this lower level, prenatal exposure is associated with developmental deficits [2]. In the U.S. annually, Trasande et al. [3] found that *c.* 300,000-600,000 children are born with mercury concentrations in cord blood that exceed 5.8 μg/L, a value associated with significant loss of IQ (intelligence quotient); the economic consequence of which is estimated to be US$8.7 billion annually in lost income.

Humans are exposed to methylmercury primarily via consumption of ocean fish [4], and while models (see review in [5]) and empirical data [6] suggest an increase in mercury content of the global ocean since preindustrial times, a corresponding increase in

methylmercury has not been shown for ocean fish. Local pollution of nearshore environments occurs where there are point-source water discharges, but for open ocean the principal source of mercury is atmospheric deposition [7]. Atmospheric mercury is now dominated by human contributions (primarily from fossil fuel combustion and artisanal gold mining), and present-day rates of mercury deposition are 3-5x greater than natural (pre-anthropogenic) rates [8]. It had been thought that mercury pollution could only result in a negligible increase in mercury concentration in open ocean waters [9], but a recent synthesis of data from water column profiles of total mercury points to a 2.6x increase (since the 1500s) in waters shallower than 1000 m, globally [6].

Here we show for a commercially important species, the yellowfin tuna (*Thunnus albacares*), a temporal increase in mercury concentration. Mercury data for Pacific yellowfin tuna from waters near Hawai'i present a unique record in that the same population/location was sampled three different time periods over 37 years, muscle tissue samples were analyzed for Hg, and data were reported in peer-reviewed literature. A network of fish aggregation devices in Hawai'i have documented the high site fidelity of yellowfin tuna [10]. The following is a report of our compilation, re-analysis, and interpretation of the mercury data.

Methods

We compiled published reports on mercury in yellowfin tuna caught from waters of the North Pacific Ocean near Hawai'i during 1971 [11, 12], 1998 [13], and 2008 [14]. From each specimen, muscle tissue was subsampled and measured for total mercury. Data are comparable across studies because of strict adherence to quality assurance/quality control (QA/QC) that ensured accuracy of data. Muscle tissue samples from 1971 [11, 12] and 1998 [13] were acid digested and analyzed by cold-vapor atomic absorption spectrophotometry. Rivers et al. [11] reported the mean recovery of spiked samples was 101% and that the result for each sample was validated by separate analysis of methylmercury, indeed confirming that nearly all of total mercury is methylmercury [15]. Thieleke [12; see also 16] had each sample analyzed by two independent laboratories, with excellent agreement between laboratories, as the maximum deviation among samples was 6%. Kraepiel et al. [13] reported detailed QA/QC procedures, including sample handling and use of duplicates (mean relative percent difference of 4.7%), spikes (mean recovery of 96.7%), and a reference sample (mean concentration of 0.128 ppm with standard deviation of 0.0098 ppm on 6 samples) for analyses. Muscle tissue samples from 2008 [14] were analyzed by thermal decomposition, amalgamation, and atomic absorption spectrophotometry. Choy et al. [14; see also 17] also reported detailed QA/QC procedures, including analysis of duplicates (data only accepted if relative percent difference between duplicates was <5%) and certified reference materials (mean recoveries of DORM-3 and High Purity Standard Trace Metal Fish were 100.3% and 96.1%, respectively).

To compare Hg concentration among the three data sets (1971, 1998, and 2008), we used the analysis-of-covariance (ANCOVA) model, with body size as the covariate. ANCOVA is the standard parametric test for comparing a characteristic of groups of subjects while controlling for the effect of another variable on that characteristic. Controlling for the effect of body size when assessing mercury concentration among groups of fish is thus a classic use of ANCOVA. Fish from 22–76 kg were included in the analysis as this size range ($\pm$ 5 kg) was common to all three data sets. It was necessary to remove the fish < 22 kg,

because these fish did not adhere to the assumption of linearity. Mercury concentrations in young tuna tend to be low, but highly variable [18]. A diet shift occurs in young tuna when a critical body mass is developed that enables endothermic capability to allow access to prey in deeper, colder water [19]. At a certain size (depending on species), likely due to this ontogenetic diet shift, the mercury concentration versus size relationship conforms to expectations (i.e, a linear relationship). Outliers, identified with Tukey box plots and confirmed by one-sided Grubbs' tests, were also removed from the data sets.

Results

The ANCOVA revealed that slopes of the relationships between mercury concentration and fish size were not statistically different among the three time periods ($F_{2,223} = 1.17$, $P = 0.31$; Fig. 1), but mercury concentrations were higher in 2008 than either 1971 or 1998 ($F_{2,225} = 11.6$, $P < 0.0001$; Tukey's HSD; Fig. 1). Accordingly, the average mercury concentration (least square mean ± std. error) was considerably higher in 2008 (0.336 ±0.023) than in 1971 (0.229 ± 0.008), or 1998 (0.218 ± 0.008). Sample size (for 1971, 1998, and 2008, n = 111, 104, and 14, respectively) is incorporated in the ANCOVA, and with the significant result, it can be ruled out – with 95% probability – that the effect of sample year was due to chance. The fourteen data points from 2008 are elevated relative to the two other data sets. Note that we conducted statistical diagnostics and found five points among the three datasets with potentially high leverage or high influence. We performed the ANCOVA without these points and found no change in the qualitative results of the ANCOVA (no difference in slopes, significant difference in intercepts).

Discussion

Our analysis, at least for the early part of the record, is in agreement with that of Kraepiel et al. [13], who found no change in tuna mercury between the same 1971 and 1998 datasets. This conclusion led Kraepiel et al. [13] to hypothesize that methylmercury forms from mercury naturally occurring in deep waters, sediments, or possibly hydrothermal vents and is therefore largely natural. Subsequently developed independent lines of evidence have suggested that vents are not strong enough sources of mercury to supply foodwebs [20], that fish do acquire methylated mercury from shallow depths in the ocean [21], and that seawater mercury concentrations are increasing ocean-wide [6], including near Hawai'I [22]. Thus, we should expect to see changes in the concentration of mercury in tuna now and in the foreseeable future.

Such changes are discernable in the recent part of the record, from 1998 to 2008, which shows an increase in mercury concentration in tuna at a rate of ≥ 3.8% per year, in agreement with recent and modeled changes in mercury cycling in the North Pacific (Fig. 2). Sunderland et al. [22] found a significant rise in mercury concentrations in seawater at all depths (0-1000 m) from 2002 to 2006. The largest increases, modeled at 3% per year between 1995-2006, are occurring in intermediate waters (150-1000 m), which in addition to receiving mercury from atmospheric deposition to surface waters above (0-150 m) also receive inputs associated with lateral flow of mercury-enriched waters from the coast of the Northwest Pacific [22, 23]. Yellowfin tuna near Hawai'i spend most of their time in surface waters or immediately below [24]. The agreement between the data/modeling by Sunderland et al. [22] and the updated tuna record compiled and re-analyzed here provides

support for the alternative hypothesis that mercury and methylmercury concentrations in the ocean are increasing due to human activity and that anthropogenic methylmercury accumulates in important commercial fish.

A criticism of the Kraepiel et al. [13] study, that effects of fishing on oceanic food webs may have affected methylmercury accumulation in tuna [25], also applies here. Data for age, growth, and trophic level are not available for the individual fish in our synthesis. In the Pacific Ocean, the largest tunas have become more rare, but no detectable change in trophic level has occurred in any population [26]. Growth statistics of yellowfin tuna appear unchanged over the past half century [27], but the data aren't ideal for determining temporal trends. One could expect an increase in growth rate, as a density-dependent response to over-exploitation of the population [28]. With increased growth, there would be "growth dilution" of mercury [29]. To sum, there are no data to suggest Pacific yellowfin tuna have different growth rates or trophic level for the study period, but if either of those two factors have changed, it would likely be in a direction that would tend to lower the concentration of mercury in tuna, masking the effect of increased mercury loading to the system.

Because fish mercury concentrations appear to be increasing in step with modeled loadings, these data indicate that mercury concentrations in open ocean fish are responsive to anthropogenic mercury releases. Lamborg et al. [6] suggested that if atmospheric mercury emissions continue to increase, the deepwater sink will become 'overwhelmed' in the coming decades. Therefore, it should be expected that the rate of increase in mercury in surface waters (0-1000 m) will be greater than the rate of increase in emissions. Mercury contamination of ocean fish is a serious global health issue, now being addressed by the UN Minamata Convention on Mercury. Current goals of the convention – that aim to reduce atmospheric mercury emissions from point sources – would result in avoided increases in rates of atmospheric mercury deposition [30]. However, even if current deposition rates are maintained, North Pacific intermediate waters are expected to double in mercury concentration by 2050 [22]. Thus, more stringent reductions in emissions are necessary.

Supplemental Data

The data reported were previously published [11-14], but can also be found in Table S1.

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Figure Captions

Fig. 1. Linear regressions of fish size (kg) versus mercury concentration (parts-per-million) for three datasets for yellowfin tuna from North Pacific waters near Hawai'i; 1971 (ref. *11*, *12*; black): Hg = -0.0799 + 0.0068 x mass, n = 111, r^2 = 0.413, P < 0.0001; 1998 (ref. *13*; red): Hg = -0.1619 + 0.0083 x mass, n = 104, r^2 = 0.375, P < 0.0001; 2008 (ref. *14*; green): Hg = -0.0718 + 0.0093 x mass, n = 14, r^2 = 0.656, P = 0.0004.

Fig. 2. Least square mean (± std err) mercury concentration in yellowfin tuna from 1971 (black), 1998 (red), and 2008 (green) from waters near Hawai'i. Overlaid on the fish data are mercury concentrations in seawater; gray squares (± std dev) represent point estimates from integrated 1000-m profiles in the eastern North Pacific, and the solid gray line represents modeled trends for intermediate waters (150-1000 m) basin wide (dashed lines represent 95% confidence interval). Seawater data and model output are from Sunderland et al. [22].

Fig. 1.

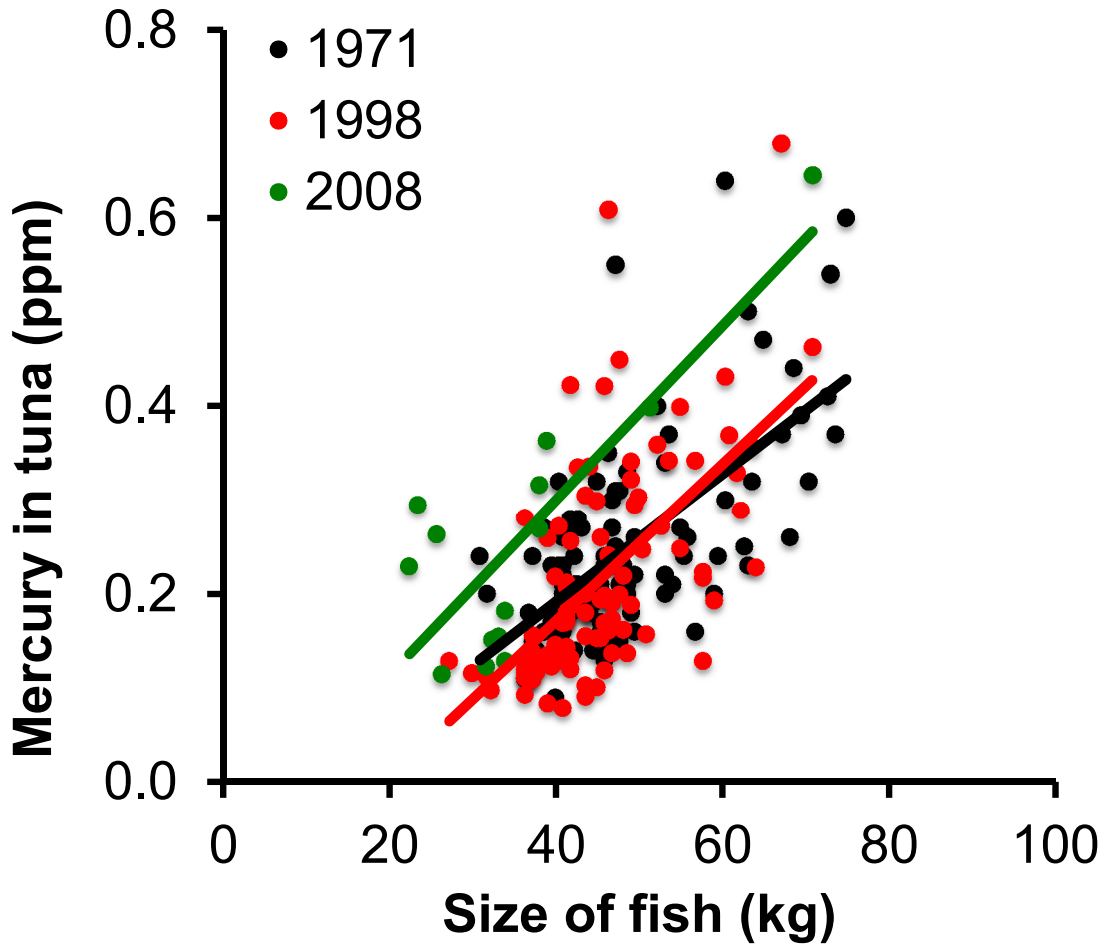
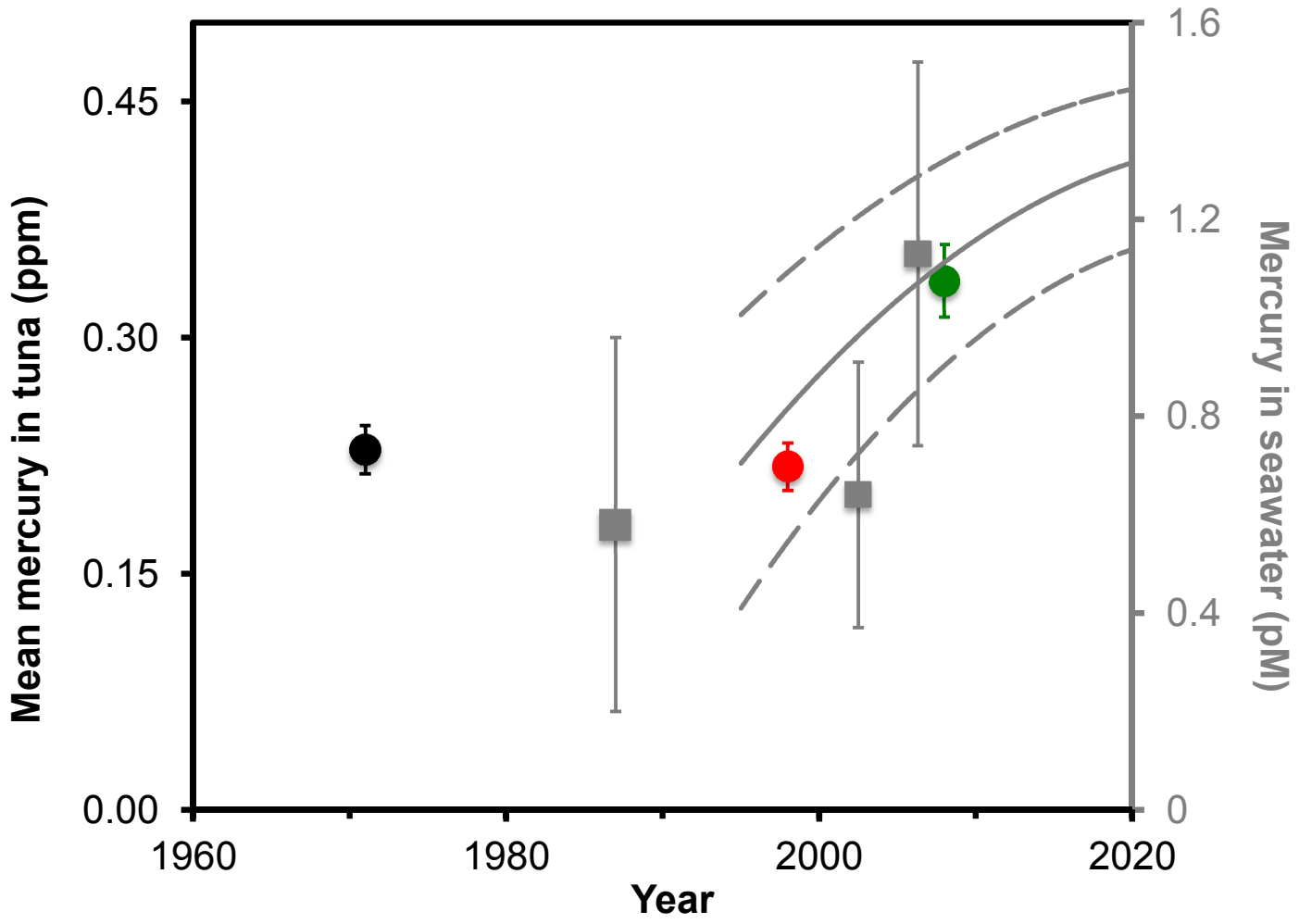

0.8
0.6
0.4
0.2
0.0
Mercury in tuna (ppm)
1971
1998
2008
0
20
40
60
80
100
Size of fish (kg)

Fig. 2.


Mean mercury in tuna (ppm)
0.45
0.30
0.15
0.00
1960
1980
2000
2020
Year
1.6
1.2
0.8
0.4
0
Mercury in seawater (pM)