

1 **Mercury flux to sediments of Lake Tahoe, California-Nevada**

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18

19 **Abstract** We report estimates of mercury (Hg) flux to the sediments of Lake Tahoe, California-
20 Nevada: 2 and 15-20 $\mu\text{g}/\text{m}^2/\text{yr}$ in preindustrial and modern sediments, respectively. These
21 values result in a modern to preindustrial flux ratio of 7.5-10, which is similar to flux ratios
22 recently reported for other alpine lakes in California, and greater than the value of 3 typically
23 seen worldwide. We offer plausible hypotheses to explain the high flux ratios, including (1)
24 proportionally less photoreduction and evasion of Hg with the onset of cultural eutrophication
25 and (2) a combination of enhanced regional oxidation of gaseous elemental Hg and transport of
26 the resulting reactive gaseous Hg to the surface with nightly downslope flows of air. If either of
27 these mechanisms is correct, it could lead to local/regional solutions to lessen the impact of
28 globally increasing anthropogenic emissions of Hg on Lake Tahoe and other alpine ecosystems.

29

30 **Keywords** mercury, sediment, Lake Tahoe, alpine

31

32 **1 Introduction**

33 There is a general lack of knowledge of the flux of atmospheric mercury (Hg) to alpine
34 ecosystems. Few empirical data exist, but a global 3-D chemical transport model (GEOS-Chem)
35 predicts high concentrations of Hg(II), the principal form of deposited Hg, over elevated land
36 (Selin et al. 2007). Indeed, recent data from Mt. Bachelor, Oregon, (2.7 km asl) showed high
37 concentrations of reactive gaseous mercury (RGM; gas-phase of Hg(II)) in free tropospheric air
38 (Swartzendruber et al. 2006). “RGM enhancements” occur at Mt. Bachelor at night when
39 downslope flows of free tropospheric air displace daytime boundary layer air.

40 Lake Tahoe, California-Nevada, (1.9 km asl) is an alpine ecosystem with special
41 protection as an Outstanding Natural Resource Water under the U.S. Clean Water Act, but from
42 sediment cores collected a decade ago Heyvaert et al. (2000) reported an exceedingly high
43 modern to preindustrial flux ratio for Hg of 24. Worldwide, this ratio is typically ~3 for sites,
44 such as Lake Tahoe, that are unaffected by point-source pollution (Lamborg et al. 2002;
45 Lindberg et al. 2007; Selin et al. 2008). Heyvaert et al. (2000) hypothesized that this unusually
46 large enhancement was a function of orographic scavenging and cold condensation processes. It
47 is unlikely, however, that these processes affect Hg flux more now than in the preindustrial past.

48 We also used a paleolimnological approach, which can in some ways compensate for a
49 lack of monitoring data (Smol 1992), to understand Hg at Lake Tahoe. However, our data do not
50 support the flux estimates or hypotheses made by Heyvaert et al. (2000); most notably, we argue
51 that the modern flux of Hg to sediment is actually ~1/3 of their estimate. A paleolimnologic
52 reconstruction can only be as good as its estimates for sediment chronology and sedimentation
53 rates, and there were irregularities in the treatment of ²¹⁰Pb data by Heyvaert et al. (2000).
54 However, even with the lower flux estimates provided in our report, the modern to preindustrial

55 flux ratio for Hg (7.5-10) is still greater than what is typical for remote regions. We offer
56 mechanistically more-plausible hypotheses to explain this phenomenon, including that, as for Mt.
57 Bachelor, Lake Tahoe is subject to RGM enhancements.

58

59 **2 Methods**

60 Two sediment cores, each 28 cm in length, were collected from the central part of Lake Tahoe on
61 18 June 2007. Core TAH-1 was taken at a location (lat. 39.1197, long. -120.0776) near research
62 buoy TB-3 from a water depth of 404 m. Core TAH-2 was taken at a location (lat. 39.1034,
63 long. -120.0133) near research buoy TB-2 from a water depth of 444 m. (The research buoys are
64 maintained by the Jet Propulsion Laboratory of the California Institute of Technology; locations
65 can be viewed at <http://remote.ucdavis.edu/tahoe_location.asp>.) The two coring sites are
66 separated by a distance of ~6 km. Coring was done aboard the University of California Davis
67 R/V *John LeConte* with an HTH gravity corer (Pylonex, Umeå, Sweden; Renberg and Hansson
68 2008). Once on board, sediment was extruded, sectioned (every half centimeter from top to 5
69 cm, every centimeter from 5 cm to 20 cm, every two centimeters from 20 cm to bottom), placed
70 in acid-cleaned polypropylene jars, and frozen.

71 In the laboratory, core sections were analyzed for water content and organic content (as
72 loss-on-ignition) (Heiri et al. 2001) and then freeze dried for determinations of ^{210}Pb , ^{137}Cs , and
73 Hg. ^{210}Pb was measured through its grand-daughter product ^{210}Po (Eakins and Morrison 1978)
74 and used to estimate age and sedimentation rates according to the c.r.s. (constant rate of supply)
75 model (Appleby 2001). Sedimentation rates were not adjusted for focusing because it appears
76 not to occur; the average ^{210}Pb flux measured in each core (determined by multiplying the excess
77 ^{210}Pb inventory by the decay constant) was about equal to the atmospheric ^{210}Pb flux (Koide et

78 al. 1972; Heyvaert 1998). ^{137}Cs was measured by gamma spectroscopy (Draut et al. in 2009) to
79 provide an additional dating marker in the sediment profiles. Total Hg was measured by thermal
80 combustion with a DMA-80 Direct Mercury Analyzer (Milestone, Shelton, Connecticut).
81 Replicate samples and certified reference materials (CRM; NIST 1515 and 1645, NRC BCSS-1)
82 were analyzed before and after every ten samples. Mean relative standard deviation for replicate
83 samples was 6.8%. Measured concentrations of CRM were within certified ranges (see
84 Electronic Supplementary Material, Table ESM1). Sediment fluxes of Hg were calculated by
85 multiplying ^{210}Pb -based sedimentation rates by Hg concentrations. Other elements were
86 measured by inductively coupled plasma-mass spectrometry after acid digestion, but only the
87 results for titanium (Ti) are reported here.

88

89 **3 Results and Discussion**

90 The stratigraphic profiles for ^{210}Pb and their interpretation are straightforward. Both cores
91 exhibit a generally smooth decrease in ^{210}Pb from the surface to the bottom of the cores, with no
92 abrupt changes in slope that might otherwise indicate hiatuses or rapid shifts in sedimentation
93 rates. The lower portions of both profiles (5-10 cm depth) are almost perfectly exponential,
94 indicating a near-constant rate of sediment accumulation for the earlier part of the record.
95 Background (supported) levels of ^{210}Pb are reached at approximately 10 cm in TAH-1 and 12 cm
96 in TAH-2 (Fig. 1a). Model estimates for age and sedimentation rates are shown in Fig. 1b and
97 Fig. 1c, respectively. Between cores, TAH-2, from deeper water, has a higher ^{210}Pb flux and
98 slightly higher sedimentation rates than TAH-1. Within cores, sedimentation rates are fairly
99 constant throughout. There is a slight increase in rates for the most recent sections (top 3 cm; c.

100 1970 - present) of both cores, and this may be a diagenetic effect or, as Heyvaert et al. (2000)
101 suggested, a result of anthropogenic erosion from rapid urbanization in the watershed.

102 Although the ^{210}Pb data of Heyvaert et al. (2000, also see Heyvaert 1998) are nearly
103 identical to ours, their interpretation resulted in questionable estimates for age and sedimentation
104 rates. The c.r.s. model and other simple models were used and reported in their study, but
105 Heyvaert et al. (2000) instead deemed as most appropriate the segmented version of the constant
106 flux with constant sedimentation rate (cf:cs) model, a more complex model that invoked massive
107 and, in our opinion, unrealistic changes in sediment accumulation. According to their approach,
108 sedimentation rates went from ~0.02 cm/yr between 1450-1860 to over 1 cm/yr between 1860-
109 1875, a period of intense logging to support mining of the nearby Comstock Lode (a putative
110 explanation). However, an increase in sediment flux of this magnitude would have produced
111 obvious changes in sediment lithology (a large increase in mineral matter, e.g., Davis et al.
112 2006), which we did not see in our cores (e.g., Ti; see Electronic Supplementary Material, Fig.
113 ESM1), nor have others (Hyne et al. 1972; Palmer et al. 1979), and a dilution of the organic
114 content of the sediment, which also did not occur (see Electronic Supplementary Material, Fig.
115 ESM2). In sediment archives from wetlands of the Lake Tahoe basin, Kim and coworkers (Kim
116 and Rejmánková 2001; Kim et al. 2001) reported changes in pollen composition (as a result of
117 altered plant communities) after logging, but no clear influence on sediment characteristics and
118 accumulation rates. Heyvaert et al. (2000) did note coherent changes in Ti and biogenic silica at
119 ~25 cm sediment depth, but it seems more likely that this depth corresponds to the termination of
120 a dry interval 600 years before present (Osleger et al. 2009). An additional problem for the
121 model used by Heyvaert et al. (2000) is that it explains the absence of measurable excess
122 (unsupported) ^{210}Pb below 8-10 cm as a result of dilution from high sedimentation. However, if

123 this were the case, other atmospheric constituents such as Hg should also decrease in
124 concentration at these depths – which they clearly do not (Fig. 2a). Heyvaert et al. (2000)
125 justified their modeling approach based on the fact that it placed the first appearance of ^{137}Cs in
126 sediment at 1955 (fallout from nuclear weapons testing began 1952). However, ^{137}Cs diffusion
127 is a well-known phenomenon in lake sediments and would tend to displace the first appearance
128 of ^{137}Cs downward in the core (Crusius and Anderson 1995), which would make ^{210}Pb dates
129 from the c.r.s. model appear too old. The problem of ^{137}Cs diffusion is exacerbated in systems
130 like Lake Tahoe with low sedimentation rates. Indeed, ^{137}Cs profiles from our cores show
131 significant upward and downward movement (see Electronic Supplementary Material, Fig.
132 ESM3), from diffusion and possibly diagenesis and/or bioturbation, although the latter is
133 unlikely to be significant below 0.25 cm sediment depth (S. Chandra, personal communication,
134 2008). In short, the ^{210}Pb profiles and other stratigraphic data from Lake Tahoe indicate
135 conformable, near-constant sedimentation; there is little evidence to suggest otherwise.

136 Concentration profiles for Hg show 7.5-10 fold increases up the cores (Fig. 2a), and
137 because of fairly constant sedimentation rates, this results in a likewise 7.5-10 fold increase in
138 Hg flux (Fig. 2b). At 9 cm in TAH-1 and 12 cm in TAH-2 (corresponding in both cores to 1850
139 and the beginnings of the industrial period and the California gold rush), Hg increases from a
140 background concentration of $\sim 0.02 \mu\text{g/g}$ dry wt and flux of $\sim 2 \mu\text{g/m}^2/\text{yr}$. This pre-1850
141 sedimentary flux is the same as reported by Heyvaert et al. (2000) and corresponds well with the
142 estimate of natural terrestrial fluxes of Hg to the atmosphere of 5 Mmol/yr (Fitzgerald and
143 Lamborg 2004), which amounts to a global average depositional flux of $\sim 2 \mu\text{g/m}^2/\text{yr}$. Increases
144 in Hg concentrations up the cores are quite remarkable in that the magnitude of the rise and even
145 the smaller details are faithfully reproduced in both cores. Such congruence from two widely

146 spaced core sites provide strong evidence for consistent changes in flux/deposition across the
147 basin. From the top of the cores, we estimate the modern Hg flux to be 15-20 $\mu\text{g}/\text{m}^2/\text{yr}$. This
148 flux is significantly lower than the estimate of 47 $\mu\text{g}/\text{m}^2/\text{yr}$ from Heyvaert et al. (2000), which
149 we argue is due to differences in models of sediment age and sedimentation rates.

150 Using our modern Hg flux estimate of 15-20 $\mu\text{g}/\text{m}^2/\text{yr}$, the modern to preindustrial flux
151 ratio (7.5-10) is still greater than the typical value of 3 for remote regions. Very little is known
152 about Hg biogeochemical cycling at Lake Tahoe and it is thus impossible to construct a robust
153 mass balance to constrain inputs and outputs. We estimate, however, from the Mercury
154 Deposition Network site in the Sierra Nevada (CA75; similar elevation and annual precipitation
155 as at Lake Tahoe), annual wet deposition of 6-8 $\mu\text{g}/\text{m}^2/\text{yr}$ (National Atmospheric Deposition
156 Program 2008) and an annual watershed contribution of 1.5-2 $\mu\text{g}/\text{m}^2/\text{yr}$ (25% of wet deposition;
157 watershed/lake area ratio ~ 1.6) (Swain et al. 1992). Together, these two inputs amount to no
158 more than 10 $\mu\text{g}/\text{m}^2/\text{yr}$. Thus, there is an additional 5-10 $\mu\text{g}/\text{m}^2/\text{yr}$ to account for even without
159 factoring in losses from river export and evasion. Dry deposition, though poorly characterized,
160 is undoubtedly another input of Hg to the lake. However, most studies of the Hg mass balance in
161 lakes require little (a couple tens of percent of total deposition) to no dry deposition to the lake
162 surface to attain closure (Watras et al. 1994). Thus, to suggest that dry deposition of Hg to Lake
163 Tahoe amounts to 5-10 $\mu\text{g}/\text{m}^2/\text{yr}$, equal to or exceeding wet deposition, requires some special
164 circumstance, such as RGM enhancements. Indeed, we posit that the high modern flux of Hg to
165 Lake Tahoe, as well as the large degree of secular change (preindustrial versus modern), are best
166 explained by a unique set of hypotheses: (1) continued recycling of Hg used in historic gold and
167 silver mining, (2) a proportional decrease in photoreduction and evasion of Hg with the onset of
168 cultural eutrophication, and (3) RGM enhancements.

169 A high modern flux of Hg from historic gold and silver mining seems unlikely, but
170 cannot be ruled out. According to estimates from the U.S. Geological Survey (Alpers et al.
171 2005), ~6 million kg of Hg were lost to the environment between 1850-1910 from gold mining
172 operations in the Sierra Nevada foothills west of Lake Tahoe but outside of its watershed, and an
173 additional ~7 million kg of Hg were lost between 1860-1900 from silver mining of the Comstock
174 Lode, a few km east of Lake Tahoe but again outside of its watershed. As Heyvaert et al. (2000)
175 pointed out, it is surprising that there is not an obvious signal from this mining contamination in
176 Lake Tahoe sediments. That is, there should be a large pulse of Hg during the late 1800s
177 (especially since Hg is not remobilized by sediment diagenesis; Rydberg et al. 2008), rather than
178 the gradual rise – more typical of sites distant from any local sources of Hg – observed in the
179 cores. Perhaps this lack of a mining signal results from Hg emissions that were likely in the
180 elemental form (Hg^0) and therefore became part of the well-mixed global pool and were
181 deposited not locally but over large areas (Strode et al. 2009). A “California gold rush” Hg
182 signal appeared to be evident in ice cores from Upper Fremont Glacier, Wyoming (Schuster et al.
183 2002), but sediment cores from a lake 12 km southwest of the glacier recorded no evidence of
184 this event (Van Metre and Fuller 2009). Indeed, no unequivocal atmospheric Hg flux from
185 historical gold and silver mining activities in North America has been found (Lamborg et al.
186 2002). Nonetheless, Nriagu (1994) argued that the volatilization of even a minor fraction of the
187 Hg from historic gold and silver mining may be partly responsible for high Hg fluxes in parts of
188 the Americas.

189 It seems likely that the rapid increase in sedimentary Hg flux during the past ~40 years
190 may, in part, be related to the well-documented reduction in water clarity of Lake Tahoe during
191 this same time period (Goldman 1998). Cultural eutrophication of Lake Tahoe has increased

192 primary production and, consequently, secchi depth and the depth of 1% light penetration have
193 both declined by 28% (Chandra et al. 2005). Sunlight plays a key role in lake water in reducing
194 Hg(II) to Hg⁰ (Amyot et al. 1994), and because lake water is often supersaturated with Hg⁰
195 (Fitzgerald and Lamborg 2004), evasive losses to the atmosphere occur. These losses can be
196 significant enough to alter fluxes of Hg in aquatic systems, with estimates that range from 7% to
197 nearly 100% of annual atmospheric deposition (Amyot et al. 1994). Recent data from the
198 METAALICUS study, which involved whole-lake additions of isotopically-enriched Hg,
199 indicated evasion of 45% of the isotopic spike (Southworth et al. 2007). Thus, it is certain that
200 some portion of the Hg(II) entering Lake Tahoe is reduced to Hg⁰ and lost via evasion, but this
201 portion is likely decreasing because of less light penetration. A very simple Hg budget, with
202 inputs balancing sedimentation and evasion (see Electronic Supplementary Material), indicates
203 50% of Hg inputs are now lost via evasion. In comparison, and according to the model, the
204 evasional flux in the preindustrial past may have been as much as 87.5% of inputs. We caution,
205 however, that these estimates are based on little data, and an empirical test of this hypothesis is
206 needed. A test could include direct measurements of photoreduction and evasion throughout the
207 year, as water clarity changes with season. Alternatively, an empirical test could take advantage
208 of the new information that, at least for lakes, Hg(II) is enriched in odd isotopes, but
209 photoreduced Hg⁰ is depleted in odd isotopes (via mass-independent fractionation; Bergquist and
210 Blum 2007). Since the likely fate of Hg(II) not reduced to Hg⁰ is sedimentation, modern
211 sediments could be less enriched than preindustrial sediments in the odd isotopes of Hg.

212 Our final hypothesis to explain the high modern flux of Hg as well as the large degree of
213 secular change is that RGM enhancements as seen at Mt. Bachelor (Swartzendruber et al. 2006)
214 also occur at Lake Tahoe and have increased in recent times as a consequence of rising levels of

215 atmospheric oxidants (Wang and Jacob 1998). From a site 30 km southwest of Lake Tahoe and
216 at about the same elevation, Murphy et al. (2006) reported the same diurnal air pattern as seen at
217 Mt. Bachelor: boundary layer air during the day and free tropospheric air at night. The study,
218 however, was not about Hg, and RGM was not measured. Importantly, relatively high ozone
219 concentrations were found in the free tropospheric air, and additional measurements by aircraft
220 indicate an enhanced regional background of ozone (California Air Resources Board 2006).
221 Ozone is believed to be a molecule involved in the oxidation of gaseous Hg^0 (GEM; comprises
222 95% of atmospheric Hg; Fitzgerald and Lamborg 2004) to RGM in the atmosphere (Calvert and
223 Lindberg 2005) and was associated with the RGM enhancements at Mt. Bachelor
224 (Swartzendruber et al. 2006). A combination of enhanced regional oxidation of GEM and
225 transport of the resulting RGM to the surface with nightly downslope flows could be driving the
226 relatively high modern Hg flux at Lake Tahoe. Indeed, an analysis of Hg wet deposition data for
227 washout curves (see Electronic Supplementary Material) from five sites in California and one in
228 Nevada (~60 km northeast of Lake Tahoe) indicates that there is generally more scavengeable
229 Hg (i.e., RGM and particulate Hg) at higher elevations (linear regression of \log_{10} -transformed
230 data, $r^2 = 0.823$, $p = 0.013$, $n = 6$; Fig. 3). Furthermore, estimates of dry deposition of Hg from
231 the site in Nevada (Lyman et al. 2007) can account for the additional 5-10 $\mu\text{g}/\text{m}^2/\text{yr}$ of Hg found
232 in the surface sediments of Lake Tahoe. Lyman et al. (2007) cautioned that these estimates are
233 derived from models with considerable uncertainty, however, and argued that, in order to assure
234 accuracy, methods to directly measure dry deposition are needed. We agree and suggest that a
235 worthwhile application would be to test whether RGM enhancements result in an increased
236 depositional flux.

237 It is possible that alpine lake sediments, through Hg records, uniquely chronicle the past
238 oxidizing conditions of the atmosphere. Contemporary observations of high concentrations of
239 RGM in the free troposphere are not unique to the study at Mt. Bachelor (Swartzendruber et al.
240 2006). High RGM has also been reported in the free troposphere at Mauna Loa, Hawaii, (Landis
241 et al. 2005) and from aircraft measurements over the Atlantic Ocean off the coast of Florida
242 (Sillman et al. 2007). However, the observation that the preindustrial accumulation of Hg in
243 Lake Tahoe sediments is comparable to that of other lake systems suggests that if RGM
244 enhancements/dry deposition are the explanation for the current flux increase, then such a
245 phenomenon was not occurring in the past. The increase in dry deposition of Hg over time
246 would probably require an increased concentration of Hg⁰ oxidizing species on a regional scale,
247 since this enhancement has been observed in Lake Tahoe and other alpine lakes in California
248 (see below), but not elsewhere. If this were true, then Hg reconstructions in high altitude lakes
249 could provide information regarding temporal changes in the atmosphere's oxidizing power at
250 regional scales, and would be a novel way to examine such behavior.

251 We strongly encourage that our latter two hypotheses be tested, as they could lead to
252 policies and management practices to lessen the impact of globally increasing anthropogenic
253 emissions of Hg (Pacyna et al. 2006) on Lake Tahoe and other alpine ecosystems. Recent data
254 from three other alpine lakes in California also have modern to preindustrial flux ratios for Hg
255 greater than 3 (Landers et al. 2008; Sanders et al. 2008). Controlling eutrophication at the local
256 level and tropospheric ozone at the regional level could result in relatively less Hg reaching sites
257 of methylmercury (MeHg) production (typically transition regions between oxygenated and
258 anoxic conditions, e.g., lake sediments). MeHg is of paramount concern in regards to both
259 human and ecological health issues of Hg. It is debatable whether MeHg contamination of

260 alpine ecosystems is high enough to affect human health (although see Blais et al. 2006), but it is
261 becoming clear that fish health is being compromised (Moran et al. 2007; Schwindt et al. 2008).

262

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267

268 **Electronic Supplementary Material**

269 Supplementary material for this manuscript includes (1) a table of quality assurance results for
270 total Hg determinations, (2) figures of Ti, organic content, and ¹³⁷Cs in sediment cores from Lake
271 Tahoe, (3) information about evasion modeling, and (4) an analysis of washout curves from
272 seven sites in California and Nevada.

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422

423 **Figure Legends**

424 **Fig. 1** ^{210}Pb data and modeling for Lake Tahoe sediment cores TAH-1 (black circles, black line)
425 and TAH-2 (grey circles, grey line); (a) cores exhibit exponential declines in ^{210}Pb with depth;
426 (b) and (c) are estimates of sediment age and mass accumulation rates, respectively, from the
427 c.r.s. (constant rate of supply) model; error bars represent 1 SD.

428
429 **Fig. 2** Total mercury (Hg_T) in Lake Tahoe sediment cores TAH-1 (black circles, black line) and
430 TAH-2 (grey circles, grey line); (a) increases in concentrations up the cores are faithfully
431 reproduced in both cores; (b) fluxes increase from $\sim 2 \mu\text{g}/\text{m}^2/\text{yr}$ in preindustrial sediment to 15-20
432 $\mu\text{g}/\text{m}^2/\text{yr}$ in modern sediment, resulting in a modern to preindustrial flux ratio of 7.5-10.

433
434 **Fig. 3** Scatterplot of elevation (meters above sea level) and mercury (Hg) washout coefficient
435 for seven sites in California and Nevada. Washout is defined as the removal of gases or particles
436 from the atmosphere by scavenging onto precipitation. The washout coefficient (a) is derived
437 from an inverse power regression of Hg wet deposition data, where precipitation amount (P) is
438 the independent variable and its measured Hg concentration (C) is the dependent variable; $C = a$
439 $\cdot P^{-1}$ (see Electronic Supplementary Material). These results indicate that the amount of
440 scavengeable Hg (i.e., reactive gaseous Hg and particulate Hg) increases with elevation. The
441 symbol “X” represents a site from the San Francisco Bay Area that is in close proximity to point
442 sources of atmospheric Hg pollution and thus is an outlier and was removed from the linear
443 regression analysis.