

Evaluation of Mercury Concentrations in Birds Collected from Great Salt Lake

PREPARED FOR: Utah Department of Environmental Quality, Division of Water Quality, & Great Salt Lake Science Panel

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Higher-than-expected selenium concentrations were found in blood of American avocets, black-necked stilts, and California gulls collected from the Great Salt Lake (GSL) during 2006, in comparison to selenium results for concurrently collected food-chain, liver, and egg samples. While the Science Panel agreed that the critical endpoints for selenium are diet and eggs, they also agreed that the question of high selenium concentrations in blood samples should be further evaluated.

It was expected that selenium concentrations in blood would be similar to those in the birds' diet during the 4 to 8 weeks before their collection. This assumption was based on feeding studies of mallards (Heinz and Fitzgerald 1993) where blood concentrations plateaued after 8 weeks at 84 percent of the 10 micrograms per gram ($\mu\text{g/g}$) and 70 percent of the 20 $\mu\text{g/g}$ dietary concentration and of American kestrels (Yamamoto et al. 1998) where blood plateaued after 77 days at 100percent of the 5 $\mu\text{g/g}$ and 98 percent of the 9 $\mu\text{g/g}$ dietary concentration. In a study in which selenium accumulation in the liver was studied (Heinz et al. 1990), liver selenium reached a peak concentration of 95 percent of the dietary concentration in 8 days and plateaued.

Blood selenium concentrations were also unexpectedly high when compared to egg concentrations in GSL birds. In kestrels fed 6 or 12 $\mu\text{g/g}$ selenium, egg concentrations were about twice the diet and blood concentrations (Santolo et al. 1999). In contrast, gull and avocet blood collected from GSL birds was 5.5 and 10 times the concentrations observed in eggs (in gulls the geometric mean in blood was 16 and in eggs it was 2.9 $\mu\text{g/g}$; in avocets the geometric mean in blood was 24 and in eggs it was 2.4 $\mu\text{g/g}$).

In a technical memorandum, Santolo and Ohlendorf (2007) suggested that the high selenium concentrations may have been due in part to elevated mercury. An inter-lab comparison was conducted to validate the laboratory results by having USGS and the project lab (LET) each analyze split samples of composite eared grebe blood for selenium and mercury (Santolo 2007). The results of the interlab comparison showed that the laboratory (i.e., LET) met the data quality objectives defined by the program. There was only an 8 percent relative percent difference (RPD) for selenium and a 13 percent RPD for mercury.

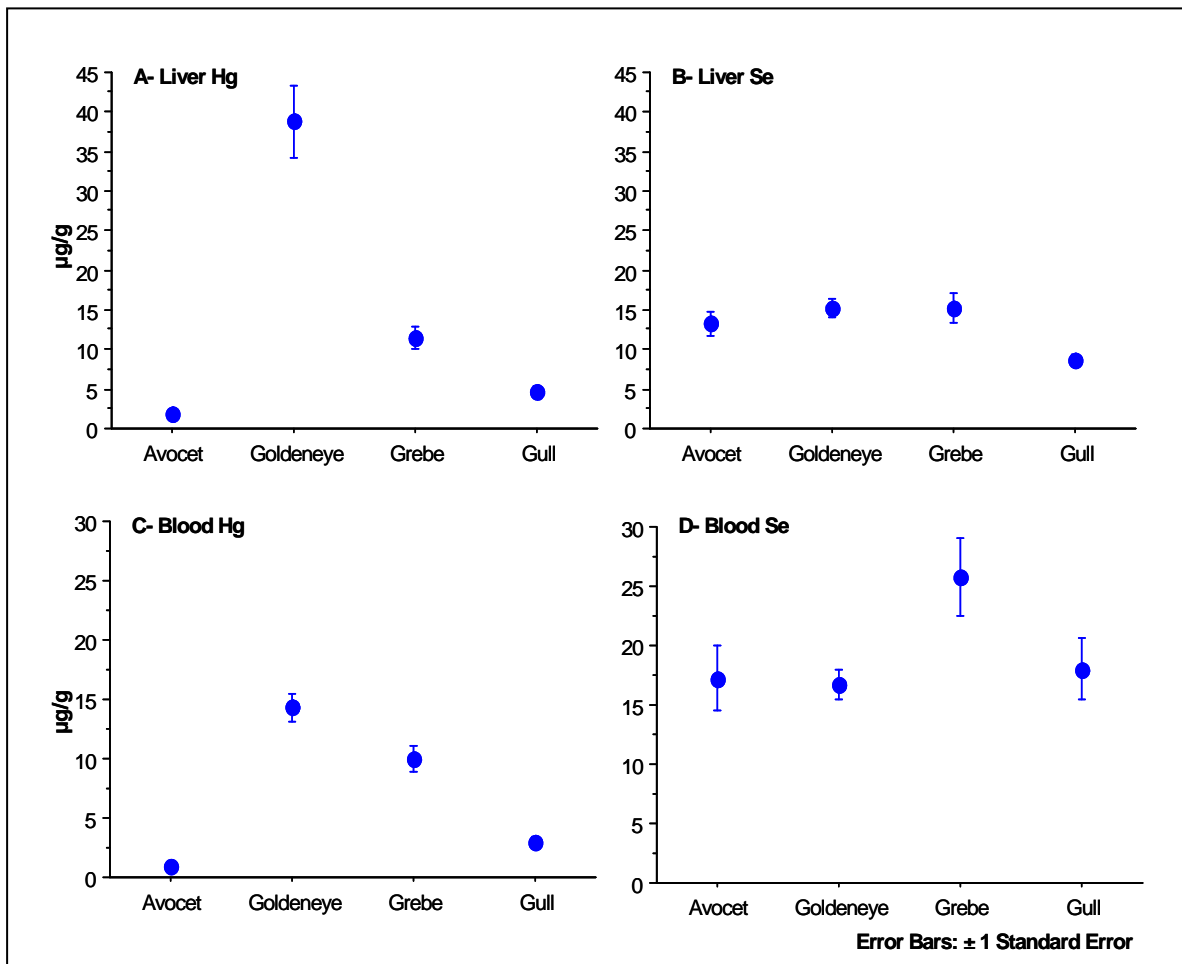
Results of sampling of avian blood, livers, and eggs for mercury and selenium have been reported by Conover et al. (2007a,b,c) and Cavitt (2007) for California gulls, common goldeneyes, eared grebes, and American avocets. For example, results of the selenium and mercury analyses of blood and liver from grebes collected near Hat Island and near Antelope Island showed a pattern of increasing mercury and selenium from fall to winter at Hat Island and higher concentrations at Hat Island than at Antelope Island (Conover 2007b). Similarly, mercury and selenium concentrations in liver and blood from common goldeneyes increased from arrival in fall through winter (Conover et al. 2007c).

This technical memorandum provides a summary and interpretation of those results from the avian mercury and selenium sampling.

Results

There are apparent species differences in mercury and selenium concentrations in liver and blood (Figure 1A-D). Mercury concentrations in livers were highest in common goldeneyes (39 $\mu\text{g/g dw}$), followed by eared grebes (13 $\mu\text{g/g}$), and lowest in California gulls (4.6 $\mu\text{g/g}$) and American avocets (1.9 $\mu\text{g/g}$; Figure 1A). However, liver selenium concentrations were similar among avocets, grebes, and ducks but lower in gulls (Figure 1B). Blood mercury concentrations were higher in wintering ducks and grebes than in breeding avocets and gulls (Figure 1C) and blood selenium was higher in grebes than other species (Figure 1D). Mercury was significantly higher in grebes sampled in November than in grebes sampled in September ($t_{18} = -3.3$, $P = 0.004$).

Figure 1. Mercury and selenium concentrations ($\mu\text{g/g dw}$) in livers and blood of American avocets, eared grebes, common goldeneyes, and California gulls sampled from Great Salt Lake.



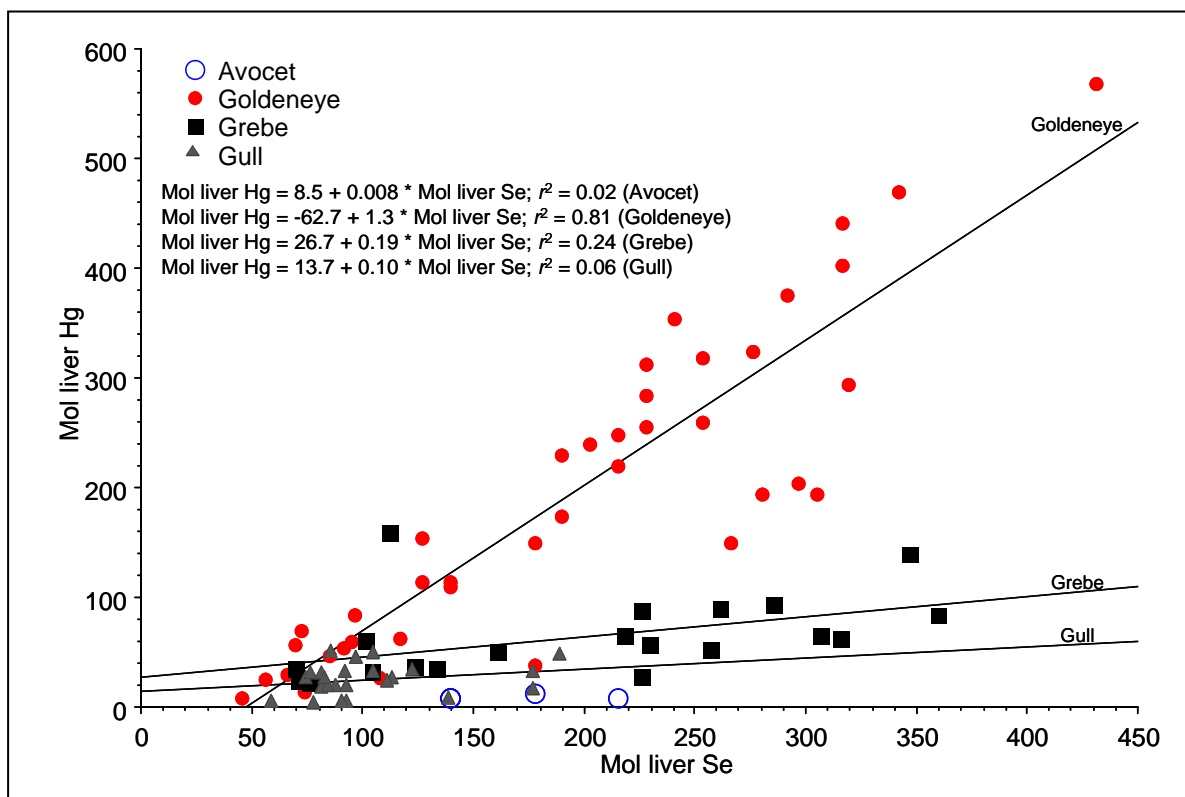
Mercury and selenium molar ratios provide the most reliable and comprehensive criteria for evaluating risks associated with exposure, so these interactions are expressed on a molar basis. There was a near 1:1 ratio in liver but not blood when mercury and selenium results from all birds (ducks, grebes, gulls, and avocets) were combined, but there were notable differences among species (Table 1, Figure 2).

The intraspecies molar relationship between mercury and selenium in liver was most significant in common goldeneyes ($r^2 = 0.81$, $F_{1,40} = 162$, $P < 0.001$), and there was slightly higher than a 1:1 ratio. Eared grebes had a low slope value with a significant relationship and a low correlation coefficient (Table 1). However, by removing a single outlier sample (i.e., likely a newly arrived juvenile with a low liver selenium and high mercury concentration), the slope value was increased to 0.24 and the significance of the relationship was increased ($r^2 = 0.61$, $F_{1,20} = 28$, $P < 0.001$). The molar relationship of mercury and selenium in gull livers from the GSL was not significant ($r^2 = 0.06$, $F_{1,24} = 1.4$, $P = 0.25$).

Table 1. Relationship between selenium (x) and mercury (y) concentrations ($\mu\text{g/g dw}$) in liver and blood of birds from Great Salt Lake collected in 2007.

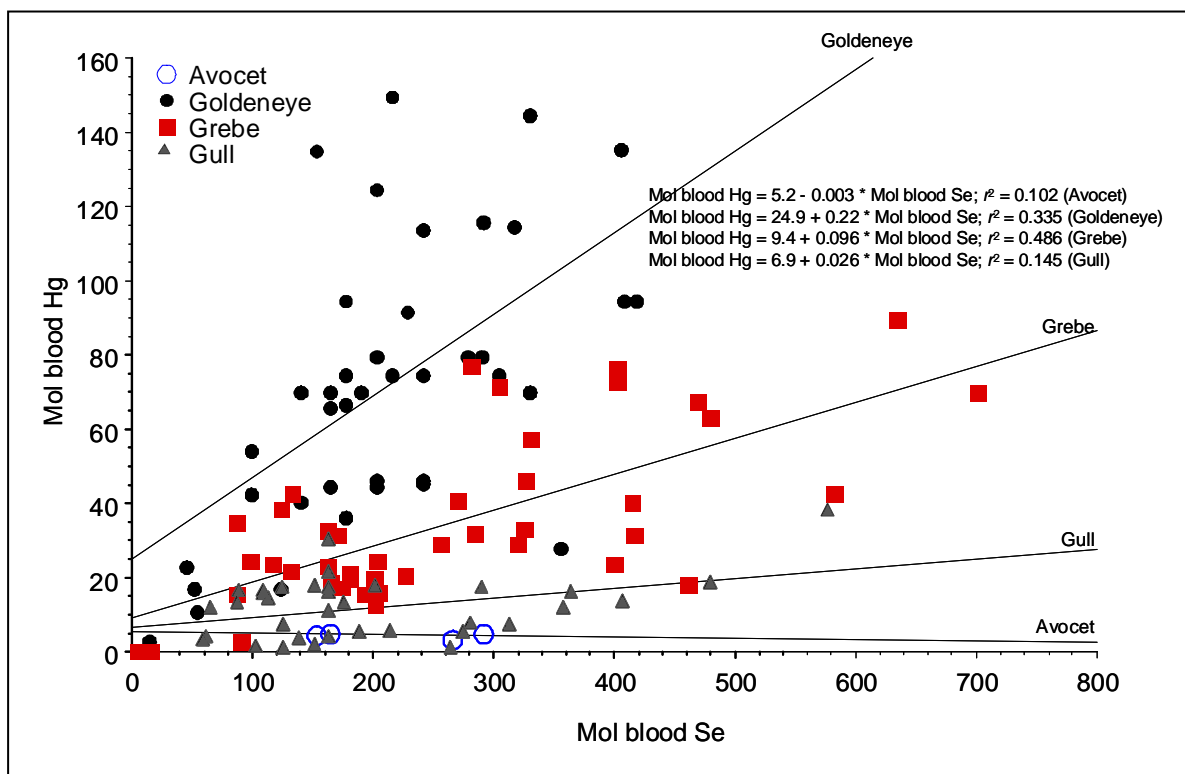
Species	<i>n</i>	Regression equation	Correlation coefficient (r^2)	<i>F</i>	<i>P</i> value	Hg Increment ¹
Liver						
All birds	89	$y = -12.1 + 2.53x$	0.52	94	< 0.001	0.99
Common goldeneye	40	$y = -12.58 + 3.36x$	0.81	162	< 0.001	1.3
Eared grebe	21	$y = 5.35 + 0.47x$	0.24	6.1	0.02	0.19
California gull	24	$y = 2.75 + 0.26x$	0.06	1.4	0.24	0.10
American avocet	4	$y = 1.69 + 0.02x$	0.016	0.03	0.87	0.008
Blood						
All birds	89	$y = -9.79 + 0.53x$	0.05	4.8	0.03	0.098
Common goldeneye	40	$y = -6.51 + 1.9x$	0.28	15.1	<0.001	0.22
Eared grebe	21	$y = 3.47 + 0.31x$	0.49	14.3	0.002	0.096
California gull	24	$y = 1.46 + 0.12x$	0.18	7.5	0.009	0.026
American avocet	4	$y = 0.99 + 0.06x$	0.43	1.52	0.34	-0.003
¹ Increment on an atomic basis (atomic weight ratio Hg/Se = 2.54).						

Figure 2. Molar relationship between mercury and selenium concentrations in livers of birds sampled from Great Salt Lake.



The intraspecies molar relationship between mercury and selenium in blood was most significant in eared grebes ($r^2 = 0.49$, $F_{1,17} = 38.8$, $P < 0.001$) and common goldeneyes ($r^2 = 0.34$, $F_{1,40} = 19.1$, $P < 0.001$), but with a low slope and correlation coefficient for both (Table 1, Figure 3). The molar relationship of mercury and selenium in gull blood also was significant, but with a lower r^2 and slope ($r^2 = 0.15$, $F_{1,36} = 5.8$, $P = 0.02$). The relationship of blood mercury and selenium on molar basis was not significant in American avocets ($r^2 = 0.10$, $F_{1,4} = 0.23$, $P = 0.68$); however, the sample size for avocets was small.

Figure 3. Molar relationship between mercury and selenium concentrations in blood of birds sampled from Great Salt Lake.



On molar basis, mercury in liver was significantly related ($r^2 = 0.83$, $F_{1,68} = 331$, $P < 0.001$) to that in blood in all birds sampled, and the ratio of liver to blood mercury was nearly 3:1 (slope = 3.1). Selenium concentrations in liver and blood also were significantly related ($r^2 = 0.37$, $F_{1,110} = 65$, $P < 0.001$) and the ratio was about 1:1 (slope = 0.95). Moles of liver selenium were significantly related to moles of mercury in the blood ($r^2 = 0.59$, $F_{1,68} = 98$, $P < 0.001$) with a high slope value (slope = 0.36). Although the relationship between mercury and selenium in blood was significant, it explained only about 16 percent of the variability in the results ($r^2 = 0.12$, $F_{1,68} = 8.8$, $P = 0.004$) with a low slope value (slope = 0.13).

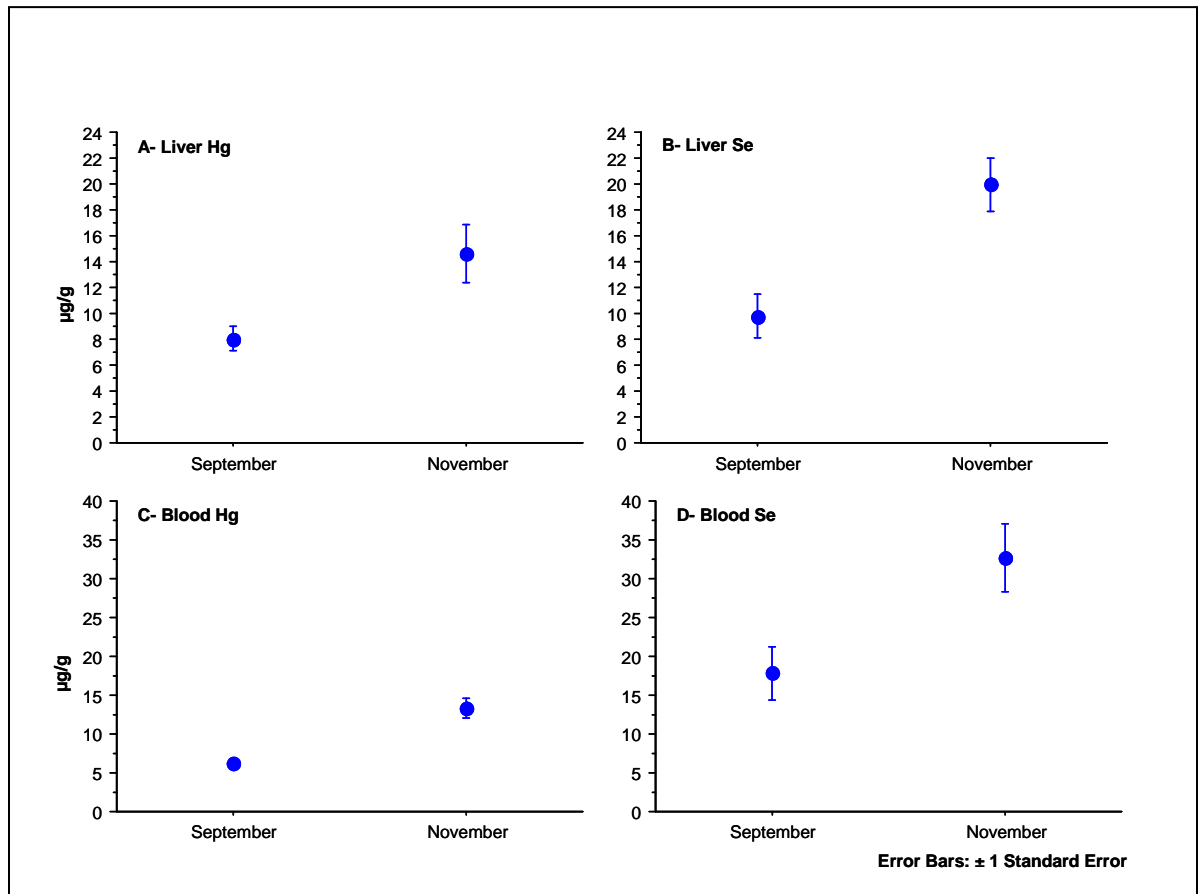
Grebes were the only species for which distinct arrival (September) and later (November) periods were sampled. In grebes, mercury and selenium concentrations in livers significantly increased from September to November (Table 2, Figure 4). Shortly after arrival, the relationship between mercury and selenium in livers was not significant ($r^2 = 0.37$, $F_{1,9} = 4.1$, $P = 0.08$) and there was a low slope value (0.14); in contrast, there was a significant relationship between mercury and selenium ($r^2 = 0.42$, $F_{1,11} = 6.6$, $P = 0.03$) and a higher slope value (0.28) in November grebes.

Table 2. Mercury and selenium ($\mu\text{g/g dw}$) in livers and blood from fall resident eared grebes in September and in November at Great Salt Lake.

Sample Period (Month)	Liver		Blood	
	GM Hg ¹ (n) Range	GM Se ¹ (n) Range	GM Hg ¹ (n) Range	GM Se (n) Range
Early (September)	7.2B (9) 4.5 - 12	8.8B (29) 5.0 - 20	4.4A (22) 0.09 - 8.6	14A (22) 0.3 - 46
Late (November)	14A (11) 5.9 - 28	13A (29) 6.4 - 28	5.7A (21) 0.05 - 18	19A (21) 1.1 - 55

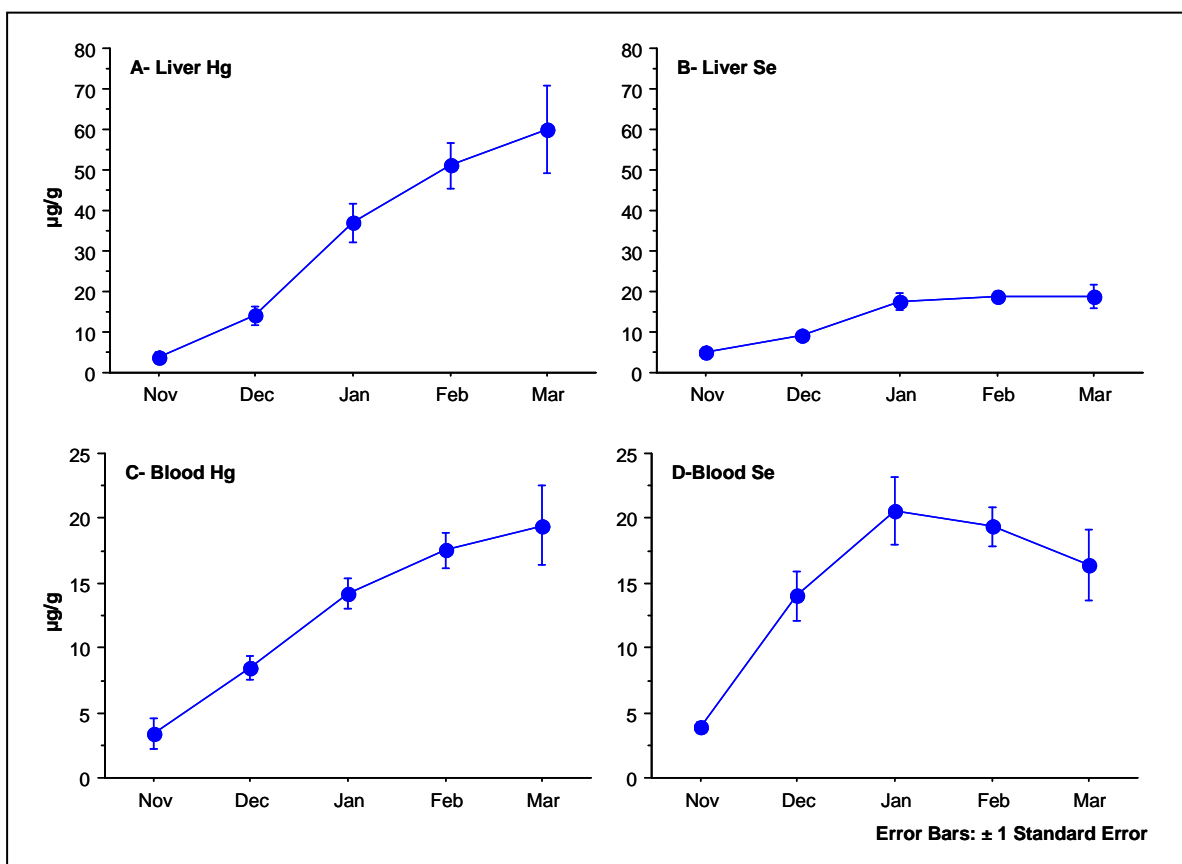
¹ Geometric means with different uppercase letters within columns are significantly different from each other (Unpaired *t*-test $\alpha = 0.05$).

Figure 4. Liver mercury (A) and selenium (B) and blood mercury (C) and selenium (D) concentrations ($\mu\text{g/g dw}$) in eared grebes collected from Great Salt Lake in September and November.



Similar but more pronounced increases in mercury and selenium in livers and blood were observed in goldeneyes collected from November 2005 through March 2006 (Figure 5A-D). For example, mercury increased in common goldeneye livers from a mean of 3.9 $\mu\text{g/g}$ dw, when they first arrived in November, to 14 $\mu\text{g/g}$ in December, 37 $\mu\text{g/g}$ in January, 51 $\mu\text{g/g}$ in February, and 60 $\mu\text{g/g}$ in March (Figure 4A). Selenium concentration in livers increased from November (GM = 5.1 $\mu\text{g/g}$) to 16 $\mu\text{g/g}$ dw in March but reached a plateau in January (Figure 5B). Mercury concentrations in blood continued to increase from November (in parallel with livers), whereas selenium concentrations in blood decreased after January (when they had plateaued in livers). The initial increase of selenium concentration in blood was much faster than in liver, and it was faster than the increase for mercury in blood (Figures 5C and 5D).

Figure 5. Liver mercury (A) and selenium (B) and blood mercury (C) and selenium (D) concentrations ($\mu\text{g/g}$ dry weight) in common goldeneyes collected from Great Salt Lake in November through March.



Mercury concentrations were lower in livers and blood of gulls and avocets than in ducks and grebes but the sample size for avocets was small (Table 3) and gulls and avocets were collected within a short period during their spring breeding season. No significant relationship was found between mercury and selenium in gulls ($F_{1,24} = 1.4$, $P = 0.25$) or avocets ($F_{1,4} = 0.03$, $P = 0.87$).

Table 3. Mercury and selenium ($\mu\text{g/g dw}$) in livers and blood from common goldeneyes, eared grebes, California gulls, and American avocets collected from Great Salt Lake.

Species	Liver		Blood	
	GM Hg ¹ (n) Range	GM Se ¹ (n) Range	GM Hg ¹ (n) Range	GM Se (n) Range
Common goldeneye	26A (40) 1.6 - 114	13A (40) 3.6 - 34	12A (40) 0.6 - 30	14A (40) 1.1 - 33
Eared grebe	11B (21) 4.5 - 32	11AB (59) 5.0 - 28	4.9B (43) 0.05 - 18	16A (43) 0.3 - 55
California gull	2.6C (24) 0.3 - 9.9	7.9B (24) 4.7 - 15	2.5C (24) 0.6 - 7.6	14A (24) 4.8 - 46
American avocet	1.9C (4) 1.7 - 2.7	13AB (4) 11 - 17	0.89C (4) 0.7 - 1.0	17A (4) 12 - 23

¹ Geometric means with different uppercase letters within columns are significantly different from each other (Tukey-Kramer $\alpha = 0.05$).

California Gull Mercury and Selenium

Gulls collected from Neponset Reservoir had significantly lower mercury concentrations in liver than gulls from Hat Island and lower blood concentrations than gulls from Great Salt Lake Minerals (GSLM) and Hat Island. However, selenium concentrations in liver and blood were similar to those observed in gulls from both Great Salt Lake sites (Table 4).

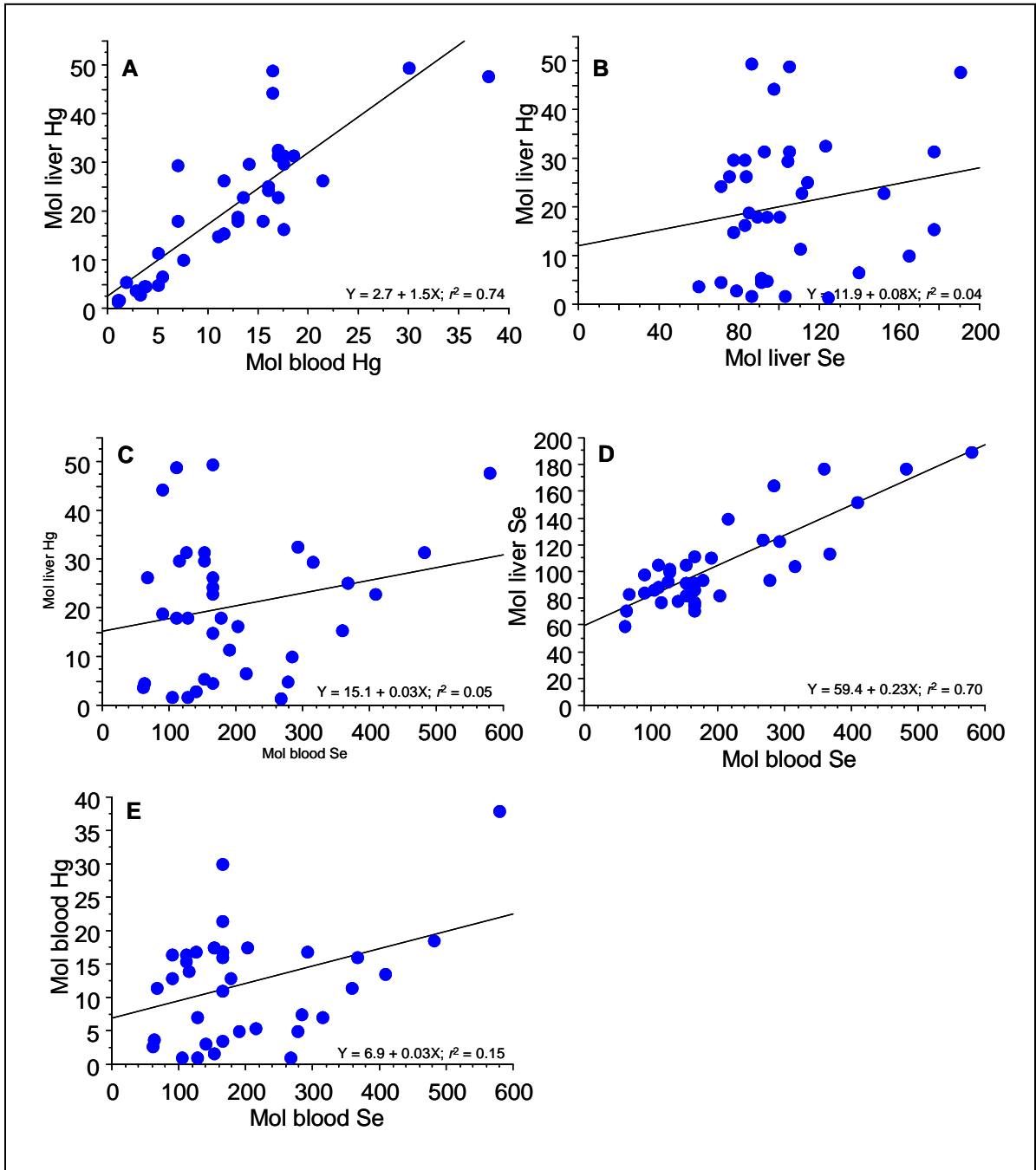
Table 4. Mercury and selenium ($\mu\text{g/g dw}$) in livers and blood from California gulls collected from Great Salt Lake and Neponset Reservoir.

Location	Liver		Blood	
	GM Hg ¹ (n) Range	GM Se ¹ (n) Range	GM Hg ¹ (n) Range	GM Se (n) Range
GSLM	2.9AB (12) 0.6 - 9.9	8.8A (12) 6.2 - 15	2.3A (12) 0.6 - 7.6	18A (12) 8.7 - 463
Hat Island	4.9A (12) 0.8 - 9.8	7.1A (12) 4.7 - 9.7	2.7A (12) 0.6 - 4.3	9.8B (12) 4.8 - 23
Neponset	1.6B (12) 0.3 - 5.9	7.9A (12) 5.6 - 13	0.8B (12) 0.2 - 3.2	14AB (12) 5.0 - 32

¹ Geometric means with different uppercase letters within columns are significantly different from each other (Tukey-Kramer $\alpha = 0.05$).

On a molar basis, gulls showed a significant positive relationship ($P < 0.001$) between mercury in liver and blood with a slope of 1.5 (Figure 6A) but did not show a significant relationships between mercury and selenium in livers ($P = 0.25$; Figure 6B) or mercury in liver versus selenium in blood ($P = 0.41$). There was a significant molar relationship ($P < 0.001$) between selenium in liver and blood with a slope of 0.23 (Figure 6D) and mercury and selenium in blood ($P = 0.02$; Figure 6E).

Figure 6. Molar relationship between mercury concentrations in liver and blood (A), liver mercury and selenium (B), liver mercury and blood selenium (C), liver and blood selenium (D), and blood mercury and selenium (E) of California gulls sampled from Great Salt Lake and Neponset Reservoir.



Discussion

We believe the most likely explanation for the higher-than-expected blood selenium concentrations is exposure to elevated mercury concentrations in GSL, as previously suggested (Santolo and Ohlendorf 2007).

Despite their common occurrence, biological effects of metal contaminant mixtures are poorly understood and difficult to predict. However, selenium and mercury are known to interact, and selenium is thought to have a protective effect against mercury by forming a stable and nontoxic complex. The interaction between these elements may also increase the retention and accumulation of mercury (Furness and Rainbow 1990) and perhaps selenium. Differences in the relationship between blood and liver selenium concentrations may be attributed to more rapid elimination from liver than blood and to binding of selenium to inorganic mercury forming an inert Hg-Se protein with a long half-life (Wayland et al. 2001). Alternatively, high mercury may cause a rapid increase in blood selenium, and faster than in liver as shown by the goldeneyes.

Studies by Henny et al. (2002) and Spalding et al. (2000) have shown high correlations of selenium with inorganic mercury on a molar basis in livers of fish-eating birds. Those authors suggested that selenium may contribute to the sequestration of inorganic mercury, thereby reducing its toxicity. This conclusion would be consistent with the results of a selenium-mercury interaction study with mallards by Heinz and Hoffman (1998) in which adults were fed 10 µg Se/g, 10 µg Hg/g, or 10 µg Se + 10 µg Hg/g (Heinz and Hoffman 1998). Female mallards fed the combination diet (10 µg Se + 10 µg Hg/g) had about 1.5 times higher liver selenium concentrations than those fed the selenium-only diet, and the male mallards fed the combination diet had almost 12 times the selenium concentration of those fed the selenium-only diet. Selenium provided a protective effect that reduced the toxicity of mercury to adult male ducks. However, when the diet contained 10 µg Se/g plus 10 µg Hg/g, the effects on reproduction were worse than for either selenium or mercury alone. The number of young produced per female and frequency of teratogenic effects were significantly affected by the combination of mercury and selenium in the diet. Selenium in eggs from mallards fed the combination diet, converted from wet weight to dry weight, were 47 µg/g, which was higher than control eggs (1.8 µg/g) and eggs from mallards fed 10 µg Se/g (38 µg/g) or 10 µg Hg/g (1.9 µg/g). Although in the GSL studies breeding gulls and avocets showed higher selenium concentrations than would be expected from dietary selenium alone, no reproductive effects were observed.

Japanese quail fed increasing concentrations of mercury with a constant concentration of selenium as selenite (i.e., 0 Hg + 6 µg Se/g, 2.5 ppm Hg + 6 µg Se/g, 5 µg Hg/g + 6 µg Se/g, 10 µg Hg/g + 6 µg Se/g, or 15 µg Hg/g + 6 µg Se/g) (nominal dw concentrations) for 20 weeks had blood selenium concentrations that increased with increasing mercury (El-Begearmi et al. 1977). This is similar to what we observed in common goldeneyes wintering at GSL.

In a field study, common eiders from the Arctic were sampled over a 2-year period (Wayland et al. 2001). From the first to second year, liver mercury concentrations more than doubled (from 1.8 to 3.9 µg Hg/g dw) but blood mercury concentrations did not change (0.23 µg Hg/dl). From the first to second year, liver selenium concentrations went from

20.1 to 18.5 $\mu\text{g Se/g dw}$ but blood selenium increased from 3.5 $\mu\text{g Se/dl}$ to 4.7 $\mu\text{g Se/dl}$. This suggests that with the higher mercury levels (i.e., in year 2), there was some proportion of selenium bound to mercury. Similarly, grebe liver mercury and selenium increased from September to November but blood mercury and selenium did not (Table 2).

In marine wading birds, selenium and mercury showed a strongly positive relationship in blood; however, this relationship was not observed in liver or kidney (Goede and Wolterbeek 1994). This suggests that the selenium concentrations observed in the blood were higher than in the diet and were caused by the higher mercury concentrations and is similar to what was observed at GSL.

Birds feeding at different sampling locations had diets that varied from saline to fresh water according to locations. Shorebirds from Ogden Bay and Saltair had at least partially freshwater diets. Gulls from Neponset Reservoir and possibly also Antelope Island and GSLM had at least partially freshwater diets. Shorebirds from Antelope Island and gulls from Hat Island likely had 100 percent diets from saline water. This may have affected exposure to mercury and selenium in individual shorebirds and gulls.

Selenium concentration in avocet blood was higher but avocets did not have a correspondingly elevated mercury concentration. However, the avocets were sampled at one point in time whereas grebes and ducks were sampled over a period of months. Avocets appear to take up selenium quickly and different species may have differences in how they process and retain selenium and mercury. For example, grebes feed only on the lake, and goldeneyes feed in marshes where more methylation of mercury occurs. Avocets eat corixids from freshwater sources (such as the KUCC outfall ditch and Ogden Bay site), as do some goldeneyes and gulls; however, grebes forage exclusively in the saline open waters.

As an essential nutrient, normal selenium concentrations in blood of birds should be about 1 $\mu\text{g/g}$, but there should normally not be any mercury in birds. Selenium and mercury in grebes almost doubled between September and November while they resided on GSL. Results of a recent study by Nathan Darnall/USGS et al. (personal communication) found a similar increase, with mercury increasing in December to almost 30 $\mu\text{g/g}$. This increase could be due to factors such as seasonal differences, stress, varying inflows, and temperatures.

There was more selenium than mercury in avocets and grebes. Although mercury undoubtedly contributes to the bioaccumulation of selenium, the elevated selenium levels may not be entirely due to mercury. In birds, uptake of selenium is faster than that of mercury, and the collected samples represent only a snapshot in time so they may not tell the entire story. Further study evaluating selenium and mercury concentrations in birds when they first arrive and then measuring those levels over time would be needed to definitively answer the question.

Generally, blood selenium concentrations were higher than liver concentrations in all birds collected at GSL. There was no significant difference among nesting colonies for selenium in livers of California gulls. However, blood selenium concentrations were significantly higher in gulls from the GSLM colony than the Antelope Island ($P = 0.007$) and Hat Island colonies ($P = 0.039$).

Laboratory analyses in samples collected during 2007 included mercury. Knowing mercury concentrations in bird samples provides us with information that can be used to identify possible reasons for the higher-than-expected blood selenium concentrations that were found. Dietary selenium influences mercury toxicity and can be directly related to mercury:selenium ratios (Kim et al. 1996, Henny et al. 2002, Nicholas et al. 2007).

The toxic effects of mercury in birds can include weight loss due to reduced food intake, weakness in wings and legs leading to difficulty flying and standing, and loss of coordination (Scheuhammer 1987). The toxic effects of selenium in adult and juvenile birds include reduced reproductive success, emaciation, and loss of feathers (Ohlendorf 2003). Mercury and selenium toxic effects were not observed in birds from GSL even though some concentrations were in the range where they would be expected. Correlation between mercury and selenium is not well established in birds, and there seem to be highly variable relationships that depend on species and concentrations. The mercury and selenium relationship may be influenced by the relative rates of accumulation (Ohlendorf 1993), the nonessential nature of mercury and essential nature of selenium, the excretion of MeHg (Kim et al. 1996), whether mercury is in the inorganic or methylated form (Henny et al. 2002), or other factors.

When there is a low concentration of mercury, a lower molar ratio is observed; however, at high mercury and selenium concentrations in the liver, most selenium binds mercury resulting in a mercury-to-selenium ratio greater than 1.0 (Kim et al. 1996). Common goldeneyes had significantly higher liver selenium concentrations than the other birds and also had a mercury increment of 1.3. This suggests that selenium plays a role in mercury detoxification for individuals with high mercury levels. However, molting and species-specific demethylation also influence the relationship.

In conclusion, both grebes and ducks showed a temporal increase of mercury and selenium in liver and blood and there was a strong molar relationship, especially in livers, between mercury and selenium for goldeneyes and grebes. In gulls, mercury concentrations were lower overall (especially in Neponset Reservoir birds) and the molar relationship for mercury and selenium was not evident. The blood selenium concentrations especially did not reflect the dietary or egg concentrations observed. A rapid increase in blood selenium appears to be associated with mercury exposure. The initial large increase in blood selenium, faster than in the liver, observed in the goldeneyes and grebes may explain the high blood selenium observed in shorebirds and gulls during 2006. Residence time for shorebirds and gulls before collection in 2006 is not known, but presumably they were in this "early exposure" phase. The grebe and duck mercury and selenium results support the hypothesis that the high selenium concentrations observed in gulls and shorebirds may have been due to the initial increase in blood selenium due to their exposure to mercury. Both dietary and egg concentrations of selenium were below levels of concern in that they were well below the EC₁₀ for effects derived in the laboratory with mallards. Dietary selenium concentrations may be too low or the exposure time too short for egg selenium concentrations to be greatly affected by the mercury-selenium interactions at GSL.

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