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Selenium mass balance in the Great Salt Lake, Utah

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ABSTRACT

A mass balance for Se in the south arm of the Great Salt Lake was developed for September 2006 to August 2007 of monitoring for Se loads and removal flows. The combined removal flows (sedimentation and volatilization) totaled to a geometric mean value of 2079 kg Se/yr, with the estimated low value being 1255 kg Se/yr, and an estimated high value of 3143 kg Se/yr at the 68% confidence level. The total (particulates+dissolved) loads (via runoff) were about 1560 kg Se/yr, for which the error is expected to be $\pm 15\%$ for the measured loads. Comparison of volatilization to sedimentation flux demonstrates that volatilization rather than sedimentation is likely the major mechanism of selenium removal from the Great Salt Lake. The measured loss flows balance (within the range of uncertainties), and possibly surpass, the measured annual loads. Concentration histories were modeled using a simple mass balance, which indicated that no significant change in Se concentration was expected during the period of study. Surprisingly, the measured total Se concentration increased during the period of the study, indicating that the removal processes operate at their low estimated rates, and/or there are unmeasured selenium loads entering the lake. The selenium concentration trajectories were compared to those of other trace metals to assess the significance of selenium concentration trends.

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1. Introduction

A numerical standard for selenium concentrations in the open waters in the Great Salt Lake is being developed to protect avian wildlife. The Great Salt Lake has been designated by the Western Hemisphere Shorebird Reserve Network as a Hemispheric Site because it supports a large population of migratory birds (Aldrich and Paul, 2002). The lake supports over 2 million shorebirds and at least 3.5 million waterfowl during spring and fall migration (Aldrich and Paul, 2002; Schussman, 1999). Over 1.5 million of eared grebes feed on brine shrimp in the lake each fall (Utah Division of Wildlife Resources, 2004).

The Great Salt Lake is the fourth largest terminal lake worldwide (Stephens, 1990) and the largest saline lake in

North America (Brix et al., 2004). The lake is shallow (maximum depth is around 9 m) with a vast surface area that varies considerably depending on its surface elevation (Baskin, 2005). The Great Salt Lake displays spatial variability among its four bays (Fig. 1), which are Gunnison Bay (north arm) and Bear River Bay in the north, and Gilbert Bay (south arm) and Farmington Bay in the south. The Southern Pacific Railroad causeway divided Gunnison and Gilbert Bays in 1959 (Gwynn, 2002; Loving et al., 2000). More than 90% of the freshwater runoff enters the south arm of the lake, via the Bear River (Bear River Bay), Weber River (Ogden Bay), and Jordan River (via Farmington Bay) (Stephens, 1990; Gwynn, 2002). The causeway separating Farmington Bay from the south arm is breached to allow bi-directional flow. In contrast to the south arm, the runoff to the north arm is very limited.

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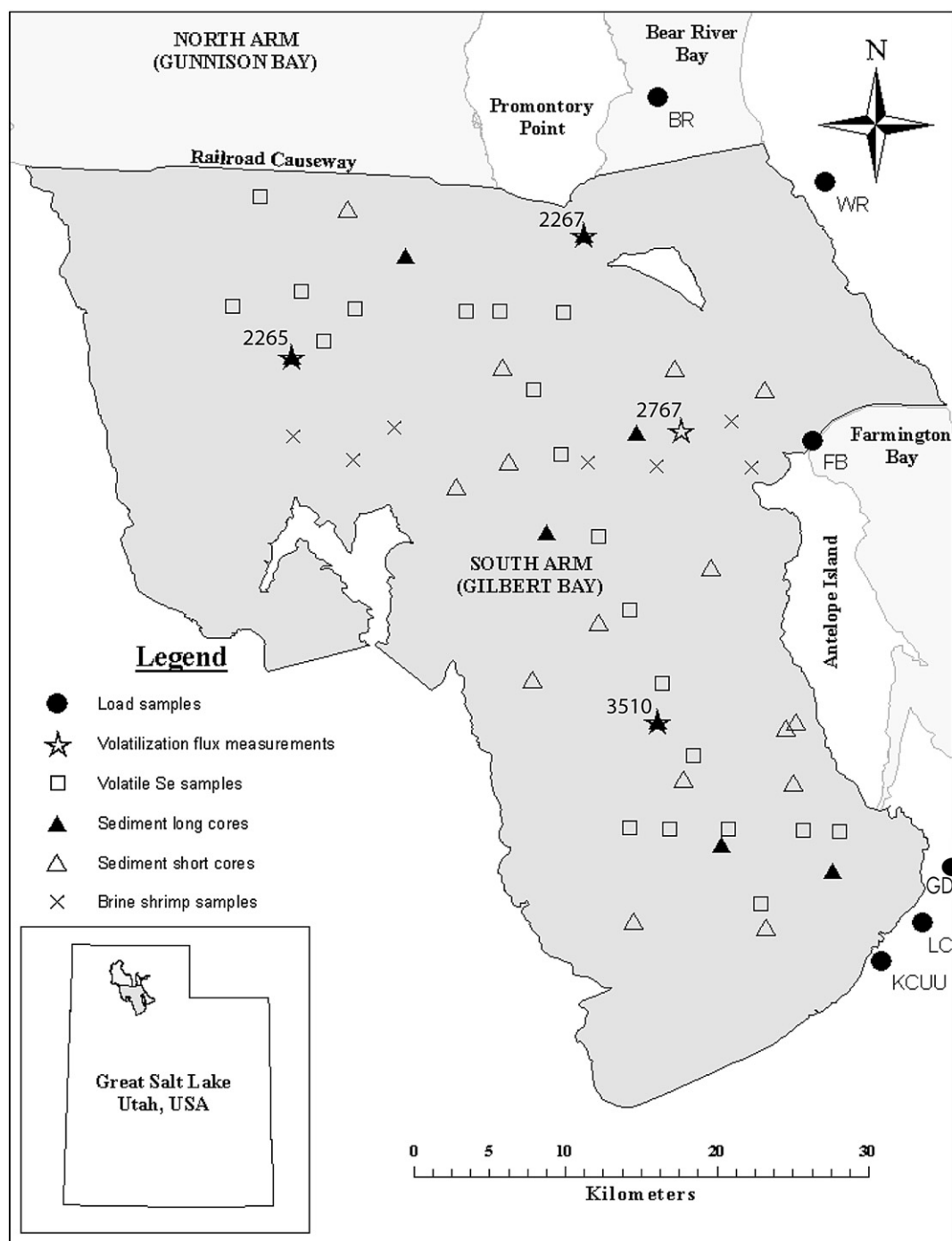


Fig. 1–Map of the Great Salt Lake showing sampling locations. Full circles stand for water inputs sampling locations. Stars stand for volatilization flux measurements. Squares stand for volatile Se samples. Triangles stand for sedimentation sampling locations (full triangles correspond to long cores and open triangles to short cores). X-symbol stands for brine shrimp sampling locations. Some sites show overlapping sampling locations, for example, volatilization flux measurement sites overlap with the long core sites in some cases, thereby making what appears to be a closed star when in fact it is actually an open star and an overlaid closed triangle.

As a result the north arm brine is more saline (16 to 29% salinity) (via evaporative concentration) and denser than the south arm brine (6 to 28% salinity) (Stephens, 1990).

Flow can be bi-directional through the Southern Pacific Railroad causeway (Gwynn, 2002; Loving et al., 2000) separat-

ing the north and south arms; less dense brine flows from the south arm to the north arm (near the surface); whereas, denser brine flows from the north arm to the south arm (at depth). The dense brine entering the south arm forms a “deep brine layer” in the south arm that extends as far south as the

southern tip of Antelope Island (Gwynn, 2002). The considerable density contrast is sufficient to yield permanent stratification despite temperature inversion during winter and spring months (Lin, 1976). The permanent stratification yields anoxia in the deep brine layer. The geochemical reactions governing Se and trace metals in the anoxic deep brine layer contrast strongly against those in the oxic shallow brine layer, yielding potential for a closed redox cycle for Se and other redox-sensitive trace elements in this density- and geochemically-stratified system (Tayler et al., 1980).

The Great Salt Lake displays striking seasonal and inter-annual variations in lake elevation, superficial area, and volume. The areal extent of the Great Salt Lake varies corresponding to variation in runoff, with highest runoff relative to evaporation occurring during late spring (snow melt), and lowest runoff relative to evaporation in late summer (Loving et al., 2000). These variations in lake stage correspond to vast changes in areal extent and volume of the lake (Fig. S1 in the Supporting Information) (Baskin, 2005). Due to inter-annual variations in runoff and evaporation, lake stage and area fluctuate dramatically over decadal time scales. The historic low elevation of the Great Salt Lake was 1277.52 m in 1963; whereas its historic high elevation (1283.77 m) was observed only 23 years later during 1986–1987 (Stephens, 1990).

Selenium is an essential nutrient for living organisms; however, the threshold between being essential and toxic is very narrow, particularly for birds and aquatic species (EPA, 1998; Lemly, 1997; Cai, 2003). Selenium concentrations between 5 and 10 $\mu\text{g/g}$ in bird diets have caused reproductive impairment (embryonic mortality and deformity) in the Kesterson Reservoir, CA (Ohlendorf et al., 1989; Ohlendorf, 2002). Previously the Great Salt Lake was protected for beneficial uses by a narrative (non-numeric standard), which states that it is unlawful to discharge any substance that “may become offensive” or produces annoyance (for instance, taste

in edible organisms) (State of Utah, 2008). Development of a numeric standard is driven by a proposal to dispose of reverse osmosis (RO) concentrate in the Great Salt Lake (Johnson et al., 2006) with a reported annual Se load to the lake of 84 to 134 kg Se/yr via a 993,755,000 to 1,591,690,000 m^3/yr stream with and Se concentration of 84 $\mu\text{g/L}$ (Geochimica, 2002).

In addition to recreation and habitat, beneficial uses of the Great Salt Lake include industry. Several companies extract salts (mainly sodium chloride), generating around \$300 million every year (Isaacson et al., 2002). The lake supports a brine shrimp industry that is valued globally for high protein brine shrimp cysts (Isaacson et al., 2002), particularly as food for Asian tiger prawns.

Upon setting a numerical standard for Se in the Great Salt Lake, the goal becomes to manage selenium loads in order to ensure that the numeric standard is not exceeded. Potential Se inputs include streams and canals; groundwater; and, wet and dry deposition. The three major runoff sources are the Bear River, Weber River and Jordan River (Stephens, 1990), whereas three additional major potential sources of Se loads include the Goggin Drain (GD), Kennecott Drain Outfall (KUCC); and the Lee Creek (LC) (Naftz et al., 2008). Net inflow from the north arm represents another potential Se input into the south arm. Selenium concentrations in groundwater entering the lake have not yet been measured, nor have wet and dry deposition of Se to the Great Salt Lake.

Potential Se outputs include volatilization, permanent sedimentation, export or migration of biota from the lake (e.g. brine shrimp harvest and migrating birds) and potential net export of Se to the north arm. Volatilization fluxes of Se from the Great Salt Lake were recently measured (Diaz et al., 2008). In a related study, permanent sedimentation fluxes of Se from the lake were also measured (Oliver et al., 2008). Selenium removal via brine shrimp export was also recently calculated (Marden, 2007). Naftz et al. (2008) provided highly preliminary estimates

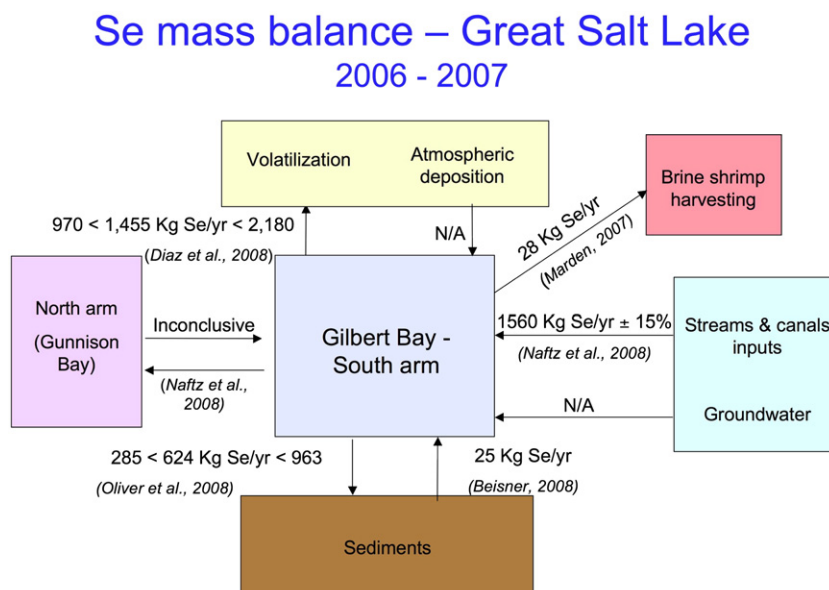


Fig. 2–Selenium mass balance for the south arm of the Great Salt Lake for 2006–2007.

of potential loss of Se to the north arm ranging as high as 880 kg Se/yr (Naftz et al., 2008). Potential storage (internal sinks and sources) of Se includes temporary sedimentation in anoxic bottom sediment and resolubilization in oxic water via resuspension or other events that place anoxic sediment in periodic contact with oxic water (Beisner, 2008). The corresponding studies comprise a geochemical sampling program in support of developing a Se standard for the open waters of the Great Salt Lake, funded by the Utah Department of Environmental Quality.

The goal of this paper is to compare recently measured Se inflows to, and Se outputs from, the Great Salt Lake, and to compare the resulting mass balance to measured concentration histories in order to assess the degree which observed concentrations reflect the measured mass balance. The mass balance was carried out within a defined control volume corresponding to the main body (open water) of the south arm of the Great Salt Lake. Shorelines in this control volume are barren except for the eastern shoreline between the Southern Pacific railroad and Farmington Bay causeways (Fig. 1). The loads to the lake were measured downstream of wetland influences.

2. Methods

The control volume boundary for the selenium mass balance was established within the open waters of the south arm (Gilbert Bay) (Fig. 1). The north boundary of the control volume is the railroad causeway separating the north and south arms (Gunnison and Gilbert Bays). The east boundary is formed by the lakeshore, except where the Farmington Bay causeway forms the southeast boundary. The southern and western boundaries are formed by the lakeshore. The control volume includes both the shallow and deep brines.

A mass balance for selenium was developed for the period May 2006 to July 2007, during which data regarding inputs and outputs to the lake were collected (Fig. 2). Aqueous chemical conditions were characterized in the field at four locations (2267, 2565, 2767 and 3510) across the main body of the Great Salt Lake (Fig. 1) and at 7 to 13 depths (varying by station), ranging from 0.2 to 8 m depth below lake surface (more details in the Supporting Information).

2.1. Load sampling locations and analysis

Se loads from the Great Salt Lake watershed tributaries were investigated recently by Naftz et al. (2008). Six continuous streamflow gages were operated by the U.S. Geological Survey (USGS) at locations where runoff enters the Great Salt Lake (Naftz et al., 2008) (Fig. 1). Locations included the bays of the Bear River (BR), Weber River (WR), and Farmington Bay (FB); as well as the Kennecott Utah Copper Corporation (KUCC) outfall, Lee Creek (LC), and Goggin drain (GD) (Naftz et al., 2008). Samples were collected monthly or bimonthly and were analyzed for Se. Se was analyzed by hydride generation and atomic fluorescence spectrometry (HG-AFS). Measured Se concentrations and discharges were integrated to yield loads (Naftz et al., 2008) using the USGS software LOADEST (Runkel et al., 2004).

2.2. Volatile Se sampling locations and analysis

Diaz et al. (2008) studied the volatilization of Se from the south arm of the Great Salt Lake during the period from summer 2006 to summer 2007. The purge and cryo-focusing trap process to collect near surface volatile Se samples was performed on the lake in order to avoid degradation of the water samples during transport and holding. Volatile Se samples were collected monthly. The cryo-focusing trap system consists of a reactor (a modified desiccator) with a diffuser connected to a helium line. The reactor can sparge 7 L of hypersaline water. The cryo trap system purges the vast majority of volatile species present via continuous sparging with inert vapor. The vapor was swept from the reactor via Teflon tubing to a glass water trap (–55 °C, dry ice/ethanol) to remove water from the flowing vapor. The vapor then entered a glass trap (–196 °C, liquid nitrogen) to trap the volatile compounds collected from the water. After collection, nitric acid was added to the glass trap to oxidize volatile Se compounds and convert them to their stable aqueous species. The closed trap was digested in a water bath at 75 °C for 3 h, and the solution was analyzed for Se by inductively coupled plasma mass spectrometry (ICP-MS) at the University of Utah Center for Water, Ecosystems, and Climate Science (CWECS) laboratory. Measured near-surface (0.2–0.5 m) volatile selenium concentrations from 4 stations (2267, 2767, 2565, 3510, Fig. 1) in the shallow brine were characterized using a sinusoidal function to provide continuous values, which along with measured temperature, wind speed and lake area allowed estimation of annual volatilization flow of Se (Diaz et al., 2008). The expression used for water transfer velocity was developed by Liss and Merlivat (1986) and modified by Livingstone and Imboden (1993) (Diaz et al., 2008). This expression was further modified to include the Schmidt number for dimethylselenide as a polynomial function of temperature (Saltzman and King, 1993), and the influence of viscosity and diffusivity according to the boundary layer model (Schwarzenbach et al., 2003).

2.3. Permanent sedimentation sampling locations and analysis

Oliver (2008) and Beisner (2008) determined that frequent sediment re-suspension yields sedimentation rates in sediment traps that are far greater than permanent sedimentation. Hence, permanent Se removal via sedimentation was estimated using sedimentation rates determined from sediment cores, as described below.

Twenty eight lake cores (20 short and 8 long cores) were taken at various sites in the south arm of the GSL during July 2006, June and July 2007 (Fig. 1) (Oliver et al., 2008). Short cores (<5 cm) were sectioned in-situ into 0–1 cm and 4–5 cm intervals, for analysis. The upper 10 cm of each of the long cores (~30 cm) was sectioned in-situ into 1 or 2 cm increments within 6 h after collection. Core slices were then chilled on ice prior to freeze-drying. A gravity coring device was used to collect all long cores, with the exception of site 3510, which was collected using a box corer, which was also used to collect the short cores. The box corer minimizes compaction to provide the best possible determination of age

as a function of depth (and sedimentation rate) in the shallowest sediment. All core slices were freeze-dried, ground with a clean ceramic mortar and pestle, and homogenized by mechanical mixing. Wet and dry weights were recorded. In the eight long cores (Fig. 1), one fraction of each core interval was analyzed for Se by hydride generation–atomic absorption spectrometry at LET, Inc. (Columbus, MO). The freeze-dried core fractions were digested using the L5-magnesium dry ash digestion procedure in which the Se was extracted from the sediment using a prescribed application of magnesium nitrate hexahydrate, HNO₃, HCl, and heat. The Se results were corrected for salt content. The core samples were analyzed for ²¹⁰Pb, ²²⁶Ra, ¹³⁷Cs, and ⁷Be by gamma spectrometry at the USGS Sediment Radioisotope Laboratory in Menlo Park, CA as described in Van Metre et al. (2004). A sediment mass accumulation rate was determined in each core based on ²¹⁰Pb decay using the constant rate of supply (CRS) model (Appleby and Oldfield, 1983). The Se sedimentation rate was then determined using the mass accumulation rates and selenium concentrations in the core samples. The Se sedimentation rates were integrated spatially using sedimentation contours derived from long-term sedimentation rates inferred from Holocene sediment thickness (Oliver et al., 2008).

2.4. Brine shrimp sampling locations and analysis

Brine shrimp samples were collected monthly during 2006 and 2007 from 7 sites and 3 depths (shallow at 1–3 m in depth; medium at 5–6 m in depth; and, deep at 8–9 m in depth) in the main body of the south arm of the Great Salt Lake (Fig. 1) (Marden, 2007). Samples were collected via plankton net hauls that were pooled vertical, or horizontal (for the 1 m sites only). All samples were sieved through 850, 500, and 125 µm opening sizes sieves (125 µm for nauplii-cysts; 500 µm for juvenile; 850 µm for adult). Each collected fraction was rinsed with pre-filtered Great Salt Lake water, saved in Whirl-pak® bags, and stored on ice for transport to the laboratory. In the laboratory, brine shrimp samples were cleaned from other zooplankton or debris; water was removed via vacuum filtering or using a pipette; and samples were frozen at –25 °C. Brine shrimp samples were sent to a contract lab (LET Incorporated, Columbia, MO) for Se analysis. Samples were freeze-dried and then acid digested previously to be analyzed using hydride generation coupled with atomic absorption spectrometry (Marden, 2007).

2.5. Concentration trajectories

The concentration trajectories of total Se (dissolved plus particulate) over the course of the study were explored by integrating the total Se concentration over time via the following mass balance:

$$[Se]_t = [Se]_{t-1} + \frac{[Se]_{load} - Se_{volatilization} - Se_{permanent\ sedimentation} - Se_{brine\ shrimp\ harvesting}}{V}$$

where $[Se]_t$ and $[Se]_{t-1}$ represent the total Se concentration in µg/L for the present and previous time steps, respectively; Se_i (i =load or removal process) represents the mass flow

(loading or removal) per unit time; and V represents the volume of the lake.

The mass balance was determined from May 19, 2006 to August 1, 2007 using daily values of fluxes (loads and removal processes). The daily values for loads and volatilization were determined as described in Naftz et al. (2008) and Diaz et al. (2008), respectively; whereas the daily values for permanent sedimentation and brine shrimp harvest were determined by division of the annual values by 365. Daily values of surface area and volume of the Great Salt Lake, for the same period, were obtained from the USGS National Water Information System (NWIS) (<http://waterdata.usgs.gov/nwis>).

3. Results and discussion

A Se mass balance was developed to determine whether measured Se removal flows match measured Se loads in the Great Salt Lake. The various loads and removal flows (measured and unmeasured) are summarized in Fig. 2. Measured total Se loads (from streams) to the south arm of the Great Salt Lake were approximately 1560 kg Se/yr, with an estimated error of 15%, during the annual period between summer, 2006 and summer, 2007 (Naftz et al., 2008). The main contributors to the loads are detailed in Table S1 in the Supporting Information (Naftz et al., 2008).

The mean Se removal flows via volatilization and sedimentation were estimated as described in the Methods. The annual volatilization flow (1455 kg Se/yr) was determined via temporal integration of hourly estimated flux rates from measured dissolved volatile Se concentrations, wind velocities, and temperatures. The annual permanent sedimentation flux (624 kg Se/yr) was estimated via spatial integration of sedimentation rates determined in discrete sediment cores using ²¹⁰Pb analyses. Uncertainties in these mean removal flows were determined by propagation of errors to yield high and low estimates around the mean (Diaz et al., 2008; Oliver et al., 2008), and these values are also given in Fig. 2. Se removal flow due to brine shrimp harvest (26 kg/yr) was estimated using measured Se concentrations in brine shrimp and documented brine shrimp harvest mass in 2006 (Marden, 2007).

The above removal flows total to approximately 2110 kg Se/yr, which exceeds the estimated loading (1560 kg Se/yr). However, the removal flow estimates represent mean values with significant estimated errors (Fig. 2). For permanent sedimentation, the arithmetic standard deviation is ±339 kg Se/yr (Oliver et al., 2008), yielding a range from 285 kg Se/yr to 963 kg Se/yr. For volatilization, the 68% confidence interval (1σ) around the geometric mean (1455 kg Se/yr) yields a range from 970 kg Se/yr to 2180 kg Se/yr (Diaz et al., 2008). Given the degree of uncertainty, it is reasonable to state that the estimated removal flows ostensibly balance the measured loads, although the uncertainties are significant, as further discussed below.

Notably, volatilization is estimated to be more important than permanent sedimentation in removing Se from the Great Salt Lake. This finding represents a major change from past conceptualizations of how the Great Salt Lake regulates trace metal concentrations. Previously the lake was assumed to

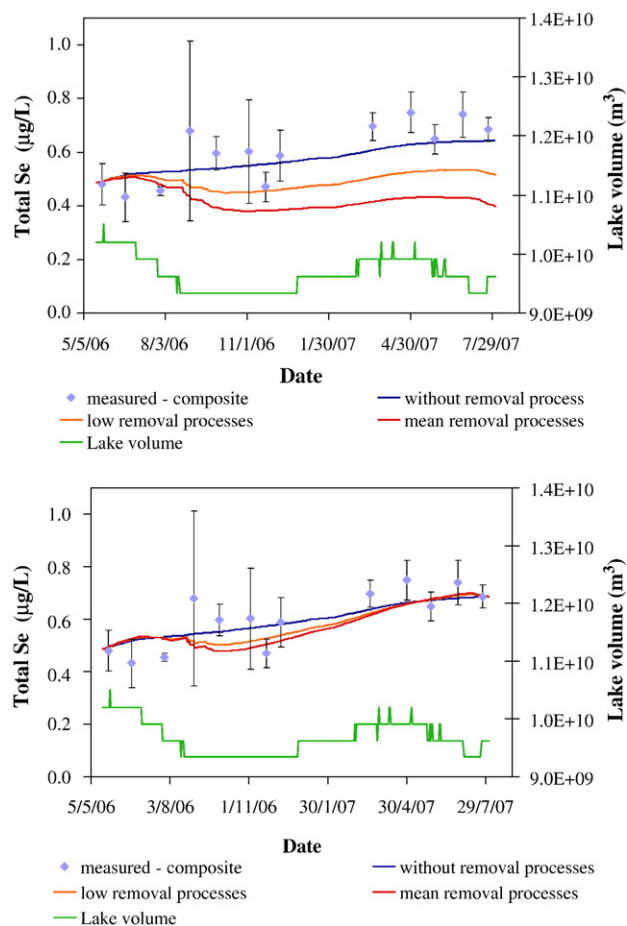


Fig. 3 – Mass balance integration of Se concentration in the Great Salt Lake. Top: using estimated loads and removal processes. Bottom: adding estimated unmeasured loads to reach the July/07 target Se concentration. [Se] measured stands for composite data from 4 sites and all shallow depths.

remove trace metals via permanent sedimentation due to formation of sulfide and other trace metal carrying particulates in the anoxic deep brine (Tyler et al., 1980).

Assuming, for the sake of analysis, that Se load and removal flows basically balance one another over an annual cycle (e.g. 1560 kg Se/yr loaded/removed), then the residence time of Se in the Great Salt Lake is on the order of about 3 to 5 years, based on the observed mass of Se in the lake, which ranges from approximately 5000 to 7500 kg, based on the observed variation in measured Se concentration and lake volume during the course of the study (Naftz et al., 2008; Baskin, 2005).

Notably, the Se concentration in the lake, as measured by hydride generation atomic fluorescence spectroscopy (HG-AFS) showed a significant increase over the course of the study in the measured total and dissolved Se concentrations (Fig. S2 in the Supporting Information). These increases were statistically significant (90% confidence interval based on Mann-Kendall test), and were observed in both the deep and shallow brine layers during the period of the investigation, constitut-

ing a net increase ranging between 0.16 and 0.34 µg/L Se in the Great Salt Lake (Naftz et al., 2008).

The significance of the observed increase in Se concentration over the period of study is also demonstrated in the corresponding mass. The error in the HG-AFS analysis is about 15% (Naftz et al., 2008), which corresponds to an error of approximately 0.08 µg/L Se based on the mean Se concentration of 0.5 µg/L Se (252 samples). This mean concentration and error correspond to a mass of Se in the lake of about 5000 kg±800 kg given the volume of the lake during the course of the study. The observed net increase in Se concentration of 0.17 µg/L over the course of the study corresponds to 1560 kg of Se (Naftz et al., 2008), indicating that this mass increase is approximately twice the analytical error.

A mass balance approach was used to examine the apparent increase in Se concentration during the course of the study in terms of the estimated loads and removal flows. The mass balance incorporated the following parameters to yield the simulated history of Se concentrations over time: 1) daily values for loads and volatilization obtained by modeling (Naftz et al., 2008; Diaz et al., 2008); 2) Daily values of surface area and volume of the lake (USGS NWIS database); 3) and daily values for permanent sedimentation and brine shrimp cyst harvest determined by division of the estimated annual values by 365.

Fig. 3 shows the averaged measured Se concentrations (4 sites, all depths in shallow layer) for samples that were collected within 2–3 consecutive days of each other. Averaging different sites and depths in the shallow layer was justified by the similar trends in the spatially averaged Se concentration with time at different depths (Fig. S3 in the Supporting Information). When removal flows were set to zero, the total selenium concentration increased during the course of the study, yielding a value (0.64 µg/L), which was only slightly lower than the HG-AFS-measured value in July, 2007 (0.69 µg/L=average of four sampling sites) (Fig. 3 top). Addition of the estimated mean removal flows (via volatilization, permanent sedimentation and brine shrimp harvest), produced a net decrease in the total selenium concentrations during the time period of the investigation, yielding a final estimated total selenium concentration (0.38 µg/L), which was low (by about 0.31 µg/L) relative to the measured value in July, 2007 (Fig. 3 top). Notably, the shapes of these simulated concentration histories reflect the temporal variation in Se removal via volatilization and loads via streams. Volatilization, which is highest during warmer months, causes the strong drop in Se concentration during summer and fall (Fig. 3). Stream loading causes the increase in Se concentration (and lake volume) during springtime. Replacement of mean removal flows by estimated low removal flow values (Table S2 in the Supporting Information) produced a negligible change in simulated Se concentration during the course of the study, and yielded a final simulated total selenium concentration (0.48 µg/L) that was somewhat below (about 0.21 µg/L) the measured value in July 2007 (Fig. 3 top). This observation indicates that the Se removal flows may operate in the low end of their estimated ranges.

Without the removal flows, the final observed Se concentration was not achieved in the mass balance model,

indicating that unmeasured loads to the Great Salt Lake exist. Potential sources of unmeasured Se load were previously described by Naftz et al. (2008), and include: (1) submarine groundwater discharge; (2) wet and dry atmospheric deposition falling directly on the lake surface; (3) resolubilization of Se from lake sediment into the overlying oxic water column; and (4) poorly characterized Se exchange between the north and south arm. Resolubilization of sediment Se into overlying oxic water was examined by Beisner (2008) who found this flow to be small, e.g., 25 kg per resolubilization event (Fig. 2). The additional loads necessary to achieve the observed concentration were 420, 1940 and 2770 kg Se/yr for no, low, and mean removal flows, respectively (Fig. 3 bottom). If the observed concentrations were less scattered, it would be

possible to discern the extent to which the expected influence of volatilization is indicated in the observed data; thereby constraining the volatilization flow. However, the scatter in the observed data precludes this assessment, and does not allow refinement of the volatilization flow based on the observed temporal trends in Se concentration.

The apparent balance between loads and removal flows (within the range of uncertainties) must also be considered in light of the timescales represented by these estimates. Specifically, the measured loads and the volatile Se flow correspond to the period of study (2006 to 2007); whereas, the sedimentation flow was estimated using sediment cores that represented timescales on the order of decades to centuries. The actual sedimentation rate during the period of

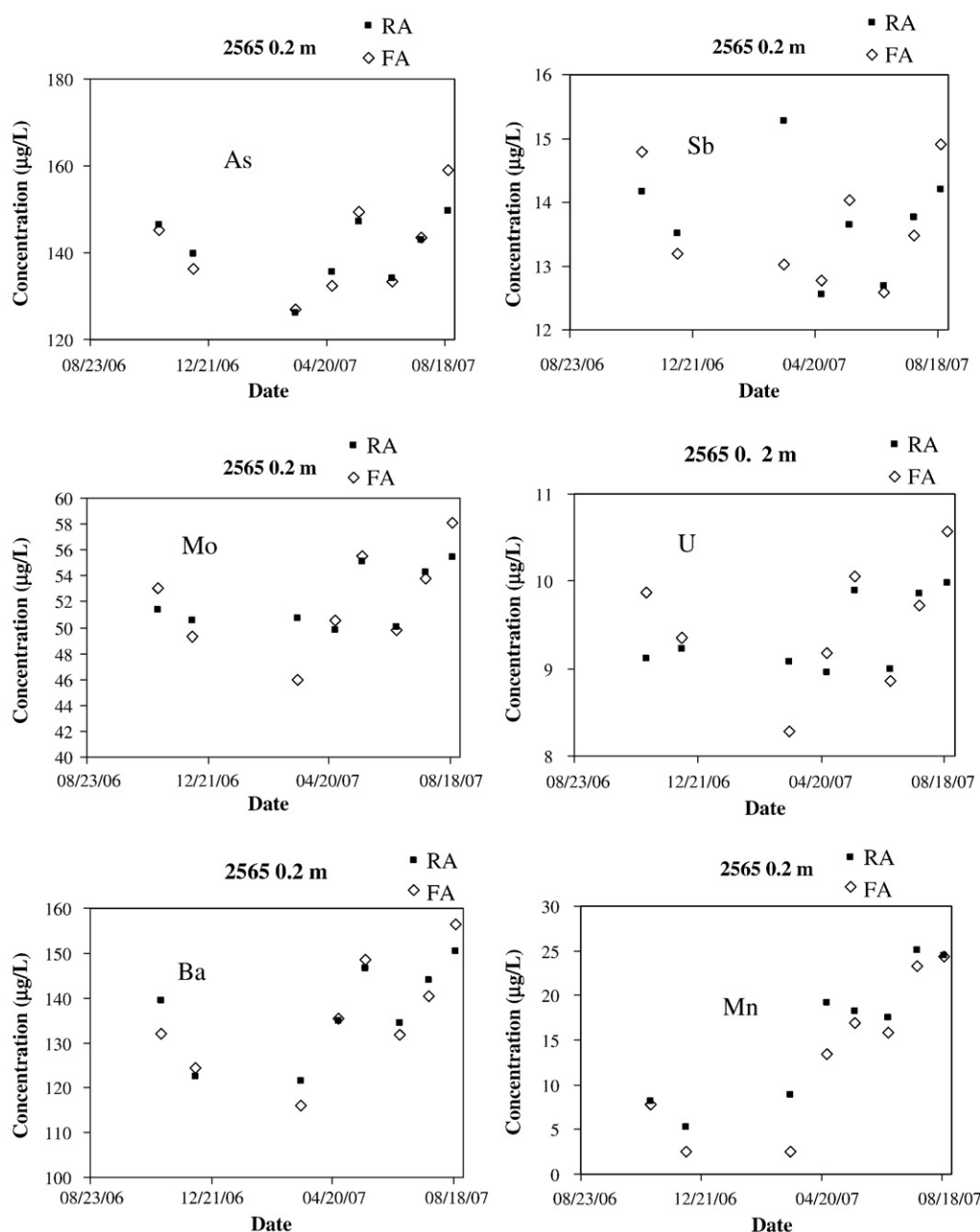


Fig. 4—Selected trace metals trajectories measured via CC-ICP-MS for shallow brines represented by site 2565 at 0.2 m.

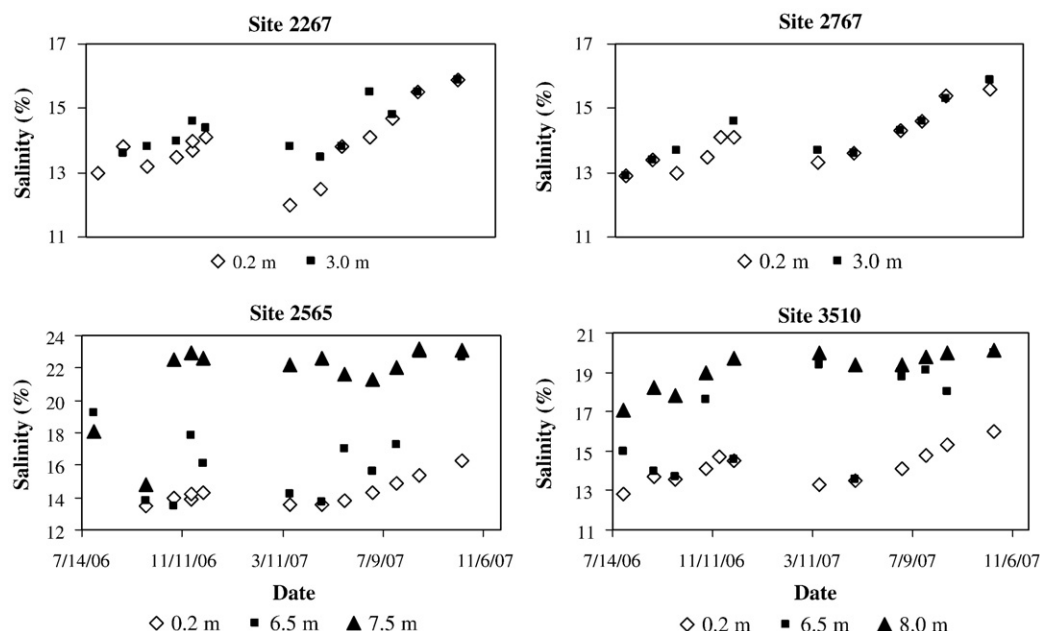


Fig. 5 – Discrete values of salinity measured during 2006–2007 in the south arm of the Great Salt Lake at four stations (2267, 2767, 2565 and 3510).

study may have differed from the time-averaged value obtained from the sediment cores, thereby influencing the balance of loads and removal flows. Sedimentation rates specific to the period of study would be potentially provided via sedimentation rates measured in sediment traps deployed in the lake (Oliver, 2008). However, sedimentation rates in sediment traps were much greater than permanent sedimentation rates, due to periodic re-suspension of bottom sediments (Oliver, 2008; Beisner, 2008), as indicated by isotopic analyses.

The concentration trajectories of Se differ from those of most of the other trace metals. Elements such as As, Sb, Mo, U and Ba showed concentrations that were slightly higher in summer–fall and lower in winter–spring (Fig. 4), suggesting seasonal controls on their concentrations. More important, they showed negligible increases in both total (RA) and dissolved (FA) concentrations over the course of the study (Fig. 4). The salinity of the lake (as shown for the four sampling sites) increased only slightly over the course of the investigation (~10%) (Fig. 5), which was far lower than the observed increase in Se concentration. In contrast, Mn concentrations increased by factors of 1.4 to 2.0 during the same period at all sites. The different trends for Se and Mn relative to the other trace metals may reflect differences in geochemical processes or different balances of loads and removal flows among these elements. In the case of Se, the mass balance indicates that loads may have exceeded removal fluxes over the course of the study, warranting continued monitoring of Se concentrations in the open water of the Great Salt Lake concurrent with continued monitoring of riverine fluxes to the lake. In order to develop a more comprehensive mass balance, it is recommended to continue or implement monitoring on the input and output fluxes that were not available for this study, such as the bi-directional flux of Se in the Pacific Railroad causeway

(north arm); the Se input via atmospheric deposition; and, the Se input through the groundwater flux.

4. Conclusions

Contrary to previous expectations, volatilization, rather than sedimentation, appears to be the dominant mechanism of Se removal from the Great Salt Lake. Given the range of uncertainties in measured Se removal flows and measured Se loads, it is reasonable to say there was a nominal balance of loads and removal flows. However, this apparent mass balance does not reflect the observed increases in Se concentration during the course of the study, which suggests that the estimated removal fluxes operate on the low end of their estimated ranges, and indicates the existence of unmeasured loads of Se to the Great Salt Lake. The trend in Se concentration warrants a long-term program to monitor Se concentrations in the lake and Se loads from contributing streams.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at [doi:10.1016/j.scitotenv.2008.11.029](https://doi.org/10.1016/j.scitotenv.2008.11.029).

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