

# Compositional changes in sediments of subalpine lakes, Uinta Mountains (Utah): evidence for the effects of human activity on atmospheric dust inputs

Richard L. Reynolds · Jessica S. Mordecai ·  
Joseph G. Rosenbaum · Michael E. Ketterer ·  
Megan K. Walsh · Katrina A. Moser

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**Abstract** Sediments in Marshall and Hidden Lakes in the Uinta Mountains of northeastern Utah contain records of atmospheric mineral-dust deposition as revealed by differences in mineralogy and geochemistry of lake sediments relative to Precambrian clastic rocks in the watersheds. In cores spanning more than a thousand years, the largest changes in composition occurred within the past approximately 140 years. Many elements associated with ore deposits (Ag, As, Bi, Cd, Cu, In, Mo, Pb, S, Sb, Sn, and Te) increase in the lake sediments above depths that correspond to about AD 1870. Sources of these metals from mining districts to the west of the Uinta Mountains are

suggested by (1) the absence of mining and smelting of these metals in the Uinta Mountains, and (2) lower concentrations of most of these elements in post-settlement sediments of Hidden Lake than in those of Marshall Lake, which is closer to areas of mining and the densely urbanized part of north-central Utah that is termed the Wasatch Front, and (3) correspondence of Pb isotopic compositions in the sediments with isotopic composition of ores likely to have been smelted in the Wasatch Front. A major source of Cu in lake sediments may have been the Bingham Canyon open-pit mine 110 km west of Marshall Lake. Numerous other sources of metals beyond the Wasatch Front are likely, on the basis of the widespread increases of industrial activities in western United States since about AD 1900. In sediment deposited since ca. AD 1945, as estimated using  $^{239+240}\text{Pu}$  activities,

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R. L. Reynolds (✉) · J. G. Rosenbaum  
US Geological Survey, Box 25046 Federal Center,  
Denver, CO 80225, USA  
e-mail: rreynolds@usgs.gov

J. G. Rosenbaum  
e-mail: jrosenbaum@usgs.gov

J. S. Mordecai  
Department of Geography, University of Utah,  
Salt Lake City, UT 84112, USA  
e-mail: jessmordecai@gmail.com

M. E. Ketterer  
Department of Chemistry and Biochemistry, Northern  
Arizona University, Flagstaff, AZ 86011, USA  
e-mail: michael.ketterer@nau.edu

M. K. Walsh  
Department of Geography, University of Oregon,  
Eugene, OR 97403, USA  
e-mail: mw Walsh2@uoregon.edu

K. A. Moser  
Department of Geography, University of Western Ontario,  
1151 Richmond North Street, London,  
ON N6A5C2, Canada  
e-mail: kmoser@uwo.ca

increases in concentrations of Mn, Fe, S, and some other redox-sensitive metals may result partly from diagenesis related to changes in redox. However, our results indicate that these elemental increases are also related to atmospheric inputs on the basis of their large increases that are nearly coincident with abrupt increases in silt-sized, titanium-bearing detrital magnetite. Such magnetite is interpreted as a component of atmospheric dust, because it is absent in catchment bedrock. Enrichment of P in sediments deposited after ca. AD 1950 appears to be caused largely by atmospheric inputs, perhaps from agricultural fertilizer along with magnetite-bearing soil.

**Keywords** Lake sediments · Mineral dust · Metals · Phosphorus · Magnetic properties · Uinta Mountains

## Introduction

Many factors influence composition of lake sediments, including sediment source, weathering and pedogenesis in the catchment, productivity, sediment-transport energy, chemical precipitation of minerals within the water column, and post-depositional alteration. Sediments, therefore, reveal aspects about environmental change that affected lake-catchment systems on time scales of years to millennia (Haworth and Lund 1984; Last and Smol 2001). Rock-derived detritus in lake deposits can commonly come from outside the catchment as atmospheric dust (Psenner 1999) that can be distinguished from locally derived sediments by chemical and mineralogic examination, a distinction important for interpretations of environmental change (Rosenbaum et al. 2009).

Human activities can have strong influence on the composition of soils, lake water, and lake sediments through hydrologic or atmospheric inputs (Oldfield and Dearing 2003). Even remote lakes can be affected by anthropogenic atmospheric inputs (Baron et al. 1986; Charles and Norton 1986; Norton et al. 1990; Shotbolt et al. 2005). Effects may include increased metal and nutrient concentrations accompanied by changes in water chemistry (Norton 1986) having potentially adverse impacts on aquatic and terrestrial biota (Mills and Schindler 1986). Soils on isolated surfaces in alpine, island, or other remote settings may contain far-traveled dust with strong

potential to influence these ecosystems (Chadwick et al. 1999). The purpose of this study is to examine sediments in two sub-alpine lakes in the Uinta Mountains (Utah) for records of atmospheric dust. Eolian silt has been recognized for its possible biogeochemical roles in soils of various elevation zones in the Uinta Mountains (Bockheim et al. 2000). High concentrations of metals, including Pb, Zn, Cu, Cd, Sn, and As, in recent sediments of Mirror Lake have been attributed by Kada et al. (1994) to smelter activities along the Wasatch Front. The Uinta Mountains setting provides the opportunity to test the hypothesis that dust generated by human activities since European settlement of western North America has changed lake-sediment chemistry and to examine the potential influence of local to regional dust sources on high-elevation ecosystems.

## Study area

Two lakes in the spruce-fir zone of the Uinta Mountains, Utah (Walsh 2002) were examined for this study (Fig. 1; ESM 1). Marshall (3,042-m elevation) and Hidden (3,152-m elevation) Lakes are located respectively about 90 and 155 km east of the center of Salt Lake City, which is located along the Wasatch Front.

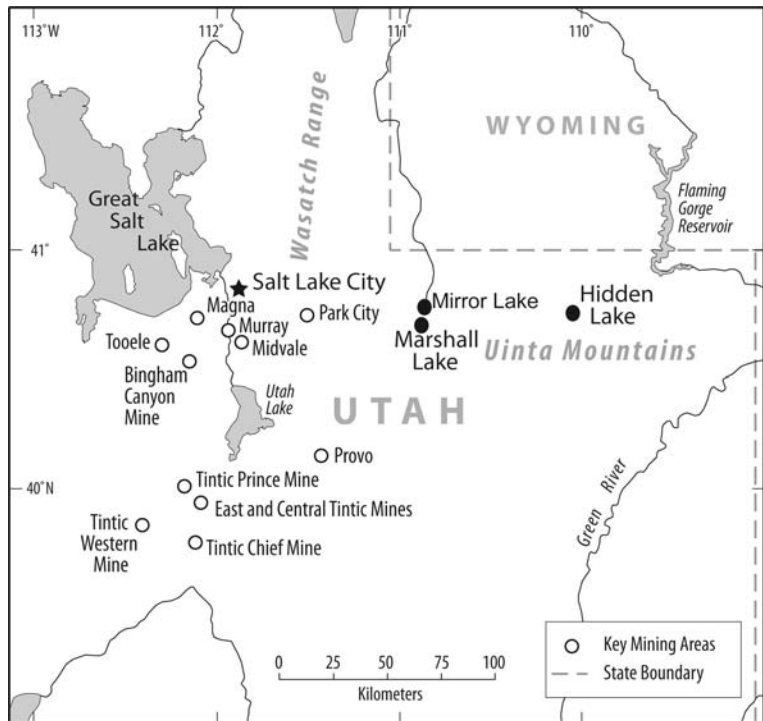
Marshall and Hidden Lakes are both nested in late Pleistocene till that covers large areas of Precambrian sedimentary rocks of the Uinta Mountain Group (UMG; Christensen and Jewell 1998; Munroe 2005). Marshall Lake has a surface area of 8 ha and maximum depth of 14.7 m. Hidden Lake covers 5 ha, with a maximum depth of 15.7 m. Rocks of the Mount Watson and Hades Pass units of the UMG (Bryant 1992) are exposed mainly in talus slopes in the Marshall Lake and Hidden Lake catchments, respectively. These units consist dominantly of quartzite and quartz-rich sandstones, with subordinate shale and arkosic sandstone (Condie et al. 2001).

## Methods

### Field methods

In 2001, a K–B gravity corer was used to take two ~50-cm-long cores from the center of each lake. Cored sediment varied among light, medium, and

**Fig. 1** Map showing locations of the Uinta Mountains, Marshall and Hidden Lakes, mining areas, and other features. The heavily urbanized swath of north-central Utah, the Wasatch Front, lies immediately west of the Wasatch Range and mountains to its south, stretching from about 30 km south of Provo to 80 km north of Salt Lake City



dark brown hues and lacked visible bedding (Walsh 2002). The cores were extruded vertically and sampled at 0.5-cm contiguous intervals. Samples of bedrock (16) and soil (7; from the top 10 cm) were also collected and analyzed for magnetic susceptibility, as described by Mordecai (2008). In addition, five samples of sediments from intermittent streams from the Hidden Lake watershed were analyzed, but such sampling near Marshall Lake was precluded by lack of stream sediment there.

### Chronology

The upper parts of the sediment cores were dated using  $^{210}\text{Pb}$  (Appleby 2001; Walsh 2002) and  $^{239+240}\text{Pu}$  techniques. The  $^{210}\text{Pb}$  analysis was done on 20 samples at 0.5-cm intervals between 0 and 9.5 cm. Samples were dried, ground to pass through a 100-mesh screen, and then analyzed by alpha spectrometry for  $^{210}\text{Po}$ , a decay product of  $^{210}\text{Pb}$ , at MyCore Scientific (Deep River, Canada). The depth-age profile was determined using the constant rate of supply (CRS) model that assumes a constant influx of unsupported, atmospheric  $^{210}\text{Pb}$ . Following methods of Ketterer et al. (2004), we analyzed 17 samples from Marshall Lake for  $^{239+240}\text{Pu}$ , which is a product

of above-ground nuclear-weapons testing that began in the early 1950s and peaked in 1963. Eleven samples represented 0.5-cm intervals from 1.0 to 6.5 cm depths, four represented 1.0-cm intervals to 10.5 cm, one came from 15.5 to 16.0 cm, and one from 20.5 to 21.0 cm. Radiocarbon dates were obtained from bulk organic carbon in sediment samples from 44.0 to 44.5 cm in the Marshall Lake core and 48.5 to 49.0 cm in the Hidden Lake core. Organic matter was separated from the sediments and converted to graphite (Jones et al. 1989) that was analyzed by accelerator-mass spectrometer methods at Lawrence Livermore National Laboratory. Ages were calculated according to the methods of Stuiver and Pollach (1977), using an assumed  $\delta^{13}\text{C}$  value of  $-25$  per mil, and are presented as uncalibrated ages.

### Magnetic properties

Magnetic and petrographic methods were combined to determine the types, amounts, and particle sizes of magnetic minerals. For magnetic property measurements, wet sediment from every other 0.5-cm-depth sample was placed in plastic cubes ( $3.2\text{ cm}^3$ ). These properties include: (a) magnetic susceptibility (MS; measured with a susceptometer with sensitivity better

than  $2 \times 10^{-9} \text{ m}^3 \text{ kg}^{-1}$ ), a measure of all magnetic material but dominantly magnetite, when present; (b) isothermal remanent magnetization (IRM; measured with an AGICO JR-5A spinner magnetometer), the first imparted in an induction of 1.2 Tesla (T) and the second in  $-0.3 \text{ T}$ ; and (c) hard IRM (HIRM =  $(\text{IRM}_{1.2\text{T}} - \text{IRM}_{0.3\text{T}}) 2^{-1}$ ), a measure of the amount of hematite. Magnetic values were normalized by dry mass. Magnetic particles were separated (Reynolds et al. 2001b) from three samples from each core, combining the 0–5, 5–10, and 25–35-cm intervals. Magnetic grains were mounted in epoxy, polished, and viewed in reflected light under oil immersion at  $400\times$  magnification.

#### Geochemistry, stable isotopes of lead, and texture

Twenty samples from each lake were analyzed for 50 major and trace elements using inductively coupled plasma (ICP) emission and mass spectrometry, following multi-acid digestion, at SGS Mineral Services (Toronto, Canada). Analyzed samples were from 1-cm increments for the top 10 cm and from 3- to 4-cm intervals for deeper sediment. Organic-matter content was determined by loss-on-ignition (LOI) and used to re-calculate elemental and magnetic values on an organic-matter-free basis. Forty elements were chosen for their possible bearing on sediment sources, biogeochemical cycling, and effects of dust on air and water quality. These elements and their detection limits are K (0.01%), Li (1 ppm), Na (0.01%), Rb (0.2 ppm), Ca (0.01%), Mg (0.01%), Sr (0.5 ppm), Ag (0.02 ppm), Cd (0.02 ppm), Co (0.1 ppm), Cr (1 ppm), Cu (0.5 ppm), Fe (0.01%), Mn (5 ppm), Mo (0.05 ppm), Nb (0.1 ppm), Ni (0.5 ppm), Sc (0.1 ppm), Ta (0.05 ppm), Ti (0.01%), V (1 ppm), W (0.1 ppm), Y (0.1 ppm), Zn (1 ppm), Zr (0.5 ppm), A (10.01%), Bi (0.04 ppm), In (0.02 ppm), Pb (0.5 ppm), Sn (0.3 ppm), As (0.2 ppm), Sb (0.5 ppm), Te (0.05 ppm), P (50 ppm), S (0.01%), Ce (0.05 ppm), La (0.1 ppm), Th (0.2 ppm), U (0.1 ppm), and Yb (0.1 ppm).

Lead isotopes ( $\text{Pb}^{204}$ ,  $\text{Pb}^{206}$ ,  $\text{Pb}^{207}$ , and  $\text{Pb}^{208}$ ) were analyzed using ICP mass spectrometry (Bower et al. 2007) to help identify sources of Pb in 22 samples from Marshall Lake sediments (1–22 cm). Samples were digested with  $\text{HNO}_3$ , HF, and boric acid, and Pb was separated with Sr-Spec resin following Bower et al. (2007).

Particle-size analysis was performed on bulk sediment after digesting organic matter using 30% hydrogen peroxide and disaggregating samples in sodium hexametaphosphate. These treatments would not have removed biogenic silica or carbonate. Particle size was determined as a volume percentage using a laser-light scattering analyzer capable of measuring particles greater than  $0.05 \mu\text{m}$ .

## Results

### Chronology

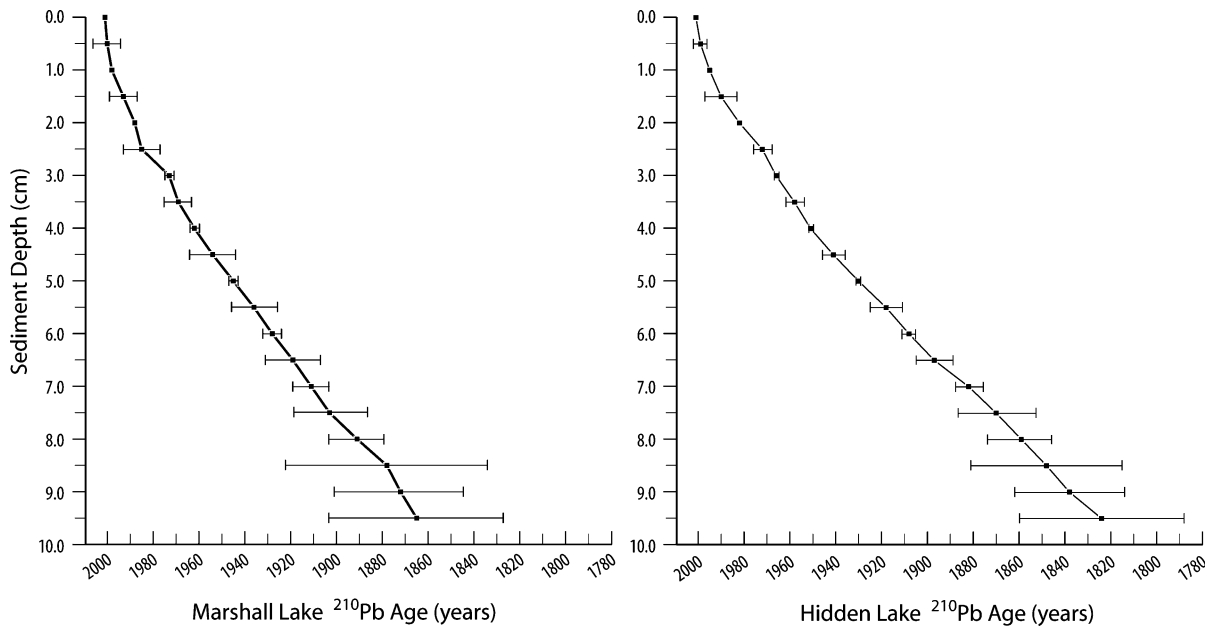
Lead-210 results indicate that sediments below 9 cm at both lakes were deposited prior to about AD 1870 (Fig. 2). The shapes of the age-depth profiles imply increases in sedimentation rates at both lakes in recent decades. For Marshall and Hidden Lakes, respectively, the average sedimentation rates are  $1.03$  and  $0.82 \text{ mm year}^{-1}$  since 1965,  $0.60$  and  $0.59 \text{ mm year}^{-1}$  for 1930–1965,  $0.58$  and  $0.46 \text{ mm year}^{-1}$  for 1900–1930, and  $0.50$  and  $0.37 \text{ mm year}^{-1}$  for 1870–1900. These estimates include the presence of organic matter and do not account for possible sediment compaction. Dating uncertainty increases toward the bottom of both cores with decay of excess  $^{210}\text{Pb}$ , and large age uncertainties overlap below depths of 8.5 cm.

The shape of the plutonium depth profile, with a maximum  $^{239+240}\text{Pu}$  activity in the 3.5–4.0-cm interval in the Marshall Lake core, indicates that the 1963-deposition peak is between 3.0 and 4.5 cm (ESM 2). The  $^{210}\text{Pb}$  age model places year 1963 at 3.81 cm. The detection of  $^{239+240}\text{Pu}$  in samples as deep as 21 cm may be related to core-surface contamination while extracting the core.

Based on  $^{14}\text{C}$  dates, the base of each core dates to more than 1500 years ago. A radiocarbon age of  $1690 \pm 40$  years was determined for a Marshall Lake sample (44.0–44.5 cm) and  $4380 \pm 60$  years for a Hidden Lake sample (48.5–49.0 cm). The causes for this large discrepancy, whether old carbon at Hidden Lake, depositional hiatus, or other factors, are unknown.

### Magnetic properties

Magnetite concentration (represented by IRM and MS values) abruptly increases at about 5.5-cm depths



**Fig. 2** Lead-210 age versus depth profiles for sediments in Marshall (*left*) and Hidden (*right*) Lakes

in cores from both lakes (Fig. 3; ESM 3). Magnetite is more abundant in sediments of Marshall Lake, in sediments both below and above the abrupt increase. On the basis of IRM values, enrichment of magnetite is nearly 6× at Marshall Lake over relatively uniform amounts in older sediment. In contrast, hematite concentrations (HIRM values) are greater in Hidden Lake sediments, and HIRM values do not exhibit systematic changes at either location (Fig. 3; ESM 3).

Petrographic observations revealed many different compositions of magnetic particles, including grains having intergrowths of magnetite, ilmenite, and other Fe–Ti oxide minerals. The internal textures and compositions of such grains indicate that they formed originally in igneous rocks and were subsequently deposited in the lakes. Most magnetic grains consist of angular magnetite and rock fragments with magnetite inclusions that range in size 4–30 μm, with most <10 μm. Sparse spherical magnetic particles, probably airborne fly ash from coal combustion (Querol et al. 1993; Goldhaber et al. 2004), are present in the top 10-cm, in which all forms of magnetite are more abundant than in deeper samples. In both cores, hematite was found as intergrowths within Fe–Ti oxide minerals and as relatively large (>100 μm) particles of specular hematite. We found no indication of post-depositional formation of

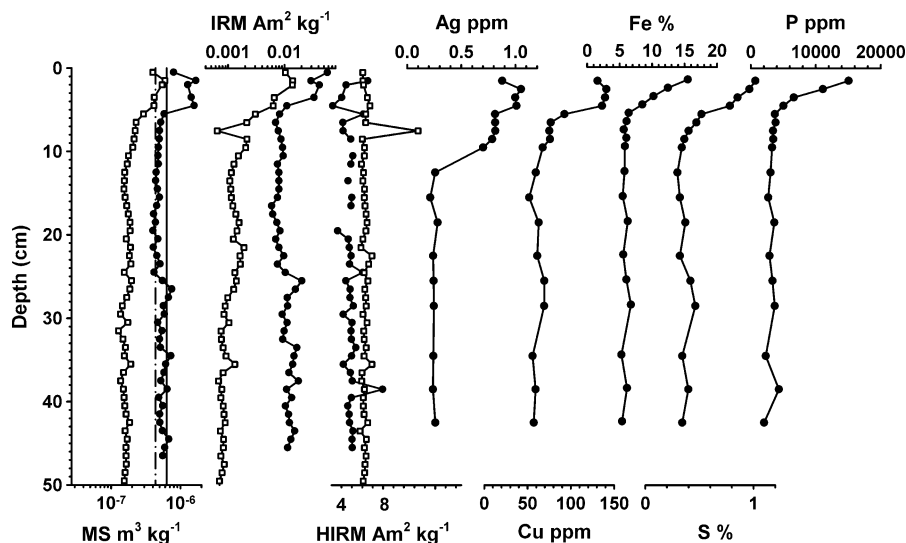
magnetic minerals, nor did we observe effects of post-depositional alteration on detrital grains (Rosenbaum et al. 1996).

Magnetic susceptibility values for lake sediments were compared with MS values of surface soil and rocks as a test for sources of magnetite (Fig. 3). Low values of bedrock MS relative to soil and lake sediment MS would strongly suggest sources of magnetite other than local bedrock (Reynolds et al. 2006). Rocks in both watersheds have low MS, ranging  $0.19\text{--}2.66 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ . In contrast, average background MS values for lake sediment below 6 cm (organic-matter-free basis) are much higher:  $5.19 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$  at Marshall Lake and  $1.81 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$  at Hidden Lake. The silt-plus-clay fractions in surface-soil samples have MS values (respective ranges of  $5.69\text{--}6.94 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$  and  $2.99\text{--}6.37 \times 10^{-7} \text{ m}^3 \text{ kg}^{-1}$ , for Marshall Lake and Hidden Lake watersheds) that are greater than the nearly uniform background MS values in the respective cored sediment (Fig. 3).

#### Geochemical and textural properties

Organic matter contents at both settings are similar, averaging 27% (standard deviation, 5%) and 31% (standard deviation, 4%) respectively in sediments at

**Fig. 3** Depth plots of magnetic susceptibility (MS), IRM (isothermal remanent magnetization), HIRM (“hard” isothermal remanent magnetization;  $\times 10^{-4}$ ) for sediments at Marshall Lake (*closed circles*) and Hidden Lake (*open squares*), and for Ag, Cu, Fe, S, and P, all on organic-matter-free basis. In the MS plot, the average MS for soil in the Marshall (Hidden) Lake watershed is denoted by the vertical solid (dashed) line. Highest MS value measured for local bedrock was  $2.6 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$



Marshall and Hidden Lakes (ESM 4–6). Organic matter increases within the top 8 cm in both cores, 23–35% at Marshall Lake and 31–43% at Hidden Lake. With organic matter removed, the lake sediments are dominantly silt or clayey silt (ESM 7). Sediment above 8 cm in Marshall Lake contains more silt (76–79%) and less clay (21–22%) than below (67–69% and 31–32%, respectively). Sediments at Hidden Lake range 73–82% in silt and 14–23% in clay, without evident trend.

Elements are divided into four groups on the basis of patterns in their organic-matter-free concentrations within the upper 25 cm (Table 1): elements that (1) increase above their background concentration (average concentration below the depths of near-surface chemical change) beginning at about 12 to 10-cm depths (Group 1); (2) increase above their background concentration beginning at or above about 6 to 5 cm, as does magnetite (Group 2); (3) decrease slightly below their background concentration at a depth shallower than about 5 cm with no well-defined trend below (Group 3); and (4) show no obvious trend (Group 4).

Group 1 elements are identical for both lakes (Table 1; ESM 4). Most Group 1 elements (Ag, Cu, Pb, As, Bi, Zn, S, Sb, Sn, and Te) also show a sharp increase at about 5 cm. Group 2 has eight elements in common for both lakes (P, Fe, W, La, Ce, Co, Mo, and Y; Table 1; S4). Group-2 behavior is also found for Yb, Sr, and Rb at Hidden Lake and for Mn at Marshall Lake. Iron (Group 2) shows additionally

small increases above 10 cm at both lakes, as does P at Marshall Lake. There are more Group-3 elements at Marshall Lake than at Hidden Lake, and Li is the only one in common. Group 4 at both sites consists of nine elements in common (Al, Ca, Sc, Ta, U, Th, Ti, Zr, and Cr). Of the elements showing upward increases in both cores, much larger increases are found in Marshall Lake than in Hidden Lake for P, Cu, Pb, Bi, S, Te, Co, Cd, Fe, Ag, Sn, In, Sb, and Mo (Fig. 4). Increases in As and La are nearly identical in magnitude for both lakes; Ni increases (by 1.5 $\times$ ) toward the top in Hidden Lake sediments but not in Marshall Lake sediment.

Background elemental concentrations in both cores are similar. Average concentrations for 31 of the 40 elements corrected for organic matter content are within 25% of baseline concentrations. The exceptions are Fe, Cu, Mo, As, and Zr that are higher in Marshall Lake sediments and Ca, Cd, Li, and Mg that are higher in Hidden Lake sediments. Magnetite is more abundant in pre-settlement sediments at Marshall Lake than in equivalent sediments at Hidden Lake.

To assess the contribution of local bedrock to the lake sediments, we used geochemical results in Tables 2 and 3 in Condie et al. (2001) for UMG quartzite, arkosic sandstone, and shale that compose bedrock for the two lakes. Similar bedrock elemental compositions in the catchments are reflected by the similar major- and minor-element concentrations (e.g. Al, K, Na, and Mg) of lake-sediment in both cores. For

**Table 1** Elemental results listed by Groups that describe patterns of variation in lake-sediment cores for Marshall and Hidden Lakes

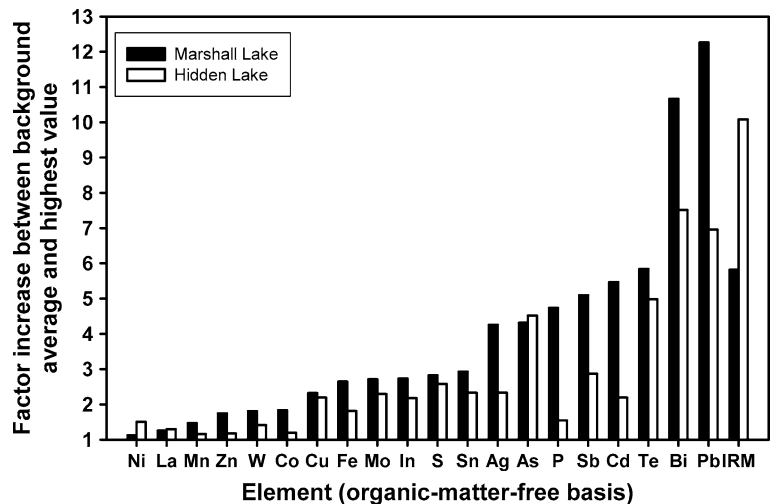
Group 1 Marshall	Group 1 Hidden	Group 2 Marshall	Group 2 Hidden	Group 3 Hidden	Group 3 Hidden	Group 4 Marshall	Group 4 Hidden
Ag <sup>a</sup>	Ag	P <sup>b</sup>	P	Li	Li	Al	Al
Cu <sup>a</sup>	Cu	Fe <sup>b</sup>	Fe <sup>b</sup>	Mg		Ca	Ca
Pb <sup>a</sup>	Pb <sup>a</sup>	W <sup>b</sup>	W	K		Sc	Sc
Zn <sup>a</sup>	Zn <sup>a</sup>	La	La	Na		Ta	Ta
As <sup>a</sup>	As <sup>a</sup>	Ce	Ce	Nb		U	U
Bi	Bi <sup>a</sup>	Co	Co	Sr		Th	Th
Cd	Cd <sup>a</sup>	Mo	Mo	Rb		Ti	Ti
In <sup>a</sup>	In <sup>a</sup>	Y	Y			Zr	Zr
Sb <sup>a</sup>	Sb <sup>a</sup>	V	V			Cr	Cr
Sn <sup>a</sup>	Sn <sup>a</sup>	Sc	Sc			Ni	Rb
Te <sup>a</sup>	Te <sup>a</sup>	Mn	Yb			Yb	Mn
S <sup>a</sup>	S <sup>a</sup>		Sr				Mg
			Ni				K
							Na
							Nb

Group 1, up-core increase in elemental abundance beginning at 12–10 cm. Group 2, up-core increase in elemental abundance beginning at or above about 6 cm. Group 3, Slight decrease in elemental abundance beginning at about 6 cm or shallower. Group 4, very little or no change in elemental abundance through the core

<sup>a</sup> Denotes element that also shows abrupt up-core increase beginning at about 5 cm

<sup>b</sup> Denotes element that also shows slight increase up-core between about 10–5 cm

**Fig. 4** Plot showing elemental increases in sediments from Marshall (solid bars) and Hidden (open bars) Lakes. The factor increase represents the difference between the average background concentration below at least 12-cm depth and the maximum value above 10 cm



example, Al concentrations average 8.9 and 10.0% in Marshall and Hidden Lakes, respectively. For the 14 elements analyzed in both bedrock and lake sediments, much higher concentrations in lake sediment at both sites are found for Ce, Y, La, Tb, Nb, Rb, Sc, V, Yb, and Ni. Additionally, Pb, Th, and U are higher at Marshall Lake, and Sr is higher at Hidden Lake.

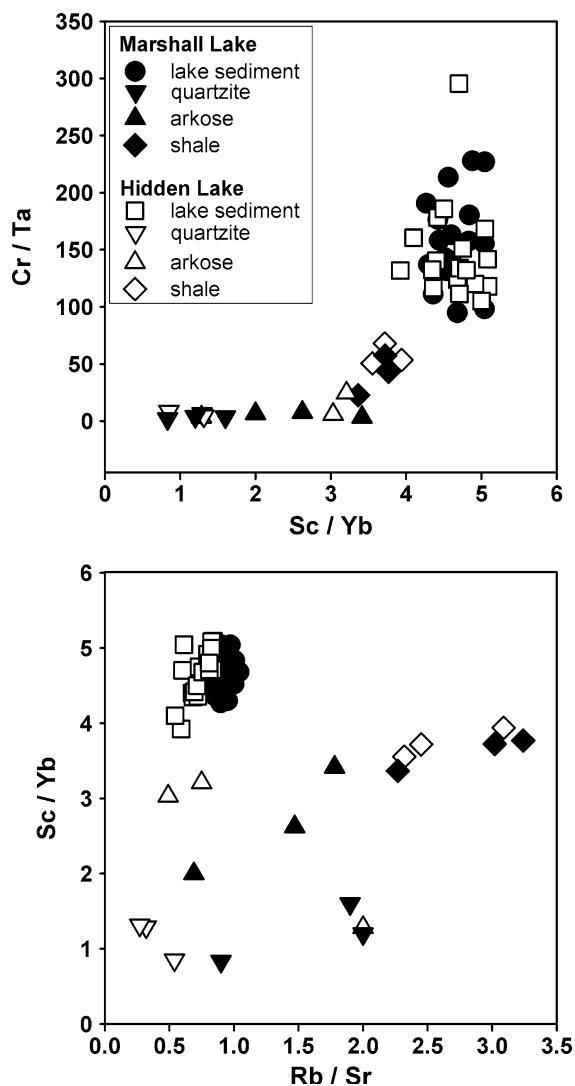
Because energies of sediment transport and deposition may influence mineralogic sorting, elemental ratios can be more informative than concentration when comparing catchment rocks and lake sediments. For example, plots of Sc/Yb against Cr/Ta and against Rb/Sr (Fig. 5) show close grouping of lake sediment samples that are segregated from UMG

bedrock, which has low amounts of Sc and Cr. The wide separation of values indicates that bedrock-derived sediment cannot account for the lake-sediment ratios. The largest separation in Rb/Sr is between lake sediments and UMG shale, suggesting that shale is a minor contributor. Similar relations are seen in a ternary plot of Sc–Th–La (ESM 8), by which a growing body of dust compositional data is represented (Muhs et al. 2008). Moreover, lake-sediment Zr/Ti values (0.036–0.077) are greatly different from bedrock values ( $Zr/Ti > 0.10$  for arkose and quartzite). A curious result, however, is

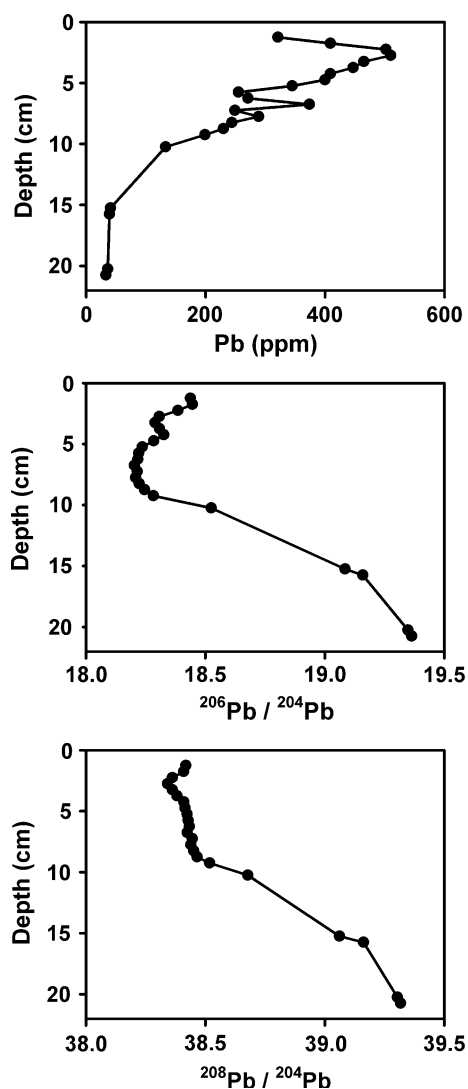
distinctly higher Zr/Ti for Marshall Lake sediments ( $Zr/Ti = 0.056–0.077$ ) than for Hidden Lake sediments ( $Zr/Ti = 0.036–0.043$ ) caused by higher Zr concentrations at Marshall Lake (ESM 4).

#### Lead isotopes

The Pb record in sediments from Marshall Lake defines four intervals (Fig. 6; ESM 4). The deepest interval (40–15 cm; predating ca. 1850) contains Pb concentrations  $< 40$  ppm (for a pre-settlement baseline of 27 ppm) and relatively radiogenic Pb



**Fig. 5** Ratio plots for sediments and rocks in the Marshall Lake and Hidden Lake watersheds. Bedrock data from Condie et al. (2001)



**Fig. 6** Plots of Pb concentration,  $^{206}\text{Pb}/^{204}\text{Pb}$ , and  $^{208}\text{Pb}/^{204}\text{Pb}$  for sediments from Marshall Lake



( $^{206}\text{Pb}/^{204}\text{Pb} \sim 19.2\text{--}19.4$  at 22–15-cm depths). In the 15–10-cm interval, Pb concentration increases sharply, with decreases in  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$ . In the 10–5-cm interval, Pb concentrations are several-fold higher than the geochemical baseline;  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  exhibit nearly constant values of  $\sim 18.2$  and  $38.4$ , respectively. The shallowest depth interval (5–0 cm) exhibits a peak Pb concentration of 510 ppm at 2.75 cm, above which Pb concentration decreases and  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  slightly increase.

## Discussion

### Evidence for dust in lake sediments

Magnetic and chemical evidence reveals a component of mineral dust in the lake sediments. The occurrence of fine-grained magnetite (most  $<10\ \mu\text{m}$ ) in lake sediments indicates the presence of far-traveled eolian mineral dust for several reasons. Firstly, local bedrock cannot be a source for magnetite in the sediments, because it is devoid of magnetite as indicated by its low magnetic susceptibility values (Fig. 3) and by paleomagnetic studies of UMG units across the Uinta Mountains (Ashby et al. 2005). Secondly, the observed magnetite in the lake sediments is similar in composition and texture to dust observed in the American west (Reynolds et al. 2001a), including that in settings in and around Bear Lake, 150 km northwest from Marshall Lake (Reynolds and Rosenbaum 2005). Dust particles of the observed size range for most magnetite particles in these lake sediments can travel hundreds of kilometers from their sources (Goudie and Middleton 2001). Finally, spherical magnetic fly ash in the shallow sediments must be an airborne component. Other origins of magnetic minerals cannot explain the observed increases in IRM and MS toward core tops. Bacteriogenic magnetite as magnetosomes (generally  $0.1\ \mu\text{m}$  and too small to be observed by our petrographic methods) would be insignificant in light of the abundance of silt-sized detrital magnetite. Moreover, petrographic observations indicate the absence of magnetic sulfide minerals, such as diagenetic greigite ( $\text{Fe}_3\text{S}_4$ ). Finally, the lack of variation in hematite concentration argues against Fe-oxide dissolution to explain magnetic property-depth profiles in the sediments.

The magnetic results suggest that sediment below  $\sim 10$  cm from Marshall Lake contains less magnetite than catchment fines in uppermost soil (Fig. 3). This difference may reflect greater amounts of dust deposition in the catchment during the past century than during prior centuries, a difference now also reflected in the elevated magnetite content at the top of the core. The higher magnetite concentrations in soil and lake sediments at Marshall Lake, compared with Hidden Lake, may be related to its closer position to dust sources in the Great Basin desert, whereby it would receive relatively more heavy-mineral dust. This possibility is supported by higher concentrations of Zr and higher Zr/Ti in Marshall Lake sediments. Similar differences have been found for mid-continent North American loess (Muhs and Bettis 2000) that have been interpreted to reflect Zr residence in relatively large particles and Ti residence in overall smaller particles capable of longer eolian transport.

Relative and absolute amounts of many trace elements in lake sediments are greatly different from those of UMG bedrock, and thereby the chemical data support magnetic interpretations about mineral-dust inputs. Large chemical contrasts have been useful for recognizing far-traveled dust in other young surficial deposits, including soils and lake sediments (Wells et al. 1987; Reheis 1990; Reheis et al. 1995; Neff et al. 2008). For the Uinta lakes, examples include the similar values of Sc/Yb, Rb/Sr, Cr/Ta and normalized Sc–Th–La in the lake sediments that are distinct from bedrock values (Fig. 5; ESM 8). These ratio values indicate compositional similarities of dust at the two sites. This observation is supported by the similar background concentrations for most elements in sediments at both lakes. The large increases in many elements toward the top of cored sediment apparently reflect recent changes in mineral dust inputs to the lake sediments related to changing dust sources, as assessed in the following section. Some degree of up-core enrichment through reduction–oxidation reactions is possible for some redox-sensitive elements but cannot account for strong enrichments in most elements.

### Timing and sources of recent atmospheric inputs

The earliest systematic elemental increases (Group 1; Table 1; Fig. 3) are for elements associated with

mined and treated ores (Ag, As, Cd, Cu, In, Pb, Bi, Sb, Sn, Te, Zn, and S). The onset of these increases occurs at about the same depths in the cores, between 12 and 10 cm, with 10 cm corresponding approximately with the middle to late 1800s in Marshall Lake. We surmise that the earliest increases for Group 1 elements at both lakes had the same causes rather than different causes and (or) the same sources at different times, and we consider that the apparent differences in timing are related to uncertainties in  $^{210}\text{Pb}$  chronologies. Mining and smelting along the western front of the Wasatch Range (Wasatch Front), beginning during the latter half of the 1800s (Blanthorn 1998), were likely causes of some of the increases in ore-related metals. Important areas included the Bingham Canyon Mine for Cu, Pb, Zn, Mo, and As; the Park City mining district for Ag, Cu, Sb, Zn, Fe, and Au-telluride; and the Tintic mining district for Ag, Pb, Cu, Zn, Cd, and Bi mainly between 1869 and 1957 (Morris 1968; James 1973, 1978; Blanthorn 1998). Smelting in other parts of the Salt Lake Valley has likely also contributed to metals in Uinta lake sediments (Raymer 1939). The relatively high concentrations of metals at more proximal Marshall Lake support the inference for sources of metal-bearing dust from the vicinity of the Wasatch.

Contemporaneous mining and treatment of ores elsewhere in western U.S. probably contributed additional metal-bearing dust. The increases in Group 1 elements show general temporal correspondence with the onset of mining production in the United States (Kelly and Matos 2008), most of it in the western part of the nation. For example, changes in Cu concentration in the lake-sediments broadly correspond with Cu production, which was dominated by the Bingham Canyon Mine (Figs. 1, 3, 7). The shift to higher As concentrations also corresponds roughly with the onset of As production, but increasing inputs from fossil-fuel combustion may have contributed to increasing As during the latter half of the twentieth while As mining waned (Fig. 7).

The onset of increases in Group 2 elements and magnetite is accompanied by additional increases in most Group 1 elements (Table 1). These simultaneously augmented inputs, apparently beginning during the late 1930s and continuing into the 1940s and 1950s, may record greater levels in human activity in the western U.S., including the introduction of aerosols related to new mining or industry.

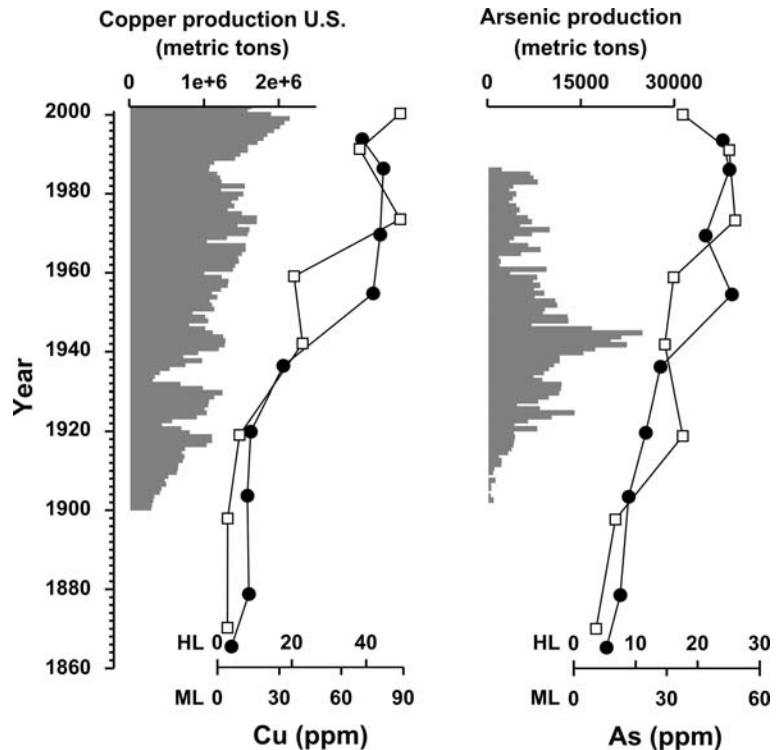
Examples of Group 2 behavior are illustrated by Mo, the mining of which began and expanded rapidly during the early 1940s, and for Y, which increases at times generally corresponding with the onset of REE mining in the U.S. in the 1950s (ESM 9).

Other sources of metals in Groups 1 and 2 probably involve the combustion of fossil fuels and perhaps refuse incineration (Nriagu and Pacyna 1988). Sediments in lakes located downwind from coal- or oil-combustion facilities commonly have increased levels of As, Sb, Cr, Zn, Hg, Mn, and Ni (Baron et al. 1986), and Sb may also be produced by fossil-fuel combustion in vehicles (Smichowski et al. 2008).

Some component of agricultural dust is suggested by the nearly coincident increases in titaniferous magnetite and P (Fig. 3), some of which may be from direct atmospheric fallout as fertilizer. The up-core concentration of P in these lake sediments has been likely influenced by redox reactions and biologic uptake (Engstrom and Wright 1984). The large difference, however, in P enrichment between sediments in the two lakes implies factors other than redox, and observed P enrichment exceeds that expected for terrestrial organic matter, on the basis of plant-P stoichiometry (Sterner and Elser 2002). The increases in P appear to correspond temporally to expansion of phosphate production and use in the American West (Fig. 8; Hein 2004; Kelly and Matos 2008) between about 1950 and 1970. Neff et al. (2008) also documented greatly increased P near the tops of alpine-lake sediment cores representing roughly the last 50 years, and they attributed such increases to expanded use of phosphate fertilizer and its atmospheric transport.

Redox reactions may also have affected the forms and distribution of Fe, Mn, S, Zn, Mo, and Cu, especially in the upper 5 cm of the cored sediment (Engstrom and Wright 1984). Several observations, however, indicate that redox is not fully responsible for these up-core increases in these elements. First, the dust signal from detrital magnetite corresponds temporally with enrichments in elements that are sensitive (P, Fe, Mn, Co, and Mo) and insensitive (the REEs, Ce, La, and Y) to redox. Second, variations for some redox-sensitive metals are different (comparing among Mo, Zn, and Cu) and appear to show strong correspondence with the history of regional mining. Moreover, sulfur concentrations do not correspond

**Fig. 7** Plots of U.S. production of copper and arsenic from mining (Kelly and Matos 2008), with age plots (from  $^{210}\text{Pb}$  models) of the respective elements in Marshall (*closed circles*) and Hidden (*open squares*) Lakes. Production of Cu is dominated by the Bingham Canyon Mine (Arrington and Hansen 1963) that started underground in 1863 and converted to an open-pit mine between 1906 and 1910



closely with organic matter content (ESM 5, 6), suggesting concentration by non-biogenic factors. The increases in S along with ore-related metals strongly imply sulfide-ore sources. Particulate sulfur and other forms of aerosolic sulfate from fossil-fuel combustion and many other North American sources have surely contributed to the lake-sediment sulfur (Malm et al. 1990).

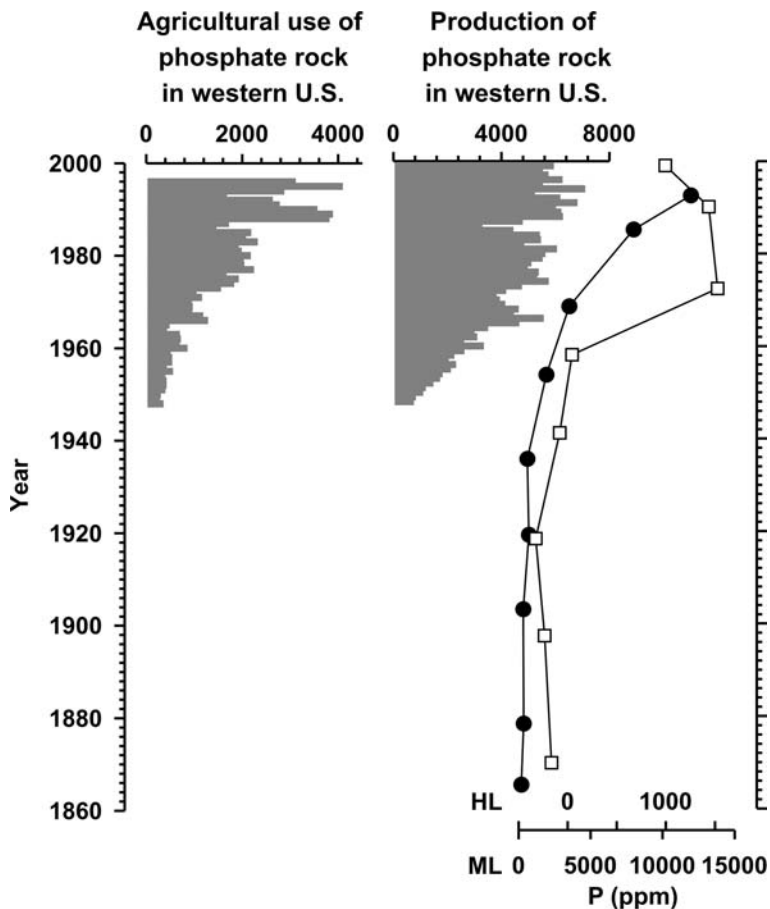
#### Anthropogenic Pb in Marshall Lake sediments—evidence from Pb isotopes

Lead isotopic compositions (Fig. 6) reveal anthropogenic sources, as documented elsewhere (Norton 1986; Van Metre and Mahler 2004). In the upper 15–10 cm of the cores, Pb is comprised of both background and anthropogenic Pb, mostly from smelting, as indicated by increased Pb concentrations and decreased  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$ . The dominant source of Pb in the 10–5-cm section is ascribed to smelting of ores from Utah because (1) maximum concentration of Pb is found in post-1945 sediments, and (2) the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio ( $18.23 \pm 0.02$ , 1 SD,  $n = 9$ ) is similar to compositions (17.6–18.6) from galena-bearing ores from Utah, such as

from the Bingham Canyon, Ophir, Park City, and Tintic districts (Stacey et al. 1968). The  $^{206}\text{Pb}/^{204}\text{Pb}$  and  $^{208}\text{Pb}/^{204}\text{Pb}$  ratios in this interval likely represent smelting emissions from Midvale, 15 km south of Salt Lake City, and Bingham Canyon. Smelters at Midvale processed ore mainly from Utah from 1871 to 1958 (U.S. Environmental Protection Agency 1995). Significant inputs from other major sources, such as the Coeur D'Alene district ( $^{206}\text{Pb}/^{204}\text{Pb} \sim 16.2$ ) or Mississippi Valley Pb ores ( $^{206}\text{Pb}/^{204}\text{Pb} > 20$ ) would produce a different Pb isotopic pattern.

Slight increases in  $^{206}\text{Pb}/^{204}\text{Pb}$  ( $18.30 \pm 0.02$ ,  $n = 5$ ) are evident in the 2.5–5.0-cm interval spanning the 1963  $^{239+240}\text{Pu}$  peak, with further increase ( $18.42 \pm 0.02$ ,  $n = 3$ ) within 1.0–2.5 cm. These changes could arise from mixing of smelted Pb with lesser amounts of gasoline-derived Pb. Increase in  $^{206}\text{Pb}/^{207}\text{Pb}$  ratios from  $1.168 \pm 0.001$  at 5–10 cm to  $1.179 \pm 0.001$  at 1.0–2.5 cm is consistent with influence (20–30%) from 1960s to 1980s U.S. gasoline (having  $^{206}\text{Pb}/^{207}\text{Pb}$  between 1.15 and 1.23; Shirahata et al. 1980) on a larger contribution (70–80%) from Utah Pb. Lead concentrations decrease in sediments deposited since about the mid-1970s probably because of cessation of smelting,

**Fig. 8** Plots of agricultural use of phosphate rock (for fertilizer) and mined tonnage of phosphate rock, both in the western U.S. (Kelly and Matos 2008), with age plots of phosphorus in Marshall (closed circles) and Hidden (open squares) Lakes



the temporary halt to mining at Bingham Canyon (1985–1987) followed by emissions controls after mining resumed, and the phase-out of leaded gasoline beginning in 1972 (Blais and Kalff 1993).

#### Other causes for changing lake-sediment composition

Evidence is lacking that soil disturbance from fires and logging in the catchments influenced lake-sediment composition, possibly through the rapid release of eolian sediment sequestered in soil. Examination of tree rings and lake-sediment charcoal (Walsh 2002) shows no evidence for historic fires in either of the catchments. Although we found no records of logging in the catchments, nearby areas were probably logged in the early 20th century. Disturbances of forest soil, however, would increase the delivery of locally derived hematite into the lakes, and this effect is not observed. Finally, chemically immobile elements (e.g.

Ti/Al) remain nearly constant through both cores and between cores, suggesting insufficient disturbance to affect preferentially the erosion, transport, and deposition of high-density minerals.

#### Conclusions

Cored sediments from two subalpine lakes in the Uinta Mountains differ in some mineralogical and chemical properties from local bedrock, mostly Precambrian quartzite having relatively uniform bedrock chemistry and mineralogy. These compositional discrepancies point to the presence of far-traveled atmospheric (eolian) dust. Striking features of the magnetic and chemical data are the abrupt and large increases in magnetite and many elements near the tops of the cores that indicate new sources of dust. The timing and composition of dust inputs strongly suggest that dust-source changes were related to the

onset and continuing period of intensive European settlement and activity in western North America that involved mining, industry, agriculture, urbanization, and transportation. Contributions to post-settlement dust by mining and smelting are revealed by lake-sediment enrichment of elements (e.g. Ag, Te, Pb, and Cd) that originated within a few hundred km of the lake sites and probably elsewhere in western United States. A record of anthropogenic Pb contamination from smelting is provided by trends in stable Pb isotopes.

The nearly simultaneous and abrupt increases in silt-sized, titanium-bearing magnetite and P may reflect wind erosion related to the expansion of intensive agriculture and use of fertilizers in the region. Increase in lake-sediment magnetite may also be associated with land disturbances by urbanization and transportation related to population growth that averaged 35% per decade from 1940 to 1980, compared with 17% per decade from 1910 to 1940, in four now-heavily populated counties (Davis, Salt Lake, Utah, and Weber) to the west of the Uinta Mountains.

The recent P additions bear on issues of ecosystem health in mountain forests and lakes. Our results also relate to ecosystem health in other ways, including potential impacts of toxic metals on aquatic organisms (Cattaneo et al. 2004). Continuing studies examine potential linkages between diatom community composition and metal concentrations in these and other lakes in the Uinta Mountains (Moser et al.).

The dust record in these lakes extends over at least the past approximately 1,700 years, on the basis of radiocarbon dating, a span that encompasses many multi-decadal droughts in western North America. Changes in dust composition are most pronounced in sediment deposited over the past approximately 140 years. These changes, which have been caused mostly by human activity far from the lakes, are much more pronounced than pre-settlement compositional changes for nearly all elements. We conclude that human activities and landscape disturbance have generated new dust sources to produce significant changes in lake-sediment composition, and, by inference, soil chemistry in the Uinta Mountains. Estimates of sedimentation rates imply increasing inputs of dust since about AD 1870. These results, combined with those of Neff et al. (2008) on lake

sediments in the San Juan Mountains (Colorado), indicate that human-induced changes in dust composition and depositional rates have been much greater than changes caused by pre-settlement conditions, including long droughts, over a large area of western North America.

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