



**CHEMICAL DATA AND LEAD ISOTOPIC COMPOSITIONS OF
GEOCHEMICAL BASELINE SAMPLES FROM STREAMBED
SEDIMENTS AND SMELTER SLAG, LEAD ISOTOPIC
COMPOSITIONS IN FLUVIAL TAILINGS, AND
DENDROCHRONOLOGY RESULTS FROM THE BOULDER
RIVER WATERSHED, JEFFERSON COUNTY, MONTANA**

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INTRODUCTION

As a part of the U.S. Geological Survey Abandoned Mine Lands Initiative (Buxton and others, 1997), metal-mining related wastes in the Boulder River study area in northern Jefferson County, Montana, have been evaluated for their environmental effects. The study area includes a 24-km segment of the Boulder River in and around Basin, Montana and three principal tributaries to the Boulder River: Basin Creek, Cataract Creek, and High Ore Creek. Mine and prospect waste dumps and mill wastes are located throughout the drainage basins of these tributaries and in the Boulder River. Mine-waste material has been transported into and down streams, where it has mixed with and become incorporated into the streambed sediments. In some localities, mine waste material was placed directly in stream channels and was transported downstream forming fluvial tailings deposits along the stream banks. Water quality and aquatic habitat have been affected by trace-element-contaminated sediment that moves from mine wastes into and down streams during snowmelt and storm runoff events within the Boulder River watershed (Buxton and others, 1997; Farag and others, 1999; Nimick and others, 1999).

Present-day trace element concentrations in the streambed sediments and fluvial tailings have been extensively studied (Aamodt, 1978; Broxton, 1980; Campbell and others, 1982; Fey and Church, 1998; Fey and others, 1999a; 1999b, 2000). However, in order to accurately evaluate the impact of mining on the stream environments, it is also necessary to evaluate the pre-mining trace-element concentrations in the streambed sediments. Three types of samples have been collected for estimation of pre-mining concentrations: 1) streambed sediment samples from the Boulder River and its tributaries located upstream from historical mining activity, 2) stream terrace deposits located both upstream and downstream of the major tributaries along the Boulder River, and 3) cores through sediment in overbank deposits, in abandoned stream channels, or beneath fluvial tailings deposits. In this report, we present geochemical data for six stream-terrace samples and twelve sediment-core samples and lead isotopic data for six terrace and thirteen core samples. Sample localities are in [table 1](#) and [figure 1](#) and [figure 2](#), and site and sample descriptions are in [table 2](#).

Geochemical data have been presented for cores through fluvial tailings on High Ore Creek (Fey and Church, 1998), on upper Basin Creek (Fey and others, (1999a), and on Jack Creek and Uncle Sam Gulch (Fey and others, (2000). Geochemical and lead isotopic data for modern streambed-sediment samples have been presented by Fey and others (1999b).

Lead isotopic determinations in bed sediments have been shown to be an effective tool for evaluating the contributions from various sources to the metals in bed sediments (Church and others, 1993, 1997). However, in order to make these calculations, the lead isotopic compositions of the contaminant sources must also be known. Consequently, we have determined the lead isotopic compositions of five streambed-sediment samples heavily contaminated with fluvial mine waste immediately downstream from large mines in the Boulder River watershed in order to determine the lead isotopic signatures of the contaminants. Summary geochemical data for the contaminants are presented here and geochemical data for the streambed-sediment samples are given by Fey and others (1999b).

Downstream from the Katie mill site and Jib tailings ([fig. 1, 1T](#)), fluvial deposits of mill tailings are present on a 10-m by 50-m bar in the Boulder River below the confluence with Basin Creek. The source of these tailings is not known, but fluvial tailings are also present immediately downstream from the Katie mill site, which is immediately upstream from the confluence with Basin Creek. Nine cores of fluvial tailings from this bar were analyzed.

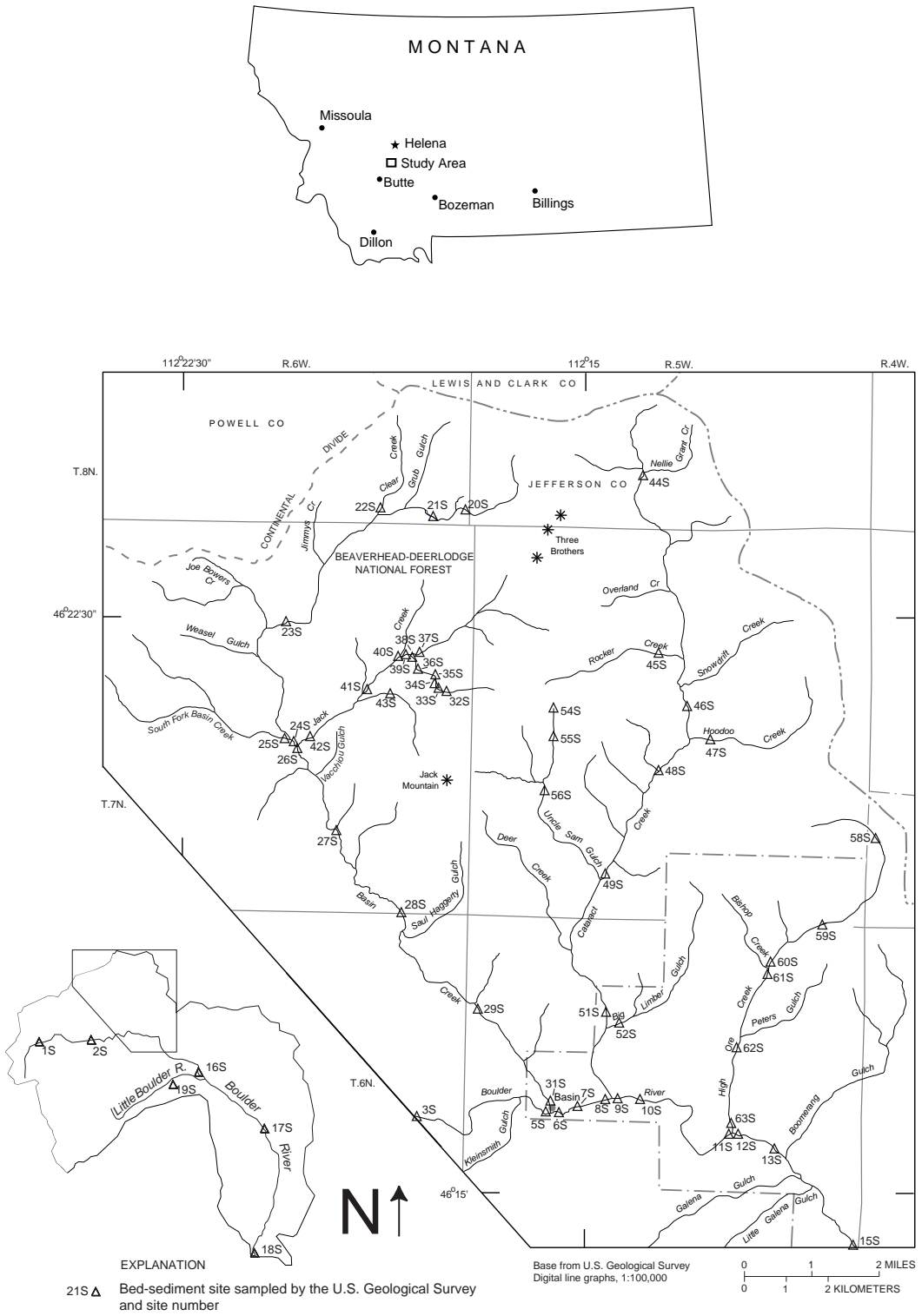


Figure 1. Index map of Montana showing Boulder River study area and sample locality map for bed-sediment sampling sites in the Boulder River Watershed.

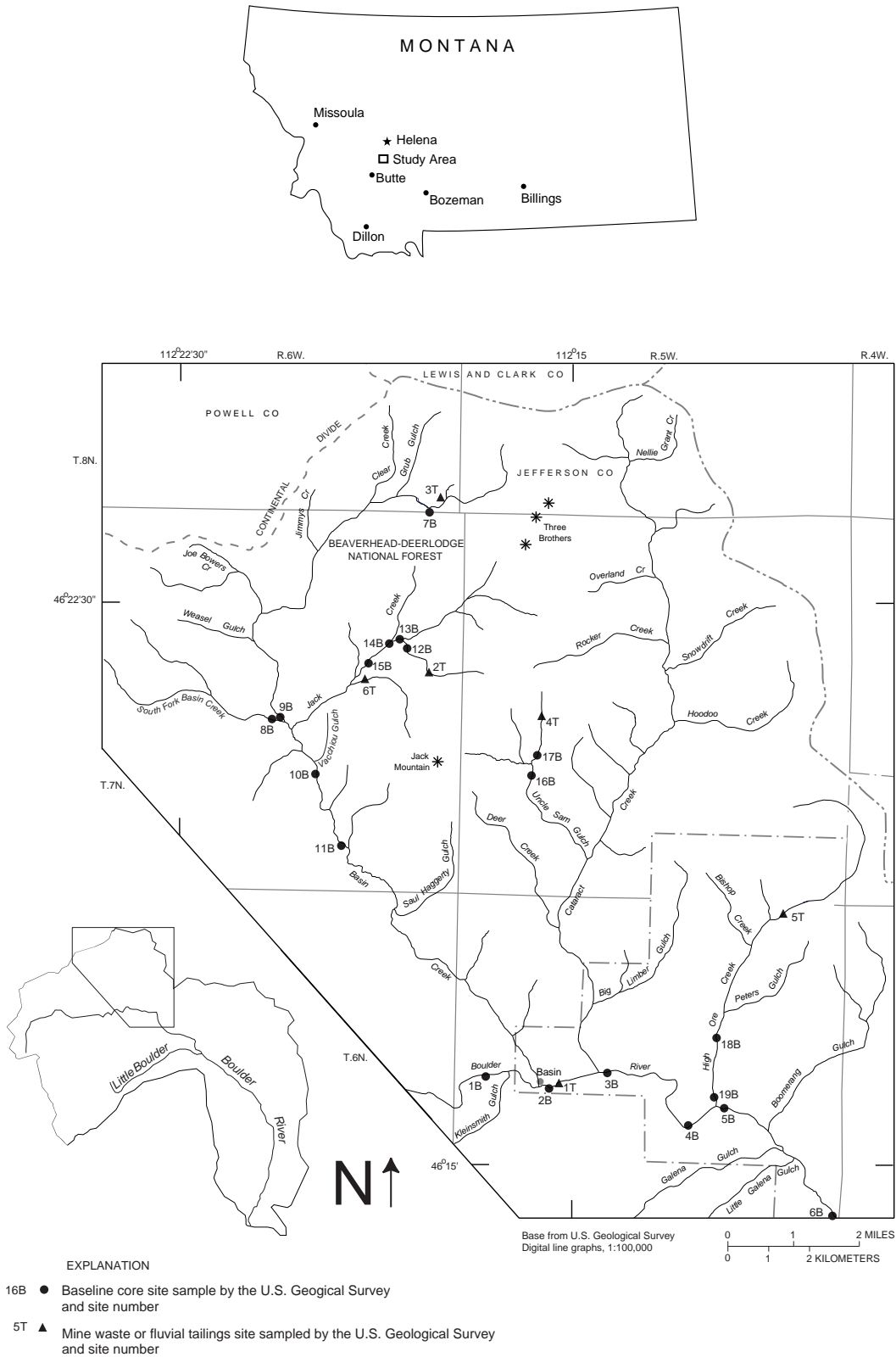


Figure 2. Index map of Montana showing Boulder River study area and sample locality map for geochemical baseline cores and mine waste and tailings sites.

Dendrochronology samples were taken at several stream terrace localities to provide age control on the stream terrace deposits. Trees growing on the surfaces of stream terraces provide a minimum age for the terrace deposits, although floods subsequent to the trees' growth could have deposited post-mining overbank deposits around the trees. Historical data were also used to provide estimates of minimum ages of cultural features and to bracket the age of events (Rossillon and Haynes, 1999).

METHODS OF STUDY

Sample collection

Streambed sediments and fluvial tailings samples

Elemental and lead isotopic data for the baseline streambed-sediment samples that have little, if any, impact by historical mining have been reported by Fey and others (1999b). Their sample localities are indicated schematically in [figure 1](#) (samples collected at nine sites; 2S, 3S, 4S, 20S, 22S, 32S, 44S, 54S, and 58S). Samples at sites 2S, 4S, and 22S were collected in October, 1996. Samples at sites 3S, 32S, 44S, and 54S were collected in July, 1997, and the remaining samples (20S, 58S) were collected in July, 1998. A second sample at site 3S was collected in July, 1998 in order to monitor yearly variations in trace-element concentrations. Sites were selected to be generally upstream from known historical mining activities. At each site, an integrated streambed-sediment sample was collected by compositing material from the active channel alluvium from 10 to 20 individual subsites within 15 m (50 ft) of the plotted sample locality. In the field, each composited sample was sieved through a 2-mm (10 mesh) stainless-steel screen, and the minus-2-mm fraction retained; the larger size fraction was discarded.

Samples of fluvial tailings collected at four sites (2T-5T, [fig. 2](#)) and the Bullion Smelter slag (6T, [fig. 2](#)) were selected for lead isotopic analyses. Three fluvial overbank-sediment samples from the Jack Creek drainage that were shown to contain tailings materials on the basis of their chemistry (35S, 37S, 40S) were also analyzed for lead isotopic compositions. These samples were collected in October 1996, in the same manner as the streambed-sediment samples, and were dry-sieved in the field to preserve the water-soluble salts contained in the samples.

Stream terraces

Samples of stream terraces were collected at six sites along the Boulder River during July 1999 (Sites 1B-6B in [fig. 2](#)). At each site, a pit was dug by hand or backhoe. Three to five subsamples were collected and given designations "a" through "e", with "a" being the highest sample stratigraphically ([table 2](#)). In general, no stratigraphy could be discerned in the gravel deposits, so each sample is really a subsample of the gravel deposit. Each subsample was sieved through a 2 mm (10 mesh) stainless-steel screen in the field. The minus-2-mm fraction for each subsample was retained separately and the larger size fraction was discarded. Each sample was treated separately, and the geochemical data evaluated to look for homogeneous distributions of ore-related trace elements throughout the terrace deposit. Samples from terrace sediment intervals having a uniform trace elemental distribution were used in this study to calculate the pre-mining geochemical baseline concentrations and determine the lead isotopic composition in pre-mining streambed sediments. This goal was achieved in streambed sediments for core 9B ([table 3](#)). Examination of the concentration data as a function of depth for

arsenic, copper, and lead clearly indicate the impact of historical mining whereas concentrations of zinc show less of an impact on streambed sediments in upper Basin Creek.

Sediment cores

Twelve sediment cores were collected in acid-cleaned sections of 2 inch (5 cm) diameter PVC pipe to determine the metal concentrations and lead isotopic compositions in streambed sediments prior to historical mining. The objective of the sampling strategy was to sample sediments deposited through the time period before the onset of historical mining activity. Thus, the concentrations of metals in the cores should decrease with depth or reach a consistent concentration providing a clear indication of the pre-mining geochemical baseline in streambed sediments. One of these samples (site 18B, [fig. 2](#)) was collected in October 1996. Four additional cores (sites 11B, 13B, 17B, and 19B) were collected in July 1997, and seven more were collected in July 1998 (Sites 7B, 8B, 9B, 10B, 12B, 14B, 15B). Subsamples from ten of the sites were selected for lead isotopic analyses on the basis of their ore-related trace element concentrations and distributions within the cores.

In addition, segments of three one-inch diameter cores collected beneath the fluvial tailings were analyzed for their lead isotopic compositions: site (3T) near the Buckeye Mine, and sites 5Ta and 5Tb on High Ore Creek below the Comet Mine. A complete discussion of the geochemistry of the cores from the Comet Mine on High Ore Creek is in Fey and Church (1998) and of the cores from the Buckeye Mine on upper Basin Creek is in Fey and others (1999a).

Fluvial tailings from site T1

Nine cores were taken on the bar in the Boulder River at site 1T ([fig. 2](#)) just downstream from the confluence with Basin Creek. Nine cores were taken at 5-m intervals along the 50 m length beginning about 7 m from the downstream end of the bar. At sites 8 and 9, the tailings had been removed by erosion and no core samples were taken. Cores 10 and 11 were taken at the upstream end of the bar. The fluvial tailings deposit at site T1 is approximately 30-40 cm thick and the top of the bar is about 1 m above the low-flow water level in the Boulder River. Core descriptions and depths of penetration into fluvial tailings are in [table 2](#). The cores were subdivided on the basis of discernable stratigraphy.

Dendrochronology

Cores of live trees were taken using a standard tree corer, the core was extracted and placed in a plastic sleeve, and most were sent to the Laboratory for Tree Ring Research, University of Arizona for analysis. Slabs of dead trees were also sent for analysis in hopes that the time of death could be determined to date the tree stump. The ages of the trees provides an estimate of the minimum age of the stream terrace.

Sample Preparation

Streambed-sediment and stream-terrace samples were dried at ambient room temperature (25°C) and sieved to minus-80-mesh (<0.18mm) prior to laboratory analyses.

Core samples were subdivided in the laboratory on the basis of mineralogy, organic content, and apparent oxidation zones. The depth assigned to each subsample is defined as the mid point for

that subsample after taking compaction into account (table 2). Individual cores were generally divided into two to ten subsamples, and ground in random order to minus-100-mesh in a vertical pulverizer.

Sample Analyses

Total digestion

The streambed-sediment, stream-terrace, and core samples were digested with a mixed-acid solution consisting of HCl, HNO₃, HClO₄, and HF. This procedure is effective in dissolving most minerals, including silicates, oxides and sulfides; resistant or refractory minerals such as zircon, chromite, and some tin oxides are only partially dissolved. Previous investigations using a variety of materials support the completeness of the digestion (Church and others, 1987; Wilson and others, 1994).

Results are reported for 34 elements analyzed by ICP-AES (inductively coupled plasma-atomic emission spectroscopy; Crock and others, 1983; Briggs, 1996). Limits of determination for the total digestion method as well as a statistical summary of mean values, standard deviations, and median values for four National Institute of Standards and Technology (NIST) standard reference materials (SRM-2704, SRM-2709, SRM-2710, and SRM-2711) are given by Fey and others (1999b). Comparisons with certified values for these standards (NIST, 1993a; 1993b; 1993c; and 1993d) are also given by Fey and others (1999b). Both analytical precision and accuracy are well within acceptable ranges.

Warm 2M HCl-1 percent H₂O₂ leach extraction

The use of a partial-digestion extraction enables one to determine concentrations of trace elements bound within different mineral phases, whereas a total-digestion extraction releases all trace elements in a sample (Chao, 1984). All sample intervals analyzed for lead isotopic compositions were subjected to a partial-digestion extraction consisting of warm (50⁰ C) 2M HCl-1 percent H₂O₂ for three hours with continuous agitation. The leachates were subsequently analyzed by ICP-AES for 32 elements. This partial extraction was designed to release trace elements associated with hydrous amorphous iron- and manganese-oxide mineral coatings and colloidal particles (Appendix III of Church and others, 1993; Church and others, 1997). Mineral coatings such as those observed in the study area can contain a significant percentage of the trace elements in a sample (Church and others, 1993, 1997).

Because we have shown that the vast majority of the contaminant lead exists within the amorphous iron- and manganese-oxide mineral coatings and colloidal particles (for example, Church and others, 1993, 1997), only the leachates were analyzed for lead isotopic compositions. This procedure presents a clearer picture of the distribution of contaminant lead and greatly reduces dilution effects from lead bound within the major mineral phases such as potassium-feldspar.

Ion exchange and mass spectrometry for lead isotope analyses

The lead separation procedure used on the 2M HCl-1 percent H₂O₂ extraction leachates to obtain the lead isotopic data is similar to those reported by Tatsumoto and others (1976) and by Unruh and others (1979). ICP-AES analyses of the selected samples provided an in-solution concentration for lead (Church and others, 1993). This concentration value is then used to calculate the volume of leachate needed to contain approximately 0.5 µg of lead; this aliquot is then evaporated to dryness in a

teflon beaker. Then 0.5 to 1.0 mL of 1.0 N hydrobromic acid (HBr) is added to the sample and warmed gently for 5 to 10 minutes. The sample is allowed to cool, is centrifuged, and the supernatant loaded onto an anion-exchange column (0.8-1.0 mL resin volume) using Dowex AG1-X8 anion-exchange resin. The column is washed with 1.2 N HBr and water, and then the lead is eluted with either 8N hydrochloric acid (HCl) or 0.5-1.0 N nitric acid (HNO₃). The sample is again evaporated to dryness and then loaded onto a second anion-exchange column with a resin volume of 0.1-0.2 mL. The column is washed with 1.2 N HBr and water, and the lead is eluted with 0.5 N nitric acid. Two or three drops of dilute (0.25-0.5 percent) phosphoric acid (H₃PO₄) are added to the sample, and it is then evaporated again to dryness.

Approximately one-half of the sample is taken up in approximately 10 µL of dilute colloidal silica gel, loaded onto a rhenium-ribbon filament, and evaporated to dryness. The filament is then loaded into a solid-source thermal ionization mass spectrometer and heated to 1150-1350°C for data acquisition. Most analyses were made using a VG Sector 54, 7-collector mass spectrometer run in “static” mode. A few samples were run on a Micro Mass 54R, single-collector mass spectrometer. No systematic biases have been observed between the two mass spectrometers (Taylor and others, 1999). Analyses of NIST SRM 981 were used to monitor mass fractionation during mass spectrometry (Cantanzaro and others, 1968; Todt and others, 1993). Methods of calculation of the analytical uncertainty are presented in the Appendix. The lead isotopic data for replicate analyses of SRM-981 are in [table A1](#).

As demonstrated by Fey and others (1999b), data from analyses of the leachates are inherently less reproducible than those from total digestion analyses. As a test of the leaching procedure for lead isotopic analyses, leachates of four NIST standards SRM 2704, 2709, 2710, and 2711 were analyzed for lead isotopic ratios. The lead isotopic analyses of replicate solutions of the four NIST standards are in [Table A2](#).

ANALYTICAL RESULTS

Geochemical Baseline Samples

Geochemical data for total-digestion analyses of the baseline stream-terrace and core samples are given in [table 3](#). Toxic ore-related trace element concentrations (for example, Pb, As, Cu) are generally similar to those found in streambed sediments upstream from known mineralization (Pb, near 60 ppm; As, about 45 ppm; Cu, about 50 ppm; Fey and others, 1999b). However, elevated concentrations of these three ore-related trace elements are found in pre-mining streambed sediments in Uncle Sam Gulch (site 17B), above the Bullion Mine (site 14B), and in High Ore Creek (site 19B). Elevated copper concentrations are found at several other sites ([table 3](#)). Zinc concentrations are quite variable throughout the study area as evidenced by the wide range in zinc concentrations among baseline streambed-sediment samples (Zn, ranges from 40 to 600 ppm; Fey and others, 1999b).

Lead isotopic data (²⁰⁶Pb/²⁰⁴Pb; [table 4](#)) show an inverse relationship with lead concentrations as is observed throughout the study area (Fey and others, 1999b). The highest ²⁰⁶Pb/²⁰⁴Pb values (approximately 18.1) found among the baseline samples are similar to those found in the streambed sediments from sites upstream from known historical mining activities. A major exception to the overall trend is the stream terrace sample collected at the confluence of Basin Creek with the Boulder River (site 2B). The ²⁰⁶Pb/²⁰⁴Pb of this sample is the lowest in the entire study area, and although it shows

low lead and arsenic concentrations (26 ppm and 35 ppm, respectively; [table 3](#)), its copper value is the highest of any of the baseline samples.

Fluvial tailings deposits from site T1

Geochemical data for core-samples of the fluvial tailings collected from site T1, the bar in the Boulder River below Basin Creek, are in [table 5](#). Ore-related trace element concentrations are generally more uniform in this fluvial tailings deposit than in those from the other mine-waste sites within the study area.

Contaminant Source samples

Lead isotopic compositions in samples of fluvial tailings and in streambed sediments collected from just below major mines are shown in [table 6](#). The overall inverse correlation between $^{206}\text{Pb}/^{204}\text{Pb}$ and lead concentrations observed among other samples from the district ([table 4](#); Fey and others, 1999b) is also observed among the samples in [table 6](#). The most contaminated samples from the Basin Creek and Cataract Creek drainages (Pb concentrations greater than 1,000 ppm) show a very narrow range of $^{206}\text{Pb}/^{204}\text{Pb}$, ranging from 17.90 to 17.93. The sample collected below the Comet Mine on High Ore Creek (site 59S, [fig. 1](#)) has $^{206}\text{Pb}/^{204}\text{Pb}$ of 18.07, a value found consistently in the High Ore Creek drainage below the Comet Mine (Fey and others, 1999b). The lead isotopic results indicate that the mineralization at the Comet Mine is from a different source and/or of a different age than the other mines in the area.

Geochemical data from the smelter slag sample from the Bullion Smelter are in [table 7](#). Geochemical data from the mine wastes collected at sites 1T-5T are summarized for the ore-related trace elements arsenic, cadmium, copper, lead, silver, and zinc in [table 8](#). Statistical summaries and ranges of compositions are given for all mine waste samples above the concentration ranges given in [table 8](#). Since the geochemical data for these elements from these sites are highly skewed distributions, the best estimate of the composition of the deposit would be represented by the median or the geometric mean concentrations. Complete published geochemical data are in the references given for each data set in [table 8](#).

Dendrochronology

The age data from tree samples collected at selected stream terrace sites are reported in [table 9](#). Unfortunately, no local dendrochronological record could be constructed from the ring structure of living trees that would allow dating of the older dead trees. The tree ring structure in living trees is complacent, that is the variation in the width of the tree rings was consistent from year to year and provided no markers in the tree ring structure that would allow ages to be assigned (written commun., Dec., 1999, Jeff Dean, Laboratory of Tree-Ring Research, Univ. of Arizona, Tucson, Ariz.). Ages from living trees therefore provide minimum ages of the terraces. These data, along with relevant historical data (Rossillon and Haynes, 1999), provide minimum ages for some of the stream terrace and core samples collected for determination of the pre-mining geochemical baseline.

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Appendix

The largest contributor to the analytical uncertainty of the lead isotopic analyses of common lead is the mass fractionation induced during mass spectrometry (Ludwig, 1980). The effects of the mass fractionation are monitored by lead isotopic analyses of NIST standard SRM-981 (Cantanzaro and others, 1968; Todt and others, 1993). Results of replicate analyses of the SRM lead standards are shown in Table A1. A mass-fractionation factor, δ , is calculated from the standard runs from each of the three isotopic ratios $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$, and $^{208}\text{Pb}/^{206}\text{Pb}$ in the following manner (Ludwig, 1980):

$$\begin{aligned}\delta &= \{(^{206}\text{Pb}/^{204}\text{Pb}_t / ^{206}\text{Pb}/^{204}\text{Pb}_m) - 1\} / 2 \\ \delta &= (^{207}\text{Pb}/^{206}\text{Pb}_t / ^{207}\text{Pb}/^{206}\text{Pb}_m) - 1 \\ \delta &= \{(^{208}\text{Pb}/^{206}\text{Pb}_t / ^{208}\text{Pb}/^{206}\text{Pb}_m) - 1\} / 2\end{aligned}$$

where the subscripts “t” and “m” refer to the true (certified) and measured values, respectively. The average of the three values is used to correct sample data for mass fractionation. Corrections to the raw sample data are applied using a linear fractionation correction in the following manner (Ludwig, 1980):

$$\begin{aligned}^{206}\text{Pb}/^{204}\text{Pb}_c &= ^{206}\text{Pb}/^{204}\text{Pb}_m * (1 + 2 * \delta) \\ ^{207}\text{Pb}/^{206}\text{Pb}_c &= ^{207}\text{Pb}/^{206}\text{Pb}_m * (1 + \delta) \\ ^{208}\text{Pb}/^{206}\text{Pb}_c &= ^{208}\text{Pb}/^{206}\text{Pb}_m * (1 + 2 * \delta)\end{aligned}$$

where the subscripts “c” and “m” denote corrected and raw measured data, respectively. Many of the standards were run at different temperatures to assess the effects of running temperature on mass fractionation. Standards run at higher temperatures (>1250°C) show a mean δ -value of 0.12 percent/amu (atomic mass unit) whereas standards run at lower temperatures indicate a larger correction of 0.14 percent/amu is required.

An uncertainty in the mass fractionation correction of ± 0.03 percent/amu is applied to the corrected sample data and represents the 2-sigma external variation of the entire data set. Overall uncertainties in the lead isotopic data reported in table 4 and 6 are calculated in the manner given by Ludwig (1980) and represent the uncertainties at the 95% confidence interval (approximately $2\sigma_m$). Because the analytical blank does not significantly affect the measured isotopic compositions, the analytical uncertainty in an individual corrected ratio, R_c , is calculated in the following manner (Ludwig, 1980):

$$\Sigma R_c^2 = \Sigma R_m^2 + (\Delta m * \sigma \delta)^2$$

where $100 * \Sigma$ is the percent uncertainty, ΣR_m is the uncertainty in the measured ratio from mass spectrometry, Δm is the nominal mass difference between the isotopes in the numerator and denominator (i.e. for $^{206}\text{Pb}/^{204}\text{Pb}$, $\Delta m = 2$) and $\sigma \delta = 0.0003$ (0.03 percent/100 from above).

As a test of the reproducibility of the leaching procedure, separate leachates of 4 NIST standards have been analyzed. Results are shown in table A2. Uncertainties have been calculated based on the procedure outlined above. The data are reproducible to within a total range of 0.2 percent in $^{206}\text{Pb}/^{204}\text{Pb}$ for each standard. Also shown are the lead isotopic data for leachates of three of the samples reported by Church and others (1997).

Table A1. Lead isotopic compositions in replicate analyses of NIST Standard SRM-981

[Uncertainties are in percent and represent 2 sigma-mean for the individual runs and 2 sigma for the mean values of each group]

Run No.	T (° C)	$\frac{^{206}\text{Pb}}{^{204}\text{Pb}}$	Uncertainty 2s mean (percent)	$\frac{^{207}\text{Pb}}{^{206}\text{Pb}}$	Uncertainty 2s mean (percent)	$\frac{^{208}\text{Pb}}{^{206}\text{Pb}}$	Uncertainty 2s mean (percent)
Certified value ¹		16.9322	0.0047	0.914561	0.0044	2.16662	0.0060
VG Sector 54 Mass Spectrometer							
981-1	1196	16.8811	0.0117	0.913193	0.0030	2.16088	0.0030
981-2	1160	16.8871	0.0099	0.913319	0.0036	2.16088	0.0030
981-2	1236	16.8918	0.0087	0.913358	0.0027	2.16129	0.0039
981-4	1203	16.8950	0.0540	0.913293	0.0087	2.16105	0.0075
981-5	1185	16.8806	0.0184	0.913163	0.0054	2.15990	0.0036
981-6	1191	16.8837	0.0099	0.913184	0.0030	2.16029	0.0030
981-7	1194	16.8852	0.0093	0.913227	0.0033	2.16036	0.0033
981-7	1237	16.8883	0.0123	0.913303	0.0033	2.16094	0.0033
981-9	1191	16.8852	0.0150	0.913168	0.0045	2.16020	0.0039
981-9	1235	16.8850	0.0120	0.913200	0.0030	2.16040	0.0036
981-10	1195	16.8854	0.0100	0.913262	0.0036	2.16041	0.0048
981-11	1203	16.8836	0.0150	0.913190	0.0039	2.15991	0.0036
981-14	1189	16.8899	0.0054	0.913313	0.0018	2.16110	0.0039
981-14	1230	16.9070	0.0060	0.913615	0.0027	2.16334	0.0027
981-15	1186	16.8844	0.0120	0.913168	0.0045	2.16012	0.0039
981-16	1200	16.8840	0.0190	0.913079	0.0036	2.16006	0.0036
981-17	1201	16.8943	0.0230	0.913288	0.0045	2.16109	0.0048
981-17	1196	16.8870	0.0210	0.913331	0.0066	2.16076	0.0114
981-18	1203	16.8821	0.0132	0.913175	0.0036	2.16031	0.0030
981-19	1176	16.8905	0.0072	0.913243	0.0021	2.16082	0.0033
981-19	1219	16.8956	0.0084	0.913376	0.0024	2.16159	0.0039
981-20	1196	16.8886	0.0150	0.913188	0.0027	2.16074	0.0054
981-22	1204	16.8950	0.0130	0.913284	0.0039	2.16141	0.0063
981-23	1203	16.8847	0.0090	0.913188	0.0039	2.16034	0.0048
981-24	1193	16.8823	0.0120	0.913137	0.0027	2.16013	0.0033
981-25	1188	16.8870	0.0220	0.913291	0.0060	2.16082	0.0042
981-26	1188	16.8943	0.0240	0.913271	0.0051	2.16110	0.0033
981-28	1193	16.8957	0.0150	0.913357	0.0036	2.16166	0.0036
981 99-1	1201	16.8885	0.0150	0.913261	0.0042	2.16082	0.0048
981 99-2	1193	16.8903	0.0096	0.913280	0.0027	2.16091	0.0027
981 99-3	1197	16.8910	0.0150	0.913323	0.0039	2.16118	0.0036
981 99-4	1199	16.8846	0.0150	0.913221	0.0027	2.16056	0.0033
981 99-5	1194	16.8889	0.0300	0.913156	0.0051	2.16031	0.0045
981 99-6	1189	16.8848	0.0168	0.913175	0.0036	2.16036	0.0039
981 99-7	1223	16.8878	0.0190	0.913237	0.0036	2.16071	0.0033
981 99-9	1189	16.8848	0.0168	0.913175	0.0036	2.16036	0.0039
981 99-10	1175	16.8860	0.0240	0.913218	0.0060	2.16023	0.0051
981 99-11	1200	16.8855	0.0190	0.913141	0.0039	2.16048	0.0042
981 99-12	1194	16.8856	0.0110	0.913192	0.0039	2.16031	0.0030
981 99-13	1189	16.8818	0.0120	0.913137	0.0039	2.16015	0.0042
981 99-14	1195	16.8808	0.0330	0.913092	0.0630	2.16017	0.0057
981 99-15	1192	16.8872	0.0150	0.913205	0.0039	2.16063	0.0039
981 99-16	1188	16.8881	0.0102	0.913236	0.0033	2.16092	0.0048
981 99-17	1197	16.8901	0.0246	0.913382	0.0045	2.16122	0.0048
981 99-18	1200	16.8926	0.0220	0.913290	0.0039	2.16109	0.0045
Mean ²		16.8873	0.0460	0.913236	0.0160	2.16066	0.0410

Table A1. Lead isotopic compositions in replicate analyses of NIST Standard SRM-981—continued

Run No.	T (° C)	$\frac{^{206}\text{Pb}}{^{204}\text{Pb}}$	Uncertainty 2s mean (percent)	$\frac{^{207}\text{Pb}}{^{206}\text{Pb}}$	Uncertainty 2s mean (percent)	$\frac{^{208}\text{Pb}}{^{206}\text{Pb}}$	Uncertainty 2s mean (percent)
981-1	1272	16.8876	0.0093	0.913253	0.0033	2.16092	0.0063
981-3	1252	16.8835	0.0087	0.913136	0.0030	2.16014	0.0042
981-3	1288	16.8900	0.0084	0.913271	0.0021	2.16102	0.0054
981-4	1316	16.8960	0.0175	0.913488	0.0057	2.16225	0.0036
981-5	1319	16.8838	0.0075	0.913174	0.0027	2.16032	0.0057
981-8	1260	16.8900	0.0084	0.913296	0.0030	2.16096	0.0057
981-10	1271	16.8988	0.0069	0.913495	0.0024	2.16210	0.0063
981-11	1271	16.8934	0.0150	0.913343	0.0048	2.16117	0.0120
981-15	1290	16.8903	0.0100	0.913265	0.0039	2.16075	0.0054
981-16	1250	16.9000	0.0078	0.913453	0.0027	2.16219	0.0051
981-17	1299	16.8936	0.0130	0.913376	0.0036	2.16136	0.0069
981-18	1288	16.8887	0.0081	0.913323	0.0030	2.16124	0.0054
981-20	1310	16.9045	0.0084	0.913578	0.0039	2.16309	0.0045
981-21	1292	16.8868	0.0300	0.913372	0.0078	2.16148	0.0118
981-22	1301	16.9021	0.0099	0.913365	0.0033	2.16210	0.0066
981-24	1288	16.8933	0.0120	0.913320	0.0036	2.16141	0.0090
981-26	1301	16.8910	0.0081	0.913213	0.0036	2.16098	0.0036
981-27	1279	16.8824	0.0190	0.913069	0.0036	2.16018	0.0036
981 99-1	1318	16.9031	0.0180	0.913515	0.0057	2.16269	0.0150
981 99-2	1299	16.8995	0.0057	0.913485	0.0024	2.16222	0.0039
981 99-3	1286	16.8986	0.0072	0.913463	0.0021	2.16214	0.0042
981 99-5	1288	16.8829	0.0150	0.913153	0.0033	2.16017	0.0039
981 99-6	1289	16.8950	0.0090	0.913382	0.0027	2.16158	0.0042
981 99-7	1302	16.8913	0.0110	0.913325	0.0024	2.16134	0.0045
981 99-8	1292	16.9085	0.0420	0.913741	0.0075	2.16411	0.0052
981 99-9	1289	16.8950	0.0090	0.913382	0.0027	2.16158	0.0042
981 99-10	1313	16.8860	0.0075	0.913244	0.0033	2.16049	0.0048
981 99-11	1302	16.9011	0.0420	0.913406	0.0072	2.16236	0.0069
981 99-13	1292	16.8938	0.0090	0.913345	0.0033	2.16160	0.0066
981 99-17	1301	16.9020	0.0051	0.913574	0.0021	2.16277	0.0021
981 99-18	1301	16.8989	0.0099	0.913442	0.0030	2.16208	0.0036
Mean ³		16.8938	0.0690	0.913360	0.0230	2.16149	0.0590
Micro Mass 54R Mass spectrometer							
981-54R-1	1245	16.8991	0.0190	0.913400	0.0120	2.16084	0.0160
981-54R-1	1310	16.9038	0.0420	0.913643	0.0120	2.16249	0.0370
981-54R-2	1235	16.8834	0.0240	0.913313	0.0090	2.16071	0.0060
981-54R-2	1335	16.8927	0.0130	0.913408	0.0060	2.16113	0.0910
Mean $\pm 2\sigma$		16.8950	0.0980	0.913440	0.0260	2.16116	0.0410

¹Certified value from Todt and others (1993)²Mean value for mass spectrometer runs at low temperature $\pm 2\sigma$ (percent); average temperature was $1198 \pm 15^\circ\text{C}$ ³Mean value for mass spectrometer runs at high temperature $\pm 2\sigma$ (percent); average temperature $1291 \pm 18^\circ\text{C}$

Table A2. Lead isotopic compositions in NIST standards SRM 2704, SRM 2709, SRM 2710, and SRM 2711

[Data are corrected for mass fractionation; uncertainties are absolute and are calculated in the manner of Ludwig, (1980)]

Sample ¹	²⁰⁶ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁴ Pb	²⁰⁸ Pb/ ²⁰⁴ Pb	²⁰⁷ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb/ ²⁰⁶ Pb
SRM-2704-1	18.777 ± 0.011	15.634 ± 0.014	38.433 ± 0.046	0.83172 ± 0.00025	2.0469 ± 0.0012
SRM-2704-2	18.819 ± 0.011	15.665 ± 0.014	38.544 ± 0.046	0.83148 ± 0.00025	2.0482 ± 0.0012
SRM-2704-3	18.799 ± 0.012	15.641 ± 0.014	38.466 ± 0.046	0.83112 ± 0.00025	2.0462 ± 0.0012
SRM-2704-4	18.788 ± 0.012	15.631 ± 0.016	38.438 ± 0.047	0.83103 ± 0.00041	2.0458 ± 0.0012
SRM-2709-1	19.087 ± 0.013	15.674 ± 0.015	38.964 ± 0.048	0.82028 ± 0.00026	2.0413 ± 0.0012
SRM-2709-2	19.077 ± 0.012	15.668 ± 0.014	38.939 ± 0.047	0.82042 ± 0.00025	2.0412 ± 0.0012
SRM-2709-3	19.060 ± 0.012	15.652 ± 0.014	38.862 ± 0.047	0.82029 ± 0.00025	2.0390 ± 0.0012
SRM-2709-4a	19.059 ± 0.012	15.652 ± 0.015	38.879 ± 0.048	0.82049 ± 0.00026	2.0399 ± 0.0013
SRM-2709-4b	19.050 ± 0.013	15.646 ± 0.016	38.864 ± 0.049	0.82057 ± 0.00029	2.0401 ± 0.0012
Church and others (1997)	19.065	15.646	38.864		
SRM-2710-1	17.818 ± 0.011	15.533 ± 0.014	38.126 ± 0.046	0.87095 ± 0.00026	2.1397 ± 0.0013
SRM-2710-2	17.806 ± 0.011	15.518 ± 0.014	38.078 ± 0.046	0.87056 ± 0.00026	2.1385 ± 0.0013
SRM-2710-3	17.800 ± 0.011	15.509 ± 0.014	38.047 ± 0.046	0.87053 ± 0.00026	2.1375 ± 0.0013
Church and others (1997)	17.819	15.537	38.141		
SRM-2711-1	17.102 ± 0.010	15.437 ± 0.014	36.947 ± 0.044	0.90165 ± 0.00027	2.1604 ± 0.0013
SRM-2711-2	17.105 ± 0.010	15.438 ± 0.014	36.956 ± 0.045	0.90176 ± 0.00027	2.1606 ± 0.0013
SRM-2711-3	17.118 ± 0.010	15.460 ± 0.014	37.021 ± 0.045	0.90233 ± 0.00027	2.1628 ± 0.0013
Church and others (1997)	17.086	15.419	36.888		
Church and others (1997)	17.097	15.430	36.936		

¹Subsample numbers 1-4 denote separate leaches. Samples SRM-2709- 4a and -4b are separate chemical separations of the same leach.