METHODS

Soil Sampling

Upland soil samples were collected at fourteen locations in the watershed in order to obtain vertical profiles of radionuclide activity (⁷Be, ²¹⁰Pb, and ¹³⁷Cs), and to characterize the total inventory of radionuclides (Figure 1). Sample sites were typically located near suspended sediment sampling locations. Most soils were collected in June 2000 although some were collected in June 1999.

Soil profiles were collected from undisturbed, relatively flat sites that appeared to be neither the site of erosion nor deposition. The upper part of the soil column was sampled with a specially-designed high resolution slot corer. This device allows sampling at 0.4 cm intervals to a depth of 35 cm with minimal compaction and smearing of the soil profile. The device allows collection of a sample with a surface area of 233 cm² which provides sufficient mass of soil for radionuclide and other geochemical measurements given the fine vertical resolution. We sampled at 0.4 cm intervals from the surface to depths of 2.0-2.8 cm; this sampling range should contain all the ⁷Be and provide the details of near surface activities of ¹³⁷Cs, ²¹⁰Pb and other geochemical constituents. Below this, we sampled at 0.8 cm intervals to approximately 7.6 cm. Additional samples deeper in the profile were typically collected at 2.5 cm intervals. Surface vegetation from the top of the core was trimmed and put into a separate bag.

Adjacent to some of the high resolution cores, soil recovery probes were used to sample soils to depths of 60 cm. These samples were sub-sectioned at 5 or 30 cm. At some sites (typically under the snowpack), soil plugs were collected to characterize the inventory of ⁷Be in the upper 4.8 cm of soil. These plugs were collected by hammering an 11.5 cm interior diameter

PVC pipe cap into the ground flush to the ground surface. A pre-drilled hole in the cap allowed air to escape. The soil was excavated along one side of the cap and the soil and cap removed.

All samples were placed in pre-tared sample bags, returned to the laboratory, dried at 80° C and prepared for radionuclide analysis. All upland soils and some floodplain soils were analyzed in total; that is, all size fractions in a soil sample were counted as one. Later, all floodplain soil samples were gently disaggregated by mortar and pestle and sieved to collect the fraction finer than 63 σ m which was analyzed by gamma spectroscopy.

Snowpack and Precipitation Sampling

We collected samples of the snowpack and precipitation in the headwaters of Soda Butte Creek in 2000 to establish the stored inventory of radionuclides released with snowmelt and to characterize the atmospheric flux during the field study. To collect samples from the snowpack, a five gallon (\sim 20 L) bucket was inverted and pounded into the snowpack perpendicular to the snow surface until the bucket was full and then it was removed. Additional buckets were filled in a similar manner as necessary to sample the entire thickness of the snowpack. Below the snowpack, three soil samples were collected to a depth of \sim 5 cm with a PVC cap as described earlier to determine the ⁷Be inventory of the soil.

The radionuclides in the melted snowpack were extracted by flocculation. Each sample was first acidified to a pH of 2 to 3 using 1 M HCl. 10 mL of FeCl₂ and 1 mL each of stable Pb and Be were added to the sample to confirm percent recovery. The pH of the sample was then raised to 8.2 by the addition of concentrated NaOH causing the flocculation of Fe(OH)₃ with the ⁷Be and ²¹⁰Pb and stable Be and Pb. The precipitate was allowed to settle for at least 8 hours at which time the majority of the supernatant liquid was siphoned off. The remaining sample was

then transferred to a 4 L separatory funnel where it sat undisturbed for another 8 hours. The precipitate was transferred to 100 mL specimen cups. A single precipitation sample was collected in a \sim 20 L bucket located in Cooke City. It was analyzed in an equivalent manner to that of the melted snow.

Daily precipitation and annual precipitation records were obtained from the National Climate Data Center (2002) and Farnes et al. (1999). Values of mean annual precipitation at sites without weather stations were interpolated from a regression ($r^2=0.98$) between elevation and annual precipitation established for the Soda Butte basin.

Suspended Sediment Sampling

Suspended sediments were collected in 2000 at about ten day intervals from nine sites along the Soda Butte Creek - Lamar River - Yellowstone River path (Figure 1). These sites extended from the headwaters of Soda Butte Creek to the Yellowstone River near Billings, Montana, a drainage length of 423 km. Occasional samples were collected at major tributaries along this reach and at Forsyth and Miles City, Montana, which are 196 and 283 km downstream of Billings, respectively. Regular sampling locations downstream from the mouth of Soda Butte Creek were at US Geological Survey gaging stations. Sediment was collected at sites at approximately 10 day intervals. The set of samples collected at approximately the same date is called a run.

Suspended sediment was collected by pumping water from the stream. The intake line to the pump was fixed in a position at least 1 m from the channel bank and at least 15 cm above the streambed. The water emitted from the output was passed through a series of 63 σ m, 32 σ m, and 20 σ m sieves that were positioned over a "32 gallon" trash can and collected. The mass of

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sediment retained on the sieves and the volume of collected water were recorded for determination of sediment and radionuclide concentrations in streamwater.

A second pump was used to draw the water from the trash can to a filter housing containing a 20.3 x 25.4 cm pre-tared sheet of 1 σ m nominal pore size Gelman Type A/E glass fiber filter paper. Filtration continued until flow rates dropped to <20% of normal rates due to sediment accumulated on the filter (0.7 to 1.2 g) whereupon filters were collected and replaced (Bonniwell et al., 1999). The process was repeated as needed to expedite collection and to ensure that enough sample was collected.

Filters were folded and placed into pre-tared 100 mL polyethylene specimen cups, dried at 60°C, and weighed to determine the mass of sediment collected. Suspended sediment concentrations were then calculated by dividing the mass of the sample by the volume of water collected. After the filters were weighed, they were transferred to ceramic crucibles and were "ashed" in a muffle furnace at 625°C for two to three hours. A maximum of three filters was ashed in each crucible. Samples were allowed to cool and were ground to a fine consistency with an agate mortar and pestle. Ground samples were then quantitatively transferred to pretared, capped petri dishes that were then sealed with electrical tape in preparation for analysis by gamma spectroscopy.

The sediments collected on the 32 σ m and 20 σ m sieve were combined and washed into a pre-tared specimen cup. The >63 σ m sediments also were washed into a specimen cup. Samples were dried and masses were obtained. The 20-63 σ m fraction was transferred into small petri dishes in preparation for analysis by gamma spectroscopy. During most sampling runs, only the two finer fractions were analyzed for radionuclide activities. All size fractions were analyzed for only runs 2 and 5.

During two sampling runs (2 and 5), the effluent water discharging from the filter housing was collected to measure the dissolved (<1 σ m) concentration of ⁷Be in the stream. Approximately 35-40 L of water were collected in two buckets. The ⁷Be was extracted in the same manner as described above for the precipitation and the snowpack samples.

Collected samples were labeled, dried, and returned to the laboratory for analysis by gamma spectroscopy. All ⁷Be activities were decay corrected to the date of June 7, 2000, the midpoint of the period of soil sampling.

In 1999, a limited set of suspended sediment samples was collected during a five day period from the headwaters of Soda Butte Creek to Glendive, MT (~850 km downstream). Approximately 20-40L of streamwater was retrieved from the stream using a bucket. The water and sediment was returned to the lab for separation of solids, drying, and analysis by gamma spectroscopy.

Streamflow

Many of the suspended sampling locations corresponded to the sites of active U.S. Geological Survey (USGS) gages (Figure 1). At these sites the USGS provided 15 minute interval records of discharge during the study period. At several sites along Soda Butte Creek (Campground, Cooke City, and Warm Creek Meadow), we established temporary gages to provide flow information. Pressure transducers were installed in slotted PVC pipe oriented vertically and anchored. Pressure was correlated to external stage plates and stage related to discharge by establishing rating curves. Standard USGS methods were used to determine flow discharge (Buchanan and Somers 1969). Stream discharge was measured over a range of stages using a Price current meter. At least 20 to 25 current velocity measurements were taken at equal

intervals along two traverses of the stream. Each measurement lasted 60 s. Stage was recorded before and after streamflow measurements. Flow discharge at the locations Above Campground, Above Republic, and Trout Lake was estimated by prorating measured discharges by drainage area.

GIS

The position of sediment sampling locations within the drainage network was characterized in several ways: longitudinal distance from the drainage divide, mean pathlength from the upstream watershed to the sampling location, and drainage area. These descriptors were taken from USGS reported values and by developing a GIS.

To develop the GIS, watersheds at the stream segment scale were delineated using the CRWR Pre-processor in ArcView 3.2. The required river data set and the Digital Elevation Models (DEM) for the study area were taken from the USGS. Some grids were only available in 30m resolution, while others were only available in 10m resolution. Consequently the 10m grids were re-sampled to 30m grids. The corresponding 1:24,000 scale river files were acquired from the National Hydrography Dataset. Using ArcMap, a geometric network of the route reach file and its nodes was created; isolated reaches and loops were removed; and mis-connections and bad geometry were corrected. The streams were burned into the DEM to ensure that the generated streams matched the actual river data set. Next, imperfections, common in DEMs, were filled to prevent incorrect flow direction. Flow directions were then calculated based on an eight-point pour model (Fairfield and Leymarie, 1991), and flow accumulations, or the number of upstream cells contributing to the catchment, were quantified. The mean flow path length from each grid cell within that catchment area to the study site was calculated.

Gamma Spectroscopy

Gamma spectroscopy for ²¹⁰Pb (46.52 keV), ⁷Be (477.6 keV), ¹³⁷Cs (661.65 keV), and ²¹⁴Bi (609.3 keV) is reasonably well-established and straightforward (Larsen and Cutshall, 1981), but the analytical details are detector and sample specific. We used two HPGe gamma detectors for the radionuclide analysis. Suspended sediment and soil samples were counted for 22-24 hours to decrease the counting errors associated with small samples and low radionuclide activities (~10-20%). Each sample was dried, weighed, and sealed in an enclosed 47 mm polystyrene petri dish for counting. The soil samples were counted after being sealed for at least three weeks to allow secular equilibrium ingrowth of gaseous 222 Rn (t_{1/2} = 3.82 days) from the decay of its 226 Ra (t_{1/2} = 1600 years) parent. Due to the low energy of the 46.7 keV 210 Pb photon, the self-attenuation correction of Cutshall et al. (1983) was used. Standards were prepared using the same petri dish and specimen cup geometries as the samples. The instrumentation and counting geometries were calibrated using commercially available mixed energies gammaemitting standards (Amersham OCY44 and NG4; NIST Standard Reference Materials 4276C). A quality assurance (QA) standard (NIST 4350B River Sediment) was run routinely to verify the accuracy of the analyses. We participated in an interlaboratory comparison program with sample splits and standards as a part of our larger QA counting procedure. All measured counts were corrected for background levels, detector and geometry efficiencies, branching ratios and decay.











