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Estimating fluxes in anthropogenic lead using alluvial soil mass-balance geochemistry, geochronology and archaeology in eastern USA

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ABSTRACT

Predicting human impact on Earth's surface requires a quantitative understanding of past human–environmental interactions. This study uses mass-balance geochemistry of buried alluvial soils, geochronology and archaeology to constrain soil age and quantify the anthropogenic addition of lead (Pb) to alluvial soils from the Delaware River valley in northeastern USA. Mass addition of Pb was calculated for two alluvial terrace landforms. The results show that for buried soils that formed during the middle and late Holocene, the addition of Pb was estimated at $1.71 \pm 0.02\ \mu g\ cm^{-2}$ and $-39 \pm 0.43\ \mu g\ cm^{-2}$, respectively. The corresponding Pb addition rates were estimated at $0.04\ \mu g\ cm^{-2}\ y^{-1}$ and $-0.28\ \mu g\ cm^{-2}\ y^{-1}$, -1, where the negative rate documents Pb loss from the soil. Conversely, the Pb addition to buried alluvial soils that formed <100 years ago were estimated at $486\ \mu g\ cm^{-2}$ and $526\ \mu g\ cm^{-2}$. The corresponding mean Pb addition rates were estimated to be $10.06 \pm 2.13\ \mu g\ cm^{-2}\ y^{-1}$ and $8.91 \pm 4.66\ \mu g\ cm^{-2}\ y^{-1}$. The recent Pb addition rates documented in this study are over 2 orders of magnitude higher than their middle and late Holocene counterparts. The >200 times increase in Pb and rapid deposition rates in the 19th–20th century alluvial soils coincide with the presence of historic Euro-American artifacts, providing support for human-induced Pb addition. Furthermore, the soil stratigraphic context of a tooth-crown bottle cap (maximum age = 58 y BP) and other archaeological age-based estimates allowed us to constrain the residence time of a historic alluvial soil to 58–39 y. The mass-balance results show that 19th–20th century Pb additions to these two alluvial profiles are unlike anything documented in the buried alluvial soils prior to the Industrial Era. Although human-induced mass influx of Pb had been previously documented in nearby lake cores and upland soils, quantification of Pb flux in buried alluvial soils, especially those with archaeological deposits, will further constrain the magnitude, extent and timing of anthropogenic Pb impact on Earth's surface.

Introduction

Quantifying human impact on Earth's surface materials is important because the mobilization of surface material may cause system-state changes on Earth that are unsustainable. One proxy for human impact is anthropogenic lead (Pb) enrichment, which could serve as both a prominent chemostratigraphic marker and a quantitative metric of human-impact on soils and sediments. For at least 2500 years human activities, e.g., ancient silver mining, wet/dry deposition of coal fly ash, leaded gasoline combustion and metal smelting plants have mobilized Pb, often resulting in elevated concentrations in soil and sediment (Lantzy and Mackenzie, 1979; Nriagu, 1983; Weiss et al., 2007 and references therein). Pb data from a Greenland ice core show increasing Pb concentrations beginning late 19th century and up to 1970 (McConnell et al., 2002). Unlike short-lived radioactive isotopes (e.g., Cs-137), increased Pb concentrations in soil and sediment will likely stay in the Anthropocene record for millennia, making it an ideal boundary marker. Thus, understanding the timing, rate and processes of anthropogenic Pb are essential to understanding the utility of this anthropogenic marker.

Intuition holds that the modification of the Pb cycle on Earth's surface could be quantified by mapping and measuring the Pb in soil. However, anthropogenic-Pb enrichment in soils can be

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confounded by biological and geomorphological setting and process(es). Soil profiles are time-averaged weathering features where Pb addition (e.g., parent material, dust, anthropogenic), loss, transformation, and translocations can affect the same profile at different intervals of time or simultaneously. The time-averaging processes of over-printing or welding can mask anthropogenic-Pb enrichment by adding or removing mass that causes apparent changes in Pb concentration. Certain types of vegetation can also concentrate Pb in high quantities in the soil. For example, roughly 0.2% of all angiosperms are heavy metal (As, Cd, Co, Cu, Mn, Ni, Pb, Sb, Se, Tl, Zn,) hyperaccumulators and metal concentrations can be 100–1000 times higher in their leaves compared to surrounding levels (Rascio and Navarri-Izzo, 2011). The organic litter from hyperaccumulators is then incorporated into the soil organic matter yielding higher Pb concentrations in the soil, which could be misidentified as anthropogenic-Pb enrichment. Conversely, experimental studies show that Pb solubility can increase with decreasing pH (Reddy et al., 1995). Solubility and leaching would deplete anthropogenic-Pb in highly acidic soils.

One way to overcome these issues is to quantify anthropogenic Pb impact on soils in depositional settings. Quantifying anthropogenic-Pb enrichment in buried and surface alluvial soils could yield more discrete episodes of pedogenesis (Holliday, 2004), unlike upland soils that often record prolonged, time-averaged episodes of pedogenesis (Saint-Laurent et al., 2010). The study of depositional sequences in alluvial settings in conjunction with the chronological resolution and behavioral context provided by archaeology is a potential solution to some of these problems, providing a clearer view of human impact on the environment. The study documented herein attempts to quantify the anthropogenic Pb flux in an alluvial environment in northeastern USA using a combined mass-balance geochemistry, geochronology and archaeology approach. The results are compared with previous research and include a discussion of regional Pb variations.

Materials and methods

Study area

Research along the Delaware River Valley, northeastern USA has shown that the floodplains and alluvial terraces consist of multistory buried soils that span the Holocene (Ritter et al., 1973; Stewart, 1990, 1991; Stewart et al., 1991; Stinchcomb et al., 2012). Because these soils have been age-constrained in past work (Stinchcomb et al., 2011, 2012, 2013), they make a good case study for comparing background Pb with anthropogenic Pb.

The study area is located within the Delaware River basin, eastern United States (Fig. 1). The middle Delaware River valley is a partly confined river valley containing relict glacial and periglacial landforms and alluvial landforms that include alluvial fill terraces, floodplains, islands, and gravel bars (Stewart et al., 1991; Witte, 2001; Schuldenrein, 2003; Witte and Epstein, 2005; Stinchcomb et al., 2012; Witte, 2012). Along this portion of the river valley, glacial recession occurred ~16 ka, which led to periglacial deposition, followed by late Pleistocene incision and then alluvial deposition around 12.8 ka (Stewart et al., 1991; Witte, 2001). A 6–5 ka incision event, followed by floodplain and terrace reworking for the past 6000 years, resulted in discontinuous alluvial terrace development throughout the reach (Stinchcomb et al., 2012; Witte, 2012). The T2 alluvial terrace comprises much of the valley bottom along the middle Delaware River valley. The surface elevation above river baseflow can vary along the T2 (11-5 m) and can be

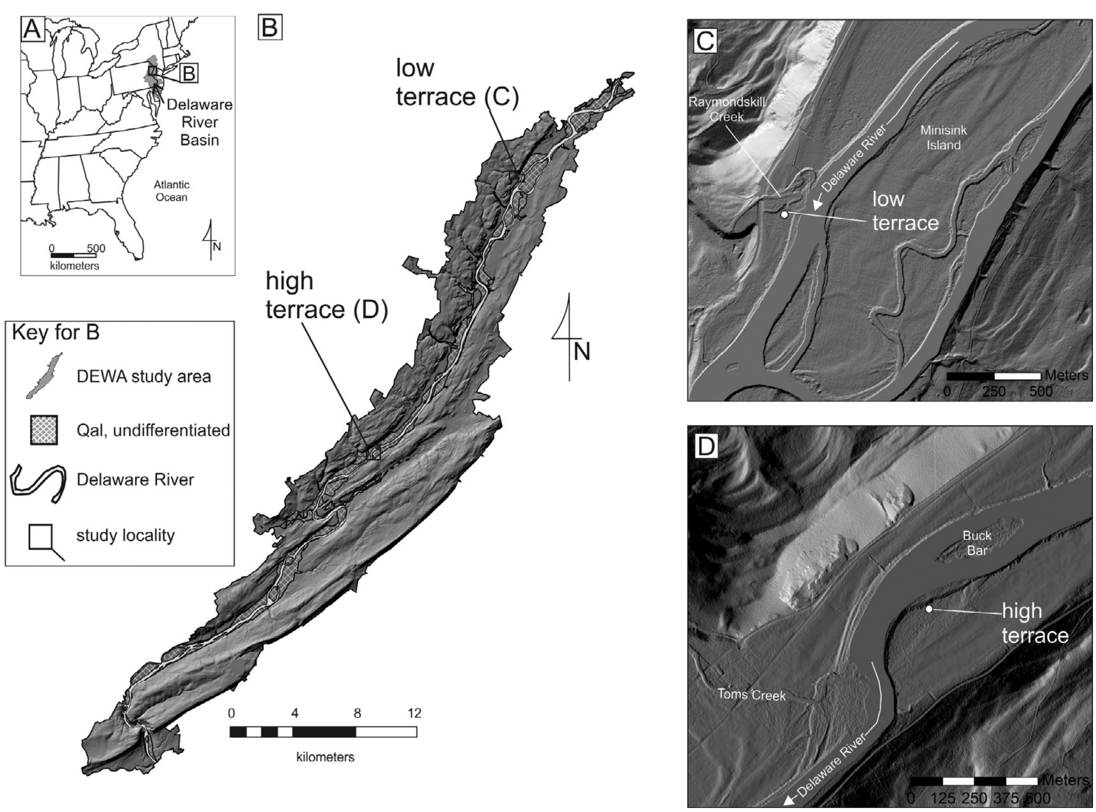

Fig. 1. Northeastern USA study area. (A) Map of northeastern USA showing Delaware River Basin. (B) Delaware Water Gap National Recreation Area (DEWA) located along the Middle Delaware River Valley. The locations of the two alluvial profiles are shown along with their corresponding alluvial terrace designations, referred to herein as high- and low-terrace landforms. The distribution of Quaternary alluvium (Qal) is also shown (Witte, 2001, 2012; Witte and Epstein, 2005). (C) and (D) show the exact locations of the high- and low-terrace landforms on bare-Earth LiDAR hillshades (1 m resolution). The Manna archaeological site mentioned in text corresponds with the low-terrace landform study area.