

Enduring legacy of a toxic fan via episodic redistribution of California gold mining debris

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The interrelationships between hydrologically driven evolution of legacy landscapes downstream of major mining districts and the contamination of lowland ecosystems are poorly understood over centennial time scales. Here, we demonstrate within piedmont valleys of California's Sierra Nevada, through new and historical data supported by modeling, that anthropogenic fans produced by 19th century gold mining comprise an episodically persistent source of sediment-adsorbed Hg to lowlands. Within the enormous, iconic Yuba Fan, we highlight (i) an apparent shift in the relative processes of fan evolution from gradual vertical channel entrenchment to punctuated lateral erosion of fan terraces, thus enabling entrainment of large volumes of Hg-laden sediment during individual floods, and (ii) systematic intrafan redistribution and downstream progradation of fan sediment into the Central Valley, triggered by terrace erosion during increasingly long, 10-y flood events. Each major flood apparently erodes stored sediment and delivers to sensitive lowlands the equivalent of ~10–30% of the entire postmining Sierran Hg mass so far conveyed to the San Francisco Bay-Delta (SFBD). This process of protracted but episodic erosion of legacy sediment and associated Hg is likely to persist for >10⁴ y. It creates, within an immense swath of river corridor well upstream of the SFBD, new contaminated floodplain surfaces primed for Hg methylation and augments/replenishes potential Hg sources to the SFBD. Anticipation, prediction, and management of toxic sediment delivery, and corresponding risks to lowland ecology and human society globally, depend on the morphodynamic stage of anthropogenic fan evolution, synergistically coupled to changing frequency of and duration extreme floods.

mercury | lowland contamination | flood frequency | climate change | morphodynamics

Hg poses contamination risks to food webs globally (1, 2), particularly where sediment-bound Hg is delivered to topographically low, depositional environments with high methylation potential. Sediment is the vehicle for ~97% of transition metal mass loading to oceans (3), and the geomorphic configuration of stored contaminated sediment on land has important implications for its subsequent remobilization (4) and delivery to the base of food webs. Sediment displaced by industrial mines is typically contaminated by toxic metals such as Hg (5–7), which threaten lowland ecosystems downstream of mining sites around the world (e.g., refs. 6–9). This contaminated sediment often infills confined river valleys (10) as massive anthropogenic fans, similar to natural alluvial fans, which undergo subsequent incision once the supply of mining sediment is stopped. Such fans are likely to increase in ubiquity and scale across the globe as demand increases for mined metals and rare earth elements.

Nineteenth century hydraulic gold mining in California's Sierra Nevada foothills (1853–1884) contemporaneously delivered ~1.1 km³ of Hg-contaminated sediment (10, 11) to downstream valleys at concentrations >10-fold higher than geographical “hot spots” of atmospheric Hg deposition (12). These immense mining sediment slugs choked Sierra rivers, which aggraded tens of meters in confined piedmont valleys, and formed anthropogenic

fans grading to the margins of the Central Valley, including the ~252 × 10⁶-m³ Yuba Fan (10) (Fig. 1 *A* and *B*). Since that time, sediment-bound Hg has contaminated fish and waterfowl of the San Francisco Bay-Delta (SFBD) (13, 14). However, the dominant modern geographical sources of Hg, as well as the processes, patterns, and time scale of its delivery to this downstream ecosystem, are debated (15–17), partly due to the lack of a generic understanding of postmining fan evolution. It is unclear whether there are modern sources of Hg to this system and by what set of physical and biogeochemical processes Hg may penetrate these sensitive ecosystems. The legacy impacts of historical mining are therefore not fully comprehended, in part, because of incomplete knowledge of the magnitude and frequency of processes contributing to fan evolution and sediment exhaustion.

Following cessation of hydraulic mining, Sierra valley aggradation was succeeded by vertical channel incision into piedmont fans, producing historical terraces along channels (10). James (18) demonstrated that ~90% of the original sediment deposit delivered to the Bear River valley (Fig. 1*A*) remained in terrace storage >100 y after mining and hypothesized that modern floods are capable of transporting it downstream. Apart from Gilbert's classic work on bed-level change (10), generalizable theory for flood-based evolution of anthropogenic fans is absent, especially over the critical management time scale of decades to centuries in this and other fluvial systems (5, 6). This precludes predictions of mining sediment exhaustion time scales and the downstream risks of heavy metal contamination to food webs.

To fill this research gap, we analyzed high-resolution historical topographical and bathymetric data collected in 1906 and 1999, and interpreted them here within the context of: historical US Geological Survey (USGS) streamflow (Q) measurements and

Significance

This paper is of fundamental interest to the millions of residents living at the downstream end of this and other global river basins beset by industrial metals mining. Sediment-bound Hg has contaminated food webs of the San Francisco Bay-Delta, but the dominant geographical sources of Hg to downstream ecosystems in this and similar river basins are debated. Likewise, the processes by which Hg is delivered to lowlands and the patterns of its floodplain deposition are poorly understood. This research addresses a gap in generic theory of postmining fan evolution that enables anticipation, prediction, and management of contamination risk to food webs.

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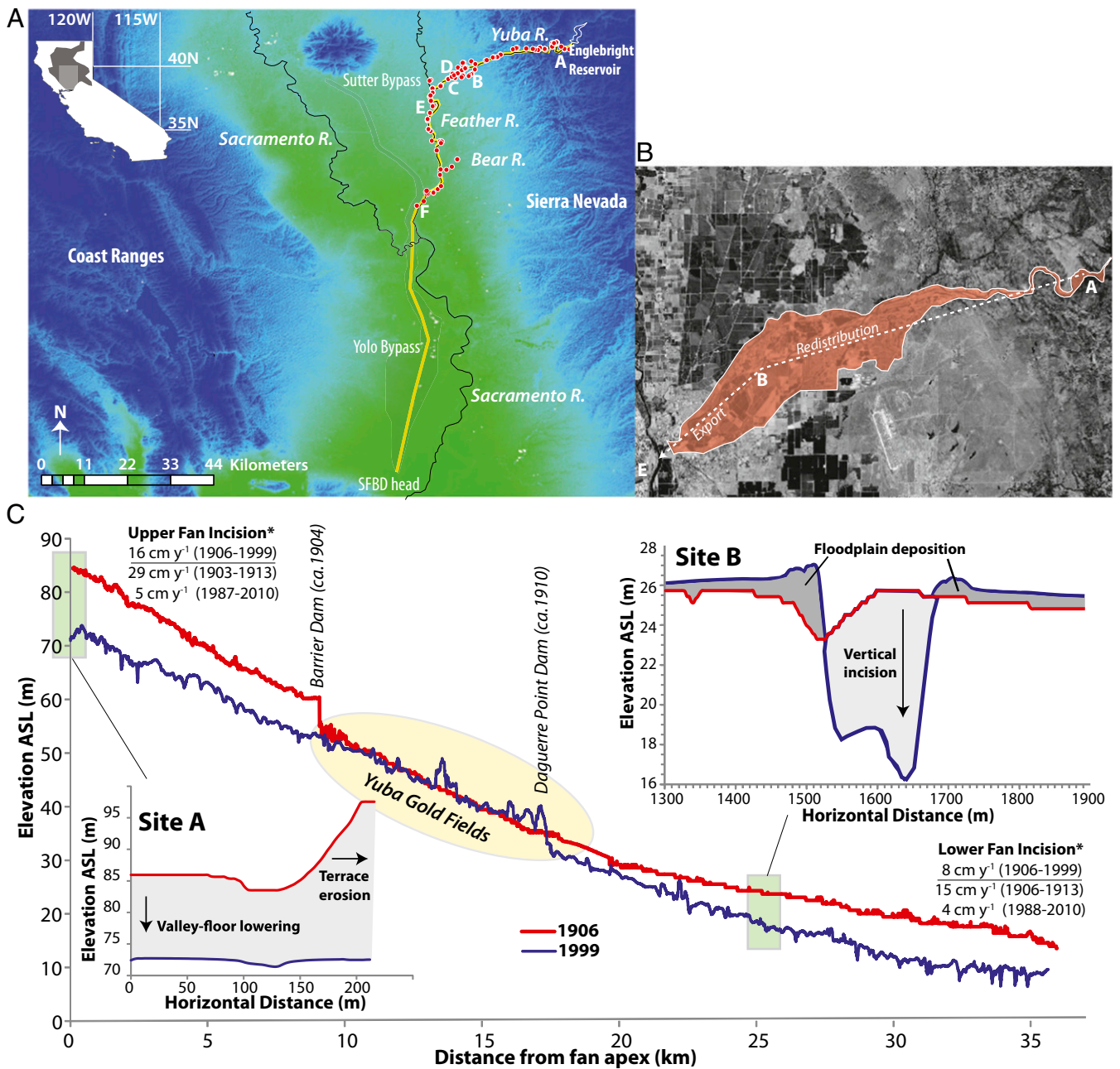


Fig. 1. Yuba Fan. (A) HgT sediment sampling locations (red circles). The longitudinal transect along which mining sediment travels from the Sierra to the SFBD is shown in yellow. Sample sites A–F are described in the main text. Base: Shuttle Radar Topography Mission (SRTM) 1 arc-second. Yuba Fan delineation and hypothesized patterns of sediment redistribution and export (B) and longitudinal elevation profiles of the Yuba River channel bed, 1906 and 1999 (C). (Insets) Plots show cross-sectional change at sites A and B. *Fan incision rates for the early (10) and late 20th century are based on trends in water surface elevation. Supporting methodological description and data on slowing incision are provided in *SI Methods* and *Table S1*. ASL, above sea level.

flood frequency; National Aeronautics and Space Administration flood imagery; and our newly developed geochemical dataset that includes total Hg concentration (HgT), Zr/Ni, and ¹³⁷Cs, supported by physically based mathematical modeling of terrace erosion and flux of mining sediment during large floods (*SI Methods*).

Results and Discussion

Terrestrial/bathymetric topographical data, collected in 1906 and 1999, reveal that the Yuba River longitudinal profile has incised systematically over the entire fan in the past century, manifesting as substantial valley-floor lowering (i.e., erosion of the channel and adjacent floodplains) and terrace erosion in the upper fan

and as vertical channel incision with modest (<1 m) floodplain aggradation in the lower fan (Fig. 1C). The riverbed level of the middle fan (“Yuba Gold Fields”) has been affected by the presence of Daguerre Point and Barrier Dams, built to trap mining sediment. Barrier Dam was destroyed by a large flood in 1907 (10), resulting in a marked adjustment of the thalweg long profile (Fig. 1C). The reservoir behind Daguerre Point Dam (built in 1910) remains completely full of Hg-laden sediment (19), so it suppresses vertical incision within the Yuba Gold Fields and has negligible sediment trap efficiency during floods.

Nevertheless, the overall centennial pattern of change appears to be systematic incision and redistribution of sediment from the

narrow upper Yuba Fan to the relatively wide lower Yuba Fan (note floodplain deposition in the lower fan, Fig. 1C) and net fan export (Fig. 1B). Streamflow measurements at USGS gauges (*SI Methods*), corroborated by repeat cross-section surveys (20) and our own field observations of bed grain size, indicate that the average incision rate has slowed considerably in recent decades (Fig. 1C and Table S1), as coarse-grained premining riverbeds were reexposed in the upper fan and a single-thread channel was reestablished in the lower fan (21). We hypothesize that once vertical incision retards, fan evacuation proceeds predominantly as lateral erosion of historical terraces by competent high flows (22).

Longitudinal patterns of sediment redistribution by floods are expressed in chemostratigraphy and measured by 176 values of HgT in the <63- μm fraction of sediment at 105 locations along the Yuba, Feather, and Bear Rivers (Fig. 1A, *SI Methods*, and Dataset S1). HgT values in the Sierra are naturally low [nonmining sediment contains HgT $\leq 0.08 \mu\text{g/g}$ (15), and Hg ore was mined and roasted in the Coast Ranges to produce elemental Hg that was transported to the Sierra for use in gold mining (11)]. Therefore, HgT in Sierra alluvium is a discriminating tracer of mining activity (15). Our HgT data range over two orders of magnitude, and thus easily distinguish stratigraphic units containing historical sediment from gold mines [regional characterization of pre- v. postmining sediment is provided by Bouse et al. (15)], constrained by documented channel change (21).

Some important generalizations emerge:

- i) Contact between pre- and postmining sediment layers consists of at least an order-of-magnitude increase in HgT in the anthropogenic sediment (Fig. 2 B–D). Furthermore, upper stratigraphic sections from relatively recent floods typically contain lower values of HgT (0.18 $\mu\text{g/g}$ or greater than

- threefold average background values) compared with underlying historical layers (Fig. 2B and Dataset S1), which apparently reflects Hg dilution through the mixing of mining and nonmining sediment, as observed in other fluvial systems (5, 6).
- ii) Deposits that were recently reworked by channel incision/migration have lower HgT than those that were not (compare Fig. 2B and E vs. C and D), because incision/migration dilutes HgT by incorporating premining sediment from upstream sources. Consequently, modern channel bed/bar sediment is generally lower in HgT than fine fractions of floodplain sediment deposited during fan emplacement.
- iii) Large stores of undiluted mining sediment in historical terraces of the upper fan contain high HgT (>1 $\mu\text{g/g}$) and are susceptible to flood-based erosion (Fig. 2A).
- iv) Great masses of sediment from various parts of Yuba Fan travel into the Central Valley during floods, as observed in bank sections along the Feather River and in large splay deposits in lowland floodplains (Fig. 2E and F, respectively), which corroborates historical suspended sediment records (23) and remote sensing imagery (24). For example, our measurements showed that the 1986 flood (25) delivered to a lowland floodway a discrete sediment deposit of $2.5 \times 10^6 \text{ m}^3$ with HgT = 0.18 $\mu\text{g/g}$, equivalent to the uppermost deposits shown in Fig. 2B and E.

Recent deposit ages identified in the chemostratigraphy are supported by our own radionuclide measurements that characterize whether upper sediment layers were stable before or after bomb testing in the 1960s, which delivered meteoric fallout of ^{137}Cs and therefore provides a marker horizon that can be used to date fluvial sediment deposits (26). For example, the uppermost surface (Fig. 2A) contains $^{137}\text{Cs} = 13.7 \text{ mBq/g}$, indicating its longer term persistence. Lower inset terrace surfaces (Fig. 2A) and upper sediment layers (Fig. 2B and F) contain 0 mBq/g,

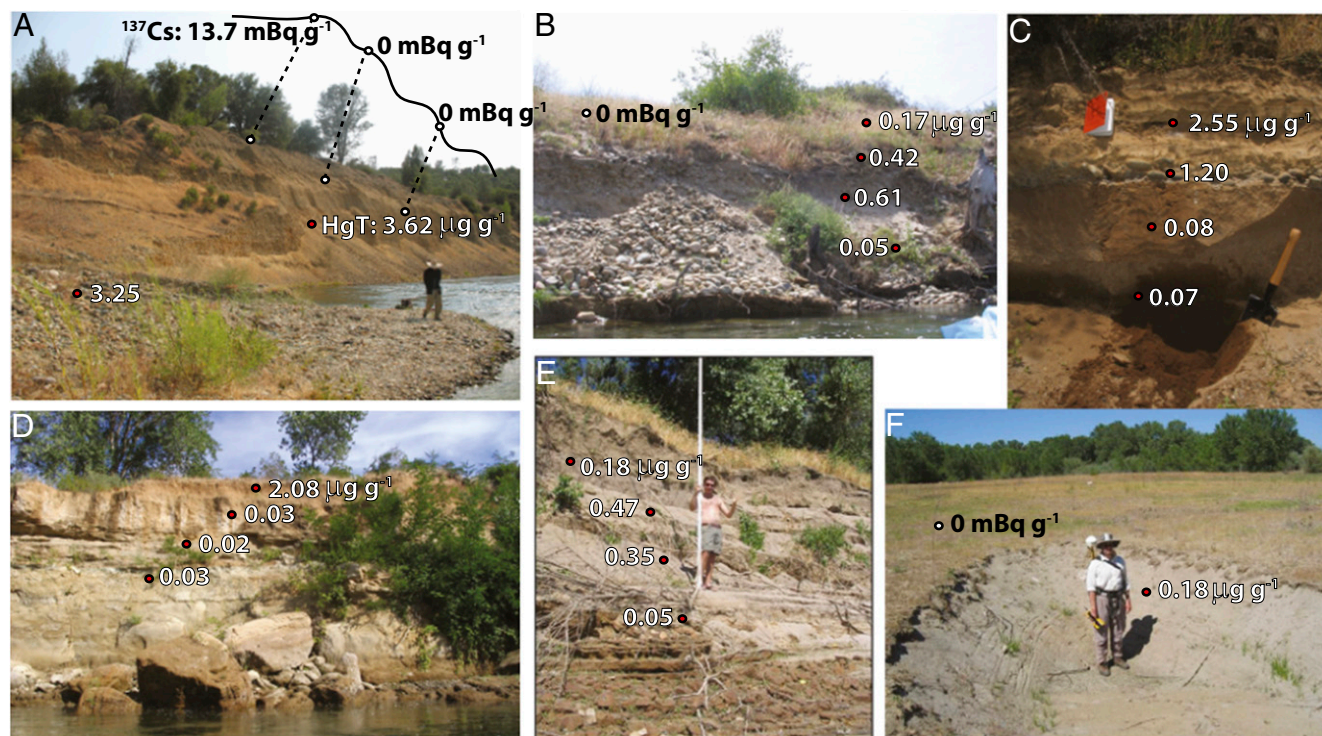


Fig. 2. Characteristic sampling sites and geochemical data arranged in downstream order. Red circles indicate approximate HgT sampling locations within the chemostratigraphy. White circles indicate approximate ^{137}Cs sample locations from surface sediments. (A, C, and D) Intact mining sediment from the original mining period. (B, E, and F) Upper layers of reworked mining sediment mixed with background (nonmining) sediment. F is adapted from Singer et al. (25). E and F represent lowland sites downstream of the Yuba Fan along the Feather River (Fig. 1A).

Our analyses highlight the importance of anthropogenic fans in the apparent lateral-expansion stage of morphodynamic evolution as primary stores, sources, and downstream exporters of mining sediment and associated contaminants. Assuming that 90% of the Yuba Fan remains in storage [equivalent to estimates for the Bear River (18)], ~0.1% of mining sediment (both fine and coarse) is evacuated per decade by a single large flood event such as those outlined here, implying a source of contaminated sediments to lowlands that will last for $>10^4$ y (*SI Methods*). This is consistent with a right-skewed model of sediment exhaustion (31) punctuated by increasingly heavy-tailed flood frequency (28, 29), which could slightly shorten mining sediment exhaustion times.

Based on modeled fluxes for flood events and assuming (*SI Methods*) conservatively low HgT, equivalent to concentrations in recent lowland flood deposits (Fig. 2F), the recent major floods exported large masses of Hg from the Yuba Fan into the Central Valley (Fig. 4 B and C). Such per flood event-based contaminant flux, predominantly derived from historical terraces relative to channel/bar sediments (Fig. 5A), represents a substantial fraction of the cumulative Hg mass (~200 kg) that is estimated to have reached the SFBD since mining began (15). Therefore, most Hg exported from anthropogenic fans is apparently still stored in lowland floodplains and floodways upstream of the SFBD. It manifests as an ~70-km long swath of Hg contamination from the Yuba Fan into the Central Valley (Fig. 5A).

This interpretation is corroborated by an independent geochemical proxy for mine-derived sediments, Zr/Ni (15). Hydraulic mining sediments in the Sierra largely derive from highly weathered Eocene sediments from which gold was extracted, and thus have high Zr/Ni ratios due to the selective removal of Ni and relative immobility of Zr (15). We analyzed our samples for these elements and found a clear down-valley trend in Zr/Ni (Fig. 5B). The HgT data show much higher variability, primarily as a consequence of sampling at various topographical positions (Fig. 2), but roughly follow a similar trend. Each elemental signature of hydraulic mining is apparently diluted downstream through the Yuba Fan, the scatter in HgT notwithstanding, and is relatively concentrated in floodplain deposits near the fan margin (Fig. 5). These data support the redistribution of mining sediment from the upper to lower Yuba Fan and its punctuated export from the entire fan into the Central Valley (Fig. 1B). They are also consistent with Hg isotope ratios in shallow SFBD sediments, which suggest a persistent upstream (Sierran) Hg source (16, 32).

Lowland floodplains are potential hotspots for Hg methylation of mining debris. Fine-grained Hg from hydraulic mining is as-

sociated with sulfide minerals (19), which are prone to oxidation (to mercury sulfate) during transport. Once delivered to lowlands, these sediments are redeposited, producing contaminated floodplain surfaces that undergo frequent wetting/drying cycles. These cycles may stimulate resident sulfate-reducing bacteria to methylate the newly arrived Hg near the sediment-water interface at very high rates (33, 34), posing risks to food webs in this and other ecosystems beset by mining-induced displacement of contaminated sediment.

The enduring legacy of mining on a grand landscape scale entails persistent lateral erosion of anthropogenic fans that continues well after the slowing of initial vertical incision and return of the channel to its original bed level. This process stage of fan evolution apparently sustains the evacuation of legacy sediment, triggered by increasingly potent floods that effectively deliver toxic sediment slugs downstream into sensitive lowlands, thus augmenting a major potential source of food web contamination in tidal wetlands of the SFBD.

These links between upland legacy mining sediment and lowland contamination, which are germane to issues of river management such as licensing for mining or dams in this and other basins around the world, are enabled by the topographical position of stored sediment and the frequency/magnitude of flood events capable of transporting contaminated sediment over tens of kilometers. Our findings suggest dangerous synergies between the morphodynamic stage of fan evolution, regional shifts in climate, and contamination risks to lowland ecosystems and human populations.

Methods

This paper presents the culmination of a multipronged approach, including assessment of historical topographical datasets, investigation of historical streamflow measurements at USGS gauging stations to determine rates of incision, flood frequency analysis, interpretation of HgT chemostratigraphy, Zr/Ni geochemistry as an independent proxy for mining sediment, and use of ^{137}Cs to establish age control for younger deposits. These approaches were combined with terrace erosion modeling based on slope stability tied to hyporheic infiltration, partitioning of failed material into suspended sediment load, and numerical modeling suspended flux through the fan (*SI Methods*).

Topographical longitudinal and cross-section profiles were extracted from historical maps and recent LiDAR tied to photogrammetry and sonar data (35). Stratified sediment samples for HgT ($n=176$) were extracted from channel bars and bank exposures at field-identified sedimentary units ($n=105$) typically in vertical sections. Samples were sieved through $<63\text{-}\mu\text{m}$ stainless-steel sieves to compare a common sediment population across all sites. The resulting material was digested at a USGS Hg laboratory in Menlo Park, California. Digested samples were harvested and analyzed on a Tekran

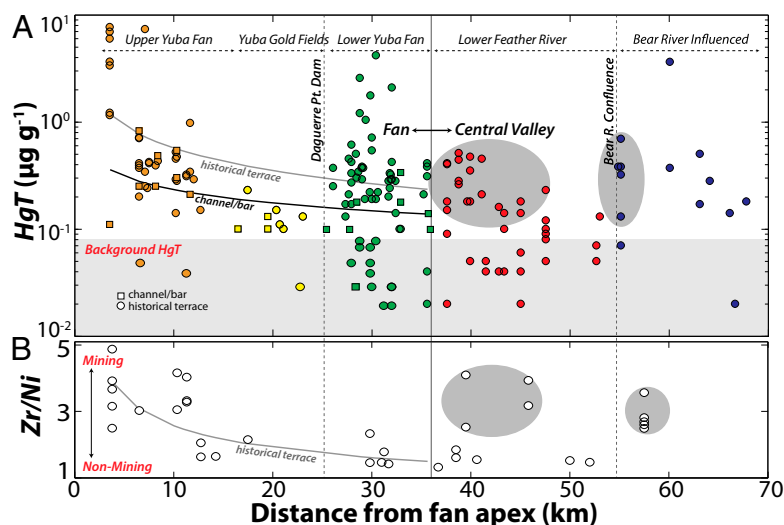


Fig. 5. Downstream patterns of geochemistry within and beyond the Yuba Fan. Measured HgT (A) and Zr/Ni (B) in sediments (*Dataset S1*). Power relationships fit to all HgT (including all stratigraphic positions from each historical terrace) and Zr/Ni data within the Yuba Fan are given in Fig. S1. Colors in A refer to geographical domains listed at the top of the figure. Shaded ovals indicate elevated geochemical concentrations downstream of the margins of anthropogenic fans.

Series 2600 Automated Mercury Analysis System according to standard procedures for quality assurance/quality control (QA/QC) (US Environmental Protection Agency method 1631, revision E) using International Atomic Energy Agency (IAEA) 405 as certified reference material. Zr/Ni analyses were conducted by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and X-Ray Fluorescence (XRF) at Activation (ACT) Laboratories Ltd. and the University of St. Andrews. The ^{137}Cs data were generated in the Exeter Radiometry Laboratory with high-purity germanium (HPGe) spectrometers.

Physically based terrace erosion modeling derives from the infinite slope stability model for riverbanks (36), wherein factor of safety (FoS) was driven by stage-discharge recorded at the upstream gauging station. This boundary flow fed into a 2D numerical model of infiltration via the Richards equation following the Dupuit–Forchheimer approximation. FoS was computed based on field-measured failure geometry and grain sizes and modeled water content. Following failure, the bed grain size distribution (GSD) was updated based on contribution from measured terrace GSD depending on the depth of failure. Sediment flux was computed iteratively for the cross-section to quantify the suspended load (Fig. 4A), based on daily flood stage and updated GSD by the Singer–Dunne equation (37). Flow and SSC through the Yuba River and its floodplain were modeled using TELEMAC 2-D (Laboratoire

National d'Hydraulique, Paris, France) and Sisyphe (a coproperty of the Centre d'Études Maritimes et Fluviales, Université de Technologie de Compiègne, Électricité de France, and the Société Grenobloise d'Études et d'Application Hydrauliques), a hydrodynamic and washload transport modeling approach (38), on a finite element mesh derived from high-resolution topographical data for the channel and overbank areas. Suspended sediment input at the upstream boundary in the model of the Yuba Fan (and corresponding uncertainty) was conservatively approximated as a point source based on an empirical relationship between sediment concentration and discharge (*SI Methods*).

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