

210Pb geochronology of flood events in large tropical river systems

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Floodplain sedimentation removes particles from fluvial transport and constructs stratigraphic records of flooding, biogeochemical sequestration and other aspects of the environmental history of river basins—insight that is enhanced by accurate geochronology. The natural fallout radionuclide 210Pb, often employed to date lacustrine and marine sediments, has previously been used to determine floodplain accumulation rates over decadal-to-century time scales using the assumption that both input concentration and sediment accumulation rates are constant. We test this model in approximately 110 cores of pristine floodplains along approximately 2000 km of the Rios Beni and Mamore in northern Bolivia; over 95 per cent of the 210Pb profiles depict individual episodic deposition events, not steady-state accumulation, requiring a revised geochronological methodology. Discrete measurements of down-core, clay-normalized adsorbed excess 210Pb activity are coupled with a new conceptual model of 210Pb input during floods: constant initial reach clay activity, unknown sedimentation (CIRCAUS). This enhanced methodology yields 210Pb dates that correspond well with (i) dates determined from meteoric caps, (ii) observed dates of river bar formation, (iii) known flood dates, and (iv) dates from nearby cores along the same transect. Similar results have been found for other large rivers. The CIRCAUS method for geochronology therefore offers a flexible and accurate method for dating both episodic (decadal recurrence frequency) and constant (annual recurrence) sediment accumulation on floodplains.

Keywords: 210Pb; geochronology; floodplain accretion; sediment accumulation; Bolivia

1. Introduction

River histories run from millions of years to hours: exceptional floods can transform a valley through avulsion, regular floods orchestrate channel migration, and even the smallest flood results in some sediment accretion. While less dramatic, the regular accumulation of sediment conveyed overbank by the turbid waters of moderate floods continually constructs basin stratigraphy—in the form

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of floodplains elevated above channel bar deposits. These processes can now be traced with a new host of dating and imaging techniques. Table 1 lists the key procedures available for dating river sediment and the dating ranges for which they are best suited, along with other logistical considerations.

The fallout radionuclide $^{210}\text{Pb}$ provides an insight into fluvial processes over the past century. To determine the timing and accumulation rates for riverborne sediment (including associated biogeochemistry and pollutants) across floodplains, sedimentation processes must be investigated for a wide expanse of locations and quantified on at least a decadal-to-century scale, so as to resolve annual variations in the spatial distribution and amount of sediment transferred from the channel to the floodplain. Until the development of $^{210}\text{Pb}$ geochronology for floodplains in the mid-1990s, researchers could only document the apparent accumulation of ‘fresh sediment’ (a lens of river sediment deposited during a single flood, covering any vegetation or developed soil) either during the event itself (sediment traps) or immediately thereafter (surveys of sediment deposits). Without a prolonged logistical effort spanning decades (e.g. repeated benchmarked topographic surveys at the same site [1]), it was unclear how the results from a single event could be integrated into a century-scale documentation of floodplain accumulation.

Most previous studies have used $^{210}\text{Pb}$ geochronology to date average sediment accumulation rates on grassy floodplains. We applied and tested such techniques for densely forested tropical floodplains along large lowland river channels in northern Bolivia (figure 1): an 800 km section of the Rio Beni and a 1200 km section of the Rio Mamore, both exhibit thick, extensive deposits of fine sediment

### Table 1. List of radionuclide techniques used for dating floodplain deposits. Half-lives of radioisotopes are shown in parentheses and the useful dating range is listed, followed by a brief summary of benefits and challenges.

<table>
<thead>
<tr>
<th>dating technique</th>
<th>age range</th>
<th>benefit</th>
<th>challenge</th>
</tr>
</thead>
<tbody>
<tr>
<td>fallout radionuclides</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{210}\text{Pb}$ (22.3 yr)</td>
<td>1–110 yr</td>
<td>cheap and fast to measure with alpha spectrometry</td>
<td>poor gamma emitter, chemistry for alpha</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$ (30.2 yr)</td>
<td>1959/63 peaks</td>
<td>short period of arrival resolves fast events</td>
<td>signal growing weak, limited temporally</td>
</tr>
<tr>
<td>$^{7}\text{Be}$ (0.14 yr)</td>
<td>&lt;0.6 yr</td>
<td></td>
<td>expensive to process, difficult assumptions, mobility</td>
</tr>
<tr>
<td>$^{10}\text{Be}$ (1.4 Myr)</td>
<td>1 kyr–5 Myr</td>
<td>date ancient sediments</td>
<td></td>
</tr>
<tr>
<td>material in deposits</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{14}\text{C}$ (5.7 kyr)</td>
<td>100 yr–50 kyr</td>
<td>simple to collect and send to accelerator mass spectrometry laboratories</td>
<td>expensive, problems with inherited age</td>
</tr>
<tr>
<td>optically simulated luminescence</td>
<td>300 yr–100 kyr</td>
<td>deposition age</td>
<td>expensive, problems with resetting</td>
</tr>
<tr>
<td>$^{10}\text{Be}$ in situ</td>
<td>1 kyr–5 Myr</td>
<td>exposure age, if shielding is known</td>
<td>expensive, problems with inherited age, shielding</td>
</tr>
</tbody>
</table>

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on their floodplains. Core samples were collected from a wide variety of channel-floodplain geometries (e.g. meander apex, straight, mobile, immobile), distances from the channel (5–2500 m) and locations (e.g. foredeep, forebulge) across the Beni and Mamore Foreland basins, and the results from approximately 110 of these cores are reported in this paper. But, unlike studies that assume constant initial concentration and a constant sedimentation rate (the CICCS model) from bulk-averaged $^{210}$Pb concentration (and sometimes grain size) for entire cores, we sampled our cores at discrete intervals and at homogenized depths of a few centimetres so that our results yield excess adsorbed $^{210}$Pb activity profiles that are normalized to clay abundance (the carrier of $^{210}$Pb). Such detailed profiles document dates and thicknesses of sediment accumulation [2] and facilitate the evaluation of the CICCS model for large, dynamic and heterogeneous fluvial systems. Apart from documenting the roles of grain size and analytical methodology in characterizing datable excess $^{210}$Pb activity, our core profiles reveal that the CICCS model assumptions generally do not apply to the Bolivian rivers. Consequently, we recommend revised laboratory and analytical procedures to determine clay-normalized absorbed excess (CNAXS) $^{210}$Pb activities and a more flexible set of assumptions for dating.
2. Previous fallout radionuclide dating of floodplain accumulation

A common approach to measuring floodplain accumulation is to determine a datable surface at some depth within the floodplain, with the bomb-derived fallout nuclide $^{137}$Cs the most widely applicable [3–6]. To be sure, such $^{137}$Cs radionuclide geochronology possesses many limitations, chief among them that the flux of both atmospheric and particle-sorbed $^{137}$Cs to floodplains is highly variable, both temporally and spatially at global and local scales. As a result, penetration and inventory [4] methods can be used commonly only to estimate the average accumulation rates, as there are few locations where atmospheric delivery has been sufficiently high and variable to resolve changes in rates or discrete sedimentation events within the 40 year geochronological window. Thus, environments in the Southern Hemisphere often exhibit low levels of $^{137}$Cs activity, owing to patterns of global winds and fallout, so that we have generally been unable to identify $^{137}$Cs in our study area in sediment or river deposits and only in trace amounts in stable upland soils.

An effective alternative is to measure the accumulation profile of $^{210}$Pb, a naturally occurring fallout radionuclide, resulting from the atmospheric decay of $^{222}$Rn, which diffuses from the lithosphere as a member of the $^{238}$U decay series. $^{210}$Pb deposition on land is primarily owing to meteoric fallout [7,8], and it is adsorbed quickly and tenaciously by the surfaces of fine sediments (and associated carbon), primarily onto clays, where, even more so than $^{137}$Cs, it is chemically immobile [9–11]. There it undergoes beta decay to $^{210}$Bi with a half-life of 22.3 years. $^{210}$Pb fallout is generally found to be constant at any given location over time scales relevant to $^{210}$Pb geochronology [6,7,12–15]. However, there are spatial variations in $^{210}$Pb fallout [7], and therefore the rate needs to be characterized for each study region (generally from inventories in soil cores from undisturbed locations). Autochthonous $^{210}$Pb activity produced by the decay of the U-series parent $^{226}$Ra (through the $^{222}$Rn daughter) within the soil or sediment is referred to as ‘supported’ activity, in contrast to ‘unsupported’ excess ($^{210}$XS) $^{210}$Pb activity from meteoric fallout. For any soils, including floodplain locations, $^{210}$Pb activity is derived from the two sources: local atmospheric fallout (‘unsupported’) and in situ production from $^{226}$Ra decay (‘supported’). In addition, floodplain deposits contain river sediment with both supported and unsupported $^{210}$Pb derived from soil erosion in the upstream portions of the catchment (e.g. fallout onto upland soils). It is the latter, often dominant source of XS $^{210}$Pb activity that facilitates measurement of floodplain accumulation rates.

For decades, $^{210}$Pb radioisotope geochronology has been applied to lacustrine [12,13,16,17] and marine [18,19] environments, which often meet the assumptions of mineralogy and grain-size homogeneity and steady-state nuclide fluxes or concentrations underpinning the two common aquatic models: constant rate of supply (CRS) or constant initial concentration (CIC), as summarized in the study of Appleby & Oldfield [12]. However, on floodplains, neither the annual input of XS $^{210}$Pb with river sediment nor the annual accumulation rates can be assumed to be constant [15], because the episodic fluxes of particle-sorbed radionuclides in fluvial systems reflect the strong spatial and temporal variability of overbank flooding [4]. These and other heterogeneities of river-floodplain systems preclude the straightforward application of conventional CRS/CIC $^{210}$Pb models for aquatic geochronology.

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A $^{210}$Pb-dating method for floodplains has been proposed by He & Walling [15], who recognized that unsupported inventories are derived from meteoric fallout at a floodplain site and the catchment-derived input of sediment. They suggested that the fallout inventory ($I_{\text{atm}}$; dpm cm$^{-2}$, at secular equilibrium, where dpm is decays per minute) can be subtracted from the whole-core radionuclide inventory ($I_{\text{total}}$; dpm cm$^{-2}$) to determine the input of river sediment. This in turn reflects the time-averaged accumulation rate, $R$ (g cm$^{-2}$ yr$^{-1}$),

$$R = \lambda_{\text{Pb}} \frac{I_{\text{total}} - I_{\text{atm}}}{A_{\text{catch}}}, \quad (2.1)$$

where $\lambda_{\text{Pb}}$ is the $^{210}$Pb decay constant (0.031 yr$^{-1}$) and $A_{\text{catch}}$ is the assumed constant concentration of $^{210}$Pb (dpm g$^{-1}$) in catchment-derived sediment that can be collected and measured. The reliability of this CICCS model depends on several important assumptions: (i) the mean $^{210}$Pb activity of river-derived sediment is relatively constant during floods (this can be documented), (ii) the accumulation rate is roughly constant over decadal time scales, (iii) atmospheric fallout is constant over decadal time scales, and (iv) the total depth of coring is sufficient to reach background supported $^{210}$Pb activity, such that $I_{\text{total}}$ is properly sampled. The CICCS procedure proved viable in tests at a small, intensively documented location (an approx. 10 000 m$^2$ expanse of grassy floodplain) along the River Culm (channel width of approx. 12 m), UK and other UK rivers [20].

Another study [4] applied CICCS to a much larger, more dynamic and heterogeneous fluvial system by measuring accumulation rates across a large floodplain/delta complex in the Bengal Basin (Bangladesh) using $^{210}$Pb geochronology for 60 cores. Comparing activities against grain size demonstrated that $^{210}$Pb was primarily adsorbed by the clay fraction of sediment, as documented previously [4,10,19,21]. Therefore, they assumed that $^{210}$Pb activity was constant only in clay and revised equation (2.1) to account for natural spatial and temporal variations in the clay content of sediment,

$$R_{\text{pb}} = \lambda_{\text{Pb}} \frac{I_{\text{total}} - cI_{\text{atm}}}{f_{\text{clay}} \rho A_{\text{clay}}}. \quad (2.2)$$

Here, $R_{\text{pb}}$ is the accumulation rate (cm yr$^{-1}$), $f_{\text{clay}}$ the clay fraction of sediment, $\rho$ the bulk sediment density (g cm$^{-3}$), $A_{\text{clay}}$ the assumed constant activity of clay-sorbed $^{210}$Pb in river sediment (dpm g$^{-1}$) and $c$ an empirical focusing factor to account for differences in the floodplain deposition of meteoric $^{210}$Pb. Goodbred & Kuehl [4] found that the clay fraction in floodplain sediment varies significantly across the Bengal Basin and that the modified CICCS approach in equation (2.2) provides good agreement between accumulation rates determined independently with the $^{137}$Cs total penetration method and the $^{137}$Cs inventory method. Their examples of $^{210}$Pb profiles from floodplain locations distal from the main channel often meet the assumptions of the CICCS approach. However, their braidbelt and channel-proximal floodplain $^{210}$Pb activity profiles do not exhibit monotonic declines (exponential or otherwise) in activity to a supported background level, suggesting that use of a simple core-averaged CICCS model might not be
appropriate for dynamic locations where grain size varies significantly, as they do in our Bolivian study area. Furthermore, the customary assumption of CIC of \(^{210}\)Pb in river sediment (or in the clay fraction of that sediment) has not been explored in detail, despite concerns that it may vary significantly between floods [15]. Therefore, the CICCS model might not be appropriate for dynamic, heterogeneous floodplain environments (where CRS/CIC models fail as well).

3. Study area and field methods

The Llanos region of northeastern Bolivia (figure 1) is an excellent locale to study active river-floodplain systems and sedimentation within foreland basins. Besides the availability of well-documented sediment fluxes and a good understanding of the neotectonic setting [22,23], the basin is essentially pristine, without artificial levees, dams, dredging, roads, significant deforestation, cultivation to affect the geochronology or other complications detracting from the study of natural floodplain and fluvial processes.

This study focuses primarily on the Rio Beni between Rurrenabaque and Riberalta (figure 1). At Rurrenabaque, the Beni has a drainage area of 70,000 km\(^2\) and a mean annual discharge of 2200 m\(^3\) s\(^{-1}\) [2,22,23], with flood discharges in excess of 15,000 m\(^3\) s\(^{-1}\) [24]. From an elevation of 190 m at Rurrenabaque, the Beni flows along approximately 800 km of channel as it crosses its foreland basin and descends to about 120 m elevation at Riberalta, where it has a total basin area of 119,000 km\(^2\). The Rio Beni has only one minor tributary, the Rio Madidi, along its entire course, so that there are no significant tributary-related changes in sediment mineralogy or \(^{210}\)Pb concentration along the entire river course. Supporting geochronological measurements are drawn from a parallel investigation along the 1200 km channel of the Rio Mamore, located east of the Beni. To demonstrate how the CIRCAUS technique can be applied elsewhere, calibration examples are presented from three other large river systems: the Ucayali River (Peru), the Mekong River (Cambodia) and the Orinoco River (Venezuela).

Twenty-five survey transects were established across the Beni floodplain in 1998–2000, involving core samples and elevation measurements approximately every 50 m for typical lengths of 200–300 m (an example is depicted in figure 2). These data were combined with system-wide surveys of topography and granulometry and an extensive geographic information system (GIS) analysis of channel migration since 1960 [23,25] to quantify rates of sediment exchange between the channel and floodplain and net sediment deposition within the Beni foreland. Cores and sediment grab samples were collected across the channel and the floodplain at standardized locations along with topographic surveys (figure 2b) across higher floodplain surfaces to avoid the more complicated infilling history possible for local floodplain lakes and depressions [23,26]. A similar number of transects were collected along the Mamore in September 2000, as part of a separate field campaign.

Cores were sampled to a depth of 65–200 cm using a hand sampling probe that collected a 2.5 cm diameter soil core within an internal polyethylene tube. The cutting tip of the soil probe sometimes jammed with a hard conical plug of material, preventing further entry of sediment into the probe and limiting
Figure 2. (a) Example of a channel location map for a Beni floodplain survey site. This site was chosen to measure sediment accumulation rates along a series of five transects spanning an active meander. In the 1999 base image (Landsat ETM+, bands 5–4–3), sand or bare earth is red, vegetation is green and water is blue. Grid spacing is 1 km. Additional GIS detail maps are presented elsewhere [23]. (b) Sketch of a typical survey transect across a Beni River meander apex taken during low-stage conditions (not to scale). Three-letter tags represent standard sampling locations for all transects. To avoid contamination from meteoric fallout of $^{210}$Pb, all grab samples were taken from the bottom of a 10 cm pit or a similar excavation of the bank face. Floodplain cores were collected at intervals of 25 or 50 m, and were surveyed by a differential global positioning system, laser and/or hand-level surveys [23]. (Online version in colour.)

core length. Laboratory X-radiographs show no obvious deformation owing to core shortening (electronic supplementary material, figure S1). ‘Grab’ samples excavated from at least 10 cm beneath the surface (below the characteristic adsorption depth of meteoric $^{210}$Pb) were collected across point bars and down the cutbanks at most transect sites. All samples were sealed in the field and flown back for cold storage.
Prior to cutting for analysis, all floodplain cores were exposed from orthogonal directions with an X-ray machine (Faxitron Cabinet X-ray Model 43855C). These images allowed documentation and evaluation of the sedimentary structures and identification of potential disturbances within cores. After imaging, 110 of the cores were then sampled at 2 cm intervals in the top 20 cm and at greater intervals deeper down, and dried at 50°C. Large organic debris (e.g. twigs, roots, leaves) were removed, although most floodplain samples were free of such material.

One dried 5 g sample from each depth was analysed for granulometry, using a combination of wet sieving at 63 or 125 μm (to determine sand fraction) and detailed analysis of the sand, silt and clay distributions (clay is defined as less than 4 μm) using Micromeritics Sedigraph 5100 particle analysers calibrated for river sediment. We have optimized the laboratory procedures for determination of the clay fraction in river sediment, including: soaking, sonic bath, stirring and drying cycles all in a weak sodium metaphosphate solution to disperse the clay nodules sometimes found in floodplain soils, wet sieving with large volumes of liquid, high volumetric sediment concentrations (approx. 5%) for Sedigraph analysis, duplicate Sedigraph runs (allowing determination of precision and identification of errors), frequent calibrations and a systematic error-checking process that triggers reruns for all questionable samples. Because Sedigraphs measure X-ray opacity of a settling column over time, they determine the mass distributions of very fine particles, including total remaining mass for particles finer than 0.6 μm—this provides precise measurement of the fine fraction available to adsorb 210Pb. The average difference of approximately 1500 duplicate runs was less than 0.5 per cent. 210Pb activity was determined on the second 5g sample with a modified 210Po method [18,19]. A 5g sample was spiked with a known activity of 209Po and subjected to sequential acid leachings (12 N HNO3 followed by 6N HCl, with a ratio of 2ml of acid per gram of sediment sample) brought to near dryness on a hot plate at 90–100°C. The samples were rinsed and centrifuged, ascorbic acid was added, and then ionic polonium auto-deposited onto a silver planchet suspended in a 0.3N HCl leachate for approximately 24h. Resulting samples were counted in alpha spectrometers (Ortec Ultra-AS) for 48–100+ h (as needed to record approx. 1000 decays) to determine the activity of the 210Po granddaughter (which reaches secular equilibrium with 210Pb after five half-lives, in approx. 1.9 yr). Care was taken to maintain consistency during all chemistry steps, especially the spiking procedure. However, our sensitivity tests and duplicate runs suggested that leaching efficiency varies only slightly with hot-plate temperature, acid volume and even number of leaching runs with HCl (see extreme example in table 2). Total analytical error averages less than 3 per cent, substantially better than the ±30–50% uncertainties common for gamma counting of lower 210Pb activities [27,28]. Supported levels of 210Pb were initially estimated from the down-core asymptote of clay-normalized 210Pb activity in numerous terrace and floodplain cores.

For particle-size experiments, grains were separated by wet sieving at 250, 125, 63, 32 and 16 μm, with repeated (three to five times) settling and decanting steps used to separate the 32–16, 16–8, 8–4, 4–2 and less than 2 μm grain sizes. For these experiments, sediment was processed in three ways to remove 210Po: (i) normal leaching, as described above, (ii) approximately five times more of the
Table 2. Total activity of $^{210}$Pb (dpm g$^{-1}$) as a function of grain size and degree of acid leaching. The sample was pumped from the floodwaters of the Rio Beni in 2001. Grain-size separation was achieved through a combination of wet sieving and repeated settling-column separations. Acid leaching is described in §3a. The coarsest size class (>250) contained some particulate organic matter, which is known to contain $^{210}$Pb, which is easily leached.

<table>
<thead>
<tr>
<th>particle size (μm)</th>
<th>normal acid (dpm g$^{-1}$)</th>
<th>5× more acid (dpm g$^{-1}$)</th>
<th>complete HF dissolution (dpm g$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt;250</td>
<td>0.27 ± 0.02</td>
<td>0.38 ± 0.02</td>
<td>0.57 ± 0.07</td>
</tr>
<tr>
<td>125–250</td>
<td>0.25 ± 0.01</td>
<td>0.29 ± 0.01</td>
<td>0.75 ± 0.08</td>
</tr>
<tr>
<td>63–125</td>
<td>0.37 ± 0.02</td>
<td>0.39 ± 0.02</td>
<td>0.97 ± 0.12</td>
</tr>
<tr>
<td>31–63</td>
<td>0.44 ± 0.03</td>
<td>0.54 ± 0.03</td>
<td>1.64 ± 0.20</td>
</tr>
<tr>
<td>16–31</td>
<td>0.50 ± 0.03</td>
<td>0.67 ± 0.05</td>
<td>1.82 ± 0.19</td>
</tr>
<tr>
<td>8–16</td>
<td>0.57 ± 0.06</td>
<td>1.03 ± 0.08</td>
<td>2.16 ± 0.22</td>
</tr>
<tr>
<td>4–8</td>
<td>0.73 ± 0.09</td>
<td>0.95 ± 0.13</td>
<td>2.19 ± 0.25</td>
</tr>
<tr>
<td>2–4</td>
<td>1.44 ± 0.19</td>
<td>1.48 ± 0.38</td>
<td>2.19 ± 0.27</td>
</tr>
<tr>
<td>&lt;2</td>
<td>1.88 ± 0.11</td>
<td>1.91 ± 0.13</td>
<td>2.18 ± 0.23</td>
</tr>
</tbody>
</table>

same acids (per unit sample weight—an extreme test of leaching sensitivity), and (iii) a mixture of hydrofluoric (HF), perchloric, nitric and hydrochloric acids was used to achieve full dissolution. All corrections for radioactive decay were made to account for the time intervals between sample collection in the field, leaching and $^{210}$Po plating, and alpha counting. One-sigma error bars were calculated for all activities. Direct gamma assays of $^{210}$Pb, $^{226}$Ra/$^{222}$Rn ($^{214}$Pb daughter) and $^{137}$Cs activities were also measured for approximately 60 samples with two calibrated LEGe detectors (Canberra model GL2020R), using attenuation corrections for the low-energy $^{210}$Pb gamma ray (46.52 keV) [29]. Replicate gamma counting was done for key profiles using ultra low background well detectors in the Exeter Radiometry Laboratory, which features ancient lead shields with cadmium liners, Ortec HPGe GWL XLB detectors with remote pre-amplifiers, stabilized digital electronics and analysed with GAMMAVISION and ANGLE software.

(b) Distinguishing adsorbed excess $^{210}$Pb activity: a revised approach

A major impediment to comparing $^{210}$Pb geochronology from different studies is that two fundamentally different techniques are used to determine total and XS activity: direct gamma counting of $^{210}$Pb decay and alpha counting of the $^{210}$Po grand-daughter. Gamma counting, which has been favoured by some researchers [12], records all $^{210}$Pb activity in the sample, the bulk of which may be locked well within the mineral lattices. In contrast, the $^{210}$Po method measures only the $^{210}$Pb activity of leachable sites (a function of chemistry). The term ‘adsorbed’ reflects the history of fallout $^{210}$Pb that was adsorbed onto the surface of a particle and the resulting $^{210}$Po that can be desorbed in a consistent way through controlled acid leaching. It is this adsorbed $^{210}$Pb that supplies the XS $^{210}$Pb activity used for dating, whereas mineral-locked activity represents only noise that must be quantified and subtracted.
The $^{210}$Po alpha-counting method offers practical advantages over gamma measurements, especially a smaller sample size (0.5–3 g) that allows for fine sectioning of cores, exceptionally low background noise, no problems with attenuation, reduced expense and simple calibration requirements. The challenge is to (i) consistently leach the adsorbed radionuclide and to (ii) characterize the supported $^{210}$Pb activity from \textit{in situ} decay of $^{226}$Ra adsorbed to the same sediment.

First, we explored the sensitivity of leaching efficiency as a function of grain size and chemical procedure for representative samples of Beni River sediment collected from floodwaters (one example is presented in table 2). In this experiment, the HF dissolution represents \textit{all} of the $^{210}$Pb activity in the sample, including the mineral-lattice-locked activity—these measurements are analogous to results of gamma counting. Silt and clay contain approximately the same total $^{210}$Pb activity, and sand contains two to four times less. However, only a fraction of the silt and sand activity (25–33\%) is released by the normal leaching procedure, with much more of the activity (66–86\%) released from the clay. Hence, the clay fraction contains most of the adsorbed $^{210}$Pb component, while the silt and sand contain mostly mineral-locked $^{210}$Pb that is not readily exchangeable. These results agree with the earlier studies [4,10,19,21] to underscore the importance of clay as the primary carrier of $^{210}$Pb activity (differences in our methods and sediment mineralogy preclude direct comparison). Our results also establish that the concentration of autochthonous, mineral-locked $^{210}$Pb is overwhelmingly larger (two to three times) than the datable sediment-adsorbed component. Gamma counting is poorly suited for Beni River sediment because of this large component of noise. Instead, we employ the $^{210}$Po alpha-counting technique to isolate the adsorbed $^{210}$Pb activity from the dominant mineral-locked activity. We also ran a third set of samples using approximately five times as much nitric and hydrochloric acid as in our normal technique (table 2). The increased $^{210}$Po leaching in response to this large volumetric increase in acid is small, suggesting that our standardized methodology for desorbing $^{210}$Po produces a consistent degree of leaching, despite variations in the grain-size distributions between samples. After conducting many similar leaching tests across a range of sediment over the past decade, we developed a procedure for leaching with nitric and hydrochloric acids that provides consistent extraction of adsorbed $^{210}$Po—hence, our procedure defines ‘adsorbed’ activity. Replicate samples from many rivers show variation of less than 5 per cent, close to our computed analytical error of approximately 3 per cent (the difference is primarily owing to sub-sampling error).

Second, we need to determine the supported activity for adsorbed $^{210}$Pb owing to local decay of the $^{226}$Ra/$^{222}$Rn/$^{214}$Pb parents near the surface of the sediment (e.g. adsorbed). This differs from the total supported activity for all $^{210}$Pb contained in a sample (as is determined by gamma spectrometry)—rather, we wish to ignore the activity of mineral-locked $^{226}$Ra/$^{222}$Rn within the crystal lattices that often dominates the signal (e.g. the mineral-locked component in table 2). What needs to be determined is how much adsorbed $^{210}$Pb activity is produced by $^{226}$Ra that is also adsorbed to mineral surfaces [30]. We measure clay-normalized supported activities directly from down-core asymptotes in $^{210}$Pb activity profiles determined with alpha counting [4,19]. This asymptote technique works best in the case of a constant supported \textit{clay-normalized} activity, meaning that local $^{226}$Ra/$^{222}$Rn activity must solely be due to clay content. Because...
Figure 3. Activities (decays per minute per gram) from (a) Beni and (b) Mamore Rivers. All core sample depths that meet two conditions are shown: (i) data are from zones of constant activity, as defined by at least three points of the same value, and (ii) sample depth must be greater than 25 cm, below which the effects of radon ventilation and XS activity from fallout ²¹⁰Pb are minimal. Solid boxes represent terrace cores, which are uplifted blocks of old floodplain that have not received sediment for centuries (and hence have only supported ²¹⁰Pb activity). Open circles are samples from the active floodplains, where most of the ²¹⁰Pb activity ‘plateaus’ have elevated, datable ²¹⁰Pb XS activity. Some floodplain samples approach the lower bound of supported activity, the envelope represented by the fitted terrace lines (solid). Solid vertical ‘initial–final’ system dating lines represent deposition and ageing trajectories for sediment with 60% clay (‘initial’ varies downstream). Other rivers show a similarly robust linear relationship between per cent clay and supported activity for old terrace sediment [31], although the slope of the fit changes between systems and there is not always a zero intercept. Three typical examples of this relationship between supported activity and per cent clay are presented for other tropical river systems: (c) the Ucayali River, Peru; (d) the Mekong River, Cambodia; and (e) the Orinoco River, Venezuela.
Figure 4. Beni floodplain cores depicting clay-normalized total adsorbed $^{210}$Pb alpha-counted (α) activity (decays per minute per gram clay) and grain size (clay, sand), and interpretation of features for CNAXS geochronology. Supported activity for all Beni cores averages 1.34 dpm g$^{-1}$ clay. (a) Site 3-A, located 1200 m from the channel when the sediment was deposited in 1954 (±2.0 years). Cap date is 1953. X-ray depicts fine laminations throughout the core. Core illustrates the typical case of a single sediment floodplain accumulation event several decades old, with no additional sediment accumulation since, and with the ‘ingrowth’ of a meteoric cap into the top 10 cm. (b) Site 3-B, located 1150 m from the channel when the sediment was deposited in 1957 (±1.8 years). Cap date is 1956. X-ray depicts fine laminations throughout the core. Core is similar to the nearby site 3-A, illustrating the general observation that nearby cores record the same event(s). (c) Site 60RFC-250, located 2000 and 2900 m from the channel when the sediment was deposited in 1974 (±1.8 years) and 1951 (±1.9 years) (the channel migrated approx. 900 m towards the coring site during the time between these two dates). Top meteoric cap is incomplete (one point with effective age 1990), and lower meteoric cap is missing. X-ray depicts fine laminations throughout the upper core, with some minor cross-bedding below 80 cm depth. This figure illustrates an example of a core with two plateaus (some of which have buried meteoric caps). Both the approximately 1973 and 1950 events are recorded throughout this survey site (depicted in figure 2a and in electronic supplementary material, table S1, transects 60–64). (Online version in colour.)

Clay accounts for most of the mineral surface area and metal ($^{210}$Pb and $^{226}$Ra) adsorption capacity in river sediment, a plot of adsorbed $^{210}$Pb activities for river deposits (without meteoric caps) versus clay fraction (figure 3a,b) characterizes the range of possible $^{210}$Pb concentrations in a system. The lower limit to this envelope is defined by samples from uplifted terraces and the oldest floodplain cores (more than 100 years old), which illustrate how supported adsorbed $^{210}$Pb activity (with no XS activity—solid boxes in figure 3a,b) varies as a function of clay content. Supported values can thus be calculated as a linear function of clay content and subtracted from the total adsorbed $^{210}$Pb activity to determine the XS $^{210}$Pb activity required for geochronological calculations.

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Figure 5. (a) Site CB-1 exhibiting constant accumulation. As supported $^{210}$Pb activity of 1.34 dpm g$^{-1}$ clay is not reached in this core, the CICCS model would underestimate the sediment accumulation rate. Line Mcs represents the best-fit CIRCACS model (CIRCA with ‘constant sedimentation’ and meteoric fallout), the predicted activity of clay-adsorbed $^{210}$Pb at the accumulation rate of 1.9 cm yr$^{-1}$. Less than 5% of cores can be interpreted as constant sedimentation (electronic supplementary material, table S1). The river has been migrating towards this site over the past 50 years so that the grain size in this core coarsens upwards—as a result, the $^{210}$Pb activity per gram total increases downwards, the inverse of the profile seen for clay-normalized activity (hence the need to normalize). Also plotted is clay-normalized XS $^{210}$Pb activity determined with gamma counting ($\gamma$). The trends are much poorer because of the inaccuracies of gamma counting and because mineral-locked $^{210}$Pb dominates our sediment (table 2). (b) Site CB-2, exhibiting a single deposition event of $\sim$160 cm dating to 1987 (±1.9 yr), due to observed channel migration. Meteoric cap date is 1993. Gamma XS $^{210}$Pb activity is not useful. ‘CB’ sites are located between sites 1 and 60. (Online version in colour.)

Conceptually, for radionuclide dating based on this diagram, packets of floodplain sediment are deposited with specific clay contents and XS $^{210}$Pb activities determined by local transport processes and supply from upstream (e.g. the open circles in figure 3a, b), as illustrated by the solid ‘initial–final’ lines.

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(‘initial’ values vary with downstream location, as we discuss later). Over time, the adsorbed $^{210}\text{Pb}$ activity of this sediment decreases without a corresponding change in clay fraction, resulting in a vertically downward trajectory of ageing along the depicted system line. This descent continues until the value intercepts the lower bound of supported adsorbed $^{210}\text{Pb}$ activity, defined by the solid ‘terrace’ lines in figure 3a,b. The adsorbed $^{210}\text{Pb}$ activities plotted for younger floodplain sediment will define a data cloud roughly parallel to and bounded below by the terrace-activity line that has no XS$^{210}\text{Pb}$ activity (open circles in figure 3a,b).

For the Rio Beni and Rio Mamore sediments, the intercepts of these supported activity lines are essentially zero, meaning: (i) in situ production of adsorbed $^{210}\text{Pb}$ is solely a function of clay abundance and (ii) the clay-normalized supported activities for these systems are uniform values that can be determined from the slopes of the lines (for Beni with zero intercept—1.34 dpm and for Mamore—1.54 dpm). Application of the technique to other river systems requires the construction of plots similar to figure 3a,b to ascertain how supported adsorbed $^{210}\text{Pb}$ activity can be defined as an empirical function of clay content. We generated such calibration curves for the Strickland [31] and Sacramento [32] Rivers, and here we present new examples of adsorbed $^{210}\text{Pb}$ versus clay for the Ucayali (figure 3c), Mekong (figure 3d) and Orinoco (figure 3e) Rivers. Other river systems can have non-clay-associated sources of $^{226}\text{Ra}/^{222}\text{Rn}$ (e.g. silt), meaning that the terrace lines have non-zero intercepts and smaller slopes.

Alpha counting with the $^{210}\text{Po}$ method generally provides smooth, interpretable profiles (figure 4) because it isolates and measures only the adsorbed $^{210}\text{Pb}$, without the considerable noise associated with: (i) the majority of $^{210}\text{Pb}$ and $^{226}\text{Ra}/^{222}\text{Rn}$ activity that is locked deep within the mineral lattices and (ii) the calibration and attenuation errors characteristic of gamma counting. The XS $^{210}\text{Pb}$ activity profiles that we have measured with gamma counting for Beni River sediment (examples are presented in figure 5) are generally noisy and difficult to interpret because of low activities and high grain-size heterogeneity [28] (perhaps, an explanation of why profiles were not published for a recent study that used gamma counting on seven Beni cores [33]). Because the high mineral-locked $^{210}\text{Pb}$ and $^{226}\text{Ra}/^{222}\text{Rn}$ contents of coarse Bolivian sediment overwhelm the adsorbed components (table 2), effective gamma counting for XS $^{210}\text{Pb}$ activity would require extremely long count times on special ultra low noise gamma spectrometers, tying up costly equipment. In contrast, the method described for alpha counting only the adsorbed $^{210}\text{Pb}$ provides a rapid and affordable means for constructing low noise activity profiles.

4. Clay normalization and the limitations of constant initial concentration and a constant sedimentation rate

For the floodplain locations surveyed across northern Bolivian Llanos, X-radiographs (electronic supplementary material, figure S1) for most cores (150 Beni and 120 Mamore imaged to date) typically indicate a moderate-to-low-energy depositional environment [23]. Core structures generally exhibit fine horizontal laminations and other micro-structures at a sub-millimetre scale, suggesting that the floodplain sediment has not been significantly disturbed.

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since deposition and that bioturbation is negligible at most locations. The rainforest trees have shallow roots, are spaced far apart and are rarely blown over (i.e. they generate limited soil mixing by tree throw). There is scarce leaf litter on the forest floor, and we have never seen evidence of infauna or epifauna that would mix the soil. The impressive numbers of tree-dwelling termites may devour all leaf litter. Because of such termites and rapid decomposition of all dead material, there are no organic litter layers, which have been found elsewhere to contain much of the XS \(^{210}\text{Pb}\) activity delivered by meteoric fallout, especially in temperate settings dominated by conifers [34,35]. The general lack of mixing and penetration of organic debris into the floodplains minimizes the complicating effects from the strong association of \(^{210}\text{Pb}\) with organic matter [36]. Hence, the floodplain material across our study basins consists mostly of undisturbed sediment deposits, and our determination and interpretation of \(^{210}\text{Pb}\) activity profiles are not complicated by crop ploughing, grazing or bioturbation.

For many of the floodplain cores cut and analysed for \(^{210}\text{Pb}\) geochronology (80 for Beni and 30 for Mamore reported here), grain size changes significantly over the depth of the core. Because \(^{210}\text{Pb}\) activity is almost entirely contained within the clay fraction of sediment (table 2 and figure 3; [4]), the sediment activity \((A_{\text{sed}})\) must also change in proportion to clay. For the case of a dynamic river that migrates across its floodplain, the clay fraction within the floodplain sediment is roughly a function of the distance from the channel—therefore, as the river migrates towards a particular floodplain location, deposited sediment will become increasingly coarser upward (figure 5). Because the Beni River has migrated towards many of the sample sites (e.g. cutbank side) over the past few decades [23] and because of heterogeneous floodplain sedimentation in general (see other profiles), many cores exhibit a considerable variation in clay fraction with depth. Therefore, the core-averaged clay-normalization strategy depicted in equation (2.2) is not applicable to such a texturally variable environment.

For example, consider the case of two 100 cm floodplain cores, the first with a uniform clay fraction of 50 per cent, and the second with 20 per cent clay for the top 50 cm and 80 per cent clay for the bottom 50 cm (both having a bulk-averaged clay content of 50 per cent over the entire core). For both cores, assume that the top 50 cm contains clay with an unsupported \(^{210}\text{Pb}\) activity of 6 dpm g\(^{-1}\), the bottom 50 cm contains clay with an activity of 3 dpm g\(^{-1}\), and below the 100 cm core depth, the unsupported activity is zero. Following a normalization procedure equivalent to that expressed in equation (2.2), the first core would have a bulk sediment activity of 2.25 dpm g\(^{-1}\), and a clay-normalized \(^{210}\text{Pb}\) activity of 4.5 dpm g\(^{-1}\) clay. The second core exhibits a much lower bulk activity of 1.8 dpm g\(^{-1}\) and a clay-normalized \(^{210}\text{Pb}\) activity of 3.6 dpm g\(^{-1}\) clay, which would result in a significantly lower average accumulation rate for the 100 cm interval according to either equation (2.1) or equation (2.2). However, both these cores have identical radiometric sedimentation histories: 50 cm of sediment was deposited, followed by a hiatus in sedimentation during which the \(^{210}\text{Pb}\) activity decayed, and then a second 50 cm of sediment was deposited. Although the preceding example is for a simplified case of two sedimentation events, cores containing many events (quasi-annual sedimentation) would exhibit the same fundamental problem with bulk averaging, if the clay fraction of the sediment varies continuously throughout the core.

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One approach that might account for variations in sediment clay fraction is to use a discrete normalization procedure for each separate sample depth, with all reported sediment activities, $A_{\text{csed}}$, normalized to $f_{\text{clay}}$, the associated sediment fraction less than 4 µm (e.g. $^{210}\text{Pb}$ activities are reported as dpm g$^{-1}$ clay),

$$A_{\text{csed}} = \frac{A_{\text{sed}}}{f_{\text{clay}}}.$$  

(4.1)

This concept is akin to considering that clay is the only subject of $^{210}\text{Pb}$ radionuclide geochronology, with the sand and silt travelling along without contributing substantially to the activity of adsorbed $^{210}\text{Pb}$. Given the very strong affinity of $^{210}\text{Pb}$ for clay particles [4,21,31], the discussion in §3, and the lack of floodplain disturbance in the Beni Foreland, it is appropriate to use clay as a datable tracer suspended within a matrix of coarser material. This approach and the determination of $^{210}\text{Pb}$ activity at discrete floodplain depths offer a flexible and robust approach for dating sediment within river floodplains because it both provides correction for heterogeneous core grain size and presents full representation of discrete depositional events. Without normalization, many profiles exhibit down-core increases or considerable variation (uninterpretable noise) in $^{210}\text{Pb}$ activity (per gram sediment), which in lacustrine environments could be mistaken as evidence for a CRS signal typical in locations where either accumulation is highly variable or all $^{210}\text{Pb}$ activity is supported [12]. Hence, all cores were processed with this clay-normalization procedure to offer the best possible insight into the processes and rates of floodplain accumulation. Because of the ease of the XS $^{210}\text{Pb}$ activity determination for the Rios Beni and Mamore (simply subtract a constant value for supported activity), for simplicity, all plots in this paper portray total adsorbed $^{210}\text{Pb}$ activity. In such depictions of total activities, the XS $^{210}\text{Pb}$ activity is equal to the total activity minus the supported activity of 1.34 dpm g$^{-1}$ clay for the Beni, 1.54 dpm g$^{-1}$ clay for the Mamore (figure 3a, b).

Of the 110 Beni and Mamore cores processed, only five show evidence for constant sedimentation (electronic supplementary material, table S1). Most cores exhibit one or more regions of relatively uniform clay-normalized $^{210}\text{Pb}$ activity, termed ‘plateaus’, separated by abrupt changes in activity (figure 4). The top 5–15 cm of many cores exhibit a considerable increase in activity, a ‘meteoric cap’ owing to the fallout of atmospheric $^{210}\text{Pb}$ in the absence of continued sedimentation (discussed later). With just one or two discrete depositional events accounting for the entire core depth over periods typically spanning several decades, it is rarely appropriate to consider floodplain sedimentation as constant. Furthermore, only two of the floodplain cores reach the supported background activity of 1.34 dpm g$^{-1}$ clay. This indicates high sediment accumulation rates across the Beni Foredeep [2,23], meaning that the cores (65–200 cm long) were usually not deep enough to reach background supported activity. As such, even in the case of uniform down-core texture, any attempts in our study area to determine $I_{\text{total}}$ by bulk averaging and application of the CICCS method would result in substantial underestimation of the average annual accumulation rates.

There are a few floodplain locations sampled within the Beni and Mamore Foreland that do indeed appear to be characterized by quasi-annual sedimentation. For example, figure 5a depicts a site that has received approximately 1.9 cm of sediment annually over the recorded depth of 160 cm.
However, in this case, the CICCS model still would not apply because the clay fraction declines up-core by a factor of 5 and the lowest $^{210}$Pb activity (2.4 dpm g$^{-1}$ clay) is well above the supported background. Despite these complications, the observed activity profile can be reconstructed using the discrete-depth approach (the line shown in figure 5), given the following assumptions: (i) sediment accumulation rate is constant, (ii) the $^{210}$Pb activity of clay in freshly deposited river sediment is constant over time for that particular reach of channel (6.5 dpm g$^{-1}$ clay here), and (iii) the meteoric fallout of $^{210}$Pb is constant (44 ± 2 dpm cm$^{-2}$ documented for many locations throughout the Beni Foreland), and is adsorbed by the surface sediment within a depth interval of 5 cm (a typical adsorption window observed in cores). This approach, which we call the constant initial reach clay activity and constant sedimentation model (CIRCACS), provides a good fit for the few cores that exhibit steady monotonic (exponential in this case) decline in $^{210}$Pb activity. Therefore, the assumption of constant sediment accumulation is appropriate in some locations, but a more labour-intensive approach than CICCS is needed to measure and model the resulting clay-normalized $^{210}$Pb activity profiles, especially for the most common case we have found of floodplain environments with non-steady accumulation.

5. Results of the river sediment survey: initial concentration variable, decreases downstream

Previous floodplain models assume that the input concentration of $^{210}$Pb is constant. As recognized by Goodbred & Kuehl [4] and previously discussed, $^{210}$Pb activity must be normalized to clay abundance at each site to account for spatial variations in deposit grain size. Here, we have demonstrated that the constant sedimentation assumption fails for the Rios Beni and Mamore, and that the clay adjustment must also be made down-core for each discrete depth, so as to account for temporal variations in the clay abundance of river sediment conveyed to each floodplain site. We next investigate how the clay-normalized activity varies both spatially and temporally for fresh river sediment, to ascertain if initial concentration in clay is indeed constant everywhere.

Figure 6 depicts activity measurements for fresh river sediment, collected from a bar within the channel along the northward-flowing Rio Beni (i.e. plotted versus universal transverse mercator (UTM) projection latitude). This sediment was most likely deposited during the late falling stages of the March–April 1999 flood, the largest on record. The activity ranges substantially in value and appears to decline downstream across the foreland basin. This trend is more evident for the grab samples taken from the tops of point bars (black circles in figure 6, from the vegetated ‘VB’ locations in figure 2b). These locations would receive sediment during earlier parts of the flood, when conditions are probably more homogeneous owing to continued erosion throughout the watershed and vigorous mixing that buffers system variations. The open circles represent samples collected from topographically lower locations on the bars, which are inherently more variable because of the vagaries of sediment input by bank collapse during the later falling river stage. Three large sediment samples were collected from the water during a moderate stage event in 2001 (below bank-full flooding), and depict no obvious downstream change in activity (probably because there
Figure 6. Down-channel transect for the $^{210}$Pb activity of clay in fresh Beni River sediment sampled from the channel, plotted versus UTM latitude (figure 1; note that the Beni flows roughly north). Left side of graph (8400 km UTM latitude) is at the sub-Andean range front, centre is forebulge (8610–8680 km UTM latitude) and right side (8780 km UTM latitude) at the confluence with the Madre de Dios River. Black circles are for grab samples taken from vegetated, topographically higher locations at the edge of the channel that receive sediments only during bank-full flooding. Open circles represent grab samples taken at topographically lower locations that receive sediment during the lower, waning stages of floods. Squares represent the activity of sediment filtered from the river water, which was collected during a single moderate-stage event that was just below bank-full discharge (February 2001). Asterisks represent activities estimated from fresh floodplain deposits (core tops from 5–50 m into the forest) and meteoric-cap-date matching using the empirical CIRCA technique (discussed later). The two key points here are that (i) the customary assumption of constant initial concentration is not necessarily valid, especially for lower, in-channel deposits (higher floodplain deposit concentrations appear to be more regular) and (ii) there is evidence for a downstream decline in activity.

was limited sediment recycling by channel migration or bank collapse at that time). Finally, the asterisks portray the clay-normalized $^{210}$Pb activity as averaged from floodplain core tops. The values are selected to decrease monotonically downstream using the empirical CIRCA technique (discussed in §5a). These deposits record $^{210}$Pb concentrations in river sediment that is conveyed onto the floodplains during infrequent rapid-rise floods of extraordinary magnitude [2,23], which epitomize system mixing and therefore characterize the most uniform conditions for radionuclide supply. These floodplain deposits are a primary source of the clay-rich sediment later re-supplied to the river by channel migration, which along with falling-stage bank collapse supplies much clay with low XS $^{210}$Pb activity to produce a variable falling-stage signal.

These data sources suggest that clay-normalized $^{210}$Pb activity of the suspended sediment in transport is neither uniform nor constant for the Beni River system, but instead varies substantially with river stage and appears
to exhibit a downstream decrease over 800 km of channel. Even the more homogeneous scenario of a large, rapid-rise flood that emplaces extensive floodplain deposits does not support the assumption of CIC of $^{210}$Pb for fresh sediment throughout the system. And, the lower stages exhibit far more temporal and spatial variability. Therefore, for dynamic river systems like the Rio Beni, a more sophisticated approach is needed to account for the processes that supply sediment and thereby to estimate the clay-normalized $^{210}$Pb activities of sediment conveyed to floodplains during large floods. We present three enhanced procedures that can be used to interpret detailed $^{210}$Pb activity profiles.

(a) Procedure 1: estimating a constant initial ‘reach’ clay activity

To develop a conceptual geochronological model appropriate for rivers and their floodplains, we seek to model the processes responsible for the conveyance and exchange of clay-associated radionuclides. The key to most effective methods of radionuclide geochronology is determining the initial concentration. Figure 6 suggests that, despite some noise for the in-channel falling-stage grab samples, a down-channel decrease in activity is clearly reflected in the overbank sediments. The following is a thought experiment for the dynamics of $^{210}$Pb concentration in sediment during floods, to explore the common ‘CIC’ assumptions for sediment conveyed to rivers (and lakes).

As quantified in earlier studies [23, 25, 26], the Beni River migrates across its floodplain, excavating enormous quantities of sediment from the cutbank side of meanders and re-depositing much of this material onto the point bars (electronic supplementary material, figure S3). The significance of this exchange process has implications for the down-channel budget of clay-associated $^{210}$Pb because the cutbanks contain material that is radioactively ‘older’ (less XS activity) than the river sediment. Therefore, the process of rapid channel migration during floods implies a continuous downstream dilution of the $^{210}$Pb activity of riverborne clay [23]—a process related to that proposed to explain an observed decline in $^{210}$Pb activity in the sub-aqueous environment of the Wilmington Submarine Canyon [37] and recently developed in more theoretical detail in the study of Lauer & Willenbring [38]. Because we have estimated (i) these migration fluxes from field and image surveys [23], (ii) average clay abundances for all types of river deposits, and (iii) the average radionuclide content of the cutbanks across the Beni Foreland (electronic supplementary material, figure S4), we can calculate an approximate budget of clay and the associated downstream decline in $^{210}$Pb activity for the Beni River.

We assume that the input concentration of $^{210}$Pb from the Andes at Rurrenabaque is relatively constant during the largest floods. However, there is undoubtedly some variation in this input activity depending on the source of the sediment eroded upstream in the source basin and the storm rainfall distribution. For example, during the early rains, erosion of surface soils high in $^{210}$Pb activity (adsorbed from meteoric fallout) is likely to dominate the signal, especially from areas with thin vegetation cover. This mechanism is supported by field observations of erosion and records from gauging stations throughout the basin [22, 39], which tend to show highest suspended-sediment concentrations during the early rains (the significance of surface erosion diminishes later in the season). As the rainy season progresses, hill slopes become more saturated,
leading to deeper mass wasting of colluvium, and river stages rise, remobilizing previously stored alluvium. These large volumes of stored material, which generally have lower activities (because they are shielded from fallout $^{210}\text{Pb}$ and are geochronologically older), would both serve to decrease and to stabilize the $^{210}\text{Pb}$ concentration at Rurrenabaque. This homogenization effect would probably be greatest at the height of the wet season during infrequent large storms, when all hillslope and fluvial processes are vigorously active. At such a time, the myriad heterogeneous $^{210}\text{Pb}$ contributions of individual failures would integrate and homogenize supply, as sediment discharges approaching 70 Mtonnes d$^{-1}$ occur at Rurrenabaque [2,23].

Once the flood passes, the sediment discharge then drops back to levels where individual hillslope failures and other stochastic processes could affect the concentration of $^{210}\text{Pb}$ in river clay at Rurrenabaque. This concept for the homogenization of $^{210}\text{Pb}$ activity during peak discharges is in agreement with observations of process and available data from floodplain deposits. In particular, the timing of floodplain sedimentation is most likely on the rising limb of large ‘rapid-rise’ floods from these large, episodic storms [2], when the XS $^{210}\text{Pb}$ concentrations from eroding soils would be expected to be high and homogenized. Near Rurrenabaque, channel sediment samples illustrate that the input concentration varies from 5.0 to 7.5 dpm g$^{-1}$ clay (figure 6), and an average value of 6.5 dpm g$^{-1}$ clay during the largest floods is a close match to most of the floodplain samples (figure 7). We believe, based on the verified dates in our floodplain cores in table S1 and the relatively constant activity levels in most floodplain ‘plateaus’ (discussed in §5b), that sediment deposited by extreme floods has a limited range (approx. 0.5 dpm g$^{-1}$ clay) of initial activities for a given reach.

Assuming this clay-normalized activity is exported from the Andes and applying the mass fluxes determined in the study of Aalto [23,25] (electronic supplementary material, figure S3) along with their associated clay content and the measured $^{210}\text{Pb}$ activity of the cutbank deposits (electronic supplementary material, figure S4), it is a straightforward accounting exercise to quantify the exchange of clay within each river reach of 10 km UTM latitude (electronic supplementary material, figure S5). The volume of material eroded from the cutbank by channel migration is multiplied by its associated density and clay fraction to determine the mass of clay returned to the channel. The mass of clay deposited within the 10 km UTM latitude reach is calculated in a similar way for sediment deposition on the bar, bed and floodplain. Because the channel migration, sediment input and floodplain deposition rates are decadal averages [23], we assume that all these process rates during flooding exhibit approximately the same ratio as their long-term averages—the decade-averaged values for sediment discharge at Rurrenabaque and the processes of floodplain deposition, sediment input and channel migration are known to be dominated by extreme floods [2]. We calculated ‘flood’ clay fluxes for any particular 10 km reach and measured the activity of the eroded cutbank clay (electronic supplementary material, figure S4). Assuming that all deposited clay in a reach has the same activity as the clay input from immediately upstream, the resulting clay flux out of the reach (electronic supplementary material, figure S5) and its associated $^{210}\text{Pb}$ activity can be determined (figure 7). The predicted downstream decline in $^{210}\text{Pb}$ activity closely matches that determined empirically from the floodplain.
Figure 7. The $^{210}\text{Pb}$ activity of Beni clay modelled at decreasing rates of river migration and floodplain (FP) deposition. Values are obtained by multiplying clay fluxes in electronic supplementary material, figure S5 by measured $^{210}\text{Pb}$ activity depicted in electronic supplementary material, figure S5. Errors of approximately 20% are expected.
The challenge of the modelled CIRCA approach is that many systems do not have a longer record of sediment fluxes like the Rio Beni, not to mention the additional effort required to measure migration rates, channel geometry, and cutbank activities and grain sizes throughout the system. Also, the effects of sediment contributed by ungauged tributaries must be considered, although this is not a problem for the Beni. In most cases, there is a simple empirical means by which to estimate the down-channel decrease in $^{210}$Pb activity during large floods. First, one must establish some radionuclide boundary conditions with fresh sediment samples from in-channel locations deposited during a recent flood, or preferably, from river sediment filtered directly from floodwaters. While potentially useful, near-channel floodplain samples may not always represent recent sediment deposition because floodplain sedimentation can be very heterogeneous and even rare at a site [2]. Second, a consideration of basin-scale channel mobility will elucidate the contribution of channel migration in terms of an expected down-channel decline in $^{210}$Pb activity. For stable channels, no significant decline would be anticipated. For more mobile channels, the CIRCA can be expected to decline monotonically downstream, at a rate directly proportional to the channel migration velocity that orchestrates mixing rates for floodplain material with lower XS $^{210}$Pb activities. The significance of any tributaries must also be considered during this step. In particular, one must assess whether the input clay activities and fluxes are sufficient to alter the clay-normalized $^{210}$Pb activity of the main channel.

The third and final step is to collect floodplain cores throughout the study area and produce and interpret activity profiles (see §5b). There is a narrow range of input concentrations for $^{210}$Pb that both match peak flood sediment deposits and provide dates that are consistent with independent means of dating (typically available for some cores). In this study, we have compared the dates of discrete sedimentation events with the independent dates of meteoric caps (discussed later), deposits of known age (from images and ground records) and a 40 year discharge record, but $^{137}$Cs geochronology could be used elsewhere where there is sufficient $^{137}$Cs activity. During our extensive fieldwork, we identified many floodplain locations that had obviously received fresh river sediment during the record flood of the preceding season—these cores provided a constraint for the $^{210}$Pb values of river sediment in those reaches (and by the ‘monotonic decline’ rule, also in nearby reaches).

In practice, the first two principles will provide a relatively narrow range of possible $^{210}$Pb activities for each reach, which the third step of calibrating to known core dates will further constrain to produce an empirical estimation of CIRCA. Because the same uniform values are used consistently for all cores within each reach and these values must decline smoothly downstream in response to documented channel migration, there is little risk of ‘fine-tuning’ the geochronology results on a core-by-core or even reach-by-reach basis. Furthermore, the more floodplain cores that are analysed, the less significant the effect of any specific core. For the Rio Beni, this empirical CIRCA method produces values that match the modelled CIRCA method (figure 7) and agrees with several independent means of dating (discussed later). For the Mamore, only the empirical CIRCA calibration has been used (figure 8), which suggests that the Mamore system exhibits no major downstream declines in CIRCA. This uniformity is probably due to two important differences between the rivers: (i)
Figure 8. Down-channel transect for $^{210}\text{Pb}$ activity of clay in Mamore River sediment plotted against UTM latitude (figure 1); symbols same as figure 6. Asterisks represent uniform down-channel activity as determined from fresh floodplain deposits (cores) and meteoric-cap-date matching using the empirical CIRCA technique. There is no significant downstream decline in activity because Mamore channel migration rates are much lower than the Beni, and tributary input is a significant source of clay with high $^{210}\text{Pb}$ activity (e.g. the Itenez River contributes clay with activity greater than 7 dpm g$^{-1}$). This example illustrates how even adjacent river systems (with similar climate and sediment sources) can have different CIRCA systematics.

channel migration rates are much slower for the Mamore than the Beni, resulting in a less significant exchange of radiometrically old sediment for new, and (ii) the Mamore receives small amounts of clay with high $^{210}\text{Pb}$ activity from dozens of lowland tributaries (basins characterized by surface erosion of high $^{210}\text{Pb}$ activity soils), compensating in part for the activity losses owing to sediment exchange during channel migration. Such modelled and empirical estimations of CIRCA during flooding are not without significant remaining questions about the exact variation of activity in rivers as a function of flood stage. However, this is an outstanding general question for many river systems—and our discussion here has provided more insight than the simple, conventional assumption of constant activity maintained for all floods throughout river systems. Consequently, this empirical approach to estimating CIRCA offers a practical means for dating floodplain sediment in mobile Bolivian rivers with unknown sedimentation, that is, a more flexible CIRCAUS method for $^{210}\text{Pb}$ geochronology that accounts for downstream variations in activity.

(b) Procedure 2: identifying and dating activity plateaus

As previously discussed, the core profiles from most floodplain locations across the Beni and Mamore Foreland exhibit regions of relatively uniform
Figure 9. Floodplain core from Beni River site LVB-6, located 2.5 km from the river. With reduced vertical resolution of clay-normalized $^{210}$Pb activity, floodplain site could easily be interpreted as having received constant sedimentation at 0.9 cm yr$^{-1}$ (line depicts best-fit CIRCACS model). However, the full vertical resolution illustrates that there are two activity plateaus and a meteoric cap. The X-ray depicts a few faint laminations, although the core appears intact. The upper plateau dates at 1983 ($\pm 1.8$ years) and the lower plateau at 1949 ($\pm 2.5$ years), both years of major floods on the Beni. The meteoric cap on the upper plateau dates at 1982. As discussed in the text, the cap on the lower plateau may actually be incorporated into the ‘shoulder’ separating the two plateaus, so that there is an approximately 5 cm zone of uncertainty between the two plateaus. This core illustrates how the CNAXS method can facilitate detailed comparison of different hypotheses for the temporal and textural nature of sediment accumulation at a site. (Online version in colour.)

clay-normalized $^{210}$Pb activity, separated by abrupt changes in activity (examples in figures 4 and 9). Whenever XS $^{210}$Pb activity is uniform over a substantial vertical distance, the CIRCA assumption implies that all of this sediment is of the same radiometric age (to visualize fully the nuances of activity profiles, it is important to plot on linear axes, not the semi-logarithmic axes often used for $^{210}$Pb plots). Because these features are so prevalent, we have termed them ‘activity plateaus’, which are uniform XS $^{210}$Pb activity zones interpreted as

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discrete sediment accumulation events. A plateau is typically bounded by an upcore rise in activity and a down-core drop, with roughly uniform activity across the plateau. The basic dating procedure is to average the value of all activities within the plateau and then to date this average with simple radionuclide decay,

\[ T_{\text{sed}} = -\ln \left( \frac{A_{\text{csed}} - A_{\text{csup}}}{A_{\text{CIRCA}} - A_{\text{csup}}} \right) / \lambda_{\text{Pb}} \]  

Here, sediment age in years \((T_{\text{sed}})\) is determined as a simple decay function of unsupported clay-normalized \(^{210}\)Pb activity (clay-normalized sediment activity, \(A_{\text{csed}}\), minus clay-normalized supported activity, \(A_{\text{csup}}\)) and the corresponding unsupported clay-normalized CIRCA value \((A_{\text{CIRCA}} - A_{\text{csup}})\).

The accuracy of the resulting date depends on how well the plateau is defined by the number and consistency of the constituent points—the standard error of the measured plateau activity is propagated through equation (5.1). In practice, dates for well-defined plateaus (±10% variability) have typical confidence intervals (including CIRCA error) of ±2 years over the past 67 years (three half-lives). Dates out to 110 years (five half-lives) or for noisy plateaus (±20% variability) are possible, but have typical confidence intervals of ±5 years (3–7 years is common). Such plateau ‘noise’ typically occurs when sample clay content is low (less than 15%), resulting in increased uncertainty in determining the clay-normalized \(^{210}\)Pb activity (owing to dividing by a small number). Dates are not particularly sensitive to deviations in the assumed CIRCA value—for example, in a reach with assumed CIRCA of 6.5 dpm g\(^{-1}\) clay, the actual initial clay activity can vary from 6.3 to 6.7 dpm g\(^{-1}\) clay without deflecting plateau dates more than 1.3 years (regardless of plateau age). Tripling that range to 5.9–7.1 dpm g\(^{-1}\) clay increases the expected error to only 3.8 years. Such relative insensitivity to variations in the initial XS \(^{210}\)Pb activity underscores the robust nature of the CIRCA assumption underpinning the geochronology.

Environments characterized by more intense bioturbation would tend to exhibit smearing of the plateau edges, but the centres should remain defined so long as the effective mixing distance is less than half of the total plateau length. Because the CNAXS method assumes nothing about the temporal or spatial nature of floodplain sedimentation, the analytical approach to selecting subsample depths is iterative. This proves to be a strength of the method because core sections initially thought to represent plateaus can be selected for additional analysis at greater resolution to investigate the hypothesized plateaus (the first laboratory run typically consists of approx. 10 discrete points along the entire core). As a result, plateaus can be ultimately defined to a vertical resolution limited only by the site bioturbation, core diameter (i.e. which limits sample mass) and the enthusiasm of the researcher. As a matter of accuracy, it is important to choose points that fully define the limits of the plateau, along with enough points across the centre to verify that the \(^{210}\)Pb activity is indeed uniform. Cores with multiple plateaus representing several accumulation events will obviously require more samples and analysis than single-event cores to interpret the complex activity profiles.

Figure 9 presents an example of the utility and detailed insight offered by this approach. In this core, \(^{210}\)Pb has been determined at every available depth interval, allowing for a good interpretation of sedimentation processes. At first
glance, sedimentation could be described by a CIRCACS model with an annual sediment accumulation of 0.9 cm yr$^{-1}$. However, further inspection reveals a plateau (dated 1983) from 18 to 41 cm and a second plateau of lower activity (dated 1949) from 41 to 65 cm. From 0 to 18 cm lies a meteoric cap, dated 1982 (see discussion in §5c). This core provides an example of how two distinct sedimentation events separated by 34 years can combine with a 27 year old meteoric cap (ingrown during the time since the last sediment was deposited at this site) to provide a false impression of constant sedimentation. A lower resolution sampling of this core would probably result in the CS interpretation (e.g. such as that in figure 5a), but a detailed activity profile illustrates that sedimentation occurs only once every few decades. This example illustrates that the utility of the CNAXS/CIRCAUS method relies upon analytical rigour in (i) selecting the appropriate sub-sample points (number and location) to fully test all reasonable sedimentation hypotheses for each core and (ii) in maintaining suitable laboratory procedures to resolve clay-normalized $^{210}$Pb activity at the necessary accuracy and vertical precision.

(c) Procedure 3: identifying and dating meteoric caps

For any given floodplain location, meteoric $^{210}$Pb falls out of the atmosphere at an approximately constant annual rate, and is rapidly and strongly adsorbed by clay within approximately 10 cm of the surface. If a surface is aggrading, this meteoric activity dose is incorporated into the most recently deposited sediment, as previously discussed for CIRCACS profiles (figure 5a). If a floodplain surface is stable, without aggradation for years to decades as is usually the case for the study rivers, the meteoric $^{210}$Pb will adsorb onto the clay nearest the surface, forming a ‘meteoric cap’, observed at most locations throughout the Beni and Mamore floodplains. The best examples of these caps are on elevated river terraces, where no deposition has occurred for hundreds or thousands of years. We collected cores from a dozen such locations across the Beni and Mamore Foreland to assess the character and the variability of the fallout process.

Terrace cores (figure 10) provide three very important pieces of information. First, they provide a means to determine the supported background activity of river sediment using the same chemistry and counting techniques that are employed for the rest of the core samples. Because the old river sediment on Bolivian terraces is mineralogically similar to the modern river sediment, this approach provides an ideal means to determine how unsupported activity varies with clay abundance. The conceptual question of how supported activity should be normalized to clay content (beyond the simple asymptote approach) was discussed earlier——while this procedure is extremely simple (constant) here, we stress that this may not be the case in regions with different mineralogy and so the normalization line should be determined for each river. The second function of terrace cores is that all unsupported $^{210}$Pb activity is in secular equilibrium with meteoric fallout, such that decay exactly equals input. Therefore, for such stable locations (undisturbed for more than five half-lives), total XS $^{210}$Pb inventory (not clay-normalized) reflects the equilibrium meteoric inventory $I_{atm} = 44$ dpm cm$^{-2}$ (±2) across 10 locations. This corresponds to an annual $^{210}$Pb fallout rate of 1.35 dpm cm$^{-2}$ yr$^{-1}$, which is higher than many other published rates [12] and therefore ideal for the dating of meteoric caps. Third, the shape of the terrace
Figure 10. Terrace core, Beni River. (a) Site 34 (3 km upstream of site 60 and proximal to floodplain site 41), located approximately 15 m above the low-stage water surface. X-ray shows no stratigraphic features, but core appears intact. Activity declines to a supported level of 1.34 dpm g\(^{-1}\) clay over a distance of 10 cm. Total unsupported activity indicates (from a depth–density integration of the cap) fallout XS \(^{210}\)Pb of \(I_{\text{atm}} = 42 \text{ dpm cm}^{-2}\). (b) Terrace core, taken 6 m away from core in (a). This exhibits similar characteristics and \(I_{\text{atm}} = 44 \text{ dpm cm}^{-2}\). (Online version in colour.)

The meteoric cap provides essential documentation of the adsorption mechanics and soil disturbance in nearby floodplain cores (including diffusion and mixing rates). For terrace locations in this study, almost all of the unsupported activity is accounted for within the top 10 cm of core, with most of that amount being adsorbed in the top 5 cm. Consequently, it follows that \(^{210}\)Pb adsorbs very rapidly upon entering the soil, and that it is thereafter immobile. Furthermore, there is limited soil mixing at these locations, as indicated by a meteoric cap that is sharply defined. Because the mineralogical, climatic and biological conditions are similar, these observations for terrace cores can be applied to any nearby floodplain surface that has been undisturbed for some period of time, even if only for a decade. The key is to be able to clearly resolve and identify a characteristic meteoric cap ‘spike’ in a core top—these are very sharp, distinctive features, rising to activity levels substantially above that of fresh river sediment and exhibiting...
at least a 2.5-fold rise in activities over approximately 10 cm. By applying these unique qualifications, it is not likely to mistake sediment accumulation for a meteoric cap. We can thereby identify distinctive meteoric caps in at least 33 per cent of our floodplain cores (electronic supplementary material, table S1). Since fallout rates ($I_{atm}$) are uniform and well defined by terrace cores, it is straightforward to date such caps.

Consider the case of a discrete floodplain sedimentation event. A fresh pulse of material arrives from the river, blanketing the location to a depth of tens of centimetres with sediment of a uniform clay-normalized activity, as resolved by the CNAXS/CIRCAUS method. As years pass without further sediment accumulation, two processes affect the $^{210}$Pb profile. First, the activity of the event plateau decays identically across the entire deposit, lowering the plateau activity. Second, continuous $^{210}$Pb fallout grows an ever-larger ‘meteoric cap’ within the top approximately 10 cm of the deposit. The total activity of this meteoric cap, which exceeds that of the underlying plateau, represents the total inventory of atmospheric $^{210}$Pb fallout within the cap, $I_{cap}$ (dpm cm$^{-2}$). Assuming relatively constant annual fallout, the age of this cap, $T_{cap}$ (years) can be calculated as

$$T_{cap} = -\ln \frac{1 - (I_{cap}/I_{atm})}{\lambda_{Pb}}.$$

Such a relationship can be used to date meteoric caps up to about five $^{210}$Pb half-lives in age. Because the process of cap growth is entirely independent from the assumptions of CIRCA and any supported activity determinations used to date the plateau in equation (5.1), this separate technique for dating meteoric caps can be used to verify the CIRCA values.

Effective cap dating requires three fundamental conditions. First, the meteoric fallout must be established and relatively uniform over multiple years, as it is for the Bolivian Llanos and many other places [7]. Second, clear evidence for a plateau must exist to a depth well beyond that of the apparent meteoric cap, and all indications (e.g. activity, grain size, X-radiographs, flooding history) must point to a single depositional event of uniform age without any further sediment accumulation. Bioturbation may occur to a limited degree and depth (e.g. due to rooting, worms and other fauna), so long as both the cap and the plateau remain clearly identifiable, such that $I_{cap}$ can be determined with confidence (high levels of mixing would smear out the high activities and sharp gradients of caps, making it difficult to rule out recent sediment deposition as a source of this activity). Third, because most of the cap activity is typically within the top 5 cm, the floodplain surface could not have been disturbed in any way such as to remove even 1–2 cm of sediment, and the coring and analytical techniques must similarly preserve and resolve this near-surface activity. In practice, across the Beni and Mamore Foreland, it is this third condition that is most commonly violated because of minor surface scour that occurs in some places. As a result, the ages determined for meteoric caps are often younger but rarely older (only one case in electronic supplementary material, table S1) than those of the underlying activity plateau. Another important consideration is that fallout of $^{210}$Pb in rain and leaf litter distribution may vary across the floodplain surface, reducing fallout reaching sediment at some locations and increasing it in others, over a spatial
distance of a few metres (a possible cause of some of the young cap values and the one older cap value in electronic supplementary material, table S1). However, we were careful to sample away from the shielding/concentrating effects of large trees (trunks and roots) and termite nests in small trees. In our extensive field experience cutting transects and seeking shelter from storms: (i) rainfall is intense in most places (in contrast to a temperate forest), (ii) rainstorms are frequent and heavy (approx. 3 m per year), and (iii) the rainforest canopy and understory are homogenous enough that the thin leaf litter is evenly distributed in most places. Therefore, while we realize that $^{210}$Pb fallout shielding or concentrating effects undoubtedly occur in some places, we believe that this is more likely to be a problem in temperate climates or on hillslopes [34]. For this study of tropical floodplains, many cores with a preserved cap show close agreement between the two ages (electronic supplementary material, table S1), including Mamore cores (figure 11). Consequently, for locations with episodic sedimentation, the proposed procedure for dating clearly defined meteoric $^{210}$Pb caps works as an effective radioisotopic geochronometer independent from the assumptions of CIRCA.

Our work raised three questions regarding meteoric-cap mechanics. First, why are caps sometimes not observed at depth on top of older sedimentation plateaus within cores with two or more sedimentation events (e.g. figures 4c and 9)? The simplest explanation is that some of the profiles of buried plateaus do not have sufficient vertical sampling resolution to properly portray these buried caps (e.g. the depth intervals were too large and homogenized the cap spikes, or all intervals were not measured—not a problem where we have done higher resolution sampling). Also, the elevated activity of the buried meteoric cap has decayed since deposition and has become less distinct from the underlying plateau. Therefore, a buried cap on an older plateau could easily be mistaken for the bottom shoulder of the overlying younger plateau, implying an approximately 5 cm zone of uncertainty at the transition between two plateaus (this may be the case for figure 9 because the two bottom samples of the upper plateau exhibit a finer grain-size distribution more characteristic of the lower plateau). Another consideration is that basal scour during the deposition of the overlying sediment could remove a few centimetres of the floodplain surface, erasing some of the previous meteoric-cap activity. Regardless of the explanation for not observing buried caps in some places, the identification and dating of activity plateaus would not be compromised for most locations.

The second question regards the prevalence of this hypothesized loss of some cap material by the scour of floodwaters flowing over the floodplain. The third question regards radon ventilation effects that diminish the $^{222}$Rn concentration and hence the supported $^{210}$Pb activity near the floodplain surface, a process similar to that described by Imboden & Stiller [41]. Both processes were not obviously common for the wet, locally inundated, densely vegetated rainforest floodplains discussed here—although in our surveys of scores of floodplain transects, we have found isolated examples of scour found in some places near the channel following exceptional floods. With regards to radon ventilation, we have not found this to be a significant problem in the Bolivian study areas, but have found elsewhere [42] that these effects can be important for diminishing supported activity near the surface of some dry, temperate floodplains.
Figure 11. Cores from a transect on the Mamore River floodplain, illustrating that the CNAXS approach of identifying sediment plateaus and meteoric caps is transportable to a nearby dynamic river system. (a) Site 12RFC-75, 100 m from the river when sediment was deposited in 1943 (±3.9 years). Cap date is 1945. Site 12 RFC-150, 175 m from the river when sediment was deposited in 1944 (±200 yr). Cap date is 1945. Cores are similar, illustrating the general observation that nearby sites record the same event(s). The Rio Mamore differs from the Beni in that channel migration rates are much lower and floodplain deposition seems to have ceased for much of the basin since the early 1970s [2]. Other than those two differences, the sediment characteristics, meteoric fallout rates and typical core activity profiles are very similar between the Rios Mamore and Beni. (Online version in colour.)

6. Independent verification of dates and summary of results

A dating methodology is only as reliable as the independent verification of its results. Many studies that use $^{210}$Pb also employ $^{137}$Cs geochronology, as previously summarized (it is convenient to gamma count both). However, we have not found measurable $^{137}$Cs activity in our own gamma counting of dozens of profiles from Beni and Mamore cores representing a diverse range of depositional environments (fallout was low in this region, with $^{137}$Cs limited to trace amounts in the most stable terrace soils, e.g. figure 10). Instead, we have verified our results
Figure 12. Accumulation occurs only during large, rapid-rise floods (determined from a daily discharge record at Rurrenabaque since 1967) and corresponds well to cold-phase ENSO events that follow warm-phase events (line shows a proxy for sea temperature) [2]. (Online version in colour.)

using five other independent means of dating. First, a third of our cores have datable meteoric caps, and these dates agree with those of the underlying activity plateau (they give the same age or younger), as shown in figures 4, 9 and 11 and electronic supplementary material, figure S3 and summarized in electronic supplementary material, table S1 (‘Cap (C)’ dates). Second, we have Landsat images and aerial photographs bracketing a sedimentation event by channel migration in seven locations, and we have field observations of sedimentation in three core locations (electronic supplementary material, table S1, ‘image (Im)’ or ‘record (R)’ dates), all 10 of which agree with core geochronology. Third, the dates of sediment accumulation on floodplains closely match the timing of large rapid-rise floods (we have a daily discharge record at Rurrenabaque since 1967) and cold-phase El-Niño southern oscillation (ENSO) events [2], summarized in figure 12. Fourth, core sets that are collected along individual floodplain transects usually exhibit the same style and timing of sedimentation (figures 4 and 11), an expected result for cores that were collected within a few hundred metres of each other. And fifth, the rates of sediment accumulation decline away from the channel as a monotonic function (out to 3 km distance), and the few cores with constant accumulation are all located within 50 m of the channel (electronic supplementary material, figure S6 and table S1). This distribution of rates and dates (in figure 12) makes good physical sense [2,23,31], so it suggests that the CIRCAUS method for $^{210}$Pb geochronology produces viable results.

The key results regarding the geomorphic and sedimentological processes characterized by CNAXS/CIRCAUS dating from these rivers were presented elsewhere [2], and therefore are only briefly summarized here (figure 12 and electronic supplementary material, figure S6 and S7). In short, sediment is conveyed onto the floodplains by ENSO-orchestrated rapidly rising floods that cause the river stage to rise much faster than the floodplain stage of ‘black water’ from local precipitation, overtopping lower regions along the natural levees. The vigorous advective flow emplaces extensive sheet sand and splay deposits into discrete locations across the higher floodplain (unlike [33] we avoided coring floodplain depressions, e.g. electronic supplementary material, figure S7), both of which we have observed during campaigns in 1999–2001, 2004 and 2011. For normal annual floods, the rain water (supplied by the torrential local rainfall

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during the flood season) fills the floodplains and spills out into the channel, preventing the sediment-laden ‘white’ river water from leaving the channel and flowing across the floodplains to emplace sediment deposits (both advective transport and lateral diffusion of sediment are also retarded by the thick rainforest vegetation). The high, variable, more regular accumulation closest to the channel represents the processes that rapidly grow the topographically prominent natural levees (we have surveyed many), and the distal, irregular rates represent the emplacement of sheet sand and splay deposits. Many nuances of temporal and spatial distributions of accumulation can be explained by consulting local rainfall and discharge records—for example, the reason that the XS $^{210}\text{Pb}$ activity values for Mamore floodplain sediment are less (figure 3b) is that accumulation ceased around 1971 because of a noted shift in climate that decreased the relative importance of the discharge for sediment-laden water from the Andes for the Mamore Foreland [2].

7. Summary

The CIRCAUS approach to $^{210}\text{Pb}$ geochronology consists of a number of conceptual and methodological enhancements that together offer a means for identifying and dating individual sediment accumulation events in the vicinity of dynamic, heterogeneous river systems. First, the approach recognizes that both the floodplain clay content and sediment accumulation rate may be highly irregular, necessitating the determination of a clay-normalized $^{210}\text{Pb}$ activity profile (by equation (4.1)) at discrete intervals for each floodplain core. We present new techniques for determining the CNAXS $^{210}\text{Pb}$ activity of sediment, which is a useful, noise-free measurement for absorbed $^{210}\text{Pb}$ activity. This is an iterative, adaptive process, with the researcher analysing additional sample points to accurately resolve, verify and date any stepped activity profiles that are initially hypothesized to represent discrete sedimentation events. Second, both a numerical and empirical means are provided to estimate CIRCA along large river systems, with explicit recognition of the processes that cause variation in the $^{210}\text{Pb}$ activity of fresh river sediment. Hence, each river system can be evaluated for processes that affect the application of $^{210}\text{Pb}$ geochronology. Third, procedures are presented to date both episodic sedimentation events (equation (5.1)) and cases of constant floodplain sedimentation (CIRCACS). This offers scientists the flexibility needed to investigate environments with a wide range of sedimentation processes and rates. Fourth, we outline an independent approach for dating meteoric caps above activity plateaus (equation (5.2)), such that the dates of discrete sedimentation events can be verified.

The techniques described are robust and transportable to many other river systems where they have worked equally well [31,32,42–47]. The concepts developed [23] for $^{210}\text{Pb}$ (downstream decline in sediment activity, accumulation event plateaus and meteoric caps) may also be applied to other clay-associated meteoric radionuclides, such as $^{137}\text{Cs}$ and $^{7}\text{Be}$, as has recently been suggested [38]. In environments with more complex delivery of sediment than in the Bolivian study area, our procedures could be combined with a numerical model [38,48] to simulate an alternating process scenario of constant sedimentation interrupted by episodic, time-dependent sediment accumulation.

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CNAXS/CIRCAUS provides an approach that can resolve complex, episodic sedimentation processes in heterogeneous, dynamic floodplain environments, providing a more detailed insight into the river’s history. Over the past several years, this refined technique for $^{210}$Pb geochronology has successfully been applied to characterize detailed floodplain/river histories on six continents. The drawback is that it is more labour intensive (many samples per core, with clay abundance determined for each sample) than previous methods, and its iterative nature requires considerable analytical investment and hypothesis testing for activity profiles before the resulting geochronology is well defined. However, once the requisite effort has been made to establish the radionuclide systematics and verify the dates for cores, additional sediment cores along the same river can be dated without much difficulty.

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References


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23 Aalto, R. 2002 Geomorphic form and process of mass flux within an active orogen: denudation of the Bolivian Andes and sediment transport and deposition within the channel-floodplain systems of the Amazonian foreland. PhD thesis, University of Seattle, Seattle, WA, USA.


