USE OF ^{210}Pb GEOCHRONOLOGY TO EXPLORE THE CENTURY-SCALE MERCURY CONTAMINATION HISTORY AND THE IMPORTANCE OF FLOODPLAIN ACCUMULATION IN ANDEAN TRIBUTARIES OF THE AMAZON RIVER

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INTRODUCTION

Mercury pollution of water and fish in the Amazon basin is a significant hazard for riparian populations, especially for people whose primary protein source is contaminated fish (Lebel et al., 1999; Maurice-Bourgoin et al., 2000; Roulet & Maury-Brachet, 2001); irreversible Hg damage has been implicated for the nervous and immune systems (Lebel et al., 1998). In the Bolivian Andes, at the headwaters of the Beni river, erosion of contaminated soils and weathering of mercury-bearing rocks during the rainy season can increase the water concentration of Hg by 50 fold (Maurice Bourgoin et al., 2001). These soils may have become contaminated through natural crustal degasification and dissolution of Hg, and also by centuries of extensive Hg use during both the colonial and post-colonial extraction of massive quantities of gold and silver (Nriagu et al., 1992). More recently, a new gold-mining boom starting in the 1970s has introduced an estimated 0.25 to 0.5 tonnes/year of Hg into the Beni River basin (Maurice-Bourgoin et al., 2000). The mercury used can be released directly into the river itself or indirectly through the atmosphere by open-air burning of the amalgam (Malm et al., 1990; Maurice-Bourgoin et al., 1999). But the major environmental impact of these mining activities is the deforestation of the exploited areas and the release of millions of tonnes of Hg-contaminated sediments and soils to affected rivers, rather than the direct release of Hg used during the last stages of the gold extraction (Maurice-Bourgoin, 2001). Numerous studies have documented the distribution of Hg in water, fish, and people throughout the Amazon basin (Akagi et al., 1995; Malm et al., 1995; Barbosa et al., 1997; Roulet et al., 1999; Silva-Forsberg et al., 1999; Maurice-Bourgoin et al., 2000), but the important link between Hg and sediment transport had not been fully explored. To investigate the sources, transport and storage mechanics, and contamination history of Hg for the Madeira River, one of the most important tributary to the Amazon in terms of sediment input, we analysed the Hg concentration and relevant geochemistry of floodplain and river sediments.

STUDY AREA

The Bolivian Amazon basin represents the Andean headwaters of the Madeira River, one of the most important tributaries of the Amazon River. In Bolivia, its drainage basin is 0,9 x 10^6 km^2, with 25% in the Andes, 27% on the Brazilian shield and 48% in the plain. The Beni and the Mamore Rivers, the largest sediment suppliers of the Madeira river (Guyot, 1993), constitute the focus of our study. Andean tributaries of these rivers drain both semi-arid areas of high altitude and areas of tropical humid forest of the sub-Andes. Heights above sea level range from 6400 m at the highest headwaters to 115 m at the Brazilian border. The Beni and Mamore Rivers (Fig. 1) transport sediments from the rapidly eroding Bolivian Andes across the floodplains of a large foreland basin. Floodplains and channels are essentially pristine, without artificial levees, dams, dredging, numerous roads, or other anthropogenic complications. At the edge of the sub-Andes, the drainage area of the Beni River at the edge of the piedmont, is 67,500 km^2 and the mean annual discharge during the sampling period, 1998-1999, averaged 2300 m^3 s^{-1}. The Beni River collects numerous Andean tributaries, half of which is exploited for their alluvial gold since the 1960s (Fig. 1). The adjacent Mamore drains a 600,000 km^2 basin, much of which is floodplain. These basins are representative of the vast expanse of Andean-Amazonian foreland basins to the north.

MATERIAL AND METHODS

One sampling survey was conducted along the entire Beni river in August-September, 1999 and another along the Mamore River in September, 2000. Water samples were collected using Teflon bottles and all procedures were performed using 'ultra-clean' techniques (Nolting and De Jong, 1994; Gaudet et al., 1995). Sediment cores were sampled 1.3 meters deep into the floodplain at different locations along these rivers (Fig. 1); they were frozen on the field and subsequently dried and processed in the laboratory. The T-Hg was determined by Atomic Fluorescence Spectrometry after reduction with Sn(II); the detection limit was 2 ng g^{-1}. The accuracy and reproducibilty of the method were calibrated by the analysis of a reference standard; the mean T-Hg concentration obtained was 92 ± 4 ng g^{-1} (dw) for a recommanded value of 92 ± 9 ng g^{-1} (dw). The concentrations of T-Hg, organic carbon and nitrogen, iron (Fe_{cdb}) and aluminium (Al_{cdb}) substituted into oxy-hydroxydes, as associated with the granulometry and

^{210}Pb activity within the same sediment cores, were used to characterise the main geochemical processes controlling the deposition of Hg within the floodplain.

A new model has been developed for ^{210}Pb geochronology on river floodplains, termed Constant Initial Reach Clay Activity and Unknown Sedimentation rate (CIRCAUS) and has been applied to more than two hundred locations throughout the Beni and Mamore systems (Aalto, 2002). The approach employs discrete down-core, clay-normalised measurements of ^{210}Pb activity and is coupled with a geomorphic model of the input concentration of ^{210}Pb in sediment during large floods. The numerical modelling approach is supplemented with an empirical determination of the supported ^{210}Pb activity, the meteoric input of ^{210}Pb, and the clay-normalized ^{210}Pb activity in fresh sediment for that reach of river. The sediment age is determined from the "decay" age of any activity plateaus (zones of uniform activity found in most cores), as well as independently calculated from the excess "meteoric cap" ^{210}Pb activity grown in above the activity of the plateau.

RESULTS AND DISCUSSION

EFFLUX OF SEDIMENTS AND ASSOCIATED MERCURY TO THE BENI FLOODPLAIN

The T-Hg concentrations in the Beni river range from 19 ng l^{-1} during the dry season to 460 ng l^{-1} at peak flood stage. The Hg associated with the particles represents 59 and 98% of the T-Hg, respectively. In this Andean tributary, the importance of soil erosion during the rainy season in controlling the mercury concentrations of freshwater has been previously demonstrated (Maurice-Bourgoin et al., 2001). For the 1999 water year, from surface water samples, we estimated a total suspended particulate matter (SPM) fflux in the Beni R. of 302 10^6 tonnes; 97% of this material was transported during the first four months of high water and about 40% is later deposited in the Amazonian floodplain, as shown in a previous study (Guyot, 1993). The total Hg (T-Hg) flux associated with sediment reached 33 tonnes, 96% of which was particulate Hg (P-Hg). During the wet season, from November to March, Hg-contaminated particles are transported from the Andean sub-basins characterised by steep slopes and by an accelerated erosion rate driven by recent agriculture practices and the ongoing construction of a major road. Due to the high adsorption capacity of mercury on fine particles (clays) in the white water rivers (in contrast to organic-rich "black waters"), most of the mercury is transported on SPM. In the Beni river basin, a depth-integrated average of 35% of the deposited sediment collected in the floodplain is composed of clays, and these contains most of the sediment-born mercury.

CENTURY-SCALE CONTAMINATION HISTORY AND IMPORTANCE OF SEDIMENT TRANSPORT AND FLOODPLAIN ACCUMULATION

For the 180 sediment samples analyzed we observe (Fig. 2) a strong correlation between T-Hg and clays content (r^2 = 0.80), Al_{tot} (r^2 = 0.65), Fe_{tot} (r^2 = 0.64) and with Fe incorporated in clay minerals, Fe_{Si}, (r^2 = 0.53) concentrations. This confirms that clays and Al and Fe oxy-hydroxides in sediments may serve as the primary carrier and facilitate the transport and storage of Hg within the Beni and Mamore river-floodplain systems.

By utilising results from a detailed model for the transport and deposition of river sediment (including clays) along the Beni river floodplain system (Aalto, 2002), we estimate, using the observed Hg-Clay relationship (Fig. 2), an annual exchange of clay-affiliated Hg between the floodplain and the channel of 2.1 tonnes Hg, primarily due to channel migration. This exchange has been estimated by subtracting the net Hg deposition of 2.4 tonnes $year^{-1}$, primarily lost into the distal floodplain, from the total deposition of 4.5 tonnes $year^{-1}$.

In many of our cores, we observe an increase of the T-Hg concentrations in sediments with depth. Applying the CIRCAUS model to date the sediment within our floodplain cores, we find that the ratio of observed Hg concentration to expected Hg concentration (as derived from measured clay abundance or from measured Al incorporated in clay minerals, Al_{Si}) is approximately 1 from 1900 to 1965 and then increases starting in the 70s (Fig. 3). This increase is most evident for the clay-normalized Hg content where we observed a best correlation between these two elements. This suggests that the clay, and in a less extend Al-normalized Hg concentration in sediment is constant from 1900-1965, and then shows an increase over the past 30 years, mostly likely due to a new input of Hg in the Andean drainage basins. This influx of Hg can be attributed to accelerated erosion of Hg enriched soils, due increased gold-mining activities, recent colonisation, and new agriculture practices (e.g. burning after deforestation), road construction, and other human activities. A substantial increase in the price of gold during the 70s, with a maximum in 1980, sparked a new gold boom in various tropical countries, and especially in Latin America (Cleary, 2000).

CONCLUSION

The substantial increase of clay-normalised Hg concentration in the sediments of an important tributary of the Amazon during the last 30 years might be explained by a recent gold mining boom and also by increased deforestation of steep hillsides and the resulting erosion of soils. This study represents the first geochronological determination of heavy metal pollution history from floodplain sediments. These results underscore the importance of river sediment as a carrier of Hg, and the key role of channel-floodplain sediment interchange in regulating the transport and accumulation of any sediment-associated pollution.

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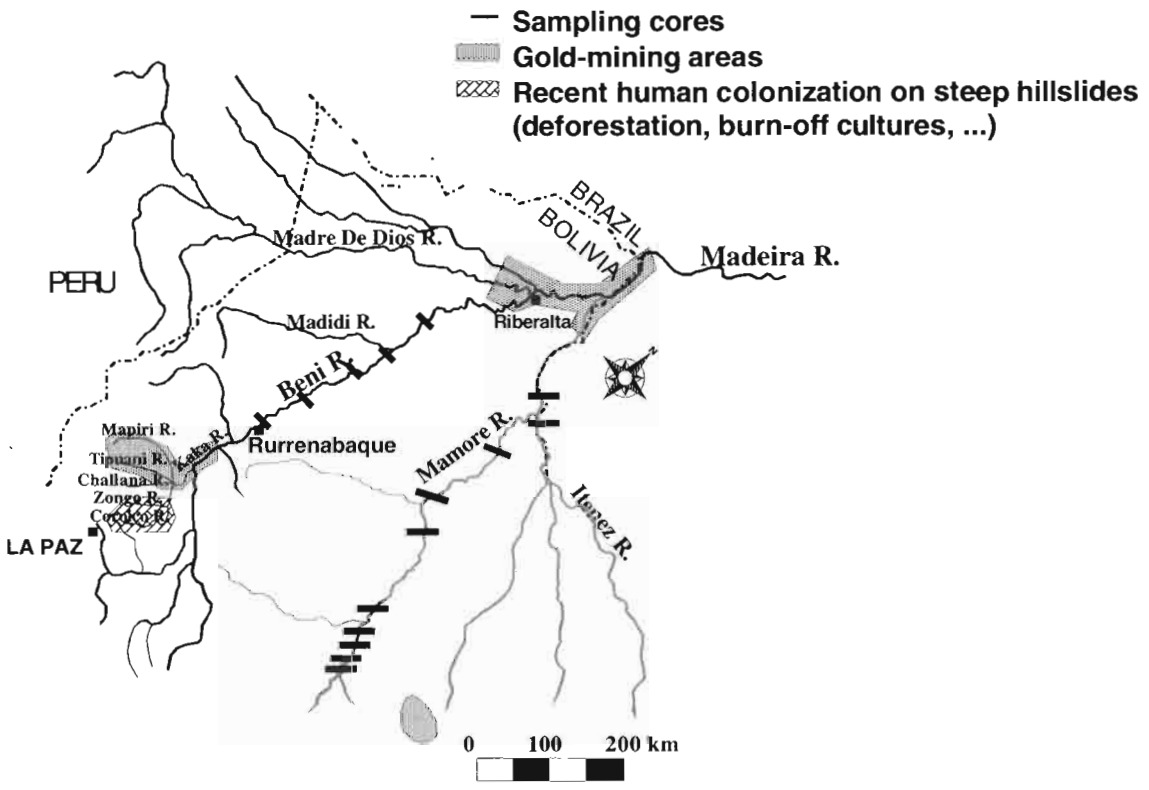

Figure 1. Location of the sampling cores and distribution of the recent anthropogenic disturbance (deforestation, mining, road construction) on the hydrological map of the Madeira tributaries.

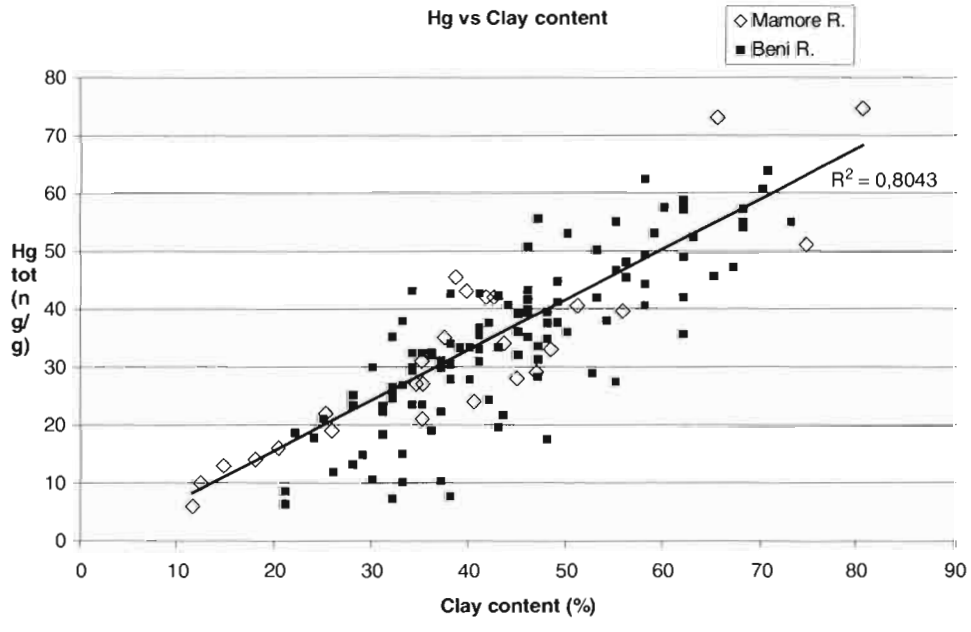

Figure 2. Correlation between total Hg concentrations and clay content in sediment samples from the beni and Mamore Rivers

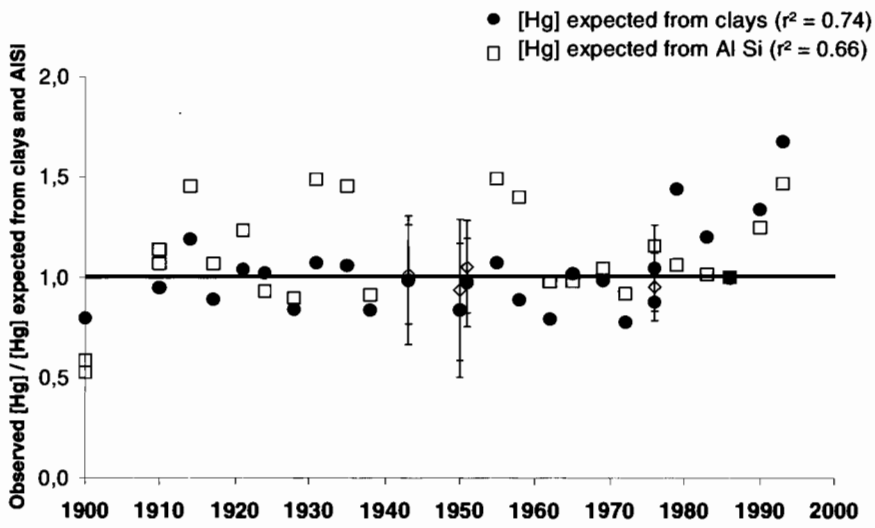

Figure 3. Observed Hg concentration / expected Hg ratio from measured clay abundance (circles), and Al incorporated in clay minerals (open diamonds), in the Beni river sediments, over the last century.

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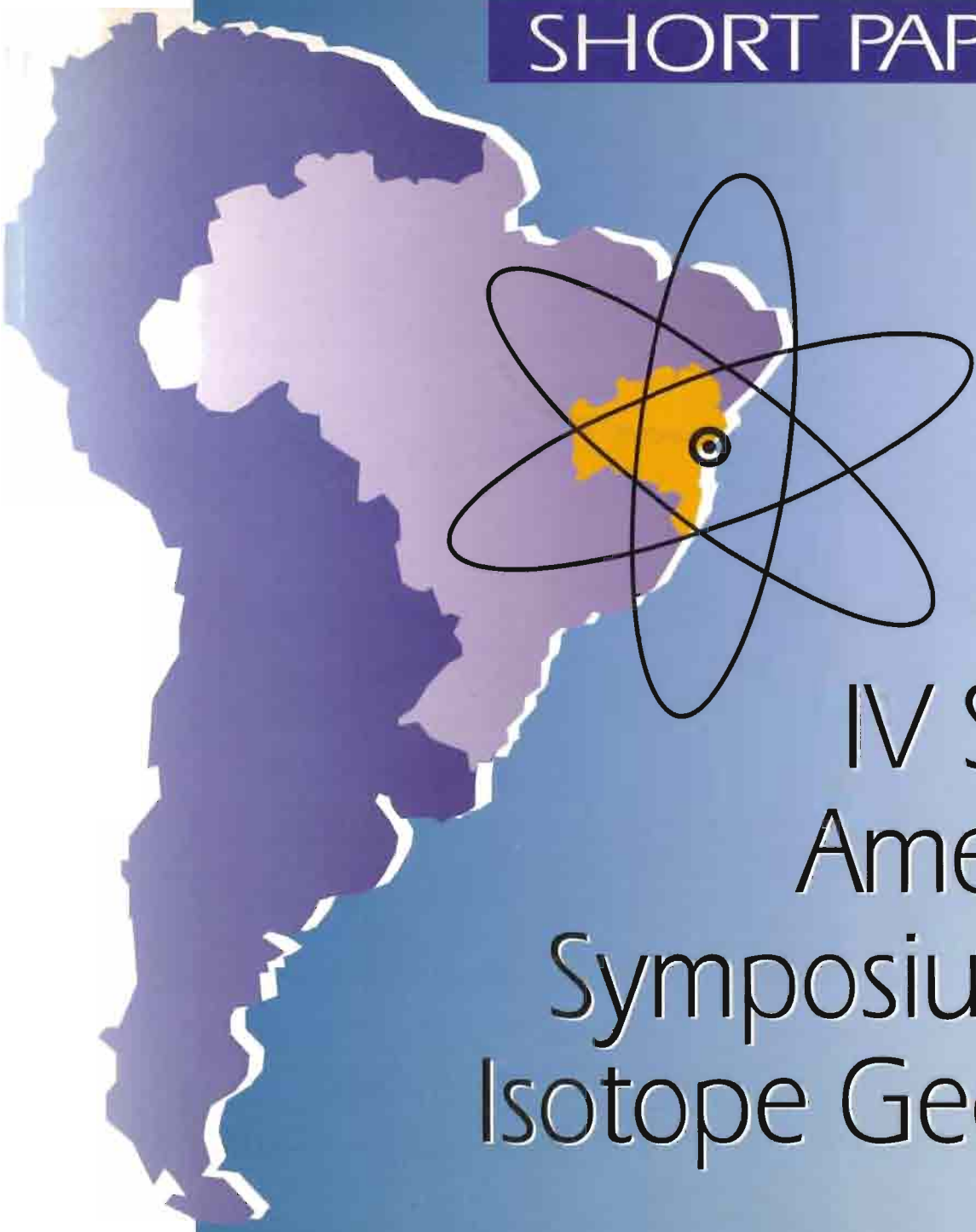

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