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1	Sediment dynamics during the rainy season
2	in tropical highland catchments of central Mexico using fallout radionuclides
3	
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14	
15	Abstract
16	Tropical regions are affected by intense soil erosion associated with deforestation, overgrazing, and
17	cropping intensification. This land degradation leads to important on-site (e.g., decrease in soil
18	fertility) and off-site (e.g., reservoir siltation, water pollution) impacts. This study determined the
19	mean soil particle and sediment residence times in soils and rivers of three subcatchments (3-12 km ²)
20	with contrasted land uses (i.e., cropland, forests, rangelands) draining to a reservoir located in
21	highlands of the transvolcanic Mexican belt. Calculations were based on rainfall amount and river
22	discharges as well as on fallout radionuclide measurements (Be-7, Cs-137, Pb-210) conducted on
23	rainfall precipitated samples, soil sampled in the catchments, and suspended sediment collected by
24	automatic samplers in the river during most storms recorded throughout the 2009 rainy season.
25	Calculations using a radionuclide two-box balance model showed that the mean residence time of
26	sediment in soils ranged between 5000±1500 and 23,300±7000 years. In contrast, sediment residence
27	time in rivers was much shorter, fluctuating between 50±30 and 200±70 days. The shortest mean

28 residence times were measured in a hilly catchment dominated by cropland and rangelands, whereas 29 they were the longest in an undulating catchment dominated by forests and cropland. Calculation of 30 the Be-7/excess-Pb-210 in both rainfall and sediment allowed gaining insight on sediment dynamics 31 throughout the rainy season. The first heavy storms of the year exported the bulk of the sediment stock 32 accumulated in the river channel during the previous year. Then, during the rainy season, the two 33 steeper catchments dominated by cropland and rangelands reacted strongly to rainfall. Sediment was 34 indeed eroded and exported from both catchments during single heavy storms on several occasions in 35 2009. In contrast, the agro-forested catchment with gentler slopes exported sediment at a constant and 36 low rate throughout the rainy season. Overall, land cover and flood type clearly proved to exert more 37 control on sediment export than slope steepness and rainfall erosivity. Our results show the priority of 38 stabilising old gully systems to prevent their extension by regressive erosion to cropland and to 39 concentrate the implementation of on-site erosion control measures in cropland and rangeland of the 40 most reactive catchments. 41 42 Keywords: tropical environments; erosion; suspended sediment; residence time; fallout radionuclides. 43 44 **1.Introduction** 45 46 47 Land degradation is particularly severe in tropical regions, such as in Mexico (Descroix 48 et al., 2008), in southern China (Barton et al., 2004) or in eastern Africa (Nyssen et al., 2004). 49 In these regions, overgrazing, deforestation, and intensification of food crop cultivation have 50 led to severe erosion and to a decline in soil fertility (Roldán et al., 2003). Furthermore, once 51 it reaches the river, sediment leads to numerous problems in downstream areas (Owens et al., 52 2005). It causes, for instance, an increase in water turbidity and a rapid filling of reservoirs. Sediment is also associated with numerous substances and contaminants (e.g., metals, 53 54 nutrients, organic compounds, antibiotics; e.g., Tamtam et al., 2008; Le Cloarec et al., 2009).

These chemicals can then be bioaccumulated by organisms such as fishes ingesting particles and they can lead to public health problems after their consumption (e.g., Sánchez-Chardi et al., 2009; Urban et al., 2009). In mountainous environments, the problems associated with sedimentation are exacerbated by the important quantities of sediment produced during storms (e.g., Meybeck et al., 2003; Mano et al., 2009).

60 Sediment supply to the river needs to be controlled to prevent these problems. However, 61 the main erosion sources fist need to be determined and the transit times of sediment within 62 rivers need to be evaluated to implement appropriate and effective erosion control measures. In mountainous areas, the transit of sediments within rivers is affected by their specific flow 63 64 regime (e.g., Dedkov and Moszherin, 1992). In tropical areas where hydrology is controlled 65 by the succession of a dry and a rainy season, we generally assume that the increase in 66 discharge at the beginning of the rainy season leads to an important resuspension of sediment 67 accumulated in the river channel (e.g., Susperregui et al., 2009). However, the dynamics of 68 this resuspension process and its implications on the sediment transit within ephemeral rivers 69 throughout the rainy season are poorly understood.

70 The use of radionuclides with different half lives (i.e., Be-7, Cs-137, and excess-Pb-71 210) can contribute to investigate this issue. Dominik et al. (1987) designed a model to 72 estimate the residence time of sediment within the Rhône River using these radionuclides. 73 Bonniwell et al. (1999) proposed another methodology based on the Be-7/excess-Pb-210 ratio 74 to estimate residence times and transport distances of sediment in a catchment dominated by snowmelt hydrology. They also applied this method in several catchments located in northern 75 76 United States (e.g., Wilson et al., 2005). Matisoff et al. (2005) proposed, for instance, to 77 calculate an indicator of the suspended sediment age by conducting this Be-7/excess-Pb-210 78 ratio based on radionuclide measurements conducted on both rainfall and suspended sediment 79 samples. More recently, Le Cloarec et al. (2007) improved the partly indeterminate two-box

model proposed by Dominik et al. (1987) by taking account of the contribution of the
dissolved radionuclide fraction to the river fluxes and by introducing the Cs-137 inventory in
soils as an additional constraint, allowing to solve the system of radionuclide mass balance
equations. These model improvements allowed calculating sediment residence times in the
Seine River lowland basin in France.

85 However, to our knowledge, this methodology has never been implemented in 86 catchments located under the tropics. Previous studies estimating radionuclide fluxes 87 generally outlined the necessity to conduct a continuous monitoring of discharge and 88 sediment concentration in rivers to strengthen the reliability of their results (e.g., Bonniwell et 89 al., 1999; Le Cloarec et al., 2007). We therefore combine the use of continuous monitoring 90 techniques (i.e., the installation of river gauging stations and automatic sediment samplers in 91 several subcatchments of a larger mountainous catchment) and the measurement of fallout 92 radionuclides in both rainfall and suspended sediment samples to quantify sediment residence 93 times in a tropical highland region of central Mexico. This region concentrates the major part 94 of the country's population (INEGI, 2006). We conducted this study in subcatchments 95 representative of potential sediment sources areas draining to a reservoir that provides 25% of 96 water distributed to Morelia city (ca. 1,000,000 inhabitants). We will use the two-box model 97 improved by Le Cloarec et al. (2007) to calculate the mean soil particle and sediment 98 residence times in those tropical subcatchments characterised by the succession of a rainy 99 season and a (long) dry season. The indicator of suspended sediment age proposed by 100 Matisoff et al. (2005) will also be calculated to outline the intra-annual sediment dynamics 101 within those subcatchments. Finally, the implications of these findings for soil conservation in 102 tropical highlands will be discussed.

103

104 **2. Materials and methods**

105

106 2.1. Study area

107

The Cointzio catchment covers an area of 630 km² located in the transverse volcanic 108 109 belt of central Mexico (Fig. 1). The catchment bedrock consists of igneous rocks generated by 110 Quaternary volcanic activities. According to the World Reference Base for soil resources 111 (FAO, 2006), soils within the catchment are mainly Acrisols on the hillslides, Andisols and 112 Cambisols in headwater areas, and Luvisols in the plains (INEGI, 1985). The Cointzio 113 catchment undergoes temperate subhumid climate conditions, according to the Köppen 114 climate classification system. In this region, rainfall mostly occurs between May and October. 115 The river network, which is mostly ephemeral, is dominated by the Rio Grande de Morelia 116 River. A dam is located at the outlet of the catchment, 13 km upstream of Morelia city (ca. 117 1,000,000 inhabitants). This dam was built in 1940 to supply water for domestic consumption as well as for industrial and agricultural activities. The Cointzio reservoir (6 km^2 ; $65 \times 10^6 \text{ m}^3$) 118 119 undergoes significant sedimentation, which leads to an increase in water turbidity in the lake 120 inducing a drastic decline in the zooplankton population disturbing the entire trophic chain 121 (Ramirez-Olvera et al., 2004) and to a 20% loss of its water storage capacity (Susperregui et 122 al., 2009). Increasing siltation of this reservoir is a major concern in the Morelia region where 123 it supplies a significant part of distributed water.

In 1975, forests occupied ca. 25% of the catchment surface. Then, after a period of deforestation (1986–1996), a significant part of the cut areas recovered, and forests were observed on ca. 30% of the Cointzio catchment area in 2003 (López-Granados et al., 2001; Mendoza and López-Granados, 2007). Soil erosion within the catchment is mainly restricted to the mountain piedmonts (Servenay and Prat, 2003). Concentrated erosion leading to the formation of long and deep gullies is mainly observed on Acrisols. In contrast, Andisols and Cambisols, which are also known to be sensitive to erosion (Poulenard et al., 2001), are mostly affected by sheet erosion (Bravo-Espinoza et al., 2009). In areas confronted with severe erosion, soil was completely removed, leading to the exposition of igneous weathered rocks and tuffs (locally referred to as "tepetates") at the ground surface (Etchevers Barra et al., 2004).

135 Three subcatchments (i.e., Huertitas, La Cortina, Potrerillos) representative of the 136 different potential sources areas supplying sediment to the Cointzio reservoir were monitored 137 in the framework of the DESIRE (Desertification mitigation and remediation land – A global 138 approach for local solutions) and STREAMS (Sediment TRansport and Erosion Across 139 Mountains) projects (Fig. 1). These subcatchments are characterised by different land uses, 140 slope gradients, and soil conditions in order to cover the range of environments found in the 141 Cointzio catchment (Fig. 2; Table 1). Each of them is drained by a deep (2-4 m) and narrow 142 (1–5 m) river channel. Riverbed material is fine-sized (clay to sand-sized) and river banks are 143 extensively colonized by seeds and shrubs increasing their cohesion and preventing their 144 collapse.

145 Huertitas (3 km²) consists of Acrisols covered by cropland and rangeland. It is 146 characterised by a hilly topography, and 6% of its surface is affected by severe soil erosion 147 (i.e., gullies and small ephemeral rills; Fig. 3). Some small-scale landslides occur sometimes 148 in steep rangeland terrains. Land use in La Cortina (9 km²) is dominated by pine-oak forests 149 (i.e., Quercus rugosa and Pinus montezumae) and cropland located on undulating hillslopes 150 underlain by Andisols. Finally, the rolling subcatchment of Potrerillos (12 km²) is underlain 151 by Acrisols and Andisols and is covered by almost equivalent proportions of pine-oak forests 152 and eucalyptus plantations, cropland, and rangeland (Fig. 2C). Gullies only affect 1% of its 153 surface (Table 1). The contribution of those potential sediment sources areas to the Cointzio

reservoir siltation and the rate at which sediment transfer operates is a subject of intensedebate for the population living in the Morelia region.

156

157 2.2. Field measurements

158

159 2.2.1. Rainfall and discharge

160 Rain gauges and river monitoring stations were installed at the outlet of the three 161 subcatchments (Fig. 1). They provided continuous precipitation and water discharge data 162 derived from continuous water level measurements (with a 5-min time step) obtained with a 163 Thalimede OTT water level gauge, stored by a data logger device (Campbell CR800), and 164 corrected using stage-discharge rating curves (see Duvert et al., in press, for details on this 165 method). Given that rainfall kinetic energy is often used as an indicator of rainfall erosivity, kinetic energy (*KE*; J m⁻²) of each rainfall event was calculated using Eq. (1) (Salles et al., 166 167 2002):

168
$$KE_{mm} = \sum_{i} 8.95 + 8.44 (\log_{10} I_i)$$
 (1)

169 where KE_{mm} is the volume-specific kinetic energy (J m⁻² mm⁻¹), and *I* is the rainfall intensity at the 170 five-min time step (mm h⁻¹).

171 Rainfall was collected during 13 storms throughout the 2009 rainy season in a 172 graduated PVC container located at the outlet of the Cointzio catchment to determine its 173 content in Be-7 and Pb-210. We poured HCl (1.2 N) into the container to prevent radionuclide 174 sorption on its sides. After each rain, water volume was measured in the container. Then, 175 dissolved AlCl₃ (~ 5 g) was added to the rainfall sample within the few days following the 176 storm and shaken for 1 h (Ciffroy et al., 2003). Pouring of NaOH to reach a pH of 8-9 177 precipitated the dissolved Al, carrying Be-7 and Pb-210 with a recovery efficiency between 178 80 and 95% (Ciffroy et al., 2003). Samples were filtered, air-dried, and sent to France for

179 radionuclide analysis.

180

- 181 2.2.2. Measurement of SSC
- 182

183 At the outlet of each subcatchment, data on suspended sediment concentration (SSC; g L⁻¹) were obtained using an automatic water sampler (ISCO 3700) triggered by water level 184 185 variations. During floods, water samples were collected after each 5-cm water level variation. 186 This sampling frequency was selected based on the mean characteristics (i.e., flood duration, 187 shape of rising and falling limbs) of the floods recorded previously (i.e., between 2006 and 188 2008; Duvert and Gratiot, unpublished data) in the subcatchments to obtain a trade-off 189 between a satisfactory event coverage and a reasonable amount of samples to collect in the 190 field (see Duvert et al., in press, for details).

191 SSC of each sample (generally ≥ 2 g L⁻¹) was estimated in laboratory by weighing an 192 aliquot of ca. 200 ml after drying it for 24 h at 60°C. For each flood, a composite sample was 193 prepared by mixing all the available individual samples. This allowed obtaining a mean 194 representative sample of each individual flood with a quantity of fine sediment (2–50 g) 195 sufficient to conduct radionuclide analyses.

196 Instantaneous suspended sediment flux SSF (t s⁻¹) was then calculated using Eq. (2): 197 $SSF = Q \cdot SSC \cdot 10^{-3}$ (2)

198

199 Suspended sediment yield (SSY; in ton, t) was also calculated for each flood using Eq. (3):

200
$$SSY = \int_{t_0}^{t_f} SSF \, dt$$
 (3)

201	with t_0 and t_f corresponding to the beginning and the end of the storm considered, between
202	May and November 2009. In total, 89 events that occurred between May and November 2009
203	were sampled at the outlet of the three subcatchments.
204	
205	2.2.3. Soil collection
206	
207	Soils representative of the different land uses observed in the three subcatchments
208	were collected. Sampling was concentrated in potential sediment source areas. For each
209	potential source, we collected five samples of surface material (top 2–5 cm) and mixed them
210	well to provide a homogeneous sample. In total, 30 representative samples were collected in
211	the field between June and November 2009 (Fig. 1).
212	
213	2.3. Radionuclide analysis
214	
215	2.3.1. Radionuclides of interest
216	
217	Be-7 is a natural cosmogenic radionuclide generated in the stratosphere and the upper
218	troposphere. Most Be-7 reaches the ground surface as wet fallout. It is strongly fixed to the
219	particles and mostly remains at the soil surface (i.e., in the uppermost 2 cm of the soil). Given
220	its short half-life ($T_{1/2} = 53$ d), it is used to document soil erosion over the short term (e.g.,
221	Sepulveda et al., 2008).
222	Pb-210 is a natural radionuclide ($T_{1/2} = 22.3$ y), which is a decay product of U-238
223	$(T_{1/2} = 4.5 \ 10^9 \ y)$. U-238 decays through a series of short-lived nuclides (e.g., Ra-226, Ra-
224	222). Ra-222 is a gas that partly remains in situ, forming "supported" Pb-210; and it partly

escapes to the atmosphere, forming "excess" Pb-210, when falling back on the soil surface bywet and dry fallout. It then strongly sorbs to soil particles.

- 227 Cs-137 is an artificial radionuclide ($T_{1/2} = 30$ y) produced by the thermonuclear bomb 228 testings conducted during the 1960s as well as, in certain regions of the world (i.e., mainly in 229 Europe), by the Chernobyl accident in 1986. It is now stored in soils, and this Cs-137 stock 230 decreases by radioactive decay and by fine sediment transfer to the rivers.
- 231 Those three radionuclides are characterised by high K_d partition coefficients (e.g.,
- 232 Wang and Cornett, 1993; Gil-Garcia et al., 2009) and allowed therefore to trace sediment

233 within catchments. The behaviour of those three radionuclides during sediment transport is

- further discussed in Le Cloarec et al. (2007).
- 235
- 236 2.3.2. Rainfall sample analysis
- 237

238 Precipitated rainfall samples were placed in polypropylene tubes. Counting was conducted 239 at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) in Gif-sur-Yvette 240 (France) using a low-background, high-efficiency, well-type Ge detector with a crystal 241 volume of 220 cm³ (GWL-220-15 Ortec). All samples were analysed within < 53 d 242 (corresponding to the Be-7 half life) after the rainfall event. Counting time reached ca. 1×10^5 s to minimise the associated errors (typically 10-30% for Be-7). 243 244 245 2.3.3. Soil and sediment analysis 246 247 Soil and composite suspended sediment samples were packed into 15- or 60-ml 248 (depending on the sediment quantity available in collected runoff) pre-tared polyethylene

specimen cups and sealed airtight to contain Rn-222 and allow ingrowth of its decay products.

Be-7 (477.6 keV), Cs-137 (661.6 keV), and Pb-210 (46.5 keV) activities in this sediment
were determined by gamma spectrometry using the seven very low-background coaxial Nand P-types GeHP detectors (Canberra / Ortec) available at the LSCE. "Excess" Pb-210 was
calculated by subtracting the supported activity (determined using two U-238 daughters, i.e.
Pb-214, by taking the average count number at 295.2 and 351.9 keV, and Bi-214 at 609.3

keV) from the total activity of Pb-210 (measured at 46.5 keV; Le Cloarec et al., in press).

255

256 When material available was insufficient (< 5 g), suspended sediment samples were 257 placed in polypropylene tubes, sealed airtight, and counted on the well-type Ge detector used 258 for rainfall precipitated sample analysis. Counting time of soil and sediment samples varied between 8×10^4 and 3×10^5 s to limit errors on activity determination (typically 5–10% except 259 for Be-7 with ca. 10–30%) and for practical reasons (i.e., to allow counting one sample d^{-1} on 260 261 each detector). All measured counts were corrected for background levels measured at least 262 every 2 months as well as for detector and geometry efficiencies. Results were systematically expressed in Bq kg⁻¹. Radionuclide activities were systematically corrected according to the 263 264 decay after the sampling period. Counting efficiencies and quality insurance were conducted 265 using internal and certified IAEA standards (i.e., IAEA-135, IAEA-375, IAEA-CU-2006-03, 266 IAEA-Soil-6, RGU-1, RGTh-1) prepared in the same specimen cups as the samples. 267 Efficiencies were interpolated for Be-7 energy.

To obtain information on the grain size of sediment, scandium was analysed by Instrumental Neutron Activation Analysis (INAA). Concentration in this trace element is widely used as a proxy of the fine grain size fraction of sediment (e.g., Jin et al., 2006; Dias and Prudêncio, 2008). Dried sediment subsamples (ca. 40–80 mg) were packed into tightly closed plastic bags, without any preliminary digestion. The subsamples were exposed to irradiation in the experimental nuclear reactor Orphée at the Commissariat à l'Energie Atomique (CEA; Saclay, France). The subsamples underwent a flux of thermal neutrons of

275 2.13 10^{13} n cm⁻² s⁻¹ during 30 minutes. After a 4-days cooling, four successive measurements 276 of gamma activities were carried out using HPGe detectors. Two reference materials (i.e., 277 IAEA SL-1 and Soil-7) were systematically used to cross-check the results. The uncertainty 278 on these measurements is $\leq 5\%$.

279

280 2.4. Calculating the radionuclide and sediment catchment budget

This work is based on a simple hypothesis stating that measuring the fallout input and the river output of radionuclides characterised by very high K_d values at the catchment scale allows tracing particles that adsorb those radionuclides (section 2.4.1). In a second step, the construction of a radionuclide catchment budget then provides a way to estimate the residence time of radionuclides and particles/sediment within the catchment (section 2.4.2).

286

287 2.4.1. Calculating the radionuclide and sediment input and output fluxes

Atmospheric deposition of Be-7 and Pb-210 was estimated based on the analysis of the rainfall precipitated samples, by multiplying the precipitate activity (Bq L^{-1}) by the corresponding rainfall amount (L m⁻²). Samples were collected all throughout the rainy season in order to take account of the temporal variations of the radionuclide flux. Atmospheric Cs-137 flux is currently considered as undetectable.

293 The mean radionuclide river fluxes (*C*) from each subcatchment were then calculated294 using Eq. (4):

$$295 \qquad C = C_{SM} \times (SSC + \frac{1}{K_d}) \tag{4}$$

where C_{SM} is the radionuclide activity per mass of dry matter (Bq kg⁻¹), SSC is the suspended sediment concentration (kg L⁻¹), and K_d is the distribution coefficient between particular and dissolved phases (L kg⁻¹). 299

 $K_{\rm d}$ values for the different radionuclides were discussed by Le Cloarec et al. (2007) who decided to use the following values: 8.4×10^4 for Cs-137, 5×10^4 for Pb-210, and 10×10^4 300 301 for Be-7.

- 302
- 303 2.4.2. Modelling the sediment residence times

304 After constructing simple one-box radionuclide balance models using Cs-137 or Pb-305 210 to estimate the residence time of radionuclides and sediment in the upstream part of the 306 Rhône River catchment (Switzerland), Dominik et al. (1987) proposed to integrate Be-7 into 307 this model to benefit from potential additional information provided by this radionuclide on 308 the rapid delivery of sediment to the river. However, because of its short half-life, the bulk of 309 Be-7 would decay in the soil of the catchment before reaching the river. To overcome this 310 problem, Dominik et al. (1987) developed a two-box balance model. The catchment surface 311 was then subdivided into two boxes: (i) a "soil-box" (which is referred to by an s subscript in 312 the remainder of the text and equations) characterised by an S_s -area, low transport velocities and hence long radionuclide residence times (the soil box comprises the uppermost soil 313 314 surface submitted to radionuclide fallout) and (ii) a "river-box" (which is referred to by an r315 subscript in the remainder of the text and equations) covering the river area and its 316 surroundings (S_r) characterised by faster exchanges and shorter radionuclide residence times. 317 Input and output fluxes of sediment and radionuclides were taken into account in each box. 318 Radioactive decay during the residence time of sediment was also considered. The initial 319 version of the model hypothesised that radionuclides were exclusively transported on soil 320 particles, whereas the version developed by Le Cloarec et al. (2007) takes into account the 321 radionuclide dissolved fractions transported in the river using the K_d values. 322 Total atmospheric input fluxes (F_a) were estimated based on the rainfall precipitated

323 samples, and the fraction present in each box was also determined (Eq. 5):

$$324 \qquad S_r + S_s = 1 \tag{5}$$

325 Mass balance equations were calculated for each reservoir. In the soil box, loss of material is 326 due to material transfer to the river box or to radioactive decay (Eq. 6):

$$327 F_a \times S_s = I_s \times (k_s + \lambda) (6)$$

328 where k_s is the rate of output transfer from the soil box (related to the residence time τ_s in the box, with 329 $k_s = 1 / \tau_s$); I_s is the radionuclide inventory in the soil box; and λ is the constant of radioactive decay. 330

331 In the river box, atmospheric inputs were added to the flux coming from the soil box. Both

fluxes were required to balance the output from the river and the radioactive decay (Eq. 7):

333
$$F_a \times S_s + I_s \times k_s = I_r \times (k_r + \lambda)$$
(7)

where I_r is the radionuclide inventory in the river box and k_r is the rate of output transfer from the river box.

336

337 The flux exported from the river box (F_r) was then calculated using Eq. (8).

$$338 F_r = I_r \times k_r (8)$$

339

In the initial version of the model (Dominik et al., 1987), Eqs. (5) to (8) were only calculated
for Be-7 and excess-Pb-210. In the version adapted by Le Cloarec et al. (2007), additional
equations were introduced for Cs-137. This simultaneous use of the three radionuclides
allowed solving the system of equations.

For each radionuclide, five variables had to be determined (S_r , k_r , k_s , I_s , and I_r). Additional assumptions were necessary to solve Eqs. (5) to (8). They were based on the comparison between λ -values and the expected k_s and k_r values. First, we hypothesised that the residence time in the soil box was expected to be an order of magnitude of decades to centuries. Most Be-7 ($T_{1/2} = 53$ d) will therefore be lost by decay and its export from the soil box was considered as negligible compared to atmospheric fluxes ($k_s \ll \lambda_{Be}$ and (I_s)_{Be}× $k_s \ll 350$ $S_r \times (F_a)_{Be}$). Second, export of material from the river box was expected to remain in the same order of magnitude as the residence time of sediment within the river, which is significantly more rapid than the decay of excess-Pb-210. Decay of this radionuclide could therefore be neglected ($\lambda_{Pb} \ll k_r$).

A relationship between the river box area (S_r) and the associated removal rate of material (k_r) could be derived based on the assumption on Be-7 and by combining Eqs. (7) and (8):

357
$$S_r = \left(\frac{F_r}{F_a}\right)_{Be} \times \left(\frac{\lambda_{Be}}{k_r + 1}\right)$$
(9)

Based on the hypothesis for excess-Pb-210 and combining the mass balance equations of Be-7 and excess-Pb-210, the residence time of sediment could be determined (Eq. 10):

361 where
$$R_r = \frac{(F_r)_{Be}}{(F_r)_{Pb}}$$
 and $R_a = \frac{(F_a)_{Be}}{(F_a)_{Pb}}$

Another relationship implicating τ_s and τ_r was derived from the mass balance equations of Cs-137. Given the atmospheric flux of Cs-137 is currently negligible ($(F_a)_{Cs} = 0$), its inventory in soils is well known (total inventory of 1130 (± 250) Bq m⁻² according to measurements carried out in 2009 in the study sites). Based on Eq. (7), we could then determine τ_s (Eq. 11):

367 with
$$M = (I_s)_{Cs} + (I_r)_{Cs}$$

Finally, the combination of Eqs. (10) and (11) allowed for determining
$$\tau_r$$
 (Eq. 12):

370
$$a(\tau_r)^2 + b(\tau_r) + c = 0$$

371 with

372 $a = -(F_r)_{Cs} \times \left(\frac{R_r}{R_a}\right) \times \lambda_{Be} \times \lambda_{Pb}$

373
$$b = M \times \left(\frac{R_r}{R_a}\right) \times \lambda_{Be} \times \lambda_{Pb} - (F_r)_{Cs} \times \lambda_{Pb} \times \left[\frac{(R_r)}{(R_a)} - 1\right] - (F_r)_{Cs} \times A \times \lambda_{Cs}$$

374
$$c = \lambda_{Pb} \times M \times \left[\frac{R_r}{R_a} - 1\right] - A \times (F_r)_{Cs} \times A = 1 - \left(\frac{F_a}{F_r}\right)_{Pb}$$

375

A sensitivity analysis of the model was carried out by Le Cloarec et al. (2007). They showed that the major uncertainty was the one associated with SSC measurements. In this study, this uncertainty reached 15 to 30% (Némery et al., 2010). Overall, the uncertainties on soil particle and sediment residence times provided by the model were estimated at 30%.

381 2.6. Estimation of suspended sediment age and fraction of new sediment

382

The methodology proposed by Matisoff et al. (2005) was followed to determine the age of sediment (*t*; Eq. 13) and the percentage of "new" suspended sediment in the river (Eq. 14):

$$386 t = \frac{-1}{\left(\lambda_{7Be} - \lambda_{210Pb}\right)} \ln\left(\frac{A}{B}\right) + \frac{1}{\left(\lambda_{7Be} - \lambda_{210Pb}\right)} \ln\left(\frac{A_0}{B_0}\right) (13)$$

387 where λ_{7Be} and λ_{210Pb} are the decay constants of ⁷Be and ²¹⁰Pb (d⁻¹); *A* and *B* are the ⁷Be and 388 ²¹⁰Pb _{xs} activities in the suspended sediment (Bq kg⁻¹); *A*₀ and *B*₀ are the ⁷Be and ²¹⁰Pb_{xs} activities in 389 rainfall (Bq L⁻¹).

390 % of "new" sediment =
$$100 \times \frac{(A/B)}{(A_0/B_0)}$$
 (14)

(12)

392	Both parameters were calculated for all the floods sampled in the three study sites throughout
393	the 2009 rainy season. This second approach was hence simpler than the two-box model. A
394	decrease in the Be-7/excess-Pb-210 ratio can then alternatively be explained by (i) an increase
395	in the sediment residence time (given Be-7 decay is faster than excess-Pb-210 decay) or by
396	(ii) the dilution of sediment enriched in Be-7 by sediment poorer in Be-7. Consequently,
397	calculation of this ratio does not theoretically allow deciding whether aging or dilution of
398	sediment occurred in the river.
399	
400	3. Results and discussion
401	
402	3.1. Main runoff and erosion trends during the 2009 rainy season
403	
404	Overall, the total rainfall depth measured in the monitored subcatchments during the
405	2009 rainy season can be considered as "normal" (593–1169 mm) compared to the long-term
406	(1954–2004) rainfall database (400–1100 mm yr ⁻¹) available for Santiago Undameo, located
407	just upstream of the Cointzio reservoir (Gratiot et al., 2010). Rainfall amount strongly varied
408	from one subcatchment to another. La Cortina received much more rainfall (1169 mm) than
409	Potrerillos (765 mm) and Huertitas (593 mm). Furthermore, two storms that occurred in June
410	2009 were unrecorded in Huertitas because of technical problems. This partly explains the
411	lower rainfall amount recorded in Huertitas compared to the two other subcatchments. Most
412	of the recorded events consisted of heavy storms associated with vertical convective cloud
413	systems and characterised by rainfall intensities reaching up to 120 mm h ⁻¹ in 5 min. Mean
414	rainfall erosivity was more than twofold higher in La Cortina (mean of 434 J m^{-2} mm ⁻¹ during
415	the season) than in Potrerillos (204 J $m^{-2} mm^{-1}$) and Huertitas (214 J $m^{-2} mm^{-1}$) (Table 2).

416 These storms generated variable runoff volumes in the three study areas. Runoff 417 coefficients were extremely low in La Cortina (mean of 0.8%), except during the first 418 significant storm of the year (3.9% on 14 July 2009). In contrast, mean runoff coefficients 419 observed in Potrerillos and Huertitas were close to 7%, with higher coefficients observed 420 during the heaviest storms (up to 14.0% in Huertitas and 17.8% in Potrerillos; Table 2). 421 Furthermore, sediment export fluctuated strongly from one subcatchment to another. Overall, specific SSY was by far the most important in Huertitas (900–1500 t km⁻² y⁻¹) and Potrerillos 422 (600–800 t km⁻² y⁻¹), whereas it was particularly low in La Cortina (30 t km⁻² yr⁻¹; Duvert et 423 424 al., in press). This major difference in erosion exports from the different subcatchments can 425 be attributed to numerous factors. First, different land uses and soil types are observed in the 426 different subcatchments (Fig. 2). A much higher proportion (52%) of the La Cortina 427 subcatchment is indeed covered by forests, which are traditionally known to produce less 428 sediment than grassland and cropland when they are not submitted to deforestation, logging, 429 or frequent fires (e.g., Inbar et al., 1998; Prosser and Abernethy, 1999). Most soils in this 430 subcatchment are Andisols, which are very permeable and generate less runoff than Acrisols. 431 Furthermore, in cropland that covers 28–46% of the surface in the study sites, the bulk of 432 erosion generally occurs when erosive storms fall on particularly erodible soils, which is 433 typically the case at the beginning of the crop growing season (i.e., May-July). In 2009, 61– 434 64% of total sediment export occurred during this period in La Cortina and Potrerillos, but 435 only 46% in Huertitas (Table 2). This is probably due to the occurrence of particularly heavy 436 storms in September in Huertitas (Table 2a).

Furthermore, SSY were higher in catchments with steeper slopes: 42% of the slopes in the Huertitas subcatchment are indeed $\geq 10^{\circ}$, compared to only 29% in Potrerillos and 21% in La Cortina (Table 1). The catchment size can also explain these SSY differences. In Potrerillos, the larger number of potential traps prevents sediment from reaching the river.

441 Collection of suspended sediment was carried out during most of the storms that occurred in
442 2009 (Table 2), corresponding to 77–87 % of the total sediment export from the different
443 subcatchments (Duvert et al., in press). We can then confidently consider that our results are
444 representative of the entire 2009 rainy season.

445

446 *3.2. Radionuclide atmospheric fluxes*

447

448 Atmospheric fluxes of radionuclides were considered as homogeneous in the Cointzio catchment, given that they are mainly controlled by latitudinal variations (e.g., Brost et al., 449 450 1991). A relationship between rainfall depth and radionuclide fallout could be derived for 451 excess-Pb-210 and Be-7 (Fig. 5). A moderate to good correlation $(0.71 < R^2 < 0.76)$, 452 significant at p < 0.005) was found between rainfall and fallout radionuclides, which is 453 consistent with the coefficients reported in the literature (e.g., Caillet et al., 2001). The linear 454 relationship between radionuclide activity and precipitation (R; mm of rainfall) allowed calculating atmospheric Be-7 (Eq. 14) and excess-Pb-210 (in GBq km⁻²; Eq. 15) depositions 455 456 (Fig. 5). (\mathbf{T}) (10^{-3}) (10^{-3})

457
$$(F_a)_{Be-7} = 10^{-3} \times (1.4R - 1.9)$$
 $(R^2 = 0.71; p < 0.005)$ (14)

458 $(F_a)_{xs-Pb-210} = 10^{-3} \times (0.09R - 0.4)$ $(R^2 = 0.76; p < 0.005)$ (15)

459 La Cortina received the highest radionuclide atmospheric fluxes, which is explained460 by the higher rainfall depth measured in 2009 in this subcatchment (Table 3).

461

462 *3.3. Radionuclide river fluxes*

463

464 Total radionuclide river fluxes calculated by combining radionuclides fixed on
465 sediment and dissolved radionuclides transported in the river water were found to be strongly

466	dependent on the suspended sediment concentration ($0.7 < R^2 < 0.8$ significant at $p < 0.001$;
467	data not shown). The variation of these fluxes can hence be partly explained by strong SSC
468	variations between the different subcatchments (Table 4). The lowest Be-7 and Pb-210 river
469	fluxes were observed in La Cortina. In this subcatchment, the contribution of the atmospheric
470	fluxes to the total radionuclide input was much more important than in the two other
471	subcatchments (Table 4). Regarding Cs-137, the river flux was also the lowest in La Cortina.
472	
473	3.4. Modelling the radionuclide content in sediment
474	
475	To strengthen the results provided by the model, we must verify that the sediment
476	particle size remained constant throughout the 2009 rainy season. Particle size, which could

481 ppm).
482 The model provided several useful insights over sediment residence times in the three

be estimated using the scandium concentrations as a proxy for the fine fraction present in

sediment collected at the outlet of each subcatchment, was constant in 2009, as demonstrated

by the low standard deviation observed around the mean scandium concentrations measured

in sediment collected in La Cortina (22±1 ppm), Potrerillos (19±2 ppm) and Huertitas (21±1

483 subcatchments (Table 5).

477

478

479

480

The model calculated a mean residence time of sediment in both rapid and slow boxes. Mean residence time of soil particles in the slow box ranged between 5000 (\pm 1500) and 23,300 (\pm 7000) years. The longer residence time was calculated for La Cortina (23,300 \pm 7000 years), whereas it was the shorter in Huertitas (5000 \pm 1500 years). In Potrerillos, an intermediate residence time was calculated (13,300 \pm 4000 years). These results were consistent with the slope characteristics and the intensity of erosion observed in these subcatchments (Table 1). Residence times were shorter in Huertitas, which is the steepest site 491 and the most affected by the development of deep gullies (Fig. 3). In contrast, residence times 492 were the longest in the most forested site characterised by the gentlest slopes (i.e., La 493 Cortina). Even though the surface of the monitored subcatchments remained in the same order 494 of magnitude $(3-12 \text{ km}^2)$, the range of residence times in the soil boxes was as wide as the 495 range calculated for different nested catchments within the entire Seine basin (between 4800 496 years in the first order catchments and up to 30,000 years in the entire Seine river basin; Le 497 Cloarec et al., 2007). Soil erosion and its consequences on residence times of particles in soils 498 were hence very variable in the three Mexican sites, which outlines their different hydrosedimentary behaviour. Within a few kilometres, erosion fluctuated indeed between low 499 rates (30 t km⁻² yr⁻¹ in La Cortina) and particularly severe rates (900–1500 t km⁻² y⁻¹ in 500 501 Huertitas).

502 Once it reached the river (i.e., rapid box of the model), sediment was rapidly exported 503 from the subcatchments. This export occurred on average in less than one rainy season, with 504 mean residence times of sediment in the river channel ranging between 50 (\pm 30) days in 505 Potrerillos and 200 (\pm 70) days in La Cortina. The intermediate mean residence time 506 estimated in Huertitas (60 ± 40 days) was not significantly different from the one calculated in 507 Potrerillos. This residence time was highly dependent on the occurrence of heavy storms, 508 leading to a significant proportion of the annual sediment yield. Mean residence times of 509 sediment estimated in the Mexican rivers remained in the same order of magnitude as the 510 ones measured in the Rhône mountainous basin (1–220 days; Dominik et al., 1987), whereas 511 they were significantly shorter than the ones estimated in upstream rivers of the lowland Seine 512 River basin (200–500 days; Le Cloarec et al., 2007).

513 The model also calculated the proportion of the subcatchment area belonging to the 514 "rapid box", i.e., the river channel and the directly connected areas. This proportion reached 515 $0.3\pm0.1\%$ of the subcatchment surface in La Cortina where the river is located in a narrow (~

516 1 m) and incised channel flowing through forests and cropland. Despite a high rainfall amount 517 in 2009 (1169 mm), SSY from this subcatchment remained low (30 t km⁻² y⁻¹); and the rapid 518 box was hence logically characterised by a small surface and rather long sediment residence 519 times (mean of 200 days compared to 50–60 days in the two other sites). Furthermore, the 520 absence of severely eroded areas in this subcatchment was confirmed by the very limited 521 surface of the rapid box calculated by the model.

522 In Potrerillos, the proportion covered by the rapid box was slightly higher $(1.5\pm0.5\%)$, 523 which is explained by the flowing of the river through a wider (2-5 m) channel across 524 rangeland, forests and cropland. In contrast, this surface proportion reached 4.5±1.5% in 525 Huertitas. This proportion of the subcatchment surface remained in the same order of 526 magnitude as the catchment proportion affected by severe erosion (6%). Hence, the model 527 associated the river bank gully systems, which are directly connected to the river, with the 528 rapid (i.e., river) box to evacuate all the sediment produced. Conceptually, we can indeed 529 reasonably consider that this gully surface sediment is rapidly transferred to the river, which 530 explains the different hydrosedimentary behaviour of the Huertitas subcatchment.

531 The particularly low content in Cs-137 (< 0.3 Bq kg⁻¹; Table 2) measured in several 532 samples collected at the outlet of Huertitas and Potrerillos led to model uncertainties specific 533 to regions with soils characterised by very low activities in Cs-137 (i.e., Mexico). Additional 534 measurements were therefore conducted on several suspended sediment samples using the 535 very low background, well-type gamma detectors available at the Laboratoire Souterrain de 536 Modane (Reyss et al., 1995). Overall, these results showed the presence of very low Cs-137 537 activities $(0.2 \pm 0.1 \text{ Bq kg}^{-1})$ in this sediment. To investigate uncertainties associated with 538 these very low activities, we attributed successively different Cs-137 activities (0, 0.1, and 0.2 Bq kg⁻¹) to the samples in which results were under the detection limits in Gif-sur-Yvette (i.e., 539 540 typically < 0.3 Bq kg⁻¹) but detectable in Modane. The model results reported in Table 5 were

541	calculated using a Cs-137 activity of 0.2 Bq kg ⁻¹ . This change of the Cs activities mostly led
542	to a ca. 15% variation of the particle residence times in the soil box. However, all variations
543	affecting the model outputs systematically remained within the 30% model uncertainties
544	mentioned above.

545

546 3.5. Suspended sediment age and fraction of "new" sediment

547

548 At the beginning of the rainy season, the rivers contained an important stock (74–88%) 549 of older sediment deposited during the previous year (Fig. 6). This older sediment stock was 550 progressively evacuated by the successive floods. Despite this feature common to the three 551 study sites, two distinct hydrosedimentary behaviours could be outlined (Fig. 7).

552 In La Cortina, sediment export was mainly achieved by the first major flood of the rainy season (14 July; Fig. 6B). This storm was intense (438 J m⁻² mm⁻¹) but not exceptional, 553 554 given that rainfall intensity reached higher levels in La Cortina later during the season (up to 763 J m⁻² mm⁻¹ on 8 September; Table 2B). During this first storm, only 12±4% of new 555 556 sediment were provided to the river. In contrast, this storm strongly contributed to the flush of 557 the sediment accumulated in the river channel during the previous season. The supply of 558 "new" sediment to the river remained continuous and stable during the rest of the season, 559 regardless of storm erosivity (Fig. 6B). Only 11-21 % of "new" sediment were observed 560 throughout the rainy season in the river channel of this site. Subsequent export of recent 561 sediment from this forested subcatchment occurred at a rather constant and low rate (Fig. 7B). 562 According to the calculation of the Be-7/excess-Pb-210, the mean residence time of sediment 563 in the river channel reached 163±14 days. This value remained in the same order of 564 magnitude as the one calculated by the radionuclide balance model $(200\pm70 \text{ days}; \text{Table 5})$.

In contrast, in Huertitas and in Potrerillos, sediment residence time and the percentage 565 566 of new sediment greatly fluctuated all throughout the rainy season. In Huertitas, sediment 567 residence time in the river channel varied between 1 and 194 days. Evolution of this residence 568 time displayed a sawtooth behaviour (Fig. 6A). For instance, the heavy storm that occurred on 569 3 September generated a dramatic supply of "new" sediment to the river (the proportion of 570 new sediment in the river channel increased from $24\pm3\%$ to $76\pm19\%$). This storm and the 571 next events led to a complete flush of the river sediment stock and to the export of older 572 sediments. Sediment residence times in the river dropped from 124 ± 11 days to 38 ± 37 days 573 between 13 August and 4 September. Conceptually, this result means that, even though most 574 sediment is gradually exported from Huertitas through a succession of deposition and 575 resuspension steps, it can be exported from the catchment by a single heavy storm. The storm observed on 3 September was indeed the most intense of the year in Huertitas (487 J m⁻² 576 577 mm⁻¹; Table 2a). A similar fluctuating behaviour was observed in Potrerillos. Complete 578 "sediment flush" of the river channel was observed twice in 2009 (on 17 August and 12 579 September; Fig. 6C). This sediment export was observed after particularly intense storms (> 580 200 J m⁻² mm⁻¹; Table 2C). Overall, Huertitas and Potrerillos were characterised by much 581 shorter sediment residence times in the river channel (111±42 days in Huertitas; 101±46 days 582 in Potrerillos; Figs. 6A-C). This finding is consistent with the mean sediment residence time 583 in the river calculated by the model (60 ± 40 days in Huertitas; 50 ± 30 days in Potrerillos; Table 584 5).

Regarding SSY, the first heavy storm of the rainy season — which traditionally
coincides with a period characterised by a sparse cover of the soil by vegetation — led to the
highest sediment export of the year. This was the case in the three monitored subcatchments.
In contrast, during the rest of the season, Huertitas and Potrerillos were much more reactive
than La Cortina (Fig. 7). Sediment stock in the river channel of these subcatchments was

hence successively renewed and evacuated during the rainy season. In contrast, La Cortina
returned to a rather "dormant" hydrosedimentary behaviour after the first heavy storm of the
season.

593

594 *3.6. Radionuclide inventories*

595

The model also provided the total radionuclide inventory in each "reservoir" (Table 6). The $(I_r/I_s)_{(Be)}$ ratio, which indicates that the Be-7 fraction originated from the atmosphere reached the river, was in the same order of magnitude as the proportion of the subcatchment surface belonging to the rapid (river) box (S_r ; Table 5).

600

601 3.7. General discussion: control of flood type, rainfall erosivity and land use on sediment
602 export and potential implications for soil conservation

603

604 We could not detect any clear effect of rainfall erosivity on sediment export. Overall, 605 in 2009, heaviest storms occurred in La Cortina (KE_{mm} between 92 and 763 J m⁻² mm⁻¹) but 606 this higher erosivity was not reflected by higher SSY. The highest SSY were indeed observed in Huertitas and Potrerillos where rainfall was characterised by lower erosivities (58-520 J 607 m⁻² mm⁻¹ in Huertitas and 42–463 J m⁻² mm⁻¹ in Potrerillos). Furthermore, high values of 608 609 KE_{mm} were never associated with high SSC peaks in the three catchments. Duvert et al. (in 610 press) conducted a factorial analysis on several parameters (e.g., flood type, antecedent 611 moisture conditions, discharge, rainfall, etc.) collected in the three subcatchments to outline 612 the driving factors of SSY. They showed that parameters associated with precipitation were 613 absolutely not related to both sediment and discharge parameters, probably because of the

high spatial variability of rainfall. In contrast, peak discharge was significantly associatedwith exported loads.

616 We can outline important insights on sediment dynamics in the river based on the 617 flood typology developed by Duvert et al. (in press). They classified the floods that we 618 analysed into three types: (i) events characterised by a lag between sediment peak and peak 619 discharge (referred to as "LaP" events); (ii) events characterised by the coincidence between 620 sediment peak and peak discharges (referred to as "SP" events) and (iii) events characterised 621 by a sediment peak leading the peak discharge (referred to as "LeP" events). Sediment 622 maintained high concentrations during the recession phase of those three flood types, 623 indicating that their export was rather "transport-limited" than "sediment-limited". Table 2 624 provides the typology attributed to each flood by Duvert et al. (in press). We could associate 625 those different flood types with indications on the sediment age and distinct hydrosedimentary 626 behaviours.

627 In Huertitas and in La Cortina, we only analysed one LaP flood. It corresponded to the 628 first significant flood of the rainy season (16 May in Huertitas, 14 July in La Cortina). This 629 flood refilled the river network with sediment and started flushing the sediment deposited in 630 2008 (74±4% of the flood export in Huertitas and 88±0.5% in La Cortina). This initial flush 631 was also associated with a limited export of sediment directly eroded from the hillslopes 632 (26±4% in Huertitas; 12±0.5% in La Cortina). In La Cortina, this event exported the bulk of 633 annual SSY, and was associated with the highest runoff coefficient of the season (3.9%). This 634 confirms the low sediment availability in the river draining this subcatchment. In Potrerillos, 635 LaP floods supplied recent sediment to the system. Proportion of "new" sediment increased 636 from 27±3% to 78±17% after these floods. In contrast, SP floods described as "overflow 637 waves" by Duvert et al. (in press) were associated with large runoff coefficients in Huertitas 638 and Potrerillos (4–16%) and exported a significant proportion of sediment in 2009 (37% of

639 SSY in Huertitas and even 77% in Potrerillos). The large dominance of SP floods (78%) in
640 Potrerillos illustrates the very reactive behaviour of this catchment. Finally, floods of LeP
641 type mostly evacuated the sediment stock available in the channel.

Given that sediment transport was rather "transport-limited" and that sediment
exhaustion was not detected in 2009, we can infer the important role played by land use on
sediment export. In the two subcatchments were gullies are found, they provided a constant
source of sediment to the river. This difference mostly explains the variation in sediment
export between Huertitas and Potrerillos — characterised by extensive gully networks — and
La Cortina.

648 Furthermore, although they were relatively low, Cs-137 activities measured in this 649 Mexican catchment were very contrasted, and they therefore allowed outlining the main 650 contributions of the different land uses to the sediment production (Table 7). In Huertitas, the Cs activity associated with gullies was extremely low (0–0.2 Bq kg⁻¹), which can be explained 651 652 by the continuous export of fine sediment to the river since the Cs-137 deposition in the 1960s 653 and by the depletion of the Cs stock in these gully systems (Fig. 3). In contrast, a significant Cs activity was measured in cropland $(2.1 \pm 0.3 \text{ Bg kg}^{-1})$. In Potrerillos, a similar Cs activity 654 655 was observed in cropland $(2.2\pm0.3 \text{ Bq kg}^{-1})$, whereas rangeland was characterised by low but detectable — Cs concentrations (0.6 ± 0.3 Bg kg⁻¹). Finally, Cs activities measured in La 656 657 Cortina were significantly higher than in the two other catchments, confirming the lower depletion of the Cs stock by erosion. They fluctuated between 3.7 ± 1.2 Bq kg⁻¹ in cropland and 658 11.9 ± 5.8 Bq kg⁻¹ under forests. 659

We can compare activities measured in potential sediment sources with the ones measured in suspended sediment at the outlet. In Huertitas, Cs-137 activities measured in suspended sediment collected at the outlet were low $(0.1-1.1 \text{ Bq kg}^{-1})$. These results confirmed the limited contribution of upstream cropland to erosion. The bulk of sediment is

664 hence produced by the historical gully networks that grow by regressive erosion and that are 665 characterised by low or very low Cs-137 activities. Gullies were probably formed due to the 666 association of traditional cropping practices with cattle grazing on hillslopes. They are now 667 partly stabilised but they keep extending by regressive erosion and conveying sediment from cropland to the river (Bravo-Espinoza et al., 2009). However, the higher contribution of 668 669 upstream cropland to erosion cannot be excluded during heavy storms. This was typically the 670 case at the beginning and at the end of the rainy season, when larger Cs activities were 671 measured in sediment collected at the outlet and when higher runoff coefficients were 672 observed (Table 2C).

673 In Potrerillos, low Cs-137 activities were also detected at the outlet, indicating that the 674 contribution of cropland to erosion must remain limited. Contribution of cropland to erosion 675 was also probably restricted to the most intense storms (e.g., on 11 September) or during the 676 evacuation of sediment eroded by a succession of erosive storms (e.g., on 23 July; Table 2A). 677 In La Cortina, Cs-137 activities were clearly higher than at the outlet of the two other subcatchments. Cs-137 concentrations (mean values of 1–3.7 Bq kg⁻¹) remained in the same 678 679 order of magnitude as the Cs-137 concentrations measured in samples collected from 680 cropland in this subcatchment (mean values of 3.1–4.1 Bq kg⁻¹).

681 Obtaining an estimation of the sediment residence times in catchments and rivers is 682 important to implement the appropriate conservation measures. Our results show that 683 stabilising the historical gully systems observed in Huertitas should be a priority, given that 684 they are characterised by much shorter residence times of sediment and that they produce the 685 bulk of sediment exported from this subcatchment. They also keep growing by regressive 686 erosion at the expense of cropland. In La Cortina, the major contribution of avocado and 687 maize fields to erosion outlines the need to carry out on-site soil conservation measures. In 688 Potrerillos, the limitation of cattle density could be envisaged to limit sediment export to the

river and to prevent the further extension of gullies to cropland. In this subcatchment, fine soil
particles are indeed removed by cattle trampling, which leads to an increase in the surface of
bare soils (Bravo-Espinoza et al., 2009).

692

693 **5.** Conclusions

694

695 This study combined for the first time the monitoring of suspended sediment transport 696 in rivers and the measurement of fallout radionuclides (Be-7, Cs-137, excess-Pb-210) in 697 rainfall, soil, and suspended sediment samples collected in three subcatchments representative 698 of the environments draining to a reservoir located in tropical highlands of the transvolcanic 699 Mexican belt. It provided a novel continuous hydrosedimentary monitoring of three 700 contrasted catchments all throughout the rainy season. Soil particle residence time calculated 701 by a radionuclide two-box balance model ranged between 5,000 (\pm 1,500) years in the 3 km² 702 steeper catchment dominated by gully networks and cropland, and 23,300 (\pm 7,000) years in the 9-km² more gentle forested catchment. Once it reached the river, sediment was 703 704 systematically exported from the catchments within a mean period ranging between 50 (\pm 30) 705 and 200 (\pm 70) days. The first heavy storm of the rainy season led to the flush of sediment 706 accumulated in the river channel during the previous season in the three study sites. Then, the 707 flood type and the land use of the different subcatchments proved to control sediment export 708 during the season. In a context of rapid siltation of the Cointzio reservoir that is crucial and 709 strategic for the water supply in the region, the local authorities should concentrate their 710 efforts to stabilise the gully systems, to prevent their extension by regressive erosion at the 711 expense of cropland, and to encourage the implementation of conservation measures in 712 cropland of the most reactive catchments (i.e., Huertitas, Potrerillos). Further research should 713 now focus on fingerprinting and quantifying the contribution of the main sediment sources

714	within the most reactive catchments in order to outline the areas where the conservation
715	efforts and the limited subsidies available should be concentrated.
716	
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894	Figure captions
895	
896	Fig. 1. Location of the monitored subcatchments (Huertitas, Potrerillos, La Cortina) within the
897	Cointzio catchment (outlet located at 19°37'46''N / 101°15'35''W).
898	
899	Fig. 2. Land uses in the study sites derived from the map drawn by Mendoza and López-
900	Granados (2007).
901	
902	Fig. 3. Partial view of the Huertitas subcatchment, showing the gully systems, cropland and
903	rangeland (picture taken on 25 June 2009).
904	
905	Fig. 4. Conceptual sketch of the radionuclide mass-balance model improved by Le Cloarec et
906	al. (2007).
907	
908	Fig. 5. Relationships between rainfall and radionuclide atmospheric fallout in the Cointzio
909	catchment: (A) Be-7; (B) excess-Pb-210.

- 910
- 911 Fig. 6. Suspended sediment age and fraction of new sediment in suspension in the three study
- 912 sites throughout the 2009 rainy season.
- 913
- 914 Fig. 7. Suspended sediment yields from the three study sites and value of the Be-7/excess-Pb-
- 915 210 ratio throughout the 2009 rainy season.

Fig. 1



Figure 2



Fig. 3



Fig. 4



Fig. 5



(A)







(C) Potrerillos





(C) Potrerillos



Altitude, soil, slo	pe, and erosion	characteristics	of the	study	sites
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Subcatchment	Area (km ²)	Altitude range (m)	Soil types ^a Slope (% of catchment area) Sever			Severely eroded areas ^b		
				0 – 9°	10 - 20	$0^{\circ} > 20^{\circ}$	(% of the catchment surface)	
<u> </u>								
Huertitas	3	2150 - 2450	Acrisols (100%)) 58%	37%	5%	6	
La Cortina	9	2250 - 2700	Andisols (100%) 79%	20%	1%	0	
Potrerillos	12	2200 - 2700	Acrisols (60%)	71%	28%	1%	1	
			Andisols (40%)					

^a According to FAO (2006).

^b Derived from the analysis of aerial photographs.

Table 2

Table 2 Rainfall, runoff, and sediment characteristics of the floods sampled at the outlet of the three study sites in 2009^a

Event date	Rainfall (mm)	Runoff (mm)	RC (%)	SSY (t)	KE (J m ⁻² mm ⁻¹)	xs-Pb-210 (Bq kg ⁻¹)	Cs-137 (Bq kg ⁻¹)	Be-7 (Bq kg ⁻¹)	Flood type
(a) Huer	titas (n = 13	5)							
16/05/2009	12.2	0.3	2.5	54	n/a	6 (±1)	0.4 (±0.1)	29 (± 1)	LaP
26/06/2009	21.0	1.9	9.2	124	220	15 (±3)	0.5 (±0.3)	29 (± 4)	SP
11/07/2009	25.8	1.4	5.3	57	263	11 (±3)	0.5 (±0.5)	45 (± 4)	LeP
24/07/2009	9.4	0.8	8.2	48	n/a	13 (±2)	< 0.3	71 (± 4)	LeP
25/07/2009	12.9	0.7	5.4	36	n/a	8 (±2)	< 0.2	70 (± 3)	LeP
04/08/2009	20.8	1.1	5.3	68	206	10 (±2)	0.4 (±0.2)	46 (± 3)	LeP
13/08/2009	8.0	0.2	2.5	6	58	16 (±4)	< 0.4	71 (± 5)	LeP
03/09/2009	29.2	1.2	4.1	71	487	4 (±2)	< 0.1	56 (± 3)	LeP
04/09/2009	14.0	2.0	14.0	124	232	< 3.0	0.3 (±0.1)	27 (± 3)	SP
09/09/2009	7.8	1.0	12.8	6	n/a	18(±4)	< 0.3	75 (± 5)	LeP
21/09/2009	35.3	4.2	11.9	87	180	12 (±3)	< 0.3	43 (± 4)	LeP
30/09/2009	12.4	0.7	5.6	5	310	12 (±4)	0.6 (±0.4)	71 (± 6)	LeP
14/10/2009	16.8	0.8	4.6	17	261	19 (±2)	0.9 (±0.2)	62 (± 4)	LeP
(b) La Co	ortina (n = 8	3)							
14/07/2009	15.2	6.1	3.9	149	438	40 (± 4)	2.8 (± 0.3)	86 (± 5)	LaP
25/07/2009	13.8	0.4	0.2	5	272	50 (± 8)	$2.5 (\pm 0.9)$	179 (± 15)	LeP
03/08/2009	19.2	0.9	0.5	15	377	55 (± 7)	3.3 (± 0.6)	156 (± 12)	LeP
05/08/2009	36.6	1.6	0.4	33	492	24 (± 3)	$2.4 (\pm 0.3)$	66 (± 4)	LeP
13/08/2009	34.4	1.4	0.4	13	347	48 (± 3)	$2.0 (\pm 0.3)$	154 (± 6)	SP
25/08/2009	41.6	1.5	0.3	14	386	39 (± 3)	$2.0 (\pm 0.3)$	$100 (\pm 4)$	LeP
04/09/2009	46.0	0.7	0.2	3	396	45 (± 4)	$0.7 (\pm 0.5)$	117 (± 7)	LeP
08/09/2009	18.8	0.8	0.4	1	763	58 (± 7)	3.7 (± 1.6)	125 (± 10)	LeP

Event date	Rainfall (mm)	Runoff (mm)	RC (%)	SSY (t)	KE (J m ⁻² mm ⁻¹)	xs-Pb-210 (Bq kg ⁻¹)	Cs-137 (Bq kg ⁻¹)	Be-7 (Bq kg ⁻¹)	Flood type
(c) Potrerillos (n = 18)									
01/07/2009	17.4	1.3	9.6	1326	195	5 (± 2)	0.5 (± 0.1)	16 (± 2)	SP
11/07/2009	9.6	1.0	13.1	290	234	8 (± 2)	< 0.2	38 (± 2)	SP
15/07/2009	18.4	1.4	9.6	394	194	< 4	$0.3 (\pm 0.1)$	31 (± 2)	SP
16/07/2009	11.8	0.8	8.2	176	158	15 (± 2)	< 0.2	39 (± 3)	SP
17/07/2009	16.0	0.8	6.4	175	223	6 (± 1)	$0.3 (\pm 0.1)$	54 (± 2)	SP
19/07/2009	21.2	1.2	7.1	240	463	5 (± 1)	$0.3 (\pm 0.1)$	37 (± 2)	SP
23/07/2009	3.6	0.2	7.2	23	94	31 (± 6)	1.1 (± 0.6)	83 (± 9)	SP
26/07/2009	5.6	0.2	4.6	29	149	14 (± 4)	< 0.3	68 (± 5)	SP
28/07/2009	10.0	0.3	3.8	50	146	17 (± 4)	< 0.4	50 (± 4)	SP
04/08/2009	11.8	0.3	3.1	39	164	19 (± 4)	$1.0 (\pm 0.5)$	85 (± 7)	LaP
06/08/2009	9.2	0.5	6.4	63	179	16 (± 3)	$0.5 (\pm 0.3)$	80 (± 4)	SP
17/08/2009	10.4	1.4	17.8	320	227	< 3	$0.2 (\pm 0.1)$	43 (± 2)	LeP
22/08/2009	11.6	1.3	14.6	280	314	< 4	$0.3 (\pm 0.2)$	40 (± 2)	SP
03/09/2009	11.2	1.4	16.0	531	161	< 3	$0.4 (\pm 0.1)$	35 (± 3)	LeP
04/09/2009	5.0	0.5	13.3	84	205	6 (± 2)	< 0.3	42 (± 4)	SP
11/09/2009	13.8	1.6	14.6	216	403	9 (± 3)	1.2 (± 0.4)	26 (± 5)	LaP
12/09/2009	10.0	0.5	6.1	74	97	< 5	< 0.2	54 (± 4)	SP
25/10/2009	8.6	0.8	9.5	125	64	7 (± 2)	< 0.2	50 (± 3)	SP

^aRC: runoff coefficient; SSY: suspended sediment yield; KE: kinetic energy; n/a: not available. Flood type as defined by Duvert et al. (in press). See the text for details.

Mean atmospheric fluxes of Be-7, Pb-210, and Cs-137 in each subcatchment in 2009

Subcatchment	Area (km ²)	Rainfall (mm)	$F_{a(\mathrm{Be})} (\mathrm{GBq} \mathrm{d}^{-1})$	$F_{a(\mathrm{Pb})} (\mathrm{GBq} \mathrm{d}^{-1})$	M _(Cs) (GBq)
Huertitas	3.0	593	0.008	0.0005	3
La Cortina	9.0	1169	0.068	0.0038	11
Potrerillos	12.0	765	0.059	0.0033	14

Mean river fluxes of Be-7, Pb-210, and Cs-137 in each subcatchment

Subcatchment	$F_{r (Be)} (GBq d^{-1})$	$F_{r (Pb)} (GBq d^{-1})$	$F_{r (Cs)} (GBq \text{ km}^{-2})$	$(F_a/F_r)_{(Pb)}$	$(F_a/F_r)_{(\mathrm{Be})}$
Huertitas	$1.7 imes 10^{-4}$	3.4 ×10 ⁻⁵	1.8×10^{-6}	14	47
La Cortina	$5.1 imes 10^{-5}$	1.8×10 ⁻⁵	1.2×10^{-6}	203	1353
Potrerillos	4.2 ×10 ⁻⁴	5.8 ×10 ⁻⁵	2.8 ×10 ⁻⁶	56	139

Model estimations of the sediment residence times in the river (τ_r) and soil (τ_s) boxes of each subcatchment; proportion of the sub-catchment area covered by the river box (S_r)

Subcatchment	hment $ au_{r}(d)$		7		<i>S</i> _r (%)	<i>S</i> _r (%)	
Huertitas	60	(± 40)	5000	(± 1500)	4.5	(± 1.5)	
La Cortina	200	(± 70)	23,300	(± 7000)	0.3	(± 0.1)	
Potrerillos	50	(± 30)	13,300	(± 4000)	1.5	(± 0.5)	

Calculated inventories of Be-7, Pb-210, and Cs-137 in the soil boxes of the different subcatchments

Subcatchment	$I_{\rm s \ (Pb)} \ ({\rm GBq \ km}^{-2})$	$I_{\rm s (Cs)} ({\rm GBq \ km^{-2}})$	$I_{\rm s (Be)} ({\rm GBq \ km^{-2}})$	$(I_{\rm r}/I_{\rm s})_{\rm (Be)}$
Huertitas	1.7	1.1	0.3	0.132
La Cortina	4.7	1.1	0.6	0.003
Potrerillos	3.1	1.1	0.4	0.008

Table 7

Radionuclide concentrations in representative samples of the different land uses observed in the different subcatchments

	Cs-137
	$(\mathrm{Bq}\mathrm{kg}^{-1})$
Huertitas:	
Gullies	< 0.2
Retreat scarp in grassland	0.8 (± 0.2)
Cropland	2.1 (± 0.3)
La Cortina:	
Woodland	11.9 (± 5.8)
Cropland (avocado)	3.1 (± 0.5)
Cropland (maize)	4.1 (± 0.3)

Potrerillos:

Grassland	0.6 (± 0.3)
Cropland (maize)	2.2 (± 0.4)