Evidence of Chlordecone Resurrection by Glyphosate in French West Indies

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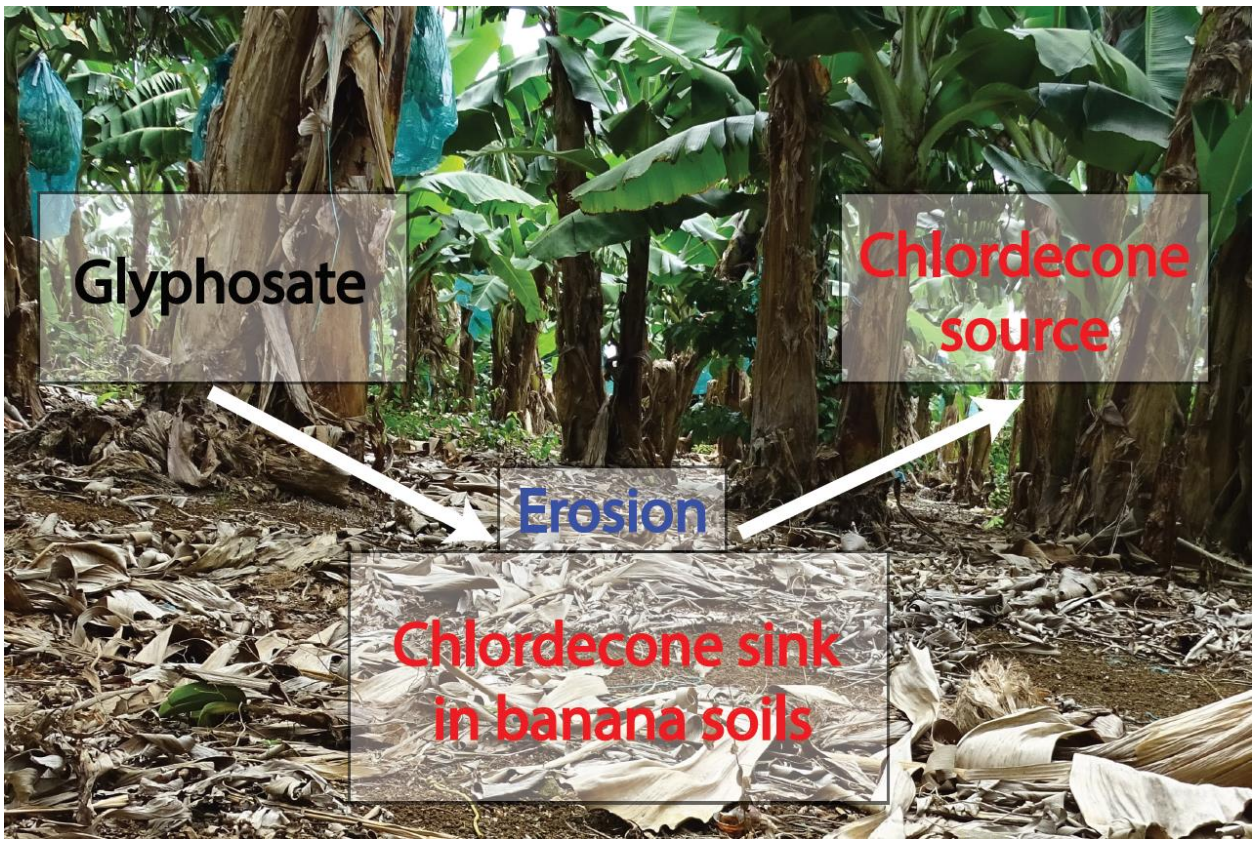
KEYWORDS : Critical zone, Glyphosate, Chlordecone, Soil erosion, Banana crops, sediment cores

ABSTRACT: The widespread use of pesticides in agriculture during the last several decades has contaminated soils and different Critical Zone (CZ) compartments, defined as the area extended from the top of the vegetation canopy to the groundwater table, and it integrates interactions of the atmosphere, lithosphere, biosphere, and hydrosphere. However, the long-term fate, storage, and transfer dynamics of persistent pesticides in CZ under a changing world remain poorly understood. In the French West Indies (FWI), chlordecone (CLD), a toxic organochlorine insecticide, was extensively applied to banana fields to control the banana weevil from 1972 to 1993, when it was banned. Here, to understand CZ trajectories we apply a retrospective observation based on marine

sediment core analysis to monitor long-term CLD transfer, fate and consequences in Guadeloupe and Martinique islands. Both CLD profiles show synchronous chronologies. We hypothesized that the use of glyphosate, a postemergence herbicide, from the late 1990s onwards induced CZ modification with an increase in soil erosion and led to the release of the stable CLD stored in the soils of polluted fields. CLD fluxes drastically increased when glyphosate use began, leading to widespread ecosystem contamination. As glyphosate is used globally, ecotoxicological risk management strategies should consider how its application affects persistent pesticide storage in soils, transfer dynamics and widespread contamination.

ABSTRACT IMAGE:



INTRODUCTION

The Critical Zone (CZ) may be defined as the reactive skin of our planet within which most of its coupled physical, chemical, biological and geological processes operating together to support life[1]. The emergence of human societies as a geological factor modified those subtle equilibria under natural forcing (climate, tectonic)[2], resulting in unprecedented biodiversity loss, biogeochemical disruptions and modifications of the erosion cycle[3] leading to potential threats to the future of humanity[4]. In recent decades, the sediment fluxes in CZ associated with the erosion of cultivated soils have greatly increased in response to changes in agricultural practices[5] such as deforestation, overgrazing, tillage and unsuitable agricultural practices with the use of herbicides[6]. Over the last several decades (last 70 years) is the use of many chemical substances to control disease (fungicides), insect damage (Insecticides), and weed competition (herbicides) in cropland dramatically rose[7]. Organochlorine insecticides such as dichlorodiphenyltrichloroethane (DDT, $C_{14}H_9Cl_5$) or chlordecone (CLD, $C_{10}Cl_{10}O$) are classified as persistent organic pollutants (POPs) by the Stockholm convention (http://www.pops.int/) but have been extensively used worldwide. Their use has been progressively prohibited since the 1970s because of their biomagnification, high toxicity, and long-term persistence in the environment. A well-known example is the contamination from the Hopewell CLD production plant in the United States between 1965 and 1975 that resulted in high worker exposure and massive pollution of over 160 km of the James River[8], which lasted for several decades[9]. Despite CLD was extensively used wordwilde, very few studies document its adverse effects on environment excepted in the Caribbean area. In the French West Indies (FWI, Fig. 1a) CLD was used to control the banana weevil during two periods: 1/ 1972–1978 under the trade name Kepone©, manufactured at Hopewell and 2/ 1982–1993 under the trade name Curlone©. CLD was banned worldwide in 1992 except in the FWI where it has

been authorized until 1993 by the French government., However, CLD is still persistent in most of CZ compartment such as soils[10], crops[11], freshwater[12], coastal[13] ecosystems until pelagic cetaceans[14], with potential for severe toxic effects to human populations[15] with one of the highest prostate cancer incidence in the world due to CLD exposure[16] and the related highest mortality rates[17]. These studies suggest that a significant amount of CLD is being transferred to the ocean, raising the question of contamination provenance and its long-term environmental fate for ecotoxicological management strategies.

In the FWI, interactions between agricultural practices such as CLD application (~3 kg/ha/yr), tillage, plant cover, irrigation and pedoclimatic conditions lead to high variability in the contamination level of soils[18,19] (Fig. 1b, c). CLD persistence in soils is explained by its 1) hydrophobicity, resulting in a high affinity for soil organic matter (high Koc 2.5 to 20 $m^3.kg^{-1}$)[10]; 2) physical sequestration in the fractal structure of soils containing allophanic clay, such as Andosols[20], which are typical of the volcanic FWI; and 3) poor biodegradability under aerobic conditions due to its chemical structure[21]. Depending on the soil type and considering negligible CLD degradation, it was estimated that CLD could remain in soil for at least several decades and up to centuries[10]. Thus, surface soil horizons act as reservoirs of CLD, gradually releasing this chemical into groundwater[19,22,23] or bound to soil particles eroded by surface runoff[24]. Thus, if erosion of cultivated soils increases in response to changes in agricultural practices, soils will be converting from sinks to sources of pesticides. It has been demonstrated, thought monitoring plots, simulation experiments and retrospective observation that the massive use of postemergence herbicides such as glyphosate since the late 1990s has a strong effect on soil erosion, as it acts on grass development and leads to permanently bare soil, as has been shown in vineyards[6,25] clementine[26] and rubber plantations[27]. The present study was designed in order to test the

79 hypothesis of enhanced CLD remobilisation due to glyphosate application through the erosion of
80 contaminated soils, as previously observed in vineyards with DDT remobilization[6].

81 To test this hypothesis, in the context of intensive glyphosate application[28], we applied a
82 retrospective observation[6,29,30] based on the analysis of marine sediment cores in order to monitor
83 long-term CLD transfer from watersheds. Indeed, CZ is submitted to processes that occur at
84 various time-scales which implies its trajectories must be documented over longer time-periods
85 much longer than direct observation experiments [31]. A retrospective observation hence allows to
86 extend into the past monitoring observations thanks to dated sediment cores and associated
87 proxies. In the present study, downcore contaminant profiles were associated with high-resolution
88 sedimentological and geochemical proxies. Short-lived radionuclides provide a precise core
89 chronology and the evolution of erosion patterns[6]. The two investigated watersheds were the Pérou
90 River[23] in Guadeloupe and the Galion River[19] in Martinique (Fig. 1d, e). These watersheds present
91 high to very high CLD soil risk contamination (Fig. 1b, c). Both watersheds are mainly occupied
92 by banana and sugar cane plantations (Fig. 1d, e). Large cropped areas in the Galion watershed
93 belong to large farms, while Pérou farmers are mostly smallholders.

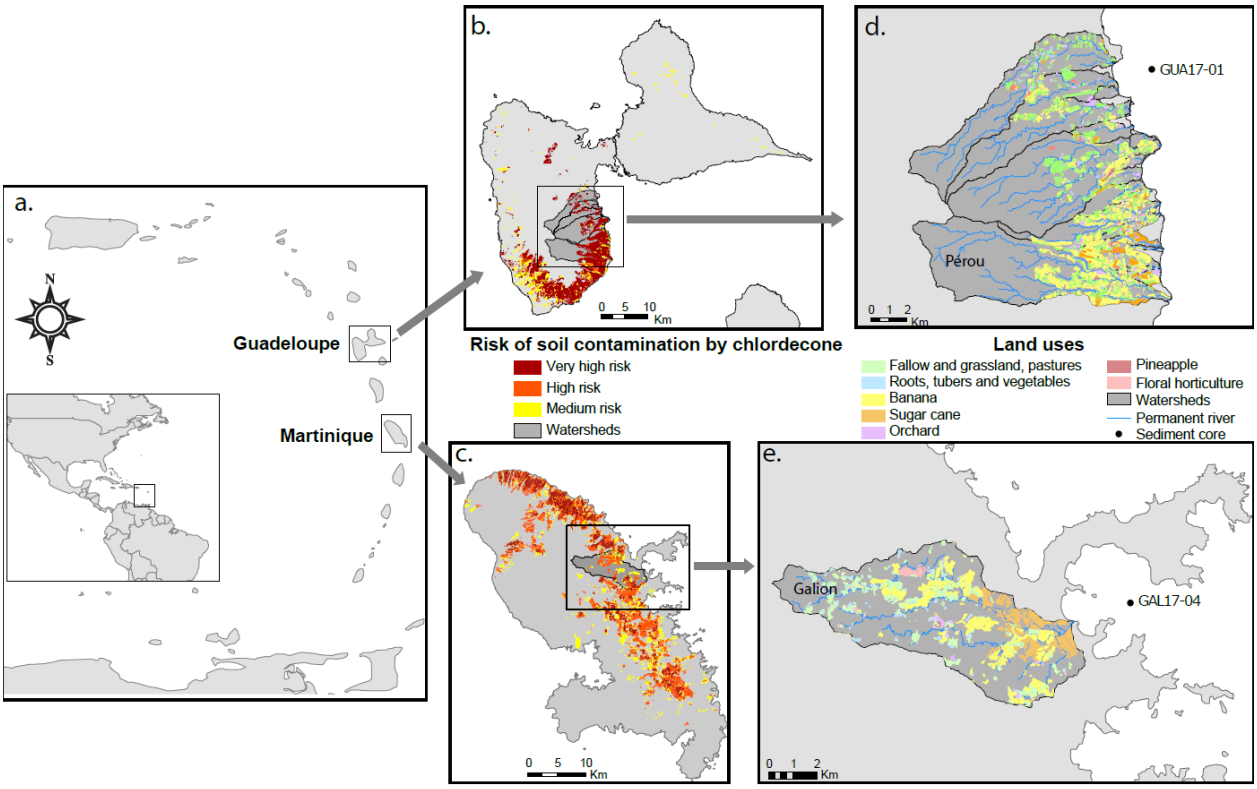


Figure 1: ***a****, Map of the Caribbean island arc with the localization of two French islands (Guadeloupe and Martinique).* ***b****,* ***c****, Maps of Guadeloupe and Martinique showing the risk of soil contamination by chlordecone (CLD; very high, high and medium risk); the dark gray area indicates the study area.* ***d****,* ***e****, Localization of the studied watershed (dark gray) according to land use. Different colors indicate the types of crops, and banana plantations are indicated in yellow. The two sampled marine sediment cores GUA17-01 and GAL17-04 are indicated by black dots. Note that the sediment core from Guadeloupe (****d****) was not collected off the estuary of the monitored Pérou River due to the absence of fine sediment in the coastal zone at this location. Pérou (Guadeloupe) and Galion (Martinique) river watersheds (d, e) were also sampled for detailed information see Figure S1. Land uses data of 2017 mad available by the French Directions of Agriculture and Forestry of Martinique and Guadeloupe (DAAF Martinique and Guadeloupe).*

Risk of soil contamination by chlordecone made available by Préfecture of Martinique and Préfecture of Guadeloupe, for their design see [19,32–34].

MATERIALS AND METHODS

Sampling. A 1.13-m-long core registered as GAL17-04 (N° IGSN TOAE0000000573) was collected in the Baie du Galion in Martinique (WGS84: 14.72801600; -60.91658800) at 15 m below sea level on April 2017. A 1.33-m-long core registered as GUA17-01 (N° IGSN TOAE0000000567) was collected in Petit Cul-de-Sac Marin in Guadeloupe (16.16857500; -61.56900800) at 13.3 m below sea level on April 2017. Both cores were sampled using an Uwitec gravity corer with a hammer from a small boat. Surface soil horizons were sampled on the Galion (Martinique, n=44) and Pérou (Guadeloupe, n=35) watersheds under different soil types (Nitisol, Andosol, Ferralsols) and land use contexts (different crop types, forest, river sediment, channel bank), as illustrated in figure S1. At the Grand Galion gauging station, 2 floods were sampled on 19 December 2013 and on 23 December 2013. During both floods, 24 water samples (330 mL each) were taken every 6 min with an automatic sampler (Sigma SD900) when the limnimetric level of the river exceeded 80 cm (i.e., 1.85 $m^3.s^{-1}$). Using this protocol, each flood was sampled for 2 h 24 min, with a 6 min resolution. Water discharges during the two floods were also measured with a pressure sensor. The discharges of both floods were provided by DEAL (The French Environment, Planning and Housing Department). By the end of each flood, 24 flasks had been collected. For both floods, due to material availability and logistical constraints, we selected 18 flasks for particle filtration. The selection retained the maximum variations in water color among the flasks and thereby the maximum variability in the particle content among the samples. Using

a vacuum pump and 0.7 µm fiberglass filters (Whatman cat no 1825-047), we separated the dissolved fraction (< 0.7 µm) from the particulate fraction (>= 0.7 µm) for each of the 18 flasks collected during each flood. The particulate and dissolved fractions were analyzed for CLD at the Laboratoire Départemental d'Analyses de la Drôme (LDA26), which is COFRAC (French Accreditation Committee) accredited. The results are presented with a +/- 30% error interval, and the limit of quantification was 0.01 µg. L^{-1} for the dissolved fraction and 10 $ng.g^{-1}$ for the particulate fraction. According to the analytical requirements of the laboratory for CLD analysis, samples were pooled (Table S1) to obtain the minimum (0.5 g) amount of solid, but the samples were kept separate according to the flood rise, flood peak and flood fall. The mass of sediment in each composite sample varied between 0.510 g and 0.891 g, while the volume of water filtered from the composite samples varied between 200 mL and 950 mL.

Logging. In the laboratory, the cores were split lengthwise, photographed, and logged in detail, noting all physical sedimentary structures. The grain size distributions of both cores were determined using a Malvern Mastersizer 2000 (Isterre) with a continuous interval of 2 cm and ultrasound during measurements. Cores were also cut at 2-cm depth intervals, and a specific volume was dried at 60°C for 4 days to determine the dry bulk density (DBD); then, the Loss Of Ignition (LOI) of each interval was measured using the protocol of Heiri[35]. The LOIs at 550°C and 950°C correspond to the percent of organic and carbonate contents of the sediment, respectively. The noncarbonate igneous residue (NCIR, express in %) of each sample was obtained by removing the LOI550 and LOI950 from the initial dry weight. The terrigenous mass accumulation was calculated as NCIR x DBD × sedimentation rate, expressed in $g.cm^{-2}.yr^{-1}$. X-Ray Fluorescence (XRF) analysis was performed on the surfaces of the split sediment cores, which had been covered with 4-µm-thick Ultralene, at 2-mm intervals using an Avaatech core scanner (EDYTEM). The

geochemical data were obtained with various tube settings: 10 kV at 0.15 mA for 15 s for Al, Si, S, K, Ca, Ti and Fe and 30 kV at 0.2 mA for 20 s for Cu, Zn, Br and Sr[36]. 3 replicates were measured every 10cm to estimate the standard deviation. Each individual power spectrum was deconvoluted into relative components (intensities), expressed in counts per second. The principal component analysis (PCA) was performed using R software.

Radionuclide measurements. Surface soil horizons were sampled in the Galion (n=44) and Pérou (n=35) watersheds at locations with different soil types and land use contexts, as illustrated in figure S1, to assess whether erosion is an important mechanism on plantations through short-lived radionuclide analyses. The two marine sediment cores collected in Petit Cul-de-Sac Marin (GUA17-01, Guadeloupe) and Galion Bay (GAL17-04, Martinique) were analyses for short-lived radionuclides to establish sediment chronology (Table S2 and S3). Prior to analyses, samples from the watershed soils and core sediment were dried at 40°C for ~48 h, the watershed samples were sieved to 2 mm, and all samples were ground to a fine powder in an agate mortar and pressed into 15 or 60 mL polyethylene containers depending on the quantity of material available for analysis. Radionuclide activities were determined with gamma spectrometry using coaxial N- and P-type HPGe detectors (Canberra/Ortec) at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). The ^{137}Cs activity (half-life: 30.17 y) was measured from the 661.7 keV emission peak. The $^{210}Pb_{xs}$ activity (half-life: 22.3 y) was calculated by subtracting the supported activity (determined by using two ^{226}Ra daughters), the ^{214}Pb activity (average count number at 295.2 and 351.9 keV) and the ^{214}Bi activity (609.3 keV)) from the total ^{210}Pb activity measured at 46.5 keV[37]. Counting efficiencies and calibration were determined using certified International Atomic Energy Agency (IAEA) standards (IAEA-444, 135, 375, RGU-1 and RGTh-1) prepared in the same containers as the samples.

Pesticide analysis. Pesticide analyses were performed on samples from cores GUA17-01 (n=30) and GAL17-04 (n=28) by an ALTHUS 30 ultraperformance liquid chromatography (UPLC) system (Perkin Elmer, USA) coupled in tandem to a triple quadrupole mass spectrometer equipped with an electrospray ionization (ESI) source (Perkin Elmer QSigth 200). For CLD (Chlordecone) and CLO (Chlordecol), 3 g of lyophilized dry sediment was extracted using an accelerated solvent extraction system (ASE200, Dionex). The organic extract was then evaporated, purified and passed through a 0.2 µm filter before analysis. The limit of detection (LOD, corresponding to a signal-to-noise ratio of 3) and limit of quantification (LOQ, corresponding to a signal-to-noise ratio of 10) were 0.22 $ng.mL^{-1}$ and 0.67 $ng.mL^{-1}$ for CLD and 0.1 $ng.mL^{-1}$ and 0.3 $ng.mL^{-1}$ for CLO, respectively. For glyphosate and AMPA (Aminomethylphosphonic acid), 1 g of lyophilized dry sediment was added to 10 mL of 0.5 M KOH and vortexed for 1 min. Glyphosate-2-13C15N and AMPA-13C15N standard solutions were added as internal standards to a final concentration of 100 $ng.ml^{-1}$ each. The extract was centrifuged, and the water extract was filtered through a 0.2 µm nylon filter and analyzed immediately after preparation. The LOD and LOQ were 2 $ng.mL^{-1}$ and 6 $ng.mL^{-1}$ for glyphosate and 1 $ng.mL^{-1}$ and 3 $ng.mL^{-1}$ for AMPA, respectively. Detailed protocols are presented in the supplementary material. Pesticides fluxes was calculated as DBD × sedimentation rate × [pesticide] and expressed in $mol.g.cm^{-2}.y^{-1}$.

RESULTS AND DISCUSSION

On land erosion. Samples under forest cover, found in upper catchment areas exposed to the highest precipitation levels, were systematically enriched in ^{137}Cs and $^{210}Pb_{xs}$ relative to channel bank samples (Fig. 2). The fallout radionuclide activities detected in cropland samples collected on banana and sugarcane plantations showed intermediate levels between those detected in forests and channel banks (Fig. 2). The results show that the sediment originates from a mix of cropland

(exposed to rainfall enriched in fallout radionuclides) and channel bank soils (sheltered from rainfall and fallout and depleted in radionuclides as demonstrated by Evrard et al. (2020)[38]. These samples show that the banana and sugarcane plantations provide a significant supply of sediment to river systems on these islands through erosional processes.

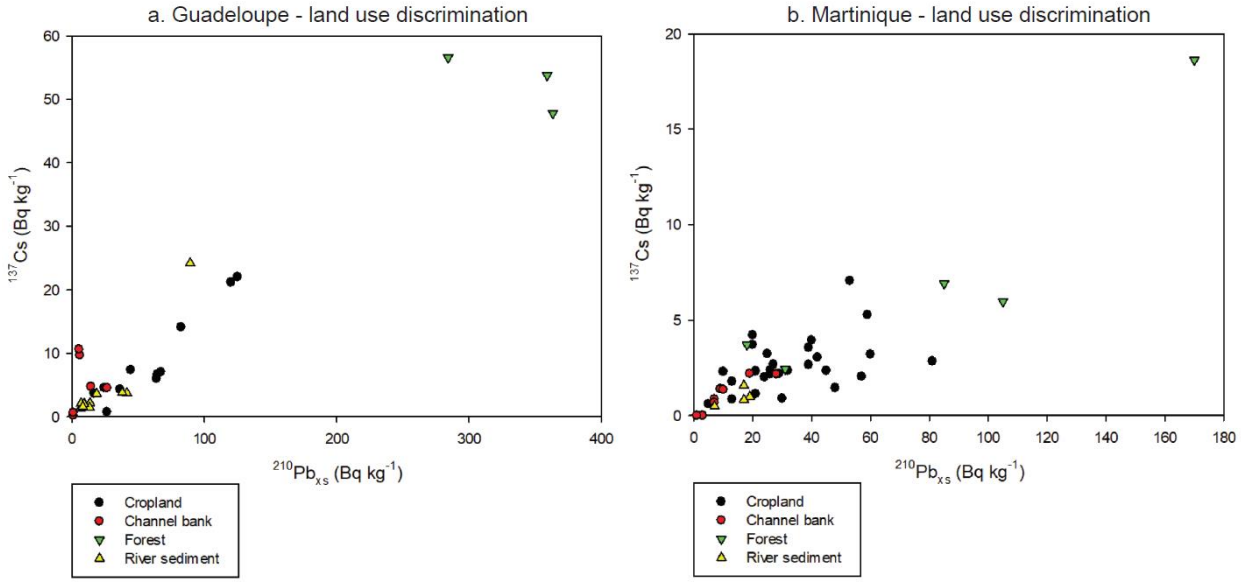


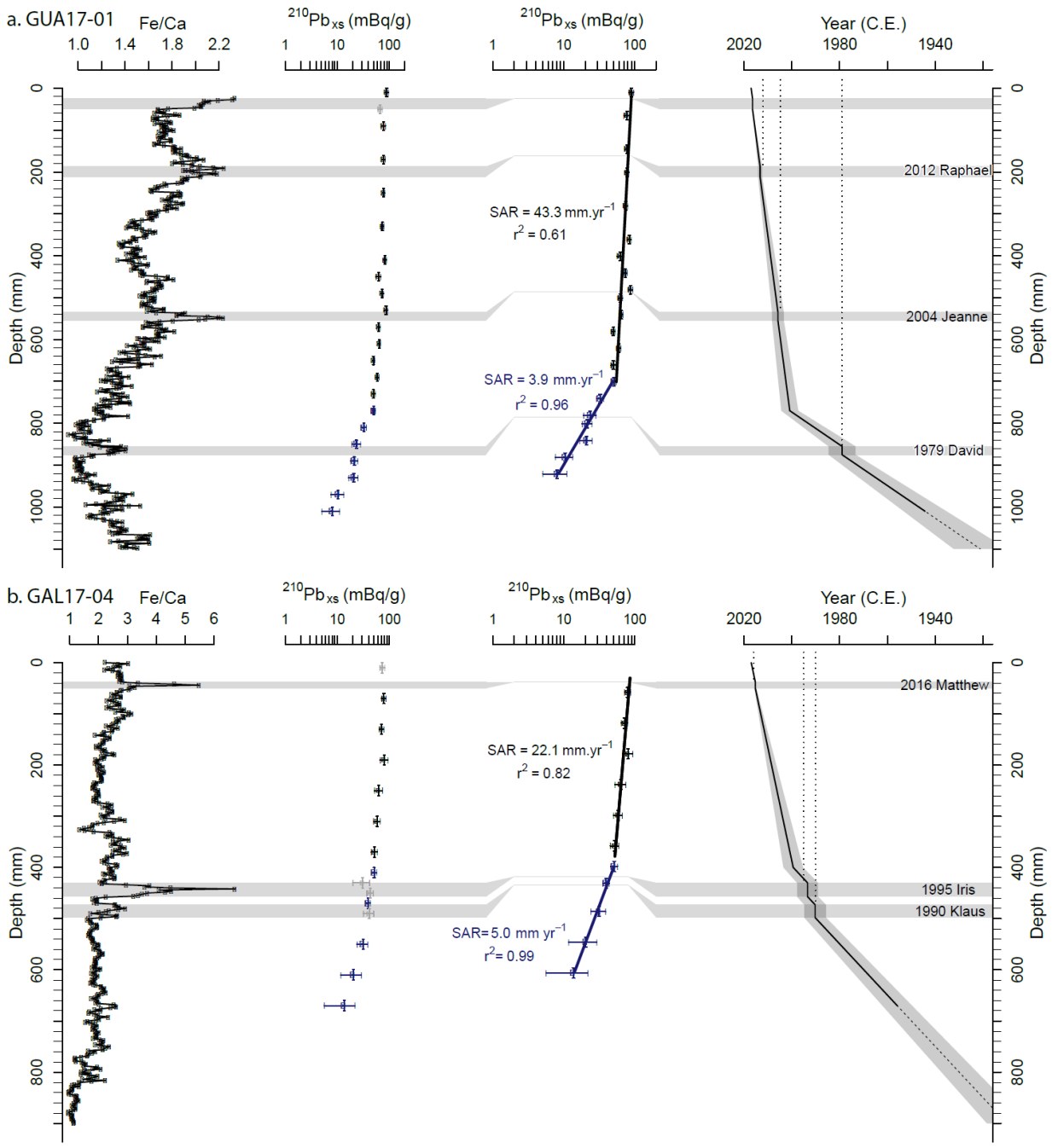
***Figure 2: a, b**, $^{210}Pb_{xs}$ versus ^{137}Cs activities in the Pérou (Guadeloupe) and Galion (Martinique) river watersheds. Data are presented for the different land use conditions: cropland, forest, river sediment and channel bank. Forest samples are enriched in ^{137}Cs and $^{210}Pb_{xs,}$ while channel banks are depleted in both radioisotopes.*

Sediment core and Chlordecone chronology. The two marine sediment cores collected in Petit Cul-de-Sac Marin (GUA17-01, Guadeloupe) and Galion Bay (GAL17-04, Martinique) (Fig. 1d, e) were characterized in terms of their particle size, LOI, XRF mineral geochemistry and pesticide contents. Both cores contained relatively homogeneous olive-brown silt sediment. No noticeable variations in either the organic content or grain size distribution were observed (Fig. S2). These parameter variations hence could not have affected the absorption/degradation of pesticides within

212 the accumulated sediment[6]. PCA on XRF data (Fig. S3) showed two sediment end-member[39]
213 inputs from the watershed, with organic matter and metallic pollutant contents and the marine
214 carbonate productivity. The geochemical data were in good agreement with the LOI indicating an
215 upward increase in terrigenous inputs from the watershed (reflected by the Fe content) while
216 carbonate productivity (reflected by the Ca content) decreased (Fig. S3). Accordingly, the Fe/Ca
217 ratio was used as a high-resolution proxy for the terrigenous fraction.

218 A chronological framework was established with the *serac* R package[40] from measurements of
219 short-lived radionuclides, constrained by the identification of historical hurricane event deposits
220 (Fig. 3, Table S2 and S3) as ^{137}Cs did not provide an interpretable profile in relation to its
221 desorption/migration in marine sediments[41]. The Fe/Ca data in the GUA17-01 and GAL17-04
222 cores led to the identification of 4 and 3 historical hurricane events, respectively, that caused heavy
223 precipitation in the region, as indicated by meteorological data (http://pluiesextremes.meteo.fr/),
224 which allow independent chronology validation. The sediment deposits triggered by these events
225 were considered as instantaneous and thus excluded from the construction of the age model, by
226 removing the depth interval and associated $^{210}Pb_{xs}$ data of each of these deposits [40,42]. The
227 logarithmic plot of the event corrected $^{210}Pb_{xs}$ activity shows a general decrease, with two distinct
228 linear trends (black and dark blue Fig. 3). According to the "constant flux, constant sedimentation
229 rate" (CFCS) model[40] applied to each segment of the profile, the levels of $^{210}Pb_{xs}$ indicate drastic
230 increases in the sediment accumulation rate (SAR) from 3.9 ± 0.6 $mm.y^{-1}$ to 43.3 ± 9.5 $mm.y^{-1}$
231 and from 5.0 ± 0.2 $mm.y^{-1}$ to 22.1 ± 5.1 $mm.y^{-1}$ with synchronous changes in 2000 (uncertainty
232 range: 1996-2005) and 1999 (uncertainty: 1995-2004) in the GUA17-01 and GAL17-04 cores,
233 respectively (Fig. 3). These two age models are well constrained by the ages of the most intense
234 historical hurricanes that have hit both islands. This concomitant increase in sedimentation rates

in sediment records located more than 170 km away from each other might only be explained by an increase in carbonate productivity in the marine system or by an increase in terrigenous inputs from the watersheds. From these age models, the terrigenous mass accumulation was calculated and interpreted as a proxy of soil erosion in the watershed, indicating that the erosion rate increased 10- and 4-fold on the basis of the GUA17601 and GAL17-04 cores, respectively (Fig. 4a, b). This interpretation rules out the hypothesis of an increase in carbonate precipitation. It is further supported by the observation of a synchronous increase in the Fe/Ca ratio for GUA core (Fig. 4a). For the GAL core we observed a short time lag (corresponding to less than 2cm) between Fe/Ca and terrigenous flux increase which could be related to sampling resolution (2cm for short-lied radionuclides not in continuous versus continuous 2mm for XRF data) or to the influence of Iris hurricane (Fig. 4b).



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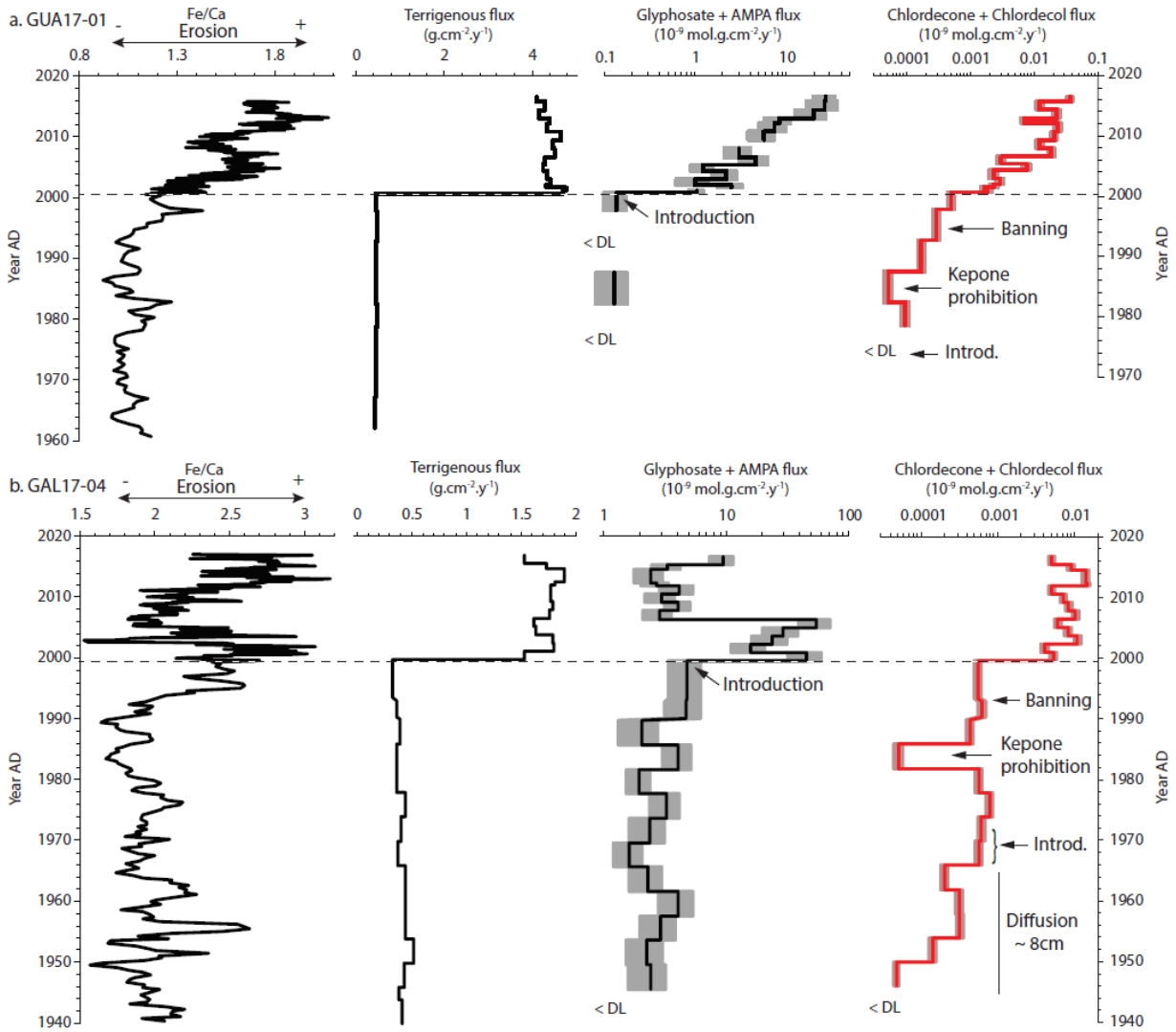
247 ***Figure 3:*** *Chronology for the GUA17-01 (**a**) and GAL17-04 (**b**) cores with the Fe/Ca, ^{210}Pbxs*
248 *activity, instantaneous event-corrected ^{210}Pbxs activity and age model with uncertainties (gray*
249 *area) realized with the serac R package[40]. The dotted lines extending from the black lines indicate*
250 *age model extrapolation. On the right part of this figure ages and names correspond to historical*
251 *hurricanes.*

Pesticide temporal variations are presented in relation to age and displayed both in concentrations (Fig. S4) and fluxes (Fig. 4). CLD and chlordecol (CLO, a CLD degradation product) concentrations present similar profiles in both cores (Fig. S4) and were thus summed and expressed as fluxes (Fig. 4) to account for the large increases in sedimentation rates. In GUA17-01, the CLD+CLO flux is first detected in 1978 +/- 7 yr, just after the introduction of CLD in 1972 and subsequently decreases, which could correspond to the period (1978-1982) between the prohibition of Kepone© in the US and the approval of the license for the use of CLD in Curlone© by the French authorities in late 1981. Then, the CLD+CLO flux increases, which continues even after its ban in 1993. In 2000 +/-5 yr, we observed a 2-fold and more than 5-fold increase in the CLD+CLO concentration and flux, respectively. In GAL17-04, CLD and CLO are first observed in very low concentrations at 8 cm (1940-1955), before its introduction dated here at 1970 +/- 6 yr (Fig. S4). This apparent preintroduction record of this compound may be explained by its possible downward diffusion in sediment, as observed for other chlorinated molecules such as DDT[6], or by bioturbation in these shallow marine environments. Then, we observe a decrease in CLD+CLO flux during the prohibition period, followed by an increase until the final ban. In 1999 +/-5 yr, we detected a 2-fold and more than 10-fold increase in the concentration and flux, respectively.

Finally, between the period of maximum CLD use and the recent high flux, we noted more than 200-fold and 20-fold CLD+CLO flux increases in GUA17-01 and GAL17-04, respectively (Fig. 4). The CLO/CLD ratio is higher in the oldest part of the two sediment cores than in the most recent sediment (Fig. S4) and in present-day soils in Martinique[43]. CLO formation probably results from the reduction of the CLD ketone group under anaerobic conditions[44], which are encountered

some centimeters below the water-sediment interface. This hypothesis is further supported by the increase in CLO/CLD with depth suggesting to long-term CLD degradation within the sediment.

Chlordecone transfer in critical zone. CLD concentrations in soils surface layers of banana plantations range from 500 to more than 2000 $ng.g^{-1}$ in the Galion watershed[19] and from 30 to more than 24000 $ng.g^{-1}$ in the Pérou watershed[23]. This spatial variability in CLD soil pollution is related both to the duration of the banana cropping period and soil characteristics[10]. Soils in which banana was never planted did not contain any CLD[19]. Samples collected in 2002 at the mouth of the Galion River contained 50 and < 10 $ng.g^{-1}$ CLD in suspended matter (>0.7 µm) and in sediment, respectively[45], reflecting CLD dilution relative to soil concentrations. In comparison, the higher CLD concentrations measured in the recent layers of the two studied sediment cores (1 to 2 $ng.g^{-1}$) were still lower than the concentrations in river samples. These differences could be explained both by terrestrial particle dilution by other sediment sources such uncontaminated fields, channel banks (Fig. 2) and marine sediments; and by particle size fractionation resulting in coarser sediment fractions in cores (Fig. S2) and finer fractions in suspended matter (higher specific surface area) collected in rivers and nearby river mouths[46].



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290 ***Figure 4:*** *Soil erosion proxies and pesticide flux chronology for Guadeloupe* ***a*** *and Martinique* ***b****:*
291 *Erosion proxies with Fe/Ca and terrigenous fluxes compared to a pesticide chronology*
292 *(logarithmic scale) reconstructed from the sum of glyphosate and aminomethylphosphonic acid*
293 *(AMPA; glyphosate degradation product) fluxes and the sum of chlordecone and chlordecol (one*
294 *of the chlordecone degradation products) fluxes. The horizontal dotted line corresponds to the*
295 *large increase in sedimentation rate identified in response to a large erosion increase synchronous*
296 *with glyphosate introduction and chlordecone flux increases. <DL indicates below the detection*
297 *limit.*

The collection of water samples during two low-intensity floods in December 2013 in the Galion watershed permitted to assess the accumulated mass of transferred CLD in the dissolved and particulate fractions (Fig. S5a, b). The CLD content in the particle-bound fraction ranged from 311 to 1059 $ng.g^{-1}$ (Fig. S5c, d). During both floods, the mass of CLD transferred by particles was higher than that transferred in dissolved form (Fig. S5), indicating that soil erosion was an important pathway for CLD transfer. In the case of very large floods, such as during storms or cyclonic events, this process could transport huge amounts of CLD from land to sea and the sediment. These results can be related to the erosion occurring in this watershed, identified by short-lived radionuclide measurements in watershed samples (Fig. 2). We hence infer that erosion of contaminated soil particles is a major CLD mass transfer process[24] which should therefore not be neglected or considered minor, contrary to recent suggestions[22].

Critical zone erosion induced by Glyphosate. The large CLD+CLO flux increases observed around 1999/2000 in both cores are synchronous with a drastic rise in erosion fluxes (Fig. 4) and thus probably have a common watershed origin. As the erosion increases synchronously in both sites located on two different islands we can discard local phenomena such as road construction or urban development. Thus, three main hypotheses could explain this observation: 1/ climate driver with an increase in precipitation, 2/ change in mechanical practice on cropland with extensive tillage or 3/ the extensive use of glyphosate leading to unprotected soil more sensitive to precipitation-induced erosion. First we can discard the climate forcing as no significant precipitation change can be identified from instrumental data during this period in this Caribbean area[47]. As these two watersheds have similar land use (Fig. 1d, e) and erosion characteristics of banana and sugar cane fields (Fig. 2), we can hypothesize that a concomitant change in agricultural practices caused this erosional increase. Since the early 1970s on banana plantations, the fields

were prepared for planting with heavy equipment to enable the planting of banana trees and the drainage of water via ditches dug throughout the fields[48]. These practices are known to contribute to soil erosion but cannot explain its large increase more than 25 yrs later.

When these fluxes increase in the two cores, glyphosate and AMPA (glyphosate degradation product) are detected in the GUA17-01 sediment for the first time, and both the concentrations and flux considerably increase in the GAL17-04 sediment (Fig. S4). Glyphosate use began in 1974, but few banana farmers (only large banana farms) probably used it from the early 1980s until 1997. In 1997, the glyphosate price (Roundup) dropped, causing 90% of farmers to use it from that year onwards. Glyphosate is still widely used in the FWI[28]. This is in good agreement with the emergence of this chemical in 1999/2000 +/- 5 yr in the two sediment chronologies (Fig. 4). In the GAL17-04 core, the glyphosate and AMPA records start earlier (1950). Even if its use in some large farms in the Galion watershed is not totally excluded, as the absence of such large farms in the Pérou watershed points the diffusion of AMPA as the most probable explanations of downcore migrations of these chemicals[6] are diffusion and/or bioturbation processes. Since its earliest appearance in GUA17-01, the glyphosate+AMPA flux continuously increased, while in GAL17-04, after a drastic increase, this flux decreased in 2006.5 +/- 2.5 yr before re-increasing in the uppermost layer (Fig. 4). This time corresponds to the entry into force of the French law on water (2006-1772), which establishes an untreated area as a buffer around a watercourse, including ditches. In Martinique, important surface drainage networks are present in banana fields. Stopping glyphosate treatment would decrease the export of this chemical through the concentrated flows in these ditches. However, glyphosate is still used in these fields, thereby probably maintaining the transfer of soil particles contaminated by CLD. The difference between our study sites could

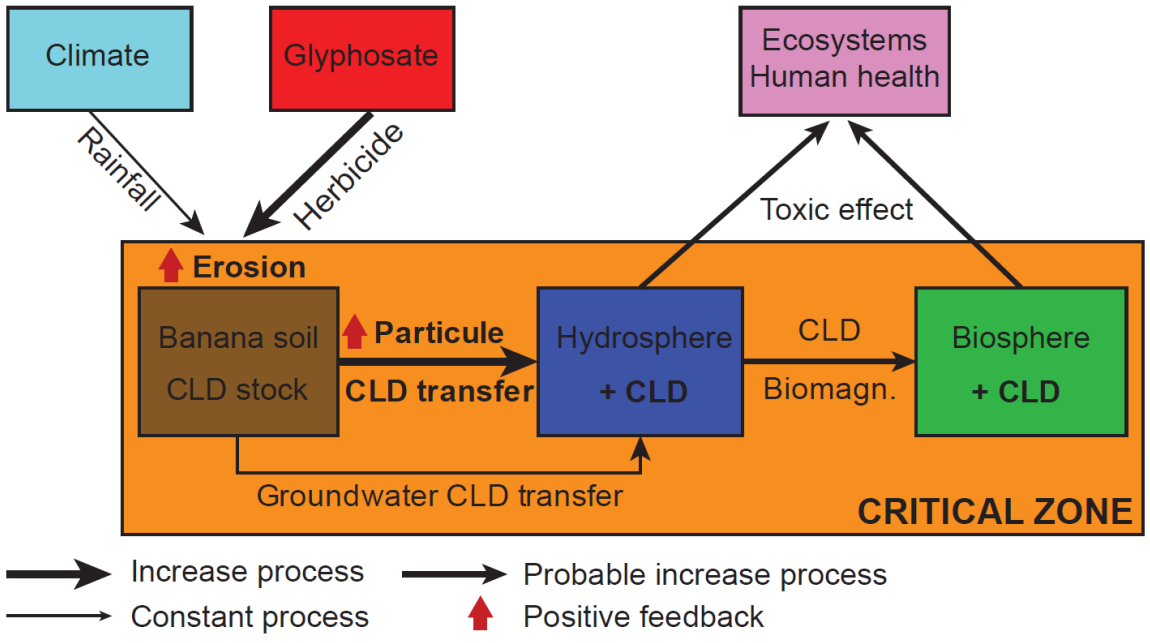
343 be linked to the fact that the large farms in the Galion watershed, which are monitored for good
344 practices, quickly adhered to the French law on water.

345 Even if the two watersheds are different in term of farm size, other important watershed
346 characteristics are the same: soils cover, type of cultures, climate. At both sites, we observed a
347 synchronous increase in CLD+CLO and erosional fluxes when glyphosate was first widely applied
348 to banana fields at the end of the 1990s. The application of glyphosate, which disrupts grass
349 development, has a strong effect on soil erosion as previous demonstrated thought monitoring
350 plots, simulation experiments and retrospective observation[6,25–27,49]. Other herbicides, such as
351 paraquat, and authorized in the French West Indies between 2003 and 2007, could have cuased the
352 same issues. Paraquat was reported to be used along with glyphosate because of glyphosate
353 resistant plants[50] and intensively used in banana plantations in Latin America[51]. While glyphosate
354 is a systemic herbicide, paraquat and other herbicides used at the early 2000s on banana plantations
355 [28] are either selective or non-systemic. As a result, they do not eliminate all plants on a field,
356 allowing them to recover. Paraquat, for instance is a non-selective but non-systemic herbicides. It
357 "burns" the leaves of plants while allowing plants to recover from stems or roots. Glyphosate acts
358 differently by killing the whole plants which leaves the soil bare and less-maintained by roots
359 leading to potential enhanced erosion rates. Erosion on bare soil might furthermore be amplified
360 by banana canopies, which exhibit high funneling ratios, favouring runoff even on soils with a
361 high infiltration rate such as Andosols, by localized rainfall redistribution and soil detachment by
362 concentrated flows[52].

363 Putting all our observations together, we argue that the widespread use of a non-specific systemic
364 herbicide (glyphosate) since the late 1990s' could be responsible of an unprecedented rise in soil
365 erosion and downstream of a major release of remnant CLD pesticides that were trapped in banana

366 fields soils since their banning in the Late-1990s' (Fig. 4). As such, we highlight that new
367 agricultural practices may induce complex interactions in CZ dynamic converting soils from sinks
368 to sources of formerly-used pesticides. This happened in FWI where CLD was probably
369 resurrected by glyphosate-induced soil erosion (Fig. 5) just as in a French vineyard where DDT
370 was resurrected by the same herbicide[6]. The eroded CLD-contaminated material was transported
371 to the marine environment bound onto fine particles, where it became a source of contamination
372 for marine organisms[13,14,46] and a potential hazard to human health through seafood consumption
373 (Fig. 5). This mechanism of old pesticides resurrection by glyphosate is now observed in banana
374 plantation and vineyard, perhaps in no tillage agro systems glyphosate-induced soil erosion could
375 be lower. We recommend future works to answer to this hypothesis. The retrospective observation
376 applied here, allowed to reconstruct long-term CZ trajectories under human forcing, hence proving
377 its strength in providing novel and complementary information to modern-day CZ observatories[31].

378



379

Figure 5*: Critical Zone dynamics and responses in FWI relation to past (CLD) and modern (Glyphosate) agricultural practices.*

Future studies of the environmental fate of pesticides in CZ should take into account these potential pesticide–environment interactions from a long-term perspective. In terms of management options, reducing soil erosion on cropland by limiting herbicide treatments would lead to the growth of understory vegetation and ultimately result in the slower leaching of the pesticides stored in soils. As glyphosate is used worldwide, it appears crucial that ecotoxicological risk assessments take into account such mechanisms of remnant pesticide mobility in the environment through herbicide-induced erosion.

ASSOCIATED CONTENT

Supporting Information.

Figure S1: Land use and sample in the investigated watersheds
Figure S2: Sedimentological and geochemical data in both cores
Figure S3: Biplot of the PCA of XRF geochemical data in both cores
Figure S4: Pesticide chronology expressed in concentration in both cores
Figure S5: Gauging station data during two flood events in the Galion watershed
Detailed pesticide analysis protocols

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Author Contributions

P.S., C.M., I.C., J.P. and O.E. conceived and designed the study. Fieldwork sampling were performed by C.M., I.C., J.P., O.E., L.D. for soil sampling, P.S. and F.A. for coring and J.P., M.L.J. and C.M. for water collecting. P.S., N.C., C.P., B.G., O.E., I.L. and A.L.D. performed the analyses. P.S. wrote the first draft of the manuscript, with subsequent contribution by all the authors.

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ABBREVIATIONS

CLD: Chlordecone

CLO: Chlordecol

AMPA: Aminomethylphosphonic acid

DDT: Dichlorodiphenyltrichloroethane

FWI : French west indies

CZ: Critical zone

LOI : Loss of ignition

XRF : X-ray fluorescence

DBD : Dry bulk density

NCIR : Noncarbonate igneous residue

CFCS: Constant flux, constant sedimentation rate

PCA : Principal component analysis

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