Evidence of chlordecone resurrection by glyphosate in

French West Indies

Pierre Sabatier^{†,}, Charles Mottes^{‡,§}, Nathalie Cottin^{||}, Olivier Evrard[†], Irina Comte^{§,#},
Christine Piot^{||}, Bastien Gay^{†,||}, Fabien Arnaud[†], Irène Lefevre[†], Anne-Lise Develle[†], Landry
Deffontaines^{‡,§}, Joanne Plet^{‡,§}, Magalie Lesueur-Jannoyer^{‡,§}, Jérôme Poulenard[†]*

[†]: Univ. Grenoble Alpes, Univ. Savoie Mont Blanc, CNRS, EDYTEM, LE Bourget du lac, France

[‡]: Cirad, UPR HortSys, F-97285 Le Lamentin, Martinique, France

[§]: HortSys, Geco, Univ Montpellier, CIRAD, Montpellier, France

^{||}: Univ. Savoie Mont-Blanc, LCME, Le Bourget du Lac, France

[†]: Univ. Paris-Saclay, UVSQ, CEA, CNRS, LSCE/IPSL, F-91191, Gif-sur-Yvette, France

[#]: Cirad, UPR GECO, F-97130 Capesterre-Belle-Eau, Guadeloupe, France

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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 life¹. 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 cycle³ leading to potential threats to the future of humanity⁴. In recent decades, the sediment fluxes in CZ associated with the erosion of cultivated soils have greatly increased in response to changes in agricultural practices⁵ such as deforestation, overgrazing, tillage and unsuitable agricultural practices with the use of herbicides⁶. 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 rose⁷. Organochlorine insecticides such as dichlorodiphenyltrichloroethane (DDT, C₁₄H₉Cl₅) or chlordane (CLD, C₁₀Cl₁₀O) 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 River⁸, which lasted for several decades⁹. Despite CLD was extensively used worldwide, 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 soils¹⁰, crops¹¹, freshwater¹², coastal¹³ ecosystems until pelagic cetaceans¹⁴, with potential for severe toxic effects to human populations¹⁵ with one of the highest prostate cancer incidence in the world due to CLD exposure¹⁶ and the related highest mortality rates¹⁷. 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 K_{oc} 2.5 to 20 m³.kg⁻¹)¹⁰; 2) physical sequestration in the fractal structure of soils containing allophanic clay, such as Andosols²⁰, which are typical of the volcanic FWI; and 3) poor biodegradability under aerobic conditions due to its chemical structure²¹. 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 centuries¹⁰. 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 runoff²⁴. 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, through 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} clementine²⁶ and rubber plantations²⁷. The present study was designed in order to test the

hypothesis of enhanced CLD remobilisation due to glyphosate application through the erosion of contaminated soils, as previously observed in vineyards with DDT remobilization⁶.

To test this hypothesis, in the context of intensive glyphosate application²⁸, we applied a retrospective observation^{6,29,30} based on the analysis of marine sediment cores in order to monitor long-term CLD transfer from watersheds. Indeed, CZ is submitted to processes that occur at various time-scales which implies its trajectories must be documented over longer time-periods much longer than direct observation experiments³¹. A retrospective observation hence allows to extend into the past monitoring observations thanks to dated sediment cores and associated proxies. In the present study, downcore contaminant profiles were associated with high-resolution sedimentological and geochemical proxies. Short-lived radionuclides provide a precise core chronology and the evolution of erosion patterns⁶. The two investigated watersheds were the Pérou River²³ in Guadeloupe and the Galion River¹⁹ in Martinique (Fig. 1d, e). These watersheds present high to very high CLD soil risk contamination (Fig. 1b, c). Both watersheds are mainly occupied by banana and sugar cane plantations (Fig. 1d, e). Large cropped areas in the Galion watershed 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 Péro River due to the absence of fine sediment in the coastal zone at this location. Péro (Guadeloupe) and Galion (Martinique) river watersheds (*d*, *e*) were also sampled for detailed information see Figure S1. Land uses data of 2017 made 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., $1.85 \text{ m}^3 \cdot \text{s}^{-1}$). 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 ($< 0.7 \mu\text{m}$) from the particulate fraction ($\geq 0.7 \mu\text{m}$) 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 $\pm 30\%$ error interval, and the limit of quantification was $0.01 \mu\text{g. L}^{-1}$ 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 Heiri³⁵. 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 $\text{NCIR} \times \text{DBD} \times \text{sedimentation rate}$, expressed in $\text{g.cm}^{-2}.\text{yr}^{-1}$. 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 Sr³⁶. 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 analysed 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 ¹³⁷Cs activity (half-life: 30.17 y) was measured from the 661.7 keV emission peak. The ²¹⁰Pb_{xs} activity (half-life: 22.3 y) was calculated by subtracting the supported activity (determined by using two ²²⁶Ra daughters), the ²¹⁴Pb activity (average count number at 295.2 and 351.9 keV) and the ²¹⁴Bi activity (609.3 keV) from the total ²¹⁰Pb activity measured at 46.5 keV³⁷. 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- $^{13}\text{C}^{15}\text{N}$ and AMPA- $^{13}\text{C}^{15}\text{N}$ 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 $\text{DBD} \times \text{sedimentation rate} \times [\text{pesticide}]$ and expressed in $\text{mol.g.cm}^{-2}.\text{y}^{-1}$.

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 $^{210}\text{Pb}_{\text{xs}}$ 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, $^{210}\text{Pb}_{\text{xs}}$ 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 $^{210}\text{Pb}_{\text{xs}}$, 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

the accumulated sediment⁶. PCA on XRF data (Fig. S3) showed two sediment end-member³⁹ inputs from the watershed, with organic matter and metallic pollutant contents and the marine carbonate productivity. The geochemical data were in good agreement with the LOI indicating an upward increase in terrigenous inputs from the watershed (reflected by the Fe content) while carbonate productivity (reflected by the Ca content) decreased (Fig. S3). Accordingly, the Fe/Ca ratio was used as a high-resolution proxy for the terrigenous fraction.

A chronological framework was established with the *serac* R package⁴⁰ from measurements of short-lived radionuclides, constrained by the identification of historical hurricane event deposits (Fig. 3, Table S2 and S3) as ¹³⁷Cs did not provide an interpretable profile in relation to its desorption/migration in marine sediments⁴¹. The Fe/Ca data in the GUA17-01 and GAL17-04 cores led to the identification of 4 and 3 historical hurricane events, respectively, that caused heavy precipitation in the region, as indicated by meteorological data (<http://pluiesextremes.meteo.fr/>), which allow independent chronology validation. The sediment deposits triggered by these events were considered as instantaneous and thus excluded from the construction of the age model, by removing the depth interval and associated ²¹⁰Pb_{xs} data of each of these deposits^{40,42}. The logarithmic plot of the event corrected ²¹⁰Pb_{xs} activity shows a general decrease, with two distinct linear trends (black and dark blue Fig. 3). According to the “constant flux, constant sedimentation rate” (CFCS) model⁴⁰ applied to each segment of the profile, the levels of ²¹⁰Pb_{xs} indicate drastic increases in the sediment accumulation rate (SAR) from $3.9 \pm 0.6 \text{ mm.y}^{-1}$ to $43.3 \pm 9.5 \text{ mm.y}^{-1}$ and from $5.0 \pm 0.2 \text{ mm.y}^{-1}$ to $22.1 \pm 5.1 \text{ mm.y}^{-1}$ with synchronous changes in 2000 (uncertainty range: 1996-2005) and 1999 (uncertainty: 1995-2004) in the GUA17-01 and GAL17-04 cores, respectively (Fig. 3). These two age models are well constrained by the ages of the most intense historical hurricanes that have hit both islands. This concomitant increase in sedimentation rates

235 in sediment records located more than 170 km away from each other might only be explained by
236 an increase in carbonate productivity in the marine system or by an increase in terrigenous inputs
237 from the watersheds. From these age models, the terrigenous mass accumulation was calculated
238 and interpreted as a proxy of soil erosion in the watershed, indicating that the erosion rate increased
239 10- and 4-fold on the basis of the GUA17601 and GAL17-04 cores, respectively (Fig. 4a, b). This
240 interpretation rules out the hypothesis of an increase in carbonate precipitation. It is further
241 supported by the observation of a synchronous increase in the Fe/Ca ratio for GUA core (Fig. 4a).
242 For the GAL core we observed a short time lag (corresponding to less than 2cm) between Fe/Ca
243 and terrigenous flux increase which could be related to sampling resolution (2cm for short-lived
244 radionuclides not in continuous versus continuous 2mm for XRF data) or to the influence of Iris
245 hurricane (Fig. 4b).

Figure 3: Chronology for the GUA17-01 (a) and GAL17-04 (b) cores with the Fe/Ca, $^{210}\text{Pb}_{\text{xs}}$ activity, instantaneous event-corrected $^{210}\text{Pb}_{\text{xs}}$ activity and age model with uncertainties (gray area) realized with the serac R package⁴⁰. The dotted lines extending from the black lines indicate age model extrapolation. On the right part of this figure ages and names correspond to historical hurricanes.

253 Pesticide temporal variations are presented in relation to age and displayed both in concentrations
254 (Fig. S4) and fluxes (Fig. 4). CLD and chlordecol (CLO, a CLD degradation product)
255 concentrations present similar profiles in both cores (Fig. S4) and were thus summed and expressed
256 as fluxes (Fig. 4) to account for the large increases in sedimentation rates. In GUA17-01, the
257 CLD+CLO flux is first detected in 1978 $\pm$ 7 yr, just after the introduction of CLD in 1972 and
258 subsequently decreases, which could correspond to the period (1978-1982) between the
259 prohibition of Kepone[®] in the US and the approval of the license for the use of CLD in Curlone[®]
260 by the French authorities in late 1981. Then, the CLD+CLO flux increases, which continues even
261 after its ban in 1993. In 2000 $\pm$ 5 yr, we observed a 2-fold and more than 5-fold increase in the
262 CLD+CLO concentration and flux, respectively. In GAL17-04, CLD and CLO are first observed
263 in very low concentrations at 8 cm (1940-1955), before its introduction dated here at 1970 $\pm$ 6 yr
264 (Fig. S4). This apparent preintroduction record of this compound may be explained by its possible
265 downward diffusion in sediment, as observed for other chlorinated molecules such as DDT⁶, or by
266 bioturbation in these shallow marine environments. Then, we observe a decrease in CLD+CLO
267 flux during the prohibition period, followed by an increase until the final ban. In 1999 $\pm$ 5 yr, we
268 detected a 2-fold and more than 10-fold increase in the concentration and flux, respectively.

269 Finally, between the period of maximum CLD use and the recent high flux, we noted more than
270 200-fold and 20-fold CLD+CLO flux increases in GUA17-01 and GAL17-04, respectively (Fig.
271 4). The CLO/CLD ratio is higher in the oldest part of the two sediment cores than in the most
272 recent sediment (Fig. S4) and in present-day soils in Martinique⁴³. CLO formation probably results
273 from the reduction of the CLD ketone group under anaerobic conditions⁴⁴, 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 watershed¹⁹ and from 30 to more than 24000 ng.g⁻¹ in the Pérou watershed²³. This spatial variability in CLD soil pollution is related both to the duration of the banana cropping period and soil characteristics¹⁰. Soils in which banana was never planted did not contain any CLD¹⁹. 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, respectively⁴⁵, 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 mouths⁴⁶.

Figure 4: Soil erosion proxies and pesticide flux chronology for Guadeloupe **a** and Martinique **b**: Erosion proxies with Fe/Ca and terrigenous fluxes compared to a pesticide chronology (logarithmic scale) reconstructed from the sum of glyphosate and aminomethylphosphonic acid (AMPA; glyphosate degradation product) fluxes and the sum of chlordecone and chlordol (one of the chlordecone degradation products) fluxes. The horizontal dotted line corresponds to the large increase in sedimentation rate identified in response to a large erosion increase synchronous with glyphosate introduction and chlordecone flux increases. <DL indicates below the detection 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 process²⁴ which should therefore not be neglected or considered minor, contrary to recent suggestions²².

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 area⁴⁷. 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 fields⁴⁸. 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 FWI²⁸. This is in good agreement with the emergence of this chemical in 1999/2000 $\pm$ 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 chemicals⁶ 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 $\pm$ 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

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

Even if the two watersheds are different in term of farm size, other important watershed characteristics are the same: soils cover, type of cultures, climate. At both sites, we observed a synchronous increase in CLD+CLO and erosional fluxes when glyphosate was first widely applied to banana fields at the end of the 1990s. The application of glyphosate, which disrupts grass development, has a strong effect on soil erosion as previous demonstrated through monitoring plots, simulation experiments and retrospective observation^{6,25–27,49}. Other herbicides, such as paraquat, and authorized in the French West Indies between 2003 and 2007, could have caused the same issues. Paraquat was reported to be used along with glyphosate because of glyphosate resistant plants⁵⁰ and intensively used in banana plantations in Latin America⁵¹. While glyphosate is a systemic herbicide, paraquat and other herbicides used at the early 2000s on banana plantations²⁸ are either selective or non-systemic. As a result, they do not eliminate all plants on a field, allowing them to recover. Paraquat, for instance is a non-selective but non-systemic herbicide. It “burns” the leaves of plants while allowing plants to recover from stems or roots. Glyphosate acts differently by killing the whole plants which leaves the soil bare and less-maintained by roots leading to potential enhanced erosion rates. Erosion on bare soil might furthermore be amplified by banana canopies, which exhibit high funneling ratios, favouring runoff even on soils with a high infiltration rate such as Andosols, by localized rainfall redistribution and soil detachment by concentrated flows⁵².

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

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

AUTHOR INFORMATION

Corresponding Author

* corresponding author : pierre.sabatier@univ-smb.fr <https://orcid.org/0000-0002-9620-1514>

401 Author Contributions

402 P.S., C.M., I.C., J.P. and O.E. conceived and designed the study. Fieldwork sampling were
403 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.
404 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.
405 P.S. wrote the first draft of the manuscript, with subsequent contribution by all the authors.

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418 ABBREVIATIONS

419 CLD: Chlordecone

420 CLO: Chlordecol

421 AMPA: Aminomethylphosphonic acid

422 DDT: Dichlorodiphenyltrichloroethane

423 FWI : French west indies

424 CZ: Critical zone

425 LOI : Loss of ignition

426 XRF : X-ray fluorescence

427 DBD : Dry bulk density

428 NCIR : Noncarbonate igneous residue

429 CFCS: Constant flux, constant sedimentation rate

430 PCA : Principal component analysis

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