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▶ To cite this version:

Rémi Bizeul, Oriane Lajoie, Olivier Cerdan, Lai Ting Pak, Anthony Foucher, et al.. Delayed environmental pollution caused by transient landscape storage - An example from the Lesser Antilles. Environmental Pollution, 2024, 366, pp.125412. 10.1016/j.envpol.2024.125412. cea-04830927

HAL Id: cea-04830927 https://cea.hal.science/cea-04830927v1

Submitted on 11 Dec 2024

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Environmental Pollution



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Delayed environmental pollution caused by transient landscape storage — An example from the Lesser Antilles

Rémi Bizeul^{a,*}, Oriane Lajoie^a, Olivier Cerdan^b, Lai Ting Pak^c, Anthony Foucher^a, Sylvain Huon^d, Thomas Grangeon^b, Olivier Evrard^a

^a Laboratoire des Sciences du Climat et de l'Environnement (LSCE-IPSL), Université Paris-Saclay, Unité Mixte de Recherche 8212 (CEA/CNRS/UVSQ), Orme des Merisiers, 91191 Gif-Sur-Yvette, France

^b Bureau de Recherches Géologiques et Minières (BRGM), Orléans, F-45060, France

^c UPR HortSys, Cirad, F-97285 Le Lamentin, Martinique, France

^d Sorbonne Université, Institut d'Ecologie et des Sciences de l'Environnement de Paris, 4 place Jussieu, Paris, 75005, France

ARTICLE INFO

Keywords: Chlordecone Banana cultivation Tropics Caribbean

ABSTRACT

The strong pest pressure on intensive banana cultivation in the French West Indies led to the intensive use of chlordecone (an organochlorine insecticide) between 1972 and 1993. Due to its high toxicity for the population and the environment, many studies were conducted on the transfer of chlordecone over the last 20 years. However, most studies focused on the dissolved fraction of chlordecone, while the particle-bound fraction was understudied. Therefore, this study reconstructs pluri-decadal erosion rates (i.e. 1980-2023) and associated chlordecone particle-bound transfers from soil and sediment cores sampled in a cultivated headwater catchment (Saint-Esprit, Martinique). Based on sediment accumulation analyses in an agricultural reservoir, high erosion rates (i.e. 10 t ha^{-1} yr⁻¹) were found in the investigated catchment during the study period, with values exceeding the estimated tolerable soil loss rate in tropical contexts (*i.e.* 2.2 t ha⁻¹ yr⁻¹). Based on the analysis of soil cores sampled along a banana plantation hillslope, this study highlights the formation of colluvial deposits with high levels of chlordecone contamination. When these areas are affected by erosion processes, this leads to massive remobilization of particle-bound chlordecone to water bodies. Indeed, in sediment sampled in the downstream reservoir, we observed a drastic increase in these transfers since 2006, synchronous with changes in agricultural practices. This study therefore highlighted the occurrence of legacy contamination at toeslope positions, which was estimated to potentially persist for 4000 to 11,000 years. Such a residence time highlights the need to implement changes in land management to effectively reduce erosion of agricultural soils, particularly in areas identified as "temporary deposition zones" for chlordecone contamination, in order to protect downstream water bodies from chlordecone transfer. To achieve this, agricultural practices that may increase soil erosion, such as herbicide application or intensive ploughing, should be minimized. Overall, this study improved our understanding of erosion and associated chlordecone transfers in tropical environments.

1. Introduction

In the French West Indies (FWI), agriculture mainly consists in intensive banana and sugarcane cultivation. The insular and humid tropical context has led to a rapid diversification of pests and weeds. In this context, chlordecone was introduced in banana plantations in 1972 to control the banana weevil (*Cosmopolites sordidus*), before being banned in 1993 in the FWI (Cabidoche et al., 2009) for health and environmental reasons. Chlordecone ($C_{10}Cl_{10}O$) is an organochlorine insecticide that has been listed as a persistent organic pollutant (POP) under the Stockholm Convention since 2009. FWI is likely the area in the world where this substance has been used most intensively and

its probably the most documented and studied location as well. In Martinique, the high contamination risk areas are estimated to cover 40% of the cultivated surfaces (Dromard et al., 2022). Chlordecone is known to have a strong affinity for organic matter as well as clay particles, which are abundant in tropical soils, explaining the chlordecone sequestration in these soils (Cabidoche et al., 2006). Despite its prohibition since 1993 in the FWI, chlordecone is still detected in several environmental compartments, including soils, water bodies (Wintz and Pak, 2021) and living organisms (Dromard et al., 2018; Méndez-Fernandez, 2018). The presence of chlordecone in the environment causes serious problems for people and ecosystems. In 2010, a study

https://doi.org/10.1016/j.envpol.2024.125412

Received 2 August 2024; Received in revised form 15 November 2024; Accepted 27 November 2024 Available online 5 December 2024

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^{*} Corresponding author. E-mail address: remi.bizeul@lsce.ipsl.fr (R. Bizeul).

showed that the average dietary intake of chlordecone per inhabitant in the FWI was around 3.3 µg per day (Maudouit and Rochoy, 2019). The presence of chlordecone in the blood can lead to neurological disorders ("Kepone shake") from 1 to 2 mg L⁻¹ (Picot and Rabache, 2012; Maudouit and Rochoy, 2019). In addition, chlordecone has had a major impact on the fishing industry in the FWI by contaminating marine ecosystems (*e.g.* phytoplankton, fish, algae) and reducing fishing areas (Dromard et al., 2022).

The use of chlordecone in banana plantations takes place in a global context of agricultural intensification and land use change (e.g.mechanization, intensive ploughing, chemical weeding). In response to these changes, an acceleration of soil erosion has been observed over the last 60 years (Owens, 2020). This anthropogenic erosion leads to an increase in riverine sediment fluxes and ultimately in sediment supply to water reservoirs (Owens et al., 2005). Small tropical island ecosystems (< 5000 km² (Browning and Sawyer, 2021)) are highly sensitive to erosion processes, particularly due to the combination of extreme climatic events (Browning and Sawyer, 2021) and steep slopes (Thiery et al., 2017). Soil erosion has a detrimental effect on agriculture by removing the most fertile surface soil horizon, incorporating denser subsoil into the surface layer, and reducing the root zone of the soil (Venkapaten, 2012; FAO, 2019). Another major consequence of soil erosion is related to the transfer of pesticides along the land-to-sea continuum (Owens et al., 2005; Sabatier et al., 2021). This is a situation that can typically be observed in the FWI. By measuring significant fluxes of chlordecone in sediments deposited in the Galion Bay (Martinique, France), Sabatier et al. (2021) highlighted the occurrence of massive chlordecone remobilization since the late 1990s, which the authors explained by agricultural intensification (e.g. glyphosate application, extensive tillage, mechanization). By further exposing the soil to erosion processes, these practices increased the chlordecone remobilization and its transfer along the land-to-sea continuum. These results highlight the important role of particle-bound transport in chlordecone transfer. Therefore, this study suggests that soil erosion from contaminated areas may increase the dispersion of chlordecone in the environment.

Several studies regarding chlordecone transfers in the FWI focused on the dissolved phase (Ponchant et al., 2020). Indeed, the highly permeable tropical volcanic soils (Charlier et al., 2008) lead to the migration of chlordecone along the soil profile, following water infiltration, which may result in groundwater contamination (Mottes et al., 2020). However, when rainwater runoff generates particle removal, the surface transfer of particle-bound chlordecone can become a significant process for its downslope and downstream dispersion (Comité de Pilotage Scientifique National Chlordécone, 2022). Nevertheless, questions remain about the fate of chlordecone in soils under banana plantations (Cabidoche et al., 2006; Comte et al., 2022), including in particle-bound form as few studies have focused on this fraction (Ponchant et al., 2020). It is therefore essential to estimate the erosion rates of chlordecone contaminated soils. In Martinique, few data are available to quantify soil erosion and arable soil loss (Saffache, 2000; Khamsouk, 2001). However, these available studies provide information for short periods (1 to 2 years) and were conducted more than 20 years ago, before the recent acceleration in the intensification of agricultural practices (Sabatier et al., 2021). To the best of our knowledge, erosion rates and associated transfers have never been reconstructed from the period of chlordecone introduction to the present day. In the absence of long-term monitoring, the study of sedimentary archives is an indirect method to reconstruct temporal variations of erosion rates and to compare them with changes in land management. Indeed, by recording sediment inputs into the reservoir over the last 40 years, the sediment archives provide relevant tools for retrospectively estimating erosion rates. This long-term reconstruction is essential to better understand the relationship between erosion and chlordecone transfers, which may serve as a scientific basis for designing relevant mitigation strategies aiming at reducing chlordecone impacts along the land-to-sea continuum. To address these issues, this study proposes to reconstruct the erosion rates and to quantify the associated chlordecone transfers over the last several decades (> 40 years) by using a combination of hillslope soil cores and sediment cores collected in a pond draining a small headwater catchment representative of the cultivated areas of the FWI. The factors influencing erosion rates over time and their role in chlordecone transfers were investigated. This study aims to improve our understanding of the dynamics of chlordecone transfers along a cultivated hillslope and, ultimately, in the sediment deposited in a downstream agricultural reservoir. By identifying high-risk areas and chlordecone transfer processes, the results of this study may provide guidelines for future land management policies, in order to mitigate the transfer of chlordecone to aquatic ecosystems.

2. Materials and methods

2.1. Study site

The Duchene headwater catchment (0.16 km²) is located in the Rivière Salée catchment (65 km²), in the southern part of Martinique Island. This part of the island is characterized by low annual rainfall (1500 mm an⁻¹) and a relatively flat topography. Regarding geology and pedology features, the studied area was characterized by hyaloclastite deposits and Ferralsols. Regarding land cover, in February 2023, the study area was covered with banana plantations in the southern part, and sugarcane in the northern and western parts (15% and 46% of the catchment area, respectively). Fallow lands were found in the western and southern parts, occupying 30% of the headwater catchment area. Two residential and farm building zones (i.e. anthropized surfaces) are also found both to the north and to the south-west, covering 9% of the headwater catchment surface. A shallow reservoir of 1.6 ha, used for irrigation, is located at the catchment outlet, in the eastern part (Fig. 1). Moreover, it should be noted that several gullies have been observed on the slopes drained by the Duchene reservoir, which indicates the occurrence of significative erosion processes. Generally, these gullies were observed in the inter-rows in banana plantations and were directly connected to the reservoir.

According to aerial images (source: Institut national de l'information géographique et forestière, IGN), the pond was created between 1974 and 1979, coinciding with the chlordecone application period (*i.e.* 1972–1993). In addition, thanks to aerial images and residents' testimonies, we know that banana cultivation took place during the chlordecone application period in the Duchene headwater catchment. No dredging operations have been conducted in the reservoir since its construction. Moreover, optimal connectivity between the slopes and the water body is ensured by the limited size of this headwater catchment, with a restricted hydrographic network and significant slopes (mean = 8.7%). Overall, this headwater catchment is a well-adapted site within the FWI for studying the long-term relationship between erosion and chlordecone transfers.

2.2. Sampling

Three sediment cores were collected in February 2023 across the reservoir (Fig. 1) using a Uwitec gravity corer equipped with a hammer from a floating platform: DUC02 (1 m-long; N° IGSN 10.58052/ IEFOU002I), DUC03 (0.89-m-long; N° IGSN 10.58052/IEFOU002J) and DUC04 (0.98-m-long; N° IGSN 10.58052/IEFOU002K). Additionally, using a Cobra TT vibrocorer, three soil cores were collected across the active banana plantation that drains into the reservoir (Fig. 1) along a transect following the main slope direction: DUCS01 (0.95m-long; N° IGSN 10.58052/IEFOU002L), DUCS03 (0,79-m-long; N° IGSN 10.58052/IEFOU002N), DUCS05 (1-m-long; N° IGSN 10.58052/ IEFOU002P). The upper part of the studied slope was covered with fallow land, which was separated from the banana plantation by a road (i.e. pathway for agricultural machinery). A second road was located in the middle part of the banana plantation. The core locations were strategically selected to cover the changes in topography along the hillslope.



Fig. 1. (A) Location of Rivière Salée catchment and Duchene headwater catchment, (B) slope map and (C) land cover in February 2023 for the Duchene headwater catchment and location of the soil and sediment cores collected across the Duchene headwater catchment.

2.3. Gamma spectrometry

Radionuclide activity (Bq kg⁻¹) was determined by gamma spectrometry using coaxial HPGe detectors at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). The efficiency and background levels of the detectors were periodically controlled with internal and IAEA soil and sediment standards. Radionuclide activities were systematically decay-corrected to the sampling date, which is indicated in the metadata for each core (Bizeul et al., 2024). The 5-cm-depth increment subsamples collected along soil cores were packed into plastic boxes (≈ 60 g) and the ¹³⁷Cs activity was measured for 24 h in order to calculate soil redistribution along the cultivated hillslope of interest. As for the sediment cores, the subsamples were packed into plastic boxes (≈ 15 g) and analyzed for $^{210}\mbox{Pb}_{xx}$ activity in order to establish a core chronology and estimate sedimentation rates in the reservoir, which is used for estimating soil erosion rates (Eq. (2), (3)). Age-depth models were computed using the serac R package (Bruel and Sabatier, 2020) (Figs. S3-S5).

2.4. Pesticide concentration measurements

Chlordecone and chlordecol (*i.e.* a chlordecone transformation product) concentrations were measured in soil and sediment cores following the method for the determination of organic micropollutants after QUECHERS-type extraction and analysis in liquid chromatography coupled in tandem with a triple quadrupole spectrometer. Chlordecone concentration measurements were conducted by the *Laboratoire de la Drôme* (Valence, France). The detection limit for chlordecone was 10 μ g kg⁻¹, while it amounted to 13 μ g kg⁻¹ for chlordecol. From

these measurements, the inventories of chlordecone and chlordecone particle-bound inputs to the reservoir were calculated:

$$Pesticide \ inventory = \sum (([Pesticide] \cdot SAR \cdot DBD) \cdot Polygon \ surface)$$
(1)

where [*Pesticide*] is the concentration in chlordecone or chlordecol (in $\mu g g^{-1}$), *SAR* is the sediment accumulation rate (in cm yr⁻¹), *DBD* is the dry bulk density (in g cm⁻³) and *Polygon surface* is the surface of the polygon defined as representative of the coring area within the reservoir, and for each core (n = 3).

2.5. Soil and sediment dry bulk density measurements

Non-calibrated sediment density was recorded every 0.06 cm along the sediment cores using the Computer Tomography scanner (CT-Scan) images obtained using the equipment (GE Discovery CT750 HD) available at the DOSEO platform (French Atomic Energy and Alternative Energy Commission, CEA Paris-Saclay, France). Grey values were extracted from the reconstructed scanner images using the free software ImageJ. These grey values were converted into dry bulk density (DBD) values and calibrated by measuring the absolute DBD (g cm⁻³) over 1.3 cm every 5 cm along the sediment cores.

2.6. Geochemical properties (x-ray fluorescence)

Geochemical properties along the cores were assessed using an AvaaTech XRF Core Scanner available at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). Measurements were performed every 1 mm along sediment cores. Results were expressed in counts per second (cps). Among the analyzed geochemical properties, variations of Ti were used as a terrigenous proxy for age-model calibration (Figs. S3–S5), as this element has been widely used in the literature to identify instantaneous sediment inputs (Bruel and Sabatier, 2020).

2.7. Organic matter properties

Total organic carbon (TOC) concentrations were measured to quantify the organic fraction of the sediment inputs into the reservoir. Moreover, to identify the signature of organic matter contained in the sediment deposited in the reservoir and to discriminate the respective contributions of sugarcane cultivation and banana plantation, total nitrogen (TN) concentration and carbon stable isotope (δ^{13} C) were also measured.

The potential occurrence of inorganic phases (carbonate minerals) was required to be determined prior to the analysis of TOC and TN concentrations and δ^{13} C composition. To this end, sediment sample aliquots were put into 100mL of deionized water and agitated using a magnetic agitator. After 24 h, water pH was measured. Samples with values rising above ca 8 indicate that carbonate minerals equilibrated with water and released bicarbonate ions, thereby reflecting the presence of minerals that can be easily dissolved such as calcite or aragonite. For sediment cores collected in the Duchene headwater catchment reservoir, the absence of carbonate minerals was confirmed. Total organic carbon (TOC) and total nitrogen (TN) elemental concentrations and stable isotope ratio (δ^{13} C) were determined in sediment core 5-cm increment depth subsamples by the combustion method using a continuous flow elemental analyzer (Elementar VarioPyro cube) coupled with an Isotope Ratios Mass Spectometer (EA-IRMS) at the Institute of Ecology and Environmental Sciences (iEES, Paris, France). Oxygen for combustion was injected for 70 s (30 mL.min⁻¹) and the temperature was set at 850 °C and 1120 °C for the reduction and combustion furnaces, respectively (Agnihotri et al., 2014). The analytical precision was assessed with repeated analysis of a tyrosine laboratory standard (n = 28 during the course of this study), calibrated against international reference standards (Girardin and Mariotti, 1991).

2.8. Soil erosion rates estimation

For each sediment core, the mass accumulation rate was estimated:

$$MAR = SAR \cdot DBD \tag{2}$$

where MAR is the mass accumulation rate (in g cm⁻² yr⁻¹), SAR (in cm yr⁻¹) is the sediment accumulation rate (derived from sediment core dating) and DBD (in g cm⁻³) is the dry bulk density (derived from CT-Scan images). Erosion rates were reconstructed by considering the three sediment cores as representative of the sediment deposited in the reservoir. In our study, we assumed, as it is traditionally done in the literature (Ben Slimane et al., 2013) for hillslope retention ponds such as the Duchene reservoir, that limited outflow occurred. Moreover, in the specific context of our study, submitted to tropical storms and a significant annual rainfall depth, with very high erosion rates occurring as demonstrated by our measurements, sediment leakage due to overflow is assumed to have little influence on the estimated erosion rates. Thus, the estimated erosion rates in this study may be considered as minimum values.

Therefore, the reservoir was divided into three polygons taking into account both sediment core sampling location and study site topography (Fig. S6). Then, an inventory of sediment quantity was computed based on MAR values (convert in t $\text{cm}^{-2} \text{ yr}^{-1}$) and polygon surfaces (in cm^2). This inventory was divided by the surface of the headwater catchment (in ha) to estimate the mean erosion rate:

$$Mean \ erosion \ rate = \frac{\sum (MAR \cdot Polygon \ surface)}{headwater \ catchment \ surface}$$
(3)

2.9. Reconstructed history of duchene headwater catchment

Over the period covered by sediment cores dating (i.e. 1980–2022), we digitized land use at different periods and selected the most intense rainfall events occurring at the nearby Saint-Esprit rainfall station. This information was used to investigate the impact of both climate variations and land management on the variations of surface erosion magnitude during the period of investigation. Based on MeteoFrance data including daily precipitation (Fig. S1) and storm reports, the major tropical rainfall events that have affected the Duchene headwater catchment between 1980 and 2023 were identified as follows : Klaus (1990), Iris (1995), Hortense (1996), Dean (2007) and Elsa (2021). Among the multiple tropical storms that occurred in the Lesser Antilles during the study period, these events were retained as their duration and intensity were considered to be of high magnitude in the study area. Events that affected other areas of Martinique were not considered in the current analysis. Land management changes within the Duchene headwater catchment were partially reconstructed during the 1980-2023 period, based on farmers' testimonies, aerial images (IGN, Fig. S2) and the available literature. Pesticide application could be reconstructed through the combination of a literature review and the compilation of French governmental data (Banque Nationale des Ventes de produits phytopharmaceutiques par les Distributeurs agréés (BNV-D)) for the whole of Martinique Island.

3. Results

3.1. Chlordecone transfers along the cultivated hillslope

According to Miller and Schaetzl's (2015) classification, the hillslope position of the soil cores was defined as : summit, backslope and toeslope. Along the core located at the summit of the hillslope. chlordecone contents ranged between 591 μ g kg⁻¹ in the upper part to 161 μ g kg⁻¹ at 40 cm depth, with a strong decrease (57%) observed at 25 cm, from 591 to 255 μ g kg⁻¹. Chlordecol concentration ranged between 27 and 13 µg kg⁻¹. Radiocesium showed activities decreasing from 2.2 to 0.5 Bq kg⁻¹ with depth. It was no longer detected below 30 cm, a depth coinciding with the strong chlordecone content decrease. Regarding the soil core collected at backslope position, chlordecone contents ranged between 576 $\mu g \ kg^{-1}$ and 218 $\mu g \ kg^{-1}$ with a decrease at 60 cm (-38%), from 493 to 304 µg kg⁻¹. Chlordecol concentration showed a maximum value at 40 cm (27.2 μ g kg⁻¹) and the substance was not detected in three layers along the core : between 5-15 cm, 25-30 cm and 60-75 cm. Radiocesium activities ranged from 3.1 to 0.9 in the 0-30 cm layer and, as for the summit core, it was no longer detected under 30 cm. The core located at toeslope position showed higher chlordecone and chlordecol contents compared to the two other cores. Indeed, chlordecone contents ranged from 2914 μg kg^{-1} in the upper part to 76 $\mu g \; kg^{-1}$ in the lowest part. These values were on average 5 times higher compared to those found in the two other cores. Chlordecol contents showed values from 160.2 $\mu g \ kg^{-1}$ at 40 cm to 18.2 μ g kg⁻¹ at the bottom of the core, these values being 4 to 10 times higher compared to those found in the summit soil core. Regarding radiocesium activities, they also showed higher values compared to those found in the two other soil cores, ranging from 5.5 Bq kg⁻¹ at 70 cm to 1.9 Bq kg⁻¹ at 45 cm. Moreover, the shape of the radiocesium profile along the toeslope core was different from that of the other soil cores. Indeed, the maximum radiocesium activity was found at 65-70 cm depth. Overall, for the three measured properties (i.e. chlordecone, chlordecol contents and radiocesium activities), an increasing trend in concentrations was observed in toeslope direction (Fig. 2). Indeed, for instance, average chlordecone contents varied from $352~\mu g~kg^{-1}$ in the summit core to $437~\mu g~kg^{-1}$ in the backslope core and even to 1241 μ g kg⁻¹ in the toeslope core.



Fig. 2. Description of the studied cultivated hillslope in 2023 (land use, slope characteristics), and variations of chlordecone, chlordecol (*i.e.* chlordecone transformation product) contents ($\mu g k g^{-1}$) and 137 Cs activities (Bq kg⁻¹) along the analyzed soil profiles. The upper core of the transect is a local highpoint, disconnected from potential upstream inputs.

3.2. Sediment accumulation in the reservoir

The temporal trends of erosion rates reconstructed in the Duchene headwater catchment (Fig. 3) showed erosion rates during the study period ranging from 7.9 to 11.9 t ha⁻¹ yr⁻¹. Moreover, significant variations were observed, following a cyclical pattern ranging from 2 to 8 years. Overall, a general increase trend (Mann-Kendall test, pvalue = 0.01) was observed between 1980 and 2023 in the study area. Indeed, the last phase of variations, from 2011 to 2019, displayed the higher erosion rates since 1980, with values ranging between 10.9 and 11.7 t ha⁻¹ yr⁻¹. The chlordecone inventory showed a global 44%increase between 1980 and 2005, from 24 g yr⁻¹ to 43 g yr⁻¹ (Fig. 3). Chlordecol was not detected in sediment until 1989. Between 1989 and 2005, the chlordecol inventory showed a slight increase, from 0.92 g yr^{-1} to 1.08 g yr^{-1} . Since 2006, a drastic increase of both the chlordecone and chlordecol inputs was observed in the sediments of the reservoir, moving from 80 g yr^{-1} to 148 g yr^{-1} for chlordecone and from 12 g yr⁻¹ to 38 g yr⁻¹ for chlordecol. Accordingly, the chlordecone and chlordecol inputs into the reservoir increased 4-fold and 45-fold, respectively, during the 2005-2023 period. This period is concomitant with the most intensive phase of erosion recorded between 2011-2019 period and the fifth phase (2006-2008). The maximum chlordecone inputs into the reservoir were observed in 2019 (162 g yr⁻¹) while – for chlordecol – they were recorded in 2018 (48 g yr⁻¹). The organic matter fluxes showed a strong decrease in the early

1980s, from 0.07 g cm⁻² yr⁻¹ in 1980 to 0.03 g cm⁻² yr⁻¹ in 1984. Between 1984 and 2005, organic inputs into the Duchene reservoir showed several phase variations, following the erosion rate tendency. In contrast, between 2006 and 2023, organic inputs increased continuously, from 0.03 g cm⁻² yr⁻¹ in 2006 to 0.06 g cm⁻² yr⁻¹ in 2023. The analysis of TOC/TN evolution gives us information concerning the source of organic matter inputs in the Duchene reservoir (Lamb et al., 2006) (Fig. S7). Two main sources of organic matter can be accumulated into the reservoir: allochtonous (i.e. from soil erosion) or autochtonous (i.e. reservoir biological activity). Based on Lamb et al. (2006) work, we know that a TOC/TN ratio above 7 indicates a main contribution of soil organic matter source while a TOC/TN ratio under 7 indicates a main contribution of organic matter from autochtonous biological activity. In the Duchene reservoir, the TOC/TN ratio shows two periods of high biological activity. The first, in the early 2000s, is synchronous with a decrease in organic matter inputs in the reservoir. The second period occurred in 2015-2018. Moreover, based on the comparison of TOC/TN and δ^{13} C values (Fig. S7, Lamb et al., 2006), it can be determined that the organic matter signature of sediment matched well with to that of soils covered with plants following the C3photosynthetic pathway (i.e., banana tree). This dominant contribution of soils under banana plantations is consistent with previous work conducted in FWI (Khamsouk and Roose, 2001). Indeed, it has been shown that this type of cultivation increases the susceptibility of soil to erosion processes.

3.3. Reconstructed history of land management and extreme rainfall events

By combining information regarding tropical storms and land use changes that took place in the Duchene headwater catchment and further, in Martinique Island, we can partially reconstruct sedimentationrelated aspects of the environmental history of the study area (Fig. 3). These results therefore allow us to investigate the links between the changes that occurred in the study area and the temporal variations of erosion rates. In 1990, tropical cyclone Klaus storm, which generated 238 mm of rainfall in 24 h (i.e., 10% of annual rainfall), was synchronous with the second increasing phase of mean erosion rates (Fig. 3). However, the other tropical storms did not match with phases of increases in erosion rates. This finding implies that the single occurrence of extreme events does not explain alone the temporal variations of erosion rates. Among the periods that can be outlined for a specific management, some of them coincide with periods highlighted by the interpretation of sediment cores as increasing phases of erosion rates. Indeed, the first reconstructed land use changes that took place in 1981 occurred during the first phase of erosion increase. At this time, aerial images showed that the major part of the Duchene headwater catchment (> 60%) was covered with bare soil under young banana trees. Three other periods with significant bare soil cover were observed at different locations across the headwater catchment: in the southern part in 2013 (i.e. ploughed banana plantations) and in the northern part in 2014 and 2020 (i.e. ploughed sugarcane plantation). These three periods of ploughing were synchronous with periods of erosion rate and pesticide inventory increases into the reservoir (Fig. 3). The introduction of glyphosate in Martinique occurred in the late 1990s (Chabrier and Queneherve, 2003) and its use grew rapidly afterwards, with a peak during the 2013-2015 period (André et al., 2024), before a decrease of its application in 2018 (Lala and Bocaly, 2020). This observation may be confronted to the increase of erosion rates observed during the 2005-2023 period and the recorded maxima of sediment yields and pesticide inputs in 2018-2019 (Fig. 3).

4. Discussion

4.1. First long-term estimates and driving factors of erosion rates

This study highlighted the occurrence of high erosion rates under banana plantations, with values (*i.e.* ~ 10 t ha⁻¹ yr⁻¹) generally exceeding the soil loss rates estimated to be tolerable in wet tropical contexts (*i.e.* 2.2 t ha⁻¹ yr⁻¹, Montgomery (2007)). Moreover, if we assume that a part of sediment originating from the subcatchment has not been trapped in the Duchene reservoir, then the erosion rates estimated in this study would provide minimum values. This high magnitude of erosion rates may potentially lead to a deterioration of soil functions and the associated ecosystem services (FAO, 2019), which constitutes a major threat to the future of agriculture on the island. Moreover, high erosion rates may contribute to pesticide transfers from agricultural land to river systems, water bodies and, ultimately, the coastal environments (Sabatier et al., 2014).

The occurrence of intense rainfall events may lead to erosion processes generating significant soil loss (Goujon, 1968; Labrière et al., 2015). In Martinique, erosion rates were also shown to increase drastically during tropical storms (Saffache, 2000). This situation was observed in the Duchene headwater catchment in 1991 during the passage of Klaus storm (Fig. 3). Although it is beyond the scope of this paper to explore the detailed relationship between rainfall and erosion, no direct relationship was found between the occurrence of intense rainfall events and erosion rates. It suggests that, in complex environments such as volcanic tropical islands exposed to heavy storms, rainfall may not be the single dominant factor driving erosion rate dynamics. Overall, the magnitude of surface erosion processes is indeed strongly linked to the joint occurrence of periods during which soils are sensitive to soil erosion and that of intense rainfall events. In particular, the sensitivity of soil to erosion processes is strongly linked to land management. The vegetation cover is a significant factor controlling the response of soils to erosion processes (Labrière et al., 2015) as well as slope steepness and soil erodibility, as conceptualized in the widely used USLE equation (Wischmeier and Smith, 1978). Under natural vegetation cover (*i.e.* forest), nutrient inputs from rainfall, dust and *in situ* biological activity are greater than organic matter losses through erosion. However, when land is cleared, this chain of accumulation is broken and the soil nutrient is disequilibrated (Roose, 1984). Under tropical climate conditions, soil erosion is concentrated spatially (bare soil, non-protected crop fields between rotations) and temporally (*i.e.* before vegetation is fully established) (Labrière et al., 2015). In the studied area, a synchronicity between the occurrence of bare soil or fields showing a sparse vegetation cover and erosion rate increases was observed, particularly during the 2006–2023 period (Fig. 3).

To control vegetation cover, several substances were applied under croplands. Among the herbicides available, glyphosate has been the most frequently used in the FWI. In 2021, Martinique was the second region in terms of glyphosate consumption in France (1.14 kg ha⁻¹, André et al., 2024). There are various uses for glyphosate: pathway cleaning (for the human passage or that of agricultural machinery), controlling weeds during cultivation (only authorized under banana plantations, and prohibited for sugarcane cultivation), when initiating an agricultural cycle (to start the cultivation on a "clean" soil) or at the end of the cycle (only under banana plantations to destroy banana trees). Accordingly, glyphosate was extensively used in Martinique since the late 1990s. Its use has increased in particular since 1998 for the destruction of banana trees (Chabrier and Queneherve, 2003). The maximum period of glyphosate sales in Martinique occurred in 2013-2015 (André et al., 2024). However, the application of this substance leading to the removal of vegetation cover increased the sensitivity of soils to erosion processes. In the Duchene headwater catchment, the period of glyphosate application was concomitant with that of the increase of erosion rates (Fig. 3). This may confirm the previously observed effect of this herbicide application on soil erosion susceptibility (Sabatier et al., 2014). Moreover, by increasing soil carbon loss (Venkapaten, 2012) and causing the mechanical disintegration of macroaggregates (Barthès et al., 2000, 1-0.2 mm soil fraction after sieving following Keeper et Rosenau (1986) procedure), the ploughing practices play also a major role on soil loss under cultivated land. In Martinique, ploughing intensity and frequency vary depending on crop types (e.g. banana, sugarcane, pineapple). Nevertheless, a global mechanization of this practice occurred in the late 1990s or in the early 2000s depending on farm size. Since this period, the use of excavators became widespread in the FWI. In the Duchene headwater catchment, a synchronous increase of organic inputs into the reservoir and erosion rates was observed since 2006 (Fig. 3). This may therefore explain soil erosion increase due to an intensification of ploughing in the Duchene croplands during the early 2000s. Among agricultural practices, irrigation can also play a major role in soil loss. The southeastern part of Martinique has the lowest annual rainfall (1250 mm yr⁻¹) compared to the other regions of the island. Crop irrigation developed strongly in the 1980s and has continued to grow ever since (Burac, 1991). It was during this period that several irrigation reservoirs were built, such as the Manzo dam located a few hundred meters from the study area. The reservoir investigated in the current research was built around 1978 (according to aerial photographs) and is also used to irrigate croplands across the catchment. The water is conveyed by pipes laid on the soil surface. This type of irrigation is known to generate massive soil loss (Sojka et al., 2015). Indeed, along a furrow irrigation structure, all three phases of erosion may be observed (i.e. detachment; transport, and deposition). These processes lead to significant particlebound transfers along the cultivated hillslope and ultimately, to the reservoir. Overall, several factors may explain the increase in erosion rates reconstructed since 2006 in the Duchene headwater catchment.



Fig. 3. Temporal variations of mean erosion rates, chlordecone and chlordecol inventories, organic inputs (black line) and TOC/TN values (ratio between Total Organic Carbon and Total Nitrogen, grey bars) in the Duchene headwater catchment as reconstructed from the analysis of sediment cores in the reservoir. Periods of application of chlordecone and glyphosate are also indicated. The five major tropical storms that occurred during the 1980–2023 period include (A) Klaus (1991), (B) Iris (1995), (C) Hortense (1996), Dean (2007) and Elsa (2021). Based on farmers' testimonies and aerial images, the land use and cover changes in Duchene headwater catchment could be reconstructed: (1) Bare soil under young banana trees (1981), (2) Fallow lands covered the total surface of the headwater catchment (1988), (3) Algal bloom in the reservoir, fallow land on the northern part (1992), (4) Initiation of sugarcane cultivation in Duchene headwater catchment (1999), (5) the entire surface of the headwater catchment was cultivated (banana and sugarcane cultivation, 2004), (6) Bare soil on the southern part (ploughed banana plantation, 2013), (7) Bare soil on the northern part (ploughed sugarcane plantation) and fallow land on the western part.

Changes in agricultural practices during this period made the soils more sensitive to erosion processes. Associated with this increase in soil loss, an acceleration in the transfer of chlordecone, chlordecol and organic carbon to the reservoir was also observed since the early 2000s.

4.2. Impacts of soil erosion on chlordecone transfers

The analysis of soil cores collected along the cultivated hillslope highlighted an intensive chlordecone transfer down the slope (Fig. 2). Thus, soil eroded from the summit of the slope is likely deposited on the toeslope following the main erosion pathways (e.g. gullies). This soil transfer is associated with a transfer of chlordecone and chlordecol, leading to the formation of a highly contaminated colluvial deposit on the toeslope (> 1000 μ g kg⁻¹ down to 70 cm). These processes are supported by the massive input of radiocesium and chlordecone in the toeslope soil profile to 80 cm, highlighting an accumulation of eroded soil material from the summit to the toeslope, close to the reservoir (i.e. 16 m from the reservoir). Ultimately, these erosion pathways transport the particles and contaminants to the reservoir, which is directly connected to the studied hillslope. Nevertheless, since 2006, a concomitant drastic increase in erosion rates, chlordecone transfers and organic inputs in reservoir sediment was observed. This means that there have been changes in the factors driving erosion. As described previously, in the late 1990s/early 2000s period, major changes occurred in agricultural practices (i.e. intensive ploughing, glyphosate application, irrigation). Based on the reconstructed history of the Duchene headwater catchment (Fig. 3), the entire surface of the headwater catchment is expected to have been covered with banana and sugarcane crops during this period. It is therefore likely that the study area was affected by these changes, which explain the increase in erosion rates. This increase was accompanied by an accelerated transfer of chlordecone and chlordecol into the reservoir. Based on this evidence, we deduced that eroded areas were more contaminated, such as the accumulation area observed in the cultivated slope. Regarding these observations, we assume that certain areas (located downslope or on flatter areas) act as "temporary deposition zones" for chlordecone and chlordecol. Indeed, this type of area was observed in the studied hillslope. When these areas are affected by intense erosion processes, which may be caused by agricultural practice changes or the occurrence of extreme rainfall events such as Klaus tropical storm, they generate a massive export of chlordecone and chlordecol. This massive export started to occur 13 years after the chlordecone ban. This observation can be linked to a recent study that has defined the concept of "pesticide resurrection" phenomena (Mottes et al., 2021). This hypothesis can be confirmed by the high concentration in chlordecol observed in the soil core collected on the toeslope and the drastic increase of chlordecol fluxes reconstructed with time based on the analysis of sediment archives from the reservoir. Moreover, the synchronous increase of organic matter inputs to the reservoir is a major indicator of the accelerated transfer of chlordecone. Indeed, due to the high affinity of chlordecone with organic matter, a decrease in soil organic matter under banana plantations may lead to further chlordecone remobilization (Cabidoche et al., 2006). Nevertheless, a part of the increase in organic matter inputs may correspond to autochtonous biological activity. Eutrophication phenomena could therefore be responsible for the low TOC/TN levels observed in the reservoir sediment.

Based on the mean estimated chlordecone application quantity in Martinique (*i.e.* 3 kg of active substance per hectare per year, Lesueur-Jannoyer et al. (2020)), we can estimate that around 990 kg of active substance was applied between 1972 and 1993 in the Duchene headwater catchment. Indeed, approximately 15 ha of banana plantations were treated during 21 years. Based on the measurements of chlordecone concentrations in sediment cores, the total quantity of chlordecone transferred to the reservoir is estimated to be around 3 kg. Accordingly, only 0.3% of the total chlordecone quantity applied in the Duchene headwater catchment was transferred to the reservoir since 1980. After

2006, the annual export ratio increased, from 0.008% to 0.015% in 2023 of the total chlordecone applied (Fig. 4). Assuming a constant ratio (*i.e.* reference year = 2023) and particulate transfer only, the total time required for the export of chlordecone applied in the Duchene headwater catchment would range between 4000 and 11000 years (Table S.1). Obviously, this simplistic calculation should not be interpreted as a result as such. However, it does provide two major pieces of information: (1) the quantity of chlordecone still present in soils remains very high, and transfers to water bodies will persist in coming decades at a high level and (2) transfers are only at an early stage, and it remains therefore very timely to make decisions to reduce them. In the literature, chlordecone persistence time in soils is currently being debated, as estimations range from decades to several centuries (Cabidoche et al., 2006; Comte et al., 2022). The wide range of estimations of the persistence of chlordecone in the FWI calls for a more systematic evaluation of chlordecone stocks in soils (Clostre et al., 2014). The method proposed in this study (i.e. using a vibrocorer, measuring chlordecone concentration in 5-cm increments down to 80 cm) provides a more comprehensive estimate of stocks. Thus, this method can be applied in contrasted cultivated slopes (*i.e.* different soil type properties, slope characteristics and land cover managements) to provide insights regarding chlordecone transfer mitigation. Overall, the issues raised by this small-scale study can be extended to a more global context. The particulate transfer of chlordecone remains an understudied subject, despite this pesticide having been used in various regions worldwide, including the FWI, Africa, and India. For this reason, studies similar to this one should be conducted not only in the FWI but also in other affected regions to better understand the factors influencing the transfer of chlordecone into the environment and to evaluate its impact on land and water resources.

5. Conclusions

The current study provided a robust estimation of soil erosion rates for banana plantations under tropical climate conditions. In addition, it provided new insights into chlordecone storage and particle-bound transfers in a cultivated headwater catchment representative of the banana plantations widely found across the FWI. The estimated rates are very high (10 t ha^{-1} yr^{-1}) exceeding tolerable soil loss rates (2.2 t ha^{-1} yr⁻¹). This is unsustainable and threatens the future of agriculture (i.e. loss of arable land) and the environment (e.g. transfer of pesticides). These results fill a major knowledge gap in the FWI regarding the quantification of erosion rates and their role in chlordecone transfers. Based on soil core analyses, we highlighted the major role of erosion on chlordecone transfers along a cultivated slope. This phenomenon led to the formation of "temporary deposition zones" (i.e. colluvial deposits) and "hotspots" of chlordecone contamination. Its vertical extent is significant, resulting in the higher contamination of the deepest layers of the soil, down to 80 cm. The acceleration of erosion during the last 20 years can be attributed to changes in agricultural practices (i.e. intensive ploughing, glyphosate application). Synchronously with this increase in erosion rates, an accelerated particle-bound transfer of chlordecone was observed in sediment cores. We assume that "hotspots" of chlordecone contamination, as observed along the studied slope, were affected by these changes and led recently to massive chlordecone remobilization (previously referred to as "pesticide resurrection", Mottes et al. (2021)). However, the stocks of chlordecone in the soil are still very high (99.7% of the applied quantity) and suggest a persistent contamination. Overall, these results represent a major advance in our understanding of chlordecone transfer processes in cultivated catchments of the FWI. By quantifying the particle-bound chlordecone transfers and highlighting the role of erosion, this work provides information that should be considered by authorities and stakeholders to help limit the transfer of chlordecone from contaminated fields to lower river systems, water bodies and, ultimately, the coastal ecosystems.



Fig. 4. Estimations of applied and transferred chlordecone quantities in the Duchene headwater catchment since 1972.

Regarding the estimated chlordecone transfer time (i.e. 4000-11000 years), changes in land management practices are essential to reduce chlordecone transfers and avoid long-term transfers within the environment. It is therefore necessary to limit erosion of agricultural soils, in particular in areas identified as "temporary deposition zones" for chlordecone contamination. To achieve this, agricultural practices such as herbicide application or intensive ploughing need to be reduced. In order to take feasible and effective measures, further studies following the protocol proposed in the current research should be carried out in other cultivated hillslopes of the FWI to provide an overall view of the potential heterogeneity of these processes. Overall, based on a coupled analysis of pesticides found in catchment soils and reservoir sediment, this work have been provided a significant step forward to improve our understanding of soil storage and environmental transfers of chlordecone in the FWI. To expand the interpretations made in this study, the analysis of contrasted cultivated hillslopes (i.e. showing different soil type properties or different slope characteristics) would be particularly insightful.

CRediT authorship contribution statement

Rémi Bizeul: Writing – original draft, Visualization, Software, Methodology, Formal analysis, Data curation, Conceptualization. Oriane Lajoie: Software, Methodology, Formal analysis, Conceptualization. Olivier Cerdan: Writing – review & editing, Supervision, Investigation. Lai Ting Pak: Writing – review & editing, Supervision. Anthony Foucher: Writing – review & editing, Investigation, Conceptualization. Sylvain Huon: Resources, Investigation. Thomas Grangeon: Writing – review & editing, Investigation. Olivier Evrard: Writing – review & editing, Supervision, Investigation, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Remi Bizeul reports financial support was provided by the Prefecture de Martinique. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by Plan Chlordecone 2021–2027 (Projet SEA9- Chlordecone, Prefecture de Martinique, France).

Data availability

The dataset is available online at Bizeul et al. (2024).

Appendix A

- A.1. Reconstructed history of land management and extreme rainfall events
- A.1.1. Precipitations data (Saint-Esprit, Martinique) See Figs. S1 and S2.

A.2. Core chronology

Based on ${}^{210}\text{Pb}_{xs}$ activities measured by gamma spectrometry and a terrigenous proxy (Ti) measured by X-ray fluorescence spectrometry, age-depth models were computed for each sediment core using the serac R package (Bruel and Sabatier, 2020). See Fig. S3, Fig. S4 and Fig. S5.

A.3. Estimation of erosion rates

In order to estimate erosion rates into the Duchene subcatchment, the reservoir was divided into three polygons. Their respective surfaces were defined taking into account sediment core sampling location and study site topography. See Fig. S6

A.4. Organic input, sediment yield and chlordecone fluxes in the duchene reservoir

See Fig. S7.

A.5. Chlordecone transfer times

Based on chlordecone inventories and the estimated quantity of chlordecone applied in the Duchene subcatchment (Lesueur-Jannoyer et al., 2020), the time required to export all the chlordecone stored in the soils by erosion can be estimated.

$$Transfer time = \frac{(Q_{applied} - Q_{transferred})}{Annual \ export}$$
(4)

where $Q_{applied}$ is the quantity of chlordecone applied in the banana plantations of the Duchene subcatchment during the 1972–1993 period, $Q_{exported}$ is the quantity of chlordecone transferred to the reservoir and *Annual export* is the quantity of chlordecone exported during one year (kg yr⁻¹). By computing all the uncertainties associated with the calculation of chlordecone inventories (*i.e.* DBD, SAR and chlordecone concentration measurements), the range of chlordecone transfer times in years can be estimated.



Fig. S1. Daily precipitations at the Saint-Esprit station (source: MétéoFrance) between 1980 and 2022.

Table S.1

Minimum, mean and maximum values estimated for (1) annual export of chlordecone to the Duchene reservoir (2) total transferred chlordecone quantity to the reservoir and (3) transfer of chlordecone in soil obtained by applying Eq. (4).

Reference	Annual export	Annual export	Annual export	CLD applied	CLD transferred	CLD transferred	CLD transferred	Transfer time	Transfer time	Transfer time
year	(inean - kg yi-i)	(iiiii - kg yi-i)	(max - kg yr-r)	Rg	(inean - kg)	(iiiii - kg)	(max - kg)	(inean - y)	(11111 - y)	(max - y)
1980	0.024	0.016	0.032	990	2.9	1.9	4.2	41 995	61088	30913
1981	0.016	0.009	0.026	990	2.9	1.9	4.2	61 315	10///3	38 381
1962	0.010	0.011	0.022	990	2.9	1.9	4.2	50162	91393	44 001
1965	0.017	0.011	0.023	990	2.9	1.9	4.2	72.625	125 /12	42130
1964	0.014	0.007	0.022	990	2.9	1.9	4.2	72023 50646	133412	44 027
1985	0.017	0.011	0.025	990	2.9	1.9	4.2	51 454	82 622	93 23 9
1960	0.019	0.012	0.029	990	2.9	1.9	4.2	51454	83 033 70 E76	34 433
1987	0.020	0.012	0.023	990	2.9	1.9	4.2	52 946	81 452	37.069
1989	0.020	0.012	0.029	990	2.9	1.9	4.2	48 724	74 444	34 311
1990	0.020	0.013	0.032	990	2.9	1.9	4.2	44 103	69.052	30.464
1991	0.022	0.019	0.041	990	2.9	1.9	4.2	34 21 4	53 080	23 808
1992	0.025	0.015	0.041	990	2.9	1.9	4.2	36.214	59 969	23 781
1992	0.027	0.010	0.041	990	2.9	1.9	4.2	25.026	55 107	25 1 97
1993	0.027	0.018	0.039	990	2.9	1.9	4.2	25 259	52 600	23187
1994	0.025	0.015	0.040	990	2.9	1.9	4.2	20 427	65 202	24 007
1995	0.025	0.013	0.038	990	2.9	1.9	4.2	29.062	41 471	20.295
1990	0.035	0.024	0.048	990	2.9	1.9	4.2	26 520	41 47 1	19742
1997	0.037	0.024	0.055	990	2.9	1.9	4.2	20 330	26 220	16 027
1998	0.041	0.027	0.058	990	2.9	1.9	4.2	23 911	30 320	10 92/
1999	0.043	0.027	0.062	990	2.9	1.9	4.2	23 223	30 696 47 E9E	13/01
2000	0.034	0.021	0.052	990	2.9	1.9	4.2	20000	47 525	16 802
2001	0.030	0.020	0.058	990	2.9	1.9	4.2	2/ 3/3	49 303	10 092
2002	0.047	0.030	0.069	990	2.9	1.9	4.2	20 903	24 122	14 240
2003	0.045	0.023	0.005	990	2.9	1.9	4.2	21 933	42 264	16 765
2004	0.039	0.023	0.059	990	2.9	1.9	4.2	234/3	42 204 25 144	16 / 03
2005	0.043	0.028	0.062	990	2.9	1.9	4.2	220/3	10 027	10021
2000	0.080	0.052	0.115	990	2.9	1.9	4.2	12,391	10 93/	7661
2007	0.089	0.057	0.129	990	2.9	1.9	4.2	11 002	17 209	7001
2008	0.067	0.055	0.128	990	2.9	1.9	4.2	11 550	17 970	10 227
2009	0.007	0.044	0.095	990	2.9	1.9	4.2	19734	10 207	10.32/
2010	0.079	0.051	0.114	990	2.9	1.9	4.2	0150	12 792	6547
2011	0.100	0.072	0.151	990	2.9	1.9	4.2	9139	12 465	6459
2012	0.000	0.075	0.133	990	2.9	1.9	4.2	9001	15 104	7025
2013	0.033	0.005	0.140	990	2.9	1.9	4.2	9942	12 217	6450
2014	0.110	0.074	0.135	990	2.9	1.9	4.2	8240	12 122	5610
2015	0.120	0.075	0.176	990	2.9	1.9	4.2	7101	10 722	5010
2010	0.13/	0.092	0.191	990	2.9	1.9	4.2	7191	10732	4045
2017	0.157	0.005	0.234	990	2.2	1.9	4.2	6277	10 168	4213
2010	0.162	0.097	0.237	990	2.2	1.9	4.2	6100	10 209	3086
2019	0.154	0.097	0.247	990	2.2	1.9	4.2	6407	9676	4554
2020	0.134	0.102	0.210	990	2.2	1.9	4.2	6665	9808	4856
2021	0.140	0.101	0.203	990	2.2	1.9	4.2	6106	0113	4301
2022	0.102	0.100	0.227	990	2.2	1.9	4.2	6680	11 646	4249
2023	0.140	0.000	0.232		2.7	1.7	7.4	0000	11040	7477



Fig. S2. Aerial images of the Duchene subcatchment (obtained from Institut national de l'information géographique et forestière, IGN).



Fig. S3. Age-depth model for the DUC02 core.



Fig. S4. Age-depth model for the DUC03 core.



Fig. S5. Age-depth model for the DUC04 core.



Fig. S6. Polygons used for erosion rates estimation.



Fig. S7. Sediment inputs (organic and inorganic fluxes) and pesticide fluxes (chlordecone and one of its degradation products, chlordecol) for the three sediment cores sampled in the Duchene subcatchment reservoir. $\delta^{13}C$ and TOC/TN ranges were also plotted for characterizing potential organic inputs into the reservoir (freshwater particulate organic carbon (POC) and soil organic matter under C₃ or C₄ terrestrial plants) compared to sediment organic matter signatures (modified according to Lamb et al. (2006)).

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