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Soil and river contamination patterns of chlordecone in a tropical volcanic catchment in the French West Indies (Guadeloupe)^{*}

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ABSTRACT

The aim of this study was to identify primary flow paths involved in the chlordecone (CLD) river contamination and quantify the CLD fluxes to assess CLD pollution levels and duration according to a typical catchment of the banana cropping area in the French Indies (Guadeloupe): the Pérou Catchment (12 km²) characterized by heavy rainfall (5686 mm year⁻¹). Three sub-catchments (SC1, SC2 and SC3) were studied during the hydrological year 2009–2010: a pedological survey combined with a spatialized hydrochemical approach was conducted. The average soil concentration is higher in the Pérou Catchment $(3400 \ \mu g \ kg^{-1})$ than in the entire banana cropping area in Guadeloupe $(2100 \ \mu g \ kg^{-1})$. The results showed that CLD stocks in soils vary largely among soil types and farming systems: the weakest stocks are located upstream in SC1 (5 kg ha^{-1}), where a majority of the area is non-cultivated; medium stocks are located in Nitisols downstream in SC3 (9 kg ha⁻¹); and the greatest stocks are observed in SC2 on Andosols (12 kg ha⁻¹) characterized by large farms. The annual water balance and the hydro-chemical analysis revealed that the three sub-catchments exhibited different behaviors. Pérou River contamination was high during low flows, which highlighted that contamination primarily originated from groundwater contributions. The results showed that only a small part of the catchment (SC2), contributing little to the water flow, comprises a major CLD contribution, which is in agreement with the highly contaminated andosol soils observed there. Another significant result considers that at least 50 years would be required to export the totality of the actual CLD soil stocks retained in the topsoil layer. The actual time for soil remediation will however be much longer considering (i) the necessary time for the chlordecone to percolate and be stored in the shallow aquifers and (ii) its travel time to reach the river. © 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Persistent organic pollutants (POPs) are organic compounds that are resistant to degradation through chemical, biological, thermal or photolytic actions (Ritter et al., 1996). POPs are also characterized by high bioaccumulation, toxicity, hydrophobicity and a long halflife, resulting in environmental persistence (Jones and de Voogt, 1999; Nakata et al., 2002). They impact many ecosystems globally, such as in the French West Indies (FWI) where banana monocropping has been practiced over large areas (Dulcire and Cattan, 2002b, a) since approximately 1980, primarily with large chemical input (Clermont-Dauphin et al., 2004).

Among POPs, Chlordecone (CLD) ($C_{10}Cl_{10}O$) was used extensively by banana growers to control the banana weevil, *Comopolites sordidus (Germar, 1984)*, during two periods: 1972–1978 and 1982–1993, under trademarks Kepone and Curlone, respectively (Cabidoche et al., 2009). Chlordecone was included in Annex A of the Stockholm Convention on Persistent Organic Pollutants (UNEP, 2001) in May 2009. Although banned in 1993, recent several surveys conducted in Guadeloupe by the French Department of Environment and the French Department of Health highlighted a CLD presence in soils, spring water, rivers and drinking water (Cabidoche and Lesueur-Jannoyer, 2011; Dumont et al., 2009). Other studies have shown subsequent food resource contamination, including fish, root vegetables and terrestrial animals products (Bocquené and Franco, 2005; Coat et al., 2011; Dubuisson et al., 2007). Multigner et al. (2010) showed that CLD exposure in







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adulthood over a 30-year period favored the development of prostate cancer in the French West Indies. The ubiquity and toxicity of this molecule therefore raises the question of its long-term environmental fate, especially in soil and water resources, which is the topic of this paper within the specific context of a volcanic tropical environment.

Studies on the fate of CLD in tropical soils and waters remain limited. Cabidoche et al. (2006) and Levillain et al. (2012) showed that soils remain a primary source of pollution nearly twenty years following the last agricultural spreading: currently, soil CLD content remains important, reaching 10 mg kg⁻¹.

The retention of CLD varies according to the organic matter content of volcanic soils (Fernandez-Bayo et al., 2013a) and the physical sequestration in the fractal structure of soils containing allophanic clays (Woignier et al., 2012). The possibility of CLD degradation in volcanic soils was demonstrated but very small and ceased after a few months, confirming the high persistence of this compound (Fernández-Bayo et al., 2013b). Accordingly, Cabidoche et al. (2009) hypothesized that the natural dissipation of soil CLD contamination occurs only through leaching. Moreover, they estimated with a simple leaching model that the complete CLD dissipation in soil would last from several decades to several centuries, according to soil type and the level of soil contamination. Contamination within water and aquatic resources was observed as indicated above, but previous studies analyzed neither the patterns of water contamination at the catchment scale nor the pathways from soils to surface waters.

Hence, the aim of this study was to identify the patterns of soil and water CLD contamination in a typical catchment within the banana cropping area in the French West Indies. Specific aims were to identify the primary pathways involved in CLD soil-to-river transfer to quantify the CLD fluxes existing in the catchment and estimate by how much the fluxes represent remaining soil CLD. Accordingly, we studied the contamination levels and transfer of CLD in the Perou catchment located on Guadeloupe Island, FWI. We first characterized soil pollution (CLD content and stocks) and then characterized basin hydrological functioning. Then, we analyzed the change in CLD concentration in the river and water tables over one year to discuss the dominant pathways of CLD at the catchment scale.

2. Materials and methods

2.1. Study site

2.1.1. Location and climate

The Pérou Catchment belongs to the volcanic island of Basse-Terre, Guadeloupe on the Lesser Antilles insular arc, NW-SE oriented (Fig. 1). The Pérou Catchment is located on the Eastern windward flank of Capesterre Mountain, between altitudes of 25 and 1400 m amsl (above mean sea level), with its upper part located closely to the Soufrière volcano crater. The Perou catchment covers an area of 12.6 km².

The Lesser Antilles are characterized by a humid tropical climate with a marine influence and two distinct seasons: a dry season from February–March and a rainy season from July to November (Morell and Jérémie, 1994). The observed mean annual rainfall over 52 years (from 1952 to 2004) was 3636 mm at the Neufchâteau research station (16°0403800 N, 61°3600400 W, 250 m amsl), located 2 km northeast of the Pérou site (Fig. 1c). February and November are statistically the driest and wettest months of the year, with mean values of 161 and 465 mm, respectively (Météo-France, 2005) (Fig. 2). The mean annual rainfall of the Pérou Catchment was 4818.5 mm from October 2009 to September 2010, as observed over a set of rainfall gauges located within the catchment. The studied year was exceptionally dry in February, October,

and November and exceptionally rainy in June and September compare to an average year (over a period of 52 years) at the NeufChâteau station (Fig. 2). Finally, an important rainfall gradient with altitude is observed (Morell and Jérémie, 1994) from 2400 mm downstream to 6000 mm upstream.

2.1.2. Geology and land-use

Basse-Terre is a volcanic island located in the recent Lesser Antilles volcanic arc (Andreieff et al., 1989; Macdonald et al., 2000). The geological setting of the Basse-Terre Island is well described in Charlier et al. (2011). The subsurface of the Pérou Catchment is almost entirely composed of Basse-Terre Axial Chain volcanic formations of submarine (hyaloclastites) and sub-aerial (massive superposed andesitic flows) origin, shallowly covered by recent Grande Decouverte volcanic formations. Six primary geological formations can be found in the Pérou area (Dumont et al., 2009) according to the observations of Charlier et al. (2011) over a Pérou subcatchment and additional observations. The six formations from oldest to most recent include i) a weathered volcanic breccia, constituting a continuous substratum; ii) superposed fracture massive lava flows with subhorizontal flow fissuring, which is a mostly unweathered nonaquifer material covering a majority of the Pérou basin; iii) nuées ardentes pyroclastic flows extending southward; iv) Lahartype formations that locally outcrop at the study site; v) recent aerial pyroclastic deposits (pumice lapilli type), which are partially weathered and interstratified with ash levels nearly covering the basin with a thickness of several meters; and vi) alluvium located in the lower part of the Pérou Catchment and colluvium in several channels. Fig. 3a) shows the extensions of formations ii), iii), iv) and vi) that outcrop over the Pérou basin.

Three primary soil types are found on the southern side of the Basse-Terre island and on the Pérou Catchment (Fig. 3b) and are formed from the recent pyroclastic deposits of the lapilli type. The soils consist of Andosols with high allophane contents, Nitisols with high halloysite contents, and Ferralsols formed from the oldest pyroclastic deposits and with high halloysite and Fe-oxihydroxide contents, according to the IUSS soil classification (IUSS Working Group WRB, 2014) and descriptions in Colmet-Daage and Lagache (1969). These soils have a high infiltration capacity (saturated hydraulic conductivity greater than 60 mm h^{-1}) and a high carbon content (over 2%, Levillain et al., 2012). These high carbon contents favor soil retention of pesticides, including organochlorine compounds (Fernández-Bayo et al., 2013a). Among the three soils, Andosols have the highest CLD-retention capacity compared to Nitisols and Ferralsols, as indicated by CLD soil/water partition coefficients normalized to the soil organic content (Cabidoche et al., 2009). Andosols at the Pérou Catchment cover the largest area, whereas Ferralsols occur locally upstream in the non-cultivated portion of the basin, and Nitisols occur in the downstream portion of the basin.

The upstream portion of the catchment contains important forest coverage (55% of the catchment area). The other areas are covered primarily with banana (Musa spp.), and the downstream portion exhibits urban patches, as presented in Fig. 3c).

Dulcire and Cattan (2002b) mention that although the farming systems within the catchment are adapted to the tropical climate and fertile soils, they have been influenced by the succession of agricultural policies since the 20th century. These policies have defined five agro-ecological zones (Fig. 3d). Large farming systems with high input levels are located in the center of the basin and downstream (zones 2 and 5, which differ according to slope) and are characterized by large agricultural plots on Andosols. Small farmers cultivate the other zones, either upstream on recently cleared Andosol containing banana (zone 1), on diversified

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