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Flood-induced metal contamination in the topsoil of floodplain agricultural soils: A case-study in Colombia

José Marrugo-Negrete[1] | José Pinedo-Hernández[1] | Enrique M. Combatt[2] | Andrea G. Bravo[3] | Sergi Díez[3]

[1] Universidad de Córdoba, Carrera 6 No. 76-103, Montería 230002, Córdoba, Colombia

[2] Faculty of Agricultural Sciences, Department of Agricultural Engineering and Rural Development, University of Córdoba, Montería 230002, Colombia

[3] Environmental Chemistry Department, Institute of Environmental Assessment and Water Research, IDÆA-CSIC, Barcelona E-08034, Spain

Correspondence
J. Marrugo-Negrete, University of Córdoba, Montería, Colombia.
Email: jmarrugo@correo.unicordoba.edu.co
S. Díez, Environmental Chemistry Department, Institute of Environmental Assessment and Water Research, IDÆA-CSIC, Barcelona E-08034, Spain.
Email: sergi.diez@idaea.csic.es

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Abstract

Anomalously cold conditions in the tropical Pacific, related to a strong La Niña event, affected numerous countries in 2010. Several areas in Colombia were severely impacted, including agricultural fields in La Mojana floodplain that were completely flooded for almost 2 years. This study assesses the effects of flooding on the levels, spatial distribution, and sources of trace metals in 222 agricultural topsoils of this floodplain. Our results show that mean concentrations of Cu 48.4 mg kg^{-1} (11–103 mg kg^{-1}), Zn 79.2 mg kg^{-1} (6–207 mg kg^{-1}), Ni 58.1 mg kg^{-1} (17–101 mg kg^{-1}), Pb 3.2 mg kg^{-1} (0.13–14 mg kg^{-1}), Cd 0.56 mg kg^{-1} (0.05–1.4 mg kg^{-1}), Mn 411 mg kg^{-1} (55–1,277 mg kg^{-1}), and Hg 0.10 mg kg^{-1} (0.05–0.22 mg kg^{-1}) were higher than the world's averages. Topsoils revealed different degrees of pollution depending on the metal, of which Pb was the most problematic element, with 82% of the soil samples classified as heavily contaminated. Principal component analysis suggests that soil contamination was primarily derived from poor agricultural practices and contaminated river overflows from upstream mining areas. Our results show that an extreme flood event might increase the bioavailability of metals (especially Pb) for crops in most (66%) of the agricultural soils, posing a potential threat to human health.

KEYWORDS
bioavailable, flooding, Hg, La Niña, Pb, trace metal

1 | INTRODUCTION

Metals are persistent contaminants that pose threats to public health (Jaishankar, Tseten, Anbalagan, Mathew, & Beeregowda, 2014, and references therein). The increased use of metals for medical, industrial, technological, domestic, and agricultural practices has raised the levels of those contaminants in the environment (Tchounwou, Yedjou, Patlolla, & Sutton, 2012). Anthropogenic activities have increased the levels of trace metals (TM) in soils (Senesi, Baldassarre, Senesi, & Radina, 1999; Lin et al., 2005; Li, Wang, Gou, Li, & Wang, 2007), which are an important sink for TM globally (Wuana & Okieimen, 2011). For example, high metal concentrations have been found in agricultural soils located in catchments affected by historical metal mining (Barać et al., 2016; Marrugo-Negrete, Pinedo-Hernández, & Díez, 2017). Moreover, the use of pesticides and chemical fertilizers during agricultural practices has also increased the levels of different contaminants in soils (Jiao, Chen, Chang, & Page, 2012). The increase of metal concentration in soils has major economic implications because land usability for agricultural and cattle production has been reduced in soils contaminated by metals in order to ensure food safety and marketability (McLaughlin, Hamon, McLaren, Speir, & Rogers, 2000).

Rapid and extreme weather events affect metal storage, mobilization, and redistribution. For example, flooding events in floodplains contaminated soils are of special concern because an increase in the soils water content results in changes in their physicochemical properties (e.g., pH and redox; Speelmans et al., 2007; Rinklebe & Du Laing, 2011). A decrease in redox and pH might result in a higher solubility of metals, becoming more bioavailable to wildlife (Antoniadis et al., 2017). The potential accumulation of TM such as Hg, Cd, Pb, Cu, Ni, Mn, and Zn into agricultural soils and their uptake by crops are of concern because it may lead to their transfer through the food chain, which can affect human health (Li, Xie, Xu, & Sun, 2006). Furthermore, agricultural soils in floodplains (i.e., alluvial soils), fertile and enriched in nutrients (IUSS Working Group WRB, 2015), are vulnerable to extreme climatic events (Fischer, Shah, & Velthuizen, 2002; Handmer et al., 2012). This is critical as future climate scenarios predict an increase in floods and droughts frequency (Fischer & Knutti, 2015; Trenberth, 2011) making greater the risks to ecosystems, property and life from heavy rain, and flooding and wind damages (Mirza, 2003). For example, only the 2010–2011 La Niña phenomenon (the positive phase of El Niño Southern Oscillation) affected four million people in Colombia and caused about US$7.8 billion economical losses related to flooding of agricultural lands, destruction of infrastructure, and payment of government subsidies (Hoyos, Escobar, Restrepo, Arango, & Ortiz, 2013). It is of utmost importance to assess the effects of these extreme and frequent climatic events on the fate of toxic metals.

The region of La Mojana in Colombia, which is more than 500,000 fertile hectares in area, is part of a wetland complex formed by hundreds of swamps that serve to contain the floods of the Magdalena, Cauca, and San Jorge rivers. The region, with areas that are suitable for agriculture and swamps with significant fishing potential (NPD/UNDP, 2008), has been subjected to extreme floods and droughts in recent years. The socio-economic and ecological implications of these climatic events in La Mojana floodplain are worrisome, as half of the maize and rice production is lost during drought and maize production is entirely lost during flood events. Moreover, floods caused by the overflowing of the rivers, which collect wastes containing toxic metals from surrounding mining areas, have led to crop losses and severe pollution (Marrugo, Benítez, Olivero, Lans, & Vazquez, 2010; Marrugo-Negrete, Olivero Verbel, Lans Ceballos, & Norberto Benitez, 2008; Marrugo-Negrete, Pinedo-Hernández, & Díez, 2015; OCHA, 2011; Pinedo-Hernández, Marrugo-Negrete, & Díez, 2015). At present, the dry season lasts longer, and the rains are torrential, causing floods that can occasionally be severe, as in the 2010–2011 La Niña phenomenon, when several areas such as La Mojana were completely flooded for almost 2 years (Vargas, Hernández, & Pabón, 2018).

In spite the risks of metals remobilization under flooding conditions and the fact that wetlands are considered priority areas for environmental preservation by the Colombian Government/Ministry (NPD, 2013), the fate of pollutants in this area remains poorly described. To fill this gap of knowledge, and in order to increase the understanding of the effects of extreme flood events on the fate of pollutants, the main objective of this study is to evaluate the effect of the 2010–2011 La Niña flooding event on the fate of metals. For this purpose, we assessed the total concentration and bioavailable fraction of selected TM in flooded and nonflooded soils and identified the sources and spatial patterns of them. Our study shall serve as a framework of reference for the planning of productive activities and mechanisms to assess pollution in other flooded areas in future studies.

2 | MATERIALS AND METHODS

2.1 | Study area and soil sampling

The Mojana region (Figure 1) is located in north-western Colombia, with an area of approximately 5,500 km^2, surrounded by three main rivers (Magdalena, Cauca, and San Jorge) and crossed by many channels that drain the area during floods. This primarily occurs in the western part of the San Jorge River Basin, which is one of the most important swampy complexes in the country. During the rainy season, these rivers receive gold mine residues from the largest gold mining area in Colombia, located outside La Mojana (i.e., south of Bolivar and northeast of Antioquia).

In the regular flood pulse, the dry season starts in December and ends in March, with more frequent rain showers occurring from April to June and again in October and November. However, in recent years, La Mojana floodplain has suffered multiple cases of severe flooding. The latest flood, a La Niña event during the second semester of 2010, followed large floods in 2005, 2007, and 2008 (Rodríguez-Gaviria & Botero-Fernández, 2013; Vargas et al., 2018). Owing to the flooding of a vast area, more than 200,000 people were affected, including traditional fishing, agriculture, and livestock communities.

Topsoil samples (n = 222) from flooded (n = 148) and nonflooded (n = 74) agricultural soils were collected from cultivated fields (e.g., corn, rice, yucca, and banana) during the dry season of 2013 (Figure 1). Following Marrugo-Negrete et al. (2017), each of the soil samples was a composite of five subsamples from a 10 × 10 m square plot collected using a stainless steel trowel.

2.2 | Chemical analysis

The analytical method used to determine the total Hg concentration in the soil samples (0.1-g dry weight) was based on thermal decomposition detected by atomic absorption spectrometry using a milestone DMA-80 (direct mercury analyser; USEPA, 1998). For the analysis of the rest of the metals, soils samples (0.5 g) were digested with HNO_3/HCl 8:2 *v*/*v* in a microwave (USEPA, 2007). Metals concentrations were determined using a spectrometer Thermo Elemental Solaar S4 coupled with flame and graphite furnace. The organic matter was determined as the percentage loss on ignition of 2 g of sediment in an oven at 450°C for 4 hours (Coquery & Welbourn, 1995).

Different parameters such as pH and cation exchange capacity (CEC) were performed as described elsewhere (Marrugo-Negrete et al., 2017). The total phosphorus was determined by Bray & Kurtz (1945) procedure.

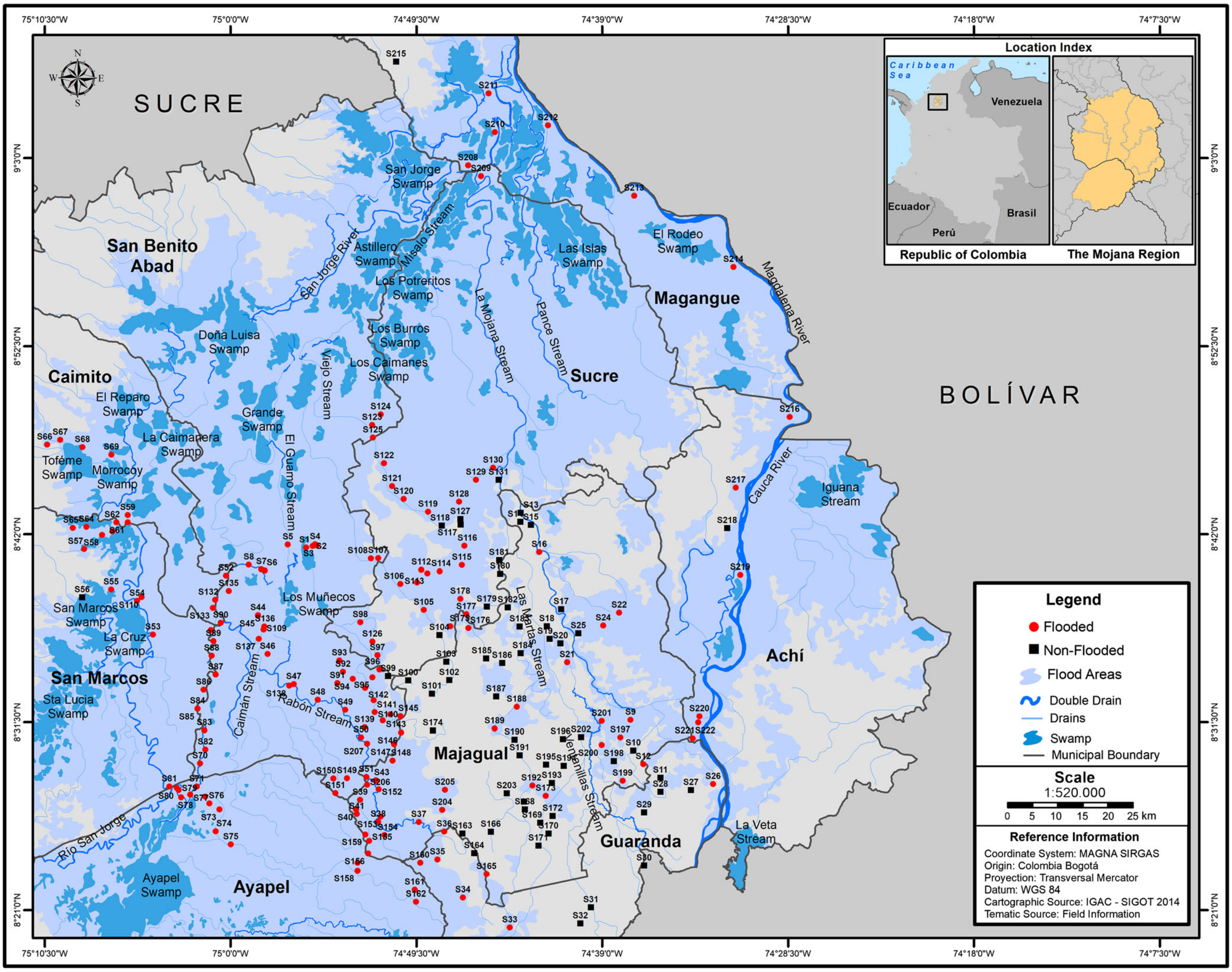

FIGURE 1 Map with the location of the sampling stations of the agricultural soils collected in the Mojana region [Colour figure can be viewed at wileyonlinelibrary.com]

Phosphate in the extract was determined colourimetrically employing a spectrophotometer UV-VIS Perkin Elmer Lambda 11 (Vorobiova, 2006). The bioavailable metal fraction, equivalent to that extracted in Step 1 of the BCR procedure (Rauret et al., 2000), was used to evaluate soluble and exchangeable metals: Cu, Zn, Ni, Cd, Hg, Mn, and Pb. To extract water-soluble, exchangeable, and carbonate-bound metals, 40 ml of 0.11-mol L^{-1} acetic acid was added to 1.0 g of the soil sample, which was then shaken (16 hours at 25°C). To this residue, 40 ml of 0.5-mol L^{-1} hydroxylamine hydrochloride was added and shaken for 16 hours to extract the reducible metals bound to Mn and Fe. For the analysis of sulphur (S) and exchangeable bases (Ca, Mg, K, and Na), extracted with calcium phosphate monobasic and ammonium acetate (pH = 7.0).

2.3 | Quality assurance/quality control

Quality control for metal determination consisted of method blank, triplicates, and the use of a certified reference material. The accuracy of the measurements was checked by analysing a certified reference material (CRM008-050; Resource Technology Corporation, USA). The percentage average recovery of metals was 97.8% (n = 3). The relative standard deviation for triplicate sample analysis was less than 10% for metals. The limits of detection for the different metals were 0.001 mg kg^{-1} for Hg, 0.008 mg kg^{-1} for Cd, 0.011 mg kg^{-1} for Pb, 0.1 mg kg^{-1} for Ni, 0.05 mg kg^{-1} for Zn, 0.05 mg kg^{-1} for Cu, 16.0 mg kg^{-1} for Mn, and 25 mg kg^{-1} for Fe.

2.4 | Determination of soil pollution

Three different indexes were used to determine the degree of soil contamination (Figure 2): the contamination factor (*CF*), which compares the measured value with the background level of the studied area (Muller, 1969); the geoaccumulation index (I_{geo}), which compares the measured value with the global geochemical background of the studied metal (Muller, 1969); and the risk assessment code (*RAC*), which assesses the bioavailability of the studied metal for food webs (Sundaray, Nayak, Lin, & Bhatta, 2011).

The *CF* was calculated by dividing the concentration of each metal (C_m) measured in the soil by mean concentration of the metal analysed in

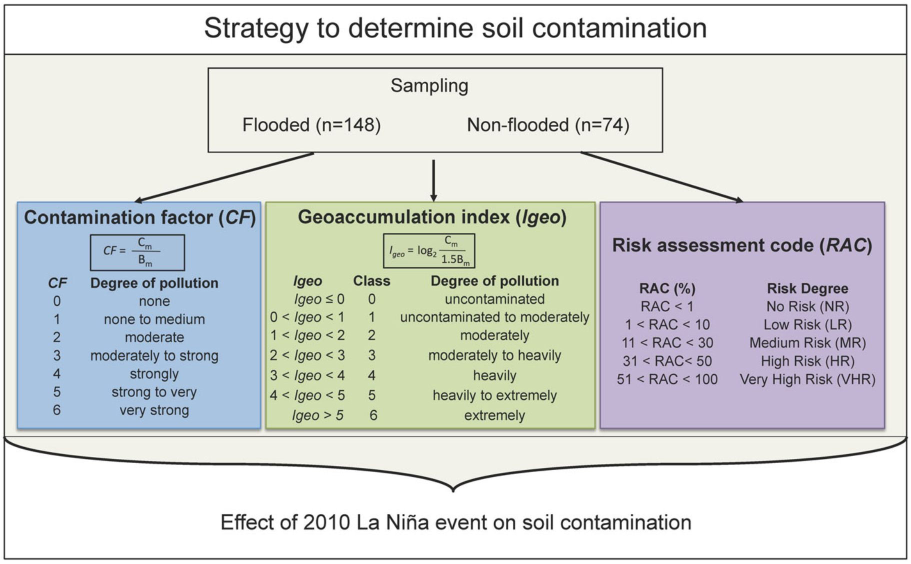

FIGURE 2 Methodology used to determine the degree of soil contamination [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 1 Concentrations of total trace metals and P (mg kg^{-1}), OM (%), CEC ($cmol_c$ kg^{-1}), and pH, in floodplain agricultural topsoils (n = 222) collected from the study area

	Cu	Ni	Pb	Cd	Zn	Mn	Hg	P	OM	CEC	pH
Mean	48.4	58.1	3.23	0.56	79.2	411	0.10	12.6	2.3	25.6	5.5
Median	47.9	58.3	3.0	0.5	79.8	397.2	0.1	9.16	2.2	25.3	5.4
SD	14.1	18.5	2.05	0.31	31.5	181	0.03	12.5	0.90	6.4	1.0
Min	10.8	16.8	0.13	0.05	6.3	55	0.05	3.74	0.17	5.3	3.8
Max	103	101	13.7	1.38	207	1277	0.22	86.3	4.6	43.8	8.9
CV (%)	29	32	64	55	40	44	29	99.8	43.3	25.0	17.4
MAC[1]	60–150	20–60	20–300	1–5	100–300	—	0.5–5				
World soil[1]	38.9	29	27	0.41	70	488	0.07				
Background soil[2]	21.2 (1.3)	23.7 (2.5)	0.23 (0.03)	0.14 (0.03)	22.1 (1.5)	178 (2)	0.051 (0.003)				

Abbreviations: CEC, cation exchange capacity; MAC, maximum allowable concentrations; OM, organic matter.

[1]Mean values compiled from Kabata-Pendias (2011).

[2]Mean values (*SD*) calculated from the concentrations of metals analysed in soils (n = 10) with no agricultural uses from other areas in Colombia.

soils (n = 5; Table 1) with no agricultural management from unflooded zones in the Mojana region (background value, B_m). The geoaccumulation index (I_{geo}) was calculated by dividing the metal concentration (C_m) measured in the studied soil samples by the geochemical background concentration of the metal (B_m) and the correction factor of 1.5, due to lithospheric effects (Muller, 1969). The RAC determines the availability of metals to be released and enters into the food chain (Sundaray et al., 2011). See Figure 2 for soil classification according to these indexes.

2.5 | Statistical analysis

The results are expressed as the mean, median, and standard deviation from triplicate set of data. Significant differences among the means at sampling stations were tested by Student's t test, with $p < 0.05$ for significance. Principal component analysis (PCA) was used to identify associations and common sources between metals. Statistical analysis was performed using SPSS v24.0.0.1.

3 | RESULTS AND DISCUSSION

3.1 | Soils characteristics

Physicochemical variables such as pH, OM, and CEC are shown in Table 1. The pH of the soil varied from 3.8 to 8.9. Most soils (n = 174) were acidic, with 78% with pH values < 6, which may induce the bioavailability of TM (Dragovic, Mihailovic, & Gajic, 2008; Kashem & Singh, 2001). Table 1 reveals topsoils ranging from 0.2% to 4.6% OM (mean, 2.3%). Most of the productive agricultural soils demonstrated OM content between 3% and 6%, and in our case, 40% of the samples had relatively low OM values between 0.2% and 2%. This

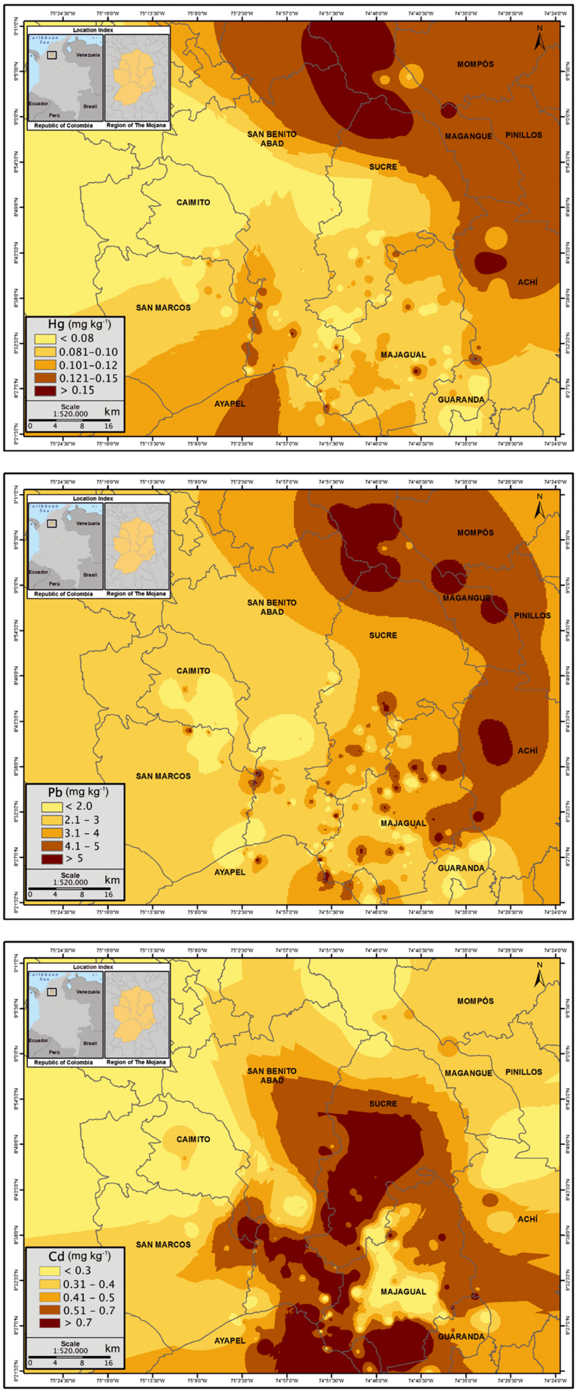

FIGURE 3 Spatial distribution of Hg, Pb, and Cd in surface soils of the study area [Colour figure can be viewed at wileyonlinelibrary.com]

means that approximately 60% of the soils had characteristics that restricted the mobility of metals, in a similar way to CEC, with most of the soils (97%) with values higher than 20 $cmol_c\ kg^{-1}$, which corresponds to soil texture for clay and clay loams.

3.2 | Occurrence and spatial distribution of TM in topsoils

Table 1 shows the concentrations of total TM in the topsoils of the floodplain. The descending order according to the mean values was Mn > Zn > Ni > Cu > Pb > Cd > Hg. The spatial distribution of different metals is presented in Figures 3 and S1. The spatial distribution of Pb and Hg reveals a similar trend, and their greatest presence is primarily in the north-eastern zones of the study area, whereas they are scarcely distributed in the central zone (Figure 3). Maximum levels of Ni, Cd, and Mn are spatially distributed in the central and southern zones. Cu and Zn are distributed throughout the study area without a marked tendency. No significant differences ($p > 0.05$) were found in metals from nonflooded and flooded topsoils, except for Mn, with higher levels in the former. Similar contents of Hg, Ni, Cd, Mn, and

Pb are generally found in the floodable areas of the Mojana region and are associated with mining wastes discharged into the streams of the main rivers.

We used different threshold and guideline international values because no limit values for metals in soils exist in Colombia. Concentrations of Cd, Pb, Cu, Ni, and Zn were observed within their respective permissible limits set by China (SEPA, 1995), the European Union (EU, 2002), India (Singh, Sharma, Agrawal, & Marshall, 2010), or Pakistan (Khan, Rehman, Khan, Khan, & Shah, 2010). The mean levels of all TM in topsoils were within the range commonly found in agricultural soils (Alloway, 2005) but higher than the corresponding values determined for the background soils (Table 1). For example, the levels of Pb and Cd were 14-times and four times higher, respectively, than those predicted in background soils.

The concentrations of metals did not present a clear trend (Figures 3 and S1); however, most of the maximum levels were found in southern floodable sites. Indeed, the highest concentrations of Cu, Ni, Mn, and Hg were found in Stations S40 (103.1 mg kg^{-1}), S207 (101.2 mg kg^{-1}), S94 (1,276.8 mg kg^{-1}), and S47 (0.22 mg kg^{-1}). The S128 station presented the highest concentration of Cd (1.4 mg kg $^{-1}$) in the middle of the region, whereas Zn reached its maximum levels at S219 (206.5 mg kg^{-1}), in the eastern part of the region, directly influenced by the flooding of the Cauca River. Finally, maximum Pb (S208; 13.7 mg kg^{-1}) was located in the northern part of La Mojana, at the margin of the river San Jorge, with significant deposition of sediments. In sum, the highest concentrations of TM were found in floodable zones, mainly associated with polluted loads from the Cauca and San Jorge rivers by mining activities. This can be explained by the fact that in recent years, there has been a significant increase in mining activities in the San Jorge River Basin. Some examples include the marked increase of ferronickel production, thanks to the installation of a new furnace to process the ore, the construction of a new thermal power station on the banks of the San Jorge River for the use of coal (NPD, 2009), the opening of new artisanal small-scale gold mines, and the recent entry of large-scale gold mining by multinational companies. The untreated wastes generated by these activities (mine tailings) are being discharged into aquatic ecosystems, which converge to La Mojana region, potentially contaminating their soils and water with metals. In addition, the Cauca River is the second most important river in the country and receives large amounts of domestic and industrial wastewater with large amounts of metals (Larmat & Soto-Duque, 2010). It also has rich gold mining areas, such as Marmato and Supía (Department of Caldas) and lower Cauca (Antioquia), where a large amount of waste (rich in metals and cyanide) is discharged either directly or via its tributaries (Prieto, 1998; Africano, 2002). During the rainy season, the river spills its waters into the Mojana region, through mouths that open on its shores as it passes (OCHA, 2011; PAHO/WHO, 2007), contaminating bodies of water with toxic elements, especially Hg (Africano, 2002; Marrugo-Negrete et al., 2008).

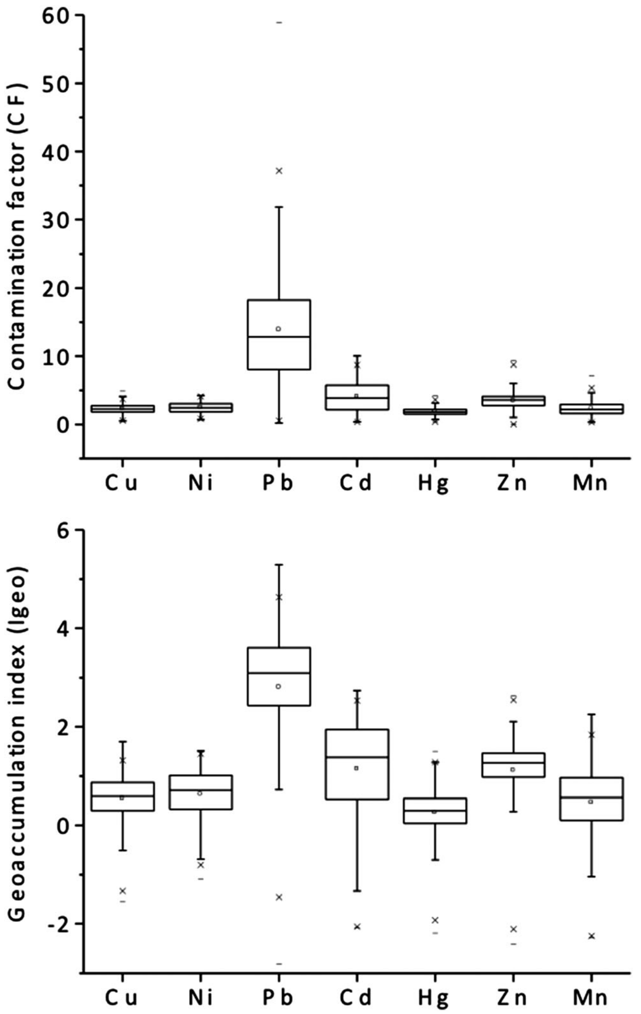

FIGURE 4 Box and whisker plots display the distributions of the different contamination indexes: contamination factor (a) and geoaccumulation index (b). Boxes depict 25th, 50th (median), and 75th percentiles and whiskers minimum and maximum values. Mean values (), outliers (x), and extremes (-)

Aside from mining activities, another source of anthropogenic contamination in the Mojana region comprises agricultural practices through the wasteful application of fertilizers (mainly phosphates) and pesticides containing traces of metals. The Mojana region is one of the major rice producers in the country, with a frequent application of superphosphates and other phosphorus fertilizers in paddy fields that contain high concentrations of Cu, Ni, Pb, Cd, and Zn, or even pesticides containing Cu and Zn.

3.3 | Assessment of soil pollution

The values of *CF* and I_{geo} in the topsoils are summarized in Figure 4. Furthermore, the different classes of *CF* and I_{geo} are summarized in Figure 2. The *CF* ranges for the different metals were 0.51–4.87 for Cu, 0.71–4.27 for Ni, 0.55–58.9 for Pb, 0.36–10.02 for Cd, 0.95–4.27 for Hg, 0.28–9.34 for Zn, and 0.31–7.17 for Mn (Figure 4a). The decreasing order according to the *CF* average for all metals was Pb (13.8) > Cd (4.1) > Zn (3.6) > Ni (2.5) > Cu (2.3) > Mn (2.3) > Hg (1.9). The average *CF* based on background values (Table 1) indicates contamination by anthropogenic sources for all metals. Indeed, 50% of the soil samples presented moderate to strong degrees of contamination for Cu (*CF*, 2–3). Moreover, 26% of the studied soils presented high levels of contamination by Ni (*CF*, 3–4). A very strong degree of contamination for Pb (*CF* > 6) is indicated in 82% of the soils, whereas 23% presented a very strong degree of contamination by Cd. A high degree of contamination by Hg (*CF*, 3–4) was seen in only 4% of the topsoils analysed and 5% in the cases of Zn and Mn. The *CF* values obtained in the present study were lower than the values recently reported for topsoils (Marrugo-Negrete et al., 2017) in the agricultural areas surrounding the Sinú River Basin of Colombia.

Figure 4b shows the values of the I_{geo} of TM. The mean I_{geo} values decreased in the order of Pb (2.8) > Cd (1.1) ≈ Zn (1.1) > Ni (0.6) > Cu (0.5) ≈ Mn (0.5) > Hg (0.3). Eighty-four percent and 70% of the samples showed uncontaminated to moderately contaminated levels of Cu and Ni, respectively. The most problematic element is Pb, with 82% of the soil samples classified as heavily contaminated (Class 4) to extremely contaminated (Class 5; Muller, 1969; see Figure 2). For Cd and Zn, 45% and 70% of the soils, respectively, were included in Class 2 (moderately contaminated). These results are consistent with *CF* values, thus corroborating the anthropogenic contribution to the soils of the Mojana region. Akin to the *CF* values, I_{geo} values are twice as low as Colombian agricultural soils along the Sinú River Basin (Marrugo-Negrete et al., 2017).

3.4 | TM sources

The relationships between TM content and surface soil properties are shown in Table S1. Aside from Cd, all of the metals were significantly positively correlated among themselves, which would suggest that they were derived from similar sources (e.g., minerals in mining activities). The only metal to have a significant correlation with Cd is P, implying a relationship between the application of P-based fertilizers and sources of this element (Grant & Sheppard, 2008). Most of the metals were significantly positively correlated with Fe and Mn (Table S1; $p < 0.01$, $p < 0.05$). These relationships of Cu, Pb, Zn, Ni, Cd, and Hg with Fe and/or Mn may suggest that metals might be absorbed and/or coprecipitated as components of crystal lattices of the minerals rather than as exchangeable ions. The results also shown that there is no significant correlation between soil pH and metal content, possibly due to the very wide pH range (Table 1), having little influence on elemental behaviour, similar than in Catalonian soils (Tume et al., 2006). However, OM was not correlated with TM, and conversely, the CEC presents significant correlation with Cu, Ni, Cd, Zn, and Mn, potentially indicating that an increase in CEC will increase the soil's ability to adsorb metals unspecifically (Bai et al., 2011).

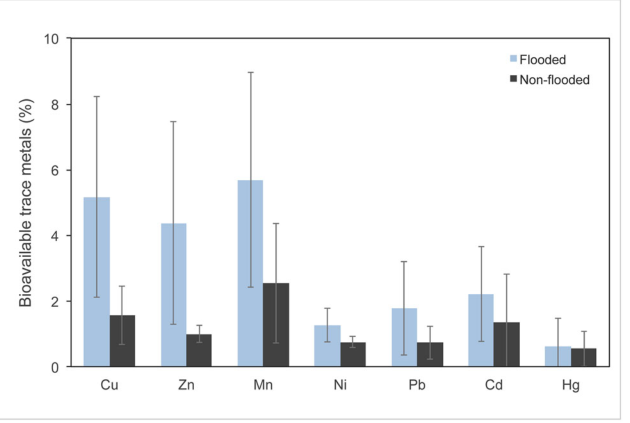

FIGURE 5 Percentage of bioavailable trace metals in flooded and nonflooded soils. Error bars are standard deviations of the average field sample [Colour figure can be viewed at wileyonlinelibrary.com]

TABLE 2 Relationships between bioavailable trace metals and pH, OM, and S by soil type

	Cu	Pb	Cd	Hg	Mn	Ni	Zn
Nonflooded topsoils (n = 74)							
pH	−0.068	−0.186	−0.551**	−0.319*	0.038	−0.023	−0.048
OM	0.047	0.083	−0.699**	−0.077	−0.036	−0.018	0.025
S	−0.161	0.258*	0.529**	0.069	0.110	−0.072	0.119
Mean bioavailable	0.70 ± 0.27_a	0.026 ± 0.014_a	0.014 ± 0.006_a	0.0010 ± 0.0003_a	11.6 ± 1.8_a	0.42 ± 0.14_a	0.73 ± 0.27_a
Mean total	48.7 ± 13.1_a	3.1 ± 1.8_a	0.5 ± 0.3_a	0.095 ± 0.03_a	463.2 ± 176.0_a	57.6 ± 18.6_a	74.9 ± 21.6_a
Flooded topsoils (n = 148)							
pH	−0.698**	−0.320**	−0.299**	−0.237*	−0.349**	−0.427**	−0.524**
OM	0.260*	0.228*	0.226*	0.113	0.266*	0.260*	0.302*
S	0.502**	0.124	−0.100	0.095	0.351**	0.471**	0.469**
Mean bioavailable	1.66 ± 1.87_b	0.030 ± 0.038_b	0.011 ± 0.006_b	0.0010 ± 0.0009_a	15.4 ± 19.1_b	0.45 ± 0.65_b	2.37 ± 2.42_b
Mean total	48.5 ± 14.6_a	3.3 ± 2.2_a	0.6 ± 0.3_a	0.1 ± 0.03_a	390.3 ± 183.8_b	58.5 ± 18.6_a	81.2 ± 35.2_a

Note. Mean concentrations expressed as mg kg^{-1}. Different letters indicate significant differences between topsoils.

*Correlation is significant at the 0.05 level.

**Correlation is significant at the 0.01 level.

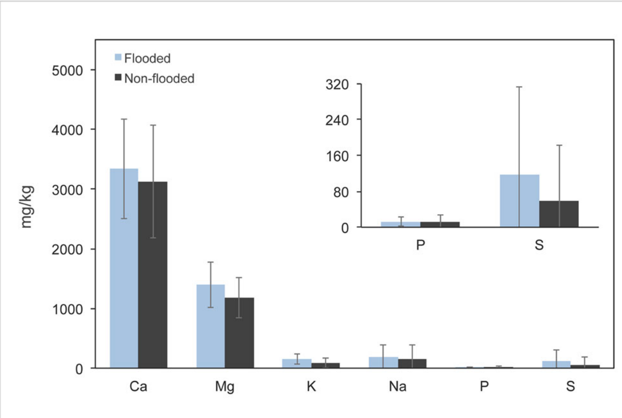

FIGURE 6 Concentration of sulphur, phosphorus, and bioavailable nutrients in flooded and nonflooded soils. Error bars are standard deviations of the average field sample [Colour figure can be viewed at wileyonlinelibrary.com]

The results of the principal component analysis are shown in Figure S2 and Table S2. Five principal components explain 69% of the data variation. Principal component 1 (PC1) accounted for 25% of variance. Ni and Fe presented the highest loadings (Table S2) in PC1 indicating the effect of anthropogenic activity in soil TM concentration and composition. Indeed, the ferronickel production in the area has been associated with high levels of Ni in wet and dry depositions (Marrugo-Negrete et al., 2017; Marrugo-Negrete, Urango-Cardenas, Burgos Núñez, & Díez, 2014). PC2 accounted for 13% of the variance. Positive loadings of PC2 (Cu, Zn, Pb, and Hg; Table S2) are associated to agricultural and mining activities that have long occurred in the area. In this sense, Cu and Pb are known markers of fertilizers and fungicides (Acosta, Faz, Martínez-Martínez, & Arocena, 2011; Besnard, Chenu, & Robert, 2001; Nziguheba & Smolders, 2008). PC1 and PC2 are therefore associated with anthropogenic activities: (a) agricultural practices such as fertilizer application (mainly Cu and Zn but also Pb) and (b) mining (ferronickel) activities (Ni and Fe), released from the mining of gold-bearing sulphide minerals (PbS). It is worth highlighting that Hg is part to this cluster, but it can be presumed that it is of a different anthropogenic origin. The presence of Hg in soils is ascribed to gold mining sites (Marrugo-Negrete et al., 2015; Pinedo-Hernández et al., 2015). PC3 and PC4 are both accounted for 11% of the variance (PC3 + PC4 = 22%) and pointed to OM, CEC, and pH as important drivers of the soil variability. PC5 (9% of the variance) was highly loaded with Cd (0.712) and P (0.720) indicating the effect of agriculture fertilizer–fungicide use in the studied soils. In this sense, owing to the highly significant correlation of Cd with P (Table S1), the main source of Cd can be attributed to the use of phosphate fertilizers applied in agricultural activities, as is well-documented elsewhere (Atafar et al., 2010). Given that phosphate rock is the primary stock material for manufacturing P-based fertilizers and may be contaminated with Cd, application of the fertilizers may contribute to Cd enrichment in the recipient soils (Jiao et al., 2012). Several studies have documented Cd accumulation in soils following the application of phosphate fertilizers (Atafar et al., 2010; Cai et al., 2012; Marin, Andrades, Iñigo, & Jimenez-Ballesta, 2016; Peris, Recatalá, Micó, Sánchez, & Sánchez, 2008).

3.5 | Flooding effects on soils

The bioavailability of TM and the ratio of S, P, and exchangeable bases (Ca, Mg, Na, and K) in flooded and nonflooded soils was evaluated by the RAC index (Figure 2). The concentration of bioavailable metals was significantly higher ($p < 0.05$) in flooded soils than in dry soils, except for Cd and Hg (Table 2). In a similar way, the percentage of bioavailable metals (Figure 5) is higher in the flooded than in dry soils, except for Hg. In fact, in the flooded soil, the mean percentage of bioavailability for each metal was 5.7% (Mn), 5.2% (Cu), 4.4% (Zn), 2.2% (Cd), 1.8% (Pb), 1.3% (Ni), and 0.6% (Hg). In nonflooded soils, the percentages were significantly lower ($p < .05$), ranging from 0.5% to 2.6%. The tailings produced during metal processing (Berkowitz, Dror, & Yaron, 2008; Trois, Marcello, Pretti, Trois, & Ross, 2007) are discharged into streams and spread over agricultural fields by the annual and regular flood pulse. Unfortunately, when a catastrophic flood event occurs, such as that of 2010 La Niña, contaminants are spread over a wider area than the usual floodable zone, because any dam is reducing the transport of sediment downstream (Teklemariam et al., 2017). Our results show that, according to the RAC (Sundaray et al., 2011), the environmental risk was low for all of the metals analysed in both flooded and nonflooded soils. However, it is important to highlight that the RAC was higher for flooded soils than for nonflooded soils.

Our results revealed no significant relationship between total TM and pH and/or OM (Table S1). In contrast, significant correlations existed between bioavailable fractions of TM and pH and OM, albeit only in flooded soils (Table 2). Our hypothesis is that as a result of soil inundation, along with the potential redox decrease and pH increase, the solubility of Fe and Mn oxides should increase via processes of reductive dissolution (Rennert, Meiβner, Rinklebe, & Totsche, 2010). Therefore, a release of the adsorbed TM occurs, which means that submerged soils increase in bioavailability.

On the other hand, the dynamics and bioavailability of nutrients depend on different processes that occur in the soil, which are accelerated during flood events. Several reports have shown the loss of nutrients by frequent flood phenomena, especially N and P (Eulenstein, Muller, & Helming, 1998; Loeb, Lamers, & Roelofs, 2008). Bioavailable nutrients (Figure 6) demonstrated a higher but not significant occurrence in flooded soils. Nutrients are easily transported in the Cauca and San Jorge rivers and reach the fields as a result of flooding (Marrugo-Negrete et al., 2015; Pinedo-Hernández et al., 2015). The increase in Ca and Mg can originate from carbonate dissolution following its mineralization, and the cation exchange of Fe^{2+}, Mn^{2+}, and NH_4^+ formed initially during the flood period (Loeb et al., 2008). The decrease of P could be related to the reduction of Fe and Mn oxides that may adsorb PO_4^{3-} (Loeb et al., 2008; Reddy, Kadlec, Flaig, & Gale, 1999). The increase in sulphur might be attributed to the reduction of SO_4^{2-} to sulphides in the soil profile as it becomes saturated with moisture (Loeb et al., 2008).

4 | CONCLUSIONS

Our study reports the chemical loading of major and TM **in** soils influenced by mining and agricultural practices, as well as their dynamics following the severe flooding in La Mojana floodplain caused by the 2010 La Niña phenomenon. TM sources for La Mojana soils were rather complex and included different anthropogenic activities. The presence of Zn, Pb, Cu, Ni, and Hg indicated mining activities, whereas the presence of Cd and P was associated to the use of phosphate fertilizers applied in agricultural activities. The indexes of contamination used in this study indicated that most of the soils were moderately contaminated, and few sites were highly contaminated. Our study is novel by showing that the flooding episode caused by the 2010 La Niña affected the available fraction of TM, which were significantly higher in flooded soils than in nonflooded soils of La Mojana, Colombian Caribbean. This is of special concern as anthropogenic activities in the study area in combination with extreme flooding events due to climate change may notably accelerate the risks of human exposure to metals. Our study highlights the relevance of assessing the TM available fraction as an index of human-risk exposure and ecosystem vulnerability to climatic changes.

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ORCID

José Marrugo-Negrete https://orcid.org/0000-0002-3181-7529
José Pinedo-Hernández https://orcid.org/0000-0002-4288-1788
Enrique M. Combatt https://orcid.org/0000-0002-8760-0089
Andrea G. Bravo https://orcid.org/0000-0002-8341-3462
Sergi Díez https://orcid.org/0000-0002-9870-2179

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