Assessment of the impact of small-scale mining on soil contamination by mercury and hydrocarbons in the kadey catchment (East Cameroon)

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ABSTRACT

The objective of the present study was to assess soil contamination by mercury and hydrocarbon products used in gold mining in the Kadey catchment area. The results obtained show that gold mining by the small mine is the cause of chemical contamination of the soil caused by hydrocarbon products with concentrations 800 times higher than the threshold value on certain points of the former mining sites. The use of mercury has led to an increase in its concentration to values that are still below the critical thresholds. It also appears from this work that organic matter plays a very important role in the retention of mercury on the soil surface. In the case of hydrocarbons are concerned, although high concentrations above are found exclusively at the surface, the rainfall contributes to their infiltration into the soil and to a horizontal redistribution of the contamination.

1. Introduction

Small-scale gold mining first appeared on the international development agenda about four decades ago [1]. It has gained considerable importance in recent years, particularly in developing countries where the lack of alternative sources of income pushes poor and poorly educated people to engage in small-scale or artisanal mining [2,3]. These activities, which take place in rural areas that are difficult to access in many parts of the world, are at least as important as large-scale mining activities, particularly in terms of the number of people employed [4]. An estimated 10–15 million people are employed in this sector [5,6], mainly in Asia, Africa and South America, and a further 85–90 million people are indirectly dependent on this activity [5]. The importance of artisanal and small-scale gold mining is also due to the fact that it accounts for 20–30% of global gold production [5]. This activity is widespread in many countries such as the Democratic Republic of Congo, China, Ghana, Tanzania, India, Ethiopia, Brazil, Pakistan, Burkina Faso, Colombia, Bolivia, … where estimates show, despite the absence of official data, that the number of people involved can be as high as 2,910,000, 2,746,000, 1,000,000, 994,000, 915,000, 728,000, 861,000, 515,000, 400,000, 268,000, 130,000 respectively [1, 7–9].

In Cameroon, small-scale gold mining has been practised since 1934. It flourished during the Second World War, but problems linked to the increase in production costs from 1946 onwards, labour difficulties and the depletion of reserves seriously affected this
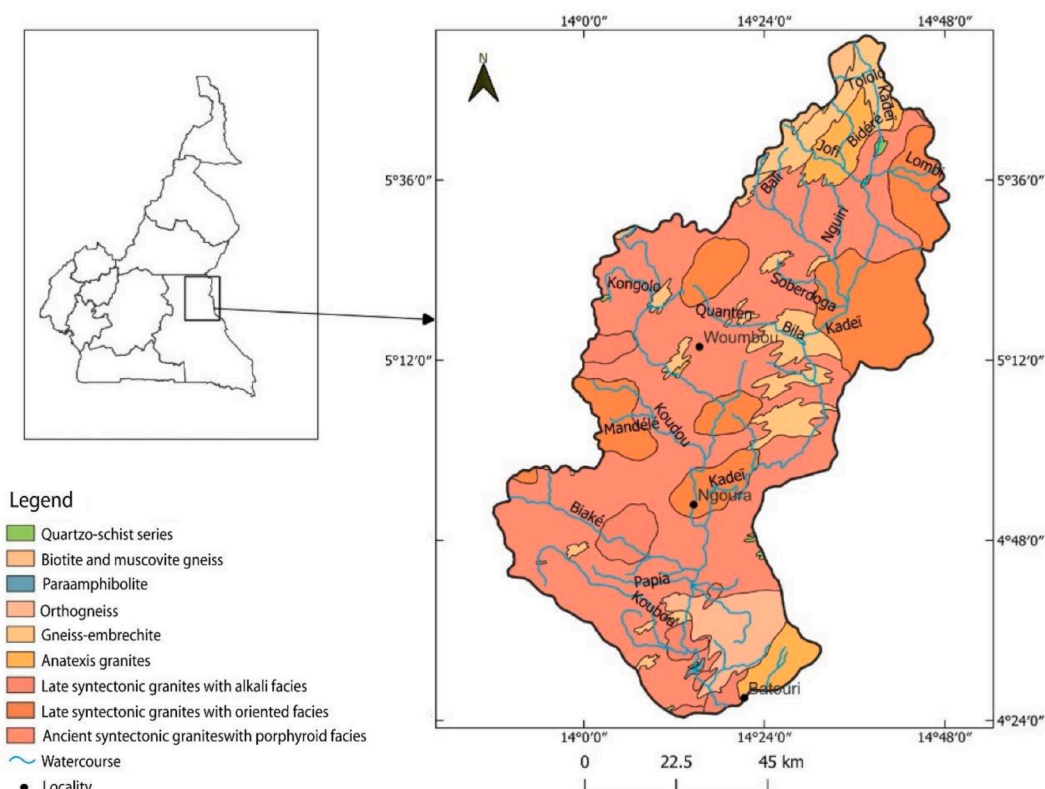
Fig. 1. Geological map of the study area (Source [33], modified).

activity, which ceased to be practised in the early years of independence [10]. The sector will continue to experience intense activity, particularly in the gold districts of the eastern region, following the sharp rise in the price of gold from 2010. This important activity, marked by the extraction of ore from alluvial and eluvial deposits and quartz veins, will entail the use of excavators, a semi-mechanised washing system and chemicals. However, as in most African countries, in the south of the Sahara, this activity has not resulted in increased mining revenues for the state or improved living conditions for local populations, particularly in Mali [11], Ghana [12,13], or Southern Africa [11]. Indeed, due to numerous dysfunctions linked to the lack of material and financial resources of the structures responsible for monitoring mining, this activity remains above all marked, as in other countries, by clandestinity, non-compliance with the regulations in force [3, 4, 12, 14], the destruction of the landscape [1] and the denaturation of river beds. However, it appears from the activities that are carried out that the crucial problem caused by mining is related to mercury and hydrocarbon contamination. The use of mercury for gold amalgamation in artisanal and small-scale mining is a critical component in the mining process [15] and the fastest emerging source of mercury contamination in the world [16,17]. According to Ref. [18], between 2010 and 2015, this activity accounted for almost 38% of mercury emissions worldwide and was the main source of emissions in South America and Sub-Saharan Africa, with 70% and 80% of emissions respectively. Artisanal gold mining is also currently considered to be responsible for more than half of global mercury emissions [19]. Mercury-contaminated terrestrial environments pose a risk to global public health, with mercury considered one of the top ten chemicals of concern [20]. The release of mercury into the environment represents a public health problem [21] that can induce serious effects on organisms depending on the time of exposure, its concentration and speciation [22]. It directly contaminates the air, soil and water, before finally being deposited in sediments where it is transformed by bacterial action into methylmercury, an organic compound of high bioavailability that can easily enter the food chain [11] after absorption by plankton [23]. Predatory species and populations are therefore exposed to high levels of concentrations in the diet through the bio-accumulation process [24]. The first to be affected are gold miners who handle and inhale mercury vapours all day long [25]. Hydrocarbon contamination at small-scale mining sites, although not yet well documented, is likely to cause serious environmental and health problems for the population [26,27] through uncontrolled use and uncontrolled dumping at most mining sites in the Kadey catchment area, as in all the major gold mining areas of Cameroon.

The assessment of the environmental legacy related to the use of mercury and hydrocarbons in small-scale gold mining is therefore still an issue of great concern in Cameroon today. The work undertaken has focused on the evaluation of the physico-chemical quality of water [28], the evaluation of the degree of contamination of water, fish or mine tailings [29,30] as well as on the impacts of gold mining on the population [31,32]. The present study specifically aims to make a quantitative assessment of soil contamination by mercury and hydrocarbons in the former gold mining sites of the small-scale mine in the upstream part of the Kadey catchment.
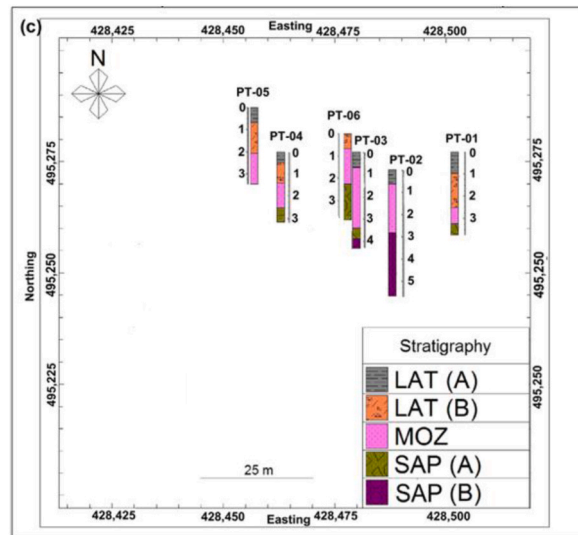
Fig. 2. Lithological description of soils [35].

2. Location of the study area and presentation of the physical environment

The study area is located in the upstream part of the Kadey catchment area, on two sites of the former base-lands of the mining operations in the locality of Woumbou (Fig. 1). Geologically, the area is part of the central part of the Pan-African Chain of Central Africa in Cameroon. The main geological formations encountered are represented by gneisses, orthogneisses, paramphibolites, syn-tectonic granites, shales and quartzites [33].

From a pedological point of view, the Kadey watershed shows a latitudinal evolution of the soils. From the South towards North, we find successively red ferrallitic soils with deep horizons locally cuirassed and abundant ferruginous concretions, ferrallitic cuirassed soils, and finally complex savannah soils, such as reworked soils with concretions and cuirass debris [34]. [35] described the litho-logical horizons in the Batouri gold district and identified five layers (Fig. 2) which are distinguished from top to bottom as follows.

- coarse nodular laterite: it is 0.5–0.99 m thick, brownish, gravelly and contains nodules varying in size from 0.5 to 5.5 cm;
- fine nodular laterite: it varies in thickness from 0.4 to 1.56 m. Its colour varies from brown to reddish brown. It is sandy to gravelly and contains nodules varying in size from 0.5 to 3.5 cm;
- the mottled zone: this is between 0.69 and 2.73 m thick. It is a silty, clayey to sandy horizon whose colour varies from yellow to reddish brown. It is gradually transformed into saprolite;
- Saprolite: varies in thickness from 0.48 to 1.66 m. This horizon is clayey with a greyish to yellowish brown colour. The texture and structure of the parent rock are partially preserved although the material is deeply weathered and friable;
- Saprock: this is the weathered bedrock with a well-preserved texture and structure. Its thickness varies between 0.43 and 1.17 m. The saprock is generally more coherent than the saprolite horizon.

Climatically, the region is influenced by subtropical and equatorial climates with two marked maxima and minima of rainfall. This results in the existence of two rainy seasons and two dry seasons. The long rainy season extends from August to mid-November and the short rainy season from March to June. The long dry season runs from November to March. The short dry season runs from June to July with sporadic showers. The average annual rainfall at the Batouri station is 1479 mm. The highest temperatures are observed during the long dry season with a maximum average of 32.1 °C in February at Batouri. The lowest temperatures are observed in January with a minimum average of 17.1 °C.

Hydrographically, the Kadey has a catchment area of 41,000 km^2 of which more than half is within Cameroon. It has its source near Garoua-Boulaï at an altitude of 1050 m and is 500 km long. It forms a natural boundary between Cameroon and the Central African Republic for about 80 km and then describes a wide loop passing a few hundred metres west of Batouri. It flows back towards the Central African Republic, crosses the border and joins the Mambéré at Nola. Its profile is irregular and its course marked by fairly large waterfalls. Its main right bank tributaries in the Cameroonian part are Bali, Bila, Oudou, Koubou and Doumé. The left bank tributaries are Boumana, Mama and Boumbé II [34].

3. Materials and methods

A field trip was conducted in December 2016 in the locality of Woumbou with the aim of taking soil samples from two former mining basements. The choice of the former base-roads is justified by the fact that it is within them that the final concentration and
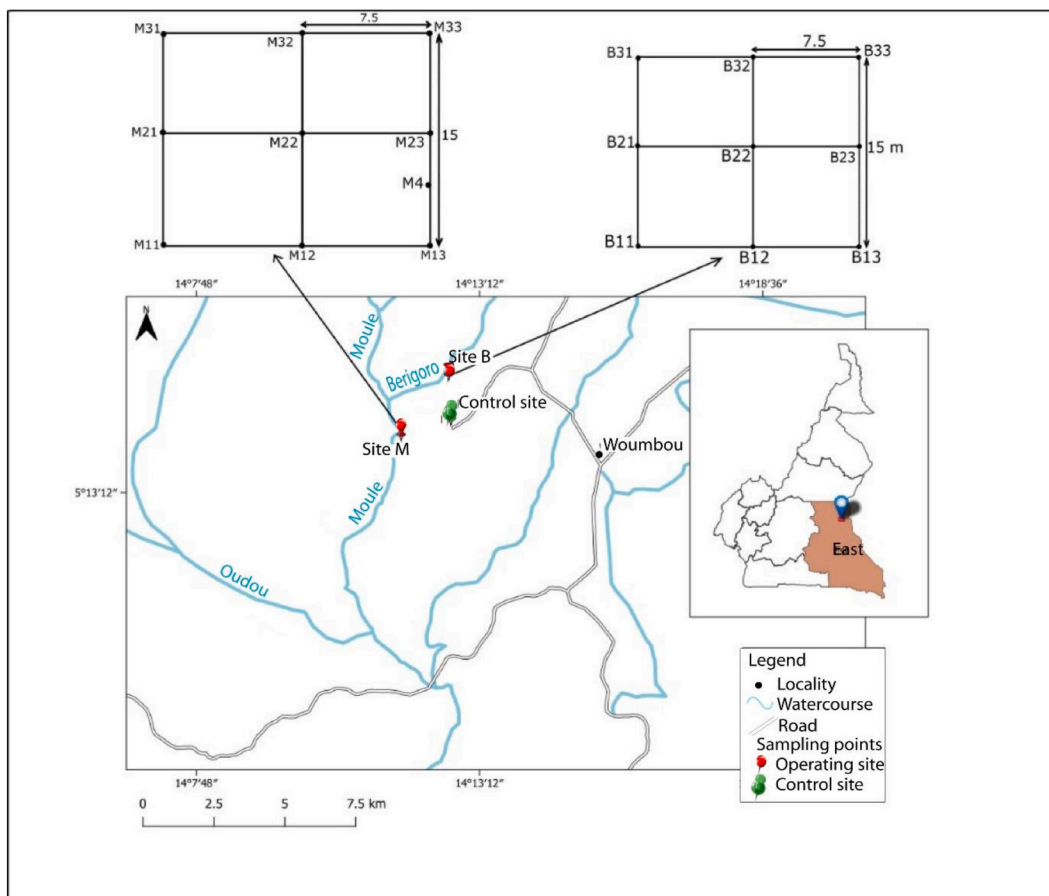
Fig. 3. Location map of sampling sites and sampling plan of sites B and M.

amalgamation processes of the gold take place and they also constitute the storage place for petroleum products intended to fuel the machines and equipment used in the exploitation. The first and second sites are located on the left banks of the Berigoro (Site B) and Moule (Site M) streams respectively (Fig. 3). Control samples were taken from a site outside the mining sites. Sites B and M are on the edge of the rivers and at a lower elevation than the control site. Site B is bare after logging and is devoid of vegetation, while site M is used for agriculture. The control site is covered by grass and free of human activity. Soil sampling at sites B and M was carried out according to the recommendations of the Cooperative Research Network on Waste and the Environment [36]. Sampling at both sites was done on the basis of a systematic square grid. In order to have a representative area of the base camp site, a 15 m square was made at each site and three transepts were drawn 7.5 m apart. On each transept three sampling points were determined: one at each end and one in the middle. A total of 9 sampling points was thus defined at each site. However, at site M a tenth point (M4) was sampled in order to analyse an area with a particularly blackish coloration. The sampling plans for both sites are shown in Fig. 3. Three samples were taken at each point, the first at the soil surface, the second at a depth of 0.5 m, and the third at a depth of 1 m. At the control site, sampling was carried out at one point as recommended by Ref. [37] for the assessment of total hydrocarbon soil contamination in Nigeria. A total of 27 samples were obtained from site B, 30 from site M and 3 from the control site (I). The samples were stored in High Density Polyethylene (HDPE) bags. Each sample was labelled with its position on the site and its depth (S, M and P standing for surface, mid-depth and depth respectively) and then placed in the cooler for transport to the laboratory. In the laboratory, the samples were dried and crushed, then digested for extraction and determination of total mercury and total hydrocarbons. Mercury and total hydrocarbon concentrations were determined at the PAC-LAB laboratory in Limbé, Cameroon. The total mercury content in the soil was measured by the graphite furnace atomic absorption spectrometry method with a detection limit of 1 ng/g. The content of total hydrocarbons was determined by the infrared spectrophotometry method with a detection limit of 100 µg/kg. The organic matter content in the soil solution was obtained by measuring the amount of total organic carbon by the colorimetric method according to ASTM D 4839. As organic matter contains 58% organic carbon, the organic matter content was calculated by multiplying the organic carbon content by a factor of 1.72 [38,39].

Table 1
Mercury and total hydrocarbon concentrations at site B, M and the control site.

| Sample	Hg (mg/kg)	HT (mg/kg)	Organic matter (in %)	Sample	Hg (mg/kg)	HT (mg/kg)	Organic matter (in %)
Surface samples							
B11S	0.050	11.7	13.55	M11S	0.040	2.6	1.51
B12S	0.050	7.4	6.82	M12S	0.050	4.4	2.32
B13S	0.030	3901.5	1.59	M13S	0.040	8130.1	9.83
B21S	0.025	11.5	2.85	M21S	0.040	3.2	7.25
B22S	0.050	6.6	7.70	M22S	0.050	37.4	11.66
B23S	0.050	7.3	1.66	M23S	0.050	1.7	7.86
B31S	0.040	5004.2	6.41	M31S	0.030	1.4	7.17
B32S	0.015	2.8	7.63	M32S	0.040	1032.7	6.27
B33S	0.050	11.5	5.69	M33S	0.050	744.2	12.48
/	/	/	/	M4S	0.013	3887.5	7.87
Mid-depth samples							
B11 M	0.015	8.5	2.51	M11 M	0.014	7.7	9.39
B12 M	0.016	13	8.13	M12 M	0.024	10.2	2.09
B13 M	0.016	7.3	7.99	M13 M	0.013	4.7	7.91
B21 M	0.014	11.7	1.11	M21 M	0.014	5.1	2.63
B22 M	0.014	7.4	7.25	M22 M	0.015	2.7	6.82
B23 M	0.011	6.8	6.27	M23 M	0.012	2.2	7.63
B31 M	0.011	3.3	7.75	M31 M	0.015	3.6	3.62
B32 M	0.015	4.3	5.03	M32 M	0.014	8.1	3.56
B33 M	0.010	1.9	5.17	M33 M	0.013	5.6	3.40
/	/	/	/	M4M	0.012	7.7	7.46
Depth samples							
B11P	0.016	6.6	7.25	M11P	0.025	3.5	1.10
B12P	0.015	9.9	2.92	M12P	0.014	6.8	4.69
B13P	0.011	8.1	4.79	M13P	0.015	2.7	3.85
B21P	0.016	8.1	7.34	M21P	0.015	3.6	2.49
B22P	0.016	13	7.17	M22P	0.012	3	2.49
B23P	0.014	11.5	6.29	M23P	0.015	3.9	3.13
B31P	0.011	6.6	8.01	M31P	0.014	3.4	2.51
B32P	0.012	5.3	5.74	M32P	0.012	3.9	1.75
B33P	0.010	3.3	4.07	M33P	0.014	2	4.00
/	/	/	/	M4P	0.023	3.4	1.13
Control samples							
IS	0.015	1.2	6.27	IS	0.015	1.2	6.27
IM	0.024	1.7	1.29	IM	0.024	1.7	1.29
IP	0.014	1.2	4.76	IP	0.014	1.2	4.76
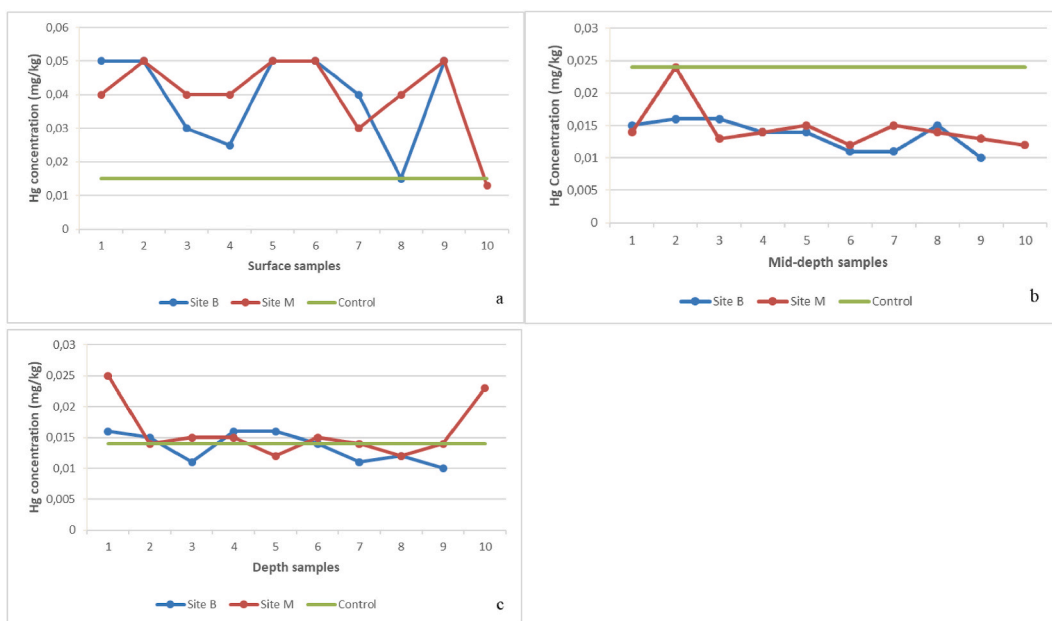
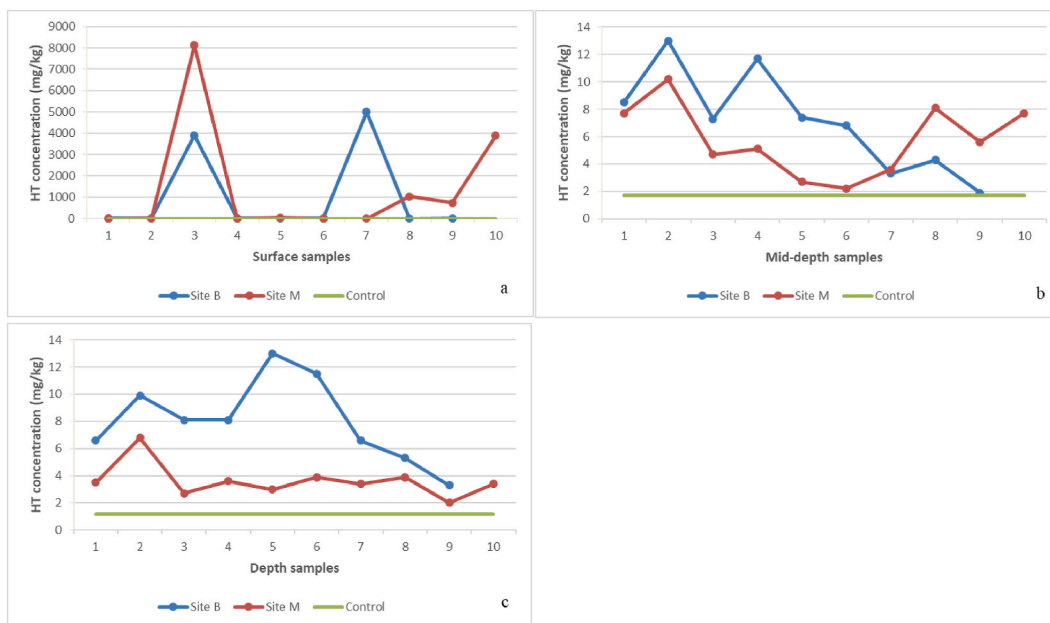**Fig. 4.** Mercury concentration: a) surface; b) mid-depth; c) depth.

Fig. 5. Concentration of total hydrocarbons: a) surface; b) mid-depth; c) depth.

4. Results

4.1. Mercury concentration in soil

Depending on the source and form of release, mercury in soil may be present concentrated in hot spots or dispersed over large areas [40]. The results of the analyses carried out on the samples taken at sites B, M and control are presented in Table 1.

4.1.1. Mercury concentration in surface samples

At site B, with the exception of sample B32S, surface concentrations range from 0.025 mg/kg to 0.05 mg/kg and 6 out of 9 samples have concentrations ranging from 0.04 mg/kg to 0.05 mg/kg (Fig. 4a). At site M, surface concentrations ranged from 0.013 mg/kg (M4S) to 0.05 mg/kg and 8 out of 10 samples had concentrations ranging from 0.04 mg/kg to 0.05 mg/kg (Fig. 4a). At the control site, the IS surface sample had a mercury concentration of 0.015 mg/kg. These results show that the surface mercury concentrations recorded at sites B and M are higher than those recorded at the control site.

4.1.2. Mercury concentration in mid-depth samples

The concentrations recorded on the mid-depth samples, vary at site B from 0.10 to 0.16 mg/kg (Fig. 4b). At site M, the concentration of 0.24 mg/kg was observed on sample M12 M while at the other samples, the mid-depth concentrations varied between 0.12 and 0.14 mg/kg (Fig. 4b). At the control site, the mid-depth sample (IM) has a concentration of 0.24 mg/kg. These results show that the mid-depth concentrations recorded at both sites are, with the exception of sample M12 M, lower than that recorded at the control site.

4.1.3. Mercury concentration in depth samples

The concentrations recorded on the depth samples vary at site B between 0.10 and 0.16 mg/kg (Fig. 4c). At site M, values of 0.25 and 0.23 mg/kg were observed on samples M11P and M4P, respectively, while concentrations on the other samples varied between 0.012 and 0.015 mg/kg (Fig. 4c). The sample taken at depth at the control site has a concentration of 0.014 mg/kg. These results indicate that the concentrations recorded at depth at both sites are the same as those observed at depth at the control site.

4.2. Concentration of hydrocarbons in soil

The analytical results for total hydrocarbon concentration at sites B, M and control are presented in Table 1.

4.2.1. Total hydrocarbon concentration in surface samples

The recorded results show that at site B, samples B31S and B13S with concentrations of 5004.2 mg/kg and 3901.5 mg/kg respectively have the highest total hydrocarbon loadings at the surface (Fig. 5a). At the other surface samples, concentrations range from 2.8 mg/kg (B32S) to 11.5 mg/kg (B11S). At site M, the highest total hydrocarbon concentrations are observed in samples M13S, M4S, M32S and M33S with values of 8130.1 mg/kg, 3887.2 mg/kg, 1032.7 mg/kg and 744.2 mg/kg respectively (Fig. 5a). The surface
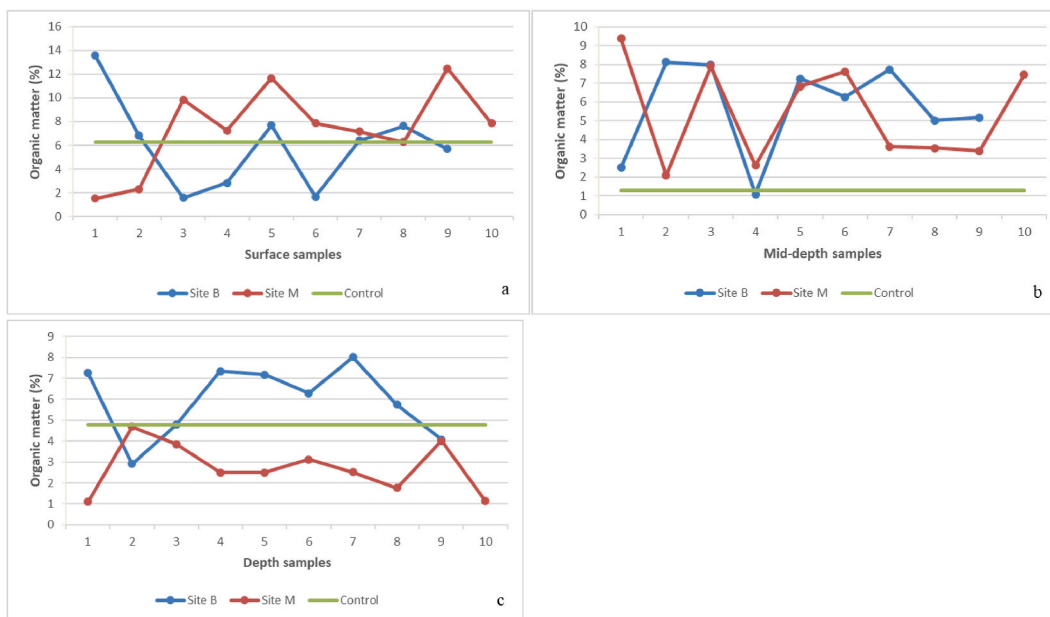
Fig. 6. Organic matter content: a) surface; b) mid-depth; c) depth.

samples that do not have high total hydrocarbon loadings at this site have concentrations that range from 1.4 mg/kg (M31S) to 37.4 mg/kg (M22S). At the control site, the recorded surface concentration (IS) is 1.2 mg/kg. It can be seen from these results that the total hydrocarbon concentrations at the control site are significantly lower than the concentrations of the samples with the highest loadings at site B and site M.

4.2.2. Total hydrocarbon concentration in mid-depth samples

At site B, mid-depth samples show concentrations ranging from 1.9 mg/kg (B33 M) to 13 mg/kg (B12 M). At site M, mid-depth concentrations range from 2.2 mg/kg (M23 M) to 10.2 mg/kg (M12 M) (Fig. 5b). The sample from the control site has a concentration of 1.7 mg/kg. This concentration is lower than those observed at sites B and M (Fig. 5b).

4.2.3. Total hydrocarbon concentration in depth samples

At depth, total hydrocarbon concentrations at site B range from 3.3 mg/kg (B33P) to 13 mg/kg (B22P). At site M, the concentrations recorded in the samples taken at depth range from 2 mg/kg (M33P) to 6.8 mg/kg (M12P) (Fig. 5c). At the control site, the concentration at depth is 1.2 mg/kg. It can also be seen that at depth, the concentrations recorded at sites B and M are higher than at the control site (Fig. 5c).

4.3. Variation of organic matter content

4.3.1. Surface organic matter content

The results obtained show that surface organic matter levels vary between 0.65% (B21 M) and 13.55% (B11S) at site B and between 1.51% (M11S) and 12.48% (M33S) at site M (Fig. 6a). In general, it can be noted that site B, which is devoid of vegetation, has low organic matter values compared to those recorded at site M where agricultural activities are carried out. At the control site, the surface organic matter content is 6.27%.

4.3.2. Mid-depth organic matter content

In the mid-depth samples, the organic matter content varies between 1.1% (B21 M) and 8.13% (B12 M) at site B and between 2.09% (M12 M) and 9.39% (M11 M) at site M (Fig. 6b). At the control site, the concentration recorded is 1.29%.

4.3.3. Organic matter content at depth

In the depth samples, the organic matter contents vary at site B between 2.92% (B12P) and 8.01% (B31P) and at site M between 1.1% (M11P) and 4.00 (M33P). At the control site, the organic matter content recorded is 4.7% (Fig. 6c).

5. Discussion

5.1. Assessment of mercury-related soil contamination

The recorded results show that at sites B and M, 15 out of 19 surface samples (78.9%) have concentrations that vary between 0.04 and 0.05 mg/kg. As a result, the surface concentrations recorded at most of the sampling points at both sites are higher than those recorded at the control point. The concentrations recorded at mid-depth at both sites are lower than those recorded at the control site. On the other hand, the concentrations recorded at depth at both site B and site M are the same as those recorded at the control site. This study shows that in the abandoned mining sites of Woumbou, the highest concentrations of mercury in the soil are found at the surface. Mercury is naturally present in soils from geological sources [41] or as result of natural events such as forest fires and volcanic eruptions [42]. Assessing mercury emissions poses serious methodological problems [43], and it is particularly difficult to distinguish between natural and anthropogenic emissions [44,45]. Nevertheless, it has been shown that the high levels of trace metals found at the surface of the soil, in contrast to the much lower concentrations immediately below, are an indication of significant inputs from human activities ([46]). From the results recorded, it can be said that:

- the concentrations between 0.013 and 0.015 mg/kg identified at the soil surface at the control point and some points at sites B and M reflect the concentration of mercury at the soil surface under natural conditions;
- The use of mercury has resulted in an increase in the concentration of mercury on the soil surface to concentrations that average 0.040 kg at sites B and M;
- mercury concentrations in deep horizons result solely from natural conditions and range from 0.010 to 0.025 mg/kg in depths of 0.5–1 m.

This study showed that soil contamination at the base-river sites remained localised on the surface horizons. The results obtained show that the mercury concentrations observed at the Woumbou mining sites are in the same order of magnitude as those recorded by Ref. [47] on the soils of the Bétaré-Oya mining sites (an important gold mining district located approximately 40 km north-east of Woumbou), where the concentrations measured vary between 0.015 and 0.057 mg/kg. However, it should be noted that mercury occurs naturally in soils at concentrations ranging from 0.03 to 0.15 mg/kg ([48]) with a mean value of 0.06 mg/kg [43]. According to Ref. [49] average mercury levels in soils vary from 0.01 to 1.5 mg/kg, depending on soil properties and proximity to an emission site. Mercury-contaminated sites often have soil concentrations that are 2–4 times higher than the geochemical background of mercury in the soil [50]. [51] defined a narrower range of 0.05–0.3, although some volcanic and organic soils, particularly in Canada, may contain higher concentrations, and near sources of industrial emissions, concentrations can be extremely high. The concentrations varying between 0.04 and 0.05 mg/kg observed on most of the samples taken from the surface of the soil of the former base camps, and which may be the result of contamination linked to the use of mercury in gold recovery, are therefore within the range of natural concentrations of mercury in the soil. The fact that the concentration of 0.015 mg/kg in the surface sample at the control site is lower than the concentrations of 0.04 and 0.05 mg/kg recorded in most of the samples from the former gold mining sites suggests that the soil concentration has increased as a result of gold mining activities at the former gold mining sites. This idea is further supported by the fact that concentrations recorded at the surface of these base-valleys are higher than those recorded at depth, whereas at the control site, the surface concentration (0.015 mg/kg) is lower than that recorded at mid-depth (0.024 mg/kg). In fact, naturally, the concentrations at depth are higher than the surface concentrations [46]. With regard to the probable contamination of the control site samples by mercury emitted at the mining sites, work by Ref. [52] in forested areas of the Amazon region shows that gaseous mercury (Hg°) emitted from mining sites into the atmosphere is deposited within 10–20 km of the source. Therefore, deposition of mercury, either gaseous (Hg°) or Hg(II) (resulting from the oxidation of gaseous mercury), from mining operations may influence soil mercury concentrations at the control site located about 2.5 km from the nearest mining sites. However, the recorded results, which indicate that the concentration observed at the surface of the control site is the same as those observed at the points with low surface concentrations at sites B and M, do not indicate contamination of the control samples from the mining sites.

In France, where the mercury concentration in unpolluted soils and outside anomaly zones varies between 0.02 and 0.1 mg/kg, work carried out on the soils of industrial sites also shows that mercury concentrations at the surface are significantly higher than those recorded at depth [53]. [53] also show that although it is quite difficult to predict the position of the mercury-polluted layer in the soil profile, deposits related to anthropogenic activities are generally found in the first 50 cm with the highest surface concentrations. The LUCAS study of more than 23,000 samples of arable soil (top 20 cm) in all European Union countries except Croatia shows an average of 0.04 mg/kg of mercury in a range from 0 to 159 mg/kg [54]. [55] results on soil profiles from gold processing sites in Tanzania indicate high mercury levels (2,495 mg/kg) in the upper 10 cm. This leads to the understanding that the mercury concentration values recorded at the soil surface of the sites studied are due to contamination related to gold mining.

Even so, the concentrations recorded are low and below the guideline values set by a number of industrialized countries, including, Australia (1 mg/kg), Canada (6,6 mg/kg) and United States of America (2,3 mg/kg) [56]. These soils would therefore not pose a health problem for people living on these sites or using them for agricultural purposes. However, the fact that mercury is observed at low concentrations in soil does not exclude that high concentrations can be detected in other environmental compartments. Soil erosion can result in massive amounts of mercury being associated with soil particles and transported to lowlands and rivers, where biogeochemical conditions favour methylation [57,58].

The low levels of mercury contamination recorded can, however, be explained by the fact that in Woumbou, mercury is only used at the end of the gold recovery process. As the ore is washed using the gravimetric method, the quantities of mercury used are lower and
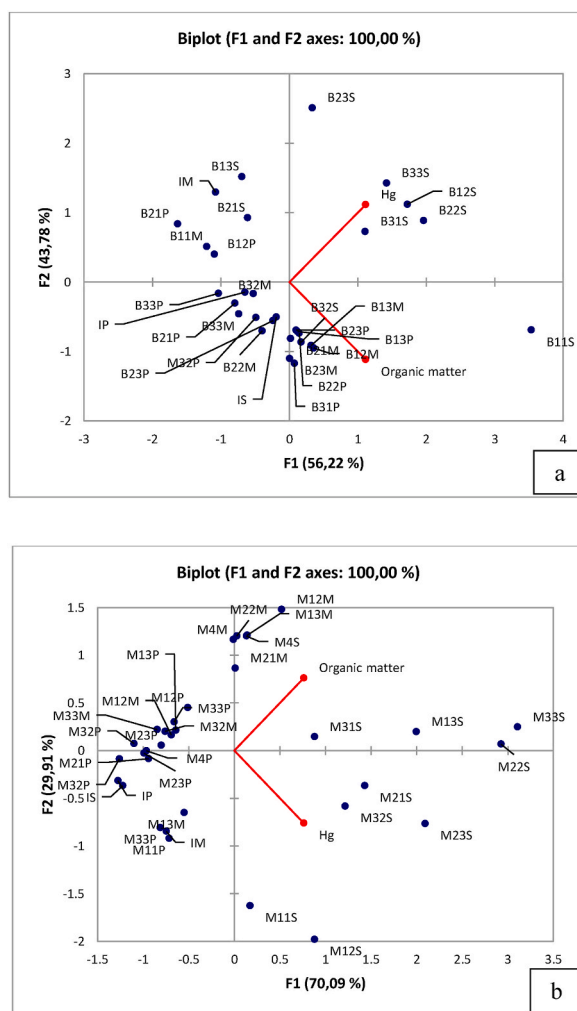
Fig. 7. Representation of the relationship: a) between organic matter and mercury on the F1 and F2 correlation diagram at site B; b) between organic matter and mercury on the F1 and F2 correlation diagram at site M.

therefore the quantities released into the environment are small. The gravimetric concentration process is indeed a practice that contributes to significant reductions in mercury consumption and emissions and only about 14 g of mercury are needed to amalgamate 1 kg of concentrate, a mercury/concentrate ratio of 1/70 [2]. In addition, the separation of gold from mercury is often done by heating the amalgam over a fire. As a result, the mercury used to recover the gold will largely evaporate [55]. in Tanzania and Zimbabwe has shown that 70–80% of the mercury released to the environment is lost to the atmosphere by heating amalgam at the mine site and in goldsmith shops in towns and 20–30% is lost to tailings, soil and water. The low levels of soil contamination at the sites studied cannot, however, be generalised to the whole Kadey catchment area and cannot be considered as definitively representative of this area. Indeed, since the year 2020, there has been a shift in mining methods to extract and recover all the oxidised part on the surface and the deeper primary formations and to use large quantities of mercury in the initial processing phase.

5.2. Assessment of hydrocarbon-related soil contamination

Petroleum hydrocarbons include lubricating oils, petrol and diesel. They are released into the ground as a result of spills during loading into machinery or following engine maintenance work, and pollute it [59,60]. Although they do not indicate the existence of a direct danger to the environment or humans, the presence of petroleum hydrocarbons in the soil helps to indicate the state of health of the soil and is an important tool in monitoring the nature of contaminants in the soil [61,62]. The analytical results show that 2 out of 9 surface samples (B31S and B13S), i.e. a percentage of 22%, at site B and 4 out of 10 samples (M13S, M4S, M32S and M33S), i.e. a percentage of 40%, at site M (Fig. 6), have total hydrocarbon concentrations well above the threshold value of 100 mg/kg set by the Dutch standard. These results show that the highest concentrations are found at the soil surface. The study by Ref. [60] on three sites in Nigeria and other studies [63,64] also show that total hydrocarbon concentrations in soils decrease with increasing depth [65]. showed

that the highest level of hydrocarbon concentration in oil exploitation areas in Indonesia is found at a depth of 0–30 cm [66]. suggest that the decrease of hydrocarbons with depth may be inherent to low soil permeability. On the other hand, the work of [67] on the vertical variation of hydrocarbon concentrations in soil profiles shows that the total hydrocarbon content has a systematic tendency to increase with depth. The observation of concentrations above the tolerable threshold on localised areas and exclusively on the soil surface shows that the particles of the clay-humus complex of the soil participate in the retention of the hydrocarbons. However, it is noted that the sub-threshold concentrations (of the Dutch standard) on the surface of sites B and M are generally higher than those recorded on the control samples. In the mid-depth and deep samples, it is also noted that the concentrations recorded at sites B and M are higher than those at the control site (Fig. 5b and c). This shows that, although the hydrocarbons have been fixed by the organic matter, the rainfall that occurs during the rainy season causes infiltration at depth and a spatial redistribution of some of the hydrocarbons from the contamination sources. This can lead to contamination of rivers and groundwater [68]. [67] have also shown that the high tendency for hydrocarbons to leach and migrate deep into groundwater is a potential hazard to groundwater.

In the United States, the Environmental Protection Agency (USEPA) classifies total hydrocarbon pollution levels into categories A, B and C with the following respective concentrations: greater than 40,000; 10,000 to 40,000; and 1,000–10,000 g/kg [69]. Total hydrocarbon pollution levels in Indonesia are also subdivided into three classes A (40,000 µg/g), B (5000 g/kg), C (1000 g/kg) [70]. In several European countries, such as the Netherlands and Spain, soils at concentrations of 5000 g/kg are considered severely contaminated [71,72]. Taking these classifications into account, samples with high hydrocarbon loads are highly contaminated and fall into contamination levels B and C of the Indonesian regulations and category C of the US standard. These high hydrocarbon concentrations create unfavourable conditions by reducing gas diffusion and increasing the number of anaerobic organisms, which contributes to reducing the oxygen available due to the anoxic conditions created by hydrocarbon films on the surface and subsurface of soils [73].

5.3. Influence of organic matter on the behaviour of mercury and hydrocarbons

5.3.1. Influence of organic matter on the behaviour of mercury

Soil plays an important role in the biogeochemical circulation of mercury. It accumulates it and is a source for other components of the environment [43]. The total amount of mercury accumulated in soils in terrestrial environments is estimated at 200,000 to 300, 000 T [74,75]. Mercury is strongly bound to soil organic matter [49]. In particular, it appears to be associated with organic matter in certain soils in tropical regions [76,77] and temperate regions [78], and its concentration in soils is a function of the rate of deposition and the carbon turnover time [43].

The evolution of mercury concentration with depth is comparable to the variation of organic matter with depth, especially at site M, where 8 out of 10 sampling points (M13, M21, M23, M22, M31, M32, M33, M4) show a decrease in organic matter with depth. On the other hand, at site B, no decrease in organic matter was observed with depth. Analysis of the correlations shows that at site B, where the correlation coefficient R^2 is 0.12, there is no correlation between organic matter and mercury. At site M, the correlation coefficient (R^2 = 0.4) shows that there is a weak correlation between mercury and organic matter. The representation of the samples in the biplot correlation diagram, defined by the F1 and F2 axes, shows, however, that at site B (Fig. 7a), mercury and organic matter are found on the positive side of the F1 axis, which represents 56% of the information. This axis reflects the influence of gold mining on the correlation between mercury and organic matter. We note that samples B11S, B12S, B22S, B31S and B33S, which are on the positive side of the F1 axis, have high mercury values and average organic matter values. These observations show that at site B, the correlation between organic matter and mercury can only be established on certain surface samples (B11S, B12S, B22S, B31S and B33S). The correlation between the two substances is not observed on the deep and mid-depth samples at this site. At site M, mercury and organic matter are also found on the positive side of the F1 axis, which represents 70% of the total information and also reflects the influence of gold mining on the correlation between mercury and organic matter. Surface samples M13S, M21S, M22S, M23S, M31S, M32S which are close to the mercury and organic matter poles have high mercury and organic matter values (Fig. 7b). This shows that there is a correlation between the two substances in the surface samples. The majority of the mid-depth and deep samples from this site are on the negative side of the F1 axis and no correlation exists between the two substances on these samples. The representation of the samples in the biplot correlation diagram, defined by the F1 and F2 axes, also shows that no correlation between organic matter and mercury can be defined on the control site samples (IS, IM and IP) (Fig. 7a and b). Other studies have also shown that the distribution of mercury in soil is correlated with organic matter [79–82]. [83] reported positive correlations between organic matter and mercury content in humus layers in eastern and northern Norway with respective correlation coefficients R^2 of 0.58 and 0.55 [84]. showed regressions between soil organic carbon content and mercury content with an R^2 coefficient of up to 47% at 14 forestry sites in the United States.

The results obtained allow us to understand that organic matter participates in the retention of mercury at the soil surface, particularly at site M. This phenomenon is also valid for site B, although the organic matter values on the surface samples are comparable to those observed at mid-depth and at depth. Indeed, at site B, the soil having been exposed, erosive movements have transported a quantity of organic matter and explain why a decrease in organic matter with depth is not observed at this site. The fact that mercury is observed in higher quantities at the surface is justified by the fact that mercury binds very strongly to organic matter and follows its variation. Thus, since organic matter is mainly concentrated at the surface of the soil, the quantity of mercury decreases with depth. Studies have shown that the distribution of mercury in soils follows that of organic matter, with the highest concentrations found in layers close to the surface and decreasing with depth [85–88]. It has been shown that adsorption, which is the dominant process that determines the fate of mercury in soil [89], is increased by the presence of organic matter due to the complexation of mercury with humic and fulvic acids, and is therefore more important in surface horizons [90].
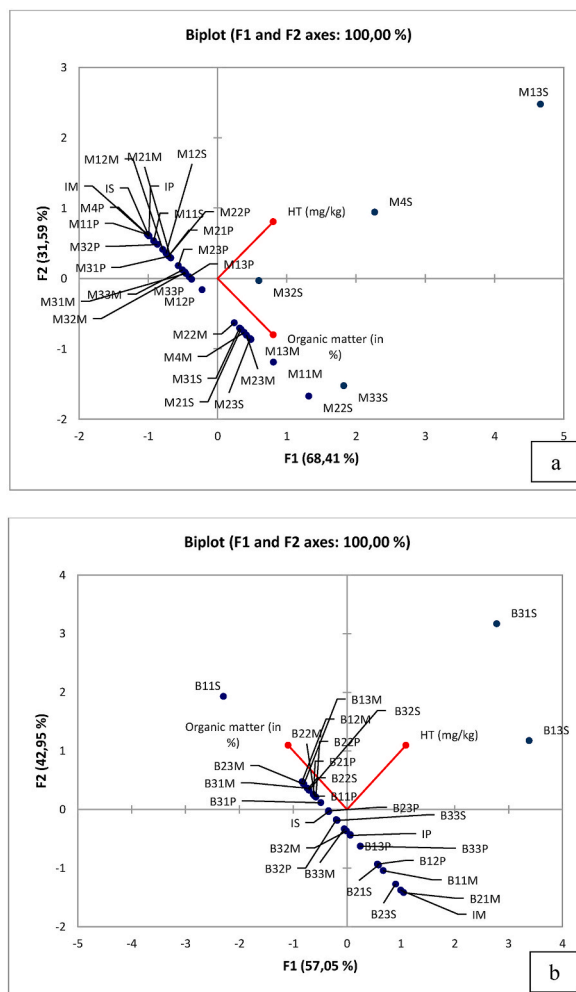
Fig. 8. Representation of the relationship between: a) organic matter and total hydrocarbons on the F1 and F2 correlation diagram at site M; b) organic matter and total hydrocarbons on the F1 and F2 correlation diagram at site B.

[53,91] estimate that more than 70% of the mercury deposits due to emissions are probably retained by the upper part of the humus-rich soil. Traces of mercury found in the lower layers come from displacement in the form of humic and fluvial complexes [53]. [92] also showed that soil organic matter can, depending on the soil matrix, interact with most mercury ionic species and immobilise mercury in acid soils. These results generally reflect the behaviour of the clay-humus complex on trace metals [93].

5.3.2. Influence of organic matter on the behaviour of hydrocarbons

The representation of the samples in the biplot correlation diagram shows that at site M, total hydrocarbons and organic matter are on the positive side of the F1 axis, which represents 68% of the information and reflects the role of gold mining activities on the correlation between organic matter and hydrocarbons. The correlation coefficient ($R^2 = 0.36$) shows that the correlation between organic matter and hydrocarbons is low. However, the position of samples M13S, M4S, M32S, M33S and to a lesser extent M22S, which are also on the positive side of the F1 axis, shows that a relationship can be established between the two substances in these samples (Fig. 8a). Indeed, these samples have high levels of total hydrocarbons and organic matter. We also note a preferential alignment with the other samples. Following this alignment, it appears that the surface and mid-depth samples with average organic matter values (from 6.82 to 7.91%) are found on the positive side of the F1 axis. Samples with low organic matter values, particularly those from the deep site as well as those from the control site, are found on the negative side of the F1 axis. These observations show that the position of these samples is influenced by the organic matter and is not influenced by the total hydrocarbon concentration.

At site B, the correlation coefficient R^2 of −0.14 shows that there is almost no correlation between organic matter and hydrocarbons. The negative value of the correlation coefficient at this site is justified by the fact that organic matter and total hydrocarbons occupy opposite positions on the F1 axis and are found on the negative and positive side respectively (Fig. 8b). This axis, which represents 57% of the total information, also reflects the influence of gold mining activities on the correlation between organic matter and hydrocarbons. The opposite positions on the F1 axis between organic matter and hydrocarbons at site B can be explained by the

fact that there is no significant difference in organic matter content between samples with high total hydrocarbon values and those with low values. A preferential alignment of samples with low total hydrocarbon values is also observed. However, at this site, there is no preferential position of surface samples over mid-depth or deep samples. This is because much of the surface organic matter at this site has been transported by water erosion, so there is no difference between the surface organic matter content and the organic matter content recorded at depth or mid-depth. This is because much of the surface organic matter on this site has been transported by water erosion.

The results obtained allow us to understand that organic matter plays an important role in the fixation of hydrocarbons. Indeed, due to their hydrophobicity, many petroleum hydrocarbons have a strong affinity with organic matter (Yang et al., 2005 [94]) and are bound to the soil by sorption of the organic matter. The sorption strength of the hydrocarbon varies depending on the nature of the hydrocarbon and the organic matter content of the soil (Langley et al., 2003 [95]). Once absorbed, hydrocarbons are difficult to remove and can remain attached to organic matter for many years before being degraded [96].

6. Conclusion

The aim of this study is to make a quantitative assessment of soil contamination by mercury and hydrocarbons in the former gold mining sites of the small mine in the upstream part of the Kadey catchment area. The results obtained show that gold mining is the cause of chemical contamination of the soil due to the spillage of large quantities of hydrocarbon products, the levels of which are 800 times higher than the threshold value at certain points on the former extraction sites. This contamination is likely to pose a serious problem for the health of people living on former gold mining sites or who carry out agricultural activities there. As far as mercury is concerned, its use in amalgamation has led to an increase in its concentration at the surface of the soil to an average level of 0.04 mg/kg. This concentration remains below critical values, but cannot be generalised to the whole catchment area. The abusive use of mercury, which has become widespread in gold mining, is likely to lead to a proliferation of contaminated sites. The study also shows that soil organic matter plays an essential role in fixing hydrocarbons and mercury to the soil surface. However, precipitation water causes infiltration and horizontal redistribution of hydrocarbons, which can contaminate groundwater and surface water. In order to carry out an in-depth assessment of the environmental legacy of gold mining and map out the risks, it will be necessary to carry out additional work at a number of sites in the gold districts, taking into account the increase in the intensity of work and changes in processing methods. It will also be necessary to carry out analyses of the speciation and bioavailability of mercury and hydrocarbons. Investigations will also have to be carried out to assess contamination linked to other substances such as cyanide, which is being used more and more frequently in gold recovery.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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