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Organochlorine and organophosphate pesticides in soils of a vulnerable area from an aquifer in Northern Colombia¹

Pesticidas organoclorados e organofosforados em solos vulneráveis de um aquífero no norte da Colômbia

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HIGHLIGHTS:

Glyphosate is the farmers' most widely used pesticide in the Morroa aquifer recharge area in Sucre, Colombia.

The highest pesticide concentration was found in the northern Corozal municipality area.

Fine textures and low pH determine the presence or absence of pesticides in the most vulnerable areas of the Morroa aquifer.

ABSTRACT: The Morroa aquifer, which is an essential source of water supply in Sucre, Colombia, with more than 500,000 users, may be at risk because of pesticide contamination that comes from different agricultural activities. The aim of this research was to evaluate the relationship between the presence of pesticides and the physicochemical properties of soils in the most vulnerable areas of the Morroa aquifer. Therefore, farmers were surveyed about pesticide usage and soil samples which were taken for analysis of pesticide and physicochemical properties. The results demonstrated that soils in these areas were relatively homogeneous in terms of texture, pH, cation exchange capacity and organic matter content. Pesticides detected in these soils were malathion, β -BCH, p-p'DDE, m-p'DDD, p-p'DDT, endosulfan, endrinetone, chlorpyrifos, dieldrin, endosulfan-sulfate, heptachlor-epoxide, and parathion. Physicochemical properties such as pH and texture of soils and intrinsic characteristics of the pesticide were found as the most influential variables according to statistical results. This study also presents a map of the spatial distribution of pesticide concentration according to the concentration of pesticides, which constitutes an important tool for the planning of agricultural activities that supports decision-making and the implementation of measures to mitigate potential impacts caused by pesticide pollution.

Key words: soil contamination, spatial distribution of pesticides, groundwater pollution

RESUMO: O aquífero de Morroa é uma fonte essencial para o abastecimento de água em Sucre, Colômbia, com mais de 500.000 usuários que podem estar em risco devido à contaminação por pesticidas que provêm de diferentes atividades agrícolas. O objetivo desta pesquisa foi avaliar a relação entre a presença de pesticidas e as propriedades físico-químicas dos solos nas áreas mais vulneráveis do Aquífero Morroa. Para isso foi aplicado um inquérito sobre a utilização de pesticidas aos agricultores e foram coletadas amostras de solo para análise de pesticidas e propriedades físico-químicas. Os resultados mostraram que os solos nestas áreas são relativamente homogêneos em termos de textura, pH, Capacidade de Troca Catiônica e Matéria Orgânica. Os pesticidas detectados nestes solos foram Malathion, β -BCH, p-p'DDE, m-p'DDD, p-p'DDT, Endosulfan, Endrinetone, Chlorpyrifos, Dieldrin, Endosulfan-Sulfate, Heptachlor-epóxido e Parathion. O pH, a textura do solo e as características intrínsecas do pesticida são as variáveis mais influentes de acordo com os resultados estatísticos. Este estudo também mostra a distribuição espacial da concentração de pesticidas em toda a área do aquífero. O mapa elaborado constitui uma ferramenta para o planejamento de atividades agrícolas que subsidia a tomada de decisões e a implementação de medidas para mitigar os potenciais impactos causados pela poluição causada pelos pesticidas.

Palavras-chave: poluição do solo, distribuição espacial de pesticidas, poluição da água subterrânea

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INTRODUCTION

The Morroa aquifer is an essential source of water supply in some municipalities of Córdoba and Sucre in Colombia as a result of the physical scarcity of surface water that affects this region located in the north of the country. This aquifer constitutes a hydrogeological richness due to the water security that offers to the population. According to Domínguez-Pérez & Mercado-Fernández (2020), more than 500,000 inhabitants rely on this source for various water use.

According to Vergara et al. (2009), 7.4% of the recharge area of the Morroa aquifer, which corresponds to rural areas of Sincelejo and Sampues, is highly vulnerable to pesticides. The massive use of pesticides on corn, cassava, yams, cotton, and tobacco crops causes soil contamination and reduces soil fertility. In addition, infiltration and leaching processes can lead to groundwater contamination, depending on the mobility of the pesticide. This impact jeopardizes water security, which affects access to good quality water for various uses (Pradhan et al., 2022; Holten et al., 2019).

It is necessary to understand that sorption, degradation, and volatilization are fundamentally governed by the action of climatic factors such as temperature and precipitation (Donga et al., 2020; Resende et al., 2022), the characteristics of the pesticide and its type of application, the nature of the soil, and the processes that take place in it (Zhang et al., 2021). On the other hand, the physicochemical properties of the soil that can determine the behavior of pesticides are texture, pH, organic matter content and cation exchange capacity (Vryzas, 2018). The aim of this research was to evaluate the relationship between the presence of pesticides and the physicochemical properties of soils in the most vulnerable areas of Morroa aquifer.

MATERIAL AND METHODS

The recharge area of the Morroa aquifer is located in the municipalities of Corozal, Morroa, Los Palmitos, Ovejas, Sincelejo, and Sampues, in the department of Sucre, Colombia (Figure 1).

The Municipal Unit of Agricultural Technical Assistance (UMATA, its Spanish acronym), due to its being, is fully aware of the agronomic management of crops in their jurisdiction. To characterize pesticide uses in this region, a representative sample of farmers, who frequently apply agrochemicals, was selected for each municipality. With this information, a non-probabilistic, intentional, extreme case sampling was designed (Zhao et al., 2020). The producers that most apply agrochemicals are the extreme sampling. Subsequently, each farm was visited to perform a survey among the selected sample, obtaining the most used pesticides with their corresponding doses with a total of 76 georeferenced surveys that were applied in the study area.

For analyzing soil characteristics and pesticide presence, 21 representative soil samples, composed of three to five sub-samples, were collected from 30 to 40 cm depths from the soil surface, taking into consideration that these depths are where the pesticides remain in the soil (Lupi et al., 2015; Sabzevari & Hofman, 2021).

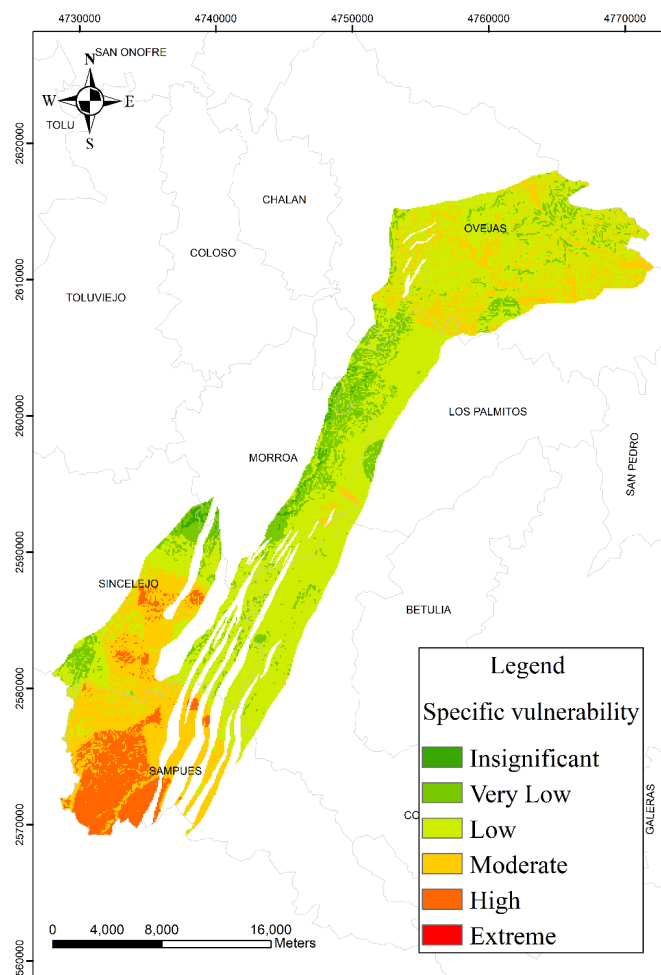


Figure 1. Vulnerability map of the Morroa aquifer

These soil samples were taken at the lowest elevation sites according to the hypsometric map, which also presents high levels of vulnerability due to the accumulation of different chemical substances carried by surface runoff (Figure 2). It is necessary to clarify that these points do not necessarily correspond to the farms surveyed due to the statistical method used.

Table 1 presents methods used for physicochemical characterization and pesticide determination in soils. For results analysis, the statistical technique of the Principal Component was carried out through R Studio. Finally, the geostatistical analysis of pesticide concentration was carried out by the kriging method in ArcMap 10.5.

RESULTS AND DISCUSSION

The results of physicochemical characteristics demonstrate that the soils located in the lower zones of the recharge area of the Morroa aquifer had a high clay content with high cation exchange capacities, the organic matter content was medium to low, and the pH ranges were neutral to acidic (Table 2).

On the other hand, the survey results reflected the main active ingredients of pesticides most used by the population in the study area. These pesticides were chosen for soil sampling in vulnerable areas, according to the map shown in Figure 2.

The survey results are presented in Figure 3, where it can be deduced that glyphosate is the most used pesticide

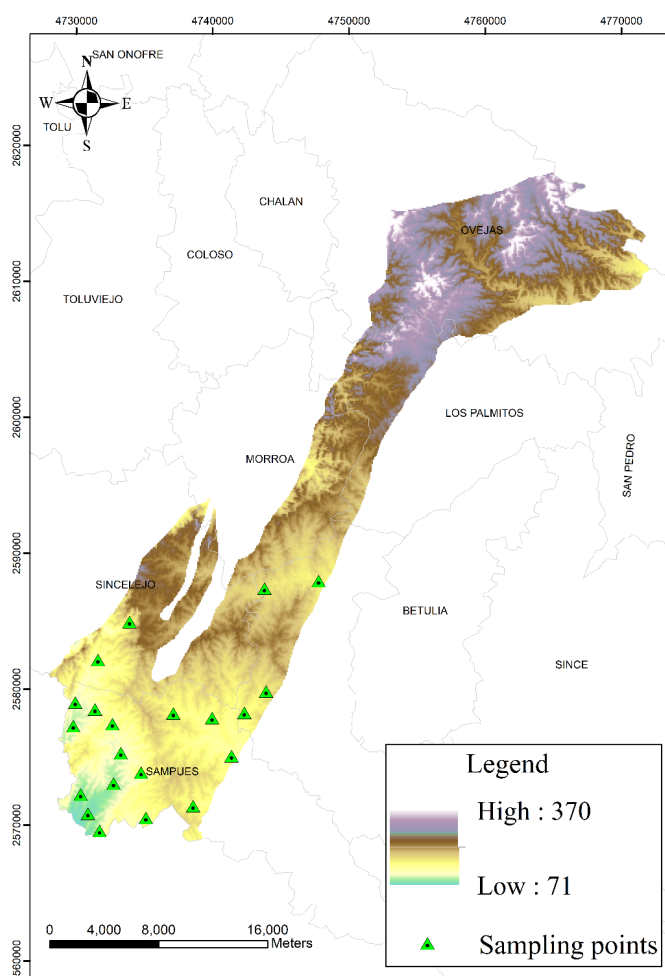


Figure 2. Sampling soil points taken in the study area

Table 1. Methods for pesticide analysis and physicochemical characterization

Pesticides	LD ($\mu\text{L } \mu\text{g}^{-1}$)	Volume	Equipment	Method
Malathion	1			
Chlorpyrifos	1			
Parathion	1			
B-BCH	1			
Heptachlor-epoxide	1			
Endosulfan	1	100 mL	GC-MS	You et al. (2004)
Dieldrin	1			
Endosulfan sulfate	1			
Endrinacetone	1			
p- p'DDE	1			
m- p'DDD	1			
p- p'DDT	1			
Physicochemical parameters	Method			
Texture	Hydrometric or Bouyoucos method (IGAC, 2006)			
Cation exchange capacity	1 Normal and Neutral Ammonium Acetate method (IGAC, 2006)			
pH	Potentiometric (IGAC, 2006)			
Organic matter	Walkley-Black (IGAC, 2006)			

by population in the study area with a 61% of frequency followed by the organophosphate insecticide chlorpyrifos, with a 42% of frequency. In order of application, it followed the herbicide paraquat, which belongs to the dipyrindyl group, and is considered a highly toxic herbicide (Niveditha & Shivanandappa, 2022). Finally, it followed dimethoate, diuron,

Table 2. Physico-chemical characteristics of soils

Sample	S	C	Si	Texture	CEC	OM (%)	pH
	(%)						
1	25.8	42.8	31.4	clay	20.5	1.1	5.8
2	29.2	32.8	38.1	clay-loam	21.5	1.7	6.0
3	47.5	21.1	31.4	loam	20.5	1.1	7.1
4	24.2	39.4	36.4	clay loam	22.0	1.7	6.5
5	25.0	36.1	38.9	clay loam	22.5	1.7	6.4
6	25.0	39.4	35.6	clay loam	23.0	1.5	6.7
7	46.7	29.4	23.9	Sandy clay loam	20	1.5	6.8
8	15.6	40.8	43.6	Silty clay	22.5	2.0	6.5
9	48.9	22.5	28.6	loam	21.0	1.1	5.1
10	32.3	40.8	26.9	clay	22.5	1.1	5.5
11	22.3	40.8	36.9	clay	19.5	2.2	6.2
12	35.6	4.2	60.3	Silty Loam	20.0	3.3	7.2
13	20.6	35.8	43.6	clay loam	20.0	2.3	6.1
14	23.9	37.5	38.6	clay loam	21.5	2.5	6.1
15	38.5	49	12.5	clay	19.7	1.81	6.33
16	43.8	35.1	21.1	clay loam	21.7	1.73	6.13
17	32.1	19.3	48.6	loam	20.3	1.22	6.53
18	37.9	30.1	32	clay loam	22.3	1.52	6.85
19	44.5	29.2	26.3	clay loam	22.7	1.37	6.42
20	40.4	34.2	25.4	clay loam	23.4	1.79	6.75
21	41.2	48.7	10.1	clay	22.34	1.21	6.84

S - Sand; C - Clay; Si - Silt; CEC - Cation exchange capacity; O.M. - Organic matter; pH - Hydrogen potential

cypermethrin, amine, methyl parathion, and malathion, respectively. These results are in agreement with Vergara et al. (2006).

Vergara et al. (2006) identifies that in the municipalities of Corozal, Morroa, and Los Palmitos (a partial area of the present study) the most frequent active ingredients used by farmers are: glyphosate, paraquat, malathion, and cypermethrin. The continuous use of these pesticides in agricultural activity generates pest resistance against their effects and alters the physicochemical properties of the soil (Boudh & Singh, 2019). The modification of properties, such as acidity and cation exchange capacity, limit the buffering capacity of soils and is related to the persistence in the use of the same contaminant pesticide.

The analyzes for pesticide detection in soils according to the methods in Table 1 revealed the presence of all the pesticides evaluated in at least one point of the 21 sampled during the field phase. Figure 4 demonstrates the spatial distribution of the pesticide's concentration and the sites where soils can be contaminated due to high levels of these substances. In order to focus the results in the sampled area, the aquifer was cut just for the southern part of it where vulnerable areas are predominant.

The spatial distribution demonstrates that the highest point concentration occurs with DDE, which reached a maximum soil concentration of $22.4121 \mu\text{g kg}^{-1}$ (Figure 4F). This value represents a punctual risk in the northern zone of the municipality of Corozal, within the aquifer area. On the other hand, pesticides such as malathion, endrinacetone and chlorpyrifos present a homogeneous dispersion of contamination associated with the detection of the pesticide in a greater number of points, generating a potential risk throughout the southern zone of the aquifer, where the recharge areas are located.

Organophosphates: Malathion (in 42.85% of the samples) and chlorpyrifos (in 33.33% of the samples), are short-lived pesticides in the soil due to their proven biodegradability (Vaishali et al., 2020) which suggests a low risk of reaching

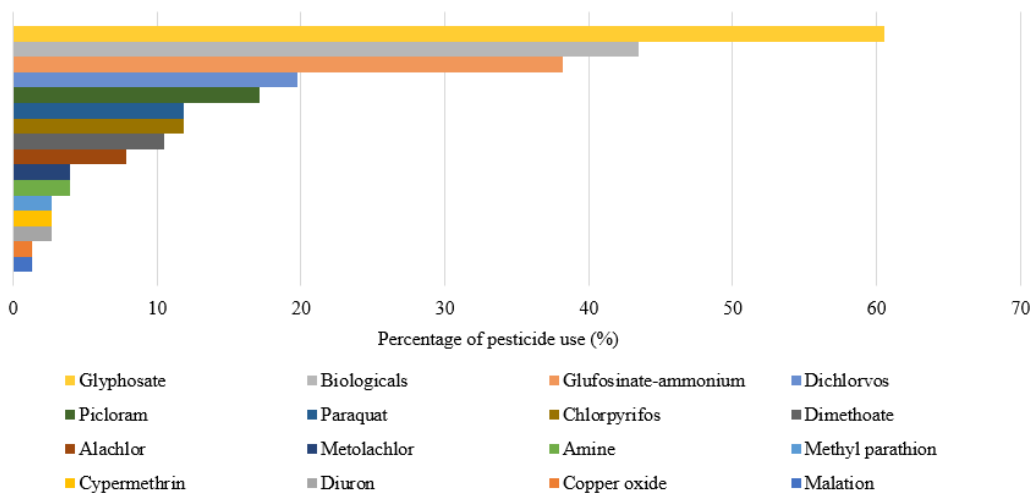


Figure 3. Pesticides used in the study area, expressed in percentages (%) according to their frequency of use

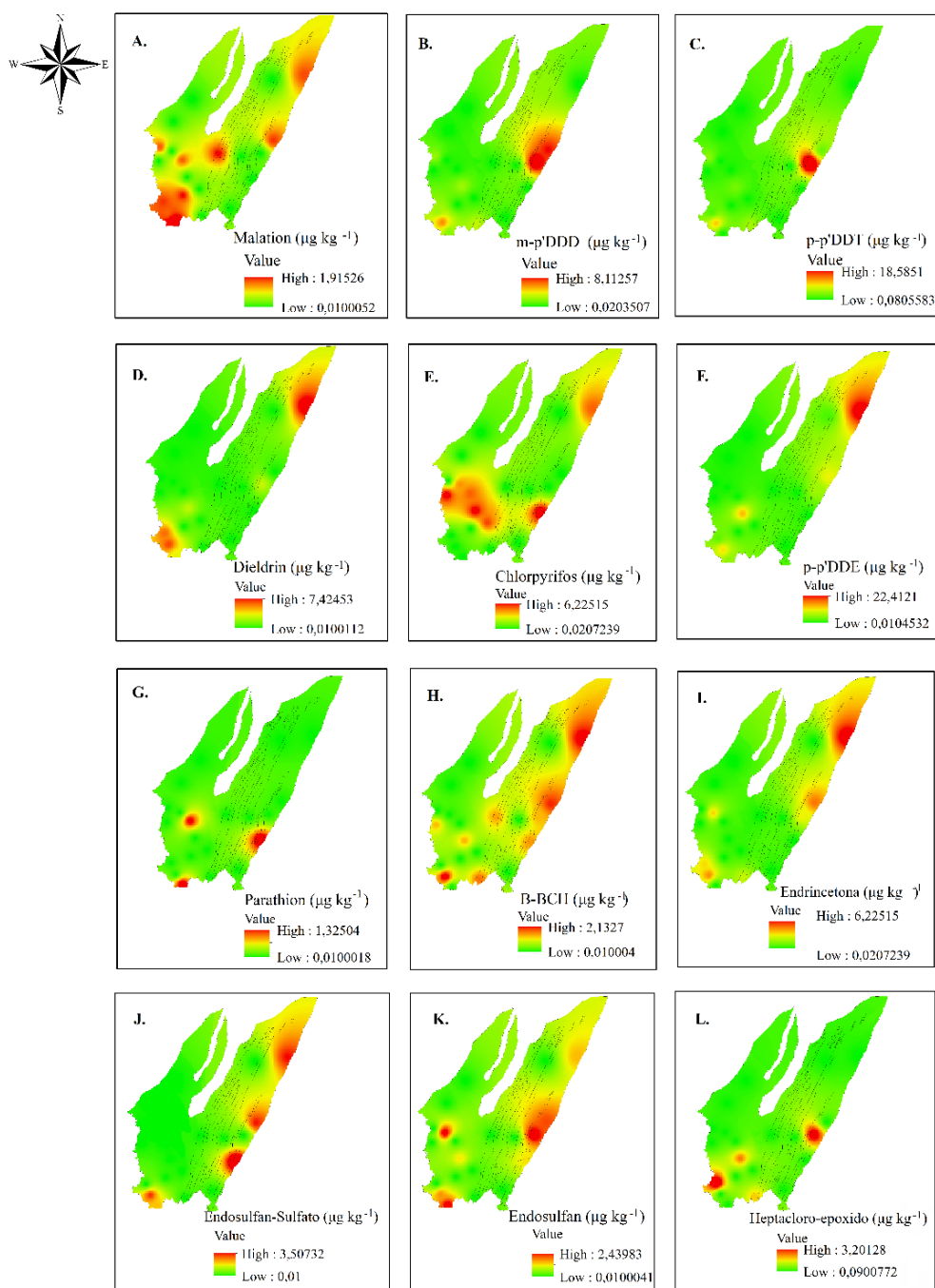


Figure 4. Spatial distribution of pesticides in the Morroa aquifer

aquifers; however, their presence has been reported in these water sources (Duttagupta et al., 2020; Vasseghian et al., 2022). For Endrinetone, despite being distributed throughout the southern zone of the Morroa aquifer, more attention is required on the potential risk that it poses to the councils of Sampues and Corozal.

The histograms (Figure 5) reflect the frequency of detection and maximum peaks reached by the pesticides endrinetone, chlorpyrifos, and malathion.

Figure 6 presents the frequency of detection of pesticides in the soil from the sampling carried out. It is important to mention that the organochlorine insecticide β -BCH, which according to the results of the surveys, is not currently used by farmers in the region and was analyzed as a control during the field phase, was detected in 42.85% (Figure 6) of the samples with a maximum concentration of $2.1327 \mu\text{g kg}^{-1}$ (Figure 4H), which demonstrates its persistent characteristics in soils and the risk associated with aquifer contamination (Perera-Rios et al., 2021). This shows that risk mitigation measures for phytosanitary contaminants shall be taken as soon as possible and carried out through legal and communicative actions that promote awareness among farmers to reduce or definitively

suspend their use, since the risk may persist over time, it may compromise food and water security in a region that is mainly supplied by groundwater.

In addition, controls such as methyl-azinphos and carbamates were not detected, as expected, since these are not applied by farmers, while the organophosphates chlorpyrifos and parathion were found in 33 and 14% of the soil samples, respectively (Figure 6). No glyphosate detected in the soil samples analyzed (Figure 6). This is an apparently paradoxical result since it was the most applied pesticide in the study area, according to surveys. Samples were taken from 30 and 40 cm depths. Glyphosate remains strongly attached between 12 -14 cm of the soil profile. Beyond these depths, the concentrations are practically null (Wang et al., 2019). Rodriguez-Gil et al. (2021) confirmed its rapid disappearance from the soil and the low polluting potential of surface and groundwater. Likewise, Giesy et al. (2000) concluded that glyphosate normally dissipates rapidly from simple ecosystems, such as agricultural ecosystems, and from more complex ecosystems, such as forestry regardless of soil diversity and climatic conditions, indicating a half-life of 32 days. In sequence, Weaver et al. (2007) determined that glyphosate has limited and transient

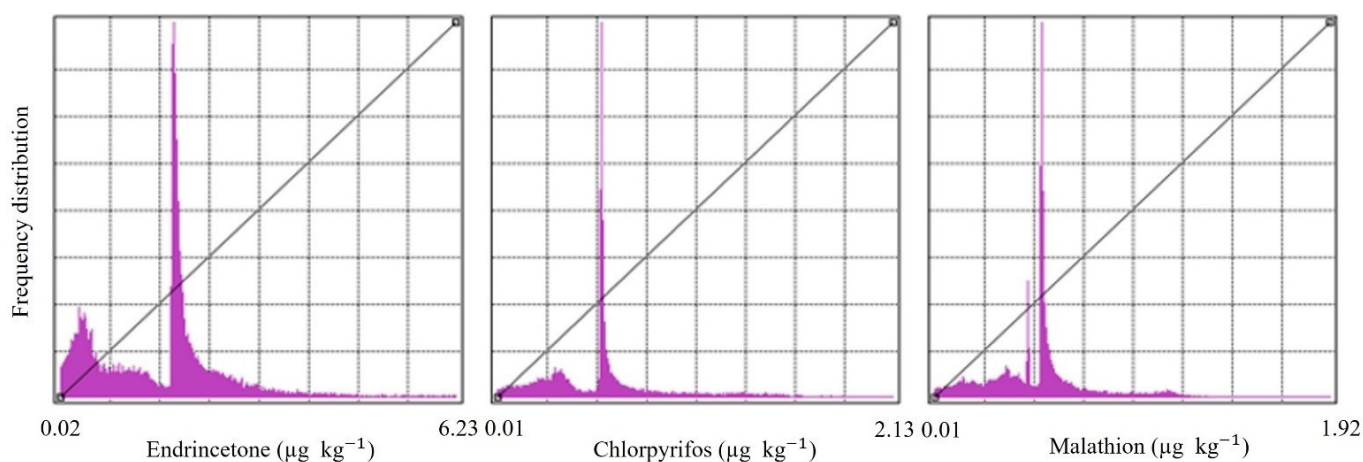


Figure 5. Concentration of pesticides histograms

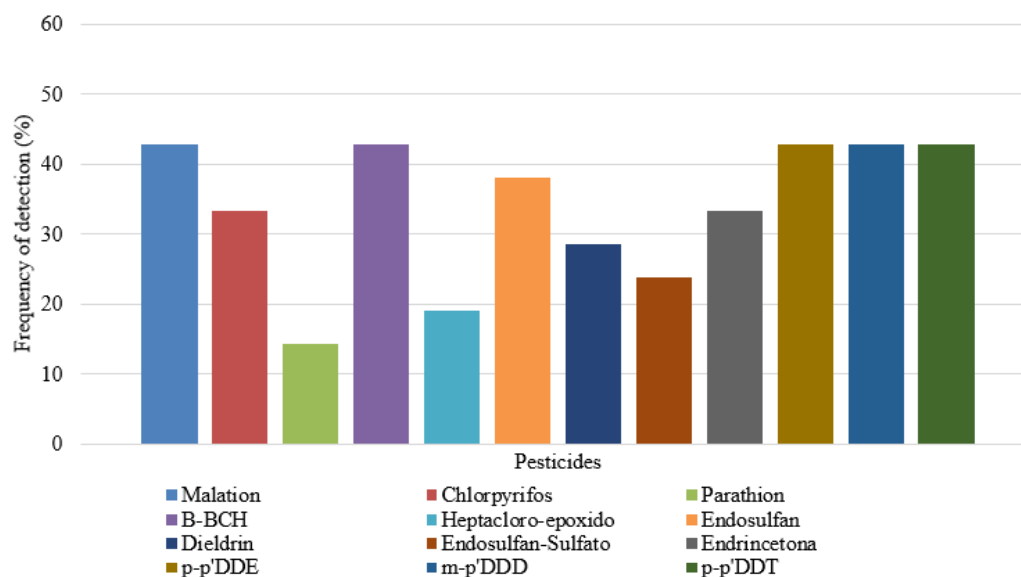


Figure 6. Frequency of detection of pesticides in soils

effects on the soil microbial community, even when applied at high doses.

However, it cannot be ruled out that glyphosate reaches groundwater, as Van et al. (2008) determined that it is found in low concentrations (below the concentrations permitted by Canadian drinking water standards) in groundwater in some regions of Canada. It is important to state that these are shallow aquifers (10 to 30 m) composed of fractured rocks or sands outside the Morroa aquifer. Furthermore, climatic factors, such as temperatures, which are less aggressive in North American countries, degrade pesticides slowly and make them more persistent in the soil (Li, 2020; Bento et al., 2019).

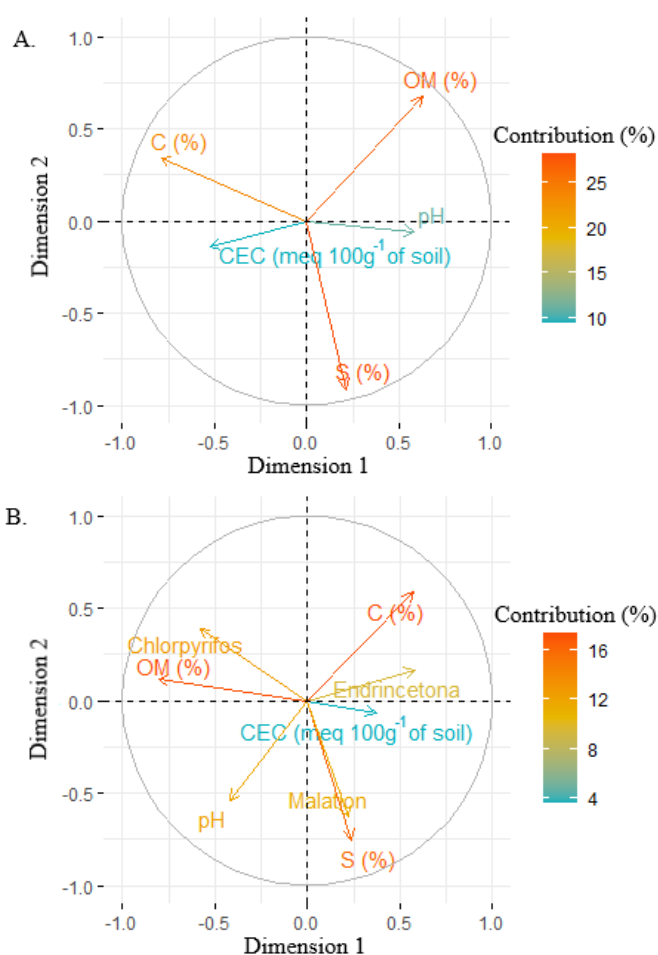
The main component A analysis of the physicochemical attributes measured in the field (%C, %S, %O.M and CEC) is presented in Figure 7A. Indicating that the cation exchange capacity and the percentage of clay are the most related, which is validated in the article by Suleiman et al. (2018). This proposes the possibility of predicting texture from CEC, since it indicates the ability of soils to retain cations, availability, and amount of nutrients to the plant and it is usually low in sandy soils. The first graph tests the consistency of the data obtained in the sampling when confronted with the theoretical framework studied. This increases the value of the analysis that can be derived from it based on the pesticides typified as the greatest interest for this study: chlorpyrifos, endrinetone, and malathion.

Figure 7B indicates the relationship between the three pesticides and the physicochemical parameters monitored. It reflects that there are relationships between the three pesticides and different physicochemical attributes. For instance, since malathion is very close to the axis of the percentage of sand, it is possible to attribute the highest concentrations of this pesticide to be found in sandier soils. Malathion would then be the least relevant pesticide since these soils are mainly clayey.

On the other hand, chlorpyrifos and endrinetone were associated with the percentage of organic matter and cation exchange capacity, respectively. It is necessary to clarify that none of the variables evaluated in the soil alone does condition the presence of pesticides in this environmental element, since it is observed in the main component analysis obtained that the factors OM, pH, texture, and CEC are far from the major axes. However, texture and pH jointly do condition the presence or absence of pesticides in the soils of the most vulnerable areas of the Morroa aquifer.

The Morroa aquifer is the only source of drinking water for several municipalities in the departments of Sucre, Bolivar, and Cordoba (Vergara et al., 2009). The presence of these pesticides in soils of the recharge areas of such a vital water body indicates a potential leaching that would end up in the human population supplied by it. Therefore, the results of the current research demonstrate a concern for environmental governance.

This research provides an important overview of the possible impacts of pesticide contamination in the Morroa aquifer, serving as a tool for planning agricultural activities to avoid the environmental and health risks associated with these pollutants.



OM - Organic matter; CEC - Cation exchange capacity; C - Clay; S - Sand; pH - Hydrogen potential

Figure 7. Principal components analysis for soil physicochemical properties (A) and pesticides and soil physicochemical properties (B)

CONCLUSIONS

1. Inappropriate activities are being carried out on this important geological arrangement like using pesticides, such as glyphosate, chlorpyrifos, paraquat, diuron, cypermethrin, amine, methyl parathion, and malathion by 61, 42, 38, 21, 16, 12, 12, 11 and 8% of farmers, respectively, and these put them at risk.

2. The concentrations of pesticides in the soil are not low, since in the case of pp'DDE, it is found in concentrations up to 22.4121 $\mu\text{g kg}^{-1}$, and it is necessary to highlight the detection of organochlorine pesticides that were analyzed as controls; therefore, all these were not expected to exist due to the reported persistent characteristics in the environment.

3. The spatial distribution of malathion, chlorpyrifos, B-BCH and endosulfan sulfate has shown higher levels according to the variability of data and geostatistical interpolation.

4. According to the main component analysis the presence of pesticides in sampling soils corresponding to the most vulnerable areas of the Morroa aquifer is due to the intrinsic conditions of these substances, also pH and texture attributes, in these specific areas.

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