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Risk estimate of water contamination by pesticides used in coffee crops

Alexandra F. S. Soares[1], Mônica M. D. Leão[2], Márcio R. Vianna Neto[3] & Sílvia M. A. C. Oliveira[4]

ABSTRACT

A study was conducted in Manhuaçu city, Minas Gerais, aiming to study the effects of pesticides used in coffee crops on surface and ground waters. Manhuaçu is located in an important Brazilian region of agricultural production. Moreover, the city possesses many different water sources directed to the public water supply, and 13 points of surface and groundwater pumping. There have also been several occurrences of cancer in the local population. A survey conducted among cities of Minas Gerais identified 122 cities with high occurrences of death due to cancer, among which was Manhuaçu. The physico-chemical properties and applied mass of the active ingredients of each pesticide studied were surveyed. Goss and GUS algorithms were applied to estimate the risk of surface and ground water contamination, respectively. Some of the most widely used pesticides in the region, whose contamination potential was estimated as high, were later detected in surface waters through LC/MS-MS. This study can help agriculturists and officials of environmental organizations select and control, respectively, less harmful products to the water.

Key words: pesticides, water contamination, GUS index, Goss' algorithm

Estimativa de risco de contaminação de mananciais por agrotóxicos utilizados em culturas de café

RESUMO

Um estudo foi realizado no município de Manhuaçu, Minas Gerais, objetivando estudar os efeitos dos agrotóxicos utilizados nas culturas de café em águas superficiais e subterrâneas. Manhuaçu está localizada em uma região brasileira importante em termos de produção agrícola. Além disso, a cidade possui vários mananciais destinados ao abastecimento público e 13 pontos de captação de águas superficiais e subterrâneas. Houve várias ocorrências de câncer na população local. Um levantamento realizado em municípios de Minas Gerais identificou 122 cidades com altas ocorrências de câncer na população, entre essas constava Manhuaçu. As propriedades físico-químicas e as massas aplicadas de cada ingrediente ativo dos agrotóxicos estudados foram levantadas. Os algoritmos de Goss e GUS foram utilizados para se estimar o risco de contaminação de águas superficiais e subterrâneas, respectivamente. Alguns dos agrotóxicos utilizados na região, cujos potenciais de contaminação foram estimados como sendo altos, foram detectados em águas superficiais por CL/EM-EM. Este estudo pode auxiliar agricultores e organizações ambientais oficiais na seleção e controle de produtos, respectivamente.

Palavras-chave: agrotóxicos, contaminação de águas, índice GUS, algoritmo de Goss

[1] Doutoranda do Programa de Pós Graduação em Saneamento, Meio Ambiente e Recursos Hídricos do Departamento de Engenharia Sanitária e Ambiental da Universidade Federal de Minas Gerais – DESA/UFMG. E-mail: asaraiva.soares@gmail.com
[2] DESA/UFMG. Av. Antônio Carlos, 6.627, Campus Pampulha, CEP 31270-901, Belo Horizonte, MG. E-mail: monica@desa.ufmg.br
[3] Graduando em Engenharia Química. Escola de Engenharia/UFMG. E-mail: marciorvneto@gmail.com
[4] DESA/UFMG. Av. Antônio Carlos, 6.627, Campus Pampulha, CEP 31270-901, Belo Horizonte, MG. E-mail: silvia@desa.ufmg.br

INTRODUCTION

During the 40s the first synthetic pesticides were made available, which later generated great benefits for crop productions. However, the adverse impacts of these products in the environment and on human health were only first discussed at the beginning of the 60s through the publication of "Silent Spring" (Carson, 1962). Since then, studies and discussions about water contamination, as well as the risks and benefits of pesticide use in agriculture, have taken place (Grützmacher et al., 2008; Vieira et al., 2009; Gonçalves et al., 2005; Corbi et al., 2006; Brondi et al., 2004; Marchesan et al., 2007). The exponential growth of the human population and the subsequent demand for food make the application of these products on crops necessary to ensure productivity at satisfactory levels.

The amount of pesticides required to fight diseases and plagues in crops represents an extremely small percentage of the total consumption. Studies report that more than 90% of pesticides applied do not reach the target organisms and, instead, end in other parts of the environment. Thus, the use of pesticides in agriculture inevitably contaminates the environment, which ultimately leads to the contamination of human beings. These undesired effects may compromise environmental quality (Werf, 1996). Still, according to Werf (1996), pesticides, depending on their own characteristics, may settle in different parts of the environment such as air, soil, surface and ground waters, which may be reached through air drift, leaching and runoff. The contamination of surface water may cause ecotoxicologic effects to aquatic flora and fauna, as well as to human health, if destined for consumption (Cerejeira et al., 2003; Bortoluzzi et al., 2006; Vanzela et al., 2010).

It is known that the introduction of toxic substances in aquatic ecosystems is among the most complex causes of potable water quality deterioration; especially, concerning pesticides, many of which are resistant to, and persistent in the face of, conventional potable water treatment (Stackelberg et al., 2007; Ormad et al., 2008; Thuy et al., 2008).

In 2001, the European Union selected 33 organic compounds, 16 of which were pesticides, as the most damaging to the control of water pollution due to their elevated toxicity, high environmental persistency and bioaccumulation potential (Council Directive, 2001). Recently, a large group of organic compounds has been marked by the American Environmental Protection Agency (EPA) and by the European Union as emergent water contaminants (Sanches et al., 2010). These factors have motivated the development of scientific research, which seeks to identify and remove these compounds, so that the requirements of the new water quality regulations, which become progressively more demanding, are met. An increase in the number of adverse effects of pesticide application has been taken into account by regulating agencies, which lead them to increase the restrictions on the use of these compounds, and even to ban their use (Werf, 1996). Although some of the most harmful pesticides have been banned, especially in developed nations, there are still several options of compounds available to the agriculturist, each one harmful to the environment to some extent. Thus, it is necessary that studies be conducted, in order to assess the potential for environmental impact of the various available pesticides.

Facing what has been exposed, a survey was conducted of the physico-chemical properties of the pesticides which are used on coffee crops, in the city of Manhuaçu, Minas Gerais, Brazil, and the risk of surface and ground water contamination was assessed.

The city of Manhuaçu is located in an important Brazilian region of agricultural production. Moreover, the city possesses many different water sources directed to the public water supply and 13 points of surface and groundwater caption. Its land relief is strong, wavy and mountainous, and its pluviometric means are high. There have also been several occurrences of cancer on the local population, which was responsible for over 12% of all deaths in the city. A survey conducted among cities of Minas Gerais, with a confidence interval of 95%, was able to identify 122 cities with high occurrences of death due to cancer. Among these was Manhuaçu (Antoniazzi et al., 2009). The research took place through a survey of the products (and their respective amounts) marketed in the region in the years from 2007 through 2010 and physico-chemical properties of their active ingredients, as well as the problems (plagues, plant diseases and weeds) which appear in coffee crop associated with each of these pesticides. Furthermore, indexes and models described in the literature, to estimate the behavior of pesticides in the environment and the expectancy of finding them in the waters, were applied. Several models were developed aiming to predict the environmental final destination of the pesticides, or to assess the risk they pose to the environment. Among the most prominent predictive models are: the EUSES model; and, the one developed by Abraham (Clarke, 2009) and (Cousins & Mackay, 2001). The latter one is based on the prediction of physico-chemical properties of active ingredients based on molecular data. Mackay (2001) has also developed a thermodynamic approach to this problem, through the use of the fugacity of each pesticide, which enables one to predict its concentration in different environmental compartments. A more conventional approach to risk estimation consists of analyzing the vulnerability of groundwater by indexes of GUS (Groundwater Ubiquity Score), and of surface water by the application of the Goss' algorithms. Studies developed in Brazil and abroad by Lourencetti et al. (2005), Milhome et al. (2009), Guzzella et al. (2006), Primel et al. (2005), Mackay & Fraser (2000) and Plese et al. (2009) are cited as examples of the application of these indexes and models, to estimate the environmental contamination risks.

This study aims to employ such risk assessment methods in the estimation of the potential for contamination of ground and surface water. Such analyses indicate which compounds present any risk of hydric resources contamination, contributing to the elaboration of a plan to monitor pesticides in waters.

MATERIAL AND METHODS

Region of study

The region chosen for this study was the city of Manhuaçu, which is located in the watershed of the Rio Doce River, Minas Gerais state, Brazil. This state is the leading national coffee producer, and Manhuaçu stands out from the other cities in agricultural production. The city's coffee harvest was over 20000 t of benefited grains (ABIC, 2010). According to data collected (IBGE, 2010), Manhuaçu disposes of a harvested area of 18150 ha. Still, according to IBGE (2010), the relief is mountainous, varying from 561 to 1760 m; its relative humidity is high (77.8%); and, the region's mean annual pluviometric index is 1860.8 mm. In this same context, during the seasons of rain, there is a high probability that the excess of these pesticides is carried away. The technological levels adopted by the producers are diverse, varying from low to high technology. Most of the coffee crops are planted in terraces. Among the main conservation practices adopted are: handling, using manual or mechanical mowers; soil analysis, for rational recommendation of soil pH correctors and fertilizers; pluvial water catch basins, for erosion controlling; and, as mentioned, planting crops in terraces. In this region, the public water supply system is composed of several independent streams (Figure 1).

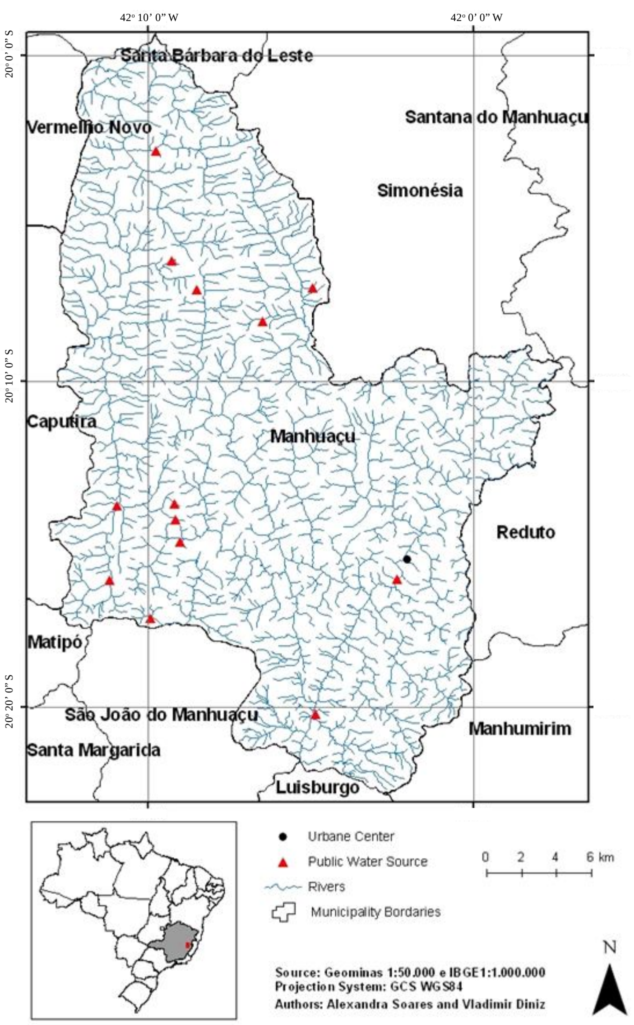

Figure 1. Location of the public water supply sources in Manhuaçu

However, there exist coffee crops adjacent to the water sources for public water supply, disrespecting the areas of permanent preservation established in the Brazilian Forest Code, and increasing the contamination risk to these hydric resources. Besides, the local population's concern with an eventual correlation between contaminated sources and cancer occurrences has led to media coverage, and it is the object of a public civil action impetrated by the Public Ministry of the State of Minas Gerais. A report presented by the Health Bureau of Manhuaçu indicated that cancer is the second greatest cause of death in the region, after undefined causes (Antoniazzi et al., 2009).

A survey of the products (and their respective amounts) marketed in the region in the years from 2007 through 2010 was conducted. The physico-chemical properties of their active ingredients, as well as the problems (plagues, plant diseases and weeds) which take place in coffee crop associated with each of these pesticides, were then stored in a database. The pesticide classes and chemical groups of the active ingredients, and their carcinogenicity, were also surveyed and stored. An exploratory LC/MS-MS analysis was run in order to detect pesticides present in surface water of the region.

Characterization and physico-chemical properties of the active ingredients

The structure and molecular mass, as well as the physico-chemical properties of each pesticide, provides information which enables us to predict the behavior of each substance in the environment. Thus, from the list of products marketed in the region from 2007 to 2010 presented by IMA (2010), the agency responsible for supervising the marketing, the storage and the use of pesticides in Minas Gerais, information regarding the formulation and active ingredients of the marketed products was obtained from the Agrolink database . After this step, the densities of the pesticides were investigated and the total amounts of active ingredients of products marketed in the region were surveyed. The main physico-chemical properties included in this study were: solubility in water at 20 °C, coefficient of adsorption in organic matter (K_{oc}), typical half-life in soil (DT_{50}), and vapor pressure (VP). The use on coffee crops and carcinogenic potential were also surveyed.

Solubility is an important parameter, as it indicates the tendency of a compound to be carried in the surface of a soil by rain or irrigation, and to reach surface waters. However, this parameter should be analyzed along with the soil-water partition coefficient (K_{oc}) to predict the substance's percolation in soil (IUPAC, 2010).

The coefficient K_{oc} indicates the tendency of the pesticide to remain adsorbed in the soil. The sorption of a pesticide in the soil reduces its mobility; and, the magnitude of this reduction also depends on the soil's physico-chemical properties as well as on the pesticide's molecular characteristics. Thus, the organic matter content of a soil is one of the items to be assessed when considering the sorption of a non-ionic pesticide. A constant of sorption, corrected by organic matter of the soil (K_{oc}), may be used to evaluate the pesticide's mobility: $K_{oc} = K_d/oc$, where K_d measures the pesticide sorption by the soil and oc is the fraction of organic carbon in the soil. A sorption constant K_{oc} so expressed permits a comparison of sorption in different soils and is very used in mobility classification methods and simulation models of pesticide behavior in soil (IUPAC, 2010; Silva & Silva, 2007).

In order to assess the environmental persistence of each substance, their respective DT_{50} values were analyzed. Half-life is defined as the time required for the amount of a given substance to be reduced to half of its initial value, which is the amount initially applied to a crop. It should be highlighted that DT_{50} is a criterion used to determine the environmental effects related to volatilization, leaching potential and degradation characteristics of several chemicals (Cabrera et al., 2008).

The software Excel® was employed to treat the data and to apply the Goss and Gustafson algorithms. The physico-chemical properties of each active ingredient were extracted from the website of the International Union of Pure and Applied Chemistry (IUPAC, 2010). The properties of the pesticides were extracted from their datasheets, available at Agrolink (AgrolinkFito) and Material Safety Datasheets (FISPQ).

Evaluation of surface water contamination potential

In order to predict the behavior of these substances in the environment, in addition to analyzing the physico-chemical properties of each active ingredient, a method developed by Goss (1992) was applied. It consists of an algorithm which classifies the contamination potential of each pesticide as high, medium or low for two different forms of pesticide transportation: dissolved in water (SL – "solution surface") and associated with suspended sediments (AD – "adsorbed surface"). The algorithms take into account the values of K_{oc}, DT_{50} and water solubility at 20 °C (Table 1). It does not take into account environmental conditions and, for this reason, it constitutes an estimate to predict the pesticides' risk.

Additionally, when assessing the ingredients which are most likely to contaminate water sources, the amount of the substance marketed during the period, carcinogenic potential and its use in coffee crops were also considered.

Evaluation of ground water contamination potential

In order to estimate the risk of groundwater contamination, physico-chemical properties of the active ingredients were used to determine the GUS indexes, which were originally developed by (Gustafson, 1989) and were later confirmed by the GLEAMS (Groundwater Loading Effects of Agricultural Management Systems) model. GUS indexes are indicators of pesticides' potential to be carried by leaching, and eventually to reach ground waters. These indexes were calculated from values of DT_{50} and K_{oc} and do not take into account environmental conditions (Table 2). Thus, the GUS indexes consist only of an estimate of environmental risk.

Table 2. Algorithms for determination of GUS indexes

Equation	Classification			
	High	Low	Very low	Medium
$GUS = \log(DT_{50}) \times (4 - \log(Koc))$	≥ 2.8	≤ 1.8	< 0 or solubility < 1 and DT_{50} < 1	All others

Source: Goss (1992)

Results and discussion

Survey of the pesticides applied to coffee crops in Manhuaçu city

Several pesticides are applied to coffee crops in the city of Manhuaçu. In total, 280 distinct products were used during the first semester (143 active ingredients), and 235 during the second semester (116 active ingredients). The most widely used were fungicides, herbicides and insecticides (Figure 2).

Most of the products (70.55%) were consumed in small amounts, less than 100 kg year^{-1}. Formulated pesticides, based on glyphosate, were the most traded in the study region, with

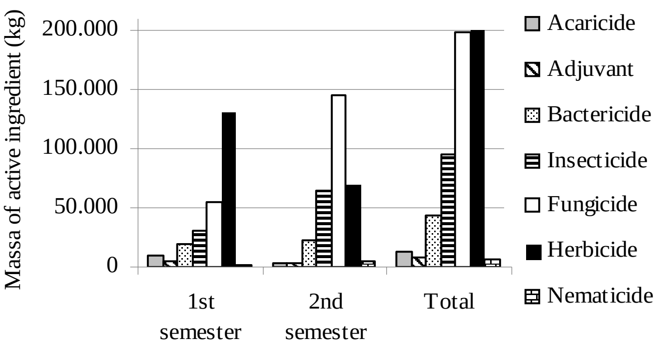

Figure 2. Most widely used pesticide classes in Manhuaçu, from 2007 to 2010

Table 1. Goss' algorithms for evaluating a pesticide's contamination potential in surface water

Transportation	Contamination potential		
	High	Low	Medium
Associated to soil (AD)	DT_{50} ≥ 40 and K_{oc} ≥ 1000 or DT_{50} ≥ 40 and K_{oc} ≥ 500 and Solubility ≤ 0,5	If DT_{50} ≤ 1 or If DT_{50} ≤ 2 and K_{oc} ≤ 500 or If DT_{50} ≤ 4 and K_{oc} ≤ 900 and solubility ≥ 0.5 or If DT_{50} ≤ 40 and K_{oc} ≤ 500 and solubility ≥ 0.5 or If DT_{50} ≤ 40 and K_{oc} ≤ 900 and solubility ≥ 2	All others
Dissolved in water (SL)	If solubility ≥ 1 and DT_{50} > 35 and K_{oc} < 100000 or If solubility ≥ 10 and solubility < 100 and K_{oc} ≤ 700	If K_{oc} ≥ 100000 or If K_{oc} ≥ 1000 and DT_{50} ≤ 1 or If solubility < 0.5 and DT_{50} < 35	All others

K_{oc} - constant of sorption based on organic carbon; DT_{50} - half-life in soil; Source: Goss (1992) annual average consumption of 19 t during the study period (2007 to 2010). But, these herbicides did not have high ground and superficial water contamination potential; also, according to IUPAC (2010), they do not cause cancer.

Over half of all the marketed pesticides were applied to coffee crops (52.58% of the total amount marketed during the first semester, and 64.52% during the second), which indicates how important coffee crop is to the region.

The chemical groups to which the applied pesticides belong are also diverse, with organophosphates (7,98%), pyrethroids (7.36%) and triazoles (8.59%) being the most widely used during the period.

Analysis of physico-chemical properties of the active ingredients

Figure 3 shows the relative frequency of 20 °C solubility of the active ingredients. The analysis of soil/water partition coefficients (K_{oc}) indicated that "non-mobile" active ingredients (> 4000 mL g^{-1}) predominate over those of moderate mobility (75 to 500 mL g^{-1}) and low mobility (500 to 4000 mL g^{-1}) – (IUPAC, 2010).

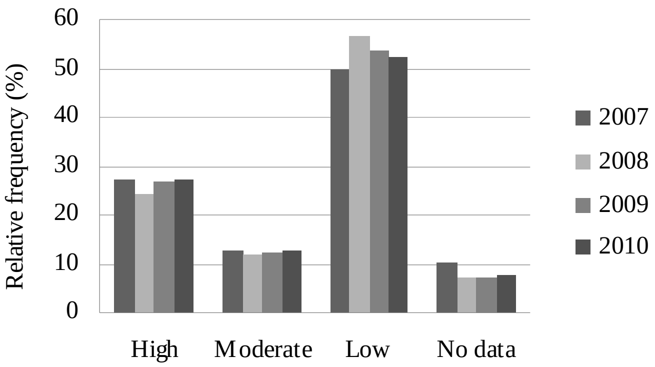

Figure 3. Relative frequency of solubility of the active ingredients in water, at 20 ºC, of the most widely used pesticide in Manhuaçu

Given that most of the products demanded in the region do not dissociate, they also tend to display reduced mobility in the environment. It is important to highlight that highly soluble molecules often display relatively low values of K_{oc} (less than 150 mL g^{-1}), which enables them to be biodegraded more quickly in soil and water (19.61% of all data).

The vapor pressure is defined as the pressure at which a substance reaches liquid-vapor equilibrium for a given temperature. It is a measure of a compound's tendency to evaporate: the greater its vapor pressure, the greater its tendency. It is the main physico-chemical property to be analyzed when predicting the entrance of a given compound, in significant amounts (volatilization), into the atmosphere. It is highly dependent on a molecule's size and functional groups. The results of vapor pressure at 25 °C (Figure 4) indicate that volatile substances predominate (vapor pressure was greater than 10^{-4} mPa for 69.70% of all data).

The DT_{50} values vary greatly among different compounds. These values may be days (organophosphates), months (triazines) or even years (diedrin and DDT). However, no unique value for pesticides half-life may be found; and, DT_{50}

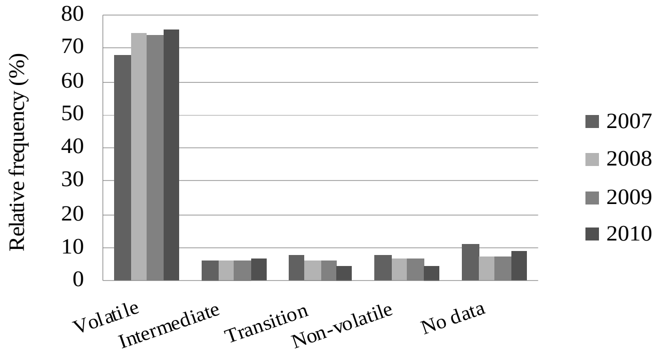

Figure 4. Relative frequency of vapor pressure of the active ingredients, at 25 °C, of the most widely used pesticide in Manhuaçu

determination is highly affected by environmental conditions such as weather, biological activity, type of soil, among others (Silva & Fay, 2004). Most of the data found in the literature were from Europe and in The United States and, thus, are not applicable to tropical countries. The so called "typical values" represent averages of DT_{50} in several different soils and, according to (IUPAC, 2010), are frequently used to guide laboratory and field studies. It is the value which is normally employed in regulatory modeling studies and should be used for aerobic conditions. Thus, for this study, typical DT_{50} values were used in the Goss and Gustafson algorithms.

By analyzing the typical DT_{50} values of each active ingredient, it was noticed that "non-persistent" (DT_{50} < 30 days) products predominate over "moderately-persistent" (DT_{50} between 30 and 100 days) (IUPAC, 2010) ones (Figure 5).

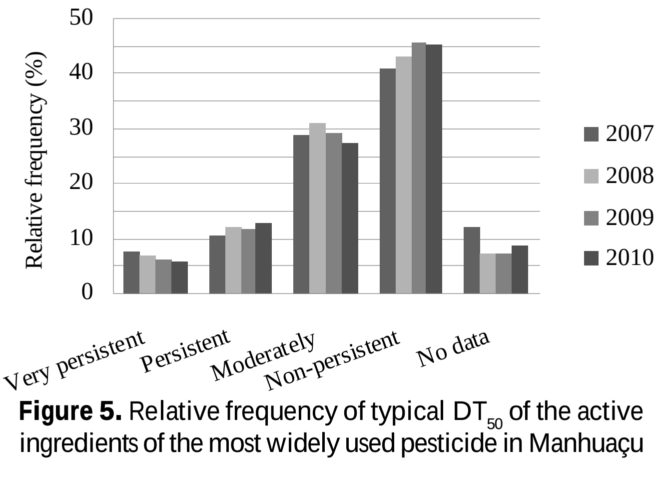

Figure 5. Relative frequency of typical DT_{50} of the active ingredients of the most widely used pesticide in Manhuaçu

Photochemical processes may be important when determining the environmental final destination of organic pollutants in aqueous environments, since they determine the rate of chemical decomposition in aquatic environments, expressed as DT_{50}, induced by light or other radiation. Other information regarding environmental conditions, such as pH, temperature and alkalinity, may influence the determination of the environmental final destination of these pollutants. Aqueous hydrolysis should also be taken into account (IUPAC, 2010).

Most of the products (42.51%) analyzed were classified as non-carcinogenic, followed by those which are potentially carcinogenic (34.56%) according to IUPAC's classification methodology. 3.05% of them were classified as carcinogenic by IUPAC, and another 15.09% may be carcinogenic as well.

It should be noticed that many pesticides may also generate metabolites, which consist of the products of pesticide biodegradation. These metabolites may actually be more toxic than their original compound.

Surface water contamination potential

In order to assess surface water contamination potential, an algorithm known as "Goss' Method" was employed. Figure 6 displays the results of transport potential associated with soil (AD) obtained when the algorithm was employed. Most of the compounds studied (40.61% annual data) displayed medium surface water contamination potential. The four pesticides with greater average annual consumption in the period of study (2007-2010) and which displayed the greatest risk of surface water contamination (AD) were: endosulfan (474.7 kg year^{-1}), chlorpyrifos (262.3 kg year^{-1}), epoxiconazole (89.0 kg year^{-1}) and diuron (70,7 kg year^{-1}) .

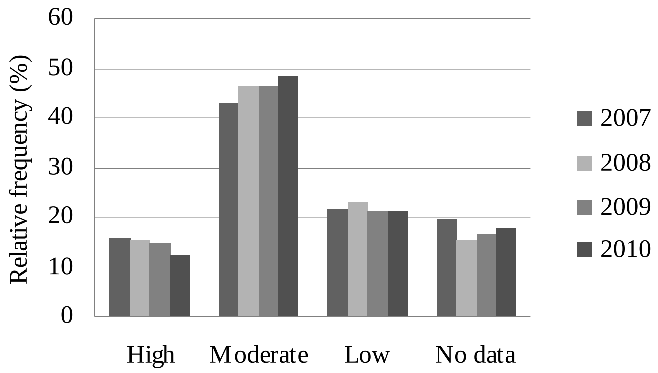

Figure 6. Relative frequency of Goss' Method – Transport potential associated with soil (AD)

Most of the compounds studied (38.18%) displayed moderate surface water contamination potential regarding water solubilization at 20 °C (SL), followed by those with high potential (30.10%) (Figure 7).

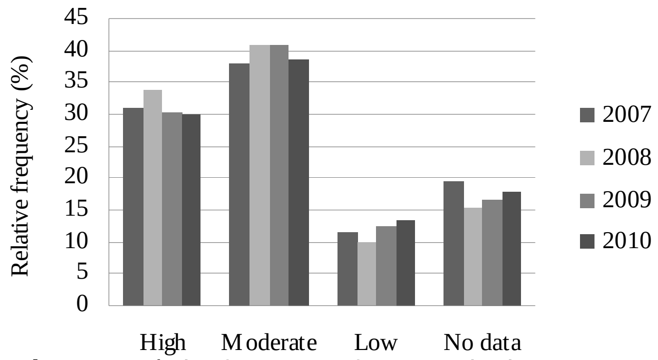

Figure 7. Relative frequency of Goss Method – Transport potential by water solubilization at 20 °C (SL)

The ten pesticides which displayed the greatest risk for surface water contamination (SL) and annual consumption were: flutriafol, chorpyrifos, triadimenol, cyproconazole, tiametoxan, thiophanate-methyl, atrazine, imidacloprid, tebuconazole and diuron.

The pesticide mancozeb displayed medium contamination potential (SL); however, it presented an annual average consumption of 952.3 kg in the period of study (2007-2010). It should be noted that this active ingredient and its metabolite (Ethylenethiourea) are classified as carcinogenic by IUPAC.

The three pesticides which displayed the greatest annual consumption and risk for surface water contamination were: atrazine, flutriafol and epoxiconazole. These compounds were also detected in an LC/MS-MS scan run on samples of water collected from the region, thus validating the methodology employed.

Ground water contamination potential

Regarding the GUS indexes analysis, most of the active ingredients (52.73%) were classified as having "low leaching" potential, presenting little risk for ground water contamination (Figure 8).

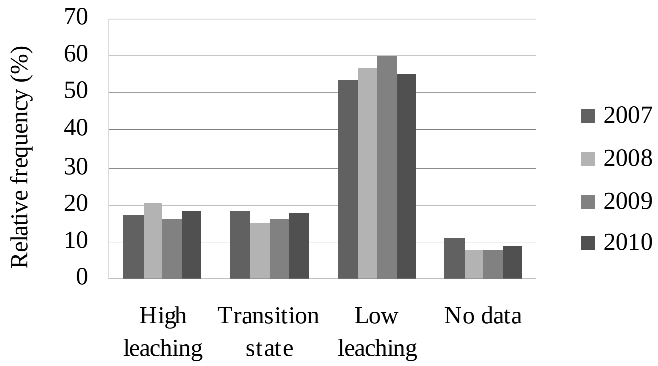

Figure 8. Relative frequency of leaching potential of the active ingredients (GUS indexes)

The four pesticides which displayed the greatest risk for ground water contamination were: picloram, thiamethoxam, cyproconazole and flutriafol.

Surface and groundwater contamination potential based on pesticide classes

The algorithms were applied to each class of the pesticides used in the region (except for the adjuvants, since the active ingredients were the focus of this work). The number of pesticides whose Goss and GUS indexes were low, medium, high or unavailable are summarized in Table 3.

Among the most widely used pesticide classes in the region, the fungicides displayed high surface water contamination potential by solubilization, medium AD and medium-low groundwater contamination potential. The insecticides displayed low leaching potential and medium surface water contamination potential.

The acaricide marketed in the region – cihexatin – has high surface water and low ground water contamination potentials. Mancozeb is used to control mites and fungi simultaneously, and it is included among the ten active ingredients most used in every year of the period of study; and, according to IUPAC (2010), it is carcinogenic. It displays

Table 3. Results of the applied algorithms for each class of pesticides in 2007

Pesticide classes	Goss								Gustafson GUS Index			
	AD				SL							
	L	M	H	ND	L	M	H	ND	L	M	H	ND
Acaricide	0	0	1	0	0	0	1	0	1	0	0	0
Acaricide/Fungicide	1	0	0	0	0	1	0	0	1	0	0	0
Acaricide/Insticide	0	2	5	0	1	4	2	0	7	0	0	0
Bactericide/Fungicide	0	0	2	2	1	1	0	2	2	0	0	2
Fungicide	1	9	4	2	0	4	10	2	7	4	3	2
Fungicide/Inseticide	0	1	0	0	0	1	0	0	1	0	0	0
Fungicide/Inseticide/Nematicide	0	1	0	0	0	0	1	0	0	0	1	0
Herbicide	3	2	2	0	1	4	2	0	3	4	0	0
Inseticide	4	6	2	2	1	9	2	2	10	0	2	2
Inseticide/Nematicide	1	0	0	0	0	1	0	0	1	0	0	0

L – low; M – Medium; H – high; ND - no data; AD – adsorbed surface; SL – solution surface

medium-low surface water and low ground water contamination potential. For mite and insect control, seven active ingredients are used. Among them, endosulfan, an organochlorate, is found. It should be highlighted that endosulfan has its withdrawal date from the Brazilian market set to 2013 (Resolution ANVISA n.28/2010).

The groundwater contamination potential of these products is low, even though their AD and SL potentials varied considerably. AD potentials can be considered high, which indicates that the ground water should be safe. The survey indicated that 4 bactericides/insecticides were used in the region. Their AD potential was high, indicated in the available data, which justified the low leaching potential indicated by the GUS index. Only one fungicide/insecticide - disulfoton - was used in the region. It has medium surface water and low ground water contamination potential. It was also found that only one fungicide/nematicide/insecticide – methyl bromide – was used. It has medium AD, high SL and high leaching potential. One insecticide/nematicide was also used – terbufos. It has low ground water contamination potential, low AD and medium SL.

Conclusions

1. Many pesticides are available to the coffee agriculturist in Manhuaçu city. Overall, they have medium surface water contamination potential (medium AD and SL) and low ground water contamination potential (low GUS index). This way, they may contaminate superficial waters mainly in the rainy period.

2. There are also several pesticides belonging to the same class (such as herbicides and fungicides).

3. This study has presented a methodology which may assist agriculturists in selecting the pesticide, whose effects are less environmentally damaging, especially concerning water pollution, to be employed on coffee crops.

Acknowledgements

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