Artigo

OCCURRENCE OF AGROCHEMICALS IN SURFACE WATERS OF SHALLOW SOILS AND STEEP SLOPES CROPPED TO TOBACCO

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Tobacco cultivation in shallow soils and steep landscape under intense use of agrochemicals contributes to environment degradation. In this study, we assessed the concentration of agrochemicals in draw wells used for human consumption and a creek in a small catchment predominantly cropped to tobacco. Chlorpyrifos, flumetralin, and iprodione were determined by gas chromatography with electron capture detection, while imidalcloprid, atrazine, simazine, and clomazone were quantified by high-performance liquid chromatography with UV detection. Considering all sampling sites, all agrochemicals were detected at least once, except for flumetralin. The occurrence of agrochemicals in tobacco crops is a consequence of their fast transfer to surface water.

Keywords: environmental contamination; pesticides; water quality.

INTRODUCTION

Agrochemical use, industrial effluent discharge, and surface runoff from solid sediments can lead to contamination of water sources, making the water unfit for human and animal consumption. The contamination of water sources can have significant negative impacts on rural communities, with reflections on the urban environment.[1] Among the pollutants in the ecosystems, agrochemicals, the generic term given for various chemical products used in agriculture, stand out owing to their active ingredients such as atrazine, simazine, clomazone, flumetralin, chlorpyrifos, and iprodione, which are widely used as prophylactic treatment in plants to maintain high agricultural production.

The southern region of Brazil is responsible for 96% tobacco production, where more than 186,000 families are involved in small- or medium-scale tobacco production, with an average farm size between 1 and 10 ha.[2] Most tobacco producing areas in the Rio Grande do Sul, southern Brazil, are located in regions having ecologically fragile soils with low clay content, steep slopes, and intensive use. However, the integration of farmers with tobacco industries to boost productivity by using improved production techniques – such using agrochemicals (fungicides, insecticides, nematicides, herbicides, molluscicides, and sucker suppressing);[6,7] unsustainable land use and management;[3] and the rampant application of nitrogen, phosphate, and potassium fertilizers[4,5] – has led to degradation and pollution of these fragile agroecosystems. The most worrisome environmental effect is that majority of these areas are the heads of water catchments and are the source of rivers that supply both rural and urban populations.[8]

In agricultural regions, agrochemical contamination of water sources occurs through point source or diffuse form. The diffuse pollution is pronounced in environments highly degraded by anthropogenic activities such as deforestation in tobacco producing regions, improper soil management, poor infrastructure, and the use of technological packages without attention to negative effects. Intensive soil tillage, pulverization, and a lack of conservation practices cause soil fragmentation and sediment transport by surface runoff.[9-13] Thus, rainfall events following the application of agrochemicals to crops may transport both those with high water solubility (imidacloprid and clomazone) and those that strongly adsorb the functional groups of soil colloids (flumetralin, iprodione, and chlorpyrifos), which are commonly applied to tobacco crops, to surface water sources.

The presence of agrochemicals and other pollutants, even in minute concentrations, is an indication of probable ecological problems and a threat to human health. Virtually, every country has established water quality legislation for both human consumption and agriculture. In developed countries, there is a strong tendency for strict regulation than in developing nations. In the European Community, for example, the maximum active ingredient present in potable water should not exceed 0.1 $\mu g\ L^{-1}$, and the sum of all active ingredients should be less than 0.5 $\mu g\ L^{-1}$.[14]

There are specific laws regarding the presence of organic molecules, especially agrochemicals. Again, the European Community was the pioneer in the establishment of maximum acceptable standards. In this norm, the maximum levels of specific synthetic pollutants, such as agrochemicals, should not exceed the limits established by DU CONSEIL Directive 98/83/EC (1998)[14] for drinking water. In 2000, it was predicted that by 2015, the water quality will be in good condition.

In Brazil, the National Environment Council (CONAMA)[15] in 2005 classified water bodies into different classes according to the quality required for their main uses, setting individual limits for each active ingredient. Similar to water for human consumption,[16] the limits established by CONAMA[15] for active ingredients of agrochemicals in different classes are milder and fail to include majority of those used in agriculture today. Besides being much more permissible,[16] the Brazilian legislation does not include the specification of some active ingredients of relevant agrochemicals, such as those classes of organophosphates and carbonates, which are widely used and highly toxic.

Accordingly, monitoring the quality of surface waters for the presence of agrochemicals in catchments may contribute to the discussions on the real impact of growing tobacco in Brazilian society.

The present study, therefore, aimed to investigate the presence of different active agrochemical ingredients in draw wells and creek

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water used for human and animal consumption, respectively, in a small catchment predominantly cropped to tobacco with a hilly landscape and shallow soils. This study hypothesized that the landscape, soil types, and crop production technology for tobacco production favor transport of agrochemicals to draw wells and creeks.

EXPERIMENTAL

Location and characterization of the catchment

The catchment discussed in this study is part of the Jacuí river basin and is located in the Central Depression of Rio Grande do Sul (Figure 1) in the municipality of Agudo, southern Brazil. The soils are classified as Entisols and Chernosols. The area is approximately 4.8 km^2, characterized by hilly relief and varying altitude, ranging from 120 to 480 m. The climate in the region is humid subtropical (Cfa type), according to the Köppen classification, with an average temperature greater than 22 °C in the hottest month and between −3 and 18 °C in the coldest. Rainfall is well distributed, ranging from 1,300 to 1,800 mm yearly. The natural vegetation belongs to the Atlantic Forest biome, and the remaining forested areas are mainly located in escarpments and close to drainage divides. The primary crop is tobacco, with most farms smaller than 10 ha.

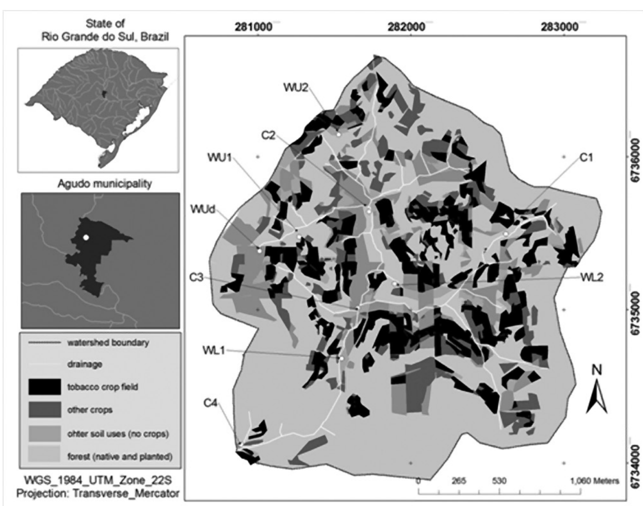

Figure 1. *Arroyo Lino headwater catchment with indications of land use, drainage system, and sampling sites. The drainage system, including all aspects of the first, second, and third order basins, is represented by solid lines, while the dotted line shows the boundary of the small basin. The sites in white represent water sampling locations used for the analysis. C1 to C4 are sampling sites along the creek under different landscape conditions, distances from tobacco to sampling sites, proportion of area under different land use, and the presence of vegetal cover at each sampling sites. WUd, WU1, and WU2 are draw wells on the upper slope, and WL1 and WL2 are draw wells on the lower slope, where "d" is a protected drainage, "1" is an open-pit draw well with no physical protection, and "2" is an open-pitt draw well with rudimentary physical protection*

The delineation of the catchment, streams, land use, and other topographical and landscape features was based on aerial photos and traverse Global Positioning System surveys. The maps were generated by a geo-referenced information system using the software ArcView 3D. Areas with different uses were identified and classified as forest (native forest and reforestation), tobacco crops, annual crops, and other crops. The total area (ha), the relative area (%) with the presence of crops and permanent vegetal cover, the average distance (m) between the nearest tobacco crops and collection sites, and the distance (m) between the nearest tobacco crops and nine sites to collect water along the catchment stream are listed in Table 1, adapted from Bortoluzzi *et al*.[6]

Sampling and analysis

Water samples were collected as described by Bortoluzzi *et al*.[7] Four creek sites (C1, C2, C3, and C4) were selected by considering the topography, the distance of tobacco crops to the collection site, the proportion of area with different land uses, and the presence of vegetal cover around the collection site. Five water sources for human consumption (draw wells) were chosen with reference to their location (three in the upper slope – WUd, WU1, and WU2; two in the lower slope – WL1 and WL2) and the physical protection system (protected drainage well- Wud; two unprotected – WU1 and WL2; and two with rudimentary physical protection – WU2 and WL2) (Figure 1). Samples were collected three times between 2005 and 2006. The first sampling was in August 2005, following tobacco transplanting. The second sampling was in January 2006, during the abscission stage. The third sampling was after the harvest in March 2006.

Prior to collection of water samples, glass bottles were decontaminated with a suitable cleaning solution, rinsed with methanol, and dried in the oven at 105 °C. Immediately after collection, the samples were packed in a cooler with ice and transported to the laboratory.

Samples were analyzed for the presence of active ingredients chlorpyrifos, flumetralin, and iprodione by gas chromatography with electron capture detection (GC-ECD), and for the presence of imidacloprid, atrazine, simazine, and clomazone by high-performance liquid chromatography with spectrophotometric detection in ultraviolet (HPLC-UV). Water samples were concentrated with cartridges of solid phase extraction containing 500 mg of C_{18} resin, followed by elution with a suitable solvent (methanol and ethyl acetate for analysis by HPLC-UV and GC-ECD, respectively) and then chromatographic determination. Detection limits in surface waters were 0.04 μg L^{-1} for imidacloprid; 0.08 μg L^{-1} for atrazine, simazine, and clomazone; 0.003 μg L^{-1} for flumetralin and chlorpyrifos; and 0.03 μg L^{-1} for iprodione.

RESULTS AND DISCUSSION

Six of the seven active ingredients studied were detected during the three sampling periods (Table 2). Flumetralin, an active ingredient and a sucker suppressor normally applied to tobacco when ripening, was not detected in any samples. However, iprodione was detected at a high concentration, with a value of 27.14 μg L^{-1} in only one site in the stream (C2), soon after transplanting, probably indicating point source contamination for this fungicide. This site is located below a small waterfall in a paddock. The creek course is parallel to the road and has no significant protection from riparian vegetation, only a few trees and many rocks. Near to this site, there were three sites for the production of tobacco seedlings using a floating system, which were coincidentally emptied immediately after transplanting the seedlings in the field. During the tobacco seedling production, various agrochemicals are applied, in particular iprodione for the prevention and control of fungal diseases. Runoff and seepages from the seedling production sites and surroundings and final dumping after seedling removal for planting in the field directly enter water bodies.

Chlorpyrifos was detected in all water samples after transplanting tobacco seedlings. According to the European Community Pollutants Classification, this active ingredient was classified as a "Priority Hazardous Substance" and that, if detected, it should be rendered attention without delay. The highest concentrations of this active ingredient were detected in water at upstream sites of the creek channel with values of 0.13, 0.1, and 0.11 μg L^{-1} for C1, C2, and

Table 1. Total area (ha), relative area (%) with tobacco crop and permanent soil cover, the average distance between tobacco and the nearest sampling site, and the distance between tobacco and the nearest stream of the nine water sampling sites in the studied catchment

Catchment characteristics	Creek (C)				Well (W)				
	C1	C2	C3	C4	WUd	WU1	WU2	WL1	WL2
Total area contributing runoff water, ha	23.5	122.2	332.3	480	4.6	3.2	1.7	3.6	7.5
Permanent vegetation cover, ha	17.6	85.7	214.1	336.1	4.3	2.4	1.1	1.6	3.7
Permanent vegetation cover (PVC), %	75	70	64	57.8	93	75	65	44	49
a) Forest, % of PVC	97	75	78	82	65	83	55	100	79
b) Pasture, % PVC	0	16	14	11	1	0	9	0	16
c) Reforestation (eucalyptus), % of PVC	3	9	8	7	44	17	36	0	5
Area with tobacco, ha	3.4	14.3	67.8	80.4	0	0.5	0.2	1.7	3.5
Area with tobacco, %	15	12	20	17	0	16	12	47	47
Other uses, ha	2.5	22.2	50.4	63.5	0.3	0.3	0.4	0.3	0.3
Average distance between tobacco fields to the nearest sampling site, m	63	42	32	10	-	60	35	1	40
Average distance between tobacco field to the nearest creek, m	1	10	1	10	-	-	-	-	-

C1 to C4 are sampling sites along the creek under different landscape conditions, distances from tobacco to sampling sites, proportion of area under different land uses, and the presence of vegetal cover at each sampling sites. WUd, WU1, and WU2 are draw wells at upper slope, and WL1 and WL2 are draw wells at lower slope, where "d" is a protected drainage, "1" is an open-pit draw well with no physical protection, and "2" is an open-pitt draw well with rudimentary physical protection. Table adapted from Bortoluzzi *et al.* [6]
-: not applicable.

C3, respectively, as shown in Table 2. Chlorpyrifos is still used to control insects at low cost, even if its use is restricted to households. It is a systemic insecticide applied to seedlings; however, its presence in water was detected in three samples (C2, WL1, and WL2) at the time of leaf ripening and after harvesting (C1), five and seven months after treatment (Table 2). Bortoluzzi *et al.*[7] detected the presence of chlorpyrifos within four months after transplanting in three small basins with similar intensive cultivation of tobacco.

Imidacloprid was detected after transplanting tobacco seedlings in R4 and after harvesting in WUD sites, demonstrating its persistence in the environment. Imidacloprid is a systemic insecticide normally used as a replacement for chlorpyrifos. This agrochemical has a low adsorption coefficient to soil organic colloids (K_{OC} of only 3.71 mL g^{-1}, compared to more than 8.151 mL g^{-1} in chlorpyrifos or 100 mL g^{-1} in atrazine), high solubility in water (610 mg L^{-1}, compared to only 1.05 mg L^{-1} in chlorpyrifos or 35 mg L^{-1} of atrazine), and a half-life that ranges from 48 to 120 days, similar to chlorpyrifos. Bortoluzzi *et al.*[6] also detected imidalcloprid in this small catchment in 2001, in higher concentrations at several sampling sites.

Atrazine, simazine, and clomazone (Table 2), all weed control chemicals, varied in their level of contamination. Atrazine was found in one draw well used for human water consumption during the first two sampling periods. Simazine was detected during the abscission stage in three out of the nine sampling sites: two from the creek and one from a draw well. After harvesting tobacco, it was also detected in samples collected at C1 and WU1 sites. Clomazone was present in the stream water at site C1 during the first sampling period, in two water samples (C1 and WL2) at the time of tobacco abscission stage, and in five samples after harvest (two from the creek and the other three from draw wells). This herbicide is highly soluble in water (1102 mg L^{-1}), and the half-life in the field varies with soil type, which can be up to 12 weeks, especially in highly contaminated soils.[17]

The use of herbicides for weed control in tobacco farming is a recent consequence of the decline in labor availability for manually controlling weed growth owing to rural exodus. Weed and pest control by chemical methods are now rampant. For example, in most tobacco farms, farmers apply glyphosate after harvest and directly sow corn on the ridges. The control of weeds during corn growth similarly requires herbicides, especially to control grasses. Continuous application of these chemicals posed a threat to the environment, as is evident by the detection of these active ingredients in surface waters of the region. This problem is intensifying owing to (i) recommendation of these and other active ingredients for the cultivation of tobacco; (ii) the shortened life cycle of new tobacco varieties; and (iii) the incentive for growing corn after tobacco.

The presence of these herbicides in waters of developed countries is the rule rather than exception, as demonstrated by the French Ministry of Health.[18] In 1991, out of 6773 water samples collected in several regions in France, more than 0.1 μg L^{-1} atrazine was detected in 37% of nontreated samples, and 25% of those distributed to consumers; 1.2% of the population received water containing more than 2.0 μg L^{-1} of this herbicide. Similarly in France, Barriuso *et al.*[19] reported that severe pollution by various agrochemicals at widely varied concentrations during the course of the year and between years depends on the date of application and the phenomena of surface runoff and leaching. A similar observation was made by Holman *et al.*[20] in North Carolina, USA, a chief tobacco producing region, who detected eleven different agrochemicals in twelve local surface water sources used for human consumption. However, agrochemicals were more frequently detected in small water basins because of short distances from crop fields. Atrazine was present in 45% of samples, and in certain seasons, the concentration was greater than 3.0 μg L^{-1}.

The widespread contamination of water bodies (creeks and draw wells) in the studied catchment is consistent with the degree of agrochemical application in tobacco and subsequently in corn directly sown in succession. The presence of agrochemicals was immediately detected in draw well water after the first rain. After transplanting, all samples from all sites were polluted by chlorpyrifos, and one particular creek site had a high concentration of iprodione. These agrochemicals were prevalent because both are widely used during this phase of tobacco cultivation. During the tobacco abscission stage, almost all sampling sites were contaminated by herbicides atrazine, simazine, and clomazone. However, chlorpyrifos was only detected in the third sampled period, which is coincidentally at the lower slope of the landscape that accumulates sediment from erosion along landscape in all catchments.[21] This may also be attributed to

Table 2. Agrochemical concentrations in water at nine sampling sites and three sampling periods during the tobacco growth cycle in the studied catchment

Agrochemical	Sampling sites								
	Creek (C)				Well (W)				
	C1	C2	C3	C4	WUd	WU1	WU2	WL1	WL2
	Concentration (μg L⁻¹)								
After tobacco transplanting									
Imidacloprid	-	-	-	0.13	-	-	-	-	-
Atrazine	-	-	-	-	-	-	-	-	0.29
Simazine	-	-	-	-	-	-	-	-	-
Clomazone	0.48	-	-	-	-	-	-	-	-
Chlorpyrifos	0.13	0.1	0.11	0.07	0.07	0.09	0.07	0.06	0.09
Flumetralin	-	-	-	-	-	-	-	-	-
Iprodione	-	27,14	-	-	-	-	-	-	-
At abscission stage									
Imidacloprid	-	-	-	-	-	-	-	-	-
Atrazine	-	-	-	-	-	0.22	-	-	-
Simazine	0.84	-	-.	0.59	-	-	-	0.44	-
Clomazone	0.34	-	-	-	-	-	-	-	0.38
Chlorpyrifos	-	0.25	-	-	-	-	-	0.06	0.07
Flumetralin	-	-	-	-	-	-	-	-	-
Iprodione	-	-	-	-	-	-	-	-	-
After harvest									
Imidacloprid	-	-	-	-	1.66	-	-	-	-
Atrazine	-	-	-	-	-	-	-	-	-
Simazine	0.76	-	-	-	-	1.10	-	-	-
Clomazone	2.90	2.50	-	-	3.89	-	0.73	-	4.40
Chlorpyrifos	0.04	-	-	-	-	-	-	-	-
Flumetralin	-	-	-	-	-	-	-	-	-
Iprodione	-	-	-	-	-	-	-	-	-

C1 to C4 are sampling sites along the creek under different landscape conditions, distances from tobacco to sampling sites, proportion of area under different land uses, and the presence of vegetal cover at each sampling sites. WUd, WU1, and WU2 are draw wells at upper slope, and WL1 and WL2 are draw wells at lower slope, where "d" is a protected drainage, "1" is an open-pit draw well with no physical protection, and "2" is an open-pit draw well with rudimentary physical protection.
-: Not detected. The detection limit for imidacloprid is 0.04 μg L⁻¹; atrazine, simazine, and clomazone is 0.08 μg L⁻¹; flumetralin and chlorpyrifos is 0.003 μg L⁻¹; and iprodione is 0.03 μg L⁻¹.

the high adsorption coefficient to organic colloids. Finally, during tobacco harvest, clomazone (a herbicide specifically formulated for weed control in tobacco fields) was detected in five out of nine sites sampled.

The presence of multiple active ingredients of agrochemicals in water bodies is a strong indicator of the inappropriate use and even exploitation of natural resources or by the problem of fragmentation of small farms, either by encouraging tobacco production, without due concern for the quality of life for the farmers and the environment. The contamination process occurs during rainfall events during which agrochemicals are carried to water bodies through surface runoff, and organic molecules and ions are transported by subsurface flow.

The configuration of the catchment sites for land use planning can be achieved by considering local conditions. Moreover, road allocation is another important factor contributing 28.1% to runoff and erosion losses.[22] Conservation of farming practices reduces the volume of water draining into streams, flood peak, production, and transport of sediments, and alters the relative contribution of sediment sources, as observed by Minella *et al*.[3] in a small catchment with similar features. These authors observed a reduction in soil erosion in a crop field with minimum soil disturbance, reduction in sediments from unpaved rural roads, and an increased erosion of stream channels and associated alluvial banks. There is a need, therefore, to maintain vegetal cover on slopes and river banks to preserve surface water quality.[6] For optimum infiltration and runoff, biopores must be sufficiently large, continuous, and open to the soil surface such that bulk density and porosity do not limit water dynamics during rainfall events.[23-25]

Although the investigated area represents a small percentage of the total catchment (Table 1), the pollution caused by an indiscriminate use of agrochemicals in tobacco production made water from the catchment unfit for human consumption.[14] As such, the use of water from these creeks and draw wells, independent of distance from tobacco fields and physical protection, poses a health threat to not only the rural dwellers but also the urban population in general. As observed by Becker *et al*.,[26] water from this catchment caused changes in the metabolic parameters of silver catfish juveniles. Therefore, awareness with regard to indiscriminate use of agrochemicals should be made a primary concern. In addition, governmental agencies should enforce rigid laws against abuse and indiscriminate acquisition and use of agrochemicals, with adequate monitoring and control measures.

CONCLUSION

Water in the catchment was contaminated to varying degrees by agrochemicals with active ingredients imidacloprid, atrazine, clomazone, iprodione, and chlorpyrifos detected in the creek and draw wells in the small headwater catchment predominantly under tobacco cultivation.

The widespread occurrence of agrochemicals in water was a consequence of the indiscriminate use of prophylactic treatment for tobacco cultivation, along with the lack of landscape and environmental planning.

Allocation of tobacco fields far from creeks or draw wells or physical protection of wells was not sufficient to avoid water contamination by agrochemicals used in tobacco production on shallow soils and steep slopes.

The use of water and its extraction in the catchment posed significant health threats. Therefore, it is essential that governmental agencies enforce rigid laws against abuse and indiscriminate acquisition and use agrochemicals, with adequate monitoring and control measures.

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