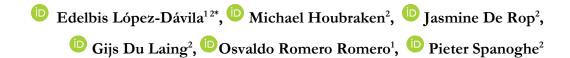
Management and environmental sustainability

Scientific and technological research article

Toxicological and ecotoxicological pressure assessment on the use of synthetic pesticides in Sancti Spíritus, Cuba



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> Subject editor: Felipe Borrero Echeverry (Corporación Colombiana de Investigación Agropecuaria [AGROSAVIA]).

> > Received: July 19, 2019

Accepted: December 15, 2020

Published: May 07, 2021

How to cite this article: López-Dávila, E., Houbraken, M., De Rop, J., Du Laing, G., Romero, O., & Spanoghe, P. (2021). Toxicological and ecotoxicological pressure assessment on the use of synthetic pesticides in Sancti Spíritus, Cuba. Ciencia y Tecnología Agropecuaria, 22(2), e1520. https://doi.org/10.21930/rcta.vol22_num2_art:1520



Abstract

A study to quantify the toxicity and ecotoxicological pressure of pesticides in the Sancti Spíritus province, Cuba, was carried out between 2011 and 2014. A longitudinal descriptive work was designed for the study period to identify potential risks to the environment and also to human health associated with the use of pesticides in the country. The Spread Equivalents (Σ Seq) and Pesticide Occupational and Environmental Risk (POCER) indicators, as well as the Toxic Load (TL) methodology of Instituto Cubano de Sanidad Vegetal, were used to determine the toxicity and ecotoxicity of pesticide use. One hundred and twentyfour active ingredients corresponding to 62 chemical families were applied in the province during the study period. Organophosphates, triazoles, sulfonylurea, pyrethroids, inorganic compounds (such as carbamates, dithiocarbamates, neonicotinoids, aryloxyphenoxypropionates, copper), and organochlorines predominated due to their use frequency. The use of toxic pesticides, and the lack of personal protection equipment, among others, made workers, residents, and applicators the toxicological modules with the highest risk of exposure. On the other hand, aquatic organisms, and the persistence of the pesticides in the soil and in groundwater, are the modules with the highest ecotoxicological pressure. By using the POCER and Σ Seq indicators, a more accurate toxicity and ecotoxicity assessment for certain pesticides can be performed in Cuba, in comparison to the one obtained when using only the TL equation currently employed in the country. In addition, substituting the most toxic pesticides (e.g., parathion, endosulfan, bifenthrin, copper oxychloride, mancozeb, paraquat, diquat, and ametryn) with less toxic ones (e.g., cypermethrin, tebuconazole, triadimenol, and bispyribac-sodium) could help reduce synthetic pesticide pressure on humans and the environment.

Keywords: ecotoxicity, occupational hazards, pesticide exposure, pesticide toxicity, toxicity

Evaluación de la presión toxicológica y ecotoxicológica del uso de plaguicidas sintéticos en Sancti Spíritus, Cuba

Resumen

Se realizó un estudio para cuantificar la toxicidad y la presión ecotoxicológica de los plaguicidas sintéticos en la provincia de Sancti Spíritus (Cuba) entre 2011 y 2014. Este trabajo puede ayudar a desarrollar políticas y prácticas de gestión para reducir los peligros del uso de plaguicidas sintéticos en el país. A través de un estudio longitudinal descriptivo, se identificaron los riesgos potenciales para el medioambiente y la salud humana asociados con el uso de plaguicidas. Para determinar la toxicidad y ecotoxicidad del uso de plaguicidas, se utilizaron los indicadores de aplicaciones equivalentes (Σ Seq) y de riesgos laborales y medioambientales (POCER, por su sigla en inglés), además de la metodología de carga tóxica (TL, por su sigla en inglés) del Instituto Cubano de Sanidad Vegetal. Durante el periodo de estudio, 124 ingredientes activos correspondientes a 62 familias químicas fueron aplicados. Por su frecuencia de uso, predominaron los organofosforados, triazoles, piretroides, compuestos inorgánicos (como el cobre), carbamatos, ditiocarbamatos, neonicotinoides, ariloxifenoxipropionato y organoclorados. El uso de plaguicidas tóxicos y la falta de equipos de protección personal, entre otros aspectos, hicieron que los trabajadores, los residentes y los aplicadores fueran los módulos humanos con el mayor riesgo de exposición. Por otro lado, los módulos de mayor presión ecotoxicológica son los organismos acuáticos, la persistencia en el suelo y el agua subterránea. Con el uso de los indicadores POCER y Σ Seq, se puede realizar una evaluación más precisa de la toxicidad y la ecotoxicidad en Cuba, en comparación con la realizada solo por la ecuación TL actualmente utilizada en el país. La sustitución de los plaguicidas más tóxicos (paratión, endosulfán, bifentrina, oxicloruro de cobre, mancozeb, paraquat, diquat y ametrina) por otros menos tóxicos (cipermetrina, tebuconazol, triadimenol y bispiribac-sodio) podría ayudar a reducir la presión de los plaguicidas sintéticos sobre los seres humanos y el medioambiente.

Palabras clave: ecotoxicidad, exposición a plaguicidas, riesgos ocupacionales, toxicidad, toxicidad de los plaguicidas

Introduction

The use of pesticides worldwide has become a basic need in various crops to ensure quantity and quality in crop production. Pesticides have been a solution in the fight against hunger and control many plant diseases that affect the breadbasket of humanity, allowing broad sectors of the population to access more quality food (Räsänen et al., 2015). There is, however, a tendency to increase its use argued in the pertinence of controlling diseases, insects, weeds, and other organisms that can interfere with crop production (Leyva et al., 2014). Although their use favors production processes, the inadequate use of synthetic pesticides, their application timing, and their use in crops in which they have not been registered, make these pesticides a potential risk to human health and the environment (Dugger-Webster & LePrevost, 2018; Mesnage et al., 2014).

The increased use of pesticides can cause specific side effects in humans (Vryzas, 2018). However, no pesticide lacks ecotoxicity; any can cause acute poisoning once they are absorbed and accumulated in organisms (De la Rosa Cruz et al., 2013), and chronic damage is the result of repeated exposure (Ventura et al., 2015). For example, there are reports of teratogenic, carcinogenic, and mutagenic diseases; damages to eyes, skin, and mucous membranes; neurotoxic damage; damage to the immune system and lungs; and infertility (López Dávila, Houbraken et al., 2020; Mwila et al., 2013; World Health Organization [WHO], 2010).

In Cuba, to increase the productivity of agricultural systems, technological packages have been introduced, including as its main component, the use of synthetic pesticides (Rosquete, 2011). In the province of Sancti Spíritus, agriculture is the leading economic sector. The need to increase yields of priority crops to reduce their imports led to utilizing synthetic pesticides (Damalas & Koutroubas, 2018).

The use of synthetic pesticides, mainly in fruits and vegetables, is a constant concern in the local population regarding the risk to human health and the environment, reflected in various journalistic reports. However, there are currently no scientific studies that evaluate this risk pressure.

Toxicity and ecotoxicity studies are useful in monitoring environmental quality (Moermond et al., 2016). Different methods and models have been developed and applied, such as the Dutch Pesticide Risk Indicator (Nationale Milieu Indicator NMI 3), the Danish Pesticide Load (PL) indicator, the German Pesticide Risk Indicator (SYNOPS), the Health Risk Indicator for Operators (IRSA), and the Toxicity Risk Indicator for the Environment (IRTE) (Kudsk et al., 2018; Oussama et al., 2015); moreover, various software or programs have also been used, such as JOVA (Tollefsen et al., 2016) and USEtox (Nordborg et al., 2017; Räsänen et al., 2013). Further, a method widely utilized is the Criteria for Reporting and Evaluating ecotoxicity Data (CRED) (Kase et al., 2016; Moermond et al., 2016).

Derived from simplified quantitative models, the Pesticide Occupational and Environmental Risk indicator (POCER) (Vercruysse & Steurbaut, 2002) and the indicator based on the sum of the annual spread equivalents (Σ Seq) (De Smet & Steurbaut, 2002) developed at Ghent University stand out as appropriate options for the Cuban context. POCER assesses the risk for a large number of environmental modules and biota, being one of the most dynamic and comprehensive models (Wustenberghs et al.,

2012). Four modules that evaluate the risk arising from occupational or other non-dietary exposure to agricultural pesticides, cover four categories of persons, including (1) risk to operators who apply the pesticides, (2) risk to workers who may be exposed through re-entry activities, such as harvest, (3) risk to residents, and (4) risk to bystanders who may be incidentally exposed during or after pesticide applications. Furthermore, six modules covering different effects and environmental compartments assess the risk to the environment. They include (1) persistence in the soil, (2) risk of groundwater contamination, (3) acute risk to aquatic organisms, (4) acute risk to birds, (5) acute risk to bees, and (6) acute risk to earthworms. The risk for each module is estimated utilizing risk indices (Vercruysse & Steurbaut, 2002).

 Σ Seq expresses the pressure on aquatic life produced by the use of pesticides (Fevery et al., 2015). This indicator has been employed since 1996 in the environmental policy of the Flemish Government (Belgium) for a regional pesticide use assessment (De Smet & Steurbaut, 2002). The use of each pesticide is weighted according to toxicity differences in aquatic organisms and the permanence time in the environment (De Smet et al., 2005).

In 1998, Cuba officially established its Environmental Law (González & Conill, 1999) regulating sustainable agriculture. Besides, in the 2007-2010 period, the Cuban Ministry of Science, Technology and Environment, established a national environmental strategy, in which by 2010, 80 % of the pest and disease control in the country must be done using natural products or biopesticides (Hernández Núñez & Pérez-Consuegra, 2012; Rosquete, 2011). However, up to date, there are no reports of compliance with this strategy. Similarly, there are no studies published in peer reviewed journals or national information articles on the level of pesticide use in this territory or the evaluation of the toxicity and ecotoxicity due to the use of pesticides; additionally, no indicators measuring these parameters were defined.

The constant concern for human health and the environment in the local population was the basis for this study using the POCER and Σ Seq indicators to evaluate the toxicity and ecotoxicity instead of the level of toxic load (TL), according to the methodology of the Cuban Plant Health Institute. The goal is to determine the risks to human and environmental health that arise from utilizing synthetic pesticides in the province of Sancti Spíritus. The study covers the period from 2011 to 2014, and aims to identify the main pesticides causing pressure (unfortunately, usage data from more recent years were not available). This will help develop policies and management practices to reduce pesticide hazards by reducing the use of pesticides that have the highest pressure on humans and the environment.

Materials and methods

The province of Sancti Spíritus, constituted by eight municipalities, is located about 400 km southeast of Havana City. It is one of the central provinces of the country and has a tropical climate. It is characterized by having an average annual temperature of 25.3 °C, average annual precipitation of 1,374.5 mm, and 78 % relative humidity (Oficina Nacional de Estadística e Información [ONEI], 2020). Sancti Spíritus has diverse agriculture, but the main crops harvested, in order of importance, are rice, tobacco, beans, roots

and tubers (e.g., sweet potato, and potato), sugarcane, vegetables (e.g., tomato, cucumber, and sweet pepper), onions, garlic, maize, and various fruits (e.g., papaya, guava, banana).

Operationalization of variables

A database with all the pesticides used in the agricultural activities registered in the accounting system of the Provincial Department of Phytosanitary Protection during the study period was collected. The pesticide use data per product were compiled according to their chemical family and biological function (per crop and year), and their toxicological reference values in humans, and terrestrial, and aquatic organisms. The hazard classification criteria of the World Health Organization (WHO, 2010) were used.

Toxic load assessment

In the Cuban agricultural context, the "Toxic Pollutant Load" indicator or simply "Toxic Load" (TL, in kg or L of the active ingredient/ha) established by the Plant Protection Department belonging to the Ministry of Agriculture of Cuba (Díaz, 2009) gives a measure of the general load on the environment resulting from the utilization of pesticides without making a distinction, and based on ecotoxic properties and differing from compound to compound. Equation 1 is used to calculate the TL in priority crops. A similar equation was employed to evaluate drinking water contamination by using pesticides in Vietnam (Chau et al., 2015).

$$TL = D * a. i. \% * NA$$

Where:

TL is the toxic load (kg or L of the active ingredient/ha), D is the dosage (kg of the commercial product/ha), *a.i.*% is the percentage of the active ingredient, and NA is the number of applications (1). This indicator was calculated for each active ingredient per crop and year, showing the total sum in each case.

Toxicity and ecotoxicity assessment

When utilizing POCER, risk indices (RIs) for human health and the environment are calculated as the predicted environmental concentration (PEC) ratio in relation to a toxicological reference value. These are well described in the study by Vercruysse and Steurbaut (2002). After assessing the relevant risk parameters, the POCER calculations can be carried out by inserting the parameters (equation 2-11) into the model and generating ten values, one for each human and environmental compartment (Claeys et al., 2005). The RI values calculated are log-transformed, and benchmarks between a lower and an upper limit were set. This generated a dimensionless value between 0 and 1 for each compartment, where 0 indicates a low risk of exposure, and 1 indicates a high risk of exposure (Vercruysse & Steurbaut, 2002).

Equation 1

Like Vercruysse and Steurbaut (2002) described, in POCER, the total risk of exposure for humans and the environment is calculated by adding the values of the different components, assuming that all components are equally important. The risk for humans is, thus, the sum of the risk for applicators, workers, secondarily exposed residents, and bystanders. The risk for the environment is calculated as the sum of risks, such as persistence, leaching to groundwater, aquatic organisms, birds, earthworms, and bees. The calculation formulas for each module are described in equations 2 to 11:

Operators:
$$RI_{operator} = \frac{IE_{operator}}{AOEL}$$
 Equation 2

IE is internal exposure during mixing/loading and application (mg/kg/day), and *AOEL* is an acceptable operator exposure level (mg/kg/day).

Worker/Re-entry workers:
$$RI_{worker} = \frac{DE*Ab_{de}}{AOEL}$$
 Equation 3

DE is dermal exposure (mg/kg/day), and Ab_{de} is dermal absorption (-)

Bystanders:
$$RI_{bystander} = \frac{DE * Ab_{de} + I * Ab_i}{BW * AOEL}$$
 Equation 4

I is inhalation exposure (mg/kg/day), Abi is inhalation absorption (-), and BW is body weight.

Residents:
$$RI_{resident} = \frac{DE * Ab_{de} + I * Ab_i}{AOEL}$$
 Equation 5

Aquatic organisms: $RQ_{aquatic organisms} = \frac{PEC_{aquatic organisms}}{minimum(norm_{aquatic organisms})}$ Equation 6

 $PEC_{aquatic organisms}$ is the predicted concentration in surface water (g/L), *minimum (normaquatic organisms)* is the lowest toxicity value of three groups of organisms (fish, *Daphnia* spp., and crustaceans) (g/L).

The lowest of the following three quotients is used as the *minimum* (*norm*_{aquatic organisms}): the lethal concentration required to kill 50 % of the population (LC₅₀) for fish/100, the half-maximal lethal concentration (EC₅₀) for Daphnia/100, and the no-observed-effect-concentration (NOEC) for algae/10.

Birds:
$$RI_{bird} = \frac{PEC_{bird}*10}{LD_{50}*BW}$$
 Equation 7

 PEC_{bind} is the estimated total daily pesticide intake (mg/day), LD_{50} is the lethal dose for 50 % of the population (mg/kg/day), and *BW* is body weight (default = 0.01 kg). Factor 10 is the criterion set by the Uniform Principles of the Commission of the European Communities established in 1994.

Bees:
$$RI_{bee} = \frac{AD}{LD_{50}*50}$$
 Equation 8

Cienc. Tecnol. Agropecuaria, 22(2): e1520 DOI: https://doi.org/10.21930/rcta.vol22_num2_art:1520 AD is the application dose (g/ha), and LD_{50} is the lethal dose for 50 % of the population (μ g/bee).

Earthworms:
$$RI_{earthworm} = \frac{PEC_{soil}*10}{LC_{50}}$$
 Equation 9

 PEC_{soil} is the estimated concentration in the soil (mg/kg), and LC_{50} is the lethal concentration for 50 % of the population (mg/kg).

Persistence in the soil:
$$RI_{persistence} = 10^{\left(\frac{DT_{50}}{90}-1\right)*2}$$
 Equation 10

 DT_{50} is the disappearance time for the first 50 % of the pesticide (days).

Groundwater:
$$RI_{groundwater} = \frac{PCE_{groundwater}}{0.1}$$
 Equation 11

 $PEC_{groundwater}$ is the predicted concentration in the groundwater (μ g/L), and 0.1 is the European drinking-water limit (μ g/L).

Based on the fact that only the total amount of pesticides and the area cultivated for each crop is reported per crop and year, the amount of each active ingredient was divided by the cultivated area, to get a dosage value per hectare (application rate). Finally, in each case (crop and year), the sum of the final values in POCER was multiplied by the total number of hectares. In this way, the crop with the highest impact can be identified at the territorial, human health, and environmental levels.

For the toxicity modules, a group of assumptions was made. These were based on the results of a farmer survey study (López Dávila, Houbraken et al., 2020). First, $IE_{operator}$ in equation 2, is strongly influenced by the use of protective clothing during mixing, loading, and spraying pesticides; for rice, aerial spraying was considered, and the use of a tractor was considered for sugarcane. Second, for re-entry workers, no protective clothing was considered. For the *resident* module, two issues were not considered; a buffer zone, as their households are located within the farm and very close to the crops, and a drift reduction due to the use of a classic nozzle.

 Σ Seq is an ecotoxicity indicator that calculates the pressure from using pesticides for agricultural and non-agricultural purposes (vector control) in aquatic organisms (De Smet & Steurbaut, 2002; Fevery et al., 2015). This indicator was included in this study, as POCER considers that the drift of pesticides mainly causes the exposure of aquatic organisms. Further, it does not consider their ability to persist in the soil, and therefore, through surface runoff and leaching, the pesticides end in water bodies. These are parameters more in line with the current Cuban agricultural context. Furthermore, the variable *minimum* (*norm*aquatic organisms) is restricted to only three ecotoxicity values (LC₅₀ for fish, EC₅₀ for Daphnia, and NOEC for algae), while the maximum allowable concentration (MAC) is established based on six different ecotoxicity values, generating more accurate results. It is calculated using the following equation 12:

$$\sum Seq = \frac{E*DT_{50}}{MAC}$$

Equation 12

Cienc. Tecnol. Agropecuaria, 22(2): e1520 DOI: https://doi.org/10.21930/rcta.vol22_num2_art:1520 Where *E* is the annual use of pesticides (kg of a.i./year), DT_{50} is the degradation time of 50 % of the a.i. in the soil (years), and *MAC* is the maximum allowable concentration for aquatic life (mg/L)

The MAC values are calculated by dividing the lowest toxicity value (representative for aquatic organisms, i.e., the acute or chronic toxicity up to three trophic levels: $EC_{50algae}$, $NOEC_{algae}$, $LC_{50crustaceans}$, $NOEC_{crustaceans}$, LC_{50fish} , and $NOEC_{fish}$) by safety factor "10", as done by Fevery et al. (2015).

Data processing procedures

The data for all the variables were summarized and tabulated. Then, a group called "vegetables" was created where tomatoes, onions, garlic, and plants belonging to the Cucurbitaceae family, among others, were included. The group called "grains," includes beans and corn. The "roots and tubers" group comprises sweet potato, malanga or taro, and potato. Finally, in the "fruits" group, coffee and banana are included. The Statistical Package for Social Sciences (SPSS) program (version 20) was employed. Pearson and Spearman's rank correlations (p < 0.01 and p < 0.05) were used to evaluate the parametric and non-parametric correlations between TL values with the POCER and Σ Seq indicators.

Results and discussion

Pesticide use in the Sancti Spíritus province in the years 2011-2014

In total, 124 active ingredients (40 fungicides, 42 herbicides, and 42 insecticides) were used in the agricultural activities of the Sancti Spíritus province during the study period. These active ingredients correspond to 62 chemical families. A similar number (69 chemical families) were applied in other provinces of equal agricultural importance in the country, according to Hernández Núñez and Pérez-Consuegra (2012). The predominant chemical families are organophosphates, triazoles, sulfonylurea, pyrethroids, inorganic compounds (e.g., copper oxychloride), carbamates, dithiocarbamates, neonicotinoids, aryloxyphenoxypropionate, and organochlorines.

The following six active ingredients (a.i.), methyl parathion, methamidophos, methiocarb, methomyl, 1,3dichloropropene, and endosulfan, are classified by the World Health Organization (WHO, 2010) as extremely toxic (Ia) and highly toxic (Ib). Moreover, 28 other compounds are in the category of moderately toxic (II). Fifty-nine percent of the products show some degree of toxicity against bees; this constitutes a significant environmental risk factor, as it can lead to declines in bee populations and the ecosystem services they perform (Fevery et al., 2016; Hladik et al., 2016). It was also found that 80 % of the pesticides are, to some degree, toxic to fish.

During the study period, the use of synthetic pesticides in the Sancti Spíritus province showed a reasonably constant use, as seen in figure 1 (except for 2014). These values contrast with the progressive

reduction strategy of the crop protection policy promoted by the Cuban Ministry of Science Technology and Environment. Its aim is to reduce the toxic pollutant load and its potential side effects on the environment and human health. At the national level in Cuba, similar results are observed (ONEI, 2020).

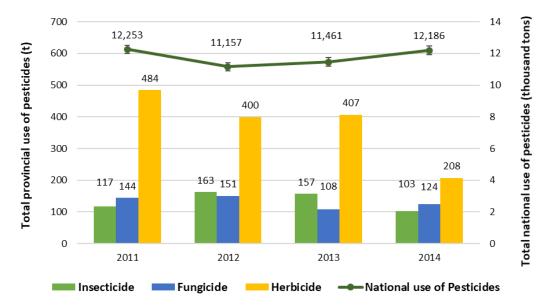


Figure 1. Pesticide use in the Sancti Spíritus province per year. Results were obtained from the database of pesticides assigned to the Sancti Spíritus province from 2011 to 2014.

Source: Elaborated by the authors

Herbicides (58 %) are the most predominantly used pesticides (figure 1). Similar results have been reported by researchers in other developing countries such as India and Ghana (Imoro et al., 2019; Sharma, Kumar, Thukral et al., 2019; Sharma, Kumar, Shahzad et al., 2019; Wumbei et al., 2019). These are followed by insecticides (21 %) and fungicides (21 %). Results are because large land extensions have been used for crops such as sugarcane, rice, and fruit trees, including banana, which requires large volumes of herbicides to control weeds. It is important to note that potato, belonging to the "roots and tubers" group was planted only in 2011 and 2012, because the agricultural strategy decision-makers of the country decided to stop planting potatoes in the province. Despite this fact, potatoes were included in this study because they represented 73 % and 33 %, respectively, of the total amount of pesticide used in 2011 and 2012 by the "roots and tubers" group. No data could be found for sugarcane in 2014.

The active ingredient primarily used in the study period was ametryn (215 t, 19 % of the total active ingredients utilized, and 30 % of the total herbicide employed), followed by 2,4-D amine salt (165 t, 14 % of the total active ingredient used, and 23 % of the total herbicide utilized), and mancozeb (100 t, 8 % from the total active ingredient employed, and 36 % of the total fungicide used). The results are in line

with the main crops that are cultivated in the territory, according to the national strategy (rice, tobacco, vegetables, grains, sugarcane, and fruits) (ONEI, 2019).

Evaluation of the toxic and ecotoxic load in the Sancti Spíritus province

Studying pesticide pressure by calculating the toxic and ecotoxic load is vital to understanding the environmental and human health risks. Once the more critical molecules are identified, actions can be proposed to eliminate or substitute them with less toxic compounds, thus, reducing their impact on the environment and human health.

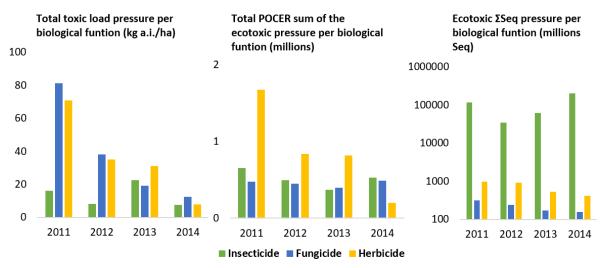


Figure 2. Ecotoxicity values during the study period in the province per evaluated indicator based on the amount of active ingredients used per biological family.

Source: Elaborated by the authors

Although herbicides were the most used pesticides in the province (figure 1), their pressure on human health and the environment was not always the highest (as seen in figure 2). The TL values were different between biological functions. In 2011, for example, the TL of fungicides was significantly higher than the TL of herbicides due to potato cultivation, reporting the highest kg of a.i. per treated area (73.2 kg a.i./ha) ratio between eight (tobacco 8.8 kg a.i./ ha) to 490 (corn 0.15 kg a.i./ha) times higher compared to other crops. Fungicides (e.g., mancozeb, chlorothalonil, and copper oxychloride) represented 48 % of the a.i. used this same year. Herbicides (e.g., ametryn and glyphosate) were in second place with 42 %. In 2012, the ratio in potatoes decreased to 14.8 kg a.i. per hectare, and in later years, potato was not planted. Another observation is the TL trend that decreased over time, although the consumption of a.i. remained fairly constant during the study period (figure 1). This is because the treated crop areas increased from 82.9 to 103.8 thousand ha, except for sugarcane (from 58.7 to 28.3 thousand ha), thus, causing a general progressive decrease in the ratio (kg of a.i. used per treated area).

According to equation 1, the TL only expresses the amount of a.i. (kg or L) applied per hectare. However, the particular toxicities for human health (NOAEL and AOEL, among others) and the environment (DT₅₀, EC₅₀, NOEC, and LC₅₀ values) are not considered, and hence, the pressure of pesticide use is not very accurate. A simple substitution by another pesticide with a lower amount of a.i. will decrease in the TL. However, if this new pesticide has higher toxicity or ecotoxicity, or both, the pressure will increase. Clear examples are shown by Wustenberghs et al. (2018). These authors found that the TL is a mere volume indicator and not a load indicator as meant by Kudsk et al. (2018). It has long and widely been acknowledged that quantities are not adequate proxies for assessing pesticide risk as Barnard et al. (1997), Stenrod et al. (2008), and Tzilivakis et al. (2004), all cited by Wustenberghs et al. (2012), reported.

Also, the POCER herbicide pressure in 2011 was quantitatively higher compared to the rest of the years (figure 2). This is due to the cultivation of sugarcane, which has the largest treated area of all crops (41 % in 2011), declining to about half in 2012 (23 %) and 2013 (24 %). Sugarcane represented 75 % of the total POCER herbicide pressure for 2011, being paraquat, hexazinone, and diuron the main a.i. due to their toxicity. In this work, the Σ Seq for insecticides increased gradually due to endosulfan use, which is the a.i. with the higher Seq-factor (DT₅₀/MAC = 1.2 * 108), i.e., 71 times higher than paraquat, the second most ecotoxic a.i. (DT₅₀/MAC = 1.7 * 106). Endosulfan was used in corn (10 kg), beans (140 kg), and onion (280 kg) in 2011. The next year, it was only utilized in onion (296 kg). In 2013, it was employed in tomato (175 kg) and onion (348 kg); in 2014, it was also applied in tomato (280 kg) and onion (925 kg), being the latter the one that exerts the highest ecotoxic pressure on aquatic organisms. Unlike TL and POCER that decreased over the years, Σ Seq increased; its values are directly related to endosulfan use, which increased over the years.

With the use of the POCER and Σ Seq indicators, considering the effect on both terrestrial and aquatic organisms, the pressure caused by a specific a.i. can be more accurately assessed (Fevery et al., 2015, 2016; Houbraken et al., 2016). Therefore, in both Σ Seq and POCER, insecticides exert significant pressure with marked differences in the case of the Σ Seq indicator. The a.i. of the used insecticides negatively impacts the environment and human health.

The trend over four years for the TL, Σ Seq, and POCER values per group of crops can be observed in table 1. A positive Pearson's correlation is found between the POCER parameters sum of toxic and sum of ecotoxic values with dependency on the year (r = 0.816, p < 0.01). In further analyses between the evaluated indicators, a correlation between the TL and POCER sum of toxic values module (r = 0.613, p < 0.01) was found, as well as between the TL and the POCER sum of ecotoxic values module (r = 0.468, p < 0.05). Moreover, no correlation was observed between the TL and Σ Seq indicators (r = 0.110, p = 0.585).

Table 1. Sum of toxic and ecotoxic values assessed from the main provincial crop groups per year. The results are based on the database on pesticides used in the Sancti Spíritus province, Cuba, for the 2011-2014 period.

n n		kg a.l.	Area (ha)	Toxic Load (kg a.i./ha)	ΣSeq (Seq)	POCER											
11 13 53 53 53 53 53 54 53 <	Year					Aquatic Organisms	Ground Water	Persistence	Earthworms	Bees	Birds	Sum Ecotoxic	Applicator	Re-entry Worker	Bystander	Resident	Sum Toxic
12100 254-04 254-04 244-04 274-04 124-04 124-04 124-04 <td></td> <td colspan="14">Sugarcane</td> <td></td>		Sugarcane															
1 2 2 2 3 2 4 2 4 1 4 1 4 1 4 1 4 1 4 1 4 1 4 1	2011	1.3E+05	5.9E+04	2.4E+01	2.5E+08	6.0E+05	2.1E+05	1.8E+05	3.5E+01	4.2E+03	1.4E+02	9.9E+05	6.3E+03	2.5E+05	3.5E+02	7.2E+03	2.7E+05
1mm 1mm 2mm 2mm <td>2012</td> <td>1.5E+05</td> <td>2.6E+04</td> <td>2.3E+01</td> <td>2.4E+08</td> <td>2.7E+05</td> <td>1.2E+05</td> <td>8.6E+04</td> <td>1.8E+01</td> <td>1.7E+03</td> <td>6.2E+01</td> <td>4.7E+05</td> <td>3.2E+03</td> <td>1.2E+05</td> <td>1.5E+02</td> <td>3.2E+03</td> <td>1.3E+05</td>	2012	1.5E+05	2.6E+04	2.3E+01	2.4E+08	2.7E+05	1.2E+05	8.6E+04	1.8E+01	1.7E+03	6.2E+01	4.7E+05	3.2E+03	1.2E+05	1.5E+02	3.2E+03	1.3E+05
Ref Second Second <td>2013</td> <td>1.5E+05</td> <td>2.8E+04</td> <td>2.1E+01</td> <td>3.7E+08</td> <td>2.6E+05</td> <td>1.2E+05</td> <td>9.3E+04</td> <td>1.4E+01</td> <td>1.5E+03</td> <td>5.3E+01</td> <td>4.7E+05</td> <td>2.0E+03</td> <td>9.2E+04</td> <td>9.6E+01</td> <td>2.7E+03</td> <td>9.7E+04</td>	2013	1.5E+05	2.8E+04	2.1E+01	3.7E+08	2.6E+05	1.2E+05	9.3E+04	1.4E+01	1.5E+03	5.3E+01	4.7E+05	2.0E+03	9.2E+04	9.6E+01	2.7E+03	9.7E+04
1 1 1 2 1	Total	4.4E+05	1.1E+05	6.8E+01	8.6E+08	1.1E+06	4.4E+05	3.6E+05	6.7E+01	7.4E+03	2.6E+02	1.9E+06	1.1E+04	4.7E+05	5.9E+02	1.3E+04	4.9E+05
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11 11 1	2012	3.3E+04	3.4E+04	9.5E-01	4.6E+07	1.6E+05	1.8E+04	2.1E+05	5.4E+00	1.2E+04	2.2E+01	4.0E+05	6.4E+02	9.3E+03	7.3E+00	3.1E+03	1.3E+04
1mm1	2013	6.1E+04	3.8E+04	1.6E+00	7.5E+07	2.1E+05	3.4E+04	2.3E+05	1.6E+01	9.6E+03	2.4E+01	4.8E+05	1.0E+03	1.3E+04	1.0E+01	3.2E+03	1.7E+04
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	Total	7.0E+04	2.4E+04	8.2E+01				1.5E+05	3.5E+01	8.8E+04	1.9E+02	5.9E+05		1.2E+05	1.4E+02	1.2E+04	1.5E+05

a.i.: active ingredient Source: Elaborated by the authors

Cienc. Tecnol. Agropecuaria, 22(2): e1520 DOI: https://doi.org/10.21930/rcta.vol22_num2_art:1520 As observed in table 1, the crop order according to the level of pressure on human health and the environment will vary among indicators. In general, the indicators evaluated point in the direction of sugarcane as the crop that exerts the highest pressure on human health and the environment, followed by rice and fruits. Vegetables and grains follow in importance, especially on Σ Seq, where their values were higher than those exerted by sugarcane. Once the crops of higher pressure are identified, the benefit of using indicators such as POCER and Σ Seq, instead of TL, is that the most affected environment and human modules are known, so decisions can be made to reverse the pressure.

From the toxicological point of view and considering the assumptions made in the calculations in POCER, re-entry workers not using personal protective equipment (PPE), and the adverse consequences derived from residents not using a nozzle with drift reduction during spraying activities, increased the risk of these modules compared to the applicator module. The ecotoxicological modules for aquatic organisms, persistence, and groundwater were the ones most at risk, due to the use of old and persistent a.i. in the environment like endosulfan, parathion methyl, methamidophos, paraquat, and ametryn, among others.

Analysis of individual hazardous active ingredients

Methamidophos (25.8 t) represented 15 % of the total amount of insecticide used (half of it was used solely in 2011) in rice, sweet potato, tobacco, grains, and vegetables, mostly. Endosulfan (3.46 t), parathion-methyl (3.02 t), and thiodicarb (1.69 t) belong to the group of insecticides forbidden for use in many countries, mainly in Europe and North America. Its use in developing countries is maintained, being Cuba an example of this (Rosquete, 2011; Vázquez & Pérez, 2017). Herbicides such as ametryn (215.3 t, 30 % of the total herbicides used), prometryn (13.41 t), hexazinone (11.60 t), and paraquat (1.28 t) are also employed.

		T	Total ΣSeq	POCER Sum (year 2011 scenario)							
	DT ₅₀ /MAC	Total kg a.i. used		Vegetables	Grains	Rice	Fruits	Tobacco	Sugar cane		
Endosulfan	117,808,219	3,482	4.1E+11	2.3E+03	2.1E+04	NR	7.4E+03	NR	NR		
Bifenthrin	193,151	3,254	6.3E+08	4.9E+03	2.3E+04	NR	NR	5.8E+03	NR		
λ-cyhalothrin	105,023	527	5.5E+07	2.8E+02	NR	2.9E+04	NR	NR	NR		
Parathion methyl	375	3,049	1.1E+06	1.4E+04	4.8E+03	NR	1.8E+04	1.4E+02	NR		
Methamidophos	4	25,791	1.1E+05	1.1E+03	3.1E+03	2.2E+04	1.1E+04	2.6E+03	NR		
Paraquat	1,667,659	1,423	2.4E+09	5.2E+03	1.5E+04	3.4E+04	1.9E+04	NR	1.8E+05		
Prometryn	2,808	12,803	3.6E+07	8.8E+02	NR	NR	1.7E+04	5.5E+03	NR		
Ametryn	1,408	215,272	3.0E+08	3.2E+02	NR	3.9E+04	6.3E+03	NR	7.7E+04		
Hexazinone	992	28,452	2.8E+07	NR	NR	NR	NR	NR	1.2E+05		
2,4-D amine salt	0.01	165,051	1.8E+03	NR	NR	7.1E+02	7.7E+01	NR	1.5E+04		
Copper oxychloride	34,247	24,943	8.5E+08	3.5E+04	2.0E+04	NR	1.9E+04	9.5E+03	NR		
Copper sulfate	769	21,065	1.6E+07	NR	NR	3.0E+04	NR	NR	NR		
Zineb	10	31,189	3.3E+05	1.6E+03	1.1E+02	NR	9.8E+03	5.8E+02	NR		
Mancozeb	4	100,229	3.8E+05	9.0E+03	8.6E+02	NR	4.7E+03	9.3E+03	NR		

Table 2. Pressure values of the active ingredients that are mainly used and those that are more ecotoxic.

DT₅₀: is the disappearance time for the first 50 % of the pesticide (days); MAC: maximum allowable concentration; a.i: active ingredient; NR: Not reported

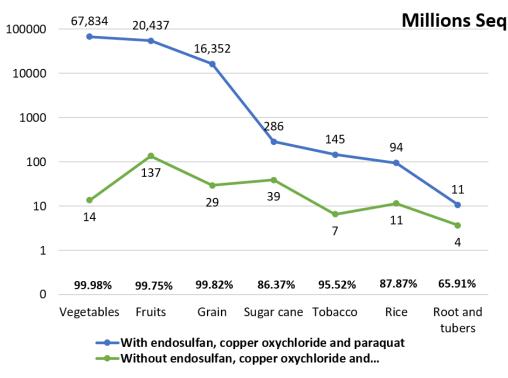
Note. The 2011 scenario was selected to illustrate the POCER case because this was the year with the highest pressure on the environment and human health.

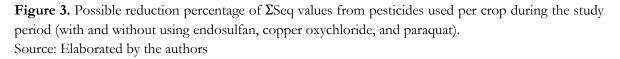
Source: Elaborated by the authors

The main active ingredients responsible for the ecotoxic pressure values are shown in table 2. As can be seen, the DT_{50}/MAC quotient is very important for Seq values. Based on this, small volumes of certain a.i. (such as endosulfan, paraquat, and oxychloride copper) can exert higher pressure than others used in large volumes. Similar results are also found for POCER. In this case, only the 2011 scenario was shown in table 2, as this was the year that exerted the most significant ecotoxicological pressure, due to the type of pesticides and quantities used. These can reduce ecotoxic pressure easier, since by eliminating or replacing a smaller amount of a.i. of higher ecotoxic pressure, reductions can be achieved. To illustrate the previous approach, some examples shown in figures 3 and 4, were developed.

A scenario with reduced average Σ Seq values for each crop is shown in figure 3. For this purpose, the contributed values from the a.i. with the highest pressure per family (endosulfan, copper oxychloride, and paraquat) were eliminated. Regarding tobacco, bifenthrin is used instead of endosulfan; and in rice, λ -cyhalothrin, copper sulfate, and ametryn are used instead of endosulfan, copper oxychloride, and paraquat, achieving reductions in Σ Seq values higher than 99 % for crops using the three latter. The percentage of reduction in sugarcane is based only on decreasing the use of paraquat, since insecticides and fungicides have not been assigned to this crop. The reduction percentage in the "roots and tubers"

group was lower because potato, the crop with the highest demand for pesticides in this group, was only cultivated in 2011 and 2012.





As the objective of POCER is to evaluate the pressure, from low to high risk, exerted by a pesticide on each of the evaluated modules, the decision-makers can either forbid the use of a high risk a.i. (table 2), alternatively, replace it with another a.i. that fulfills the same plant protection function with less pressure. The results of POCER concerning ecotoxicities from organophosphates and others like imidacloprid, bifenthrin, and β -cyfluthrin were higher than the pressure from endosulfan in some scenarios. On the other hand, by using POCER, endosulfan remained just as in Σ Seq, i.e., as one of the active ingredients that received the highest score from the aquatic organisms.

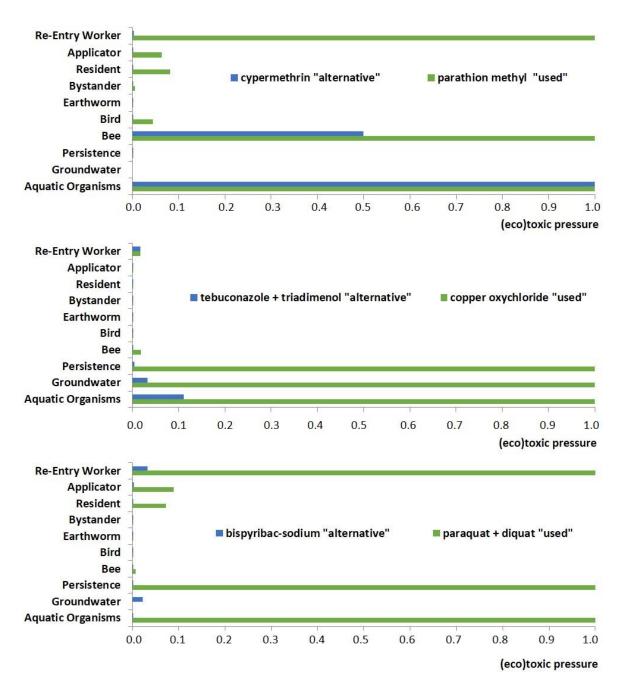


Figure 4. Proposal obtained with POCER to substitute high-risk pesticides to reduce the pressure evaluated in human (re-entry worker, applicator, resident, and bystander) and environmental modules (earthworms, birds, bees, persistence, groundwater, aquatic organisms).

Source: Elaborated by the authors

Cienc. Tecnol. Agropecuaria, 22(2): e1520 DOI: https://doi.org/10.21930/rcta.vol22_num2_art:1520 The organophosphorus compounds play an essential role as a whole due to their toxicity. As seen in figure 4, possible substitutes for the highest-scoring products are cypermethrin instead of parathion methyl, potentially reducing the risk by 50 %. A mix of tebuconazole and triadimenol under the commercial name Silvacur Combi[®] EC 30, reduced by 95 % the risk of copper oxychloride; on the other hand, bispyribac-sodium reduced by 98 % the risk of the paraquat-diquat mix (Doblete[®] LS 20).

Effects of using pesticides

Herbicides are the most used pesticides mainly due to the development of monocultures in large land areas, for example, in cereal grains (Beasley, 2020) and fruits, as is the case in the Sancti Spíritus province of Cuba. Cereal grains and fruits are the main crops cultivated in many countries that suffer from the highest pesticide load (Böcker & Finger, 2016; Chau et al., 2015; Schreinemachers et al., 2015; Shil Cha et al., 2014). The tendency to decrease the use of pesticides shown in this province for the study period follows the strategies to reduce the use of synthetic pesticides in the other provinces of agricultural importance in Cuba (Rosquete, 2011). However, in other provinces, there was a sustained increase in the use of pesticides (Hernández Núñez & Pérez-Consuegra, 2012). The pressure of pesticide use agrees with other tropical regions of the world (El Salvador, Brazil, Taiwan, Cambodia, Vietnam, and Tanzania) (Cremonese et al., 2014; Schreinemachers et al., 2015). Pesticides such as organophosphates, pyrethroids, carbamates, dithiocarbamates, neonicotinoids, and organochlorines used during the study period constitute an important risk to human health and the environment (Chau et al., 2015). Some of the compounds used (endosulfan, methamidophos, parathion methyl, mancozeb, paraquat, ametryn, and hexazinone) exhibit also chronic effects such as high mutagenic (Chaves et al., 2017), carcinogenic, and teratogenic potential (Cremonese et al., 2014), and high tendency to accumulate in fat-rich tissues. Furthermore, they are highly persistent in the soil and food, with a high capacity for biomagnification in food webs (European Food Safety Authority [EFSA], 2017).

Long-term environmental effects of pesticide use are recognized worldwide (López-Dávila, Ramos Torres et al., 2020; Mendonca et al., 2016). Lethal and sublethal effects on wild and managed bees are well documented (Fevery et al., 2016; Hladik et al., 2016; Parrilla et al., 2015), and exposure occurs via direct contact with pesticide spray or dry residues, as well as indirect contact contamination of nectar and pollen. In aquatic ecosystems, pesticides constitute a potential threat to aquatic biodiversity (Levine & Borgert, 2018; Pérez-Parada et al., 2018). The presence of highly toxic compounds can lead to a decrease in the number and varieties of fish, or alter phytoplankton communities, affecting, in turn, other trophic levels (Beasley, 2020).

Toxic load associated risks

There is now a perception that pesticide use is increasing (López Dávila et al., 2020), despite developments in biological control and the implementation of ecological pest management in the peasant sector promoted by the government. In other provinces, crop production (rice, cucurbits, beans, sweet potato, and tomato) used amounts of pesticides similar to those reported in this study, where the final

amount of pesticide used was up to 4.2 times higher than what had been planned (Hernández Núñez & Pérez-Consuegra, 2012).

It has been recommended that farmers become informed of the risks to which they are exposed, and the importance of using personal protective equipment and a drift reduction nozzle to minimize pesticide exposure and prevent intoxication (Yarpuz-Bozdogan & Bozdogan, 2016). Another way to reduce risk is to use a.i. of lower toxicity (Pesticide Action Network Europe, 2010). The Food and Agriculture Organization of the United nations (FAO) recommends in its Code of Conduct on Pesticides that those of category Ia and Ib (WHO, 2010), and if possible, Class II, should not be used in developing countries (World Health Organization & Food and Agriculture Organization of the total pesticides applied is included in the possible, probable, or human carcinogen and endocrine disruptor category. From the 124 a.i. applied, four (paraquat, methyl parathion, methamidophos, and benomyl) are included in international conventions (PIC, COP, and LRTAP), aiming at eliminating or limiting their use. Forty-one a.i. are listed with a classification in cancer categories (possible, probable, or human carcinogen) by United States Environmental Protection Agency, European Union, and the International Agency for Research on Cancer (IARC). Thirty-two a.i. are potential endocrine disruptors in humans and wildlife (WHO, 2010), posing a high risk for human health and the environment.

In both the European Union and North America (the United States and Canada), 15 of these products still used in Cuba were banned, and two have strict regulations implemented ten years ago (EFSA, 2017; Roberts & Routt, 2013) as they cause damages to human health and biodiversity (Pesticide Action Network Europe, 2010). The active ingredients endosulfan, methamidophos, methyl parathion, benomyl, and thiram are part of Annex III of the Rotterdam Convention, updated in 2017 (Food and Agriculture Organization of the United Nations & United Nations Environment Programme, 2017). Although endosulfan (causes chronic toxic effects on the nervous and immune systems, endocrine disruptive action and inconclusive evidence of its mutagenic and genotoxic action) was also included in Annex A of the Stockholm Convention (United Nations Environment Programme, 2009), it is still used in Cuba (Hernández Núñez & Pérez-Consuegra, 2012).

Ecotoxicity tests

The results from the Σ Seq indicator in Sancti Spíritus showed increased ecotoxic output over time, from 118 billion Seq to 259 billion, in contrast to a developed country like Belgium, which reduced the ecotoxicity values caused by pesticides. The Σ Seq values for the Cuban provinces for 2011 were more than 10 times higher than those obtained by Fevery et al. (2015) for that same year in Flanders (10.56 billion Seq). As these authors mentioned in their paper, the use of endosulfan was responsible for the high ecotoxicity values recorded. Endosulfan represented in the current study between 94.83 % (for beans in 2011) and 99.97 % (for onion in 2014) of the ecotoxicity indicator outcome for the crops where this product was used. However, it is necessary to eliminate the use of this insecticide, as was done in most developed countries in the European Union (EFSA, 2017).

An example of the positive change in ecotoxicity values when the use of endosulfan is eliminated, is what was experienced in the Flanders region in Belgium. When it was discontinued in 2012, its Σ Seq value decreased by 71 % compared to the 2009 values (Fevery et al., 2015). Paraquat and copper oxychloride are also responsible for high pesticide pressure values. Their pressure can be eliminated or replaced by other pesticides with less impact on the environment (Fevery et al., 2015).

Several authors agree that, due to the persistence of some pesticides in the soil and their ability to leach into groundwater and surface water bodies, aquatic organisms from the POCER indicator are the main modules at risk as a consequence of the use of highly toxic herbicides like paraquat and prometryn, as well as organophosphate insecticides (Bozdogan et al., 2015; Fevery et al., 2016; Yarpuz-Bozdogan & Bozdogan, 2016). Furthermore, in a citrus-growing region of Spain, the organophosphate chlorpyrifos followed by copper oxychloride were the most ecotoxic of the commonly applied pesticides for aquatic organisms (Cunha et al., 2012).

As Fevery et al. (2016) mentioned, 1 kg of a particular pesticide can exert a different pressure than the same amount of another pesticide. It is necessary to weigh the use of pesticides in relation to the toxicity coefficients for the various environmental compartments to quantify the risk of exposure to pesticides.

The POCER indicator has already proven its usefulness in Belgium and in other European countries (Claeys et al., 2005; Cunha et al., 2012; Bozdogan et al., 2015; Yarpuz-Bozdogan & Bozdogan, 2016) as a toxic pesticide reduction planning tool. POCER can be used as a decision-making tool for choosing alternative pesticides with respect to pressure on human health and the environment (Wustenberghs et al., 2018). It can also assess the impact of all pesticide applications related to a crop within a year and evaluate alternative cropping systems. The feasibility and effectiveness of policy measures, and the best practice at the farm level coupled with economic models, can be evaluated without jeopardizing profitability (Vercruysse & Steurbaut, 2002; Wustenberghs et al., 2018).

Conclusions

The study shows the suitability of POCER and Σ Seq indicators as important tools for decision-makers, as they can make more accurate toxicity and ecotoxicity assessments due to pesticides use, compared to the TL equation currently used in Cuba. The POCER indicator helped visualize that the human modules of re-entry workers (due to not using personal protective equipment) and residents (due to the negative consequences of not using drift-reducing nozzles during spraying activities), were the highest risk modules, even higher than the applicator module. Concerning the environmental modules, the POCER and Σ Seq indicators helped visualize aquatic organisms, persistence in soil, and groundwater as the most affected modules. The POCER and Σ Seq analyses showed how ecotoxic pressure can be reduced by more than 50 % by removing or replacing the more ecotoxic active ingredients, especially those still in use, such as endosulfan, bifenthrin, lambda-cyhalothrin, paraquat, prometryn, ametryn, hexazinone, and copper oxychloride. These results are directly related to the reduction goals promoted by the national government. To conclude, the indicators evaluated point in the direction of sugarcane as the crop that exerts the highest pressure on human health and the environment, followed by rice and fruits.

Acknowledgments

The author would like to thank Mr. Eris Perdomo, a specialist in signage and crop protection of Instituto Provincial de Sanidad Vegetal, for the data provided to carry out this study.

Disclaimers

The authors declare that they have no conflict of interest.

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