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Insecticide contamination in organic agriculture: Evidence from a long-term farming systems comparisons trial

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A B S T R A C T

Synthetic pesticides applied in conventional agriculture to control pests tend to compromise ecosystem services, and their residues may contaminate organic agriculture. To understand the significance of this contamination, also in small-scale farming systems in sub-Saharan Africa, quantitative data is required. Therefore, we compared synthetic insecticide and botanical/biopesticide residues in conventional and organic agricultural production systems after nine years of continuous cultivation of a maize-based crop rotation system at two sites in Kenya. Our results show high detectable concentrations of synthetic insecticide residues (imidacloprid, acetamiprid, chlorpyrifos, and chlorantraniliprole) in conventional plant produce and soil. Furthermore, the organophosphate chlorpyrifos was detected at concentrations above European Union Maximum Residue Limits (MRL) for plant produce, indicating potential risks for human health. Additionally, we detected imidacloprid, acetamiprid, chlorpyrifos, and chlorantraniliprole concentrations in the soil, indicating potential environmental harm. No residues of biopesticide/botanicals were detected in any of the production systems. However, we detected imidacloprid and chlorantraniliprole in organic plots. The findings indicate that the MRLs can be crossed even if synthetic insecticides are applied according to or below the recommended rates on the conventional plots. Thus, synthetic insecticides potentially risk human health and the environment, while botanicals and bio-pesticides represent a safe alternative.

1. Introduction

Globally, synthetic pesticides are used in conventional agricultural production systems to eliminate pests known to contribute to food losses (Asfaw et al., 2009). This is common for small-scale agricultural production systems in sub-Saharan Africa (SSA) (Wandiga et al., 2002). The trend illustrates that importing pesticides to sub-Saharan African countries, including Kenya, has recently increased (Route to Food Initiative, 2019). Even though pesticide usage per hectare is relatively low in SSA compared to other countries (Repetto and Baliga, 1996), most horticultural farmers tend to use higher rates and prefer highly toxic chemicals (Snyder et al., 2015). According to the WHO, pesticides can harm farmers' health (FAO, 2011). Studies have confirmed the concern that globally, 44 % of farmers experience acute pesticide poisoning yearly (Boedeker et al., 2020). At the field level, sprayed pesticides enter the environment via various matrices, including plants, water bodies, animals, and soil, where over time, some may degrade through chemical or microbial pathways (Pretty, 2012). The same applies to insecticides, which are used to eliminate insect pests. Residues of certain insecticides can harm non-target organisms and the environment and threaten sustainable agriculture management (Carr et al., 1997).

Insecticide exposures can be avoided by adopting sustainable

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agricultural production systems such as organic farming, which relies on botanicals and bio-pesticides. However, synthetic insecticide drift from neighbouring conventional farms and persistent synthetic residues in soil and crops from previous seasons from conventionally cultivated fields (Yurtkuran and Saygı, 2013) could contaminate organic agricultural systems. Moreover, how long insecticides remain in the farming system matrix (soil and water) depends on the physicochemical properties of the soil and water systems and type of insecticide, and other factors such as climate (Ware and Whitacre, 2004).

This study evaluated synthetic- and bio-pesticides (insecticide) residues in soil and plant samples from conventional and organic agricultural production systems at low and high input levels after nine years of continuous cropping. We hypothesized that insecticide residues would be more persistent in conventional agricultural production systems than in organic ones. In contrast, residues of botanicals and bio-pesticides will be degraded in the latter agricultural systems. Additionally, we hypothesized that residues of synthetic insecticides from conventional would contaminate organic experimental plots.

2. Materials and Methods

The study on insecticide residues was conducted in the ongoing Long-term Farming Systems Comparisons (SysCom) trials at Chuka in Tharaka Nithi County, about 150 km away from the capital Nairobi (37°04.747′ N; 1°0.231′ S) and Thika in Murang'a County about 50 km away from Nairobi (37°38.792′ N; 0°20.864′ S), situated in the sub-humid zones of the Central Highlands of Kenya. A bimodal rainfall characterizes both sites in a long and short rainy season, with total annual precipitation of about 1373 mm at Chuka and 840 mm at Thika. In addition, soil types further distinguish both sites, with Chuka having a more fertile soil classified as Humic Nitisol compared to Thika soil classified as Rhodic Nitisol based on FAO world reference base for soil resources (IUSS Working Group WRB, 2006; Wagate et al., 2010a, 2010b). However, both sites have a similar annual mean temperature (~20 °C) and altitude (~1500 m asl). More details on sites and setup can be found in an earlier publication (Adamtey et al., 2016) or on the project website (FiBL, 2022).

At each site, conventional (Conv) and organic (Org) farming systems are being compared at two levels of input: high input (High) using the recommended rates of fertilizer and pesticides as well as supplementary irrigation (which represents commercial production), and low input (Low) using limited amounts of fertilizer and pesticides (representing smallholder production, mainly for subsistence). The type and quantity of inputs applied in the high-input systems were based on the recommendations of the Kenyan Ministry of Agriculture (MOA). The inputs in the low input system were based on a survey conducted within the areas of the two experimental sites (Musyoka, 2007; Székely, 2005).

The management practices of these four farming systems were applied on experimental plots of 8 × 8 m (64 m^2; net plot 6 × 6 m) arranged in a Randomized Complete Block Design (RCBD), replicated four times at Chuka, and five times at Thika. The crop rotation consists of maize, beans, leafy vegetables, and potatoes, planted in a 3-year-6-season crop rotation with a long and a short season. An overview of fertility, water, and pest management can be seen in Table 1.

Table 1
Fertility, water, and pest management of all farming systems in the long-term trials at Chuka and Thika, Central Highlands of Kenya.

Treatment	Fertility Management	Pest management	Water management
Conv-Low	Organic & synthetic fertilizer (45 kg N ha^{-1} $year^{-1}$ & 60 kg P_2O_5 ha^{-1} $year^{-1}$)	Synthetic pesticides	Rainfed
Org-Low	Organic fertilizer (45 kg N ha^{-1} $year^{-1}$ & 60 kg P_2O_5 ha^{-1} $year^{-1}$)	Biopesticide/ Botanicals	Rainfed
Conv-High	Organic & synthetic fertilizer (225 kg N ha^{-1} $year^{-1}$ & 286 kg $P_2O_5ha^{-1}$ per year)	Synthetic pesticides	Irrigation
Org-High	Organic fertilizer + rock-PO_4 (225 kg N ha^{-1} $year^{-1}$ & 286 kg $P_2O_5ha^{-1}$ per year)	Biopesticide/ Botanicals	Irrigation

Conv-Low: Conventional low input system; Org-Low: Organic low input system; Conv-High: Conventional high input system; Org-High: Organic high input system.

The data on insecticide residues were obtained from an analysis of soil and plant materials sampled after nine years of continuous cropping during the short rainy season of 2016. The soil samples were taken in the net plot in triplicates at two depths (0–25, 25–50 cm) before the planting (BP; November 2016) and during the last harvest of the crops (harvest sampling; see below). The BP sampling was carried out only at the Thika site. This was not done at the Chuka site due to logistical problems. Plant samples were taken from the leafy vegetable cabbage (*Brassica oleracea* subsp. *capitata* L.) in the case of high input systems, and from kale (*Brassica oleracea* subsp. *acephala* DC.) and Swiss chard (*Beta vulgaris* var. *cicla* L.) in the case of low input systems, which were grown in this season. Sampling time was at the last harvest of the crops in March 2017 at Chuka and in January/February 2017 at Thika. For cabbage, the crop was sampled after separating the whole crop into two parts: the marketable cabbage head for human consumption and the remaining parts, which are typically used for animal feed and composting. All samples were transported in polyethylene bags and stored at −20 °C in the laboratory. Before storage, plant samples were cut into smaller sizes and homogenized with a blender.

The "QuEChERS" method developed by Anastassiades et al. (2003) was used to extract and clean up the synthetic insecticide residues acetamiprid, imidacloprid, chlorpyrifos, chlorantraniliprole, and the biopesticide/botanical residues azadirachtin and pyrethrin. Residues from these insecticides were selected for the study because they were applied either during the sampling season or during the last cropping year (short rainy season 2015 – short rainy season 2016; see Table 2 Supplementary material).

A multi-residue approach similar to the one used in Irungu et al. (2016), using LC-MS/MS as described below for screening, was adapted to detect the chemical contaminants against the targeted pesticides and biopesticides/botanicals. Data analysis was carried out by monitoring two transition ions. The most dominant transition ion was used for quantification, whereas the second most intense ion was used as a qualifier for confirmation purposes. To generate calibration curves used for quantitation, matrix-matched calibration standards were prepared at seven calibration levels (from 0.1 to 100 ppb including the zero point) in blank extracts of the respective matrices (soil and respective plant matrices for cabbage, kale and Swiss chard). The resulting calibration curves were used to determine the method's limit of quantification (LOQ) and limits of detection (LOD). The LOQ was set as the minimum concentration that could be quantified with acceptable accuracy and precision.

The analysis was performed using ultra-high-pressure liquid chromatography (UHPLC) Agilent 1290 series coupled to a 6490 model triple quadrupole mass spectrometer (Agilent Technologies), with an ifunnel JetStream electrospray source operating in the positive ion mode. The electrospray ionization settings were gas temperature, 120 °C; gas flow, 15 l/min; nebulizer gas, 30 psi; sheath gas temperature, 375 °C; sheath gas flow, 12 l/min; capillary voltage, 3500 V; nozzle voltage, 300 V. The ifunnel parameters were high-pressure RF 150 V and low-pressure RF 60 V. Nitrogen was used both as a nebulizer and as the collision gas. Data acquisition and processing were performed using Mass Hunter Data Acquisition; Qualitative and Quantitative analysis software (Agilent Technologies, Palo Alto, CA, v.B.06 and v.B.07). The chromatographic separation was performed on a Rapid Resolution reverse phase column-C18 1.8 μm, 2.1 × 150 mm column (Agilent Technologies). A gradient elution at a flow rate of 0.4 ml/min was used with water and acetonitrile. Each contained 5 mM ammonium formate in 0.1% formic acid as mobile phases A and B respectively.

The data retrieved from the analysis were entered into a database. The statistical analysis was done with the statistics software R (R Core Team, 2020) after separating the dataset into different sampling materials (soil, plant), trial sites, and sampling dates. Initial exploitation was done with the NADA package (Lee, 2020) and the functions *censummary* to derive the mean and standard deviation with a Regression on Order Statistics (ROS.) as data contained left-censored values (Helsel, 2005). The same package was also used (function *cendiff*) to compare soil samples means of the organic and conventional farming systems within each site, depth, and within/between input levels. The average residue concentration of plant samples of the organic and conventional farming systems was compared within each crop, crop part, and input level. The significance level was set at $p < 0.05$. In addition, we used the *tidyverse* package to compile data, derive tables, and generate the figures (Wickham et al., 2019).

3. Results

3.1. Residues of synthetic pesticides in conventional systems

Generally, our results show that residues of synthetic insecticides were detected in both conventional farming systems (high and low input) in soil and plant samples. In the soil, the highest concentrations of acetamiprid (164 μg kg^{-1}), imidacloprid (361 μg kg^{-1}), chlorantraniliprole (8 μg kg^{-1}), and chlorpyrifos (386 μg kg^{-1}) were always found in Conv-High at the topsoil during harvest at Chuka (Fig. 1, Fig. 2).

The detailed results showed a significant increase of the acetamiprid and chlorpyrifos residues in soil from BP to harvest sampling (measured at Thika) ($p < 0.01$) in the topsoil in Conv-High (Table 3). Both active ingredients were part of insecticides sprayed during the season (Table 3). In contrast, residue concentrations of imidacloprid and chlorantraniliprole were similar or decreased significantly in soil from BP to harvest ($p < 0.05$).

The residue concentrations in the subsoil (25–50 cm) of Conv-High were generally either lower compared to the topsoil, e.g., imidacloprid 10 to 120-fold lower, or not detected at all (<LOQ). Also, in Conv-Low, most concentrations of residues were several times lower compared to Conv-High. Thus, the highest concentrations of acetamiprid (0.5 μg kg^{-1}), imidacloprid (23 μg kg^{-1}), chlorantraniliprole (6 μg kg^{-1}), and chlorpyrifos (1 μg kg^{-1}) in Conv-Low in soil were found at various depths and sites (Fig. 1 and Table 3).

The high values in soil for Conv-High were also reflected in the plant samples showing the highest concentrations at Chuka for acetamiprid (406 μg kg^{-1}), imidacloprid (433 μg kg^{-1}), chlorantraniliprole (0.7 μg kg^{-1}), and chlorpyrifos (232 μg kg^{-1}) (Fig. 3, Fig. 4, and Table 3).

Notably, the concentration of chlorpyrifos in the edible plant sample grown in Conv-High at Chuka and Thika (232 μg kg^{-1} and 105 μg kg^{-1}) exceeded the Maximum Residue Level (MRL) for cabbage determined by the European Union applicable during the sampling time (10 μg kg^{-1} see also Table 3) (Website). The residue concentrations in Conv-Low in soil and plant samples were generally lower than Conv-High. However, at Chuka and Thika, the concentrations detected in Conv-Low of chlorpyrifos in kale (16 μg kg^{-1} and 19 μg kg^{-1}) and in Swiss Chard (13 μg kg^{-1} and 16 μg kg^{-1}) also exceeded the MRL (10 μg kg^{-1}) (Website).

3.2. Residues of biopesticide/botanicals and synthetic pesticides in organic systems

No residues of biopesticide/botanicals were detected in the soil and the plant samples obtained from the organic farming systems. Neither Org-Low nor Org-High showed residue concentrations of the biopesticides/botanicals. However, residues of imidacloprid and chlorantraniliprole were also detected in the organic farming systems. Imidacloprid was detected in soil and plant samples with the highest concentration during harvest in Org-Low in topsoil at Chuka (1.4 μg kg^{-1}) and in Org-High in edible plant samples at Chuka (0.7 μg kg^{-1}; Figs. 1 and 3). These concentrations were several times lower (significantly; $p < 0.05$) than the concentrations found in Conv-Low (15-fold) and Conv-High (600-fold) at the same sites and sampling points (Table 3). In addition, the residue concentrations were below MRLs. In

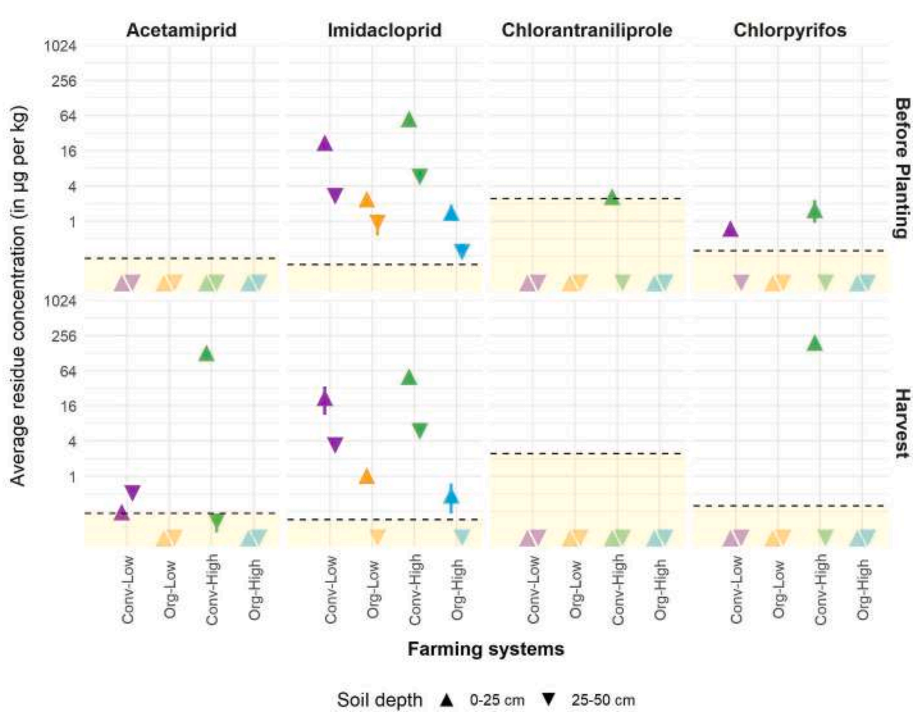

Fig. 1. Average residue concentration (in μg per kg) of acetamiprid, imidacloprid, chlorantraniliprole, and chlorpyrifos in two soil depths (0–25 cm, 25–50 cm) before planting (BP) and during harvest sampling at Thika, Central Highland of Kenya (Note: dotted line is the limit of quantification (LOQ); all transparent symbols for average residue concentration at the bottom of the graph indicating values below LOQ).

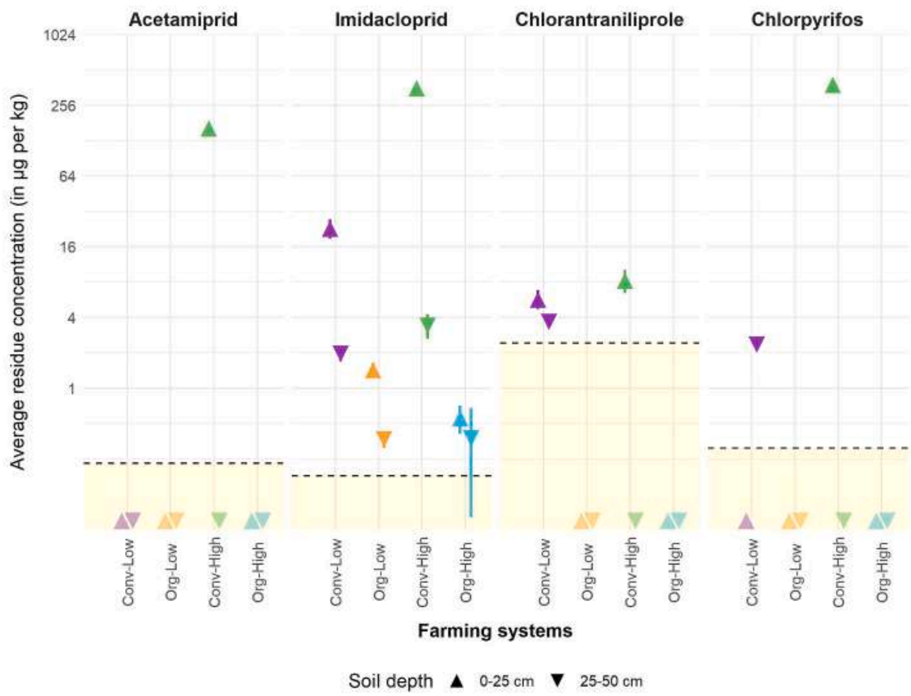

Fig. 2. Average residue concentration (in µg per kg) of acetamiprid, imidacloprid, chlorantraniliprole, and chlorpyrifos in two soil depths (0–25 cm, 25–50 cm) during harvest sampling at Chuka, Central Highland of Kenya (Note: dotted line is the limit of quantification (LOQ); all transparent symbols for average residue concentration at the bottom of the graph indicating values below LOQ).

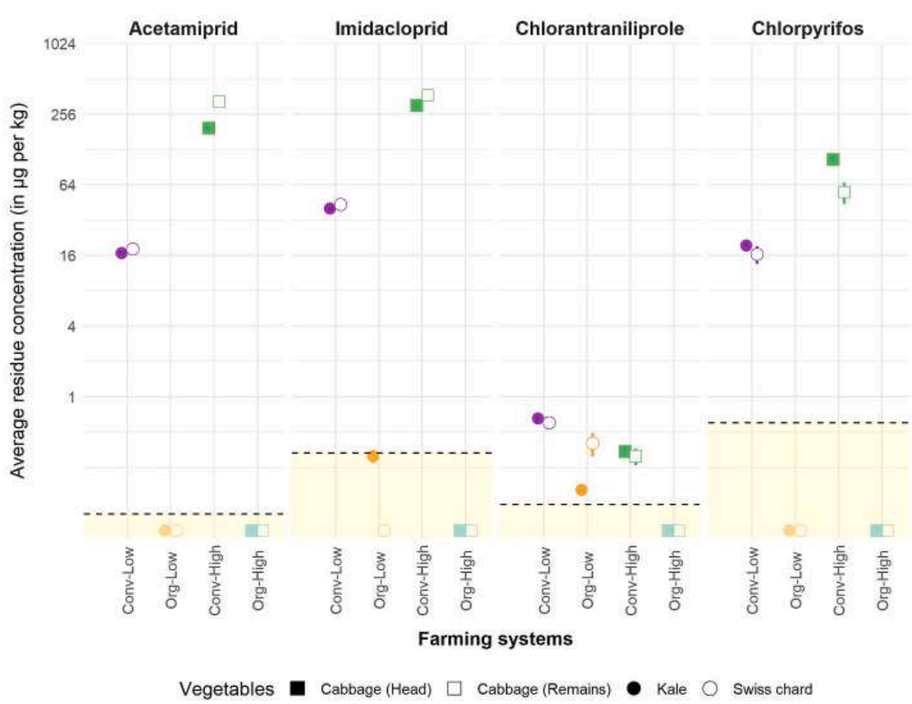

Fig. 3. Average residue concentration (in µg per kg) of acetamiprid, imidacloprid, chlorantraniliprole, and chlorpyrifos in four plant parts (cabbage head, cabbage remains, kale, and Swiss chard) at Thika, Central Highlands of Kenya (Note: dotted line is the limit of quantification (LOQ); all transparent symbols for average residue concentration at the bottom of the graph indicating values below LOQ).

Org-Low, only residues of chlorantraniliprole were detected in the plant samples at Chuka and Thika (0.1 and 0.3 µg kg^{-1}; Fig. 3). Also, these concentrations were several times lower than the concentration found in Conv-Low.

4. Discussion

Synthetic insecticide residues, detected in both soil and plant samples, were highest in the conventional systems. This also applies to the neonicotinoids imidacloprid and acetamiprid residues that were detected highest in the topsoil of Conv-High. Neonicotinoids are perceived

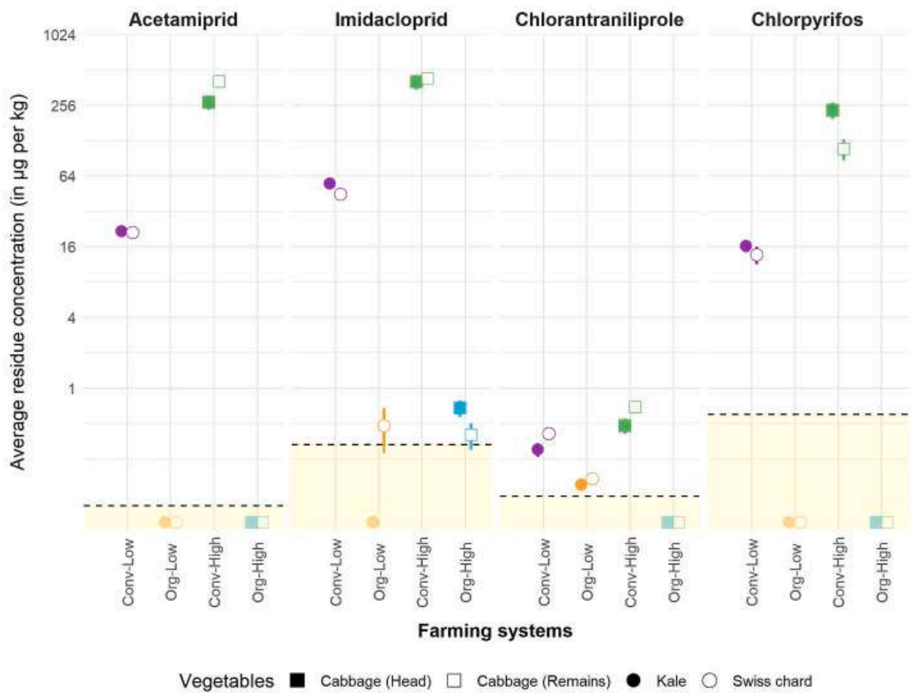

Fig. 4. Average residue concentration (in µg per kg) of acetamiprid, imidacloprid, chlorantraniliprole, and chlorpyrifos in four plant parts (cabbage head, cabbage remains, kale, and Swiss chard) at Chuka, Central Highlands of Kenya (Note: dotted line is the limit of quantification (LOQ); all transparent symbols for average residue concentration at the bottom of the graph indicating values below LOQ).

critically because of their potential risks to human health and the environment. According to several authors, Imidacloprid is toxic to pollinators and natural enemies (de Lima e Silva et al., 2017; Tan et al., 2014). The active ingredient of neonicotinoids interferes with the insects' nervous system and does not differentiate between target and beneficial insects. Therefore, the imidacloprid application in the conventionally treated study plots, especially in Conv-High, may harm pollinators and natural enemies. Notably, imidacloprid has been banned in the European Union since 2018 for outdoor usage (European Commission, 2018). It is also known that imidacloprid reduces biochemical and microbial soil functioning, such as the total bacteria population in the soil (Cycoń and Piotrowska-Seget, 2015). More recently, concerns were raised by scientists about neonicotinoid risks for agricultural use in Africa (Burnside, 2020). Another factor why neonicotinoids are perceived critically is their persistence in the environment. A previous study showed that neonicotinoids such as imidacloprid could remain in the soil for more than a year, especially in tropical soils with half-lives of over 150 days (Dankyi et al., 2018). Interestingly the residues did not increase significantly between the BP and the measurement at harvest even though imidacloprid was sprayed this season. We assume that the high residue amounts already present at the BP measurement caused this incident.

The neonicotinoid acetamiprid was detected in soil and plant samples of conventional farming systems, highest in Conv-High. The detected residues increased significantly between the BP and the harvest measurement and could be explained by the pesticide application in the season. The detected residues can represent an environmental risk, especially in Conv-High since it is also classified as moderately hazardous to pollinators (Yue et al., 2018). Nonetheless, the MRL determined by the European Union for acetamiprid applicable during the sampling time was not exceeded by the pesticide residue detected (Website). However, the residue concentration in kale in the Conv-Low system exceeded the current applicable MRL (10 µg kg^{-1}, Website) and although acetamiprid was applied below recommendations for the Conv-Low system (see Materials and Methods section), the residue concentration at this level indicate a risk to human health. It was shown that acetamiprid could cause acute risks such as crouching, tremors, and convulsion, but also long-term risks such as neurotoxicity effects through direct exposure (Alavanja et al., 2004). Furthermore, acetamiprid is found to cause reproductive toxicity in mammals, including humans through chronic exposure (Zuščíková et al., 2023).

The studied active ingredient chlorpyrifos was detected in both soil and plant samples, and was highest in the Conv-High system. The detected residues increased significantly between the BP and the harvest measurement and could be explained by the pesticide application in the season. Chlorpyrifos is an organophosphate classified as highly toxic to bees and aquatic organisms (Giddings et al., 2014) and therefore represents an environmental risk if applied as in the Conv-High plot. Furthermore, the chlorpyrifos residues exceeded the MRL threshold of the European Union both in Conv-High and Conv-Low systems. Chlorpyrifos can cause acute and long-term health effects in humans (Testai et al., 2010) since it is neurotoxic with reproductive and developmental toxicity and is classified as "Moderately hazardous" for humans by the World Health Organization (World Health Organization, 2020). Consequently, chlorpyrifos was withdrawn from the European market in 2020 (European Commission, 2020).

The synthetic insecticide chlorantraniliprole is very persistent in water, soil, or sediment (World Health Organization, 2020). Chlorantraniliprole residues were detected in the topsoil in the conventional systems, even though it was applied two seasons before the sampling (short rainy season 2015). Chlorantraniliprole is very toxic to aquatic organisms (USEPA, 2008). Thus, a run-off or leaching of it into water bodies may have negative implications for aquatic organisms.

The finding that residues from synthetic insecticides were detected in soil and plant samples in the conventional systems throughout the season, whereas in the organic systems, no residues of biopesticides/botanicals were found, confirms our hypothesis that synthetic insecticide residues in conventional systems can be more persistent than botanicals/bio-pesticides in organic systems. In addition, the residues also exceeded MRLs for some crops in the conventional systems, even though the spraying was done at recommended rates (Conv-High) or even lower (Conv-Low). A possible reason for the exceeding value could be the inadequate knowledge base on the fate, effect, and risk of pesticides in tropical environments (Lewis et al., 2016). Thus, a decision on recommended rates (by, e.g., governmental organizations) for pesticide use of farmers is probably based on the effectiveness of the pest control and not on environmental or human risk because detailed data is not available. This is also shown by the current state of published peer-reviewed papers on the topics: only 478 articles were identified for the whole of Sub-Saharan Africa (SSA) by a recent study (Fuhrimann et al., 2022). However, another explanation is that the MRLs are designed for the EU and might be only partially adaptable to SSA. A study focusing on risk exposure might be more helpful like Serbes and Tiryaki (2023).

None of the active ingredients in the biopesticides/botanicals used in this study were detected in the samples, indicating that they are non-persistent in the environment and, as indicated in the literature, do not pose a health risk to humans or non-target organisms (Akbar et al., 2010). This confirms our hypothesis that residues of the tested botanicals and bio-pesticides will be degraded faster and can be a safe alternative for pest management practices. However, residues of synthetic pesticides were detected in soil and plant samples in all the agricultural production systems, including the organic systems, which confirms our hypothesis that residues of synthetic insecticides from conventional can contaminate organic experimental plots. This finding is in line with studies conducted in Europe, e.g., by Tiryaki (2017) or Schleiffer and Speiser (2022), where the authors state that synthetic pesticide residues were regularly found for organic produce. The contamination in organic production can happen due to either obstinate environmental contamination (e.g., irrigation water), application drift or illegal use of pesticides (Tiryaki, 2017). In this controlled study, the illegal use of synthetic pesticides in the organic plots can be excluded as a contamination source, but there was no information collected about the residue concentration of inputs like irrigation water or organic fertilizers. However, it can be assumed that most contamination appeared through application drift due to the proximity of the trial plots (2 m distance from each other). This would be in line with the study of Dalvie et al. (2014) that showed increasing residue concentrations in air during the time of pesticide application in Western Kenya.

An important limitation of this study is that sampling was carried out for insecticides only once in replicates after nine years of continuous planting. As such, a more detailed sampling over a long period for different pesticides (insecticides, herbicides, fungicides, etc.) is required to robustly determine the full potential of fate and contamination potential in the two different farming systems. In addition, research on pesticide metabolites and pathways, for example, through water, drift, or animal feeding under tropical conditions, may be needed to provide scientific evidence for detailed policy recommendations.

5. Conclusions and recommendations

After nine years of continuous cropping at the two study sites in Kenya, high amounts of synthetic pesticides were detected in plant and soil samples in the conventional farming systems. In conclusion, our results have demonstrated that pesticide residue of plant parts grown in conventional plots can exceed the Maximum Residue Limits given by the European Union (both for low and high input systems). In addition, pesticide residues were detected in soil samples, especially in the Conv-High plots. This indicates that the synthetic insecticides, which were applied according to the recommendations (Conv-High) or below the recommendations (Conv-Low), can still harm the environment and human health, while the studied botanicals and bio-pesticides represent a safe alternative. However, more research on pesticide fate and impact in different farming systems under tropical conditions must be done to provide clear recommendations for policymakers.

CRediT authorship contribution statement

Ivonne Kampermann: Formal analysis, Writing – original draft, Writing – review & editing, Visualization. **David Bautze:** Formal analysis, Writing – original draft, Writing – review & editing, Visualization, Project administration, Funding acquisition. **Millicent Mapili:** Investigation, Data curation. **Martha Musyoka:** Conceptualization, Resources, Data curation, Writing – review & editing, Project administration, Funding acquisition. **Edward Karanja:** Conceptualization, Resources, Data curation, Writing – review & editing, Project administration. **Komi K.M. Fiaboe:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Project administration. **Janet Irungu:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision. **Noah Adamtey:** Conceptualization, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cropro.2023.106529.

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