

## Pesticide Residue and Health Risk Analysis of Six Commonly Used Herbal Medicinal Products in Kumasi, Ghana

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### Abstract

*Background:* Ghana does not have reliable surveillance programme for ensuring safety of food and herbal products on the market. Data from previous studies have shown the presence of pesticide residues in food products and its associated health risk. In this present study we assessed the residual pesticide content of six most patronized herbal preparations in Kumasi, Ghana and the attendant risk of such contamination to public health.

*Methodology:* The herbal preparations were cleaned up using Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) method for insecticide analysis. Analysis was carried out on GC-MS using 33 external standards.

*Results:* The total pesticide content in most of the herbal preparations was below the maximum residue limit (MRL). However, the concentration of pirimiphos-methyl in herbal product 'E' (HPE) (0.082 mg/kg) was higher than the MRL (0.05 mg/kg). In addition, the hazard indices' (HI) for five herbal products were less than '1' and safe for consumption without any possible future health risk. However, the HI for HPE was determined to be greater than '1'. Aldrin, dieldrin and chlordane which are banned in Ghana were also identified in 50% of the studied herbal preparations.

*Conclusion:* Data from this study reveals that HPE is contaminated with pirimiphos-methyl pesticide and unsafe for consumption. Also, the presence of banned pesticides in 50% of the studied herbal preparations calls for concern and urgent intervention by relevant regulatory authorities.

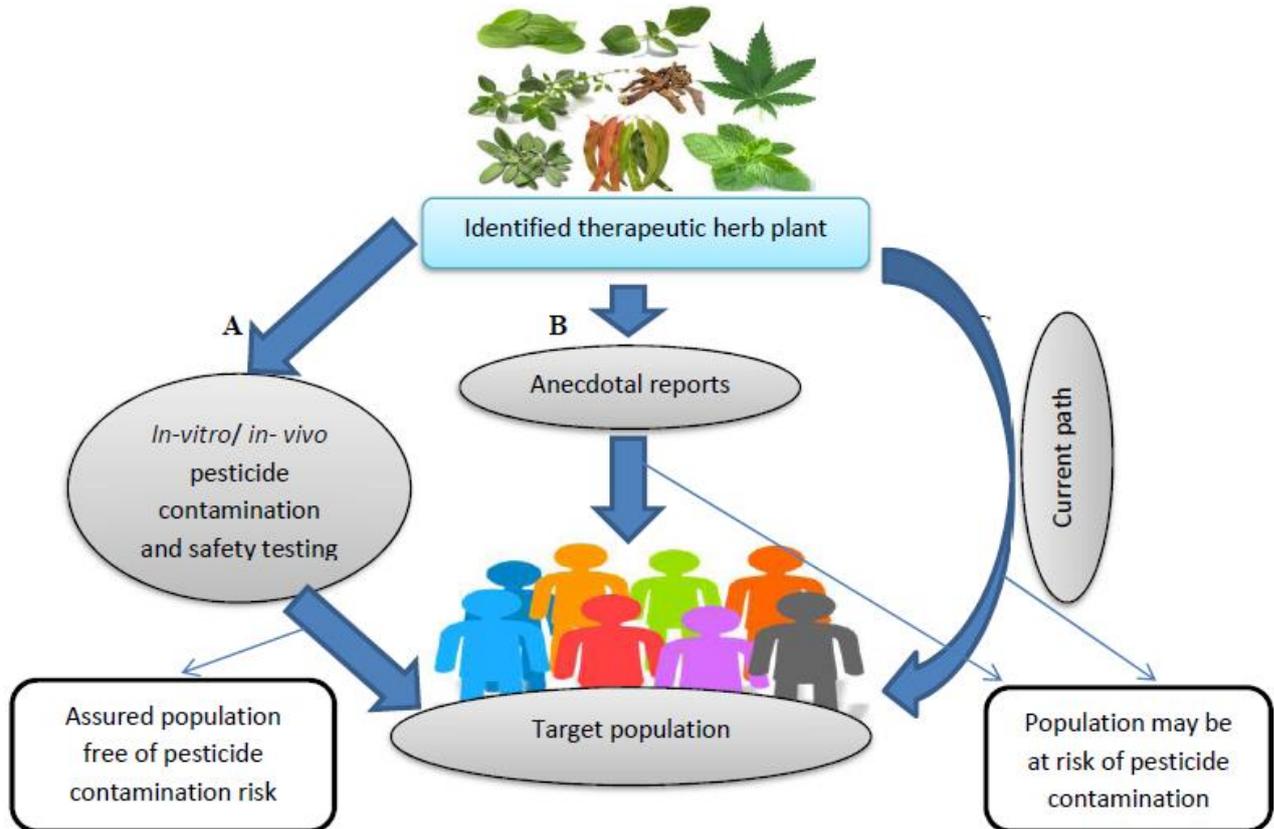
**Keywords:** Herbal medicinal preparation, hazard index, pesticides, contamination, health risk, safety.

### Introduction

Close to 80% of the Ghanaian population rely on the herbal medicines (HM) for their basic health needs. They (HMs) are used to cure minor ailments, treat chronic diseases and for preventive purposes. Pesticide-contaminated herbal preparations may have detrimental effects and it belittles the intended health benefits of the herbal product. Population growth has resulted in increased use of pesticides to increase crop yield, reduce post-harvest and storage losses and increase shelf life of farm produce, (Chouaibou et al., 2016; Johnson et al., 2013). Residual pesticides may end up in the tissues of these medicinal plants. Elevated pesticide residues in medicinal plants is, therefore a major health concern, (Obiri-Danso et al., 2011). Though in Ghana, most herbal medicinal materials are obtained from medicinal plants grown in the wild, research has shown that pesticide uptake by non-targeted plants through indirect pesticide application is significant, (Essumang et al., 2013).

Pesticide residues in herbal products and other food stuff on the Ghanaian market are poorly monitored. There is also apparently no regular surveillance programme to regularly monitor the residual pesticide content in food and multierbal products on the market putting the health of the Ghanaian populace at pesticide contamination health risk. In addition, there is also currently no published work on pesticide residues in herbal preparations in Ghana, as a result, residual pesticide data on herbal preparations in Ghana are lacking. The few published studies from the Ghanaian context were cited on vegetables (Amoah et al., 2006; Darko and Akoto, 2008; Osei-Fosu et al., 2014; Akoto et al., 2015a), fruits, (Fosu et al., 2017), beans (Di Bella et al., 2017), cereals (Akoto et al., 2015b), honey (Darko et al., 2017), fishes (Kuranchie-Mensah et al., 2013) and dairy products (Darko and Acquah, 2008). Most of these studies suggested pesticide contamination health risk to

consumers. Though studies on residual pesticide content on herbal preparations have been published in some European, Asian and American countries, geographical, exposure, and other variations in anthropogenic activities makes it not generalizable to the herbal products on the Ghanaian market, (Naithani and Kakkar, 2004; Oh, 2009; Mosaddegh et al., 2014; Yang et al., 2017). It is imperative, therefore, that herbal medicinal products on the Ghanaian market are regularly monitored to assess possible contamination and health risk and ensure public safety. The main aim of this study, therefore, is to determine the pesticide content and assess the human health risk of the six-most patronized herbal medicinal products on the Kumasi market of Ghana.



**Figure 1.** Schematic route of herbal medicine from field identification to target population for human consumption.

Path 'A' is the recommended scientific path, but paths 'B' and 'C' are the observed paths for most HMPs in Ghana these days. Most herbal materials and products are not tested for pesticide contamination before they reach the target population in Ghana. The dominant paths currently in Ghana are the paths 'B' and 'C' which put the population at risk of possible pesticide contamination and health risk. Path 'A' which is scientifically recommended path is somehow rarely used by few herbal companies in the country.

## Methodology

### Sample selection



**Figure 2.** Pictorial map of Ghana showing the sampling city

A mini survey was carried out among some randomly selected herbal medicine users and pharmacy shops that also sell herbal medicine on wholesale or retail. The mini survey was carried out in Kejatia, Bantama and Ashtown districts in the Kumasi Metropolis of Ghana, (Fig. 2). Herbal medicine users were asked about the anti-malarial, anti-hypertensive and anti-diabetic herbal medicinal products they go for when unwell. Also, the wholesale and retail pharmacy shops involved in the study were asked about the most patronized antimalarial, anti-diabetic and anti-hypertensive HMPs in their collections. The list was compiled and tallied. The top 3 antimalarial, top 2 anti-diabetics and the top 1 anti-hypertensive HMPs which were most patronized according to the survey data were selected for the pesticide study. The six HMPs investigated were Alive Diabalex Mixture<sup>®</sup> for diabetes, Osompa D.P.<sup>®</sup> for diabetes and high blood pressure, Time Herbal Mixture<sup>®</sup> for malaria, Tetewobika Garlic Bitters<sup>®</sup> for hypertension, Adutwumwaa Malamix<sup>®</sup> for malaria and Taabea Herbal Mixture<sup>®</sup> for malaria. The six HMPs were randomly coded for ethical reasons and henceforth shall be represented by their random codes; HPA, HPB...HPF.

## Extraction and clean-up of the liquid herbal mixtures for GC-MS pesticide analysis

Herbal preparations were extracted in n-hexane-ethylacetate system (3:7) and clean-up using Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS) sample preparation method for GC-MS multi-residue pesticide analysis. The procedure as described by Nguyen and Kimaru was applied (Kimaru, 2014). Standard pesticide solutions of 33 pesticides with known concentrations were used as external standards. Results were presented in chromatographs, tables and figures.

### Quality control parameters of the GC-MS

The detector type of the GC-MS was set to 3800 (10 volts), Bus address was set to 44, channel was set to ECD, sample rate was set to 10 Hz and a run time of 32.968 minutes. A total of 33 external pesticide standards were used and the instrument was set to analysis mode. Pesticide content was calculated using peak area measurement and the offset time was set to 0.000 minutes. Duplicate injections were made at a delay time of 0.8 minutes.

### Health risk estimation

#### Estimated daily intakes (EDIs) of the pesticides identified in the herbal products

The estimation of EDI of each of the identified pesticide present in the mixture was determined by the following equation, (Ou et al., 2016).

$$EDI = \frac{E_D \times C}{W_{AB}}$$

Identified pesticides present in the herbal preparations were compared to international acceptable reference dose levels.  $EDI = [C \text{ (content of pesticide)} \times E \text{ (daily dosage)}] / (BW)$ . BW is the Ghanaian average body weight; (65 kg adults, 24 kg children). (Ekhatior et al., 2017; Ou et al., 2016). Estimated daily intake for Chlopyrifos is 0.1 mg/kg Bw acute and 0.01 mg/kg Bw chronic. (Skovgaard et al., 2017)

#### Targeted hazard quotient (THQ)

THQ for pesticide contaminants without established RfDo shall have their THQ estimated using the formula below. Where C is the concentration of the contaminant pesticide in the HMP (mg/kg), MRL is the maximum residual limit in  $\text{mg.kg}^{-1}.\text{day}^{-1}$ .

$$THQ = C / MRL$$

#### Hazard index (HI)

The chronic hazard index (HI) is the sum of more than one hazard quotient for multiple toxicants. Harrison and Chirgawi, (Harrison and Chirgawi, 1989) reported that exposure to two or more pollutants may result in additive and/or interactive effects. THQs can be summed across constituent pesticides to generate a hazard index (HI) for an oral dosage-pathway combination.

$$HI = \sum_{n=1}^{\text{total}} THQ_n$$

## Results and discussion

The total pesticide content in most of the herbal preparations was below the MRL limit. However, the concentration of pirimiphos-methyl in HPE (0.082 mg/kg) was higher than the MRL limit (0.05 mg/kg). This means that HPE do not meet the legal requirements for pirimiphos-methyl to be on the market and used by patients and may pose health risk.

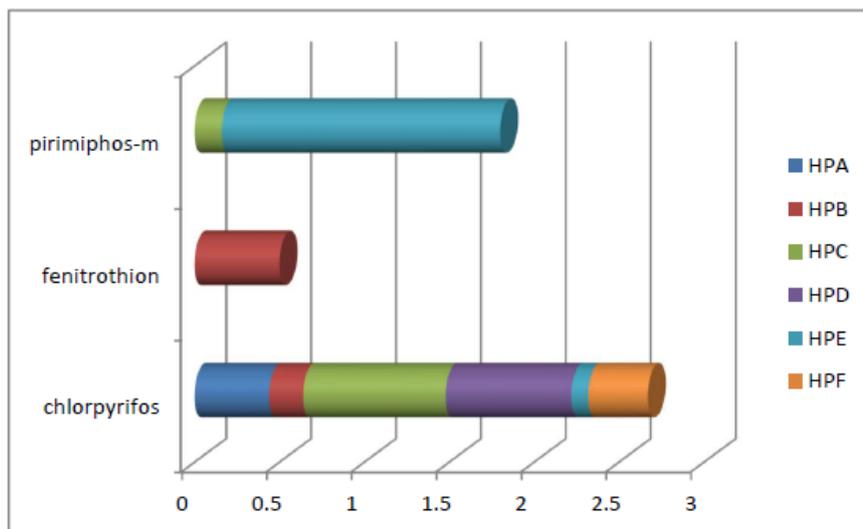
**Table 1.** Pesticide residues detected and quantified from the herbal products

Sample	Pesticide class	Pesticide name	Conc. In Sample (mg/kg)	Retention time	Mean Conc. (mg/kg)	MRLs
HPA	organophosphorus	Chlorpyrifos	0.022	10.72	0.022	0.05
HPB	organophosphorus	Chlorpyrifos	0.01	10.715	0.01	0.05
	organophosphorus	Fenitrothion	0.05	11.09	0.05	0.1
HPC	pirimiphos-m	pirimiphos-m	0.008	10.486	0.008	0.05
	organophosphorus	Chlorpyrifos	0.042	10.711	0.042	0.05
HPD	organophosphorus	Chlorpyrifos	0.037	10.706	0.037	0.05
HPE	organophosphorus	pirimiphos-m	0.082	10.476	0.082	0.05
	organophosphorus	Chlorpyrifos	0.005	10.703	0.005	0.05
HPF	organophosphorus	Chlorpyrifos	0.0175	10.702	0.0175	0.05

Chlorpyrifos pesticide was identified in all the six herbal products used in the study. This indicates that Chlorpyrifos is a commonly used pesticide in Ghana. Though its' use is legal, care must be exercised during usage to reduce human exposure and health risk. Some legally banned pesticides in Ghana including Aldrin, dieldrin and chlordane, ("EPA Bans 25 Dangerous Agro-Chemicals," 2008) were also identified in 50% of the herbal products. This may indicate their continue use in the country despite the ban of their usage in the country Authorities are therefore called on to regulate their importation and usage in the country.

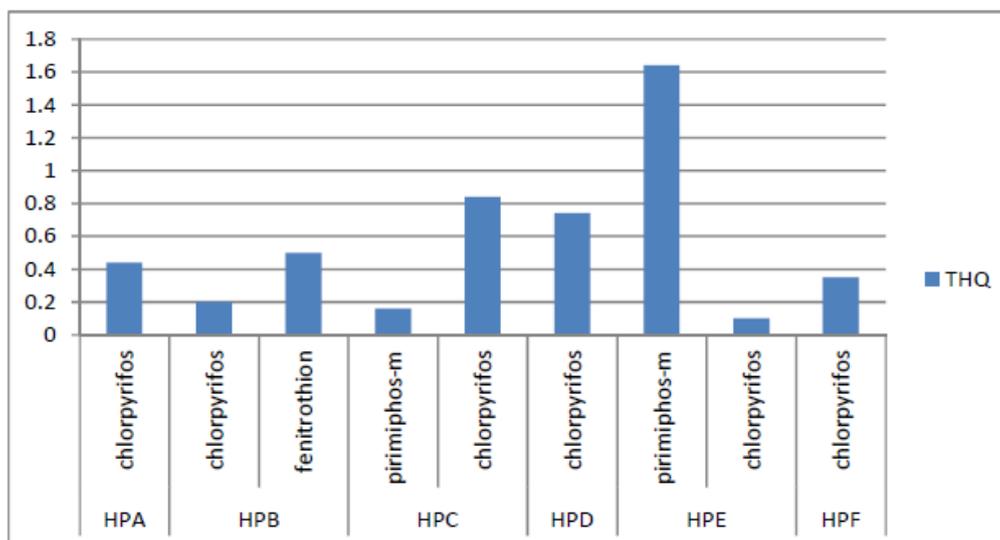
**Table 2.** Qualitatively Identified Pesticides from the herbal preparations

HPA	HPB	HPC	HPD	HPE	HPF
<b>Chlorpyrifos</b>	Chlorpyrifos	gamma HCH	chlorpyrifos	Allethrin	gamma HCH
	fenitrothion	aldrin		heptachlor	heptachlor
		dieldrin		Aldrin	chlorpyrifos
		lambda-cyhalothrin		Permetrin	pirimiphos-m
		permetrin		chlorpyrifos	aldrin,
		pirimiphos-m		pirimiphos	gamma chlordane
		Chlorpyrifos			dieldrin ppDDE
					lambda-cyhalothrin
					cypemethrin



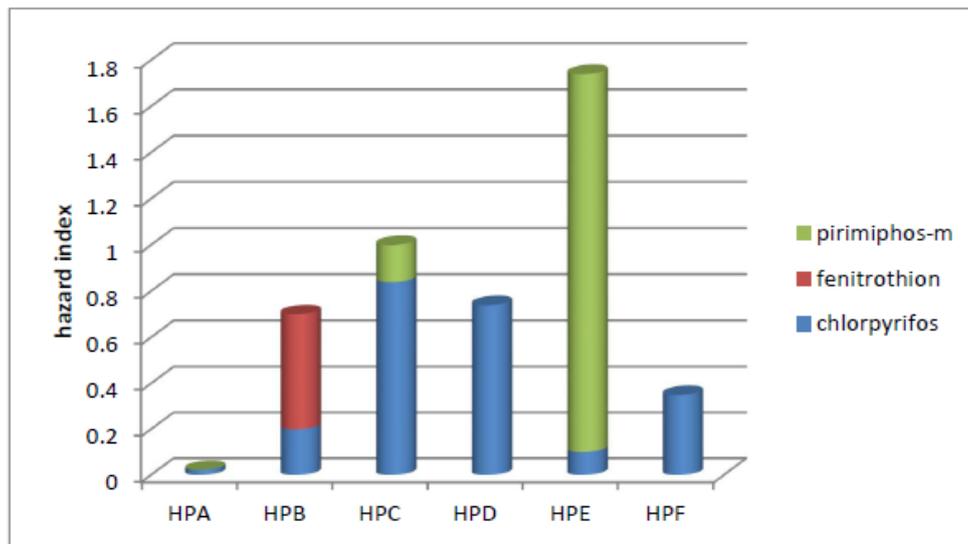
**Figure 4.** Pesticides quantitatively measured from HPs

Identification and quantitative estimations based on external standards were carried out on the pesticides. It was realized that pirimiphos-methyl was quantitatively identified in HPA and HPC, Fenitrothion in HPB only and chlorpyrifos in HPA-HPF. Exposure to one or more of these pesticides at high concentrations has been associated with differing health conditions, (Ngoula et al., 2007). Fenitrothion has been reported to have toxic effects to the liver, brain, lung, and kidney, (Abdel-Ghany et al., 2016) Exposure to one or more of these pesticides should therefore be monitored to avoid any serious health problems.



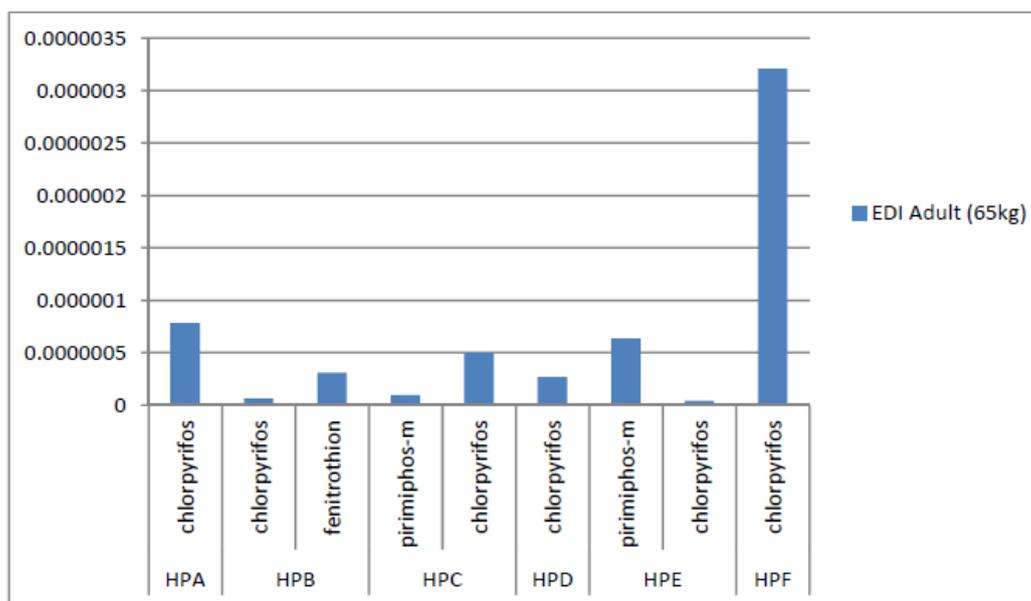
**Figure 5.** The targeted hazard quotient (THQ) of the individual pesticide present in the herbal products

THQ is used to express the health risk posed by a pesticide to a unit '1'. If the THQ is equal to or higher than 1, (Wang et al., 2005) there is a potential health risk to the exposed population to the related pesticide and related interventions and protective measures needed to be taken to protect the population. In this study however, most of the THQ values for most of the pesticides were less than 1. This indicates that the individual pesticides in the preparation do not pose health risk over the long term exposure to the herbal preparations. However, the THQ for pirimiphos-methyl for HPE was 1.6. This is far above the safe level of 1.0 and indicates that, the population administering HPE are at risk to pirimiphos-methyl risk over a long-term exposure.



**Figure 6.** Hazard index (HI) of the pesticides present quantitatively in the herbal preparations

The chronic hazard index (HI) is the sum of more than one hazard quotient for multiple toxicants present in a herbal preparation. THQs can be summed across constituent pesticides to generate a hazard index (HI) for an oral dosage-pathway combination. The Total hazard risk of the herbal products based on pesticide contents were assessed and referenced to '1'. If the HI value is less than 1, the exposed population is unlikely to experience any adverse health hazard in their lifetime. However, if the THQ is equal to or higher than 1, (Wang et al., 2005) there is a potential health risk to the exposed population. The HI for all the herbal products was less than '1' and safe for consumption without any future health risk. But, the HI for HPE was greater than '1' probably due to the high pirimiphos-methyl content. This observation indicates the population using HPE are at risk of possible adverse health consequences due to long-term exposure to pirimiphos-methyl.

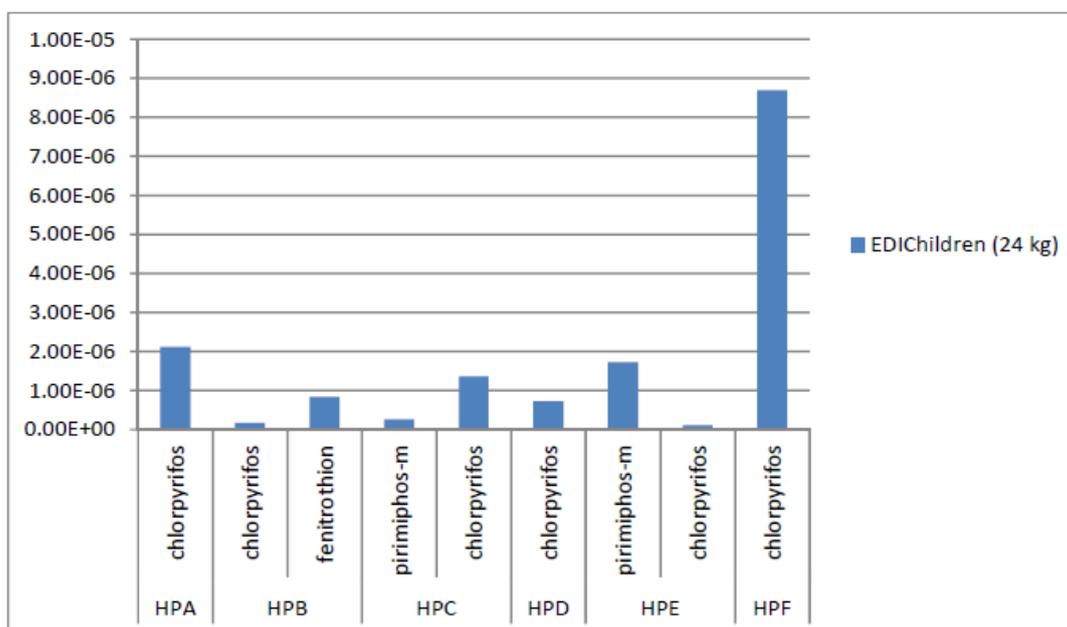


**Figure 7.** Adult estimated daily intakes (EDI) for the pesticides

**Reference values**

Pesticide	ARfD	ADI
Chlopyrifos-methyl	0.1 mg/kg bw	0.01 mg/kg bw/day, (Pubchem, n.d.)
Pirimiphos-methyl	0.15 mg/kg bw	0.004 mg/kg bw/day, (Pubchem, n.d.)
Fenitrothion	0.013 mg/kg bw	0.005 mg/kg bw/day, (Pubchem, n.d.)

The intake of the pesticides on the daily basis based on the dosage of the herbal preparations and the adult body weight was generally low. The EDI values were all below the international short-term oral reference dose limits for Chlopyrifos (0.1 mg/kg bw), Pirimiphos-methyl (0.15 mg/kg bw) and Fenitrothion (0.013 mg/kg bw). This indicates that the intake of the herbal products based on the prescribed dosages on the product bottles may not pose any acute health risk. The intake concentrations were also within acceptable intake values; Chlopyrifos (0.01 mg/kg bw/day), Pirimiphos-methyl (0.004 mg/kg bw/day) and Fenitrothion (0.005 mg/kg bw/day). This observation indicates that intake of these herbal preparations under investigation may not pose any mid/ long-term health problems to the public.



**Figure 8.** Estimated daily intakes (EDI) of the pesticides for Children

**Reference values**

Pesticide	ARfD	ADI
Chlopyrifos-methyl	0.1 mg/kg bw	0.01 mg/kg bw/day, (Pubchem, n.d.)
Pirimiphos-methyl	0.15 mg/kg bw	0.004 mg/kg bw/day, (Pubchem, n.d.)
Fenitrothion	0.013 mg/kg bw	0.005 mg/kg bw/day, (Pubchem, n.d.)

The child intake of the pesticides on the daily basis based on the dosage of the herbal preparations and the children’s body weight (24 kg) was generally low. It was observed that, the intake concentrations were also within acceptable intake values; Chlopyrifos (0.01 mg/kg bw/day), Pirimiphos-methyl (0.004 mg/kg bw/day) and Fenitrothion (0.005 mg/kg bw/day). This indicates that intake of these herbal preparations under investigation may not pose any mid/ long term health problems to the public. The EDI values were all below the international short-term oral reference dose limits for Chlopyrifos (0.1 mg/kg bw), Pirimiphos-methyl (0.15 mg/kg bw) and Fenitrothion (0.013 mg/kg bw). This observation indicates that the intake of the herbal products based on the prescribed dosages on the product bottles may not pose any acute health risk.

## Conclusion

Except for HPE, the total pesticide content of the herbal preparations mostly patronized and used for managing malaria, diabetes and hypertension in Ghana was within internationally acceptable safety limits. The concentration of pirimiphos-methyl in HPE was higher than the MRL. Data from our study suggest that consumption of HPE may predispose to pirimiphos-methyl exposure and possible health hazard following chronic use of this herbal preparation. Similarly, some banned pesticides including Aldrin, dieldrin and chlordane were identified in 50% of the studied herbal preparations. This should be of public health concern and therefore calls for urgent intervention by relevant regulatory authorities to forestall possible adverse health consequences.

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