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Quantification, distribution and cancer risk assessment of pesticides in mango orchards of Kerala, India

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ABSTRACT

Muthalamada in the Western Ghats has been a prominent mango-cultivating region of Asia for the last three decades. In the present study, soil samples from Muthalamada mango orchards were analysed for pesticide contents and estimated associated cancer risk indices. Sixteen pesticides, including organochlorines, organophosphates, synthetic pyrethroids, and carbamates were detected in the region. The concentration of chlorpyrifos (2.05–720.27 mg/kg) was the highest level reported from any agroecosystem in India. Community health risk assessment indicates high carcinogenic risk and greater susceptibility in children. The study demonstrates the need for immediate interventions to reduce the ecological and human cancer risks in the mango orchards.

KEYWORDS
Pesticide; soil; carcinogen; Muthalamada: health risk

Introduction

With the development of agricultural production, large quantities of pesticides are being deliberately released into the environment. By the year 2050, the use of pesticides will be approximately 2.7 times greater than that of the year 2000 [1,2]. The indiscriminate application of pesticides in agricultural fields causes contamination of environmental matrices and results in bioaccumulation and biomagnification through the food chain [3,4]. Some of the organochlorines and organophosphates are banned owing to their persistency, toxic degradation products, and lipophilic characteristics. It is reported that less than 0.1% of the pesticides applied reach the target pest, leaving 99.9% in the environment [5–11]. Pesticides are retained in soil mostly by adsorption to a solid surface, the presence of polar and non-polar groups in the soil, van der Vaal dispersion forces, ion exchange, interaction with metallic ions, charge transfer, and hydrophobic effects, and degradation occurs through physical, chemical, and biological processes. The degraded pesticide metabolites generally show less bioactivity than the parent pesticides,

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but in exceptional cases, metabolites showing greater bioactivity have also been observed [12]. These contaminants are considered hazardous to aquatic organisms, birds, and human beings [13–19].

The pest problem is more severe in tropical countries, as the climate provides favourable temperature and moisture conditions [20], resulting in increased demand for pesticides. Pesticide residues have been detected in a wide range of matrices, from drinking water to human breast milk from India [21–23]. A few studies have reported organochlorines and their metabolites from different parts of Kerala [24–28].

Soil properties play a major role in the dispersion and degradation of pesticides in agricultural areas [29]. Pesticides get absorbed into the soil components, transported to different ecological matrices, including the food chain, and ultimately reach the human body [30–32]. In agriculture fields, farmers are exposed to pesticide-contaminated soils through different pathways such as dermal contact, direct ingestion, and inhalation [33]. The probability of cancer can be estimated using indices such as hazard quotient (HQ), incremental lifetime cancer risk (ILCR), etc. based on the concentration of pesticide present in the soil [34–36]. There is growing evidence of human health risks from soil pesticide pollution in agricultural regions [37,38]. In the Kerala context, studies have shown a higher cancer risk associated with non-dietary ingestion of pesticides [39]. Mango orchards in the Western Ghats provide a livelihood for thousands of people. So far, no attempts have been made to estimate the ecological and human health risks associated with intensive pesticide application in the region.

Mangifera indica Linn is one of the most popularly cultivated fruits of the tropical and sub-tropical regions and is grown in more than 100 countries. Muthalamada Mango Orchard (MMO) is the only mango orchard in Kerala, located in Palakkad district close to the Tamil Nadu-Kerala border. Community health surveys conducted among farmers reported health impacts from pesticide exposure among local people [40]. Since the households of the MMO are located inside the mango orchards, the human population is constantly exposed to the contaminated environment. The available literature shows that studies on pesticide contamination and its ecological and human health impacts in several agro-ecosystems in Kerala are limited and extremely scarce. The present study is an attempt to analyse the pesticide residues in soil both qualitatively and quantitatively and associated risk factors along the Muthalamada mango-cultivating regions.

Materials and methods

Study area

The Muthalamada Mango Orchards (MMO) are situated near the western side of Palakkad District in central Kerala, India (10° 38′0″N and 76° 48′ 0″ E) (Figure 1). The region is a part of the Western Ghats, an important biodiversity hotspot. The area has a humid tropical climate and receives approximately 1500 mm of rainfall annually, with a major contribution from the southwest monsoon. The temperature of the region ranges between 28°C and 33°C, and extreme heat events are frequent during the summer seasons. Traditionally, paddy was the major crop in this region, along with vegetables, millets, and grains. During the past 20 years, most of the paddy fields have been replaced by mango plantations. Currently, nearly more than 3000 acres of mango orchards spread

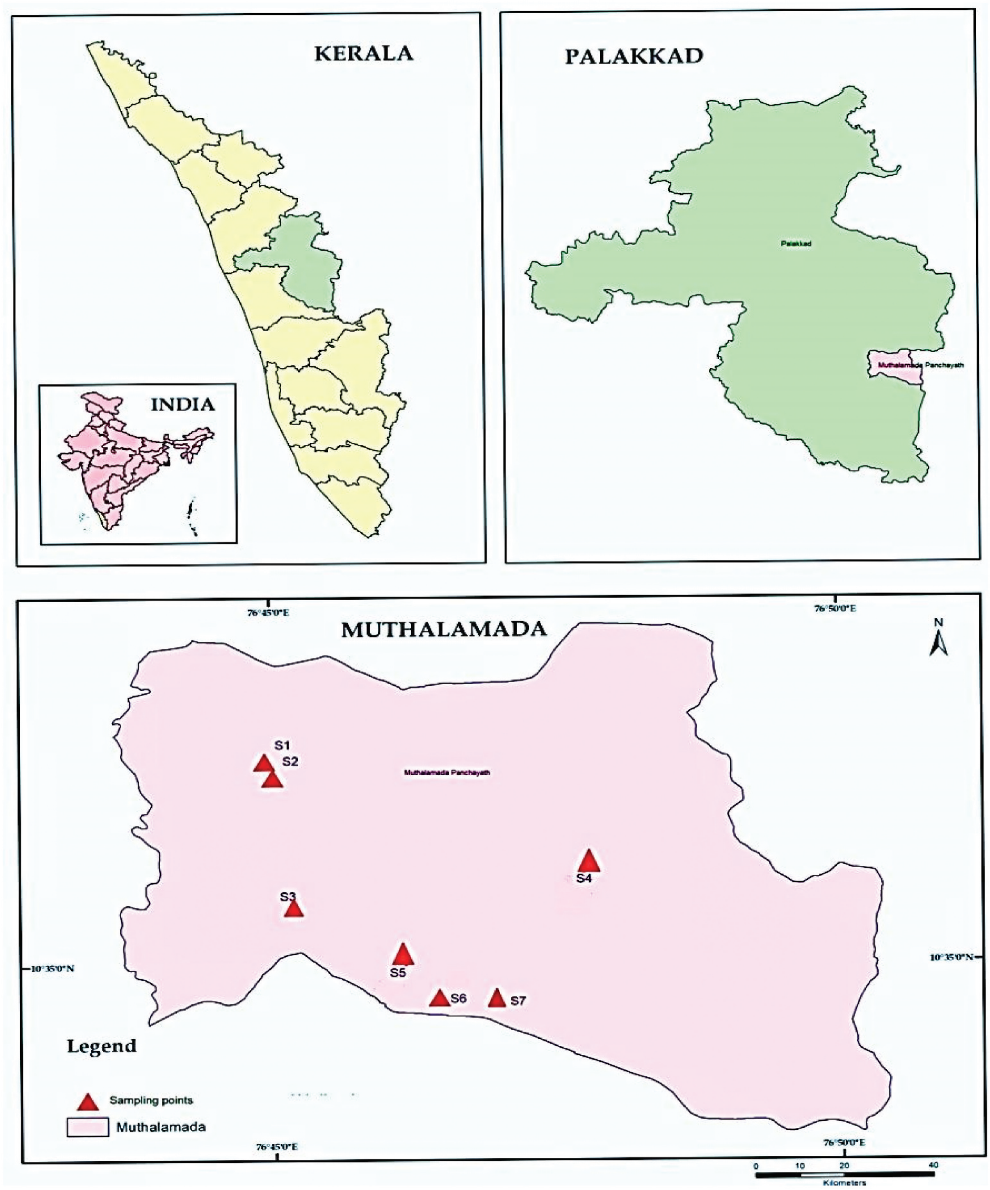

Figure 1. Map of study area showing sampling locations.

over this region produce about 20,000 tonnes of mangoes every year, which are valued at about three billion Indian rupees. Alphonso, Sindhooram, Benganapalli, and Neelam are the major grafted mangoes cultivated in the orchards. Most of the plantations are operated by large-scale planters by leasing land from local people, who provide the labour. Soil samples were randomly collected from six different locations in Muthalamada Grama Panchayat. One sample from the forest area was taken as the control site (pre-harvest time). About 500 g of soil were collected from each location at 0 to 15 cm depth and one metre away from the main tree trunk [41]. Samples were

Table 1. Details of soil sampling stations of Muthalamada mango orchards.

Sl no	Station	Location	Remarks
1	Station 1	10.61649°N, 76.74899°E	20-year-old mango plantation. Weeds were removed as part of intensive weed management.
2	Station 2	10.61386°N, 76.75011°E	Near an irrigation dam. Un-weeded area
3	Station 3	10.59178°N, 76.75096°E	5 to 10 year old mango plantation. Practising intensive weed management.
4	Station 4	10.59178°N, 76.75844°E	25-year-old mango plantation in a reclaimed paddy field
5	Station 5	10.57423°N, 76.77423°E	5-year-old mango plantation. Practising intensive weed management.
6	Station 6	10.57423°N, 76.77423°E	20 year old mango plantation. The area was un-weeded.
7	Station 7	10.57427°N, 76.774230°E	Control site. Near to the forest area

packed in plastic bags, brought to the laboratory on the same day, and preserved until the analysis. Table 1 shows descriptions of sampling sites. The samples were air-dried at room temperature, ground, and sieved (2 mm mesh).

Pesticide analysis

Acetone-shake flask method was used for the extraction of pesticides from the soil samples. An amount of 10 g of air-dried soil was taken in an Erlenmeyer flask and suspended in 20 ml of high performance liquid chromatography (HPLC) grade acetone, followed by shaking for 2 h. The suspension was filtered through Whatman No. 41 filter paper, centrifuged, and passed through a chromatographic column (sintered) packed with high-purity anhydrous sodium sulphate, silica gel, and activated alumina [42]. Subsequently, the extracts were concentrated in a rotary vacuum evaporator at 50°C at 500 psi, transferred to pre-cleaned 100 µl glass vials, sealed using parafilm to avoid further contact with air, and refrigerated until further analysis. Pesticide detection was done using a 7693 Auto sampler, Agilent Technologies Series gas chromatograph with helium carrier gas (99.9995) for separation of the pesticide.

Instrumental analysis

The pesticides were analysed by gas chromatography/mass spectrometry (GC-MS) (7693 Auto sampler, Agilent Technologies) and liquid chromatography – mass spectrometry (LC- MS MS) (ULTIVO 6465) facilities available at Sea Lab Pvt. Ltd., Kerala (accredited NABL, as per the ISO 17,025:2005 Standard). The samples were analysed for quantification of organochlorine pesticides (OCP), organophosphates (OP), synthetic pyrethroids (SP), and carbamates. The instrumental conditions are given as supplementary data.

Sample preparation and clean-up procedure

Working standard solutions of appropriate concentrations were prepared from stock solution (1000 µg/mL) by serial dilution, which was used for instrument calibration. The pesticide concentrations were quantitatively determined by the external standard method using the peak area of the sample. The peak identifications were based on the accurate retention time and m/z value of each analyte. Linearity was assessed using a series of pesticide standards (organochlorine, organophosphate, carbamates, and synthetic pyrethroids) of increasing concentration and acquired for all the pesticides (r^2 ranged from 0.987 to 0.997). A standard pesticide solution and solvent blank were injected into the GC-MS and LC-MS MS before the analysis of soil sample extracts. Concentrations above the limit of quantitation (LOQ) limit were taken for calculation, and those below the LOQ were taken as zero.

Method of quantifying recovery rate

The average recovery experiments were carried out by weighing 10 g of soil in a 50-mL centrifuge tube and spiked with different concentrations of mixed pesticides. The soil was shaken to make a homogeneous mixture of pesticides. The pesticide-spiked soil samples were air-dried at room temperature, kept for a day, and then analysed to assess the recovery rate. The sample was extracted with 1% acetic acid in acetonitrile, followed by the QuEnChERS extraction method with extraction pouches containing 6 g of anhydrous magnesium sulphate and 1.5 g of sodium acetate. Extraction was followed by dSPE clean up with 150 mg anhydrous magnesium sulphate, 50 mg PSA (primary secondary amine), 50 mg C18, and 7.5 mg GCB (graphitised carbon black), and finally injection of GC-MS and LC-MSMS. The target compounds were not detected when the blank samples were analysed. The average recovery of pesticides ranged from 70% to 120%. The limit of detection (LOD) of individual pesticides ranged from 0.1 ng/g to 1.08 ng/g, respectively, and the LOQ was 10 ng/g.

Health risk assessment

Health risk assessment predicts the probable effects of toxicants on human beings over a specified period. Individuals can be exposed to contaminated soils through several pathways, including ingestion, inhalation, and dermal contact. In this study, a health risk assessment model recommended by the U.S. Environmental Protection Agency (USEPA) [42] was applied to calculate the carcinogenic risks for adults and children from non-dietary exposure to pollutants. This method is widely used for assessing health and cancer risk in human beings associated with pesticide exposure in various agricultural regions of the world [43]. The model uses the concentrations of pesticides present in the soils to assess human health risks; thus, the number of variables is minimal, which provides a more accurate estimation of risk. For organochlorine and carbamate pesticides, cancer risk through ingestion, dermal contact, and inhalation of soil particles was estimated using the following equations:

$$CR_{Ingest} = C_{soil} \times IngR \times EF \times ED/BW \times AT \times CF_{soil} \times SF_{soil}$$

$$\begin{aligned} CR_{Dermal} = {} & \left(C_{soil}1 \times Surface\ rea\left(cm^2\right) \times Skin\ Adherence\ \left(mgcm^2\right)\right. \\ & \left.\times Dermal\ Absorption\ factor/BW \times AT \times CF \times SF_{soil}\right) GIABS \end{aligned}$$

$$CR_{Inhale} = \left(C_{Soil} \times SA \times InhR \times AFInh \times EF \times \frac{ED}{PEF} \times AT\right) IUR$$

The particle emission factor (PEF) is the concentration of toxins adsorbed to inhalable particles (PM10). The assessment of cancer risk through ingestion, dermal contact, and inhalation was based on a human life span of 70 years. Soil ingestion rates ($I_{ng}R$) of 100 mg d^{-1} and 200 mg d^{-1} were used for adults and children, respectively. Exposure duration (ED) of 70 years for adults and 12 years for children was based on the average lifespan, and an assumed exposure frequency (EF) of 350 days/year, excluding 15 days of holidays, was adopted. The average time (AT) was calculated as 25,550 (70 × 365) days for adults and 2190 days for children. A body weight of 70 kg for adults and 27 kg for children was selected. The contact surface area of skin with soil was set at 3300 cm^2, assuming the hands and arms were exposed to the soil. The skin adherence factor for soil (AF) was 0.2 mg cm^2. The inhalation rate was 15.8 m^3d^{-1} for adults. The value of the fraction of contaminants absorbed in the gastrointestinal tract (GIABS and $AF_{Inh)}$ was set at 1 for the preliminary risk assessment. Dermal absorption factor (DBF) is chemically specific; its value is 0.13.

Lifetime Average Daily Dose (LADD), Incremental Lifetime Cancer Risk (ILCR), and Hazard Quotient (HQ) were calculated by the following equations:

$$LADD = CS \times IR \times CF \times EF \times ED/BW \times AT$$

$$ILCR = LADD/CSF$$

$$HQ = LADD/RfD$$

Details of different parameters used for risk assessment are given as supplementary data.

Results

The pesticide analysis revealed the presence of pesticide contamination in the soils of MMO, encompassing various categories such as organophosphates, carbamates, synthetic pyrethroids, and organochlorines. Sixteen pesticides were identified within the MMO soil samples, and their enumeration is provided. The chromatograms depicting the specific pesticides are included in the appendix for reference.

Significantly, the investigation identified a range of key pesticides prevailing in the region. These prominent pesticides encompass Aldicarb-Sulphonate (also known as aldoxycarb), Hexachlorobenzene (HCB), Alpha-Benzene Hexachloride BHC-α, Beta- Benzene Hexachloride BHC-β, chlorobenzilate, O,P'- Dichlorodiphenyldichloroethylene (O.P'-DDE), malathion, bifenthrin, cyhalothrin (lambda), imidacloprid, diuron, and chlorpyrifos. No pesticide residues were detected in the soil samples collected from the control site during the study.

The results of the One-way Analysis of Variance (ANOVA) presented in Table 2 indicated statistically significant differences ($p < 0.01$) in pesticide concentrations among the designated stations. This finding provides empirical support for the alternative hypothesis, indicating that there are substantial variations in pesticide concentrations across the different stations. Subsequent analysis using the Least Significant Difference (LSD) test further affirmed the existence of significant distinctions in pesticide concentrations among the stations. When denoted by the same alphabetical values, no statistically significant differences were observed. Conversely, divergent alphabetical values indicated significant variations. Notably, specific, significant differences were identified for various pesticides at specific stations. These include Diuron at S6, Cyhalothrin at S5

Table 2. Concentrations of pesticides (mg/kg) in the soil samples collected from Muthalamada mango orchards.

		Pesticide concentration (mg/kg)						
Sl no	Pesticides	S1	S2	S3	S4	S5	S6	Pr(>F)
1	Diuron	0.21[b]	0.21[b]	0.22[b]	0.22[b]	0.22[b]	1.21[a]	<0.01
2	Aldicarb-sulphonate (Aldoxycarb)	5.61	ND	ND	ND	ND	ND	-
3	Cyhalothrin (lambda)	2.94[c]	0.42[d]	2.19[c]	0.34[d]	8.33[a]	5.22[b]	<0.01
4	Thiamethoxam	*	1.51	*	*	*	*	-
5	BHC alpha isomer	0.14[b]	0.00[a]	0.14[b]	0.00[a]	0.00[a]	0.00[a]	<0.01
6	HCB	0.17[a]	0.00[b]	0.00[b]	0.00[b]	0.17[a]	0.00[b]	<0.01
7	BHC Beta isomer	0.00[b]	0.00[b]	0.00[b]	0.00[b]	0.20[a]	0.00[b]	<0.01
8	Malathion	0.01[b]	0.00[c]	0.00[c]	0.04[a]	0.00[c]	0.00[c]	<0.01
9	Imidacloprid	*	6.48	*	*	*	*	-
10	Chlorpyrifos	24.68[b]	2.05[b]	3.40[b]	720.27[a]	37.72[b]	55.63[b]	<0.01
11	O,P-DDE	0.00[b]	0.00[b]	0.35[a]	0.00[b]	0.00[b]	0.00[b]	<0.01
12	Metalaxyl	*	0.02	*	*	*	*	-
13	Bifenthrin	5.43[c]	0.80[d]	10.03[b]	13.90[b]	28.47[a]	24.99[a]	<0.01
14	Triazophos	*	0.07	*	*	*	*	-
15	Thiophanate-methyl	*	1.02	*	*	*	*	-
16	Chlorobenzilate	2.00[a]	0.00[b]	0.00[b]	0.00[b]	0.00[b]	0.00[b]	<0.01

ND: Not detected * Below Detection Level, Pr F value: probability of F value, a b c d: Least Significant Difference symbols for comparison pesticides values for comparison.

and S6, BHC alpha isomer at S1, S2, and S3, HCB at S1 and S5, BHC Beta isomer at S5, Malathion at S4, Chlorpyrifos at S4, O,P'-DDE at S3, Metalaxyl, Bifenthrin at S5, and Chlorobenzilate at S1. These findings underscore the varying levels of pesticide contamination across the sampling stations.

Chlorpyrifos was detected in high concentrations across all studied stations. Station S1 showed contamination with BHC (0.14 mg/kg), HCB (0.17 mg/kg), chlorobenzilate (2.00 mg/kg), organophosphates (chlorpyrifos (24.68 mg/kg), malathion (0.01 mg/kg)), and synthetic pyrethroids including diuron (0.21 mg/kg), cyhalothrin (lambda) (2.94 mg/kg), and bifenthrin (5.43 mg/kg). Aldicarb-sulphonate was found in high concentration (5.61 mg/kg). BHC-α (0.14 mg/kg), O, P-DDE (0.35 mg/kg), chlorobenzilate (2.00 mg/kg), BHC-β (0.20 mg/kg), and HCB (0.17 mg/kg) were noticed in stations S1, S3, and S5. Pyrethroid pesticides, such as cyhalothrin (lambda), bifenthrin, imidacloprid, diuron, thiamethoxam, triazophos, and metalaxyl, were found in all stations. Station S5 had a high concentration of bifenthrin (28.47 mg/kg) and cyhalothrin (lambda) (8.33 mg/kg). Chlorpyrifos contamination was observed in the order of S4>S6>S5>S2>S1. Additionally, elevated concentrations of chlorpyrifos (720–37.72 mg/kg), diuron (2.22–2.22 mg/kg), and bifenthrin (13.90–28.47 mg/kg) were detected in all stations. Soil contamination was also evident with diuron (1.21 mg/kg), Thiophanate-methyl (1.02 mg/kg), and triazophos (0.07 mg/kg). Detected pesticide GC MS and LC MS MS chromatograms are provided as supplementary data.

Health risk assessment

This study assessed exposure to BHC metabolites, Dichlorodiphenyltrichloroethane (DDT) metabolites, chlorobenzilate, HCB and aldicarb-sulphonate through the soil ingestion pathway; because these compounds are hydrophobic and have a higher affinity for soil particles. Therefore, contaminated soil was considered the major route of exposure for the human population. The farmers in Muthalamada are expected to have a comparatively higher exposure to the pesticides through soil ingestion. Human health risk as incremental lifetime cancer risk (ILCR) and non-cancer hazard quotient (HQ) were evaluated by calculating the incremental lifetime average daily dose (LADD) of each pesticide, and the results are provided in Tables 3 and 4. According to USEPA [42], lifetime average daily dose (LADD) is an amount of assumed chemical intake by a person (per kg of bodyweight per day) that has adverse health effects when absorbed into the body over a long period of time. The estimated LADD of adults for BHC- α and BHC- β were 0.2×10^{-6} and for HCB, 0.001×10^{-6}–0.24×10^{-6}, and that of children was 1.03×10^{-6} to 1.48×10^{-6}. The estimated LAAD of O,P'-DDE for adults and children was $0.5 \times 10^{-6}$and 2.59×10^{-6} respectively and chlorobenzilate and aldicarb sulphonate showed very low risk from exposure. The incremental lifetime cancer risk (ILCR) for adults and children ranged between 0.02×10^{-3} to 0.2×10^{-3} for BHC α and β. For O,P'-DDE it was 0.7×10^{-3} for adults and 0.37×10^{-3} for children. The Hazard Quotient indicates the degree of exposure potential for developing non-carcinogenic health effects in human beings over an average lifespan. The hazard quotients of HCB for adults and children were estimated as 5×10^{-3} and 0.36×10^{-3}.

According to the USEPA standards, when the chronic daily intake (CDI) is greater than the reference dose (RfD) of the contaminant via each exposure route, this would lead to an adverse human health effect. The present study showed that non-carcinogenic risks to adults and children by OCP pose moderate risk, with no dietary exposure [44]. For OCPs, the health risk from dietary exposure was far higher. The permitted value of ILCR is between 10^{-6} and 10^{-4}, denoting less potential for cancer risk. The ILCR value of 10^{-6} indicates that it is virtually safe, and 10^{-4} indicates low health risk [45]. In the study, BHC-α, BHC-β, O,P'-DDE, chlorobenzilate, and aldicarb-sulphonate showed a moderate risk. A similar trend was earlier reported from India and other parts of the world [46–51] which indicates that the level of contamination in MMO is significantly high and should immediately be cut. LADD of pesticide showed a decreasing trend in the order of Aldicarb>HCB>O,P'-DDE, PDDE>BHC-α>BHC-β>Chlorobenzilate. The results indicated that contaminated soils could pose a greater cancer risk to children than adults.

Discussion

The levels of carbamates, synthetic pyrethroids, and organophosphates were high compared to organochlorines, probably because of restrictions or bans on several organochlorine compounds. The variation of pesticide concentration in samples collected from different stations can be attributed to the mode of application by farmers (usually, they mix more than five pesticides in a tank and spray on the mango trees during flowering time). The un-weeded locations showed higher pesticide content, probably because photo degradation was less. Factors such as the slope of the area, drainage, surface characteristics, etc. also affect the concentration of pesticides in the soil.

Monocrotophos, chlorpyrifos, malathion, and phosphamidon were detected in human blood samples collected from the agricultural regions of Punjab. Levels of 8.37 ppb of O, P-DDE were reported from the Andaman Islands [16]. Another study from Assam reported 0.288 ± 0.26 mg/kg of O,P-DDE in the agricultural soil [13]. Kumar et al. [52] reported 0.034 ± 0.0058 mg/kg of BHC α and 0.00071 ± 0.0002 mg/kg of O,P-DDE from

Table 3. Carcinogenic risk assessment of organochlorine and carbamate pesticides in adults of MMO.

Sl no	Pesticide	CRingest	CRderml	CRinhale	LADD	ILCR	HQ
1	BHC-α	0.14×10^{-6}	0.22×10^{-6}	0.9×10^{-9}	0.2×10^{-6}	0.02×10^{-3}	0.1×10^{-6}
2	HCB	0.02×10^{-6}	0.08×10^{-6}	0.06×10^{-9}	0.001×10^{-6}	0.14×10^{-6}	5×10^{-3}
3	HCB	0.48×10^{-6}	1.37×10^{-6}	1.12×10^{-9}	0.24×10^{-6}	34×10^{-6}	0.12×10^{-6}
4	BHC-β	0.57×10^{-6}	1.13×10^{-6}	1.32×10^{-9}	0.2×10^{-6}	0.02×10^{-3}	0.1×10^{-6}
5	O,P′ -DDE	1.00×10^{-6}	13.2×10^{-6}	2.31×10^{-9}	0.5×10^{-6}	0.7×10^{-3}	0.25×10^{-6}
6	Chlorobenzilate	0.02×10^{-6}	0.03×10^{-6}	0.06×10^{-9}	0.01×10^{-6}	0.001×10^{-3}	0.005×10^{-6}
7	Aldicarb- sulphonate	16.02×10^{-6}	1.04×10^{-6}	37.14×10^{-9}	1.37×10^{-6}	0.19×10^{-3}	0.45×10^{-3}

CRingest - Cancer Risk via ingestion, CRdermal - Cancer Risk via dermal contact, CRinhale - Cancer Risk via inhalation, LADD - Life time Average Daily Dose, ILCR - Incremental Lifetime Cancer Risk, HQ - Hazard Quotient.

Table 4. Carcinogenic risk assessment of organochlorine and carbamate pesticides in children of MMO.

Sl no	Pesticide	CRingest	CRderml	CRinhale	LADD	ILCR	HQ
1	BHC-α	2.06×10^{-6}	0.42×10^{-6}	0.9×10^{-6}	1.03×10^{-6}	0.14×10^{-3}	0.5×10^{-6}
2	HCB	0.14×10^{-6}	0.15×10^{-6}	0.06×10^{-9}	0.07×10^{-6}	10×10^{-6}	0.36×10^{-6}
3	HCB	2.51×10^{-6}	2.56×10^{-6}	1.12×10^{-9}	1.25×10^{-6}	0.17×10^{-6}	0.62×10^{-6}
4	BHC-β	2.96×10^{-6}	2.10×10^{-6}	1.32×10^{-9}	1.48×10^{-6}	0.2×10^{-3}	0.74×10^{-6}
5	O,P′-DDE	5.18×10^{-6}	24.60×10^{-6}	2.31×10^{-9}	2.59×10^{-6}	0.37×10^{-3}	1.29×10^{-6}
6	Chlorobenzilate	0.86×10^{-6}	0.07×10^{-6}	0.06×10^{-9}	0.07×10^{-6}	0.01×10^{-3}	0.35×10^{-6}
7	Aldicarb- sulphonate	83.1×10^{-6}	32.58×10^{-6}	197.22×10^{-6}	41.55×10^{-6}	5.93×10^{-3}	13.85×10^{-3}

CRingest - Cancer Risk via ingestion, CRdermal - Cancer Risk via dermal contact, CRinhale - Cancer Risk via inhalation, LADD - Life time Average Daily Dose, ILCR - Incremental Lifetime Cancer Risk, HQ - Hazard Quotient.

intensive agricultural soils in North India. These values are lower than those in soil samples collected from the MMO. The presence of organochlorines and their metabolites indicates the historical application and higher quantity during the past decades. Similar findings can be observed for DDT and organochlorine metabolites reported from various parts of the world [53–55]. The OCP residues were still detectable in the soil samples more than 20 years after the bans for DDT, BHC, and HCB, demonstrating their persistence in the environment [56–61]. Pesticides such as aldoxycarb, HCB, BHC, chlorobenzilate, chlorpyrifos and malathion were listed as genotoxic and banned in several countries, including India. Akoijam and Singh [62] detected imidacloprid residues from the soils of the Punjab paddy fields up to 0.18 mg kg^{-1}, which is lower than the value obtained in the present study.

Another study found pesticide residues of chlorpyrifos (0.025–0.64 mg/kg), lambda-cyhalothrin (0.05–0.06 mg/kg), bifenthrin (0.14–1.23 mg/kg) and quinalphos (0.03–0.41 mg/kg) from cardamom-cultivated regions of Kerala [63]. Pesticide residues were estimated from mango orchards in Ratnagiri district, and reported imidacloprid residue in soil samples ranging from 0.8–2.3 ppm, carbendazim, monocrotophos, phorate, and hexaconazole [64]. Levels of 0.028 mg/kg of malathion, 4.35–30.15 mg/kg of diuron and 5.35–31.41 mg/kg of chlorpyrifos were detected from the pineapple farms in Kerala [65]. Several organochlorine pesticides were quantified from the surface soils of Palakkad district, Kerala. The dominant insecticides were endrin (64 ng/g), α-BHC (1.9 ng/g), β-BHC (9.7 ng/g), heptachlor (2.4 ng/g), HCB (15.9 ng/g) and DDT metabolites (5.4ng/g) [27]. The results of the present study exceed those detected in all the previous studies, especially in Kerala. Table 5 compares the levels of concentration (mg/kg) of the organochlorine pesticides in the soil from different parts of the world to those found in the present study (ND indicates the pesticides below detectable levels and NA represents

Table 5. Comparison of OCPs and Aldicarb – sulphonate concentration in the soils of MMO with other contaminated regions reported.

Area	BHC-α (mg/kg)	HCB (mg/kg)	BHC-β (mg/kg)	O,P-DDE (mg/kg)	Chlorobenzilate (mg/kg)	Aldicarb-sulphate (mg/kg)	Reference
Aligarh, UP, India	0.00179 ± 0.04735	NA	NA	0.00–.0071	NA	NA	Nawab et al. [12]
Assam, India	0.098 ± 1.945	NA	NA	0.288 ± .261	NA	NA	Mishra et al. [13]
Andaman Islands	NA	NA	NA	0.00–.00837	NA	NA	Murugan et al. [16]
Eloor-idayar region, Kerala	689µgm/gm	NA	690µgm/gm	NA	NA	NA	Divya et al. [18]
Kuttanad agro ecosystem-Kerala-India	0.00001–0.00955	NA	NA	0.00–.00218	NA	NA	Sruthi et al. [19]
Zhangzhou City, China	0.0043 ± 0.00529	NA	NA	0.00197 ± .00210	NA	NA	Dan et al. [66]
North India	0.03496 ± 0.00587	NA	NA	0.00071 ± .00027	NA	NA	Kumar et al. [52]
Argentina	0.0023 ± 0.0007	NA	NA	0.0035 ± .0006	NA	NA	Gonzalez et al. [53]
Jordan	NA	0.04 ± 0.01	NA	0.46±.05	NA	NA	Al-Mughrabi et al. [54]
North west China	NA	0.01171(ng/g)	NA	0.00052	NA	NA	Huang et al. [55]
Present study	ND-0.02	0.05 ± 0.03	ND-.03	ND-.05	ND-.001	ND-5.61	Present study

ND-Not detected, NA- Not analysed.

those not analysed in the referred study). The available literature clearly indicates that the ecological and human health risks from pesticide residues are higher in MMO than in several agroecological regions.

Commonly applied insecticides in mango orchards are endosulfan, parathion methyl, chlorpyrifos, cypermethrin and fenvalerate organochlorines such as BHC α, β, γ, δ-HCH, pp-DDE, op-DDT, pp-DDD and pp-DDT, which were frequently detected from mango orchards in Lucknow, India [67,68], and deltamethrin residues were detected from a mango orchard in Bangalore [69]. Phorate, malathion, chlorpyrifos, pendimethalin, butachlor, endosulfan-I, DDT isomers, lambda cyhalothrin, fipronil, atrazine, pretilachlor, propiconazole, and triazophos were detected in the soil samples taken from Haryana [70]. The present study is a pioneering attempt to quantify pesticides in mango orchards in Kerala. The assessment revealed a high concentration of pesticides compared to the soil samples assessed from other mango orchards in India. MMO soils, characterised by an acidic and basic pH (5–8.5) and moderate organic matter content, might have imparted more stability to pesticide molecules and enhanced the rate of adsorption. Stations that detected the prominent presence of organochlorine pesticides indicate their historical application and persistence, which is directly influencing the retention, transport, and degradation processes within the soil [71]. The absence of residues in the control samples indicates that the mango cultivation practices are the major source of pesticide contamination in the soil.

In several regions, the general population has reportedly shown detectable levels of pesticide metabolites in urine, indicating potential exposure from indirect sources, including dietary (drinking water, food) and non-dietary (dust, breathing air) exposures [72,73]. Since soil is the main source of various exposure pathways, the MMO region poses a health risk to the local population. The increasing trend in cancer incidence over the last 50–60 years may be largely attributed not only to the ageing of the population but also to the diffusion of carcinogenic agents in occupational environments [74–80]. A non-significantly higher risk of breast cancer was reported in women exposed to DDE [81]. The present study also reported the presence of DDE in the soil, indicating a similar risk exists in the area. Apart from DDE, HCB, BHC-α and BHC- β also pose carcinogenic risks in children [82–85], which are quantified in the present study. Among the organophosphate and carbamate pesticides detected, chlorpyrifos reportedly induces the proliferation of cancer cells [86]. Exposure to the carbamate insecticide aldicarb modulates human oestrogen and progesterone receptor activity in breast and endometrial cancer cells [87–89]. Studies have reported diuron and chlorpyrifos as carcinogenic in human beings, and these chemical substances were also reported in high concentrations in MMO soil. It is found that the occurrence of multiple myeloma was higher in male pesticide handlers, whereas that of melanoma was higher in females. Multiple myeloma, a haematopoietic malignancy of the plasma cells, is one of the most common haematological cancers in men and women and it is frequently expressed in pesticide applicators, although the causal factors are still poorly understood. Table 6 provides a list of published literature on the type of cancer reported from pesticide residues included in the current study [90–106].

Table 6. Type of cancer reported by the environmental exposure of pesticides (organochlorines, carbamates, organophosphate and synthetic pyrethroids) in different regions of the world.

Slno	Country	Specific population	Type of cancer reported	References
1	New Zealand	Pesticide applicators	Multiple myelomaNon-hodgkin's lymphoma Non-hodgkin's lymphoma	Mannetje et al. [90]
2	Iceland	Pesticide applicators	Adenocarcinoma	Zhong and Rafnsson [91],
3	Netherlands	Pesticide applicators	Multiple myeloma	Swean et al. [92]
4	United States	Pesticide applicators	Melanoma	Alavanja et al. [93]
5	Australia	Pest control workers	Pancreatic cancer	Beard et al. [94]
6	United States	Pesticide applicators	Multiple myeloma	Blair et al. [95]
7	United Kingdom	Farm residents	Multiple myeloma	Bradbury et al. [96]
8	Australia	Farm residents	Multiple myeloma	Depczynski et al. [97]
9	South Korea	Farm residents	Multiple myeloma	Lee et al. [98]
10	Spain	Farm residents	Multiple myeloma	Parron et al. [99]
11	Israel	Farm residents	Multiple myeloma	Atzmon et al. [100]
12	Florida	Citrus workers	Multiple myeloma	Nigg and stamper [101],
13	Egypt	Farm residents	Multiple myeloma	Tchounwou et al. [102]
14	North Carolina	Pesticide workers	Breast cancer	Duell et al. [103]
15	Eastern Slovakia	Farm residents	Multiple myeloma	Pavuk et al. [104]
16	India	Farm residence	Breast cancer	Rusiecki et al. [105]
17	India	Farm residents	Breast cancer	Mathur et al. [106]

Cancer risk assessment

The cancer risk for children and adults in the study area was evaluated by considering ingestion, inhalation, and dermal exposure routes [107]**. A value below to 1×10^{-6} is considered a negligible risk for cancer. OCPs such as BHC-α, BHC-β, HCB, O,P'-DDE, chlorobenzilate and aldicarb-sulphonate (aldoxycarb-carbamate pesticide) were taken for the health risk assessment. The study revealed that present levels of OCPs and aldoxycarb in MMO soils pose a higher risk to children than adults. Total ILCR values from the study sites ranged from 0.01×10^{-3} to 5.93×10^{-3} for children, which exceeds the target risk level of 1×10^{-6} [42], indicating potentially high health risk, especially for Aldicarb- Sulphonate and HCB. A similar trend was also observed for adults with a moderate level of cancer risk. The health risk assessment factors like LADD, ILCR, and HQ are very high among children in other agroecosystems of India, Iran, Kuwait, Southeast China, Southern Italy, Nigeria and Kenya [32,108–112]. Thus, community health managers need to focus on pesticide pollution in MMO. There is a higher cancer risk in children through interactions including mango fruit collection from the orchards and group activities such as playing and bathing in nearby public ponds in and around the mango orchards.

The current study indicates the need for an epidemiological survey to estimate cancer incidents among farmers and residents of MMO. Such a survey can only be conducted by the state health department or local self-governing departments (LSGDs). The data presented in the current study can serve as a baseline for epidemiological analysis and cancer risk mitigation in the study area. The results of this study concerning agricultural management imply that the OCPs and carbamate pesticides can be remobilised, thereby resulting in bioaccumulation and contamination of groundwater. A similar case was reported from Kasargod, Kerala, India [113]. Thus, there is a need for continuous monitoring of pesticides in agricultural soils as an initial control measure to reduce environmental and health risks.

Since high pesticide toxicity is prevalent in MMO, alternative pest control methods such as microbial pesticides and biopesticides are advisable. Studies showed that a microbial consortium can effectively eliminate the small leafhopper *Empoasca flavescens* (Homoptera: Cicadellidae) in mango plantations [114,115]. Integrated Pest Management (IPM) using trapping techniques such as lures or baits and clean cultivation methods can minimise the infestation of pests like mango nut weevil [116–118]. Implementation of such an alternative method will be helpful for minimising existing and future environmental risks as well as human health risks in MMO.

Conclusion

The present study demonstrates the ecological and human health risks associated with the contamination by pesticides in the soils of Muthalamada mango orchards. The mango cultivation in the region generates huge revenue for the farmers and provides a livelihood for thousands of people. High concentrations of pesticides in the soil are probably caused by their indiscriminate application and low mobility. There is an urgent need to cut pesticide use, follow manufacturers' advice faithfully, and investigate the cancer risk for adults and children in the region.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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