INNOVATIVE ENERGY TECHNOLOGY SYSTEMS AND ENVIRONMENTAL CONCERNS: A SUSTAINABLE APPROACH
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Pesticide Residue in Environment and its Management

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Abstract

Advantages associated with the application of pesticides in enhancing the agricultural productivity must be weighed against the possible health hazard arising from the toxic pesticide residues in food. First and foremost the application of pesticides should be in compliance with good agricultural practices, using only the required amounts. Further the current shift in world opinion from 'chemical farming' towards 'organic farming' is a sustainable approach to minimize the damage posed by widespread contamination of environment by pesticides [11]. However, the challenge lies in achieving food safety in developing countries where the indiscriminate application of pesticides results in the presence of residues in food commodities. However, due to several socio-cultural and technical reasons, diffusion and acceptance of this approach among the farming community in developing countries like India has been very slow.

1. Introduction

Pesticides (insecticides, fungicides, etc.) are used globally for the protection of food, fiber, human health and comfort [21]. However, their excessive use/misuse especially in the developing countries, their volatility, long-distance transports eventually results in widespread environmental contamination. In addition many older, non-patented, more toxic, environmentally persistent and inexpensive chemicals are used extensively in developing nations, creating serious acute health problems and local and global environmental impacts [9]. Further while remarkable progress has been made in the development of effective pesticides, the fact remains that a very small fraction of all applied pesticides is directly involved in the pesticidal mechanism. This implies that most of the applied pesticides find their way as 'residue' in the environment into the terrestrial and aquatic food chains where they undergo concentration and exert potential, long term, adverse health effects [21].

Contrary to popular opinion, chemicals have been used for thousands of years to control unwanted animals, plants, and microorganisms - the "pests." Well into the age of synthetic organic compounds, mankind was so preoccupied with the effectiveness of his so-called "pesticides" that there was little thought of what eventually would happen to the steadily increasing volume and number of these chemicals released into the environment. The term "pesticide" encompasses chemical agents used to control a wide variety of creatures inimical or just bothersome to man insects, mites, rodents, nematodes, weeds, fungi, and others. However, these "active ingradients" are formulated into thousands of mixtures- dusts, wettable powders, emulsifiable concentrates, etc-which are the products of commerce. For proper perspective, one must appreciate the volume of pesticides used now and in the past. The decades after World War II saw the large-scale production of cheap, longlasting synthetic

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petrochemicals, e.g. pesticides, dielectrics, plasticizers, and fire retardants. The realization that persistent chemicals could be both uncontrollable and unexpectedly toxic is usually attributed to Rachel Carson, who published *Silent Spring* in 1962 [6].

Food is the basic necessity of life and food contaminated with toxic pesticides is associated with severe effects on the human health. Food legumes are an important part of the human diet, as these are good and inexpensive sources of proteins, carbohydrates and dietary fibres. Stored grains are highly contaminated with pesticides as these are stockpiled and periodically treated with pesticides to control pest infestation. It is a usual practice to store grains for long term (3-36 months) at ambient temperatures in large warehouses where several doses of insecticides (chlorpyrifos being used as a termiticide) are applied to reduce the losses from storage pests. Grains treated with chemical pesticides show presence of bound residues even after fairly long periods of storage contri-buting to dietary intake of pesticides [14]. Pesticides are significant group of xenobiotics affecting the biota. Regulation 396/20 of the European Parliament and of the Council established values of the maximum residue levels (MRLs) of pesticides in products of plant and animal origin [19]. Humans are exposed to small amounts of pesticide residues after pesticides are metabolized by plants or decomposed by environmental agents, trace amounts of pesticide residing in the human body for long periods can cause chronic diseases and can lead to cancer [5].

Honey bees can bring many pollutants deposited on plants into the hive. Therefore, plant protection products used in agriculture can not only cause mass poisoning of bees but may also enter bee products, especially honey [1] affecting its quality, properties and posing a particular threat to human health [16]. Residual pesticides in water can be destroyed by, say, Fenton oxidation, electrochemical oxidation, TiO2 catalytic treatment, and UV photolysis [22]. Food is the basic necessity of life and food contaminated with toxic pesticides is associated with severe effects on the human health. Hence it is pertinent to explore strategies that address this situation of food safety especially for the developing countries where pesticide contamination is widespread due to indiscriminate usage and a major part of population lives below poverty line. It is therefore of significance to evaluate simple, cost effective strategies to enhance food safety from harmful pesticides for poor populace. Food processing at domestic and industrial level would offer a suitable means to tackle the current scenario of unsafe food. The processing of food commodities generally implies the transformation of the perishable raw commodity to value added product that has greater shelf life and is closer to being table ready [8]. Unit operations normally employed in processing food crops reduce or remove residues of insecticides and other pesticides that are present in them. These operations such as washing, peeling, blanching and cooking play a role in the reduction of residues (Elkins, 1989).

2. Fate of Pesticide in Environment

2.1. DDT (dichlorodiphenyltrichloroethane) and PCBS (polychlorinated biphenyls) and organochlorines in public health

Organochlorines are a diverse group of persistent synthetic compounds, some of which are detectable in nearly everyone. Many organochlorines are endocrine disruptors or carcinogens in experimental assays. p,p'-DDE (dichloro diphenyl dichloroethene) and PCBs (polychlorinated biphenyls) comprise the bulk of organochlorine residues in human tissues. Authors [15] reviewed relevant human data cited in the 1991–1995 Medline database and elsewhere. High-level exposure to selected organochlorines appears to cause abnormalities of liver function, skin (chloracne), and the nervous system. Of more general interest, however, is evidence suggesting insidious effects of background exposure. Of particular concern is the finding of neonatal hypotonia or hyporeflexia in relation to PCB exposure. The epidemiologic data reviewed, considered in isolation, provide no convincing evidence that organochlorines cause a large excess number of cancers. A recent risk assessment that considered animal data, however, gives a cancer risk estimate for background exposure to dioxin and dioxin-like compounds (e.g. some PCBs) with an upper bound in the range of 10−4 per year.

2.2. Pesticide residues in IPM and non-IPM samples of mango (*Mangifera indica*)

Shashi *et. al.* (2008) conducted experiment to analyze the residue of commonly used pesticides viz. methyl parathion, chloropyrifos, endosulfan, cypermethrin, fenvalerate, carbendazim, imidacloprid and carbaryl in mango, Dashehari variety, integrated pest management (IPM) and non-IPM samples were collected from the IPM and non-IPM orchards, Lucknow, India. Also presented a method for the simultaneous determination of these pesticides in mango samples. Residues of methyl parathion, chloropyriphos, endosulfan, cypermethrin, fenvalerate were extracted from the samples with acetone: cyclohexane: ethyl acetate in the ratio 2:1:1 followed by cleanup using neutral alumina. Analysis was performed by gas chromatography electron capture detector (GC-ECD) with a megabore column (OV-1). Residues of carbendazim, imidacloprid and carbaryl were extracted with acetone and after cleanup, analysis was performed by high performance liquid chromatography (HPLC) using photo diode array (PDA) detector. Recoveries of all the pesticides ranged between 72.7 – 110.6%, at 0.1 and 1.0 μg g−1 level of fortification. The residues detected in non-IPM samples of mango were found to be below the prescribed limits of maximum residue limit (MRL) while IPM samples were free from pesticide residues.

A small difference was observed in both the IPM and non-IPM samples of Dashehari mango. The reason behind this may be due to the different agricultural practices under which pesticides were applied according to the farmer knowledge in non-IPM field while in IPM field pests were controlled by practicing IPM knowledge. It is concluded that the pesticides in the samples were non detectable or within the permissible range of MRLs prescribed by FAO/WHO and may not present harm to public health. The standardized modified extraction method of GC-compatible pesticides for Dashehari mango may be useful for the routine analysis of mango fruits, which provide the Estimated method detection limit (EMDL) well below the MRLs of respective pesticides.

2.3. Persistence and Metabolism of Fipronil in Rice (*Oryza sativa* Linnaeus) Field

Rice is one of the most important food crops worldwide. However, it is also a valuable tool in assessing toxicity of organic and inorganic compounds. Fipronil insecticide has been widely used to control rice pests. The research was conducted to evaluate the fate of fipronil in rice field [18]. Persistence and metabolism of fipronil in rice is studied by applications of Regent 0.3G@45 and 180 g a.i. ha-1 was made 7 days after transplanting of paddy. Samples of paddy plants were collected at 7, 15, 30, 45, 60, 90 and 120 days after the application of insecticide. The samples of rice grains, bran, husk and straw were collected at the time of harvest. The samples were extracted and cleaned up by following a standardized methodology. Fipronil and its metabolites were quantified by gas liquid chromatography and confirmed by gas chromatography mass spectrometer. The total residues of fipronil and its metabolites in paddy plants after 7 days of its application at recommend and four times of recommend doses were found to be 6.60 and 19.85 mg kg-1, respectively. Among fipronil metabolites, sulfone

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derivative had maximum residue concentration followed by other metabolites viz. sulfide, amide and desulfinyl. The residues were reached below the detectable limit (0.01 mg kg-1) after 45 and 90 days at recommend and four time of recommend doses, respectively. At harvest, the samples of paddy straw, rice grains, bran and husk did not reveal the presence of fipronil and its metabolites.

2.4. Persistence and Dissipation of Quinalphos in/on Cauliflower and Soil

Persistence and dissipation of quinalphos residues in/on cauliflower was studied [20] after giving spray applications at two concentrations, i.e. recommended dose of 500 g a.i. ha-1 and double the recommended dose of 1,000 g a.i. ha-1. Residue analysis of cauliflower curds was carried out after the third spray over a period of 15 days. Initial residues of quinalphos on cauliflower from the two treatments were 1.19 and 1.842 mg kg-1. The residues persisted up to 15 days from both the treatments. The residues of quinalphos dissipated from both treatments with the half-life of 4.8 and 5.3 days. Based on the persistence study and maximum residue limit value of 0.05 mg kg-1 the safe pre-harvest interval was worked out as 17 and 22 days from treatment at the recommended and double the recommended dose, respectively. Analysis of soil samples was carried out on the 15th day of sampling and residues were found to be 0.013 and 0.044 mg kg-1.

2.5. Pesticide residue dissipation upon storage in chickpea legume

Over the years, contamination of food commodities with harmful pesticide residues has led to serious concern due to health hazards and environmental implications. In this background, effect of pesticide residue dissipation on chickpea legume under simulated storage conditions was investigated by Geetanjali and Poonam (2012). The dissipation pattern of chlorpyrifos and its metabolites under storage conditions for five months showed that the chlorpyrifos residues were four and five times (at the recommended and double recommended doses, respectively) above the MRL, highlighting the concern regarding safety of such stored grains for human consumption.

2.6. Pesticide residues levels in honey from apiaries

Authors [23] estimated the concentration levels of 30 pesticide residues in honey samples collected from apiaries in northern part of Poland (Pomerania) using method based on the QuEChERS extraction followed by liquid chromatography-tandem mass spectrometry with electron spray ionization (LC-ESI-MS/MS). In 29% of the samples were found positive for at least some of the target compounds. Concentration of bifenthrin, fenpyroximate, methidathion, spinosad, thiamethoxam, and triazophos exceeded maximum residue levels (MRL) in five samples (11%), the kind of the residues being correlated to agriculture practices in the region. The maximum values of these pesticides were 14.5, 16.3, 25.7, 20.6, 20.2 and 20.3 ng/g respectively. Profenofos was the most abundant at concentration ranged from <LOQ to 17.2 ng/g.

3. Detection of Pesticide Residue

3.1. Liquid Chromatography-Mass Spectrometry in Pesticide Residue Determination in Food

Authors [4] enlightened the use of liquid chromatography (LC) in pesticide residue determination was usually limited to groups of compounds or single compounds for which no

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suitable gas chromatographic (GC) conditions were available. However, recent developments have significantly enlarged the LC scope in this field of analysis. One of the most important advances was the on-line coupling of efficient LC separation with mass spectrometry detectors (LC-MS and LC-MS/MS) that makes this technique an excellent method for the determination of pesticides and their transformation products in complex matrices such as food.

The common feature of different approaches to pesticide chemistry is the appearance of compounds increasingly more polar. For this reason, LC techniques, especially RP (Reverse Phase), are useful in their separation without any pre-treatment. Moreover, the contribution of MS, with its high sensitivity and considerable diagnostic power, is fundamental. The relatively high number of publications on the analysis of pesticides in food samples by LC coupled to MS/MS shows that this technique has become a powerful tool in the quality control of food and food safety issues. Although reported results are controversial, ESI is the most frequently utilized ionization source because of its higher sensitivity for most important group of pesticides. Deeper studies into the different source designs are required to find a relation between sensitivity and ionization source. The benefits of LC-M for pesticide residue analysis in food are widely recognized. However, some critical aspects need to be taken into account in relation to quantification, especially the matrix effect is an unresolved question yet. Some approaches to overcome this effect have been commented on and discussed thoroughly in this paper. The tendency is to perform an appropriate quantification with matrix-matched standards. triple Quadrupole (QqQ) is the instrument most commonly used in this field because the lower level of detections (LODs) achievable permit compliance with the strict MRLs established by governing authorities. Both QqQ and quadrupole ion trap (QIT) are frequently used in this field. The use of high performance mass analyzers such as TOF or quadrupole time-of-flight (QqTOF) is also expected to increase in the next few years. This increased use will be for unknown metabolite identification, for non-target pesticide screening methods and for analyte confirmation in positive samples.

4. Pesticide Residue Management

4.1. Pesticide residue removal from vegetables by ozonation

A novel machine was developed [7] to remove pesticide residues from vegetables using ozone. This domestic-scale vegetable cleaner consists of a closed cleaning chamber, an ozone generator, a water re-circulation pump, and an oxidation–reduction potential (ORP) electrode. Two vegetables, Chinese white cabbage and green stem bok choy, and three pesticides, permethrin (trans-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate), chlorfluazuron (1-[3,5-dichloro-4-(3-chloro-5-trifluoromethyl-2-pyridyloxy)phenyl]-3-(2, 6-difluorobenzoyl) urea), and chlorothalonil (tetrachloroisophthalonitrile) were used in tests. Cleaning for 15 min with pump recirculation removed 51% of chlorfluazuron and 53% of chlorothalonil. When the ozone production rate was 250 mg/h, removal efficiencies were 60% for chlorfluazuron and 55% for chlorothalonil, increases of 2–9% over pump recirculation only. When the ozone production rate was 500 mg/h, removal efficiencies were 75% for chlorfluazuron and 77% for chlorothalonil; increases of 24% over pump re-circulationonly. After the ozone treatment, all the pesticide residuals met the Standards for Pesticide Residue Limits in Foods.

4.2. Food processing a tool to pesticide residue dissipation

Authors [11] during 2008 appraised on food safety is an area of growing worldwide concern on account of its direct bearing on human health. The presence of harmful pesticide residues in

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food has caused a great concern among the consumers. Hence, world over to tackle food safety issues, organic farming is being propagated. However, due to several reasons, diffusion and acceptance of this approach in developing countries has been very slow. Therefore, it is important in the transient phase that some pragmatic solution should be developed to tackle this situation of food safety. Food processing treatments such as washing, peeling, canning or cooking lead to a significant reduction of pesticide residues. The processes include: baking, bread making, dairy product manufacture, drying, thermal processing, fermentation, freezing, infusion, juicing, malting, milling, parboiling, peeling, peeling and cooking, storage, storage and milling, washing, washing and cooking, washing and drying, washing and peeling, washing peeling and juicing and wine making. Extensive literature review demonstrates that in most cases processing leads to large reductions in residue levels in the prepared food, particularly through washing, peeling and cooking operations.

4.3. Bioremediation

The process of bioremediation enhances the rate of the natural microbial degradation of contaminants by supplementing these microorganisms with nutrients, carbon sources or electron donors (Table 1). This can be done by using indigenous microorganisms or by adding an enriched culture of microorganisms that have specific characteristics that allow them to degrade the desired contaminant at a quicker rate. Ideally, bioremediation results in the complete mineralization of contaminants to H_2O and CO_2 without the build up of intermediates. Bioremediation processes can be broadly categorized into two groups: *ex situ* and *in situ*. *Ex situ* bioremediation technologies include bioreactors, biofilters, land farming and some composting methods. *In situ* bioremediation technologies include bioventing, biosparging, biostimulation, liquid delivery systems and some composting methods [10].

4.4. Phytoremediation

Currently, a significant amount of research is being conducted on the interaction between microorganisms and plants in the rhizosphere and the potential to use this for the remediation of pesticide-contaminated media [10] (Table 2).

5. Lessons in Environmental Health

Environmental health has evolved rapidly in recent decades, drawing largely on new analytic technologies, advanced data acquisition and modeling, mechanistic studies in toxicology, and the conceptual framework of risk assessment. The latter combines toxicologic and epidemiologic data with improved techniques for quantifying exposure, producing estimates of risks from environmental hazards or conditions to selected target populations. The public governments have become increasingly concerned with

environmental health and quality. The major lessons have been

(*a*) environmental-health scientists must participate in policy debates

Table 1: Bioremediation of Pesticides

Experimental Design	Contaminant(s)	Experiment Duration	Results/ Remediation Efficiency	Source
White Rot Fungus biodegradation	DDT	30 days	69% of DDT degraded. 3% mineralized to CO2	[17]

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White Rot Fungus biodegradation several species of fungi	DDT	30 days	Species Dependent. Approx. 50% degradation. Approx. 5 to 14% of DDT mineralized to CO2	[3]
White Rot Fungus biodegradation - Phanerochaete chrysosporium	Mirex, aldrin, heptachlor, lindane, dieldrin, chlordane.	21 days	Chlordane: 9% to 15% metabolized to CO2. Lindane: 23% metabolized to CO2. No other compounds were significantly degraded.	[13]

Table 2: Phytoremediation of Pesticides

Experimental Design	Contaminant(s)	Experiment Duration	Results/ Remediation Efficiency	Source
Examined the use of the herbicide tolerant species, Kochia, to enhance rhizosphere degradation	Atrazine, metolachlor and trifluralin	14 days	Enhanced microbial degradation was observed in rhizosphere. 45% reduciton of atrazine. 50% reduction of metolachlor and 70% reduction of trifluralin.	[2]

(*b*) environmental health problems are exceedingly complex and require interdisciplinary research and

(*c*) environmental health is a global issue

The globalization of commerce, the untested impact of international trade agreements, increased migration, and especially increased population, have profound impact on the quality as well as availability of air, water, land, and food. Global atmospheric transport of pollutants and the effect on atmosphere and climate are two examples of globalization of environmental health [12].

6. Conclusion

Pesticide residues in food are influenced by storage; handling and processing which is post-harvest of raw agricultural commodities but prior to consumption of prepared foodstuffs. Extensive literature review demonstrates that in most cases processing leads to large reductions in residue levels in the prepared food, particularly through washing, peeling and cooking operations. Washing with water and various chemical solutions for domestic and commercial use are necessary to decrease the intake of pesticide residues. Freezing as well as juicing and peeling are necessary to remove the pesticide residues in the skins. Cooking of food products helps to eliminate most of the pesticide residues. Residues of post-harvest insecticide treatment on stored grains generally decline only rather slowly [11]. However even in those processing into foods results in large losses. Removal of residues in food by processing is affected by type of food, insecticide type and nature and severity of processing procedure used. Hence a combination of processing techniques would suitably address the current situation in food safety.

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