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Review Article

Decontamination of Pesticide Residues for Sustainable Agriculture

George F. Antonious*

Division of Environmental Studies, Kentucky State University, USA

***Corresponding author**

George Antonious, Professor, Kentucky State University, College of Agriculture, Division of Environmental Studies and Sustainable Systems, 104 Chappell Building, Frankfort, KY 40601-2355, USA, Tel: 502-597-6005; Fax: 502-597-6763; Email: george.antonious@kysu.edu

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Abstract

Sustainable Agriculture satisfies human food and fiber needs, makes the most efficient use of on-farm resources and integrates natural biological cycles, enhances the quality of life for farmers and society as a whole, and sustains the economic viability of farm operations. A farm that is able to produce continuously, yet has negative effects on environmental quality elsewhere, does not support sustainable agriculture. Over-application of synthetic pesticides and fertilizers or animal manure, which can improve productivity of a farm but can pollute nearby rivers and coastal waters does not contribute to sustainable agriculture. The lack of pesticide field dissipation data and the increasing concern about agricultural chemicals and their impact on surface and ground water quality have made this an international issue. Pesticide adsorption to soil is related more to soil organic matter than to other soil chemical and physical properties, therefore, addition of recycled waste as soil amendments having high organic matter content is a management practice that should be exploited to trap pesticides *in situ* in agricultural fields that would have been transported into rivers and streams contaminating natural water resources. The use of recycled waste for land farming could decrease dependence on synthetic fertilizers and provide alternatives to farmers dealing with the escalating production costs associated with increasing costs of energy. Soil management practices that bind pesticides and excess nutrients to soil particles, reducing pesticides, nitrate, nitrate, and phosphate in runoff and seepage water are vital to sustainable crop production systems and environmental quality. Soil organic matter has an important effect on the bioavailability, persistence, biodegradability, leaching, and volatility of chemicals in soil that could intercept pesticide-contaminated runoff from agricultural fields and provide a potential solution to pesticide contamination of surface and seepage water from farm- lands. Mitigation and removal of excess pesticide residues before they runoff and enter bodies of water is the main focus of the research plan at Kentucky State University (KSU)/Environmental Science at the College of Agriculture. Studies at KSU research farm indicated that bio filters installed down the field slope reduced runoff water volume by 44% and herbicides (trifluralin and clomazone) residues in runoff water by 55% and 38%, respectively. These findings revealed that bio filters offer a viable means of treating pesticides in runoff water arising from agricultural fields.

INTRODUCTION

The utilization of organic waste in crop production is an integral part of sustainable agriculture. Optimized soil management represents an important strategy for sustainable agricultural systems. Since 1991, Kentucky State University (KSU) Environmental Toxicology program has been engaged in many field and laboratory studies to investigate the relationships between soil farming practices, soil erosion processes, runoff water, fate of pesticides, and environmental quality. The Karst topography in Kentucky is defined as a geological region underlain by soluble bedrock (limestone) characterized by ground depressions, sinkholes, caves, and underground drainage. Karst aquifers are highly susceptible to contamination from the soil surface. Various agricultural and management practices have been used at the KSU/Environmental Toxicology Research to mitigate or eliminate environmental pollution by pesticide residues. Planting living fescue strips against the contour of the land slope reduced pesticide residues in runoff water. However, this practice has the disadvantage of increasing the potential of soil infiltration by pesticides [1,2]. Unfortunately, plastic mulch, which can cover between 50-70% of a field surface, increased surface water runoff from rainfall and irrigation [3]. This means much of the pesticides applied in living fescue or in plastic-mulched fields may seep into groundwater or leave the agricultural fields as surface runoff. In agriculture, composting provides an organic amendment useful for improving soil structure and nutrient status [3-6] and it generally stimulates soil microbial activity [7,8]. In addition to the beneficial components of soil amendments, such as sewage sludge organic matter and nutrients, sewage sludge also contains heavy metals (Cd, Cr, Ni, Pb, Zn, Cu, and Mo) that may transfer into surface water [9] or accumulate in edible portions of plants at harvest [10,11] and present human and aquatic life-toxicity risks. Excessive amounts of these heavy metals in soil can be toxic to plants, may adversely affect organisms that feed on these plants, may result in bioaccumulation in animals and humans, and may enter water systems through surface runoff and leaching.

Sustainable agriculture promotes methods that prevent environmental contamination, and requires that agricultural practices be achieved in ways that do not cause harm to the environment or workers. Examples of harmful agricultural practices include over-application of inorganic pesticides and fertilizers that pose a risk to consumers, farm workers, and surrounding communities. Environmentally and economically viable agriculture requires the use of cultivation practices and innovative technologies that maximize agrochemical efficacy while minimizing off-site movement. In Kentucky, pesticides are applied according to guidelines set by the Environmental Protection Agency (EPA) and the Kentucky growers guidelines [12]. However, pesticides application periods usually coincide with seasonal rainfall events. As a result, soil erosion and pesticide wash-off from pesticides-treated soil and plant foliage increase the likelihood that pesticides enter the Kentucky River watershed and impact surface water quality. Runoff from agricultural watersheds carries enormous amounts of pesticides.

There is an urgent need to develop long-term, low-energy, biological, self-sustainable systems of farming. Application methods of these systems must be simple, inexpensive, safe, energy conserving, and be able to reduce soil erosion. Soil erosion reduces productivity by decreasing soil depth, removing nutrients required for plant growth, and altering soil physical properties resulting in less water infiltration, poor crop establishment and root penetration. Yield decline due to erosion is usually masked by advances in technology such as use of fertilizers, use of high-yield crop varieties, and improved planting technology. Intensive use of pesticides in many parts of the U.S. increases the potential for serious non-point source contamination of soil and receiving water. The occurrence of pesticides in runoff and infiltration water [4,13-15] and on treated vegetables [16] increased the potential impact on human health and environmental quality. According to the USEPA, over 441 million kg of conventional pesticides were used in the U.S. in 2001 [17]. Of that total, 77% were used in agricultural applications, and 11% were used for home and garden purposes. Nationwide, approximately 1,200 water body impairments have been attributed to pesticides [18]. Agricultural productions are frequently conducted in close proximity to lakes, reservoirs, and streams. Herbicides are usually applied to bare soil before crops are planted, incorporated to 1 to 2 inch depth for the control of grasses and broad leaf weeds in agricultural crops, residential grass lawns, and golf courses. Agricultural production is an important part of the nation's economy and pesticide use on crops is extensive [19,20].

There is a concern over the risks of contamination of food and drinking water by residues of synthetic agrochemicals, and the negative impact of agrochemicals on the countryside. A central hope in these concerns is the safe use of agrochemicals, development of new soil management practices, and use of mitigation techniques [21, 22]. Additional technological and infrastructural solutions are needed to reduce pesticides release and protect the environment and human health. For agricultural purposes, these technologies and systems must be cheap, reliable, and easy to apply with low labor and time input. In addition, the escalating production costs associated with the increasing costs of energy and fertilizers to U.S. farmers and the problems of soil deterioration and erosion associated with intensive farming systems have generated considerable interest in less expensive and more environmentally compatible production alternatives such as recycling wastes from several processing operations for use as soil conditioners which could provide high-quality organic amendments for soil improvement and crop production. In the U.S., approximately 6.2 million dry metric tons of municipal sewage sludge is produced annually by wastewater treatment facilities [23] and the tonnage is expected to increase due to population growth, increasing urbanization and industrialization. In addition, the rapid growth of the poultry industry has resulted in significant manure generation. More than 11.4 million tons of poultry litter was generated in the U.S. [24].

Significance of the Problem

Approximately 41,511 water body impairments across the U.S. have been attributed to pesticides and of that total 1,300 water body impairments occurred in the state of Kentucky [25]. According to the United Nations World Water Development Report, some two million tons of waste per day are disposed to near receiving waters, including industrial wastes and chemicals, human waste, and agricultural wastes such as fertilizers and pesticides [26]. A significant association was found between the season of elevated agrichemicals in surface water in April-July and higher risks of birth defects in live births in the U.S. [27]. Concentrations of atrazine, nitrates and other pesticides also were higher in the months of April-July. Chlorophenoxy herbicides are widely used in the U.S. for broadleaf weed control in grain farming and parking maintenance. Most of the spring and durum wheat produced in the U.S. is grown in Minnesota, Montana, North Dakota, and South Dakota, with more than 85% of the acreage treated with chlorophenoxy herbicides such as 2,4-dichlorophenoxyacetic acid (2,4-D) and 4-chloro-2-methylphenoxyacetic acid (MCPA). Schreinemachers [28] found that infants in these four wheat-producing states conceived in April-June, the time of herbicide application, were more likely to have circulatory/respiratory (excluding heart) malformations compared to births conceived during other months. Mitigation of pesticide residues in soil and natural water resources could be achieved using modern management practices such as bio-filters [21], adsorption techniques [22,29], biochar [30], and recycling waste for use as soil amendments.

Ksu/College of Agriculture Current Research in Pesticides Mitigation

Kentucky State University's College of Agriculture is currently engaged in innovative research in various aspects of pesticide mitigation techniques. The division of environmental studies has provided the leadership in pesticide mitigation techniques by implementing a new technology using bio-filters (bio-beds) to reduce the impact of field application of pesticide residues on environmental quality. Kentucky farmers have been provided with different models of soil management practices that could be used for growing vegetables on erodible lands and for increasing farm productivity and profitability, while reducing pesticides mobility and off-site movement. KSU is currently using sewage sludge "bio-solids" and chicken manure as alternative fertilizers and some farmers in Kentucky are currently using sewage sludge for growing vegetables for commercial production. This alternative to inorganic fertilizers resulted in a considerable savings for limited-resource farmers who face the escalating production costs associated with the rising costs of energy and fertilizers. The Metropolitan Sewer District in Louisville, Kentucky (Figure 1) has turned human and pre-treated industrial waste into dried pellets that can be used as fertilizer. Under rules set by the US EPA, it can be spread on crops used for human consumption as well as those used for animal feed (Lexington Herald-Leader, 2005) [31]. The waste pellets are available at several hardware, lawn and garden stores. In addition to Kentucky and Indiana region, the new product known as "Louisville Green" (Figure 2) has been licensed for use on farms in South Carolina, Georgia, Tennessee, Missouri, Arkansas, and Mississippi. In addition, the University of Kentucky Poultry Research Facility in cold stream farm, Lexington, KY discards significant amounts of broiler and chicken manure waste that could be used as organic fertilizer. At KSU Research and Demonstration Farm (Franklin County, KY), researchers are currently using these two organic amendments to decrease dependence on synthetic fertilizers.

The following techniques are currently utilized at KSU/ College of Agriculture (Frankfort, KY) to mitigate and reduce environmental pollution by synthetic pesticides:

1- Enhance SOM Content: Humic substances in soil organic matter (SOM) have an aromatic framework that contains hydrophobic (fate-soluble) and hydrophilic (water-soluble) sites. Several hydrophobic sites on SOM could be explored. These

Figure 1 Metropolitan Municipal Wastewater Treatment Plant, Louisville, KY.

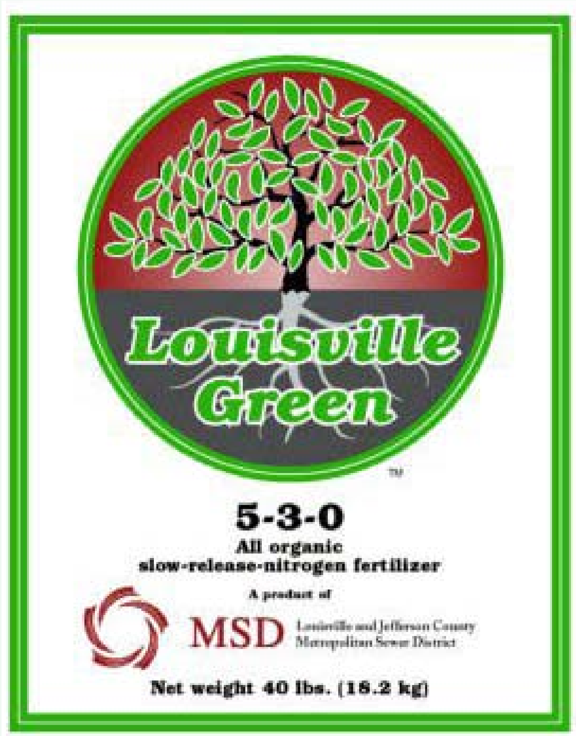

Figure 2 Municipal sewage sludge "Louisville Green" NPK (5:3:0) used for growing vegetables in commercial growers farms (Meade County, KY).

include fats, waxes resins, and aliphatic side chains. Bonding of non-polar pesticides to SOM is likely a pesticide–lipid interaction. Because lipids are associated with soil humus (a sticky brown insoluble organic matter), pesticide adsorption by soils depends on the SOM content [29]. Accordingly, the increased organic matter in soil due to compost addition (compost-amended soil) plays an important role in the adsorption of non-polar pesticides such as bensulide (an herbicide) and pyrethrins (a group of insecticides). This management practice could be used for bonding (trapping) non-polar pesticides to reduce surface water and groundwater contamination by pesticides. Studies indicated that concentration of pyrethrins and piperonylbutoxide (PBO), a pyrethrum synergist, adsorbed by compost-amended soil was significantly higher than that adsorbed by no-mulch bare soil [29]. Based on soil analysis, the SOM content of compost-amended soil (5.7%) was nearly twice that of the no-mulch soil (2.7%). These results are in accordance with the work of Singh and Singh [32] that reported higher adsorption of endosulfan (a non-polar insecticide) in natural soils compared to oxidized soils and are in accordance with Krohn [33] who reported that pesticide sorption on compost can be characterized by the adsorption coefficient (Kd) values, which tends to increase with increasing SOM content.

2-Enhance the Activity of Soil Microbes: Soil microbes (bacteria, fungi, protozoans, and algae) excrete a variety of enzymes (ureases, invertases, dehydrogenases, cellulases, amylases, phosphatases) that are recognized as a primary means of degrading pesticides in soil and water ecosystems. Soil microorganisms also produce sticky substances (polysaccharides) that help soil particles adhere to one another and help the soil to resist erosion. Specific emphasis has been given to soil enzymes in relation to reclamation and the enzymatic processes that play a significant role in bioremediation for sustainable agriculture. Remediation of pesticide-contaminated soils is based on the degrading activity of soil microbiota. The addition of organic amendments such as yard waste compost [34], straw [35], tree leaf mulch [36], and chipped wood from twigs [37] reduced the negative effects on soil microbial populations and soil enzyme activities due to the increased SOM and its role in sorption processes as a result of the presence of humic substances containing carbonyl, carboxyl, phenolic, and alcohol functional groups [38].

3-Implement Biobed Systems: A biobed (biofilter) is a cavity that filled with a mixture of composted organic matter that provides a potential solution to pesticide contamination arising from agricultural chemicals. Biobeds were built at the end of runoff field plots at the KSU Research Farm using locally available materials (chopped wheat straw, peat moss, and top soil). Peat and straw provide numerous sites for pesticide sorption. They help maintain aerobic conditions when combined with sufficient humidity or moisture owing to its high water-holding capacity. Straw acts as an additional food source for lignin-degrading soil microorganisms that produce enzymes catalyzing the degradation of a broad spectrum of chemicals [39]. The topsoil acts as an inocoulum of soil microbes for the system and is likely to vary in terms of its physical, chemical, and biological characteristics from one farm to another. Repeated use of certain pesticides over a number of seasons can result in enhanced degradation of pesticides due to adaptation and proliferation of specific soil microbial communities that utilize the pesticides of toxic compound as an energy source, and thus, degrade it more easily.

4-Use Constructed Wetland Microcosms: Constructed wetland (CW) microcosms systems were built at KSU (Figure 3) to treat wastewater containing pesticide residues by filtration, settling, and bacterial decomposition in a natural-looking lined marsh. CW systems have been used nationally and internationally with good results, but performance levels decrease in cold climates during winter months. CWs may be described as soil/plant systems for wastewater treatment in which pollutant removal is based on the general principles of nutrient transformation. CWs could be used as a low-cost alternative method for controlling water pollution from point and nonpoint sources. Plants have been used for wastewater treatment, and their ability to remove xenobiotics like pesticides has been attributed to the microorganisms associated with their roots. The literature review showed that CWs have been proposed for retaining agricultural pollutants as a potential best management practice to mitigate the effects of pesticide-associated agricultural runoff [40]. More than 60 reports on CWs show that CWs have been widely used to control both point- and nonpoint-source pollution by pesticides in surface waters. CW microcosm systems were effective in decreasing concentrations of chlorpyrifos and chlorothalonil pesticides in stimulated storm water runoff [41]. Results from several studies indicated that CW microcosms could be part of an efficient mitigation plan for treatment of pesticide mixtures in contaminated wastewater. Use of aquatic plants has a great potential to function as *in-situ* on-site biosinks and bio-filters of aquatic pollutants. These plants are useful in phytoremediation because of their ability to degrade environmental contaminants via their exudates (enzymes) released to the contaminated sites [42].

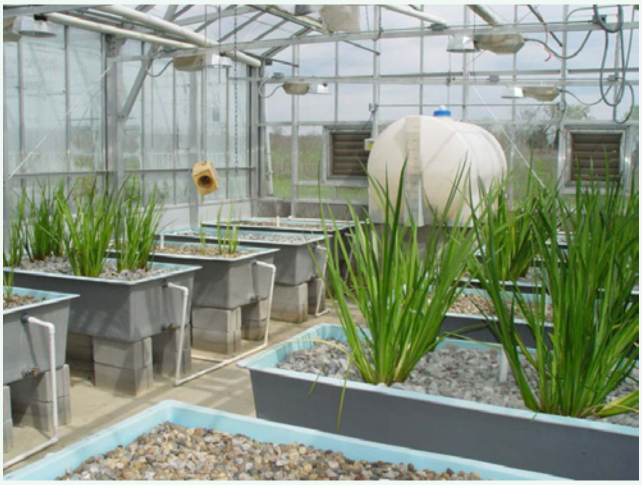

Figure 3 Greenhouse wetland microcosms (n=12) of 182 × 72 × 43 cm each (capacity of 157 gallons) containing two types of bedrocks, crushed limestone and river gravel, and planted with iris plants, *Iris versicolor* as the plant test.
Note that a 500 gallon supply tank installed inside the greenhouse is used to distribute water equally to each of the 12 microcosms used to breakdown pesticides.

In addition to the outlined environmental remediation techniques described above, sustainable agriculture promotes the use of natural products for pest control as alternatives to synthetic pesticides. The use of natural products for reducing the impact of synthetic pesticides is one of the main objectives at KSU, College of Agriculture. Farmers from many developing countries have used plant extracts to protect food and fiber from insect damage. Chili pepper powder deterred ovi position of the onion fly, *Delia antique* [43]. Similarly, capsaicin in hot pepper has been reported to reduce larval growth of the spiny bollworm, *Eariasinsulana* [44] and the use of oleoresin from capsicum as a repellent against cotton pests has been reported [45]. Water extracts of some hot pepper accessions (genotypes) were highly toxic to the cabbage looper larvae *Trichoplusia ni* (Hubner), the most difficult pest of crucifer crops to control during the past decade. At KSU/ College of Agriculture, Antonious *et al.,* [46] investigated the composition of hot pepper fruit extracts that might explain the observed differences in toxicity and repellency to spider mite, *Tetranychusurticae* (Koch), among fifty-two accessions of hot pepper fruits of plants grown from seeds obtained from eight countries of origin (Figure 4) through the USDA National Germplasm Collection. Three decanoic acid methyl esters (pentadecanoic acid methyl ester, hexadecanoic acid methyl esters, and octadecanoic acid methyl ester) predominated pepper fruit extracts of most toxic accessions to the cabbage looper [47]. Methyl esters are aliphatic long-chain saturated fatty acids that are common components of plant lipids. Extracts from pepper fruits might provide an opportunity for use in crop protection as alternative to synthetic pesticides.

Published work at KSU on natural products also indicated that wild tomato leaf extracts of *Lycopersiconhirsutum* plants (Figure 5) could be explored as an alternative to synthetic pesticides [48-50]. Three methyl ketones (2-tridecanone, 2-dodecanone, and 2-undecanone) were effective against the tobacco hornworm, *Manducasexta* L. 2-Tridecanone was the most effective methyl ketone against tobacco hornworm and tobacco budworm, *Heliothisvirescens* [48]. 2-Tridecanonealso was effective against adults of the green peach aphid, *Myzuspersicae*, and required a significantly lower dose than 2-undecanone. 2-Tridecanone, which has an herbaceous spicy-like odor, was found toxic to a number of insect species by contact ingestion, or vapor action [51]. The short persistence of 2-tridecanone on the leaves of the greenhouse vegetables tested [52] could be recognized as a desirable chemical characteristic. Ideally, safe pesticides remain in the target area long enough to control the specific pest and then degrade into harmless compounds. Such alternatives, which have few or no side effects on the environment, low toxicity to warm-blooded animals including humans, high efficacy against insects and spider mites [49] and low potential for insect resistance development, are in great demand. Products from hot pepper fruits and wild tomato leaves could be explored and used to develop natural products for use as biodegradable alternatives to synthetic pesticides.

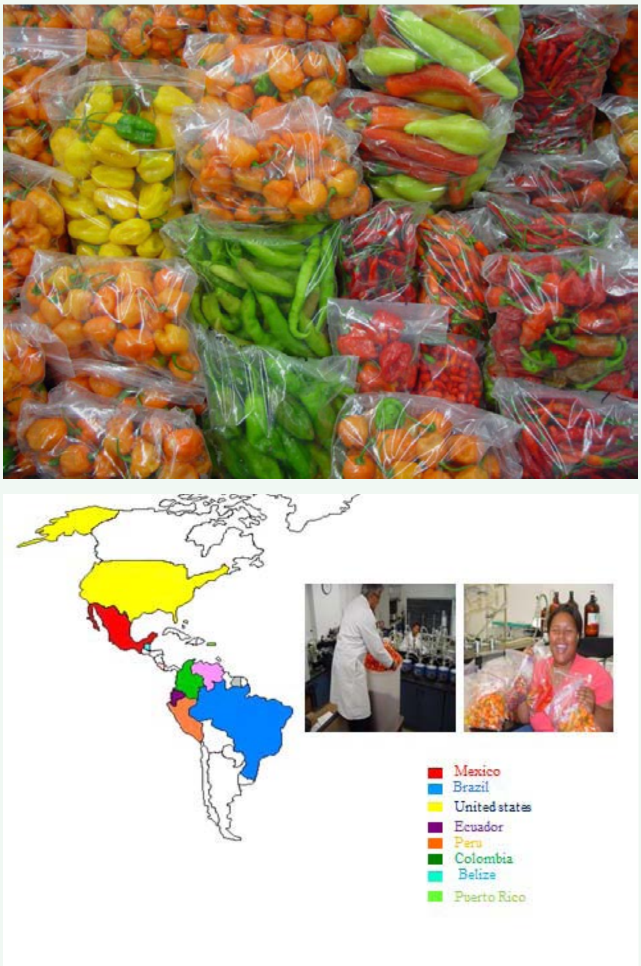

Figure 4 Hot pepper fruits of plants grown from world-wide seeds obtained from the USDA/ARS (Griffin, GA) and planted at Kentucky State University Research Farm (Franklin County, KY).

Figure 5 Wild tomato plants, *Lycopersicon hirsutum f. glebratum* grown at Kentucky State University green houses for developing new insecticides of natural origin.

CONCLUSION

Environmental pollution by pesticides is a matter of growing concern in the U.S. and worldwide. Mitigation and removal of excess pesticide residues in soil before they run off and enter bodies of water is the main focus of the research plan at KSU College of Agriculture. Research has shown that soil amendments (i.e., sewage sludge and chicken manure) and the installation of biobed systems at the end of the field slope at the KSU Research Farm, successfully reduced runoff water, sediment loss, and pesticide mobility at the edge of the field and provided an acceptable yield. Biobeds are a new technology in the U.S. that could be implemented in land farming to reduce pesticides mobility into runoff and seepage water arising from agricultural fields. KSU has provided Kentucky farmers with leadership in different farming alternatives including cropping systems, planting living fescue strips, use of sewage sludge as soil management practices to reduce dependence on synthetic fertilizers. These practices would increase farm profitability and contribute to local economic development. Use of soil amendments help enrich soil, increase crop yield, and provide additional environmental benefits in reducing stress on landfills, where bio-solids would usually disposed. Future research objectives will include the development of novel formulations of capsaicin and methyl ketones derived from the plant accessions characterized in these studies.

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