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Research Article

Evaluation of the environmental exposure risks of pesticides used in vegetable production in Türkiye

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ABSTRACT

In this study, first, a list of pesticides that can potentially pose environmental exposure risks was compiled by analyzing the recent literature on residue levels in fresh vegetables produced in Türkiye. Then, by using the fundamental environmental partitioning properties of these pesticides, their potential multi-media environmental distributions were assessed. Acetamiprid, chlorpyrifos, and pyridaben were among the pesticides that frequently exceeded the residual limit values. Multi-media environmental modeling was conducted for these three pesticides using an evaluative four-compartment (air, soil, water, sediment) model. Compartmental distributions, inter-compartmental mass transfer rates, advective, and reactive losses were estimated for the selected pesticides after their simulated application to soil. The ranking of overall persistence among the pesticides was found to be pyridaben > chlorpyrifos > acetamiprid. The percentage mass distribution of acetamiprid in water was higher due to its low volatility and high solubility. The overall persistence of chlorpyrifos was limited by its higher partitioning to air although it is more persistent than pyridaben in other compartments. To investigate the residue dynamics of the three pesticides in tomato crops, temporal changes in harvest fractions were compared using the regression equations of the crop model dynamiCROP. Acetamiprid was estimated to be taken up at higher rates in tomatoes after initial application. The residue dynamics of chlorpyrifos and pyridaben were found to be similar. The quantitative methods in this study can be used to assess the environmental risks associated with commonly used pesticides in Türkiye and to address the issue of exceeding residue limits in agricultural products.

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INTRODUCTION

Türkiye is one of the leading countries in the global vegetable production. According to the data from the Food and Agriculture Organization of the United Nations (FAO), in 2021, Türkiye ranked fourth in total fresh vegetable production, following China, India, and the United States [1]. Regarding tomatoes, which are among the most widely produced vegetables globally, Türkiye holds the position of the third-largest producer, following China and India. Examining the FAO's data on total vegetable exports for the year 2021, Türkiye ranks eighth in terms of export quantity (2.34 million tons) and twelfth in terms of export value ($1.9 billion) [2]. The vegetable product that Türkiye exported the most in 2021 was tomatoes, with an export quantity of approximately 606,583 tons and a value of around $357 million. By enhancing agricultural production efficiency and product quality, Türkiye has the potential to obtain a larger share in global agricultural trade.

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Globally, agricultural areas experience approximately a 35% loss due to diseases, pests, and weeds [3]. Plant protection processes need to be an essential part of agricultural production. The fundamental principle is to protect agricultural crops from diseases, agricultural pests, and weed infestations without causing harm to the natural environment. Pesticides are used in agriculture to prevent agricultural losses, achieve high yields, and meet the increasing demand. According to the study conducted by Doğan and Karpuzcu [4], pesticide use in Türkiye is approximately 1.66 kg/ha, which is not significantly higher compared to countries with similar magnitude of land areas devoted to agricultural production. However, pesticide use in Türkiye is concentrated in the Mediterranean Region, where agricultural activities are intensively conducted [5].

Pesticide use in agriculture has the advantage of increasing productivity and production quantity. However, the environmental pollution caused by pesticides and the presence of pesticide residues in agricultural products have negative implications in terms of health and economics. The European Union utilizes the Rapid Alert System for Food and Feed (RASFF) portal to monitor all food and animal feed products imported from non-EU countries, including animal feed, animal-derived products, milk and dairy products, honey, plant-based products, and medicinal plants. Türkiye is the country with the highest number of notifications compared to other countries exporting to the EU [6]. According to the 2020 report published by the RASFF portal, there were 405 notifications indicating that pesticide residue values in products imported from Türkiye exceeded the maximum residue level (MRL) [7]. Notifications regarding pesticide residues can be queried in real-time through the RASFF Window website [8].

Numerous studies have been conducted in Türkiye investigating the residue levels of pesticides in agricultural products. A review study conducted by Tiryaki [9] compiled the results of pesticide residue analyses in fresh and/or dried fruits and vegetables, as well as processed agricultural products in Türkiye. In the study by Tözün and Akar [10], 35 studies conducted after 2010, analyzing pesticide residues in food items were examined. It was reported that the maximum residue level (MRL) was exceeded in approximately half of the food samples.

The adverse effects of all harmful chemicals, including pesticides, on humans can vary depending on the level and duration of exposure, and also, on the toxic effects of the chemical. Human exposure to pesticides can occur during their production, storage, transportation, and use, and via consumption of products containing chemical residues. Pesticides can enter the human body through inhalation, skin contact, and gastrointestinal pathways [11]. Pesticide exposure in humans can result in acute effects such as respiratory problems, headaches, skin issues, and nausea, while long-term effects such as neurotoxicity, endocrine disruption, and cancer have also been observed [12].

After application, pesticides can transport to environmental media beyond the intended agricultural target. The fate and transport of the pesticide, once released into the environment, depend on the chemical and environmental characteristics in addition to the application rate. Pesticides can contribute to atmospheric pollution through volatilization. They can also reach surface waters through surface runoff and, in some cases, contaminate groundwater through leaching. Pesticides can undergo biological, chemical, and/or photolytic degradation in/on plant tissues and other environmental compartments. They may become sorbed to the soil particles depending on the properties of the pesticide and the soil. Multi-media environmental pollution caused by pesticides can have adverse effects on non-target vertebrates, invertebrates, and plants [13, 14].

Multi-media environmental fate and transport models are used to predict the distribution and movement of chemicals in environmental systems with multiple environmental phases and compartments. An overview of the history of multi-media environmental models is provided by Rong-Rong et al. [15]. In the late 1950s, studies on pollutant fate and exposure assessment focused on evaluating the behavior of a single pollutant in a single environmental media. In the 1970s, multi-media mass balance models were developed for regionally and globally distributed metals such as lead, mercury, arsenic, and cadmium. In the late 1970s, studies were conducted on multi-media mass balance models for organic pollutants [15]. Multi-media fate models can quantitatively analyze the intermedia transfers, accumulation, and persistence of pollutants in the environment. The model outcomes can be used to investigate the potential pollution scenarios caused by the chemical, and to analyze the environmental mechanisms underlying the existing pollution conditions.

Pollutant chemicals can be transported to and accumulate in various plant parts, such as roots, stems, leaves, tubers, fruits, and flowers [16]. Several models have been developed to explain the plant-environment relationships regarding chemical pollution. In the 1990s, models were developed to represent the uptake processes of organic chemicals by plants in the environment. Some models focused on root uptake, while others addressed uptake through leaves, and some models considered both pathways [16]. In recent years, models have been developed to describe the interaction of specific crops with environmental pollution [17]. Steady-state calculations are commonly preferred in models due to their simplicity and relatively low data requirements. However, most environmental conditions are characterized using dynamic processes. Additionally, the emission inputs into the real environment are almost never constant [18]. Recently developed plant uptake models take into account dynamic processes [16, 19, 20].

In this study, multi-media environmental models were used to investigate the environmental fate and transport of pesticides that are widely used in fresh vegetable production in Türkiye. First, pesticides frequently exceeding the maximum residue limits in fresh vegetables produced in Türkiye were determined through a literature survey. Then, the physicochemical properties of these pesticides were identified. The subsequent model-based investigation focuses on examining the environmental risks of commonly used pesticides applied at high doses in Türkiye using multi-media fate models and studying the dynamics of pesticide levels in tomato crops using a crop-specific multi-compartment plant uptake model. The proposed quantitative methods in this study have the potential to be used further in evaluating environmental risks associated with widely used pesticides in Türkiye and addressing the issue of exceeding residue limit values in agricultural products.

MATERIALS AND METHODS

Determination of Pesticides Excessively Used in Fresh Vegetable Production

In this study, scientific publications containing the analysis results of pesticide residues in food consumption products in Türkiye after 2010 were examined. During the literature search, relevant studies were accessed using the terms "pesticide," "residue," or "pesticide residue," and "Türkiye" in databases such as Web of Science, TR-Dizin, ScienceDirect, Google Scholar, and Scopus. Studies conducted before 2010 and residue analyses in animal-derived food products were not included in the survey. Recent studies focusing on cases where maximum residue level (MRL) values were exceeded in agricultural foods were identified. The literature research revealed that pesticide residue analysis studies frequently focused on fresh vegetables, and especially on tomatoes, peppers, and cucumbers. Considering the high daily consumption quantities of these three food products, the pesticides exceeding the MRL values in them were determined and listed. The physicochemical properties influencing the environmental fate of the listed pesticides, such as molecular weight, water solubility, vapor pressure, and half-life in environmental compartments, were compiled from the literature.

Multi-Media Fate Modeling

Multi-media fate models are tools used to estimate the chemical pollution levels in different environmental compartments, quantitatively analyze inter-compartmental transport processes, and describe the accumulation and persistence properties of chemicals. Multi-media models conceptualize different environmental media as interacting compartments with defined volumes and surface areas. The assumption of complete mixing is made for each compartment. Chemical equilibrium processes and advective transport, diffusive transport, and reactive processes are mathematically defined. Mass balance equations are formulated for each compartment and the phases constituting them [21]. The model outputs provide quantitative information on the tendency of chemical pollutants to accumulate and persist in the considered environmental media. The principles of multi-media fate and transport models are detailed by Parnis and Mackay [22]. Depending on the problem to be addressed, multi-media environmental models can be constructed at different scales and at different levels of detail. In this study, calculations were performed using three different multi-media fate models with varying detail and scope to examine the environmental exposure risks of pesticides used in fresh vegetable production.

Chemical Space Diagram

Chemical space diagram is a conceptual diagram drawn using the output of the simplest possible mass balance equation based on the equilibrium assumption between different phases within a closed system [23]. The diagram illustrates the tendency of chemicals to be present in air, water, or organic matter phases based on their fundamental partitioning characteristics. In the model that generates the diagram, the organic matter phase is represented by octanol, serving as an indicator of the tendency of organic chemicals to associate with solid phases such as soil and sediment. In the calculations, volumes ratios of 656,000:1,300:1 were used to represent the typical volumes of the air, water, and octanol phases, respectively, as suggested in the original study by Gouin et al. [23].

Equilibrium Criterion (EQC) Model

The Equilibrium Criterion (EQC) Model is a multi-media fate model developed by the Canadian Environmental Modeling Centre (CEMC) to be used as a tool for assessing the environmental exposure risks of chemical pollutants [24–27]. The model is developed using fugacity-based mass balance equations to represent the equilibrium and loss processes, advective inputs and inter-compartmental transport of a chemical pollutant. The model allows for calculations of different levels of complexity, but all calculations involve the assumption of steady-state conditions. The modeled environmental system includes compartments of air, water, soil, and sediment. The environmental properties of these compartments, such as volume, interfacial area, density, and organic carbon content, were standardized by the model developers to represent a typical terrestrial region. The model program does not allow users to modify the environmental properties, thereby enabling calculations focused on assessing the characteristics of the chemical pollutant using a "unit world" approach [28]. As input to the model, the physicochemical properties of the pollutant, such as melting point, water solubility, vapor pressure, octanol-water partition ratio (K_{OW}), and half-life in environmental compartments are required. Additionally, emission information needs to be specified.

Plant Uptake Model

In this study, a dynamic multi-compartment plant uptake model, dynamiCROP, was used for the analysis of pesticide residue levels in tomatoes. The DynamicCROP model was developed to evaluate pesticide applications in food crops and exposure effects to pesticide residues, in a life-cycle assessment context [16, 19]. Chemical properties, crop characteristics, and environmental properties constitute the inputs to the model. The model considers inter-compartmental transport and degradation processes. The most fundamental result provided by the model is the harvest fraction (hF), which represents the residue amount in the harvested crop relative to initially applied pesticide mass [16]. Crop-specific regression models were also developed for dynamiCROP with the aim of reducing the input requirements and facilitating practical usage [29, 30]. The regression models calculate hF values for three different compartments: soil, crop, and crop surface. During the construction of the regression models, parameters that significantly affect the model results were identified. It was observed that the inter-phase partitioning ratios of chemicals, lipid contents of crops, and soil parameters significantly influence the dynamiCROP model output. Therefore, these parameters were assigned as independent variables in the regression equations. In the regression models, the harvest fraction is calculated according to Equations (1) and (2) [29].

$$hF_i = \alpha_{0i} \times \exp\left(\beta_i \left(\Delta t \times k_i\right)\right) \quad (1)$$

$$hF = hF_{soil} + hF_{crop} + hF_{crop-surface} \quad (2)$$

In Equation (1), hF_i represents the harvest fraction calculated for compartment i (i = {*soil, crop, crop-surface*}). Therefore, the parameters α_{0i}, β_i, k_i are computed for each compartment. Regression equations that enable the calculation of these parameters for different plant species are provided by Fantke et al. [29]. Δt is the time (days) passed since the pesticide application. In this study, regression equations developed for tomatoes were used. The parameter α_{0i} represents the initial pesticide amount in the compartment and, for tomatoes, it is dependent on the molecular weight of the pesticide chemical. The k_i value (elimination coefficient) is a measure of the elimination rate from the respective compartment. For the soil compartment, the elimination coefficient is dependent on the persistence value in the soil compartment; and for the crop compartment, it can be calculated using the half-life of the chemical in tomatoes. For the crop-surface compartment, the log K_{OW} value and molecular weight of the chemical are used to calculate the k_i value. β_i used in Equation (1) is a coefficient that takes a constant value for each crop. Once the hF values are calculated for each compartment, the total harvest fraction (hF) is obtained by adding them (Equation (2)).

RESULTS AND DISCUSSION

Pesticides Exceeding the Residual Limit Values in Fresh Vegetable Crops

In this study, 13 different pesticide residue analysis studies conducted after 2010 were examined. In the published studies, a list of pesticides exceeding the Maximum Residue Level (MRL) was compiled for tomatoes, peppers, and cucumbers produced in Türkiye (Table 1). It was observed that the MRL was exceeded for a total of 34 different pesticide residues. Approximately 44.1% of the chemicals exceeding MRL are included in the banned active substances list prepared by the Ministry of Agriculture and Forestry of the Republic of Türkiye, dated March 3, 2022 [31]. It is expected that the use of these pesticides has been discontinued. Comparing the results of future residue analysis studies with the contents of Table 1 can provide information about the effectiveness of pesticide bans in practice. The most frequently exceeded MRL values were observed for acetamiprid, chlorpyrifos, and pyridaben (Table 1).

The pesticide types and substance groups that frequently exceeded the MRL in fresh vegetables produced in Türkiye were identified. Additionally, MRL values and pre-harvest intervals for pesticides were compiled from the Ministry of Agriculture and Forestry's Plant Protection Products Database [32] (Table 2). The pre-harvest interval refers to the time between the last pesticide application and harvest. It is determined by the Ministry of Agriculture and Forestry for each pesticide type and plant species [32].

The properties of chemical constituents in pesticides play a crucial role in the environmental fate and plant uptake of these pesticides. The physicochemical properties of pesticides that exceeded MRL at least once in fresh vegetable products (Table 2), were compiled from the Pesticide Properties Database (PPDB) [33] (Table 3). The molecular structures of pollutants determine their partitioning behavior and degradation rates in environmental compartments [34]. The molecular weight of a pollutant affects the interphase diffusion coefficient [35]. The interaction between vapor pressure and water solubility values determines the partitioning behavior between air and water phases, while a high log K_{OW} value indicates a preference for the organic phase (hydrophobicity) compared to the water phase [36]. Half-life represents the time required for the concentration of a chemical in environmental compartments to decrease by half. Since the half-life can vary not only based on the chemical properties but also on the environmental characteristics, the values obtained from the literature should be used as rough estimates for approximate calculations [22].

In addition to their toxic effects, the persistence of pesticides in environmental compartments is also an important factor when regulating their use. A significant portion of the persistent organic pollutants (POP) listed in the Stockholm Convention and subjected to international measures are pesticides with high persistence [34]. Chlorpyrifos, who had been frequently detected to exceed MRL (Table 1) and had been recently banned in Türkiye, is among the chemicals proposed for listing as a POP under the Stockholm Convention [51]. Highly persistent pollutants can reach high concentrations in environmental compartments, thereby increasing exposure risks. When the relationship between the banned status of pesticides and their half-lives in Table 3 is examined, it can be observed that the average half-lives of banned pesticides in all compartments are higher than those of non-banned pesticides. However, the difference between the averages of the two groups is not statistically significant ($p>0.05$).

Environmental Fate Analysis

Locations of the Pesticides on the Chemical Space Diagram

The positions on the chemical space diagram of all the pesticides that exceeded MRL values in fresh vegetables (Table 3) have been determined (Fig. 1). It can be said that this group of pesticides, which are widely used and can reach high concentrations in vegetable products, tend to distribute between the

Table 1. Pesticide residue analysis studies focusing on fresh vegetables in Türkiye (Published scientific studies since 2010)

Reference	Year	Agricultural product	Pesticides exceeding the MRL	
[37]	2011	Tomato Pepper	Carbendazim Ethion Triazophos Oxamyl	
[38]	2014	Tomato Pepper Cucumber	Acetamiprid Alpha -Endosulfan Beta – Endosulfan Carbendazim Chlorpyrifos Clofentezine Cymoxanyl Dichlorvos	Dimethomorph Imidacloprid Malathion Methomyl Oxamyl Tebuconazole Triadimenol Trifloxystrobin
[39]	2016	Tomato	Acetamiprid Beta – Endosulfan Chlorpyrifos Tetradifone	
[40]	2017	Tomato	No active substance exceeding the MRL was detected	
[41]	2018	Tomato Pepper Cucumber	No active substance exceeding the MRL was detected	
[42]	2018	Tomato	Acetamiprid	
[43]	2018	Tomato	Acetamiprid Imazalil Iprodione	
[44]	2019	Tomato Pepper	Acetamiprid Bromopropylate Chlorpyrifos Cyproconazole Dichlorvos Etofenprox Etoxazole Fenarimol	Fenazaquin Formetanate -HCI Methomyl Metrafenone Omethoate Pendimethalin Pyridaben
[45]	2016	Tomato Pepper	No active substance exceeding the MRL was detected	
[46]	2021	Cucumber	Imidacloprid	
[47]	2022	Pepper	Acetamiprid Chlorpyrifos Etofenprox Etoxazole Fenazaquin Formetanate -HCI Methomyl Metrafenone Pyridaben	
[48]	2022	Tomato Cucumber	Pirimiphos Chlormequat chloride Pyridaben Chlormequa	
[49]	2022	Pepper Cucumber	Metrafenone Pyridaben	

Table 2. MRL values and pre-harvest waiting periods of pesticides that exceed residue limit values in fresh vegetables produced in Türkiye [32]

Pesticide active substance	Pesticide type	Substance group	MRL (mg/kg)			MRL (mg/a pre-harvest waiting period kg)		
			Tomato	Pepper	Cucumber	Tomato	Pepper	Cucumber
Acetamiprid	Insecticide	Neonicotinoid	0.5	0.3	0.3	3 days	3 days	3 days
Alpha - Endosulfan	Insecticide	Organochloride	Banned (2010)			–	–	–
Beta - Endosulfan	Insecticide	Organochloride	Banned (2010)			–	––	–
Bromopropylate	Acaricide	Diphenole	Banned (2011)			–	–	–
Carbendazim	Fugacide	Carbamate	Banned (2011)			–	–	–
Chlormequat chloride	Plant Growth Regulator	Quarternary ammonium compound	–	–	–	–	–	–
Chlorpyrifos	Insecticide	Organophosphate	Banned (2020)			–	–	–
Clofentezine	Acaricide	Tetrazine	–	–	0.2			3 days
Cymoxanyl	Fugacide	Cyanoacetamide	0.4	–	0.08	3 days	–	3 days
Cyproconazole	Fugacide	Triazole	No data available			No data available		
Dichlorvos	Insecticide	Insecticide	Banned (2011)			–	–	–
Dimethomorph	Fugacide	Morpholine	1	1	0.5	7 days	1 day	7 days
Ethion	Insecticide	Organophosphate	Banned (2010)			–	–	–
Etofenprox	Insecticide	Pyrethroid	0.7	-	-	3 days	–	–
Etoxazole	Acaricide	Diphenyl	0.07	0.01	0.02	3 days	3 days	3 days
Fenarimol	Fugacide	Pyrimidine	Banned (2011)			–	–	–
Fenazaquin	Acaricide	Quinazoline	0.05	–	–	3 days	–	–
Formetanate -HCI	Insecticide	Formamidine	0.3	–	0.01	14 days	–	7 days
Imazalil	Fugacide	İmidazole	0.3	0.01	0.5	3 days	3 days	3 days
Imidacloprid	Insecticide	Neonicotinoid	0.5	–	–	–	–	–
Iprodione	Fugacide	Dichlorophenyl	Banned (2018)			–	–	–
Malathion	Insecticide	Organophosphate	0.02	–	–	7 days	–	–
Methomyl	Insecticide	Carbamate	Banned (2021)			–	–	–
Metrafenone	Fugacide	Benzophenone	0.6	2	0.5	3 days	3 days	1 day
Omethoate	Insecticide	Organophosphate	Banned (2012)			–	–	–
Oxamyl	Insecticide	Carbamate	Banned (2012)			–	–	–
Pendimethalin	Herbicide	Dinitroaniline	0.05	–	–	Pre-planting	–	–
Pirimiphos - Methyl	Insecticide	Organophosphate	0.01	–	0.01	7 days	–	7 days
Pyridaben	Insecticide	Pyridazinone	0.15	0.3	0.15	3 days	3 days	3 days
Tebuconazol	Fugacide	Triazole	0.9	0.6	0.6	7 days	3 days	3 days
Tetradifone	Acaricide	Diphenyl	Banned (2011)			–	–	–
Triazophos	Insecticide	Organophosphate	Banned (2010)			–	–	–
Triadimenol	Fugacide	Triazole	Banned (2021)			–	–	–
Trifloxystrobin	Fugacide	Strobilurin	0.7	0.4	0.3	3 days	3 days	3 days

water and octanol phases instead of being present in the air. The distribution characteristics between the water and octanol phases vary depending on the values of the octanol-water partition ratio for these chemicals. However, most pesticides are in the intermediate region where significant distribution between both the water and octanol phases is expected.

The positions of acetamiprid, chlorpyrifos, and pyridaben, which frequently exceeded the MRL values in recent residue analysis studies (Table 1), are highlighted in red on the chemical space diagram (Fig. 1). These three pesticides are in three separate regions on the diagram, indicating that their environmental partitioning behaviors are different.

Table 3. Physicochemical properties of pesticides that exceed residue limit values in fresh vegetables produced in Türkiye [33]

Pesticide active substance	Molecular weight (g/mol)	Octanol– water partition ratio ($\log K_{OW}$)	Solubility in water (mg/l, 20 °C)	Vapour pressure (mPa, 20 °C)	Half-life (days)		
					Soil	Water	Sediment
Acetamiprid	222.67	0.8	2950	$1.73*10^{-04}$	1.6	4.7	337.5*
Alpha - Endosulfan	406.93	4.74	0.32	8.3	50	–	–
Beta - Endosulfan	406.93	3.83	0.45	–	–	–	–
Bromopropylate	428.1	5.4	0.1	0.011	59	4	63
Carbendazim	191.21	1.48	8.0	0.09	40	7.9	33.7
Chlormequat chloride	158.07	-3.47	886000	$1.0* 10^{-03}$	27.4	0.5	3.75
Chlorpyrifos	350.58	4.7	1.05	1.43	386	5	36.5
Clofentezine	303.15	4.09	0.0342	$6.0* 10^{-04}$	69.8	2.1	9.6
Cymoxanyl	198.18	0.67	780	0.15	1.7	0.3	0.3
Cyproconazole	291.78	3.09	93	0.026	142	–	1000
Dichlorvos	220.98	1.9	18000	2100	2	–	0.22
Dimethomorph	387.86	2.68	28.95	$9.7* 10^{-04}$	72.7	10	38
Ethion	384.48	5.07	2	0.2	90	–	–
Etofenprox	376.49	6.9	0.0225	$8.13* 10^{-04}$	11	5.7	13.3
Etoxazole	359.42	5.52	0.07	0.007	19.3	1.45	79.5
Fenarimol	331.20	3.69	13.7	0.065	250	4	Stabil
Fenazaquin	306.40	5.51	0.102	$1.90* 10^{-02}$	45	–	–
Formetanate -HCI	257.8	-0.0014	822000	$1.60* 10^{-03}$	12.9	0.3	0.3
Imazalil	297.18	2.56	184	0.158	76.3	7.8	117
Imidacloprid	255.66	0.57	610	$4.0* 10^{-07}$	191	30	129
Iprodione	330.17	3.0	6.8	0.0005	36.2	2.0	4.0
Malathion	330.36	2.75	148	3.1	0.17	0.4	0.4
Methomyl	162.21	0.09	55000	$2.13* 10^{-06}$	7	2.9	3.7
Metrafenone	409.27	4.3	0.492	0.153	200.9	3.9	9.3
Omethoate	213.2	-0.9	500000	19.0	0.1	–	4.5
Oxamyl	219.26	-0.44	184100	0.018	5.3	–	0.7
Pendimethalin	281.31	5.4	0.33	3.34	182.3	4	16
Pirimiphos - Methyl	305.33	4.2	11	$2.00* 10^{-03}$	39	–	–
Pyridaben	364.93	6.37	0.022	0.001	55	1.18	17.5
Tebuconazol	307.82	3.7	36	$1.30* 10^{-03}$	63	42.6	365
Tetradifone	356.06	4.61	0.078	$3.20* 10^{-05}$	112	–	–
Triazophos	313.3	3.55	35	1.33	44	35	35
Triadimenol	295.76	3.18	72	0.0005	250	53	91
Trifloxystrobin	408.37	4.5	0.61	$3.40* 10^{-03}$	0.34	1.1	2.4

*: Acetamiprid sediment half-life value was obtained from EPI Suite [50].

Acetamiprid has the lowest log K_{OW} value among the three pesticides and its vapor pressure is low. Therefore, it is expected to be present in the water phase. Acetamiprid's relatively high solubility increase its likelihood of transport into plants through water uptake from soil. Additionally, the risk of surface water and groundwater pollution by Acetamiprid should be considered.

Chlorpyrifos is located to the right of the middle region of the diagram, indicating a tendency for presence in the octanol phase with non-negligible partitioning in the air and water phases (Fig. 1). Chlorpyrifos has a higher air-water partition coefficient compared to the other pesticides shown in the diagram. Due to its high half-life values in both sediment and soil, Chlorpyrifos is expected to remain and accumulate in the soil compartment where it is applied. Chlorpyrifos is a widely used pesticide that can cause environmental contamination in air, water, and soil, and it has

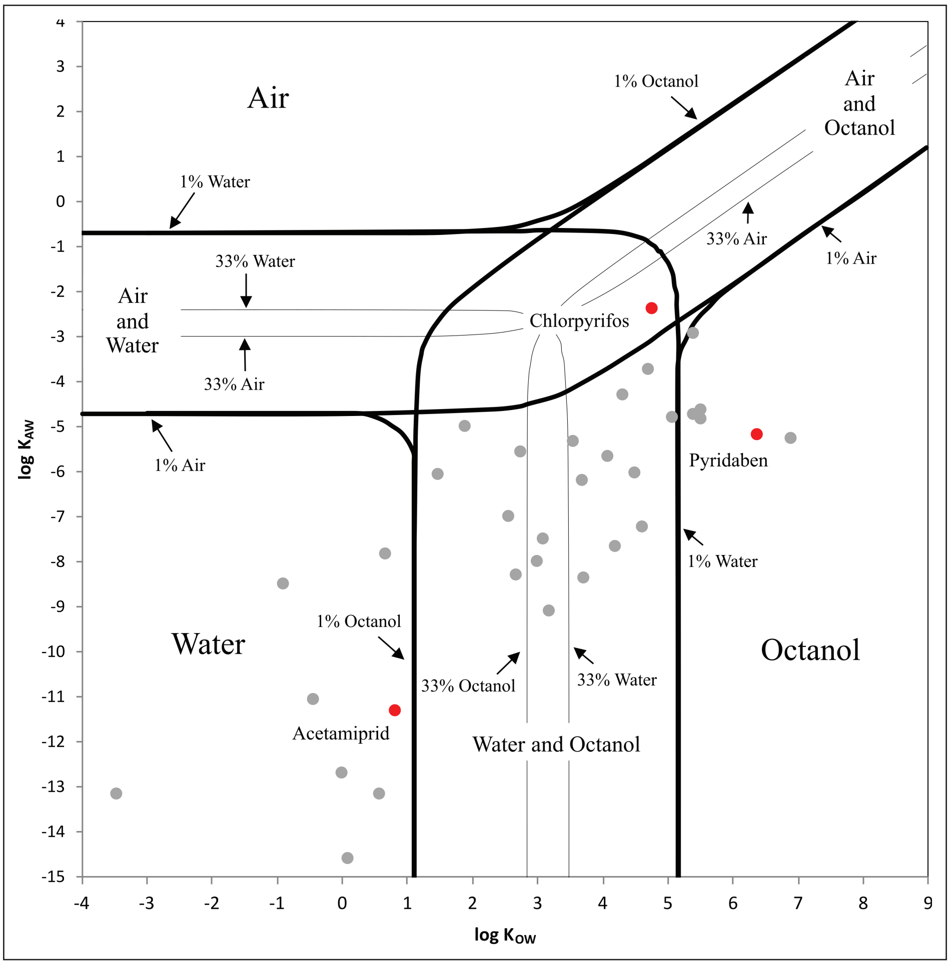

Figure 1. Chemical Space Diagram (Volatile substances tend to locate on the upper-left, water-soluble substances tend to locate on the lower-left, hydrophobic substances tend to locate on the lower-right [23]. The lines indicate constant percentages between the air, water, and octanol phases).

been associated with various health issues, including endocrine disruption [52]. Its use in Türkiye has been banned.

Pyridaben has the highest log K_{OW} value among these three chemicals. It is expected to tend to be present in the octanol phase. This characteristic may contribute to its sorption to soil and accumulation in the lipid tissues of plants. However, a more detailed modeling approach is necessary to assess the risk associated with its transport to surface water and groundwater.

Equilibrium Criterion (EQC) Model Results

Acetamiprid, chlorpyrifos, and pyridaben were subjected to Level III calculations using the EQC model. Level III mass balance calculations assume a steady-state condition but do not assume equilibrium between compartments, taking into account advection, diffusion, and reactive transfer processes. The properties of the Level III standard evaluative environment of the EQC model are given in Hughes et al. [27]. It was assumed that the three modeled pesticides enter the standard environmental system through emission to soil. The same emission rate (1000 kg/hour) was assigned to each pesticide, and the results were evaluated comparatively. The half-lives of acetamiprid, chlorpyrifos, and pyridaben were assigned the values of 3.36, 4.38, and 6.06 hours, respectively, as obtained from EPI Suite [50]. The modeling results are summarized in Figure 2–5.

The mass distributions of the three pesticides among the environmental compartment were calculated (Fig. 2). Soil, the compartment where the emissions occur, contained the largest mass fraction for all the pesticides. Acetamiprid was found in the water compartment in significantly high amounts, and it had almost no presence in the air compartment. The pesticide that had the largest fraction in the soil compartment was Chlorpyrifos. Also, a significant amount of Chlorpyrifos was present in the air compartment. Pyridaben's compartmental mass distribution was similar to Chlorpyrifos, but it had a higher fraction in water and a lower fraction in air. These results were in accordance with the information obtained from the chemical space diagram. However, none of the pesticides were distributed to the sediment compartment in significant amounts. This result is related with the intermedia mass transfer properties of the pesticides. And, it can change if there are direct emissions to the compartments other than soil. Also, although the total mass in the sediment compartment is low, since sediment volume is small compared to the other compartments, the concentrations may still reach dangerous levels.

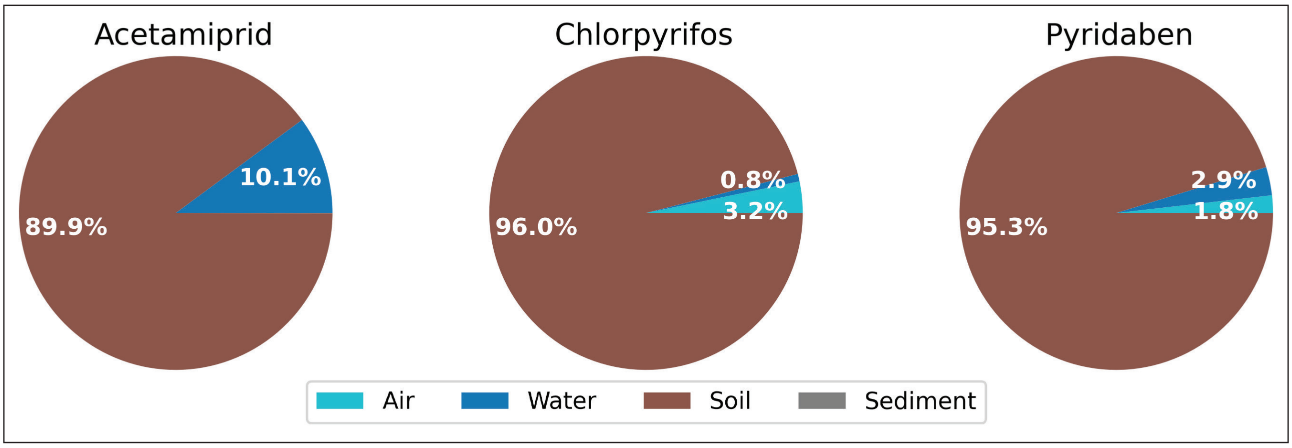

Figure 2. Compartmental mass distributions of the pesticides in the EQC model environment.

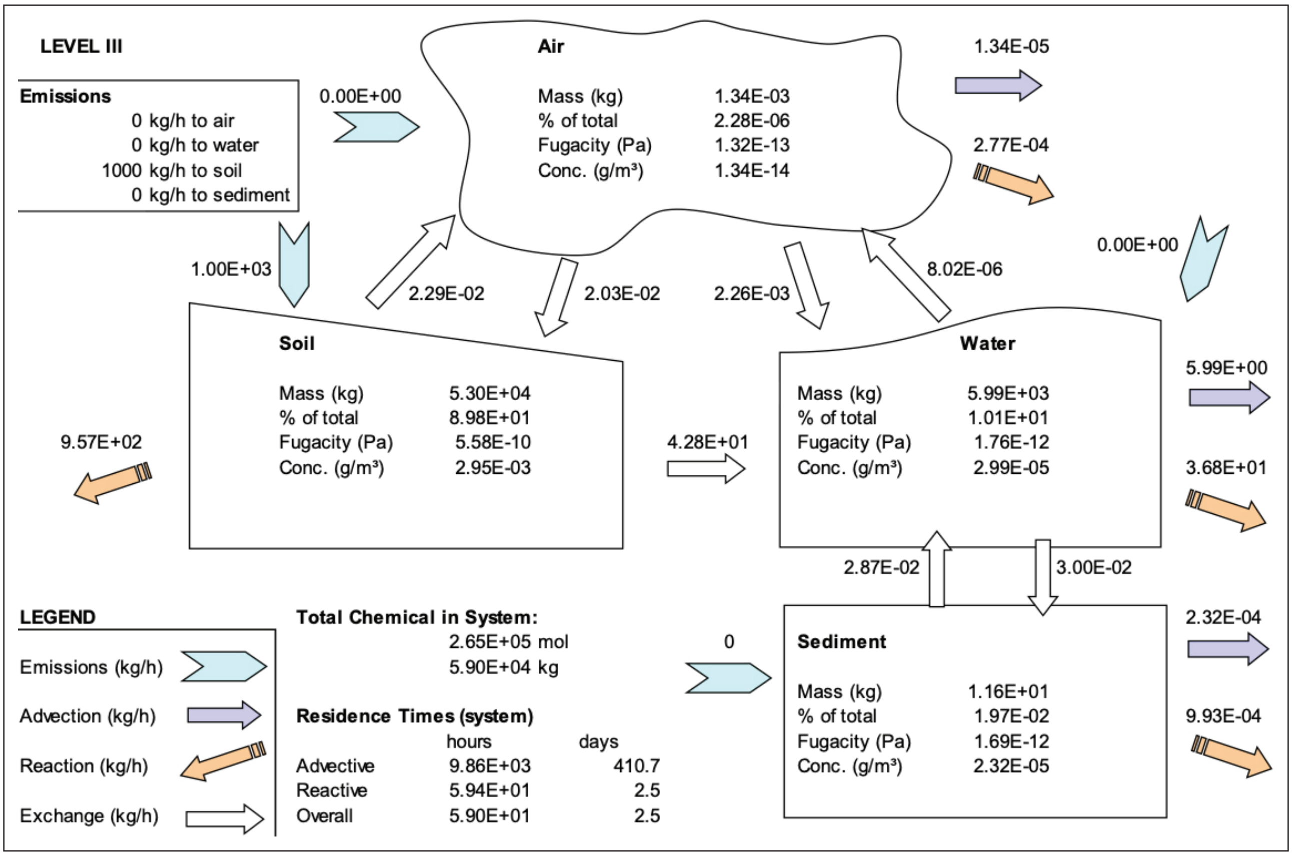

Figure 3. EQC model results for acetamiprid.

Mass transport rates and the residence times calculated by the EQC model are provided in the summary diagrams in Figure 3–5. Acetamiprid's overall persistence in the EQC environment is 2.5 days, lower than the other two pesticides (Fig. 3). When the model results for Chlorpyrifos is examined, it's high soil-to-air transfer can be seen (Fig. 4). Correspondingly, there are significant reactive losses in the air compartment. Despite its high hydrophobicity, chlorpyrifos tends to transfer to the air compartment, increasing its advection and reactive losses in the system, resulting in a total persistence value of 7.5 days. The intercompartmental distribution behavior of pyridaben is similar to Chlorpyrifos (Fig. 5). However, it shows less tendency for transfer to the air compartment, resulting in relatively higher accumulation in the water and sediment compartments. This limits the advection and reactive losses, causing pyridaben to have a slightly higher total persistence in the system compared to chlorpyrifos. The total persistence of pyridaben is calculated as 11.9 days. The main loss mechanism is reaction for all the three pesticides.

The net intermedia transfer rates of the three pesticides are given in Table 4 as a fraction of the total emission rate (1000 kg/h). A comparative examination of the intermedia transfer rates reveals the dominant direction of movement for the pesticides after being released to the soil compartment. It is seen that the dominant movement of acetamiprid in

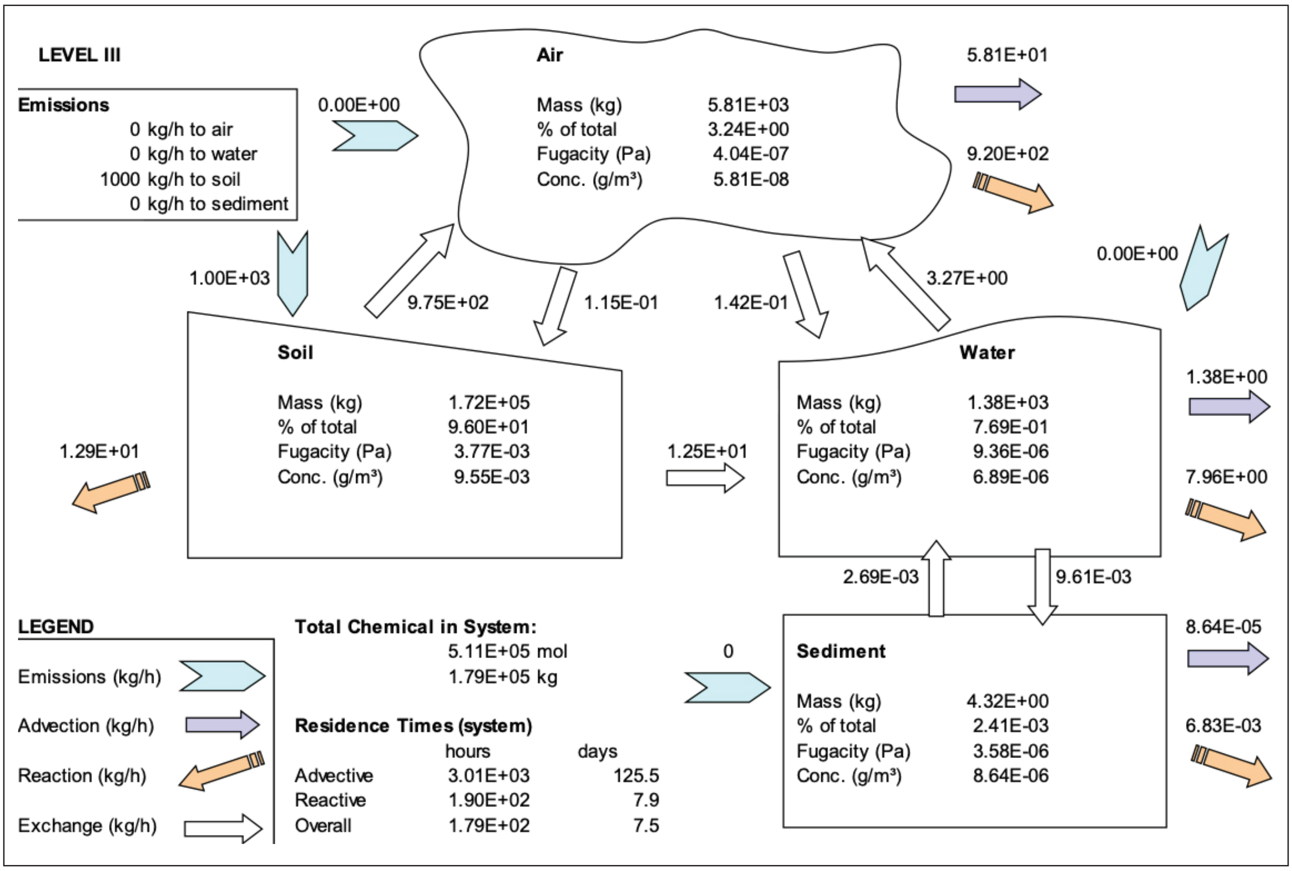

Figure 4. EQC model results for chlorpyrifos.

Table 4. Net intermedia transport rates for the three pesticides after being emitted to soil (fractional rates to the total emission rate) (EQC Model Results)

	Acetamiprid	Chlorpyrifos	Pyridaben
Soil-to-air	2.54E-06	97.47%	63.76%
Soil-to-water	4.28%	1.25%	21.92%
Water-to-air	-2.25E-06	0.31%	0.60%
Water-to-sediment	1.23E-06	6.92E-06	1.51E-05

the environmental system is from soil to water. Acetamiprid experiences very limited transfers between the other compartments. The potential of acetamiprid to pollute surface waters and groundwater should be further investigated using more detailed models. For chlorpyrifos, the dominant transfer is from soil to air, where it is lost from the system mainly by degradation reactions and advection. Although degradation in air is the main loss mechanism for chlorpyrifos, advective loss through atmospheric transport is not negligible. Long range transport potential of chlorpyrifos through atmosphere is a significant concern supported by monitoring and modeling studies [53]. Long-range transport potential is one of the criteria for chemicals to be classified as a persistent organic pollutant (POP), and chlorpyrifos is a POP candidate under the Stockholm Convention [51]. For pyridaben, soil to air transfer is dominant, but its soil to water transfer is also significant. Since EQC model results indicates higher persistence for pyridaben compared to chlorpyrifos, the potential for pyridaben to reach remote regions through atmospheric and aquatic transport should be further investigated.

Time Dependent Residue Values in Tomato

DynamicCROP model's regression equations were used to calculate the time-dependent harvest fractions for acetamiprid, chlorpyrifos, and pyridaben in tomato products (Fig. 6). Acetamiprid's harvest fraction, and consequently, its residue amount, exhibits a faster decline compared to the others. On the other hand, acetamiprid's initial harvest fraction is higher compared to the other two pesticides. This is due to its lower molecular weight value, which facilitates its incorporation to the tomatoes after being applied. As a result of the high persistence values of chlorpyrifos and pyridaben, a slower decrease in the harvest fraction is observed for these two chemicals. In tomato production, the recommended pre-harvest waiting period for acetamiprid and pyridaben is three days, and the MRL values are 0.5 and 0.15 mg/kg, respectively (Table 2). After three days, a decrease in residue levels is observed for all pesticides. However, it is predicted that Acetamiprid's residue rate will still be relatively high when compared to the initial residue values. For these two pesticides, re-evaluating application doses and pre-harvest waiting periods through more specific model calculations and controlled experiments can provide useful information for preventing the exceedance of MRL values.

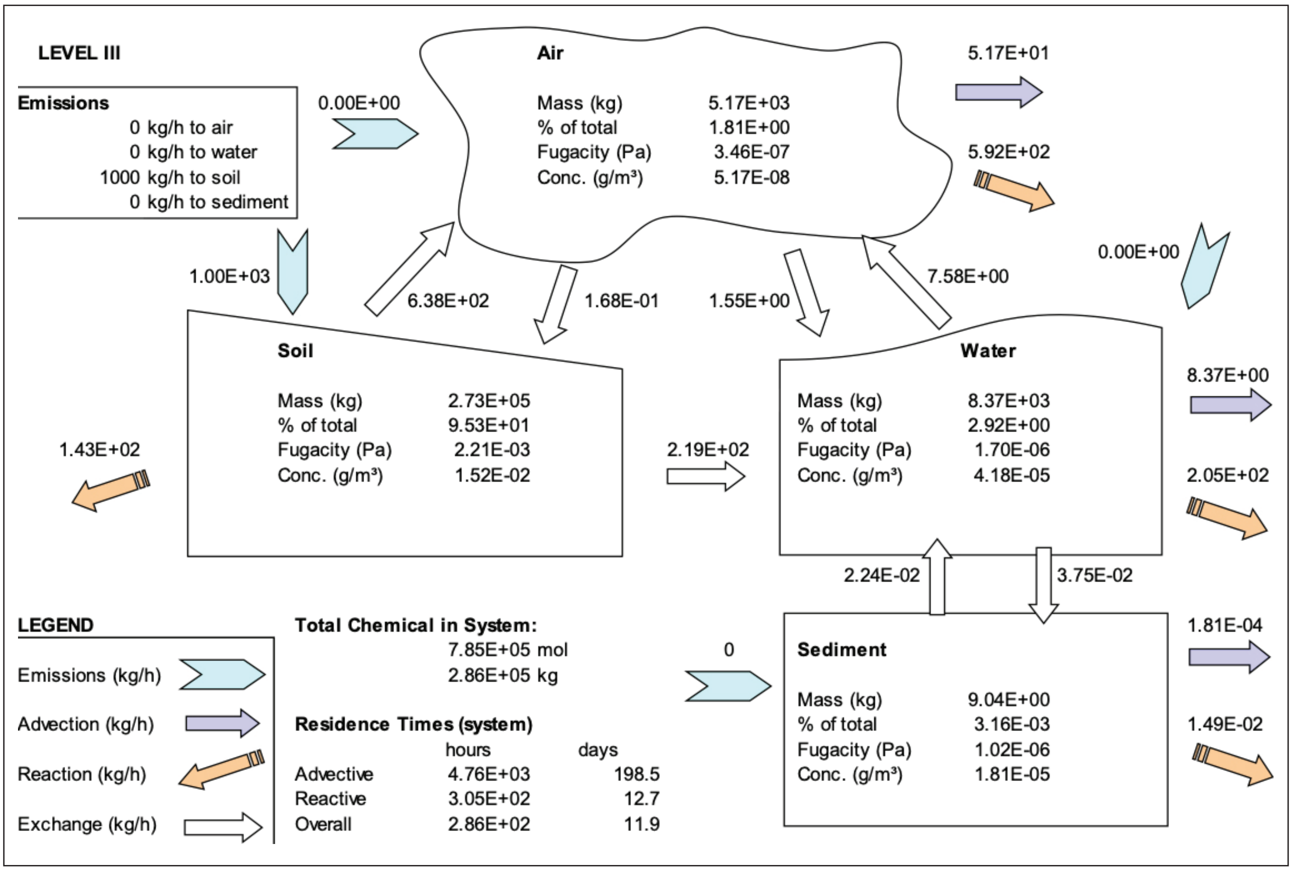

Figure 5. EQC model results for pyridaben.

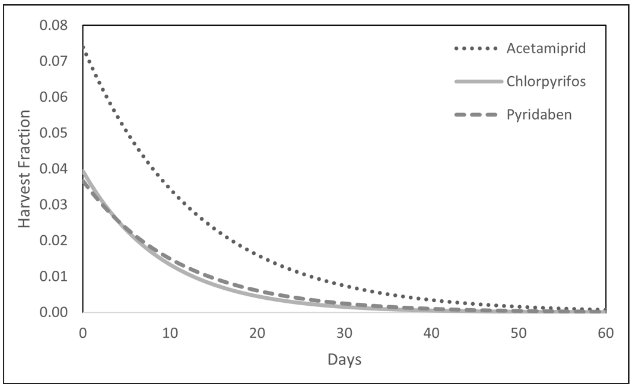

Figure 6. Time dependent harvest fractions estimated by the plant uptake model.

CONCLUSION

This study presents modeling methods for assessing the environmental exposure risks of pesticides. Pesticides, widely utilized in agricultural practices due to their efficacy and cost-effectiveness, can pose potential risks to both the environment and human health when misused or applied improperly. Also, the residue levels exceeding the regulated MRL values causes significant economic losses. In the reviewed scientific studies, there were many instances of pesticide residues exceeding MRL values and banned pesticides were also identified in the products. This indicates that existing preventive measures may not be entirely adequate.

In the first part of this study, recent published research on pesticide residues in fresh vegetables produced in Türkiye was examined. The reviewed research findings showed a significant occurrence of pesticide residues exceeding the MRL values. The frequent exceedance of MRLs in agricultural products can lead to health risks for consumers and may cause environmental problems in agricultural regions. Moreover, the disposal of agricultural products containing pesticide residues above permissible levels, results in economic losses. The quantity of pesticide residues in fresh vegetable products is influenced by various factors, including the mode of pesticide application, dosage, timing, and the physicochemical properties of the pesticide active ingredients. In this study, the physicochemical properties determining the fate of problematic pesticides in environmental compartments and plants, along with the recommended pre-harvest intervals, were identified referring to up-to-date sources.

The second part of the study involved conducting calculations to demonstrate the applicability of multi-media fate models in evaluating the environmental and health risks of pesticides. A chemical space diagram was used to assess the environmental partitioning behavior of the pesticides. The diagram provided valuable insights into how pesticides would distribute among air, water, and solid organic phases upon release into the environment. It was observed that most pesticides associated with exceeding regulatory limits tend to accumulate in the water and organic solid phases. Further detailed evaluations were performed for three pesticides (acetamiprid, chlorpyrifos, pyridaben) that were found to exceed MRL values relatively more frequently. These three pesticides have different locations in the chemical space diagram. Acetamiprid tends to be present in the water phase, while pyridaben shows an affinity for the organic solid phase. On the other hand, chlorpyrifos, which is currently banned from use in Türkiye, demonstrates a relatively higher affinity for the air phase compared to the other pesticides.

The Equilibrium-Criterion Model (EQC), which includes the air, water, soil, and sediment compartments, were applied to assess the multi-media environmental fate and transport characteristics of the selected three pesticides. Level III steady-state model calculations were performed under the same environmental conditions and emission scenarios, enabling a comparative evaluation. The accumulation levels in the four compartments, inter-compartmental transfer rates, and losses from the system due to advective and reactive processes were calculated for all three pesticides. It was observed that all three pesticides reached the highest accumulation in the soil compartment, where they were initially released to the environment. When comparing the environmental persistence of the three pesticides, the order is as follows: pyridaben > chlorpyrifos > acetamiprid. Degradation reactions were identified as the main loss process for all pesticides. Despite chlorpyrifos having higher reaction half-lives in water and soil, pyridaben showed higher environmental persistence. This can be attributed to pyridaben's higher hydrophobicity but lower volatility compared to chlorpyrifos. The use of more detailed multi-media fate models based on EQC but tailored to represent specific agricultural regions can help assess pesticide related environmental pollution. The model results can guide field and sampling studies. Model simulations can be conducted to contribute to agricultural pollution management efforts.

Finally, calculations were performed using a dynamic plant uptake model, called dynamiCROP, for the selected three pesticides. In these calculations, dynamiCROP's regression equations were used, and the time-dependent changes in the pesticides' harvest fractions were compared. The results of the calculations indicated that acetamiprid had a higher initial uptake into tomatoes compared to the other two pesticides. This finding was interpreted to be due to acetamiprid's relatively lower molecular weight, which may result in higher diffusion into tomatoes after application. On the other hand, acetamiprid residues in tomatoes were found to decline faster than the other two pesticides. The dynamics of chlorpyrifos and pyridaben residues were similar to each other. Conducting simulations using the full version of the dynamiCROP model would allow more detailed evaluations. Additionally, similar simulations can help in the determination of the pre-harvest waiting time and the initial pesticide dosage in agricultural applications.

The use of environmental fate models in the production and authorization processes of pesticides can reveal the potential environmental issues at an early stage enabling the application of preventive measures. This approach can provide guidance when determining the pesticide's dosage, application method, pre-harvest waiting period, and potential residue levels. Furthermore, the integration of environmental fate and transport models with plant uptake models can enable comprehensive analyses at field and regional scales. Similar models can be employed to aid remediation plans in regions where pollution problems exist.

DATA AVAILABILITY STATEMENT

The author confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

CONFLICT OF INTEREST

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

ETHICS

There are no ethical issues with the publication of this manuscript.

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