



Human Exposure to an Organophosphate Pesticide During Spraying – an Experimental Model

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ABSTRACT *This study assessed the human exposure to chlorpyrifos-methyl (organophosphate pesticide) in experimental conditions. We conducted the study on an uncultivated field by measuring pesticide's concentrations at adult's and children's respiratory level, during a spraying session. Also the daily intakes in humans were calculated. The data obtained showed that pesticide dispersion dependent on wind speed leads to the loss of a significant amount of substance, which may pose health hazards at least equal or greater for the population outside the spraying area, in comparison to the people directly exposed. The daily intakes calculated for the similar concentrations of the pesticide were higher in children compared to adults and for males compared to females. For both children and adults, the daily intakes of chlorpyrifos-methyl calculated in our experiment model were lower than ADI. The weather conditions, such as wind speed, humidity and temperature should be considered in spraying timing.*

Introduction

Organophosphates (OPs) are a group of synthetic pesticides used to control various insects on crops and in homes (Levine, 2007; Tadeo et al., 2008), and are widely used in agriculture. These pesticides are highly toxic cholinesterase-inhibiting compounds, but tend to degrade in the environment with exposure to sunlight and water. The lack of knowledge about how OPs should be applied and poor monitoring of their use have been associated with various human health problems, while environmental pollution by OP residues has accumulated over time (CDC, 2009; Levine, 2007; WHO, 2005). Human exposure to OP insecticides is widespread. Occupational use of OP insecticides, primarily in agriculture, represents the largest class of exposure. However, many populations, including children, have been shown to be widely exposed to OP insecticides through their diet, residential use, by living close to farms and from para-occupational exposures via parents working in agriculture (Coronado et al., 2006; Naeher et al., 2010; Munoz-Quezada M.T. et al, 2012).

Air pollution due to pesticides is a persistent problem in modern agriculture. Pesticides contaminate the atmosphere through various pathways. (Gil & Sinfort, 2005). Spray drift (pesticide's movement in the air away from the spraying site) is a complex problem in which equipment design and application parameters, spray physical properties and formulation, as well as meteorological conditions interact and influence pesticide loss. Weather conditions such as wind, temperature, relative humidity and precipitation influence the effectiveness of spray applications and the potential for wastage by run-off and drift. (Deveau, 2009)

Highly concentrated agrochemical residues generated during spray application can move (drift) beyond the target foliage (or in some cases, target soil) to non-target receptors including water, plants, animals and humans. Non-target receptors may be acutely exposed and therefore face the greatest risk of adverse effects during and immediately after the spray application. In addition to agrochemical residues movement in turbulent air masses downwind of application site, residues can also become concentrated in inversions or stable air masses and transported long distances. Similarly, agrochemicals can volatilize from plant and soil surfaces in

comparatively high concentrations for several days after application. These secondary drift residues also pose a hazard to nearby non-target receptors. (Felsot et al, 2010)

A previous work of Lovász and Gurzău (2013) conducted in a rural area showed that 76% of all persons who worked with pesticides have never used protective equipment.

The present study was conducted outside the built-up area of Sâncraiu locality on an uncultivated field in order to monitor under field conditions the dispersion in the air of chlorpyrifos-methyl (O,O-dimethyl O-3,5,6-trichloro-2-pyridyl phosphorothioate) a compound belonging to the organophosphate pesticides group.

Since inhalation may be an important route of exposure to pesticides, the present study assessed farmer's exposure to chlorpyrifos-methyl during an experimental spraying session.

Materials and methods

For measuring pesticides concentrations in the air during spraying, 3 series of manual pump (Knapsack sprayer) spraying were performed of about 20 minutes each with RELDAN 22 EC insecticide, containing chlorpyrifos-methyl 225 g/l as active substance. Spraying solution (0.2%) was prepared on the spot before each spraying. Manual pump spraying was performed at a height between 0,5m and 1,5m from a fixed point in wind direction. The air samples were collected using sampling pumps with absorbent tubes (OVS-2 tube: 13mm quartz filter, XAD-2 140/270 mg). The sampling pump has been set and calibrated at a flow rate of 1 L/min by means of a rotameter prior to each sampling series. The sorbent tubes were fixed at heights of 0.9 (respiratory level for children) and 1.5 m (respiratory level for adults) and at distances of 0m, 2m and 5m downwind from the spraying site. Meteorological conditions (temperature, humidity, pressure, wind speed and direction) have been monitored during each experiment by means of Pro X USB weather station (Irox) at 1.5m height.

The sampling method for measuring the pesticides concentration in air was in compliance with NIOSH manual of analytical methods, number 5600 (NIOSH 5600, 1994). The quartz filter and XAD-2 frontal section were transferred to a 4 ml

vial and the short socket of polyurethane foam together with XAD-2 spare section was transferred to a separate 4 ml vial. The desorption solvent (2 mL acetone/toluene solution: 1/9) was added to each vial and allowed to rest for 30 minutes. The sample was then extracted in an ultrasonic bath for 30 minutes. By means of a Pasteur pipette the liquid phase was transferred in 1.5 ml sample vials and analyzed by a gas chromatograph coupled with mass spectrometer GSMS-QP2010 Plus.

The detection limit of the method (MDL) for chlorpyrifos-methyl was 0.00018 mg/mc. Control samples determined for each set of analysis fell within the standard deviation for the average value ($0.030 \pm 0.0023 \mu\text{g/ml}$). Quality control was made through specific methods, to evaluate the accuracy of our analyses, meeting the Romanian Accreditation Association standards and certifications.

In the statistical processing ANOVA, Student's t test and test „r” (Person-Bravais) was used.

The daily intake of chlorpyrifos-methyl was calculated using a calculation model developed by ATSDR (Agency for Toxic Substances and Disease Registry within the Center for Disease Control and Prevention, part of the U.S. Department of Health and Human Services) in 2008.

Results

During the experiment 18 samples have been collected for the determination of chlorpyrifos-methyl concentrations in air. The results are presented in Table 1.

Table 1: Chlorpyrifos-methyl concentrations in air and meteorological conditions during pesticide RELDAN 22 EC spraying

Spraying	Distance (m)	Concentration at 0.9m height (mg/m ³)		Concentration at 1.5m height (mg/m ³)		Meteorological conditions			
		Wind speed (m/s)	Wind direction	Temperature (°C)	Humidity (%)	Pressure (kPa)			
I.	0m	0.0017	0.0016	1	South-West	13	75	93.8	
	2m	0.0201	0.0219						
	5m	0.0139	0.0103						
II.	0m	0.0006	0.0012	1.3	South-West	13	75	93.9	
	2m	0.0102	0.0160						
	5m	0.0086	0.0119						
III.	0m	0.0003	0.0003	1.6	South-West	15	70	93.9	
	2m	0.0163	0.0108						
	5m	0.0116	0.0176						

Chlorpyrifos-methyl concentrations measured in air during the three spraying sessions were ranging between 0.0003 mg/m³ and 0.0219 mg/m³. The peak concentration during the three spraying sessions (0.0219 mg/m³) was measured at the height of 1.5m and at a distance of 2m from the application point. The lowest concentration (0.0003 mg/m³) was recorded at a wind speed of 1.6m/s at the application point (at 0m), at the height of 0.9m and 1.5m.

Increasing wind speed decreased the measured chlorpyrifos-methyl concentrations in air both at the height of 0.9m and 1.5m, at the application point (0m). At the distance of 2m and 5m, the concentrations changed only at the height of 1.5m, decreasing in the first case and increasing at the distance

of 5m. Air temperature, humidity and atmospheric pressure were relatively constant in all of the situations. (Figure 1)

The amount of spray drift is directly proportional to wind speed and inversely proportional to relative humidity. Since relative humidity is related to temperature, the value of spray drift is directly proportional to temperature too. (Nuytens et al, 2006).

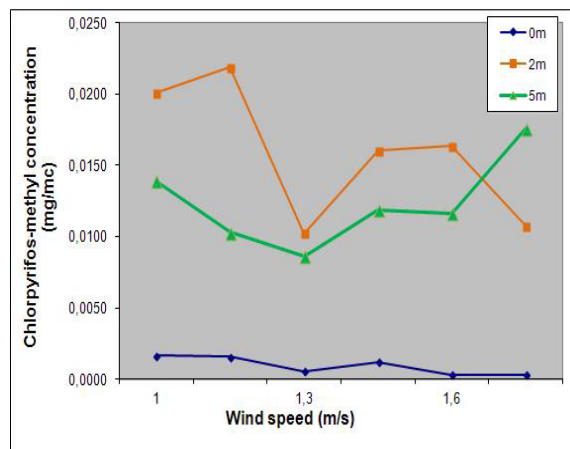


Figure 1: Chlorpyrifos-methyl concentration dependent on wind speed

ANOVA test for a single factor shows that there are no significant differences between sprayings (p=0.73) referring to the measured concentrations of chlorpyrifos-methyl in air.

The correlation test „r” (Pearson-Bravais) shows that there are no correlations between chlorpyrifos-methyl and the height of sampling (r=0.064)

Student's t-test between average concentrations measured at 0.9 m and 1.5 m shows there are no significant differences (p=0.8).

On the other hand, student's t-test of differences between average concentrations shows that there are very significant differences between the samples collected at 0m, 2m, 5m, respectively (p<0.001). The same thing is demonstrated by applying ANOVA test for a single factor (f=33.6 and p<0.001).

Figure 2 shows that the concentrations of chlorpyrifos-methyl at 0m and 2m decrease dependent on the wind speed (R²=0.73, R²=0.42) while at 5m distance concentrations increase at 1.3 m/s and 1.6 m/s, respectively (R²=0.19).

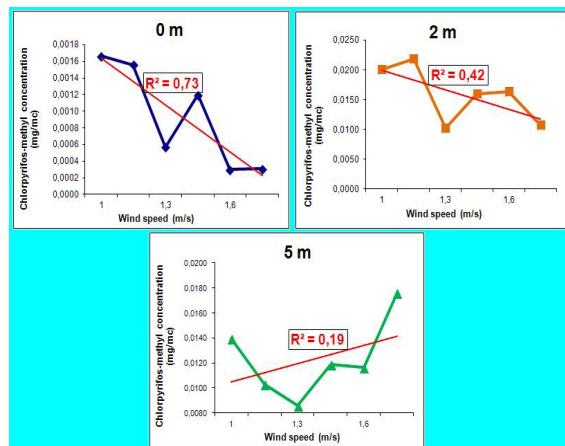


Figure 2: Regression line for chlorpyrifos-methyl concentrations depending on height, distance and wind speed.

Chlorpyrifos-methyl is classified as a noncarcinogenic organophosphate substance used against insects, which inhibits acetylcholinesterase activity. The acceptable daily intake (ADI) of chlorpyrifos methyl is 0–0.01 mg/kg body weight (bw), determined at the Joint FAO/WHO Meeting on Pesticide Residues (JMPR), where the compound was assessed in 1975, 1991 and 1992. (WHO, 2009)

Based on the lowest, highest and mean measured concentrations of chlorpyrifos-methyl during the experiment we have calculated the daily intake through inhalation for children (6-8 and 12-14 years old) and adults. The obtained daily intakes in each situation were compared to the acceptable daily intake (ADI) (WHO 2009).

	Concentration (mg/m ³)		Inhalation rate (m ³ /zi)	Intake (mg/kg/day)	% Acceptable daily intake (ADI) of Chlorpyrifos-methyl
Children (6-8 years, 25 kg)	Highest	0.0201	10	0.00804	80.4
	Lowest	0.0003		0.00012	1.2
	Mean	0.009		0.00344	34.4
Children-male (12-14 years, 49 kg)	Highest	0.0201	15	0.00615	61.5
	Lowest	0.0003		0.00009	0.9
	Mean	0.009		0.00263	26.3
Children-female (12-14 years, 49 kg)	Highest	0.0201	12	0.00492	49.2
	Lowest	0.0003		0.00007	0.70
	Mean	0.009		0.00211	21.1
Adult- male (19-65 years, 70kg)	Highest	0.0219	15.2	0.00476	47.6
	Lowest	0.0003		0.00007	0.7
	Mean	0.010		0.00224	22.4
Adult- female (19-65 years, 70kg)	Highest	0.0219	11.3	0.00354	35.4
	Lowest	0.0003		0.00005	0.5
	Mean	0.010		0.00166	16.6

Table 2: Chlorpyrifos-methyl Intake Dose Via Inhalation

The comparisons between the intake dose calculated for children and the adults and the recommended acceptable daily intake (ADI) values are presented in Table 2.

The results showed that the daily intake related to different concentrations decreases with age, this aspect being mostly evident in the case of the pesticide's highest concentrations. At the similar concentrations of the pesticide, the children's daily intake is higher compared to adults. In our experimental model the daily intake doses do not reach the ADI, the calculated values representing a different percent from ADI (80.4% for children aged 6-8, 61.5% or 49.2% for children aged 12-14 and under 50% for adults).

In terms of gender, the same exposures is related to higher daily intake for males versus females, both for children and adults, due to the inhalation rate.

Conclusions

During our experiment, using the regular dilution of the active substance, the chlorpyrifos-methyl concentrations were measured in air ranged from 0.0003 mg/m³ to 0.0219 mg/m³.

Measurements have shown the significant effect of wind speed on the amount of spray drift (dispersed). Related to this, people can be exposed to pesticides even at large distances from the spraying point, causing potential health effects.

The measured concentration of chlorpyrifos-methyl at 1.5 height (adult respiratory level) shows that the exposure level for adults can be greater than for children, which not necessarily leads to a higher level of risk.

Consequently, the daily intakes calculated for the similar concentrations of the pesticide were higher in children compared to adults and for males compared to females. Children could face higher risks of adverse effects than adult farmers when exposed to chlorpyrifos-methyl during spraying.

For both children and adults, the daily intakes of chlorpyrifos-methyl calculated in our experiment model were lower than ADI.

Even so, weather conditions, such as wind speed, humidity and temperature should be considered in spraying timing.

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