Science of the Total Environment 584–585 (2017) 856–868

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Human exposure and risk assessment to airborne pesticides in a rural French community

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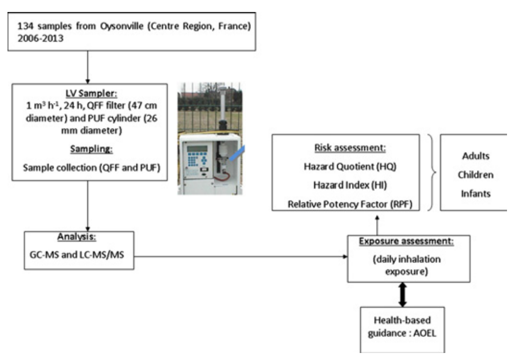
HIGHLIGHTS

- Human inhalation risk assessment of outdoor airborne pesticides was performed.
- The levels of 41 detected pesticides ranged from 0.12 ng m^{-3} to 1128 ng m^{-3}.
- All measured reported Hazard Quotients <1
- The cumulative risk for the two pesticide types assessed is acceptable.
- For infants the estimated cancer risk was lower than 8.93×10^{-05}.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 29 September 2016
Received in revised form 19 January 2017
Accepted 19 January 2017
Available online 1 February 2017

Editor: D. Barcelo

Keywords:
Pesticides
Exposure assessment
Risk assessment
Atmospheric concentrations

ABSTRACT

Outdoor air samples collected during the pesticide agricultural application period (spring and summer) from a rural community in the Centre Region (France) were analyzed to investigate temporal variation of atmospheric pesticide levels (2006–2013) and human inhalation exposure in adults, children and infants.
The most frequently detected pesticides were herbicides (trifluralin, pendimethalin), fungicides (chlorothalonil) and insecticides (lindane and α-endosulfan). The three currently-used pesticides most frequently detected presented concentrations ranging from 0.18 to 1128.38 ng m^{-3}; 0.13 to 117.32 ng m^{-3} and 0.16 to 25.80 ng m^{-3} for chlorothalonil, pendimethalin and trifluralin, respectively.
The estimated chronic inhalation risk, expressed as Hazard Quotient (HQ), for adults, children and infants, was <1 for all measured pesticides. Likewise, the cumulative exposure for detected organophosphorus and chloroacetamide pesticides, was estimated using the Relative Potency Factor (RPF) and Hazard Index (HI) as metrics, which was indicated that no risk was observed. The cancer risk classified as likely or possibly carcinogen was estimated to be <8.93 E−05 in infants, for the detected pesticides.

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http://dx.doi.org/10.1016/j.scitotenv.2017.01.132

1. Introduction

A wide variety of pesticides can be applied in agriculture and their identity depends on a range of factors including the specific pest and crop of interest. During 2010, about 208.000 t of pesticide active ingredients were used in Europe (EU-15) (ECPA, 2010) and >300 active substances are nowadays authorized by the European Union for their application on various crops according to the Regulation (EC) 1107/2009 (EU, 2014). Several studies in Europe have reported pollution of ambient air by agricultural pesticides in concentrations ranging from few pg m^{-3} to several ng m^{-3} (Schummer et al., 2010; Borrás et al., 2011; Coscollà et al., 2010; Hart et al., 2012; Raeppel et al., 2014). The potentially adverse effects of exposure to pesticides on the general population, and specifically on the more susceptible groups such as infants and children, are a public health concern (Marks et al., 2010; London et al., 2012). Apart from ingestion of foods, drinking water and dermal exposures, inhalation of ambient air could also be a relevant pathway of exposure to pesticides.

A major proportion of the applied pesticides are emitted to the atmosphere. Post-application emissions that involve volatilization from soil and plants, and wind erosion of soil particles containing sorbed pesticides represent further significant pesticide input into the troposphere for several days or weeks after pesticide application (Scheyer et al., 2007). Therefore, accurate information on active substance usage and inventories of emissions are relevant for assessing the exposure and risk to human populations (Sarigiannis et al., 2013).

As a result of agricultural use, a large number of pesticides have been detected in outdoor ambient air. Hart et al. (2012) detected 24 CUPs (currently used pesticides) in the particulate matter, in concentrations between 3.2 and 1196 ng m^{-3} in the atmosphere of the Valencian region (Spain). These authors also reported the levels of 17 more polar CUPs in the PM10 with concentrations ranging from 6.8 to 2898 pg m^{-3} in the same area (Coscollà et al., 2013). Moussaoui et al. (2012) assessed the contamination level of pesticides in ambient air of northern Algeria, detecting concentrations levels from 16 pg m^{-3} to 11 ng m^{-3}. In a more recent study, Yusà et al. (2014) analyzed 56 samples from April to October of 2010 from a rural station in the Valencian region. Twenty out of the 82 pesticides searched were found at this rural site. The average concentrations ranged from 5.75 to 117.01 pg m^{-3}, with maximum concentrations lying between 15.54 and 758.60 pg m^{-3}. Coscollà et al. (2014) mainly found carbendazim, metalaxyl, terbuthylazine and myclobutanil in concentrations from 16 pg m^{-3} to 174 pg m^{-3} in ambient air (June–July 2013) at a rural station located in the same area. Mahmood et al. (2014) assessed the organochlorine pesticide contamination and its probable hazardous effects on human health such as carcinogenic, reproductive, neurological, immunological and other adverse effects. They collected samples from Gujranwala, Punjab Province, Pakistan detecting concentration ranges from 123 to 635 pg m^{-3}.

Although a guidance on pesticide exposure and risk assessment for operators, workers, bystanders and residents was recently developed (EFSA, 2014a), the methodologies focused on the exposure and risk assessment of air pesticides on the general population are scarce. Yusà et al. (2014) proposed a screening approach for inhalation chronic risk assessment of CUPs present in ambient air, based on the concentration of these compounds in the inhalable particulate matter (PM10). They applied this approach in a pilot case study for the risk assessment of an agricultural community in Valencia, Spain. In California, the Department of Pesticide Regulation from the Environmental Protection Agency implemented an air monitoring network for measuring pesticides in various agricultural communities to provide data for assessing potential health risks. However they only collect the gas phase fraction (AMN, 2012).

To perform an inhalation risk assessment to airborne pesticides, it is necessary to know the total concentrations of the active substances (gas + particulate phases). In this paper we present the temporal variations (2006–2013) of pesticide levels in the atmosphere of a rural site in the Centre Region (France). Using these concentrations we have estimated the human inhalation exposure and the risk assessment of the general population living near the sampling site.

2. Experimental

2.1. Pesticide selection

In 2011, around 60,000 t of pesticides were sold in France, which is the first consumer country in the European Union (Ministere de l'Écologie, du Developpement durable et de l'énergie, 2014). The farming activities are intensive in the use of pesticides in the Centre Region (France), consuming a total of 4179 tons of active ingredients in 2001 (5.5% of the national consumption) (FREDEC, 2004). Fifty-eight pesticides were studied, of which CUPs and some banned pesticides, constituted the vast majority. A total of 17 insecticides, 23 fungicides and 18 herbicides were investigated. CUPs approved by EU regulations were applied into arable crops such as maize, barley, and wheat, and their selection was based on the amount applied in the Centre Region (France). In addition, some banned persistent pesticides, previously detected in this region, (2,4′ DDD, 4,4′ DDE, α-endosulfan and γ-HCH) were also studied, because some of them could still be present in outdoor ambient air mainly due to their persistence. Table 1 shows the usage and legal status of the measured pesticides.

2.2. Sampling and site characterisation

The present work is part of a broader study on characterisation of contemporary pesticides in the atmosphere of Centre Region (France), which is one of the country's largest regions in France. We previously carried out a monitoring study of five sites (three rurals and two urbans) during three sampling campaigns between 2006 and 2008 (Coscollà et al., 2010). The present work focused on risk assessment and the data from one of the sites (Oysonville) was used with additional data collected at this site between 2009 and 2013. The sampling station was located in a rural area in northern Centre Region, in a town called Oysonville. Oysonville (48°23′35″N, 01°56′57″W) had 501 inhabitants in 2010 and it is situated 55 km from the north of Orléans city (Eure-et-Loir, Région Centre, France). The density population was 52 inhabitants/km^2. The minimum altitude is 143 m and maximum altitude, 154 m, and the total surface is 9.63 km^2. Location map is shown in Fig. 1.

The station is placed in the outskirts of the village, in the countryside, surrounded at many arable crops. About 1 km is the distance between the sampler and the first houses of the town. The most extended crops in this area are wheat, rape and barley. The orchards crop production varies according to the season. There are some spring crops such as wheat, barley, maize and rape; and some summer crops such as beet, peas, potatoes and rape. Samples were collected at the human breathing zone (approximately 1 m from the ground level).

A total of 134 samples were collected (26 in 2006, 13 in 2007, 12 in 2008, 16 in 2009, 13 in 2010, 18 in 2011, 18 in 2012 and 18 in 2013). The sampling weeks coincide with the application period of pesticides for this region (spring and summer seasons) (see Fig. 2).

A low-volume sampler (Partisol 2000) from Thermo Electron Corporation (East Greenbush, NY, USA) sampling both gaseous and particulate (Total Suspended Particles, TSP) phases was employed. Samples were collected using quartz fiber filters (QFFs, 47 mm diameter) (particle phase) and PUF cylinder (polyurethane foam, 26 mm diameter × 76 mm length with 22 mg·cm^{-3}) (gas phase). The samples (QFFs and PUF) ran for 168 h at a flow rate of 1 m^3 h^{-1}. The total volume collected was approximately 168 m^3.

2.3. Sample analysis

QFFs and PUFs samples were extracted together in stainless-steel cells in the same Accelerated Solvent Extraction (ASE) 300 PLE system