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Identification of the main pesticide residue mixtures to which the French population is exposed

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ABSTRACT

Owing to the intensive use of pesticides and their potential persistence in the environment, various pesticide residues can be found in the diet. Consumers are therefore exposed to complex pesticide mixtures which may have combined adverse effects on human health. By modelling food exposure to multiple pesticides, this paper aims to determine the main mixtures to which the general population is exposed in France. Dietary exposure of 3337 individuals from the INCA2 French national consumption survey was assessed for 79 pesticide residues, based on results of the 2006 French food monitoring programmes. Individuals were divided into groups with similar patterns of co-exposure using the clustering ability of a Bayesian nonparametric model. In the 5 groups of individuals with the highest exposure, mixtures are formed by pairs of pesticides with correlations above 0.7. Seven mixtures of 2–6 pesticides each were characterised. We identified the commodities that contributed the most to exposure. Pesticide mixtures can either be components of a single plant protection product applied together on the same crop or be from separate products that are consumed together during a meal. Of the 25 pesticides forming the mixtures, two—DDT and Dieldrin—are known persistent organic pollutants. The approach developed is generic and can be applied to all types of substances found in the diet in order to characterise the mixtures that should be studied first because of their adverse effects on health.

1. Introduction

The use of pesticides, which has become essential over the last few decades owing to intensive farming, results in the widespread presence of residues in water, soil and foodstuffs. In its last report, the European Food Safety Authority acknowledged that pesticide residues were detected in 46.7% of the 67,887 food samples analysed throughout the European Union in 2008. Residues of at least two pesticides were found in 26.7% of the analysed samples, one third of which contained more than 4 pesticide residues (EFSA, 2010a). Consumers are thus exposed to complex mixtures of pesticides which are a suspected risk to human health (EFSA, 2008; WHO, 2009). Currently, there is an international consensus on the need to consider chemical mixtures when characterising the risk of human co-exposures (ACROPOLIS, 2010–2013; European Commission, 2011; Meek et al., 2011). The risk assessments conducted worldwide focus on chemicals belonging to the same chemical family (carbamates, organophosphorus pesticides, triazoles) and/or sharing the same mechanisms of action (ACROPOLIS, 2010–2013; Boobis et al., 2008; Bosgra et al., 2009; EFSA, 2009; EPA, 2002, 2007; Müller et al., 2009; Reffstrup et al., 2010). They are based on cumulative exposure and on the cumulative effects of these substances, assuming that interactions between substances are unlikely to have a significant effect on health. However, interactions between pesticides cannot be ruled out (EFSA, 2012). Moreover such studies are of limited interest for health risk assessment, as they may not reflect real mixtures of pesticides likely to occur in actual diets. Indeed, individuals are probably exposed to pesticides from different chemical families.

The PERICLES research programme coordinated by the French Agency for Food, Environmental and Occupational Health & Safety, aims firstly to identify pesticide mixtures to which the French population is exposed through diet and secondly to investigate the possible combined effects of their components on human cells. Within this programme, an original Bayesian nonparametric model was developed to define the combination of pesticides. The main advantage of the Bayesian nonparametric approach is that no assumption is made on the form of the distribution of co-exposure to pesticides while it is based on a random mixture of parametric distributions. The number of components of the mixture is automatically inferred from the data in contrast with the classical approaches that require the specification of the

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number of mixture components. This eventually allows representing any kind of complex distributions, possibly multimodal. Pesticides forming mixtures are selected based on their probability of occurring simultaneously, considering both the pesticide residue patterns in food and the population's dietary habits. In Crépet and Tressou (2011), the mathematical description of the model was specified and applied to co-exposure of the French adult and child population to 79 pesticides. For the adult population, this study resulted in the selection of 20 mixtures composed of 3–17 pesticides. In this first application, the non-detected values of pesticide residues in food were assumed to be uniformly distributed between 0 and the limit of reporting, which corresponded to the analytical limit of detection or quantification. However this major assumption was unrealistic when pesticides were not used on crops. New calculations were then conducted, assuming an absence of residues in food samples with analytical values below the limit of reporting. This paper describes the mixtures obtained on the basis of this assumption, the proportions of each pesticide composing the mixtures and the foodstuff which contributed most to exposure to the different mixtures.

2. Material and methods

2.1. Pesticide selection and residual concentration

The data on pesticide residues in food and drinking water are based on annual control and monitoring programmes implemented in 2006 by the French ministries in charge of consumer affairs, agriculture and health. These programmes provide analytical results for up to 300 different pesticide residues screened for in about 150 types of raw agricultural commodities, water included. Sampling of monitoring programmes was carried out in order to collect commodities representing those sold on the French market. Results from control programmes are not representative but were included because for most commodities the levels do not differ from those of the monitoring programmes (ANSES, 2009a, 2010a). The year 2006 was chosen because it corresponds to that of the food consumption survey. Residues of 79 pesticides, for which at least 10% of analytical results were quantified, were retained for the study. They were analysed in 120 raw agricultural commodities, representing 306,899 analytical results and 8364 food/residue combinations. For each combination, the observed residue levels were modelled using a histogram distribution. Each histogram interval was constructed using two consecutive observed residue levels, and the probability of falling within the interval was estimated using the proportion of corresponding observed values. A null residue level was attributed to analytical results reported as “below the limit of reporting” as recommended by the European Food Safety Authority (EFSA, 2010b) when the proportion of censored data exceeded 80%.

2.2. Individual food consumption data

Consumption data was extracted from the second “Individual and National Study on Food Consumption”, INCA2 survey, carried out by the French Food Safety Agency (ANSES, 2009b; Dubuisson et al., 2010; Lioret et al., 2010). This survey was conducted between 2005 and 2007 and took into account seasonal variations. Two independent population groups were included in the study: 2624 adults aged 18–79 and 1455 children aged 3–17. Participants were selected using a three-stage random probability design stratified by region of residence, size of urban area and population category (adults or children). Each participant was asked to complete an anonymous seven-day food diary. Foods were subsequently categorised into 1280 “as consumed” food items (INCA2 classification). To match the consumption data with the pesticide residue levels in food, the “as consumed” foods were broken down into 181 raw agricultural commodities. To do this, 763 standardized recipes defined by the French Food Safety Agency that take into account industrial processes, home cooking habits and edible portions for the INCA2 survey, were used (ANSES, 2010b). Based on the sampling weights provided for each individual which represented their frequency in the entire French population, two samples of 1898 adults and 1439 children were built from the original samples, by carrying out random trials with replacement. Only individuals whose energy needs are covered by the declared consumption were included. The time window for short-term exposure calculation was a 24-h day. To avoid autocorrelation problems, 1 day was randomly selected for each individual from the records of 7 consecutive days.

2.3. Dietary co-exposure assessment

For each individual surveyed in INCA2, the consumed quantities of commodities containing a given pesticide were multiplied by the associated residue levels and cumulated across commodities to obtain the total daily exposure to the pesticide. Exposure was expressed in micrograms of the chemical per kilogram of consumer body weight for the selected day (μg/kg BW/d). A series of 100 possible daily exposure values to the 79 pesticides was calculated for each individual by randomly selecting residue levels from each commodity/residue histogram. The above series of exposures represented the total uncertainty of the exposure of an individual during a given day. One hundred values are a sufficient number to accurately estimate the exposure distribution as was tested in Crépet and Tressou (2011). To make comparisons between exposure levels possible, exposures were log-transformed, centred around the mean and rescaled by the standard deviation for each pesticide and across individuals. Finally, the 95th percentile of exposure to each pesticide for each individual was retained to form the final data set.

2.4. Individual co-exposure clustering

As diet is a combination of various foods consumed by individuals, the univariate distribution of the population exposure to a pesticide is multimodal. This implies that the population can be clustered into groups according to the level of exposure to a pesticide. Moreover, each individual is exposed to several pesticides each day. The clustering should be performed by considering the levels of exposure to the 79 selected pesticides. Therefore, individuals can be clustered into a certain number of groups which are homogeneous in terms of exposure profile. Using the Bayesian nonparametric approach detailed and validated in Crépet and Tressou (2011), co-exposure was modelled as a mixture of multivariate normal distributions that made it possible to account for the correlations between pesticides in each group of individuals. The mixing distribution indicating the group to which each individual was assigned was modelled with a Dirichlet process (Ferguson, 1973; Lo, 1984). Dirichlet processes are the natural extension of Dirichlet distributions used to model vectors of proportions summing to one when the length of the vector is unknown. In our case, this implies that each individual will belong to a certain group of the population in terms of its co-exposure to pesticides, the number of groups being unknown. The means and the covariance matrices of the multivariate normal distributions are drawn from a Wishart-Normal distribution as this is the classically chosen conjugate prior for the multivariate normal distribution. This is a technical choice simplifying computation that does not imply any specific assumption regarding the form of the distribution of co-exposure.

2.5. Correlations between pesticide exposures

For each group of individuals, the model produced a specific vector of mean exposures to the 79 pesticides and a matrix of covariance. Groups of individuals with higher exposure, i.e. with a majority of means exceeding the population mean, were selected to define mixtures of pesticides. The covariance matrices associated with the selected groups were studied to determine the pesticides to include in the mixtures. For each selected group of individuals, pesticides with at least one correlation above an arbitrary fixed value of 0.7 were selected. This choice is discussed in Section 4. The proportion of each residue from a mixture was calculated using the ratio of its mean exposure level and the sum of the mean exposures of all residues composing this mixture. For mixtures shared by several groups of individuals, the mean of the different proportions obtained from each group was used.

2.6. Main food contributors to the pesticide exposure mixtures

In order to determine the raw agricultural commodities that explain exposure to the mixtures, a principal component analysis was performed for each mixture while considering exposure of the general adult population. Individual exposure to a pesticide was then broken down into individual exposures by raw agricultural commodities used as supplementary variables in the analysis. The commodities most correlated with the main axes were those that most explain the population's exposure to the mixture (Fisher test with p-value < 0.05). Therefore, there was a strong relationship between exposure to the mixture and the consumption of a particular raw agricultural commodity. This was either due to the presence of a high level of a single pesticide of the mixture in this commodity, or because there were several pesticides of the mixture in this commodity. Finally, for each selected commodity mean exposures to each pesticide forming a mixture were summed to identify the commodities with the highest levels of exposure to a mixture. Please note that this sum had no biological significance but was useful to identify the contribution of a specific commodity to the total mixture exposure.