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Quantifying dietary exposure to pesticide residues using spraying journal data

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ABSTRACT

Relatively few studies are available on the combined risk of realistic dietary pesticide exposure. Despite available studies showing low risk, public concern remains. Recent methods used to estimate realistic exposure levels have a number of drawbacks, and better methods are needed. Using a novel approach, we estimated the combined exposure in the Danish population, resulting from pesticide usage in Danish agriculture. The complete Danish spraying journal data from 2014, and supervised trial residue levels reported by EFSA, were used in combination, generating residue estimates in 25 crops. Cumulative risk assessments were made for six typical Danish consumer diets. In terms of intake of cereals, sugar, fruits and vegetables, the 25 crops included accounted 70% of the diets of Danish consumers. The Hazard Index (HI) method was used to assess the consumer risk. Despite the conservative (cautious) approach, low HI values where obtained. Highest HI was 14% of the Acceptable Daily Intake (ADI) for Children. The main advantages of the new exposure estimation method are 1) comprehensive use data not relying on random samples, 2) coverage of all pesticides used, and 3) more precise estimates of residues that are below the standard reporting limits in the national monitoring program.

1. Introduction

Current regulation of pesticide residues in food focus mostly on single pesticide toxicity data, pre-approval residue measurements in harvested crops and post approval monitoring of compliance with maximum residue levels (MRLs) (Commission, 2016; EFSA, 2016). In reality however, the consumer is over the long term exposed to low amounts of hundreds of different pesticides. Hence, there is public concern that these residues might add up and jointly pose a risk for the consumer (Boobis et al., 2008). There is scientific consensus that the risk of multiple residues should be addressed by grouping pesticides into cumulative assessment groups (CAGs), grouping pesticides that share toxicological target/mode of action, has similar chemical structure, and similar mechanism of pesticidal action (CAG:s) (Boobis et al., 2008) but these groups are not yet defined in a regulatory context. There are reports in the literature on risk assessment of combined pesticide residue exposure, with varying approaches, like grouping insecticides (Boon et al., 2008; Jensen et al., 2009; Wong et al., 2014) or endocrine active pesticides (Jensen et al., 2013). There is one report in which the full dietary residue exposure was quantified, accompanied with a cumulative risk assessments grouping all pesticides together with no regard to mode of action, using the dose addition model (Jensen et al., 2015).

In quantifying the total mean exposure, previous studies all struggle with some basic methodological problems: 1) they are based on a limited number of food samples, originating from National Monitoring Programs (NMPs), or taken by researchers 2) the sampling programs usually do not analyze all pesticides possibly present in the foods 3) the quantification limits (LOQ) are usually in the 0.01–0.05 mg/kg range, and residues below this level cannot be quantified. These problems leave researchers unable to precisely define the exposure level, instead forcing them to report in terms of high and low bound exposure scenarios (Jensen et al., 2015; Nougadere et al., 2012). Thus, there is a need for alternative data

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sources that can overcome these limitations and provide more accurate and robust exposure estimates.

In Denmark, by law farmers have to record all their pesticide use in spraying journals. Since 2012, this reporting is done electronically and aggregated data is available from commercial data suppliers. The requirement concerns farmers with a cultivated area of more than 10 ha and greenhouse/horticultures, orchards, open field vegetable producers and plant nurseries having an annual turnover of 50.000 DKK (appr. 7.000 €) or more (EPA, 2017). These criteria effectively include all Danish food growers of relevance to the overall food supply. Producers below these limits are likely to be non-professional producers. Such producers are less likely to even use any pesticides at all, since usage of pesticides in Denmark requires a spraying certificate, that is granted by the authorities and requires formal training. The spraying journal has to contain information about crop, trade name of pesticide product, total amount used, number of hectares treated, and has to be reported to the authorities once per year. The authorities make around 600 farmer inspections annually where spraying journals are checked, and violations of rules are punishable by law, and can lead to heavy fines and reduced EU subsidiaries (EPA, 2017). Violations in this context means use of the products outside of the permitted use pattern (crop, dose and timing of application) as defined in the approval from the Danish Environmental Protection Agency (Danish EPA).

European pesticide regulation requires that a company seeking to register a pesticide for use in a certain crop has to perform at least 4–8 residue trials in that crop over two seasons, or one season if trials are widely enough spread climatically, in the climatic zone (EU north or EU south) where registration is sought (Commission, 2016). In such trials the pesticide has to be applied in worst case conditions with regards to residues formation in the harvested goods. That means the maximum dose rate needed to control the pest has to be used, applied at the latest crop growth stage for which registration is sought. These trials have to comply with Good Laboratory Practice (GLP), which ensures high data quality. Using validated analytical methods, the objective is to determine the residue level at harvest. Crop metabolism studies are conducted to establish the relevant residue definition. The European Food Safety Authority (EFSA) publishes the results of these residue trials as part of their review of Maximum Residue Levels (MRLs) according to article 12 of EU regulation 396/2005, or as part of evaluation of active substances according to EU regulation 1107/2009. Results are presented as so called “Supervised Trial Median Residue” (STMR), “Highest residue” (HR) resulting from use according to the so called “critical GAP” (GAP = Good Agricultural Practice), which equals use at worst case conditions with regards to residue formation. The STMR is the median residue level from the whole trial package, and the HR is the highest single value.

The purpose of this study was to develop and evaluate a novel method to more accurately estimate realistic pesticide residue exposure, by using spraying journal and residue trial data in combination. The goal was to estimate average residue levels that are appropriate for use in chronic cumulative risk assessments (Boobis et al., 2008). The potential advantages are higher accuracy in estimating residues below detection limit, and more comprehensive coverage of used pesticides. The purpose was also to estimate the cumulative risk to Danish consumers resulting from domestic agricultural pesticide use. We used six typical Danish diets (Pedersen et al., 2010) assuming 100% domestic produce consumption of 25 crops. The cumulative chronic health risk of the estimated exposure was assessed using the Hazard Index (HI) method, which is based on the dose addition model (Boobis et al., 2008; Kortenkamp et al., 2012; Reffstrup et al., 2010; Wilkinson et al., 2000).

2. Method

2.1. Selection of crops for typical diets

Six different diets were used, representing consumption pattern in the Danish population: Adult, Man, Woman, Child and Male + Female High Fruit & Vegetable (HFV) consumer (Table 1). The diets were based on consumption data reported by the National Food Institute, Technical University of Denmark (Petersen et al., 2013). The data originated from the Danish National Dietary Survey 2003–2008 (Pedersen et al., 2010). This cross-sectional survey included 2700 participants aged 4–75 years old drawn from the Danish Central Person Register. The participants were characterized as closely representative of the Danish population, and the data has been used in a previous pesticide residue exposure study (Jensen et al., 2015).

All crops that contributed at least 0.1% by weight of any of these diets were included in the analysis. (Table 2). No products of animal origin were included, since no pesticide residues have been detected in the 477 animal product samples taken in Denmark in 2013 and 2014(Fødevarestyrelsen, 2013; 2014). Aggregated monitoring data from entire EU published by EFSA does report pesticide findings also in products of animal origin (EFSA, 2016). However, a closer look shows that these findings are low levels of banned, persistent compounds like DDT, that due to their nature may be found almost ubiquitously. As the present project focused on current pesticide usage and due to the fact that spraying journals has no value in studying exposure to no-longer- in-use, persistent environmental pollutants, all products of animal origin were excluded.

The total crop area cultivated in 2014 in Denmark for the included crops where obtained from Danish Statistics web database (www.dst.dk). The consumption of sugar was estimated from the results of the national food survey 2011–2013, published by Danish Technical University (Pedersen et al., 2015). The consumption of beer was estimated from statistics published by Danish Statistics (www.dst.dk) and the Brewers Union (www.bryggerforeningen.dk). In terms of intake of cereals, sugar, fruits and vegetables, the 25 crops account for around 70% of the diets of Danish consumers.

2.2. Selection of pesticide treatments for inclusion

All pesticides were included, also the ones used at a crop growth stage where none or very little residue in the harvested crop is to be expected (e.g. early season herbicides). However, seed treatments were only included for potatoes, as information on treatment of imported seeds was lacking. Seed treatments are applied before a crop is sown and therefore give minimal or no residues in the harvested goods. Due to the very large number of different pesticide products used, a limitation was set that only products used on at least 5% of the total treated area were included. However, in practice many product uses occurring on less than 5% of the area where also included in the calculations. In total, 671 different pesticide/crop combinations and 93 different active substances

Table 1
Consumer groups from which crop consumption data was derived (Petersen et al., 2013). Detailed consumption data can be found in appendix 1.

	Adult	Male	Female	Child	Male HFV[a]	Female HFV[a]
Age	15–75	15–75	15–75	4–6	15–75	15–75
Bodyweight (kg)	75.1	83.5	68.2	21.8	84.4	69
Number	1599	721	878	106	118	258

[a] HFV = High Fruit & Vegetables, consuming higher than average amount of fruits and vegetables, e.g. vegetarians.