Pesticide residues intake of French adults under increased consumption of fresh fruits and vegetables – A theoretical study

SAIDA BARNAT¹, MICHEL BOISSET², FRANCINE CASSE¹, MICHEL CATTEAU¹, JEAN-MICHEL LÉCERF³, DANIEL VESCHAMBRE¹ and ALAIN PERIQUET³

¹Aprifel, Agence pour la recherche et l'information en fruits et légumes, Paris, France
²Administration Déleguée régionale Paris 7 Saint-Lazare, Paris, France
³Département de Nutrition, Institut Pasteur de Lille, Lille, France
⁴Centre Technique Interprofessionnel des Fruits et Légumes, Paris, France
⁵Université Paul Sabatier, Toulouse, France

The increase of fresh vegetable and fruit (FVF) intake could contribute to the prevention of obesity and several major pathologies and thus represents a major public health goal in industrial countries. Nutritional recommendations for increased consumption of FVF could lead to ingestion of unwanted amounts of pesticides. This study was undertaken to evaluate theoretical exposure of French adults to pesticide residues under increasing amounts of FVF intake. Balanced menus with 200 – 400 – 600 – 800 and 1200 g FVF/day were established. Amounts of active substances brought by every FVF vector, at the maximum residue level (MRL) were summed up to determine the intake for each active substance. Values were compared to the Acceptable Daily Intake (ADI) to point out any potential over exposure. A maximalistic approach was adopted and no reducing factor due to processing was taken into account. It was found that under minimal recommended consumption of fruit and vegetables (400 g/d), no active substance reached the ADI, but 18 vs 144 were over 10% of the ADI. Raising FVF to 600 g/d increased the number of active substances over 10% of the ADI to 24, but again in no case was the ADI exceeded. Doubling the intake to 800 g/d gives values over the ADI for 2 active substances only (cyhexatin and thiram). It was concluded that the increase in fruit and vegetables up to 800 g per day should not expose adults over the ADIs for a majority of authorized pesticides. In this regard, residues levels of ten active substances in fruit and vegetables should deserve particular attention.

Keywords: Pesticide; exposure; dietary intake; adult diet; fruit; vegetables.

Introduction

The increase of fresh vegetable and fruit (FVF) intake represents a major public health goal in industrial countries. Such a change in dietary habits could contribute to the prevention of obesity and major diseases such as diabetes, cardiovascular diseases, osteoporosis and cancer.

In France, 55% of men and 64% of women aging 45–60 years are considered as poor fruit consumers. These percentages rise up to 72% and 64% respectively for vegetables. The first objective of the health nutrition plan 2006–2008 is to lower these figures by one fourth within 4 years.

Consumers are prone to recognize the nutritional benefits of eating higher amounts of FVF but still question the safety of these foods due to the presence of residues of plant treatment products. This fear is periodically highlighted by media and organic agriculture lobbies on reproductive and cancer risks from chronic exposure to plant treatment products. It appears thus appropriate to evaluate any change in risk level that such increase of consumption pattern may elicit.

Risk evaluation and management of pesticide residues relies on internationally accepted, scientifically based concepts and methods allowing the derivation of acceptable daily intakes (ADIs) for chronic effects and, when appropriate, acute reference doses (ARDs) for short-term effects. In Europe, the Directive 91/414/CE has set stringent requirements concerning the placing on the market of plant protection products, including the exposure assessment of the consumer to legal pesticide residues.
Pesticide residues intake and diet

Accurate exposure assessment of the consumer remains a difficult task due to the great number of active substances used on fruit and vegetables and various analytical difficulties. Further, processing by the consumer (peeling, washing, cooking, etc.) drastically reduce the intake, depending both on the pesticide molecule and the process itself. Probabilistic approaches based on Monte Carlo models have been used in recent years but are strongly dependent on the knowledge of both consumption and contamination distributions. They have been used mostly to assess dietary intake of organophosphate and carbamate compounds.\(^{[2-3]}\)

For small-size children, exposure to organophosphate pesticides has been assessed by urine biomonitoring.\(^{[4-6]}\) The results of these studies suggest that ADIs for these compounds may be exceeded, but indoor use of biocides may cause overestimation of dietary intake.

The aim of this study was to assess by a deterministic approach, changes in potential exposure of French adults to pesticides when the fresh and vegetables consumption are increased and to compare potential exposure to the allowable daily intake of these compounds.

Materials and methods

Estimations of intakes

Five daily balanced menus with increased proportion of fruit and vegetables (200, 400, 600, 800, 1200 g per day) over a period of four weeks were designed. Within a set of menus (200 g per day for example) the quantities of each fruit and vegetable consumed over the past four weeks were added.

The quantities of active substances brought by every fruit and/or vegetable vector, at the maximum residues levels (MRL), were summed up and this figure was expressed as mg/kg body weight/day. The French MRLs are only from fresh, refrigerated and frozen products and were drawn from the French regulation on December 31\(^{st}\) 2004. Canned products have not been taken into consideration. Such a conventional approach grossly overestimates the intake for the following reasons:

(i) Residues were considered at the MRL, although it is accepted that using the good agricultural practice in specific crop, only 30% of the crop present levels between the limit of the analytical determination and the MRL,

(ii) No reduction factors associated with industrial or domestic processing of vegetables or fruit (washing, brushing, peeling, cooking, etc.) were taken into account,

(iii) All the active substances authorised on a specific commodity were supposed to be present simultaneously in that commodity. This is a very unlikely situation because a few of these possible substances are used in a campaign depending upon the kind of parasite, the infestation threshold and weather conditions,

(iv) All fruits and vegetables proposed in the menu were ingested.

Menu composition

Five menus with increasing FVF levels (200, 400, 600, 800, and 1200 g) per day were selected in order to conform the recommendation to increase the daily portion of these food commodities in the daily intake. These menus were established on a four-week period for each level of fruit and vegetable intake, in the dual concern of maintaining both diversity, seasonality, and balance of foods.

The structure of meals was identical. The quantities of FVF varied as did proteins (meat, fish, eggs), carbohydrates (starchy foods, bread), fats (oils, butter) in order to maintain a similar average nutritional balance. Potato, due to its of starchy food status was not initially included in the study. Some French nutritionists however, classify potato as a vegetable because it is highly consumed in parts of France with low intake of FVF and we have subsequently carried out calculations including the potato. Mean energy and macronutrient composition of menus over 4 weeks are given in Table 1.

Calculations of pesticide intake

The MRLs were obtained from peer-reviewed databases. European and French values are essentially identical but when different, the lowest figure was retained for calculations. The percentage of ADI likely to be ingested on a daily basis via each of fruit and vegetable was calculated with the simplified formula:

\[
\frac{Q \times MRL \times 10^{-4}}{ADI \times 1.68}
\]

where

\( Q = \) Factor of monthly consumption of each fruit or vegetable, expressed in grams

\( MRL = \) Maximum Residue Levels for a given fruit or vegetable, expressed in mg/kg foodstuff

\( ADI = \) Acceptable Daily Intake of a given active substance, expressed in mg/kg body weight and per day.

Table 1. Mean energy and macronutrients composition of menus.

<table>
<thead>
<tr>
<th>Fruit and vegetable level (g/day)</th>
<th>Kilocalories</th>
<th>Proteins (%)</th>
<th>Carbohydrates (%)</th>
<th>Lipids (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>2706</td>
<td>14</td>
<td>55</td>
<td>31</td>
</tr>
<tr>
<td>400</td>
<td>2688</td>
<td>13</td>
<td>55</td>
<td>32</td>
</tr>
<tr>
<td>600</td>
<td>2688</td>
<td>14</td>
<td>54</td>
<td>32</td>
</tr>
<tr>
<td>800</td>
<td>2685</td>
<td>14</td>
<td>54</td>
<td>32</td>
</tr>
<tr>
<td>1200</td>
<td>2681</td>
<td>14</td>
<td>54</td>
<td>32</td>
</tr>
</tbody>
</table>
In the calculations, the length of monthly consumption was 28 days (4 weeks). Body weight was 60 kg, European standard for toxicological risk evaluation.

When the amount of ingested active substance was lower than $10^{-6}$ mg (<1 mg), its contribution to residue intake was considered negligible (ex: acetonifen, azoxysterine and butraline on garlic in the 200 g/d menu). The benchmark of 10% of the ADI was retained for direct comparison purpose with results of previous French estimations using duplicate meal method or simulation methods which have shown that theoretical or estimated maximum daily intake did not exceed 10% ADI in the great majority of cases.

**Results**

**Daily intake of fruit and vegetables**

Fifty-six items were represented in our menus (17 fruits and 39 vegetables, excluding potato). Intake of each fruit and vegetable over the four-week period for each type of ration (200, 400, 600, 800 and 1200 g) and derived daily values are presented in Table 2.

**Active substances used on fruit and vegetables**

At least 200 active substances are authorized for fruit and vegetables in France, but only 10% of them are widely used, according to the authorities. Cross-checking of the active substances used on fruit and vegetables and the nature of foodstuffs composed the menus showed that no less than 162 substances are likely to be used in different treatment schedules. Nine substances exhibit more than 20 different uses such as chlorothalonil and azoxysterin (22 uses), pyrimicarbis (24 uses), copper (25 uses), fluazinof-p-buty (31 uses), iprodion (31 uses), mancozeb (32 uses), deltamethrin (33 uses) and lambda-cyhalothrin (38 uses). The number of authorized substances of potential use on each fruit and vegetable included in the menus is given in Table 3.

**Estimates of intake of active substances via fruit and vegetables**

As expected, the amount of active substances ingested increased when the ration of fruit and vegetables increased in the menu from 200 g/day to 1200 g/day and mean percent of the ADI increased almost linearly with the weight of fruit and vegetables in the menus (Table 4). The intake ranged from $10^{-6}$ to $10^{-3}$ mg for the 200 g/day menu of fruit and reached $10^{-2}$ mg in a few cases in the 1200 g/day menu (e.g. 0.0189 mg for maleic hydrazide on carrot). In this study, elemental sulphur was a noticeable exception with daily ingested amounts in the range $10^{-3}$–$10^{-2}$ mg whatever the menu.

In the low fruit and vegetable menu (200 g/d), 10 active substances among 162 were found to possibly exceed 10%
of the ADI, considered as a benchmark level by various risk evaluation committees (Fig. 1). Six active substances (Iprodione, mancozebe, flusilazole, ethoxyquin, lambda-cyhalothrin, and phosalone) were evaluated at levels from 10 to 15% ADI whereas four active substances—carbaryl, dicofof, thiram and cyhexatin reached or exceeded 20% ADI.

Eighteen active substances were shown to potentially exceed 10% of the ADI in the 400 g/day fruit and vegetable diet. In addition to substances already identified in the 200 g/day, 8 new pesticides were over the 10% benchmark level: carbendazim (10.7%), chlorothalonil (15.7%), chlorpyrifos-ethyl (16.2%), dithianon (11.4%), fluazinam (14.3%), procyonidone (16%), phosmet (20%) and propinebe (13.6%).

Among 24 substances identified in the 600 g/d fruits and vegetable diet, 6 additional compounds were found to be present over 10% of the ADI: carbofuran (16.1%), cyfluthrin (11.5%), flonicamidone (10.9%), propargite (16.1%), pyrimicarb (11.4%) and tebufenpyrad (10.2%).

Moreover, intake of carbaryl, dicofof and thiram was over 50% ADI.

When the daily amount of fruit and vegetables in the menu was raised to 600 g/two substances (cyhexatin, and thiram) potentially exceeded the ADI and dicofof reached 52% of the ADI. Carbaryl, ethoxyquin and flusilazole accounted for 87%, 75% and 67% of the ADI, respectively. Another group of 8 active substances were the 10% benchmark level (alphamethrin, beta-cyfluthrin, captane, endosulfan, flufenoxuron, formethane, iprovalicarbe, and myelobutani).

In the menu with the highest proportion of fruit and vegetables (1200 g/day), 47 substances were present at levels greater than 10% benchmark level. Further, carbaryl, cyhexatin, ethoxyquin, flusilazole and thiram were substantially over the ADI (133%, 184%, 116%, 105% and 160%, respectively). Ten substances may reach or exceed ADI when fruit and vegetable ration is greater than 800 g/d carbaryl, cyhexatin, dicofof, ethoxyquin, flusilazole, iprodi- one, lambda-cyhalothrin, mancozebe phosalone and thiram. These substances should deserve greater attention in view of potential health impact in view of increased FFV consumption. Among these, carbaryl, flusilazole and iprodi- one are classified as suspected carcinogens (Carc. Cat.

### Table 4. Mean percent of acceptable daily intake (ADI) in the various menus.

<table>
<thead>
<tr>
<th>Fruit and Vegetable (g/d)</th>
<th>Mean% ADI a</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>2.9</td>
</tr>
<tr>
<td>400</td>
<td>5.9</td>
</tr>
<tr>
<td>600</td>
<td>8.7</td>
</tr>
<tr>
<td>800</td>
<td>11.5</td>
</tr>
<tr>
<td>1200</td>
<td>18.1</td>
</tr>
</tbody>
</table>

*mean percent of ADI for 162 active substances.

### Table 3. Number of authorized active substances on fruits and vegetables included in the menus.

<table>
<thead>
<tr>
<th>Number of authorized substance</th>
<th>Fruits and vegetables in the menus</th>
</tr>
</thead>
<tbody>
<tr>
<td>0–10</td>
<td>10 Avocado, beet, dill, fennel, fig, kiwi fruit, mushroom, parsley, spinach, turnip</td>
</tr>
<tr>
<td>10–15</td>
<td>13 Artichoke, basil, carrot, celery, celeriac, chicory, corn salad, leek, lemon, mint, tarragon, watercress</td>
</tr>
<tr>
<td>15–20</td>
<td>20 Garlic, annas, eggplant, broccoli, cabbage, Brussels sprout, cauliflower, red cabbage, green cabbage, chive, cucumber, zucchini, shallot, green bean, lettuce, let, batavia lettuce, orange, pepperoni, pomelo, pumpkin</td>
</tr>
<tr>
<td>20–30</td>
<td>6 Banana, quince, onion, watermelon, pear, plum</td>
</tr>
<tr>
<td>&gt;30</td>
<td>6 Strawberry, peach, nectarine, tomato, apple, table grapes</td>
</tr>
</tbody>
</table>

Fig. 1. Distribution of pesticide estimated levels compared with acceptable daily intake ADI in diets with 200, 400, 600, 800, 1200 g of fruit and vegetable by day.
Table 5. Estimated intake of pesticides compared to acceptable daily intake (ADI) for all active substances in the menus with and without potato.

<table>
<thead>
<tr>
<th>Fruit and Vegetable (g/d)</th>
<th>Estimated Daily Intake (% ADI)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>With potato *</td>
</tr>
<tr>
<td>200</td>
<td>3.5</td>
</tr>
<tr>
<td>400</td>
<td>6.5</td>
</tr>
<tr>
<td>600</td>
<td>9.0</td>
</tr>
<tr>
<td>800</td>
<td>11.7</td>
</tr>
<tr>
<td>1200</td>
<td>18.1</td>
</tr>
</tbody>
</table>

*not different from the menus without potato at p = 0.05 (Mann-Whitney Wilcoxon test).

3) under Directive 67/548 CE. Moreover, fluzilazole, like other triazole fungicides, exhibit teratogenic properties in rats in vitro and is classified as a possible developmental toxin in man (Repr. Cat. 2) according to European Union.

Incidence of the incorporation of potato in the menus

Some 35 substances are authorized for the potato, which is one of the most vulnerable foods along with strawberry, nectarine, peach, and tomato and apple (Table 3). Carbaryl, linuron, diquat, ethoprophos, glufosinate, lambda-cyhalothrin and fluufenacet estimated intake was clearly enhanced when potato was incorporated into the diet at 200 g/d and 400 g/d of fruit and vegetable a day. A similar trend, although of smaller magnitude, was also observed for 17 other substances (fluazifop-p-butyl, copper, flutolanil, metribuzine, maleic hydrazide, quialfop-ethyl, cymoxanil, carbosulfan, rimsulfuron, imazalil, deltamethrine, manebe, zoxamide, thiabendazole, fenithrothion, penicycuron and aclonifen. As shown in Table 5, estimated daily intake of pesticide was not significantly altered when potato were included in the diet.

Discussion

This study is the first attempt to evaluate the consequences of implementing nutritional recommendations for enhanced fruit and vegetable consumption on pesticide residues ingestion. Estimation of ingested amounts of pesticides remains a difficult task partly due to geographical and seasonal variations. Normal dietary variation of the French population remains rather poorly characterized. One of the few studies conducted in France included 64 fruit and vegetable items (23 fruits and 40 vegetables) whereas we have introduced fifty-six food commodities in the different menus (17 fruits and 39 vegetables including potato). The variety of fruit and vegetables is thus similar in both studies. The number of fruit and vegetables varied from 2 portions in the 200 g/day menu to 6–8 portions in the 1200 g/day menu. Previous French studies have shown that mean intake of food and vegetable was between 300 and 400 g/day, including potato.

The MRLs vary largely between active substances and between the crops for which it is authorized. For example, MRL values range from 0.005 mg/kg cyhexatin when used on peach and nectarine to 0.05 mg/kg on tomato and 0.3 mg/kg on plum. Such variations can be explained by the fact that MRLs should be set as high as possible to eradicate the target parasite or maintain an acceptable threshold of infestation on the one hand, and kept as low as possible to protect consumer health on the other hand. Sound estimation of pesticide intake must take into account properly both the variety and the amount of fruit and vegetables consumed. It must be kept in mind that we intently considered that all the authorized substances could be present simultaneously in the proposed menus which represents an unrealistic situation. Six commodities could bear 20–30 active substances, and five commodities more than 30 active substances of which tomatoes (34), potatoes (35), strawberries, nectarines, peaches (37), apples (54) and table grapes (73). Our results highlight the most vulnerable—and consequently the most protected—commodities.

Several hypotheses of this theoretical study lead to a marked overestimation of pesticide intake. Nevertheless, calculated daily intakes for a great majority of substances would be kept within 10% of the ADI if the daily ration of fruit and vegetables is raised to 600, and 800 g. For the 1200 g fruit and vegetable menu 115 substances from 162 would remain below this benchmark value and 142 would be lower than 30% of the ADI. Although 10% and 30% benchmark values are devoid of toxicological significance they are widely accepted in risk evaluation of dietary contaminants. Our results clearly indicate that for the great majority of active substances ADIs are not likely to be reached or exceeded even when dietary intake of fresh vegetable and fruits are substantially increased. This conclusion is strengthened by the fact that a dramatic decrease in concentration for many active substances is afforded by simple washing and peeling of fruit and vegetables. In this view, Rawn et al. have observed a decrease from 74.5% to 97.9% in organophosphate insecticides (azinphos methyl, phosalone and phosmet) in Canadian apples.

In a duplicate diet study of meals collected in mass eating establishments, Leblanc et al. have shown that estimated daily intakes for ten substances ranged from 0.2% (procymidine) to 4% (triazophos) of the ADI. For these compounds estimated intakes were respectively 23.7 and 37.5 times lower than the theoretical maximum intakes for French adults calculated by the “Observatoire des Consommations Alimentaires” with a deterministic method. For Italian adults, analysis of ready meals showed that intake accounted for 2.6% of ADI on average. The 20th Australian Total Diet Survey carried out in Australia indicated that the intake of 8 substances solely including
carbaryl, and iprodione was above 0.2% of the ADI for young adults.

Conclusion

This theoretical study shows that an increase in fruit and vegetables up to 800 g per day should not lead to exposure of adults to a majority of authorized pesticides over the ADIs. In this regard, carbaryl, cyhexatin, dicofol, ethoxyquin, flusilazole, iprodione, lambda-cyhalothrin, mancozebe phosalone and thiram residue levels in fruit and vegetables should deserve particular attention. However, several hypotheses retained for the evaluation of pesticide intake produce a gross overestimation of actual exposure. Determination of these target compounds in the various menus should substantiate the conclusion that consuming 5 to 10 different fruit and vegetables does not lead to undue pesticide exposure. This study is now underway by our group.

Acknowledgments

The work was partially supported by a research grant from Aprifel, a French non-profit organization of fresh food and vegetables producers. The authors wish to thank Mrs A Deniaud and Mrs L Bouherbachene for their expert assistance in the preparation of this manuscript.

References


