Essais d'évaluation des résidus de produits phytosanitaires

dans les fruits et légumes

Professeur Alain Périquet – Université Paul Sabatier – Toulouse 3

D'après :

Etude Théorique 2005- 2006

&

Etude pratique 2007-2009

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

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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 exceeded. Doubling the intake to 800 g/d increased the number of active substances over 10 % of the ADI to 24, but again in no case was the ADI exceeded. Thus, 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[1] 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 (ARfDs) 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.

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Realistic approach of pesticide residues and French consumer exposure within fruit & vegetable intake

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The increase of fruit and vegetable (F&V) intake contributes to the prevention of chronic diseases, but could also significantly increase pesticide exposure and may thus be of health concern. Following a previous pesticide exposure assessment study, the present study was carried out to determine actual levels of pesticides within 400 g of F&V intake and to evaluate consumer risk. Forty-three Active Substances (AS) exceeding 10 % of the Acceptable Daily Intake (ADI) in balanced menus established for our previous theoretical study were considered. Fifty-six pooled food samples were analyzed: 28 fruit samples and 28 vegetable samples. Pesticide values were compared to Maximum Residue Levels (MRL) and to the "toxicological credit" derived from ADI. It was observed that 23 out of the 43 retained AS were never detected, 5 were detected both in F&V samples, 12 only in fruits and 3 only in vegetables. The most frequently detected AS were carbendazim, iprodione and dithiocarbamates. When detected, AS were more frequently found in fruit samples (74 %) than in vegetable samples (26 %). A maximum of 3 AS were detected at once in a given sample. Overall, we observed 8 and 14 overruns of the MRL in 1204 measures in pooled vegetable and fruit samples, respectively (0.7 % and 1.2 % of cases, respectively). Chronic exposure for adults was the highest for dithiocarbamates but did not exceed 23.7 % of the ADI in F&V. It was concluded that raising both F&V consumption up to 400 g/day (~5 F&V/day) according to recommendations of the national health and nutrition plan, does not induce pesticide overexposure and should not represent a risk for the consumer.

Keywords: Pesticide residue; exposure; fruit; vegetable; maximum residue level (MRL); acceptable daily intake (ADI)

Introduction

Increasing dietary intake of fresh Fruit and Vegetables (F&V) may significantly contribute to the prevention of major chronic diseases, such as diabetes,[1,2] cardiovascular diseases[3,4] osteoporosis[5,6] and cancer.[7,8] Nevertheless, on one hand the health benefits of F&V are recognized, on the other hand they represent a source of exposure to pesticides for the consumers. Such a change in increasing F&V intakes could induce a rise in pesticide exposure and thus may be of health concern.

A recent report from the European Commission Directorate General for Health and Consumers (DGSANCO) including national data sets showed that, in 8 % of unprocessed F&V sampled in France, Active Substances (AS) levels exceeded Maximum Residue Levels (MRL).[9] This report however, did not point out any chronic risk for the consumer.

In a previous theoretical study using deterministic approach (exposure assessment with AS levels set at MRL), we have shown that exposure in French adults consuming up to 800 g F&V per day should not lead to expose adults consumers above the Acceptable Daily Intake (ADI) for all the AS studied (except for cyhexatin and thiram).[10] Residue levels of 10 AS should deserve particular attention because they exceeded 10 % of the ADI in menus containing only 200 g of F&V: carbaryl, cyhexatin, dicofol, ethoxyquine, flusilazole, iprodione, lambda-cyhalothrin, mancozeb, phosalone and thiram.

Our previous theoretical study lead to gross overestimation of actual exposure of the consumer for two reasons: AS levels were set at MRL levels and we did not take into account processing and culinary practices. These treatments can cause losses of AS depending on their physicochemical properties and the nature of treatment. In a meta-analysis, Keikotlhaile et al.[11] showed that processes which

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Pourquoi ces études?

- **Augmenter sa consommation** quotidienne de fruits et légumes (F&L) est une recommandation de Santé Publique

- Dans le contexte actuel, les membres du Comité Sécurité Alimentaire ont estimé qu'il convenait d'y **associer une démarche sécuritaire**

- **Afin d'évaluer le niveau de risques** représentés par les résidus de pesticides dans le cadre d'une consommation croissante de F&L

Etude1

les fruits et légumes frais
Aprifel

Etude Théorique 2005-2006

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

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Principes de l'étude théorique

- **ETAPE 1 : Élaboration de menus quotidiens équilibrés dont la proportion de F&L allait croissant :**
  (200 – 400 – 600 – 800g par jour) et répartis sur 4 semaines

- **ETAPE 2 : Addition à l'intérieur d'une même catégorie de grammage** (200 g par jour par exemple) **des quantités de chacun des F&L consommés** pendant les 4 semaines et ramenées à la consommation journalière.

- **ETAPE 3 :** Calcul des quantités maximales de résidus de **chaque produit phytosanitaire** pouvant se retrouver **sur chacun des F&L** afin de connaître la quantité de chaque matière active ingérée quotidiennement.

- **ETAPE 4 : Comparaison de cette valeur à la DJA** qui représente la quantité de matière active qu'un individu peut ingérer chaque jour, pendant toute sa vie, sans qu'il en résulte d'inconvénient pour sa santé
  → **afin de connaître la part du Crédit Toxicologique utilisée.**

Cette démarche se situe dans une hypothèse maximaliste car nous avons considéré que

- **les niveaux de résidus sont équivalents aux LMRs**
  alors qu'il est classiquement admis que les bonnes pratiques agricoles aboutissent à un niveau résiduel qui se situe autour de 30% de la LMR.

- **toutes** les substances actives les plus utilisées **sur chacune des denrées** devaient être **prises en compte,** alors qu'il est très peu vraisemblable que, au cours d'une même campagne, toutes ces molécules soient mises en œuvre.

- **les facteurs de réduction** inhérents aux procédés industriels ou ménagers (lavage, brossage, épluchage, cuisson, etc.) **ne devaient pas être pris en compte**

- **l'intégralité des menus** proposés était **totalement consommée**

Résultats

Les substances actives les plus utilisées sur les F&L

183 substances ont été répertoriées

165 ont été prises en compte

23 ont été identifiées comme susceptibles d'être très souvent utilisées

Pourcentages moyens des DJA représentés par les quantités de substances actives ingérées par les vecteurs F&L

Quantité de F&L par jour (g)	Toutes les substances actives (%) n=165
200	2,85
400	5,81
600	8,54
800	11,26

En comparant ces résultats à ceux de l'enquête DGAL, il apparaît que :

- **les conditions maximalistes des 2 études sont similaires**
- **Le facteur de consommation était établi à partir des déclarations d'achats des ménages, alors que nous l'avons calculé à partir de menus**
- **le nombre de substances actives** dépassant **10% de la DJA est identique (21 versus 22 molécules)** pour le régime à 400 g de fruits et légumes par jour,
- **Dépassement de la DJA (enquête DGAL) Vs. aucun dépassement de DJA**

Etude2

Etude pratique 2008-2009

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Pourquoi l'étude analytique?

Vérifier les résultats de l'étude théorique

Dosage des substances actives

Prendre en compte les habitudes alimentaire et d'achats

Intégrer les facteurs de réduction inhérents aux procédés ménagers : « aliments tels que consommés »

Méthodologie (1) : Menus

- Constitués dans un double souci de diversité et d'équilibre alimentaire
- Validés par le service Nutrition de l'Institut Pasteur de Lille
- Quantité de F&L : 2 fois celle recommandée (soit 800g)
- Protocole d'échantillonnage :
  - fruits d'une part / légumes d'autre part,
  - 2 échantillons/j pendant 4 semaines
  - soit 2 x 7 jours x 4 semaines = 56 échantillons

Méthodologie (2) : 43 SA recherchées
(d'après l'étude théorique)

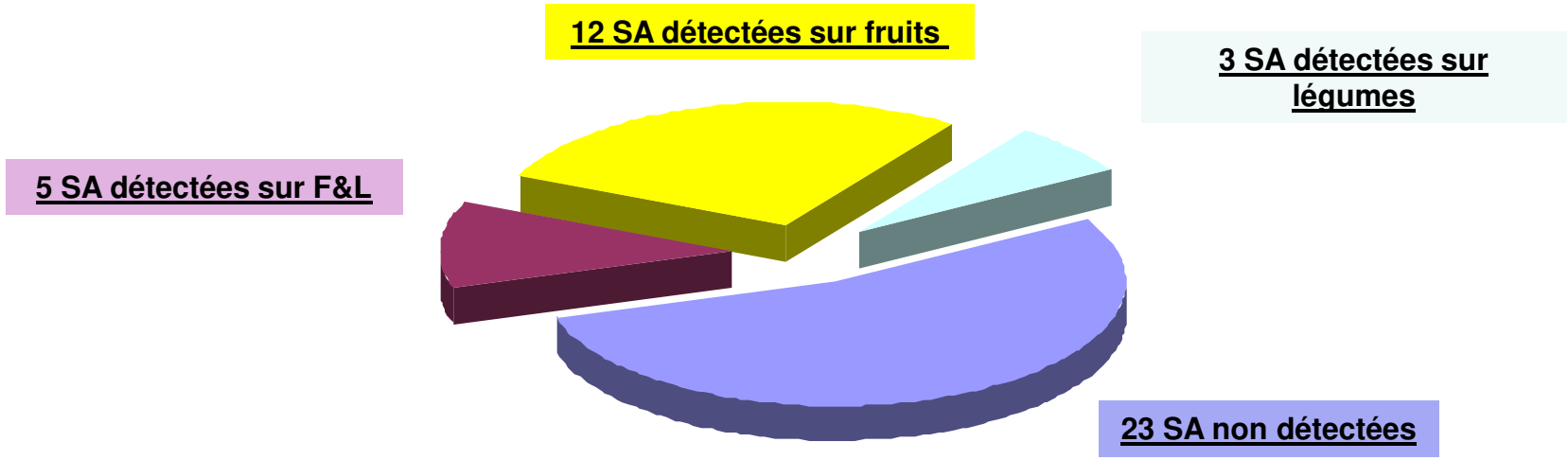
Insecticides/Acaricides	Fongicides	Herbicides
Carbaryl	Captane	Fluazifop-p-butyl
Carbofuran	Carbendazime	Flufénoxuron
Chlorfenvinphos	Chlorothalonil	Linuron
Chlorpyriphos-Ethyl	Difénoconazole	
Cyfluthrine	Diphénylamine	
Cyhexatin	Dithianon	
Cyperméthrine	Dithiocarbamates*	
Deltaméthrine	Ethoxyquine	
Dicofol	Famoxadone	
Endosulfan	Fluazinam	
Fénitrothion	Flusilazole	
Forméthanate	Iprodione	
Lambda-cyhalothrine	Iprovalicarbe	
Oxydéméton-méthyl	Mépanipyrim	
Phosalone	Myclobutanil	
Phosmet	Procymidone	
Propargite	Thiabendazole	
Pyrimicarbe	Thiophanate-méthyl	
Roténone	Vinchlozoline	
Tébufénozide		
Tébufenpyrade		

* *(mancozèbe, manèbe, métam sodium, thirame, zirame)*

Résultats

Détection et quantification des SA

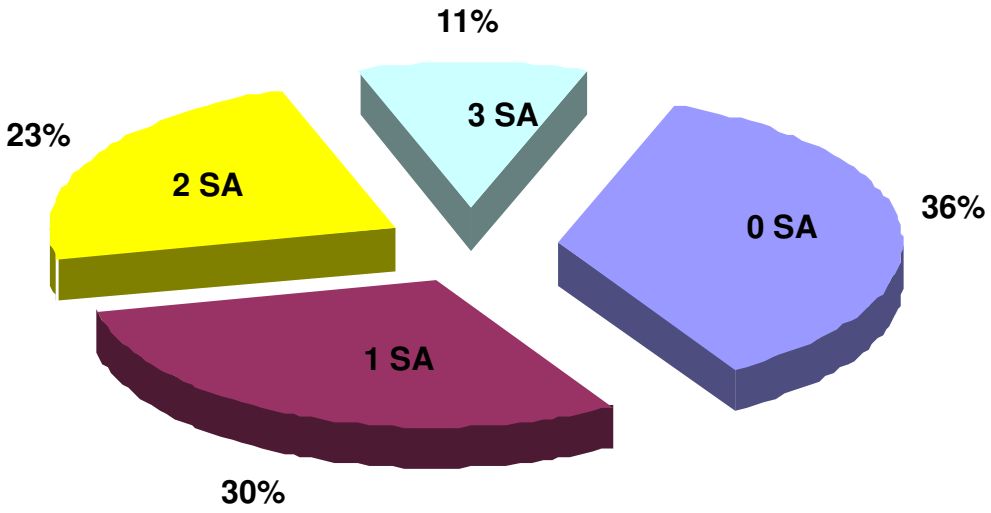
Nombre de SA détectées sur 43 SA recherchées



Sur les 43 SA recherchées, 23 ne sont pas détectées, 12 sont détectées uniquement sur les fruits et 3 uniquement sur les légumes, 5 sont détectées à la fois sur F&L

Détection et quantification des SA

Nombre de SA détectées dans un même échantillon



36% des échantillons ne contenaient aucune SA

11% des échantillons contenaient 3 SA

Aucun échantillon ne contenait plus de 3 SA détectables

Comparaison aux LMRs européennes

Echantillon de légumes

	Valeur moyenne détectée (mg/kg)	LMR Europe (mg/kg)	% de la LMR
Difénoconazole	0,012	0,05	24 %
Iprodione	0,011	0,02	53%
Carbendazime	0,010	0,1	10%
Deltaméthrine	0,01	0,05	20%
Lambda-cyhalothrine	0,014	0,02	69%
Dithiocarbamates	0,068	0,05	**136%**
Thiophanate-méthyl	0,01	0,1	10%
Chlorfenvinphos	0,011	0,02	54%

En valeur moyenne : pas de dépassement de LMR sauf pour les dithiocarbamates

Comparaison aux LMRs européennes

Echantillon de fruits

	Valeur moyenne détectée (mg/kg)	LMR Europe (mg/kg)	% de la LMR
Carbendazime	0,015	0,1	15%
Dithiocarbamates	0,044	0,05	89%
Iprodione	0,013	0,02	63%
Thiophanate-méthyl	0,01	0,1	10%
Phosalone	0,009	0,05	19%
Myclobutanil	0,01	0,02	52%
Chlorpyriphos-éthyl	0,01	0,05	20%
Lambda-cyhalothrine	0,01	0,02	50%
Carbaryl	0,016	0,05	31%
Cyperméthrine	0,01	0,05	21%
Procymidone	0,018	0,02	88%
Pyrimicarbe	0,011	0,5	2%
Thiabendazole	0,012	0,05	24%
Flufénoxuron	0,011	0,05	21%
Propargite	0,01	0,01	100%
Diphénylamine	0,01	0,05	20%
Tébufénozide	0,01	0,05	20%

Evaluation du risque consommateur

Paramètres pris en compte

- La **DJA** : caractérise le risque résidu (mg/kg poids corporel /jour)
- Le **poids corporel (PC)**

→ **Crédit Toxicologique (mg/j) = DJA x PC**

- La **quantité de F&L consommée** : 400g de F&L/j (200g F + 200g L)
- La **quantité de résidus (R)** mesurée dans les F&L (mg/kg)

→ **Exposition (mg/kg/j) = R x 200/1000**

L'évaluation du risque chronique consiste à comparer le crédit toxicologique à l'exposition

→ **% du Crédit Toxicologique = Exposition / Crédit Toxico x100**

Evaluation du risque consommateur

Echantillons de légumes

	Valeurs moyennes détectées (R) (mg/kg)	DJA (mg SA /kg PC/j)	Crédit toxicologique (adulte) (mg/j)	Exposition (200g) (mg/kg/j)	Pourcentage du crédit toxicologique
Difénoconazole	0,012	0,01	0,58	0,002	0,42%
Iprodione	0,011	0,06	3,45	0,002	0,06%
Carbendazime	0,01	0,02	1,15	0,002	0,18%
Deltaméthrine	0,01	0,01	0,58	0,002	0,35%
Lambda-cyhalothrine	0,01	0,005	0,29	0,002	0,71%
Dithiocarbamates	0,068	0,001	0,058	0,01	23,73%
Thiophanate-méthyl	0,01	0,08	4,60	0,002	0,05%
Chlorfenvinphos	0,011	0,0005	0,029	0,0021	7,45%

Aucun dépassement de la DJA pour les légumes

Le % du crédit toxicologique le + élevé est de 24% pour les dithiocarbamates

Evaluation du risque consommateur

Echantillons de fruits

	Valeurs moyennes détectées (R) (mg/kg)	DJA (mg SA /kg PC/j)	Crédit toxicologique (adulte) (mg/j)	Exposition (200g) (mg/kg/j)	Pourcentage du crédit toxicologique
Carbendazime	0,009	0,02	1,15	0,0017	0,15%
Dithiocarbamates	0,044	0,001	0,06	0,0089	15,47%
Iprodione	0,025	0,06	3,45	0,0025	0,07%
Thiophanate-méthyl	0,01	0,08	4,60	0,0019	0,04%
Phosalone	0,009	0,01	0,58	0,0019	0,33%
Myclobutanil	0,01	0,02	1,15	0,0021	0,18%
Chorpyriphos-éthyl	0,01	0,01	0,58	0,002	0,35%
Lambda-cyhalothrine	0,01	0,005	0,29	0,002	0,70%
Carbaryl	0,016	0,008	0,46	0,0031	0,68%
Cyperméthrine	0,01	0,05	2,88	0,0021	0,07%
Procymidone	0,018	0,025	1,44	0,0035	0,24%
Pyrimicarbe	0,011	0,035	2,01	0,0022	0,11%
Thiabendazole	0,012	0,1	5,75	0,0024	0,04%
Flufénoxuron	0,011	0,0035	0,20	0,0021	1,06%
Propargite	0,01	0,007	0,40	0,002	0,50%
Diphénylamine	0,01	0,02	1,15	0,002	0,17%
Tébufénozide	0,01	0,02	1,15	0,002	0,17%

Aucun dépassement de la DJA pour les fruits

Le % du crédit toxicologique le + élevé est de 15% pour les dithiocarbamates

Conclusion

- **Pour les légumes :**
  - **8 SA détectées** sur les 43 SA recherchées
  - 16 détections de SA sur 1204 dosages, soit **1,3% de détections**
  - En valeur moyenne, seuls les **dithiocarbamates** dépassent la LMR
  - **Pas de dépassement de DJA**

- **Pour les fruits :**
  - **17 SA détectées** sur les 43 SA recherchées
  - 45 détections de SA sur 1204 dosages, soit **3,7% de détections**
  - En valeur moyenne, seule la **propargite** atteint la LMR
  - **Pas de dépassement de DJA**

- Dans un même échantillon ont été retrouvées au **maximum 3 SA**

- Les dithiocarbamates méritent plus grande investigation (technique de dosage particulière- source de biais)

Constat- Recommandations

- **Sécurité du consommateur bien assurée : aucun dépassement de DJA**

- **Respecter les Bonnes Pratiques Agricoles**

- **SA à surveiller** :

Dithiocarbamates	Thiabendazole	Chlorfenvinphos
Iprodione	Carbendazime	Carbaryl
Lambda-cyhalothrine	Myclobutanil	Procymidone
Difénoconazole	Flufénoxuron	
Propargite		

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