#### Dietary intake of pesticide residues in Italy: results of 2005 - 2008 study.

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Abstract. This study, promoted by the Environmental Agencies' Pesticides Working Group (abbreviated to gruppo AAAF in Italian), seeks to assess the quantity of pesticide residues in pre-prepared (ready-to-eat) meals (comprising a first course, side dish, fruit, bread and wine), to quantify the amounts consumed and allow comparison with Acceptable Daily Intake (ADI) and Acute Reference Dose (ARfD). The data provided by 17 laboratories showed an average number of 3.4 pesticides in each meal and a maximum of 13 pesticides. The most frequently found substances were: pirimiphos-methyl (93 times), procymidone (70), cyprodinil (42), chlorpyrifos (36), iprodione (36), metalaxyl (36) and pyrimethanil (26).The distribution of residues in each dish making up the meal was also examined, and the results showed that on average in the period 2005-2008 : 51.7 % of residues were present in fruit, 27.4 % in wine, 8.7 % in the side dish, 7.6 % in the main course and 4.5 % in bread. The daily intake of pesticide residues was calculated and compared with the ADI values. In the case of adults, in 2005-2008

the average daily intake of pesticides in relation to ADI was 3.5 %; for teenagers it was 6.2 % and for children it was 12.5 %. The quantity of pesticide residues present in the meal was calculated and compared with the ARfD values. The maximum levels observed were 88% in 2005, 43% in 2006, 17% in 2007 and 21% in 2008 for a child weighing 20 kg.

Riassunto. Lo studio progettato dal gruppo di lavoro Fitofarmaci delle Agenzie Ambientali (abbreviazione gruppo AAAF) permette: di quantificare i residui di fitofarmaci in un pranzo pronto per essere consumato (comprendente il primo, il contorno, la frutta, il pane ed il vino), di accertare la quantità assunta e di compararla con Acceptable Daily Intake (ADI) and Acute Reference Dose (ARfD). I dati forniti da 17 Laboratori evidenziano una media di 3,4 residui per pranzo ed un massimo di 13 residui. I fitofarmaci rilevati con maggior frequenza sono: pirimiphos-methyl (93 volte), procymidone (70), cyprodinil (42), chlorpyrifos (36), iprodione (36), metalaxyl (36) and pyrimethanil (26). E' stata studiata la distribuzione dei residui di fitofarmaci nelle diverse portate del pranzo ed i risultati del periodo 2005-2008, calcolati come valore medio, valutano che il 51.7 % dei residui è presente nella frutta, 27.4 % nel vino, 8.7 % nel contorno, 7.6 % nel primo piatto e 4.5 % nel pane. L'assunzione giornaliera di residui di fitofarmaci è stata confrontata con i valori di ADI. Nel caso di un adulto, l'assunzione giornaliera media comparata al valore ADI è pari al 3.5 %, per un ragazzo il 6.2 % e per un bambino il 12.5 %. La quantità di residui di fitofarmaci presenti in un

Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31 pranzo è stata confrontata con i valori di ARfD. Il valore massimo riscontrato per un bambino di 20 kg, è pari a 88 % nel 2005, 43% nel 2006, 17% nel 2007 e 21% nel 2008.

Key Words: pesticide residues; daily intake of pesticide residues, dietary exposure to pesticide residues, dietary intake of pesticide residues.

Parole chiave: residui di fitofarmaci, assunzione giornaliera di residui di fitofarmaci, esposizione con la dieta a residui di fitofarmaci, assunzione con la dieta a residui di fitofarmaci.

### **INTRODUCTION**

The Italian monitoring programmes for pesticide residues in food (Decree of the Ministry of 23 December 1992) are a useful tool for ensuring that consumers are not exposed to unacceptable pesticide residue levels and that authorised pesticides are correctly applied to food crops (application rate and pre-harvesting intervals). The results of the monitoring programme were taken into consideration to evaluate the Estimated Daily Intake (EDI) of pesticides from food, on the basis of knowledge of the diet and factors relating to the processing of the food, bearing in mind preparation (cooking, for example) and the removal of certain parts (for example the skin of bananas and oranges). Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31
In Italy (Camoni *et al.*, 1998; Camoni *et al.*, 2001; Gaidano and Fabbrini, 2000; Tasiopoulou *et al.*, 2007), U.K.( Harris and Gaston, 2004), Spain (Berrada *et al.*, 2006), Poland (Nowacka and Gnusowski, 2007) and Ghana (Darko and Akoto, 2008) numerous studies have been carried out to evaluate EDI and long-term (chronic) hazards resulting from ingestion of levels of pesticide residues in the diet.

Total Diet Studies as defined by the World Health Organization (WHO 1997), consisting of analysis of a representative `market basket' of foods, usually prepared for normal consumption have not been carried in Italy.

The results of Total Diet Studies carried out in the United States (Pennington et al., 1987,

Pennington et al., 1996), Kuwait (Saxaya et al., 1999), Japan (Maitani, 2004), Canada (Rawn et al.,

2004) and Cameroun (Gimou et al., 2008) are available.

For estimation of the dietary intake of pesticide residues in France (Leblanc *et al.*, 2000) the duplicate diet method was applied, consisting of analysis of exact duplicates of prepared meals eaten and drunk during a period varying from 1 to 7 days.

The Environmental Agencies' Pesticides Working Group (abbreviated to AAAF in Italian) devised, planned and carried out this study, which set itself the objective of estimating pesticide intake from residues present in the diet. This paper presents the results of pesticide residues in samples, collected during the period 2005-2008. Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31 Seventeen Italian laboratories were involved in sampling and analysing first courses, side dishes, fruit, bread and wine. The intake per meal was calculated taking into consideration the concentration of pesticide residues found and the quantities of the first course, side dish, fruit, bread and wine in kg, consumed during a meal. Considering that two meals per day are normally consumed, the daily intake was obtained by multiplying the intake per meal by two.

In order to compare intake (daily or for the meal) with the Acceptable Daily Intake (ADI) or Acute

Reference Dose (ARfD), we considered an adult weighing 60 kg, a 40 kg teenager and a 20 kg

child.

The values calculated for daily intake and intake from the meal per kg of body weight were compared with the ADI and ARfD levels established by the European Union (EU Pesticides database).

## Materials and methods

## Laboratories participating in the study

From 2005 to 2008 the analytical data was supplied by the 17 public Italian laboratories as listed in Table 1.

The criteria used to choose the laboratories and the analytical methods have been described in detail

previously (Lorenzin, 2007).

# Sampling

The samples were collected at different times during the year and on different days of the week.

The dates when the samples were taken are shown in Table 2.

Sample preparation, Analysis, Daily intake of pesticide residues and Ratio between the daily

intake of pesticide residues and ADI.

The procedures have been described in detail previously (Lorenzin, 2007).

# Calculation of the quantity of pesticide residues in the meal.

The laboratory determined the concentration (in mg/kg) of pesticides in the first course, side dish, fruit, bread and wine.

The quantity of pesticides (in mg) present in the first course, side dish, fruit, bread and wine was calculated by multiplying the concentration found (in mg/kg) by the weight (in kg) recorded in the

prepared sample.

The quantity found in wine was not considered in calculations for the teenagers and children, as it was assumed (or rather hoped) that wine is not consumed by these categories. The intake from one meal per kg of body weight (in mg/ kg bw) was calculated by dividing the data for the quantity found in the meal by the average weight of an adult (60 kg), teenager (40 kg) and child (20 kg).

The calculation was performed for each pesticide found in the meal.

# Calculation of the ratio between intake of pesticide residues from the meal and ARfD.

The values for the estimated intake from the meal per kg of body weight (in mg/ kg bw) for each pesticide was compared with the ARfD levels established by the European Union (EU Pesticides database).

The comparison was undertaken by dividing the values for the intake from the meal by kg of body weight, and comparing this result with the ARfD levels for each of the pesticides found.

# **Results and discussion**

#### **Pesticides in Meals**

The laboratories analysed 200 meals, determining pesticide residues in the first course, side dish, fruit, bread and wine.

Overall pesticide residues were measured in 158 meals (79%), with a maximum of 13 different pesticides and with an average number of 3,4 pesticide found for each meal.

The average number of pesticides found in the meal (Table 3) reached its highest level in 2007 and 2008, with 3.8 pesticides per meal. Figure 1 shows the average number of pesticides found in each dish and in the meal, subdivided by the years in the study. It can be noted that the average number of pesticides in the meal was lower than the total of averages for each dish. This situation arises because the same pesticide may be present in 2 or 3 dishes.

91 pesticides were detected in the meals: 2-phenylphenol, acetamiprid, azinphos methyl, azoxystrobin, benalaxil, benomyl, bifenthrin, boscalid, bromopropylate, bupirimate, buprofezin, captan, carbaryl, carbendazim, chlorfenson, chlorothalonil, chlorpropham, chlorpyrifos, chlorpyrifos methyl, cyfluthrin, cymoxanil, cypermethrin, cyproconazole, cyprodinil, cyromazine, deltamethrin, diazinon, dichlofluanid, dicloran, dicofol, difenoconazole, dimethoate, dimethoate, dimethomorph, diphenylamine, ditalimfos, ensodulfan, ethofenprox, fenarimol, fenazaquin, fenhexamide, fenitrothion, fenthion, fludioxonil, folpet, hexaconazole, imazalil, iprodione, Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31 iprovalicarb, kresoxim methyl, lambda cyhalothrin, linuron, lufenuron, malathion, metalaxyl, methidathion, methiocarb, methomyl, methoxychlor, myclobutanil, nuarimol, oxadiazon, paration, penconazole, pendimethanil, phosalone, piperonyl butoxide, pirimicarb, pirimiphos methyl, pp-DDD, procymidone, prometryn, propamocarb, propanil, propargite, propiconazole, pyrazophos, pyrimethanil, quinoxyfen, spiroxamina, tebuconazole, tebufenpyrad, tecnazene, tetrachlorvinphos, tetraconazole, thiabendazole, tiophanate methyl, tolylfluanide, triadimenol, trifloxystrobin, vinclozolin.

Pirimiphos-methyl was the pesticide most frequently found in the meal as a whole (93 times), in bread (79 times) and in the first course (22 times). In many cases pirimiphos-methyl residues were detected both in bread and the first course. The pesticide most frequently found in wine was procymidone (40 times), while in fruit it was chlorpyrifos (27 times) and in the side dish it was chlorpropham (12 times).

Table 4 shows the number of pesticides found in meals only for the 24 pesticides most frequently detected. Over the four years of the study, the trend was for an increase in pirimiphos-methyl, metalaxyl and fenhexamide, whereas there was a decrease in fenitrothion and procymidone. In particular, one can observe a considerable decrease in the values for procymidone in 2008, due to the cancellation of its use from 31 December 2007. The number of times cyprodinil and metalaxyl were detected increased considerably from 2008 as these were the pesticides substituted for

procymidone in crop treatments.

The number of times procymidone was detected also fell in fruit in 2008, whereas the presence of new pesticides authorised in Italy, such as boscalid, increased.

By multiplying the concentration of pesticide residues by the weight of the food, the quantity of pesticides in the first course, side dish, fruit, bread and wine was obtained: the data obtained was used to calculate the arithmetic mean.

Table 5 gives the average quantity of pesticides found in the first course, side dish, fruit, bread and wine. The 2005-2008 data shows that 51.7 % of the total quantity of pesticides found came from fruit, followed by wine (27.4 %), the side dish (8.7 %), the first course (7.6 %) and bread (4.5 %). Fruit contributes in a decisive manner to the dietary intake of pesticide residues. In 2005 the percentage was 77.3 %, whereas in subsequent years the percentage fell to 34.6 %. In 2008 the considerable increase in the percentage for wine should be noted, arriving at a level of 47.2 %. The maximum quantity of pesticides found in the meal (Table 6, column 7) was 10 – 50 times the average quantity (Table 5, column 7), with the highest level (1505  $\mu$ g) being detected in 2005, whereas in other years the maximum quantity of pesticides fell within the range of 151-375  $\mu$ g. In 2005, 2006 and 2007 the largest quantity of pesticides was always contained in fruit; in 2008 the highest levels were observed in wine.

To calculate the intake of pesticides for a teenager and child the contribution from wine

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consumption was not considered. The average number of pesticide residues thus fell from 3.4 to 2.5.
For teenagers and children 70.5 % of pesticides came from fruit, 12.7 % from the side dish, 10.3 % from the first course and 6.5 % from the bread.

### Comparison of the daily intake of pesticide residues with ADI

The daily intake per kg of body weight for each individual pesticide must be compared with the

ADI for that pesticide.

The result obtained, expressed as a percentage of ADI. Values greater than 100% indicate that the ADI has been exceeded, whereas values lower than 100%, indicate that the daily intake per kg of body weight is less than the ADI.

The results of analysis carried out in 2005-2008 allowed comparison between the daily intake per kg of body weight and the ADI, 680 times in the case of the adult and 498 times for a teenager and a child (Table 7, column 5). The difference in the number of comparisons depends on the fact that the contribution of wine was not considered for a teenager and a child.

The data comparing the daily intake per kg of body weight and the ADI established by the European Union, expressed as a percentage of ADI; gives a maximum value (Table 7, column 2) of 78 % - 51 % - 155 % - 349 % for adults, an average value (Table 7, column 3) of 3.7 % - 2.3 % - 4.4 % - 3.5 %

Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31 and a median level (Table 7, column 4) of 0.6 %– 0.6 %– 0.7 %– 0.4 %. For teenagers the maximum value was 117 %– 44 %– 232 %- 523 %, the average value 6.9 %– 3.5 %– 7.5 %– 6.9 % and the median level 1.1 %– 1.1 %-1.0 %-0.6 %. For children the maximum value was 234 %– 88 %– 464 %- 1046 %, the average 13.8 %– 7.1 %– 15.0 %– 13.8 % and the median level 2.2 %- 2.3 %– 2.0 %– 1.3 %. The data refers respectively to 2005 - 2006 - 2007 – 2008.

The number of values exceeding 100% (Tables 7, columns 6), is very low: in the case of children, only on 1.8 % of occasions (9 times out of 498) was this level exceeded over the four years of the study. Almost half the figures are under 1 %, leading to a median value much lower than the average.

Comparison of the values in this study with the results of studies to calculate the National Estimated Daily Intake (NEDI) and Total Diet Studies is very difficult, because different calculation systems have been used. It is possible to compare average daily intake as a percentage of ADI estimated by this study with the maximum value as a percentage of ADI estimated by other studies. The maximum values for NEDIs relative to pesticide residues for official monitoring studies calculated by Camoni *et. al.* (1998 e 2001) from 1993 to 1997 in Italy, have been estimated to be 4.8 % in 1995, whereas the other NEDI/ADI ratio values are almost always less than 1%. Similar considerations can be made as regards comparison of the values for the NEDI/ADI ratio estimated by Gaidano and Fabbrini (2000) with pesticide residues from private organisations, which

obtain data comparable with the data of Camoni et. al.(1998 e 2001).

During the period 2002 -2005 Tasiopoulou *et al.* (2007) observed chronic dietary exposure of the Italian population (adult, children and toddlers) to dicofol, with levels of 3.5 % for adults and 5 % for children and toddlers.

Using the most conservative model of chronic intake in U.K., Harris and Gaston (2004) observed dietary intake of glyphosate residues of 11 % of acceptable daily intake.

The results of the monitoring programme were taken into consideration by Berrada et al. (2006) to evaluate whether the estimated daily intake (EDI) of pesticides through the fruit consumed by the Spanish adult population is a cause for heath concern according to the recommended intake of the FAO/WHO. The maximum EDI/ADI values observed , as a percentage of ADI, were 8.61 % for citrus fruit.

Chronic dietary exposure of consumers to pesticide residues from Polish crops (Nowacka e Gnusowki, 2007) was estimated and supported by a national survey. This was shown not to be high and the maximum values observed in apples were 1.3 % for adults and 7.5 % for toddlers. Darko e Akoto (2008) calculated the Estimated Average Daily Intake (EADI) in Ghana, by multiplying average pesticide concentration by food consumption rate. The hazard indices were obtained by dividing EADIs by the corresponding values for FAO/WHO acceptable daily intake. In tomatoes the maximum value of the hazard indices was 7.9 %, increasing to 17.4 % in aubergines. Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31
In a study (duplicate diet method) carried out in France (Leblanc *et al.*, 2000) for 10 pesticides in 1998-1999, the daily intake estimates were generally low, representing a maximum of only 4% of ADI for pesticide residues.

FDA Total Diet Studies (Pennington *et al.*, 1987; Pennington *et al.*, 1996) estimated the mean daily intake for eight age-sex groups from 1984 to 1991. In the first study (1984-1986) the maximum value for the mean daily intake estimated for the group of males aged 14-16 (body weight 60 kg) was 5.8 % as a percentage of ADI, whereas in the second study (1986-1991) the maximum value was 3 % as a percentage of ADI.

In a Kuwaiti Total Diet Study, Sawaja *et al.* (1999) observed dietary intake of pesticides as a percentage of ADI. This was variable, with a maximum value of 15.9 %.

In Japan (Maitani, 2004), a Total Diet Study for pesticides from 1977 to 2002 highlighted average levels of daily intake as a percentage of ADI in 2002 much lower than 1 %

A Canadian Total Diet Study published by Rawn *et al.* (2004) calculated the dietary intake of pesticides for 10 groups of people aged from 1 to over 65 and recorded a maximum average daily intake of 2.9 % as a percentage of ADI for all ages.

In a Cameroonian Total Diet Study, Gimou *et al.* (2008) estimated mean dietary exposure to be from 0.24 % to 3.03 % of acceptable daily intake (ADI).

The results of studies to calculate the National Estimated Daily Intake (NEDI), the duplicate diet

Pubblicato sul La Rivista di Scienza dell'Alimentazione – Journal of Food Science and Nutrition Numero 3 Luglio – Settembre 2011 Pagine 19 - 31 method study and the Total Diet Studies showed maximum dietary intake of pesticides as a percentage of ADI ranging from 1 % to 17 % and did not highlight any trend for an increase or decrease over time.

The results of this study showed dietary intake of pesticides as a percentage of ADI, to be from 2 % to 4 % for adults and from 7 % to 15 % for children (Table 7, column 3, 2005-2008). The range found in this study and the ranges reported in the literature are very similar, even if referred to a long period of time and very different timescales to this study.

### Comparison of pesticide residues intake from meals with ARfD

The results in this study, compared intake from meals (acute intake per kg of body weight) with the ARfD values established by the European Union, expressed as a percentage of ARfD. In no case does the value exceed 100 %, namely when acute intake is greater than the ARfD values. The data comparing the intake from meals per kg of body weight and ARfD, expressed as a percentage of ARfD; gave a maximum value (Table 8, column 2) of 29 %- 25 %- 6 %- 7 % for adults, an average value (Table 8, column 3) of 1.1 %- 0.9 %- 0.4 %- 0.2 % and a median level (Table 8 column 4) of 0.2%- 0.1 %- 0.1 %- 0.04 %. For teenagers the maximum value was 44 %- 22 %- 10 %- 11 %, the average value 1.4 %- 0.7 %- 0.5 %- 0.4 % and the median level 0.1 %- 0.1

% - 0.1 % - 0.06 %. For children the maximum value was 88 % - 43 % - 17 % - 21 %, the average 2.7 % - 1.5 % - 1.1 % - 0.7 % and the median level 0.3 % - 0.3 % - 0.2 % - 0.1 %. The data relates to the following years respectively: 2005 -2006 -2007 -2008.

The number of values exceeding 100% (Tables 8, columns 6) was zero. More than 70 -80 % of values, expressed as a percentage of ARfD, were below 1 %, giving rise to a median value much lower than average.

The maximum values, expressed as a percentage of ARfD, were within the range 6 - 29 % for adults,

9-44 % for teenagers and 17 - 88 % for children.

Very similar values were observed by Jensen et al. (2003) and Nowacka e Gnusowki (2007).

The results of the Danish pesticide residue-monitoring programme were taken into consideration by

Jensen et al. (2003) to evaluate acute exposure risk assessment. ARfD for an adult or for a

5-year-old girl was not exceeded. The maximum values were 27 % (adult - percentage of ARfD)

and 69 % (child - percentage of ARfD).

Acute dietary exposure of consumers to pesticide residues from Polish crops was evaluated by Nowacka e Gnusowki (2007). The maximum exposure for adults was lower than 20 % of ARfD, while for toddlers it slightly exceeded 40 % of ARfD value.

## Conclusions

The 2005 - 2008 study devised and planned by the Environmental Agencies' Pesticides Working Group (AAAF group) allowed intake of pesticide residues through the diet to be estimated, by analysing a first course, side dish, fruit, bread and wine.

Two hundred meals were analysed by 17 laboratories and the data showed the presence of 3.4

pesticide residues on average in one meal.

Comparison between the estimated daily intake and ADI showed the average value, expressed as a

percentage of ADI, to be in the range 2 - 4 %, 3 - 7 % and 7 - 15 % respectively for adults,

teenagers and children.

In no case was acute intake greater than the ARfD values established by the European Union and the maximum values observed for children in the period 2005-2008 were 88 %-43 %-17 %-21 %, expressed as a percentage of ARfD.

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Figure 1. Average number of pesticide residues in the each dish and in the overall meal in the





Number	Laboratory	Years
1	ARPA Sicily Ragusa	2005-2006-2007-2008
2	ARPA Sicily Palermo	2005-2006
3	ARPA Sicily Catania	2005
4	ARPA Puglia Bari	2005-2006-2007-2008
5	ARPA Campania Napoli	2005-2006
6	ARPA Sardinia Cagliari	2005-2006-2007-2008
7	ARPA Marche Macerata	2005-2006-2007-2008
8	ARPA Tuscany Arezzo	2005-2006
9	ARPA Emilia Romagna Ferrara	2005-2006-2008
10	ARPA Liguria La Spezia	2005
11	ARPA Piemonte Turin	2005-2006
12	ASL Lombardia Bergamo	2005-2006-2007-2008
13	ARPA Veneto Verona	2005-2006
14	ARPA Friuli Venezia Giulia Pordenone	2005-2006-2007-2008
15	ARPA Friuli Venezia Giulia Gorizia	2005
16	APPA Trento	2005-2006-2007-2008
17	ARPA Valle d'Aosta	2006-2007-2008

**Table 1**. List of laboratories participating in the study.

Year	1 <sup>st</sup> sampling	2 <sup>nd</sup> sampling	3 <sup>rd</sup> sampling	4 <sup>th</sup> sampling
2005	8 Feb (tue)	26 May (thu)	24 Oct (mon)	21 Dec (wed)
2006	16 Mar (thu)	29 May (mon)	6 Oct (fri)	12 Dec (tue)
2007	30 Mar (fri)	30 May (wed)	8 Oct (mon)	11 Dec (tue)
2007 bis*	17 Apr (tue)	8 Jun (fri)	11 Oct (thu)	19 Dec (wed)
2008	7 Mar(fri)	15 May (thu)	15 Sep (mon)	5 Nov (tue)
2008 bis*	13 Mar (thu)	27 May (tue)	26 Sep (fri)	17 Nov (mon)

**Table 2.** Dates of sampling carried out in the period 2005-2008.

\* Further optional sampling

Table 3. The number of pesticide residues found in the meals: average and maximum levels in the

period 2005-2008.

	2005	2006	2007	2008	2005-2008
					average
Average	2.4	3.6	3.8	3.8	3.4
Maximum	10	9	13	8	10

nesticide	2005	2006	2007	2008	2005-2008
pesticide	2003	2000	2007	2008	total
pirimiphos-methyl	20	15	27	31	93
procymidone	17	23	24	6	70
cyprodinil	7	12	11	12	42
chlorpyrifos	6	16	7	7	36
iprodione	7	13	11	5	36
metalaxyl	5	8	8	15	36
pyrimethanil	7	7	7	5	26
carbaryl	2	4	12	5	23
diphenylamine	6	1	6	9	22
captan	2	3	8	7	20
chlorpyrifos-methyl	5	5	6	4	20
clorprofam	4	7	5	3	19
fludioxonil	2	6	5	5	18
fenhexamide	1	4	4	8	17
fenitrotion	6	6	5		17
diclofluanide	4	3	3	6	16
malation	1	6	5	1	13
imazalil	2	5	1	4	12
piperonil butossido	1	4	1	6	12
tiabendazolo	2	4	5	1	12
bromopropilato	3	1	3	1	8
azinfos metile	2	1	4		7
propamocarb		2	5		7
boscalid				6	6

**Table 4**. Number of pesticides found in the meals (for the 24 pesticides detected most frequently).

	First Course	Side dish	Fruit	Bread	Wine	Complete Meal
Years	μg	μg	μg	μg	μg	μg
2005	0.9	0.6	23.4	0.8	4.5	30.2
	(3.0 %)	(2.1 %)	(77.3 %)	(2.8 %)	(14.9 %)	(100 %)
2006	1.4	2.1	8.4	0.4	5.5	17.6
	(7.8 %)	(11.7 %)	(47.4 %)	(2.3 %)	(30.9 %)	(100 %)
2007	1.8	1.4	5.4	0.9	1.9	11.5
	(15.6 %)	(12.1 %)	(47.5 %)	(7.9 %)	(16.8 %)	(100 %)
2008	0.5	1.0	3.8	0.6	5.2	11.0
	(4.1 %)	(8.9 %)	(34.6 %)	(5.1 %)	(47.2 %)	(100 %)
2005-2008	1.1	1.3	10.2	0.7	4.3	17.6
average	(7.6 %)	(8.7 %)	(51.7 %)	(4.5 %)	(27.5 %)	(100 %)

**Table 5**. Average quantity of pesticides found in the meal.

\* The value in brackets is the quantity of pesticides as a percentage of the quantity found in the complete meal.

	First Course	Side dish	Fruit	Bread	Wine	Meal
Years	μg	μg	μg	μg	μg	μg
2005	21.6	52.2	1505.3	29.3	75.0	1505.3
2006	88.0	145.5	228.6	31.9	180.0	228.6
2007	92.9	40.0	151.1	41.2	67.0	151.1
2008	11.7	64.3	238.0	13.4	375.0	375.0

**Table 6**. Maximum quantity of pesticides found in the meal.

	Maximum (%)	Average (%)	Median (%)	Number of values Total	Number of values over 100 %	Number of values over 50 %	Number of values over 10 %	Number of values over 1 %		
	2005									
Adult	78.3	3.7	0.6	121	0	2	11	46		
Teenager	117.3	6.9	1.1	84	1	2	11	43		
Child	234.5	13.8	2.2	84	2	6	18	54		
				20	06					
Adult	51.1	2.3	0.6	170	0	1	9	66		
Teenager	44.0	3.5	1.1	120	0	0	11	65		
Child	88.0	7.1	2.3	120	0	3	19	77		
				20	07					
Adult	154.8	4.4	0.7	200	2	2	14	82		
Teenager	232.2	7.5	1.0	158	2	5	18	79		
Child	464.4	15.0	2.0	158	5	10	30	90		
				20	08					
Adult	348.8	3.5	0.4	189	1	1	8	50		
Teenager	523.3	6.9	0.6	136	1	2	12	51		
Child	1046.4	13.8	1.3	136	2	4	21	80		
				2005-	-2008					
Adult	51-349	2 - 4	0,4 – 0.7	680	3	6	42	244		
Teenager	44- 523	3 - 7	0.6 – 1.1	498	4	9	52	238		
Child	88 - 1046	7 - 15	1.3 – 2.3	498	9	23	88	301		

**Table 7**. Ratio of daily intake per kg of body weight and ADI levels.

	Maximum (%)	Average (%)	Median (%)	Number of values Total	Number of values over 100 %	Number of values over 50 %	Number of values over 10 %	Number of values over 1 %		
	2005									
Adult	29.3	1.1	0.2	71	0	0	2	11		
Teenager	44.0	1.4	0.1	56	0	0	1	8		
Child	87.9	2.7	0.3	56	0	1	2	10		
				200	)6					
Adult	25.0	0.9	0.1	108	0	0	3	14		
Teenager	21.7	0.7	0.1	88	0	0	1	14		
Child	43.3	1.5	0.3	88	0	0	1	18		
				200	)7					
Adult	5.8	0.4	0.1	133	0	0	0	9		
Teenager	8.7	0.5	0.1	115	0	0	0	15		
Child	17.4	1.1	0.2	115	0	0	3	25		
				200	)8					
Adult	7.0	0.2	0.04	104	0	0	0	4		
Teenager	10.5	0.4	0.06	93	0	0	1	9		
Child	20.9	0.7	0.12	93	0	0	1	12		
				2005-2	2008					
Adult	6 - 29	0.2 – 1.1	0.04 - 0.2	416	0	0	5	38		
Teenager	9 - 44	0.4 - 1.4	0.06 – 0.1	352	0	0	3	46		
Child	17 - 88	0.7 - 2.7	0.1-0.3	352	0	1	7	65		

**Table 8**. Ratio of intake from meal per kg of body weight and ARfD levels.