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Chronic dietary exposure to pesticide residues in the United States

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Abstract

Background: Discussions as to the extent of pesticide residue contamination in the food supply often rely on results of government residue monitoring programs focusing primarily upon the percentages of samples containing pesticide residues and the number of violative residues identified. Such an approach does not adequately convey the likelihood of pesticide residues posing consumer risks since residue regulatory limits are not safety standards and violative pesticide residues rarely constitute residues of health concern. It is more appropriate to develop estimates of actual dietary exposure to pesticides and to compare such estimates to established toxicological criteria such as the Chronic Reference Dose (RfD). The U.S. Food and Drug Administration's (FDA) Total Diet Study (TDS) previously provided such information but last published its findings in 1995 to estimate dietary exposure to pesticides detected between 1986 and 1991. This paper provides updated estimates of dietary exposure to pesticides in the United States using the most recent TDS findings on pesticide residues.

Results: A total of 77 specific pesticides were detected from market basket samples of 2240 TDS food items analyzed by FDA in 2004 and 2005. All estimated exposures to the 77 pesticides for the General US population were well below chronic RfD levels. Only 3 of the 77 pesticides showed exposures greater than 1 % of chronic RfDs, while 14 showed exposures between 0.1 and 1 % of chronic RfDs and 19 had exposures between 0.01 and 0.1 % of chronic RfDs. The remaining 41 pesticides had exposures below 0.01 % of chronic RfDs.
Compared with 1986–1991 findings, dietary exposure to six environmentally persistent chlorinated hydrocarbon insecticides were reduced by factors of 47 to 96 % in 2004–2005. Exposures to 15 different population subgroups were estimated and indicated that children, particularly two year-olds, frequently receive higher exposures to pesticide residues in their diets than do adults.

Conclusions: Chronic dietary exposure to pesticides in the diet, according to results of the FDA's 2004–2005 TDS, continue to be at levels far below those of health concern. Consumers should be encouraged to eat fruits, vegetables, and grains and should not fear the low levels of pesticide residues found in such foods.

Keywords: Pesticides; Food safety; Risk assessment; Exposure assessment; Total diet study; Reference dose

Background

Residues of pesticide chemicals are frequently detected on the edible portions of foods and consumers are routinely exposed to pesticide residues in their diets. The extent of pesticide contamination in the food supply is often discussed in terms of the results of regulatory monitoring conducted by bodies such as the U.S. Food and Drug Administration (FDA) and the European Food Safety Authority (EFSA). In 2012, FDA monitoring results of 1,158 domestic samples showed that 57 % had no detectable residues while 2.8 % contained violative pesticide residues (FDA 2015). Among the 4,365 imported food samples analyzed, 66 % had no detectable residues while violations were detected on 11.1 % of the samples. EFSA reported on the analysis of 80,967 food samples collected in 2013 and concluded that 54.6 % of the samples had no detectable residues while 2.6 % of the samples resulted in violations (EFSA 2015).

Such findings may be confusing to consumers since the majority of samples contained no detectable residues of pesticides although pesticide residue violations also frequently occurred. Unfortunately, while counterintuitive, the recitation of findings from regulatory monitoring programs

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is of little value in terms of assessing the potential health risks posed by pesticide residues in foods. This is primarily due to the fact that the allowable levels are not indicators of safety but rather reflect enforcement tools to assess whether Good Agricultural Practices have been followed (Winter, 1992). As such, residue violations often indicate breaches of Good Agricultural Practices but only in very rare circumstances represent cases of health concern. To clarify the differences between allowable pesticide residue levels and levels of health significance, a companion system to establish levels of safety concerns for specific pesticides on specific food items has been proposed (Winter and Jara, 2015).

The potential health risks posed by pesticide residues in foods can best be assessed by developing estimates of dietary exposure to pesticides and comparing exposure estimates to toxicological indicators of health concern such as the Chronic Reference Dose (RfD) or the analogous Acceptable Daily Intake (ADI). An accurate estimation of dietary pesticide exposure requires data on specific levels of pesticide residues detected (not just whether the residues were legal or violative) as well as estimations of consumption amounts of all foods for which residues are detected.

The FDA has conducted the Total Diet Study (TDS) annually since 1961 (Gunderson 1995). The TDS uses a market basket approach in which FDA inspectors purchase market baskets of 280 food items from retail locations in various regions of the country. The food items are prepared for consumption and then analyzed for potential food contaminants such as radionuclides and pesticide residues as well as for metals and selected nutrients. Food consumption estimates for the TDS diets are derived from national food consumption databases. By multiplying the levels of pesticide residues found on the food items by food consumption estimates, it is possible to obtain an estimate of chronic dietary exposure to pesticide residues in foods.

Historically, FDA released its estimates of chronic dietary exposure to pesticides obtained from TDS findings and compared exposure estimates to ADI levels. Unfortunately, the last public report of such findings was made in 1995 and estimated dietary exposure to pesticides from TDS market baskets collected between 1986 and 1991 (Gunderson 1995). Thus, while FDA continues to analyze TDS samples for pesticide residues, it has not made its findings of TDS consumer exposure to pesticides available for two decades.

At the same time, FDA does release the analytical data resulting from subsequent TDS market baskets as well as food consumption estimates for each of the TDS foods. It is therefore possible to estimate dietary exposure to pesticides from TDS data and to compare exposure levels to chronic RfD or ADI levels.

This paper provides estimates of dietary exposure to pesticides based upon the most recent two years (2004 and 2005) for which TDS pesticide residue data are available. Exposure estimates are compared with chronic RfD values to determine the potential risks associated with the exposures. In addition, comparisons between exposures from the 2004–2005 and the 1986–1991 market baskets are made to identify longer-term trends in dietary exposure to pesticides.

Methods

Pesticide residue levels on the 280 TDS foods were obtained from FDA's summary of pesticide residue findings for Market Baskets 2004–1 through 2005–4 (FDA 2014). While analytical methods were capable of detection of more than 300 pesticides and/or pesticide metabolites or breakdown products, residues were detected for 77 specific pesticides. According to FDA guidelines, a value of 0 was assigned for residues below the analytical method's limit of detection while trace findings were reported at the measured value.

Food consumption estimates for each of the 280 TDS foods were acquired by downloading FDA's "TDS Diets, Version 3" (FDA 2013). Food consumption estimates were derived from the results of the United States Department of Agriculture's 1994–96 and 1998 Continuing Survey of Food Intakes by Individuals. Consumption estimates were expressed in terms of grams of food per day and were separated into 15 population subgroups: Total US population, infants 6–11 months, children 2 years, children 6 years, children 6–10 years, males (14–16 years, 25–30 years, 40–45 years, 60–65 years, 70 years) and females (14–16 years, 25–30 years, 40–45 years, 60–65 years, 70 years).

To estimate dietary exposure to specific pesticides for each of the population subgroups, the mean residue level (expressed in mg/kg) found on a specific TDS food was multiplied by the food consumption estimate to yield an estimate of the daily exposure to the pesticide on that food. If, as was common, a pesticide was detected on more than one TDS food, contributing exposures from each food were combined to yield a daily exposure estimate to the pesticide.

To allow direct comparisons between chronic RfD values and exposure estimates, daily pesticide exposure levels were divided by body weight levels estimated for each population subgroup (EPA 2011). Final chronic dietary exposures to specific pesticides for each population subgroup were then expressed in terms of nanograms pesticide/kg body weight/day (ng/kg/day).

The methods used to estimate chronic dietary exposure to pesticides represent a simplistic deterministic approach in which residues are considered to be present at mean levels and food consumption estimates are chosen that represent the 50th percentile for each of the 280 foods considered. As such, the approach attempts to most accurately characterize typical chronic dietary exposure to pesticides. The use of higher percentiles of residue level or food consumption estimates would serve to artificially inflate exposure estimates. Deterministic methods using higher percentiles of residue levels and/or food consumption estimates are useful, and necessary, in assessing acute exposure to pesticides in foods but their use in a chronic exposure assessment is not warranted.

Chronic RfD values for specific pesticides were primarily obtained from the Integrated Risk Information System (IRIS) used by the United States Environmental Protection Agency (EPA). The chronic RfD represents an estimate of the amount of a chemical an individual could be exposed to daily throughout the individual's lifetime that is likely to be without an appreciable risk of harm (Winter and Francis 1997). In some cases, chronic RfD values were not available through IRIS but could be identified through documents discussing EPA Pesticide Reregistration Eligibility Decisions for specific pesticides. Chronic RfD values were not obtained for the pesticides azinphos-methyl, chlorpyrifos-methyl, dicloran, dicofol, fenhexamid, omethoate, phenylphenol-ortho, quinoxyfen, tecnazene, thiabendazole, triadimefon, and trifloxystrobin; ADI values from the literature were chosen as chronic RfD surrogates for each of these pesticides.

Results and discussion

The FDA analyzed a total of 2240 food items (8 market baskets x 280 items per market basket) in its 2004 and 2005 TDS for residues of more than 300 pesticides and/or metabolites and breakdown products. Residues of 77 pesticides were detected.

The estimated chronic dietary exposures to the 77 pesticides for the General US population are compared with chronic RfD values in Table 1. Exposure levels to all 77 pesticides were below chronic RfD values. Exposures to 3 pesticides exceeded 1 % of the chronic RfD values while exposures between 0.1 and 1 % of chronic RfD values were noted for 14 pesticides. Another 19 pesticides demonstrated exposures between 0.01 and 0.1 % of the chronic RfD values, while exposures for the other 41 pesticides were below 0.01 % of the chronic RfD. The median exposure relative to the chronic RfD for the 77 pesticides was for the fungicide captan, with an exposure of 0.0074 % of the chronic RfD.

The determination of chronic RfD values requires assessment of the No Observed Adverse Effect Level (NOAEL), which represents the highest continuous dose given to the most sensitive animal species that does not demonstrate any noticeable toxicity (Winter and Francis 1997). The NOAEL levels are typically reduced using uncertainty factors (100 or, in some cases 1000) to produce the chronic

Table 1 Comparison of General US dietary exposure to pesticides with Chronic Reference Doses (RfDs)

Pesticide	General US Exposure (ng/kg/day)	Chronic RfD (ng/kg/day)	%RfD
Acephate	18.4281	4000	0.4607
Atrazine	0.0072	35000	<0.0001
Azinphos-methyl	11.1229	5000	0.2225
Azoxystrobin	1.1496	180000	0.0006
Benomyl	9.2942	50000	0.0186
Bifenthrin	0.9997	15000	0.0067
Biphenyl	0.3963	500000	0.0001
Captan	9.6072	130000	0.0074
Carbaryl	10.2073	100000	0.0102
Carbofuran	0.3657	5000	0.0073
Chlordane (total)	0.0992	500	0.0198
Chlordane, cis	0.0479		
Chlordane, trans	0.0125		
Nonachlor, trans	0.0367		
Octachlor epoxide	0.0020		
Chloropropham	287.7204	200000	0.1439
Chlorothalonil	0.1205	15000	0.0008
Chlorpyrifos	2.3760	3000	0.0792
Chlorpyrifos-methyl	12.7964	10000	0.1280
Clopyralid	0.8296	150000	0.0006
Coumaphos	0.0068	700	0.0010
Cyfluthrin	0.2220	24000	0.0009
Cypermethrin	8.9699	10000	0.0897
Cyprodinil	12.9971	37500	0.0347
2,4-D	0.6738	10000	0.0067
DCPA	0.2398	10000	0.0024
DDT, total	6.6482	500	1.3296
DDE, p, p′	5.9085		
DDT, o, p′	0.0120		
DDT, p, p′	0.1471		
TDE, o, p′	0.0258		
TDE, p, p′	0.5547		
DEF	0.1086	100	0.1086
Diazinon	0.2394	200	0.1197
Dicamba	0.7488	30000	0.0025
Dichloran, total	2.9128	2500	0.1165
Dichloran	2.8691		
2,4-Dichloro-6-nitrobenzeneamine	0.0437		
Dicofol, total	0.3547	200000	0.0002
Dicofol, o, p′	0.0345		
Dicofol, p, p′	0.3201		

Table 1 Comparison of General US dietary exposure to pesticides with Chronic Reference Doses (RfDs) (Continued)

Dieldrin	0.9841	50	1.9683
Dimethoate	1.5595	200	0.7797
Diphenylamine	79.6903	25000	0.3188
Endosulfan, total	9.3737	6000	0.1562
Endosulfan I	1.6268		
Endosulfan II	2.1831		
Endosulfan sulfate	5.5638		
Endrin	0.0228	300	0.0076
Esfenvalerate	0.0281	1800	0.0016
Ethion, total	0.4912	500	0.0982
Ethion	0.4869		
Ethion, oxygen analog	0.0043		
Ethoxyquin	0.5940	20000	0.0030
Famoxadone	0.1185	1400	0.0085
Fenarimol	0.0196	65000	<0.0001
Fenhexamid	0.9919	200000	0.0005
Fenitrothion	0.0132	1500	0.0009
Fenoxaprop-ethyl	0.0002	15000	<0.0001
Fenpropathrin	3.3847	25000	0.0135
Fenvalerate	0.3790	25000	0.0015
Fludioxonil	21.1253	30000	0.0704
Heptachlor	0.0593	13	0.4563
Imazalil	0.5742	13000	0.0044
Iprodione, total	6.4931	40000	0.0162
Iprodione	6.1491		
Iprodione metabolite	0.3441		
Lambda-cyhalothrin	0.6892	5000	0.0138
Lindane	0.0693	300	0.0231
Linuron	0.2379	2000	0.0119
Malathion	29.6982	20000	0.1485
Metalaxyl	1.0858	60000	0.0018
Methamidophos	7.9984	50	15.9968
Methidathion	0.9529	1000	0.0953
Methomyl	1.9347	25000	0.0077
Methoxychlor, total	0.0428	5000	0.0009
Methoxychlor, o, p′	0.0173		
Methoxychlor, p, p′	0.0255		
Mirex	0.0020	200	0.0010
Omethoate	0.8798	300	0.2933
Oxamyl	0.0466	25000	0.0002
Permethrin, total	73.4122	50000	0.1468
Permethrin, cis	35.4246		
Permethrin, trans	37.9876		
Phenylphenol, ortho	11.3419	20000	0.0567
Phosalone	0.0491	2000	0.0025
Phosmet	4.7271	20000	0.0236
Piperonyl butoxide	0.0029	6300000	<0.0001
Pirimiphos-methyl	6.3524	10000	0.0635
Procymidone	0.0247	35000	0.0001
Propargite	11.6608	20000	0.0583
Propiconazole	0.0002	13000	<0.0001
Quinclorac	1.0475	380000	0.0003
Quinoxyfen	0.2185	200000	0.0001
Quintozene	0.0743	3000	0.0025
Tecnazene	0.0008	20000	<0.0001
Thiabendazole	77.7198	100000	0.0777
Triadimefon	0.0033	10000	<0.0001
Triclopyr	0.0398	50000	0.0001
Trifloxystrobin	0.3277	100000	0.0003
Vinclozolin	0.8230	25000	0.0033

RfD level. In the case of dietary exposure to a pesticide that represents 0.01 % of the chronic RfD, the exposure level (assuming the use of a 100 fold uncertainty factor in deriving the RfD) is one million times lower than levels that do not cause any noticeable effects in animals dosed continuously with the pesticide throughout their lifetimes. Thus, findings from this study reinforce the notion that typical consumer exposure to pesticide residues is at levels far below those of health concern.

The highest exposure of a pesticide relative to the chronic RfD was for methamidophos, which showed an exposure to the General US population of 16 % of the chronic RfD. Methamidophos is an organophosphate insecticide that shares a common toxicological mechanism with several other common insecticides. The estimated exposure to methamidophos for the General US population, 8.0 ng/kg/day, is actually lower than estimated exposures to other organophosphate insecticides such as

Table 2 Comparison of chlorinated hydrocarbon insecticide dietary exposure for two year-olds, 2004–05 vs. 1986–91 [a]

Pesticide	2004–05 (ng/kg/day)	1986–91 (ng/kg/day)	Percentage reduction
DDT	23.18	43.8	47
Dicofol	1.52	35.5	96
Dieldrin	2.7	7.2	63
Heptachlor	0.15	2.5	94
Lindane	0.26	3.2	92
Methoxychlor	0.1	0.9	89

[a]1986–91 data obtained from Gunderson (1995)

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a]

Pesticide	US General	6–11 month	M/F 2 years	M/F 6 years	M/F 10 year	F 14–16
Acephate	18.4281	17.2302	**42.6352**	29.8366	17.0595	8.5112
Atrazine	0.0072	0.0103	**0.0203**	0.0096	0.0052	0.0047
Azinphos-methyl	11.1229	32.5191	**82.2131**	47.7940	26.9670	9.1684
Azoxystrobin	1.1496	0.8092	**4.6546**	2.2937	2.1211	1.1600
Benomyl	9.2942	57.5515	**70.8385**	34.5814	17.4725	7.3732
Bifenthrin	0.9997	**2.8008**	2.1272	1.6090	0.6109	0.3124
Biphenyl	0.3963	0.8070	**1.4732**	1.1553	0.6452	0.4489
Captan	9.6072	7.0953	**72.0200**	34.5332	21.3902	9.9809
Carbaryl	10.2073	64.3193	**83.9221**	41.4619	18.4712	11.6797
Carbofuran	0.3657	0.2989	**1.2256**	1.3252	0.4160	0.1333
Chlordane (total)	0.0992	0.0698	**0.1761**	0.1504	0.1270	0.0739
Chlordane, cis	0.0479	0.0220	0.0914	0.0790	0.0598	0.0378
Chlordane, trans	0.0125	0.0146	0.0234	0.0172	0.0198	0.0086
Nonachlor, trans	0.0367	0.0296	0.0585	0.0511	0.0461	0.0267
Octachlor epoxide	0.0020	0.0036	0.0028	0.0032	0.0012	0.0007
Chloropropham	287.7204	242.2743	**981.7293**	785.8608	528.4143	351.6736
Chlorothalonil	0.1205	0.0000	**0.2162**	0.1754	0.1824	0.0259
Chlorpyrifos	2.3760	2.5690	**9.8882**	5.5129	3.8547	2.2513
Chlorpyrifos-methyl	12.7964	12.4004	**40.6889**	37.3143	23.2516	13.4883
Clopyralid	0.8296	2.3563	**5.2877**	3.2069	1.8738	0.6758
Coumaphos	0.0068	0.0025	0.0218	0.0109	0.0014	0.0028
Cyfluthrin	0.2220	0.0079	0.1208	0.1013	0.1664	0.0467
Cypermethrin	8.9699	8.1437	**17.1238**	10.4601	3.8659	6.0168
Cyprodinil	12.9971	11.9101	**119.2924**	47.1034	31.9188	14.6596
2,4-D	0.6738	0.5266	**2.1795**	1.8683	1.1665	0.5975
DCPA	0.2398	0.1450	**0.5591**	0.3654	0.2523	0.1717
DDT, total	6.6482	8.0532	**23.1850**	18.0708	11.8551	6.3338
DDE, p, p′	5.9085	7.3662	21.6413	16.9921	11.1399	5.7689
DDT, o, p′	0.0120	0.0062	0.0156	0.0098	0.0070	0.0100
DDT, p, p′	0.1471	0.3271	0.6385	0.3535	0.2306	0.1427
TDE, o, p′	0.0258	0.0530	0.0241	0.0231	0.0076	0.0122

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a] (Continued)

TDE, p, p′	0.5547	0.3008	0.8654	0.6923	0.4700	0.4000
DEF	0.1086	0.0153	**0.4647**	0.3953	0.2353	0.1265
Diazinon	0.2394	0.0658	**0.3905**	0.3455	0.2217	0.2152
Dicamba	0.7488	2.2202	**4.8366**	3.0227	1.8074	0.6574
Dichloran, total	2.9128	**6.4696**	5.3965	3.9703	3.9514	1.6418
Dichloran	2.8691	6.4696	5.3180	3.9066	3.8853	1.6323
2,4-Dichloro-6-nitrobenzeneamine	0.0437	0.0000	0.0784	0.0636	0.0662	0.0094
Dicofol, total	0.3547	0.4972	**1.5264**	0.9061	0.6697	0.2927
Dicofol, o, p′	0.0345	0.0465	0.2390	0.1394	0.0934	0.0248
Dicofol, p, p′	0.3201	0.4508	1.2874	0.7667	0.5763	0.2679
Dieldrin	0.9841	1.5196	**2.7181**	2.1000	1.5493	0.7663
Dimethoate	1.5595	**9.5053**	6.1889	3.5457	1.8318	1.0376
Diphenylamine	79.6903	98.6515	**606.5610**	395.4847	180.9575	70.1601
Endosulfan, total	9.3737	11.0237	**21.0158**	16.0028	10.7486	6.8948
Endosulfan I	1.6268	1.8331	4.4537	3.0479	2.2759	1.4302
Endosulfan II	2.1831	2.4196	6.1116	4.1611	2.9966	1.8655
Endosulfan sulfate	5.5638	6.7710	10.4504	8.7938	5.4761	3.5992
Endrin	0.0228	**0.0627**	0.0158	0.0187	0.0025	0.0075
Esfenvalerate	0.0281	**1.1880**	0.1006	0.0843	0.0328	0.0416
Ethion, total	0.4912	0.1987	1.3184	**1.3622**	0.9313	0.6354
Ethion	0.4869	0.1979	1.3153	1.3588	0.9288	0.6332
Ethion, oxygen analog	0.0043	0.0008	0.0031	0.0035	0.0025	0.0022
Ethoxyquin	0.5940	0.0848	0.7868	**1.6565**	0.8585	1.0186
Famoxadone	0.1185	0.0042	0.0645	0.0541	0.0888	0.0249
Fenarimol	0.0196	0.0104	**0.1187**	0.0392	0.0066	0.0055
Fenhexamid	0.9919	0.8467	**5.9062**	2.5504	2.0440	1.0660
Fenitrothion	0.0132	0.0118	0.0191	0.0261	0.0146	0.0120
Fenoxaprop-ethyl	0.0002	**0.1076**	0.0002	0.0000	0.0000	0.0000
Fenpropathrin	3.3847	15.5940	**25.9598**	13.3071	7.3449	3.1080
Fenvalerate	0.3790	**3.0032**	1.0053	0.5161	0.2748	0.2500
Fludioxonil	21.1253	33.0302	**66.5408**	56.8803	13.5351	26.1458
Heptachlor	0.0593	0.0534	**0.1478**	0.1303	0.0978	0.0489
Imazalil	0.5742	0.1826	0.3263	0.1806	0.1094	0.0859
Iprodione, total	6.4931	11.0094	**26.2975**	18.0616	5.8599	7.8378
Iprodione	6.1491	10.3505	25.2904	17.2475	5.6553	7.4416

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a] (Continued)

Iprodione metabolite	0.3441	0.6589	1.0071	0.8142	0.2046	0.3961
Lambda-cyhalothrin	0.6892	**3.5221**	1.2727	0.5294	0.2707	0.6808
Lindane	0.0693	0.0272	0.2586	**0.2933**	0.1840	0.0942
Linuron	0.2379	0.2362	**0.5644**	0.5572	0.3341	0.1655
Malathion	29.6982	24.4021	**94.4458**	89.6674	57.0931	29.6243
Metalaxyl	1.0858	0.7104	**9.1932**	3.6071	2.3705	1.1077
Methamidophos	7.9984	13.9201	**15.9612**	9.7545	7.1500	4.5228
Methidathion	0.9529	0.7878	**5.6458**	3.3187	2.2614	1.3605
Methomyl	1.9347	1.1424	**6.5387**	3.6508	3.0433	1.7831
Methoxychlor, total	0.0428	0.0244	**0.1006**	0.0846	0.0622	0.0199
Methoxychlor, o, p′	0.0173	0.0000	0.0000	0.0000	0.0000	0.0000
Methoxychlor, p, p′	0.0255	0.0244	0.1006	0.0846	0.0622	0.0198
Mirex	0.0020	0.0010	0.0040	0.0024	**0.0042**	0.0021
Omethoate	0.8798	**3.6634**	1.7655	1.0999	0.8271	0.5772
Oxamyl	0.0466	0.0024	0.0764	**0.0775**	0.0538	0.0211
Permethrin, total	73.4122	88.3346	**101.6469**	80.6081	28.9863	22.4073
Permethrin, cis	35.4246	42.4138	48.9996	38.7410	14.0356	11.0349
Permethrin, trans	37.9876	45.9208	52.6472	41.8671	14.9507	11.3723
Phenylphenol, ortho	11.3419	**36.9188**	22.7534	17.2762	13.4283	7.9713
Phosalone	0.0491	**0.1417**	0.0232	0.0477	0.0617	0.0561
Phosmet	4.7271	17.3239	**19.7964**	15.2866	4.9499	5.3718
Piperonyl butoxide	0.0029	**1.5171**	0.0307	0.0000	0.0000	0.0000
Pirimiphos-methyl	6.3524	2.0673	19.2953	**19.6057**	15.6757	4.3406
Procymidone	0.0247	0.0127	0.0508	**0.0672**	0.0407	0.0377
Propargite	11.6608	16.8366	**55.0796**	33.4737	6.9992	
Propiconazole	0.0002	**0.1076**	0.0002	0.0000	0.0000	0.0000
Quinclorac	1.0475	2.4162	**3.8001**	2.5547	1.6717	0.9916
Quinoxyfen	0.2185	0.1696	**2.1860**	0.8194	0.5358	0.2512
Quintozene	0.0743	0.0952	**0.2157**	0.1604	0.1027	0.0518
Tecnazene	0.0008	0.0003	0.0009	0.0008	0.0010	0.0008
Thiabendazole	77.7198	297.9485	**615.7219**	346.4239	157.2699	67.4992
Triadimefon	0.0033	0.0090	**0.0348**	0.0024	0.0058	0.0052
Triclopyr	0.0398	0.1179	**0.2211**	0.1905	0.1436	0.0520
Trifloxystrobin	0.3277	0.2543	**3.2789**	1.2290	0.8038	0.3767
Vinclozolin	0.8230	0.9713	**2.9116**	1.5535	1.1562	0.7111

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a] (Continued)

M 14–16	F 25–30	M 25–30	F 40–45	M 40–45	F 60–65	M 60–65	F 70	M 70
16.6332	16.2023	15.7001	21.6464	16.1272	18.6599	17.3764	20.7385	24.0702
0.0047	0.0051	0.0053	0.0066	0.0067	0.0068	0.0085	0.0117	0.0138
8.3018	6.9344	7.9611	9.1004	7.1628	6.4063	10.6951	10.3453	12.4859
0.9070	1.0642	0.3536	1.3359	0.6003	1.4125	1.0705	1.1712	1.1343
5.7494	8.0872	5.3303	7.9251	5.6981	15.7172	8.8867	11.2131	9.4220
1.0640	0.8047	0.8643	0.7484	0.6973	1.7146	1.0494	1.3084	0.9522
0.4603	0.2670	0.2977	0.3111	0.3271	0.2688	0.3726	0.4180	0.4281
5.6579	7.1469	4.0346	9.5369	5.7625	7.9005	8.7611	9.7644	9.2733
10.4841	8.4267	6.1624	7.8695	7.7365	9.6286	8.2916	10.5855	8.9616
0.6874	0.2739	0.0836	0.3614	0.2877	0.2016	0.2547	0.3386	0.4815
0.0870	0.0689	0.0728	0.0768	0.0817	0.1271	0.0962	0.1116	0.1194
0.0416	0.0325	0.0372	0.0370	0.0383	0.0589	0.0503	0.0539	0.0607
0.0125	0.0091	0.0084	0.0095	0.0113	0.0177	0.0094	0.0132	0.0120
0.0313	0.0256	0.0257	0.0287	0.0306	0.0464	0.0347	0.0418	0.0441
0.0016	0.0016	0.0015	0.0016	0.0015	0.0042	0.0018	0.0026	0.0026
525.9805	249.3211	375.2243	191.8389	269.2749	116.9667	224.2651	158.1203	195.8510
0.1221	0.1160	0.0956	0.1034	0.0905	0.3776	0.1823	0.1505	0.1970
2.3484	1.8354	1.9704	2.3210	2.0765	4.0656	2.2079	2.4321	2.1384
19.1060	9.3327	12.5135	10.0469	11.2686	9.4714	10.4018	10.5705	11.0058
1.0531	0.5116	0.6111	0.4960	0.6298	0.6130	0.7019	1.0500	1.0474
0.0059	0.0055	0.0048	0.0056	0.0029	**0.0589**	0.0034	0.0073	0.0102
0.1485	0.1835	0.3236	0.3696	0.2331	**0.3776**	0.1847	0.1690	0.2134
7.3911	5.9054	7.3996	7.4440	5.7677	16.1349	12.9267	13.8612	9.2338
4.4096	9.4482	3.2211	12.4248	8.1982	13.2693	11.2770	15.4098	12.2943
0.8322	0.5248	0.6807	0.5106	0.5936	0.2281	0.6328	0.6007	0.6054
0.1457	0.2279	0.1898	0.2575	0.1982	0.2792	0.2255	0.2900	0.2151
10.0929	5.3405	5.8316	4.8189	5.9851	5.9422	6.5278	5.2919	5.3867
9.2297	4.6048	5.4149	4.1668	5.2854	5.1620	5.5193	4.4752	4.6449
0.0122	0.0092	0.0099	0.0120	0.0098	0.0188	0.0136	0.0129	0.0123
0.2212	0.1143	0.1368	0.1157	0.1290	0.1260	0.1384	0.1317	0.1387
0.0166	0.0240	0.0127	0.0230	0.0196	0.0365	0.0280	0.0404	0.0352

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a] (Continued)

0.6132	0.5882	0.2573	0.5015	0.5414	0.5990	0.8284	0.6318	0.5557
0.1867	0.0914	0.1604	0.0800	0.0989	0.1099	0.0761	0.0407	0.0376
0.1779	0.2044	0.1988	0.2714	0.2196	0.3073	0.2134	0.1736	0.2062
1.0057	0.4768	0.5371	0.4497	0.6017	0.5380	0.6243	0.8806	0.8722
3.5281	2.6613	2.5448	2.4112	2.4526	6.1333	3.5399	2.9938	3.5544
3.4838	2.6193	2.5101	2.3737	2.4197	5.9964	3.4738	2.9392	3.4830
0.0235	0.0245	0.0219	0.0210	0.0225	0.0755	0.0320	0.0532	0.0526
0.2921	0.2439	0.2535	0.2669	0.2621	0.4260	0.3315	0.4221	0.3968
1.2978	0.7177	0.8500	0.7267	0.8265	1.1578	0.8707	1.0342	0.9787
1.3173	1.3197	1.0581	1.6276	1.3318	2.3255	1.2750	1.3512	1.5124
65.1788	48.8023	64.6768	70.0400	50.1053	21.8490	78.2527	64.3824	81.6502
10.0291	7.2010	8.0732	8.6182	7.8821	11.6984	8.2978	9.3896	9.2824
1.8862	1.2782	1.4997	1.4701	1.4373	1.5188	1.3303	1.3647	1.3876
2.7102	1.7453	2.0406	2.0020	1.7985	2.3578	1.8384	1.8595	1.8490
5.4327	4.1775	4.5329	5.1461	4.6463	7.8219	5.1291	6.1655	6.0459
0.0108	0.0202	0.0118	0.0198	0.0153	0.0323	0.0191	0.0384	0.0349
0.0329	0.0152	0.0231	0.0183	0.0161	0.1703	0.0291	0.0398	0.0277
0.9142	0.4462	0.5556	0.4021	0.4089	0.5313	0.2811	0.2328	0.2838
0.9111	0.4409	0.5519	0.3982	0.4049	0.5255	0.2792	0.2307	0.2810
0.0031	0.0053	0.0037	0.0039	0.0040	0.0057	0.0019	0.0020	0.0028
0.7313	0.6021	0.7855	0.6930	0.6076	2.0313	0.3458	0.2589	0.3347
0.0793	0.0980	0.1728	0.1973	0.1244	**0.2016**	0.0986	0.0902	0.1139
0.0032	0.0145	0.0184	0.0167	0.0156	0.0521	0.0155	0.0212	0.0312
0.6234	0.8525	0.2541	1.0779	0.5421	1.1354	0.8992	1.0622	0.9554
0.0091	0.0111	0.0110	0.0176	0.0114	**0.0307**	0.0082	0.0065	0.0057
0.0000	0.0000	0.0000	0.0000	0.0000	0.0130	0.0000	0.0000	0.0004
2.0885	2.1623	2.1035	3.0221	2.2667	3.5484	3.2033	3.4172	3.5806
0.2464	0.2754	0.2897	0.3427	0.3419	0.6703	0.4263	0.6357	0.7162
13.5739	18.9030	7.6395	24.7306	19.2154	18.2427	20.6759	27.8568	17.7320
0.0816	0.0474	0.0569	0.0443	0.0543	0.0740	0.0428	0.0483	0.0468
0.4188	0.3255	0.2403	0.5650	0.4071	0.6250	0.7987	1.5949	**1.7592**
3.7946	5.6403	2.2778	7.4015	5.6484	6.2786	6.2383	8.3525	5.5698
3.5852	5.3320	2.1398	6.9979	5.3389	5.9016	5.8986	7.9255	5.2641

Table 3 Dietary pesticide exposure estimates for specific population subgroups (ng/kg/day) [a] (Continued)

0.2094	0.3083	0.1380	0.4036	0.3095	0.3771	0.3397	0.4270	0.3057
0.2008	0.4422	0.4518	0.8299	0.4960	1.7703	0.9220	0.8178	0.7633
0.1176	0.0677	0.0548	0.0509	0.0754	0.1302	0.0425	0.0358	0.0397
0.1302	0.2156	0.1544	0.2746	0.2104	0.3906	0.2221	0.1815	0.2134
42.7260	21.6705	29.3563	22.6826	26.6826	18.7036	25.9992	24.2481	25.5719
0.3646	0.7334	0.4357	1.0752	0.7592	1.1266	0.9553	1.2109	1.0349
6.0969	6.0924	7.5724	8.8198	7.6102	8.5927	8.3945	9.4966	10.7210
1.5486	0.7018	0.9954	0.6535	0.6442	0.2578	0.7235	0.9699	0.7849
1.5201	1.6876	1.0198	2.2454	1.3298	1.9276	1.7928	1.9009	1.8352
0.0331	0.0443	0.0364	0.0434	0.0402	0.0474	0.0454	0.0268	0.0340
0.0005	0.0215	0.0150	0.0220	0.0194	0.0042	0.0214	0.0118	0.0162
0.0326	0.0228	0.0214	0.0214	0.0208	0.0432	0.0241	0.0150	0.0178
0.0012	0.0017	0.0014	0.0016	0.0016	0.0021	0.0022	0.0021	0.0019
0.6311	0.7045	0.7028	0.9551	0.7554	1.4615	0.8879	0.8878	0.8731
0.0387	0.0272	0.0453	0.0432	0.0400	0.0714	0.0536	0.0423	0.0630
80.2934	57.2709	66.1862	56.3328	48.4414	130.2292	82.2046	101.4862	72.1103
38.5280	27.6290	31.8599	27.3468	23.4409	62.8005	39.6630	48.7888	34.8039
41.7654	29.6419	34.3263	28.9859	25.0005	67.4286	42.5416	52.6974	37.3064
23.6654	7.2615	6.2418	10.2150	9.1555	27.4469	13.1577	17.8501	14.0892
0.0464	0.0734	0.0799	0.0449	0.0500	0.0703	0.0111	0.0071	0.0156
3.2591	3.8788	2.2576	5.1557	3.9565	4.5583	4.6251	5.6095	4.2228
0.0000	0.0000	0.0000	0.0000	0.0000	0.1823	0.0000	0.0000	0.0000
8.1045	5.2557	6.2427	5.4557	4.8230	8.8276	5.5365	3.4975	3.7984
0.0648	0.0205	0.0188	0.0278	0.0245	0.0266	0.0167	0.0188	0.0194
6.6315	9.0565	5.8634	12.0346	9.6961	19.5833	10.0678	15.6229	13.0497
0.0000	0.0000	0.0000	0.0000	0.0000	0.0130	0.0000	0.0000	0.0004
1.2727	1.1870	1.1458	0.9132	0.9463	0.4484	0.7726	0.5402	0.4990
0.0542	0.1524	0.0542	0.2101	0.1428	0.2083	0.1836	0.2548	0.1979
0.0740	0.0637	0.0553	0.0663	0.0544	0.0693	0.0682	0.0857	0.0783
0.0013	0.0006	0.0009	0.0007	0.0008	0.0005	0.0008	0.0008	0.0009
70.3642	49.8231	61.0063	63.9146	52.3844	42.6089	75.2598	78.1656	84.9818
0.0013	0.0032	0.0041	0.0019	0.0024	0.0052	0.0017	0.0018	0.0020
0.0901	0.0397	0.0396	0.0265	0.0275	0.0339	0.0194	0.0150	0.0138
0.0813	0.2287	0.0813	0.3152	0.2141	0.3125	0.2754	0.3822	0.2968
0.5821	0.7231	0.3974	0.8727	0.5666	0.9401	0.8155	0.9626	0.9968

[a]Levels in **bold** represent highest exposed population subgroup

malathion (29.7 ng/kg/day), acephate (18.4 ng/kg/day), chlorpyrifos-methyl (12.8 ng/kg/day), and azinphos-methyl (11.1 ng/kg/day). The reason that methamidophos poses a much greater exposure relative to the chronic RfD than do the other organophosphate insecticides is that it has an extremely low chronic RfD of 50 ng/kg/day. This low RfD is derived partly from the use of a 1000 fold uncertainty factor (rather than the typical 100 fold) and also from a toxicologically suspect low NOAEL of 50 μg/kg/day. Comparisons of the LD50 values for methamidophos and some of the most acutely toxic organophosphate insecticides do not indicate such disparities in toxicity.

The only other pesticides showing dietary exposure greater than 1 % of the chronic RfD were dieldrin (2.0 % of chronic RfD) and DDT (1.3 % of chronic RfD). Both of these pesticides are chlorinated hydrocarbon insecticides that have been banned from use for decades; their presence in food results from low environmental degradation and uptake from contaminated soil by plants.

Several other chlorinated hydrocarbon insecticides were also detected in the 2004–2005 TDS. To indicate trends in exposure to chlorinated hydrocarbon insecticides over time, it is possible to compare these findings with those of prior years. Table 2 shows the reduction in dietary exposure levels for two year-olds (the population subgroup associated with the greatest dietary exposure to pesticides) from dietary estimates using the 1986–1991 TDS data (Gunderson 1995) to dietary estimates using the 2004–2005 TDS data. Dietary exposures were reduced by between 47 and 96 % from 1986–1991 to 2004–2005 for the six chlorinated hydrocarbon insecticides studied.

Chronic dietary exposure estimates to the 77 pesticides for each of the 15 population subgroups is shown in Table 3. While there is significant variation in the exposure estimates for specific pesticides among the population subgroups, all exposures to all population subgroups were below chronic RfD levels. Table 3 also indicates that the highest exposures are typically observed in the younger population subgroups, as is expected since younger members of the population consume more food relative to their body weights than do older members of the population. For 51 of the 77 pesticides, population subgroup exposure was highest for two year-olds, while another 13 pesticides showed highest exposure for 6–11 month infants. Exposure to six year-olds was highest for 6 pesticides and exposure to 60–65 year-old females was highest for 4 pesticides. Three other population subgroups (10 year-olds, 14–16 year-old males, 70 year-old males) showed the highest dietary exposure to one pesticide each.

This paper provides estimates of chronic exposure to pesticides based upon mean residue levels found in the TDS and reliance on average food consumption patterns for the General US population and for population subgroups. As such, efforts were not made to assess acute toxicity risks from pesticide residue exposure to determine the likelihood that a single person's dietary exposure to a pesticide could exceed the acute RfD on a given day. More sophisticated risk assessment methods, using probabilistic models to study variability in pesticide residue levels and food consumption data, would be needed to achieve this task. Such approaches are commonly performed by the EPA, however, and pesticides are not allowed to be registered for use unless EPA concludes that the pesticides pose a "reasonable certainty of no harm" when considering potential increased susceptibility for specific population subgroups, aggregate exposure to pesticides (water, food, and residential exposure) and cumulative exposure to families of pesticides possessing common mechanisms of toxicological action such as the organophosphate insecticides. For acute pesticide exposure, a pesticide poses a "reasonable certainty of no harm" when it can be established that acute exposure for sensitive population subgroups has at least a 99.9 % chance of being below the acute RfD (EPA 2014).

Conclusions

The findings from analysis of TDS pesticide residue data confirm that while pesticide residues are frequently detected in a variety of food products, chronic dietary exposure to pesticides continues to be at levels far lower than levels considered to be of health concern. Consumer fears from pesticide residues provide the potential for consumers to reduce their consumption of fruits, vegetables, and grains, negating the positive health benefits attributed to consumption of large amounts of such foods in one's diet. Findings from this study also indicate that the potential health benefits from further reducing one's exposure to pesticide residues through purchase of organic foods may not provide any appreciable benefit given the very low level of pesticide residues consumers are typically exposed to from conventionally produced foods and the finding that organic foods commonly have been shown to contain pesticide residues as well, although at lower frequency than their conventional counterparts (Winter 2012).

While the findings from this paper represent the most recently released TDS data for pesticide residues, such findings are still at least ten years old and may not reflect pesticide residue levels that are encountered today. Food consumption data is similarly dated, representing results from dietary surveys conducted in 1994–96 and in 1998. Fortunately, the methodology to develop exposure estimates for pesticides can be easily adapted to more contemporary residue and food consumption data when such data become available. At the present time, however, the methodology used in this paper provides a convenient and comprehensive approach to estimate chronic dietary exposure to pesticides and to compare such exposures with chronic RfD levels. Unless significant changes in pesticide use patterns and food consumption behavior become evident, it is unlikely that the use of more contemporary data will alter the conclusion that chronic dietary exposure to pesticides is typically well below chronic RfD levels.

Competing interests
The author declares that he has no competing interests.

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