

Uptake of Copper (Cu), Lead (Pb), Cadmium (Cd), Arsenic (As) and Dichlorodiphenyltrichloroethane (DDT) by Vegetables Grown in Urban Environments

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Environment Protection & Heritage Council

This paper was presented
at the Fifth National
Workshop on the Assessment
of Site Contamination

Proceedings of the Fifth National Workshop on the Assessment of Site Contamination

Editors: Langley A, Gilbey M and Kennedy B

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Printed version ISBN 0-642-32355-0 Electronic (web) ISBN 0-642-32371-2

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Uptake of Copper (Cu), Lead (Pb), Cadmium (Cd), Arsenic (As) and Dichlorodiphenyltrichloroethane (DDT) by Vegetables Grown in Urban Environments

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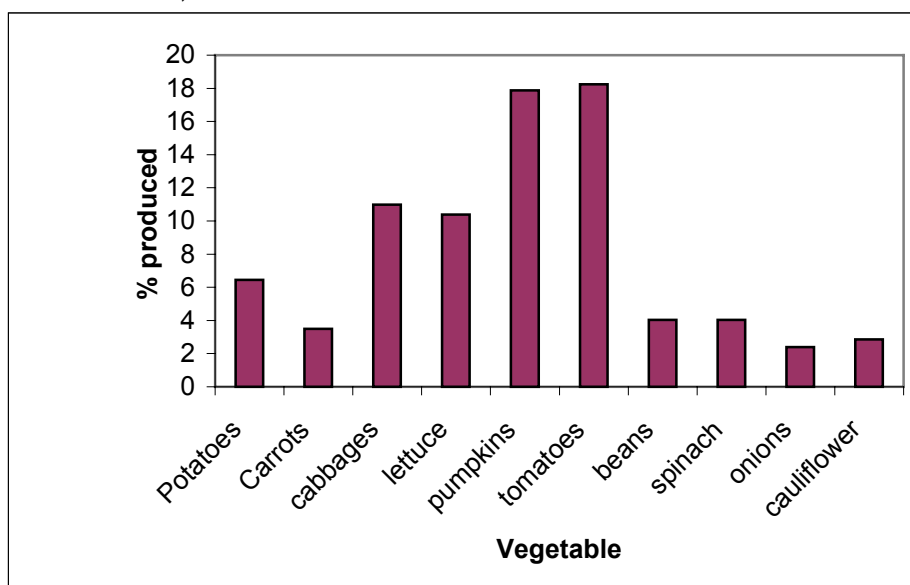
1 INTRODUCTION

In Australia one of the major pathways of adult exposure to the contaminants copper (Cu), lead (Pb), cadmium (Cd), arsenic (As) and dichlorodiphenyltrichloroethane (DDT) is through the food chain. While the main source of vegetables consumed by the majority of the Australian population is from retail outlets some groups within the population produce and consume large quantities of homegrown vegetables. This discussion details the current state of knowledge about production and consumption of homegrown vegetables by Australians and potential risks associated with vegetables grown in urban environments.

2 HOME PRODUCTION OF VEGETABLES BY AUSTRALIANS

There is very limited data on the home production of vegetables by Australians. Vegetable production by Australian home-owners was assessed over a 12 month period (1991-1992) by the Australian Bureau of Statistics (ABS 1994) and is discussed in detail in Cross and Taylor (1996). The production determined in the 1991-92 survey is now used to provide data on estimated home production in the Apparent Consumption of Foodstuffs produced annually by the ABS. The major crops grown by homeowners in the 1991-92 period is shown in Fig. 1.

Fig. 1 Percentage of main vegetables grown by homeowners in the 1991-1992 survey. (Data from ABS, 1994)



Total production of vegetables by homeowners for the 12 month period was 152,684.80 tonnes (ABS, 1994).

The most prevalent homegrown vegetables based on weight are tomatoes, pumpkins, cabbages and lettuce. The total percent of homes growing any vegetable ranged from 23.4% for ACT to 51.4% NT with the average for Australia of 35.3% (Cross and Taylor 1996).

3 CONSUMPTION OF BACKYARD-GROWN VEGETABLES BY AUSTRALIANS

The mean contribution to the adult diet in produce-growing houses was derived by Cross and Taylor (1996) based on ABS data for each state on quantity of total home food production for each vegetable type and number of houses producing those items in each state (Table 1).

Table 1 Estimated average percent contribution by mass of homegrown produce to the average daily adult diet, in households producing that food group (from Cross and Taylor, 1996).

	NSW	VIC	Qld	SA	WA	Tas	NT	ACT	Aus
Vegetable	22	22	25	23	19	35	20	19	23

This derivation assumes the homeowners consume all homegrown produce and that all household members are adult. Cross and Taylor (1996) indicate the errors in this assumption and suggest that for the purpose of risk assessment it may be more appropriate to make site-specific assessment of risk based on the wide site-specific differences in both environment and human behaviour.

When considering risk to consumers of elevated contaminant intake through the consumption of homegrown vegetables it is also necessary to consider the distribution of the contaminant through the plant and differences between vegetables in their ability to accumulate contaminants. Generally higher metal concentrations are found in leafy vegetables and root crops than in other vegetables (Tiller *et al.* 1976; Spittler and Feder 1979). There is very limited data about contaminant accumulation in herbs. An Austrian study of contamination of food found herbs had a particularly high Pb content (Pfannhauser and Pechanek 1977). A study of trace elements in meadow plants in Switzerland found cadmium accumulated in broadleaved herbs up to 8 times higher than in grasses (Stunzi 1998).

4 SOURCES OF CONTAMINATION OF CU, PB, CD, AS AND DDT IN URBAN AUSTRALIA FOR HOMEGROWN VEGETABLES

Contamination of vegetables in urban soils occurs through plant uptake of contaminants when grown on a contaminated soil and/or surface contamination through aerial deposition. Sources of contamination in Australian soils are covered in greater detail in Barzi *et al.* (1996). Generally, vegetables grown in urban soils in Australia become contaminated as a result of the following activities or scenarios:

- Use of industrial slag as residential land fill eg Windang Beach Garden Estate, NSW
- Proximity of residential areas to mines or smelters eg. Residences located near the Pasmenco refinery in Hobart, residences in Port Pirie, SA and Mt.Isa, Qld

- Proximity of residential areas to motorways. This was a greater problem before the introduction of Pb-free petrol
- Location of housing on areas that were previously used for agricultural purposes (eg As/DDT contamination of soil due to location of dip sites or orchard soils that have been contaminated with lead arsenate) or were previously zoned for industrial uses (eg decommissioned railway yard or gasworks sites)
- Contamination of soil surrounding old houses that have been painted with Pb-based paints.

In a detailed appraisal of the characteristics of the urban soils, Craul (1985) points out that soils in urban and suburban areas are frequently disturbed and subject to mixing, filling and contamination with heavy metals and organic contaminant residues. Therefore the assessment of such contaminated sites and contaminant behaviour including their phytoavailability is often a contentious issue given the wide micro and macro variability in urban soil types. Implications of fill material and leachates from such material on contaminant release and mobility and their potential for plant availability and ground water contamination are neglected areas that require much work to assist regulators to set policies. Moreover, current assessment of contaminated sites places increasing emphasis on methods that have been traditionally used for the assessment of rural agricultural soils. Such methods may not be reflective of contaminant distribution and their bioavailable fraction in urban soils.

5 FACTORS CONTROLLING PHYTOAVAILABILITY IN AUSTRALIAN URBAN SOILS

The availability of metal or organic contaminants in soil to plants is controlled by many factors, which are covered in greater detail in Adriano (1986), Brusseau and Kookana (1996), Graham-Bryce (1981), Helmke and Naidu (1996), Jones and Jarvis (1981), Smith *et al.* (1998), Weber *et al.* (1993) and Alexander (1995).

The dynamic equilibrium between metals in solution and soil-solid phase is determined by the properties of the soil and composition of the soil solution. This equilibrium in turn controls the availability of contaminants to plants for uptake. The major soil factors controlling the equilibrium are soil pH, ionic strength, and presence of cations in soil solution that may compete for sorption, presence of ligands in soil solution that may affect sorption, soil organic matter and dissolved organic material. The effects of these factors on the phytoavailability of the contaminants being discussed is summarised below (Table 2).

Table 2 Effects of soil factors on phytoavailability of Cu, Pb, Cd, As and DDT.

Soil factors	Cu	Pb	Cd	As	DDT
↓ soil pH	↑	↑	↑	↓	No effect
↑ competing cations in soil solution	↑	↑	↑	↓	No effect
↑ concentration of ligands in soil solution			↑*	↑	
↑ Dissolved organic material	↑	↑	↑	↑	↑

* Cd availability in the presence of ligands varies depending on the nature of substrate; in materials containing oxidic minerals (eg. Fe-oxyhydroxides) certain ligand ions reduce Cd availability while in some soils (eg. Xerals) commonly found in Southern Australia, ligand ions enhance Cd availability.

Soil pH is considered to be the most important factor controlling phytoavailability of certain metal contaminants, such as Cd, Pb and Cu (Tiller 1988 and 1989). Increased sorption of metal ions with increasing soil pH is due to both increased negative surface charge of soils containing large quantities of iron and aluminium oxides (Naidu *et al.* 1994) and the increased concentration of the MOH⁺ species in the soil system (Hodgson *et al.* 1964). However, for metals that form oxyanions and metalloids such as As, increasing the pH decreases their sorption by soils.

The optimal pH for a contaminated home-garden is suggested to be pH_w > 6.5 because this would ensure strong binding of most metal contaminants (Tiller 1988). However problems may be encountered with raising the pH above this level such as decreased availability of micronutrients and the onset of deficiency symptoms. Soil pH is an important factor controlling availability only for polar species that may become charged with changes in soil pH and then become sorbed to charged clay particles within the soil.

The effect of ionic strength on sorption of metal ion (M²⁺) by soils depends on the surface properties of the soil particles. Naidu *et al.* (1994) found that sorption of Cd decreased with increasing ionic strength in soils with a net negative charge. Sorption of cations by soils is a competitive process. Cadmium, Cu and Pb all form divalent cations so the presence of other divalent cations (eg Ca²⁺, Co²⁺, Cr²⁺) will retard the sorption of the contaminants of concern due to competition and hence increase their phytoavailability (Garcia-Miragaya and Page 1976). Competitive interactions have also been reported between nitrogen and As for uptake by silver beet tops (Merry *et al.* 1986).

Ligands, such as chloride (Cl⁻), sulfate (SO₄²⁻), phosphate (PO₄³⁻), and nitrate(NO₃⁻), in soil solution can affect the nature of the metal species in soil solution, which in turn can affect the bioavailability of metals in soils. Increased Cd uptake by potatoes has been observed when the irrigation water was saline, which was attributed to elevated Cl⁻ concentrations in soil solution (McLaughlin *et al.* 1994). The presence of phosphate has also affected root uptake of arsenate (Woolson *et al.* 1973). While salinity may not be an issue in most urban environments, it is well recognised that many suburban regions are used for semi-commercial vegetable production and it is here that secondary treated effluents containing significant levels of Cl⁻ and other ligands ions are being used as irrigants.

In the vicinity of plant roots the concentrations of dissolved organic material may be quite high due to root exudates and microbial activity. The role of these exudates on metal

availability requires more work but some studies indicate the presence of root or microbial exudates enhance metal uptake.

In contrast to metals, only limited information is available on the plant uptake of organic contaminants and none is related to the urban environment. Information on organic contaminant transfer from soils to growing plants especially food chain crops is critical to risk evaluation. While there is some information available on pesticide uptake by certain plant species (eg Lickfeldt and Branham 1995; Kamal *et al.* 1988; Orfanedes *et al.* 1993) research on pesticide uptake by vegetables and the mechanism of uptake is limited. Crops may become contaminated with pesticides in several ways including: direct application of the insecticide to crops leading to sorption and persistence in the plant; contamination by pesticide drift or dust particles that contain adsorbed pesticides; root uptake of pesticides and absorption of pesticides vaporised from the soil. Uptake of pesticide was related to the water solubility of the two chemicals. They found that less atrazine (5.5%) than alachlor (12.5%) was taken up by the corn plants due to the more limited solubility and mobility of atrazine in the aqueous phase. They also found that corn was able to biotransform both atrazine and alachlor in the treated plots. The portion of the pesticides taken up was proportional to the water volume transpired by the plants on the plot. Nash and Beall Jr. (1980) studied the major pathways of chlorinated hydrocarbon insecticides uptake by soybean plants using a glasshouse study. Their study was designed to permit the investigator to distinguish between the contribution of root uptake and sorption of vapours by aerial parts. They grew soybeans in nonsterile soils subjected to either a surface or a subsurface treatment of 20 mg kg⁻¹ (about 45 kg ha⁻¹) of DDT, dieldrin, endrin or heptachlor. They protected the stem from the soil by growing through a glass tube and splashing was avoided during irrigation. Following harvesting of the plants, they estimated radioactive counts for the organochlorine pesticides. They found that the major environmental source of DDT residues in the soybean plants arises from vapour movement from contaminated soil surfaces while the major pathway for the other organochlorines was primarily from root uptake and translocation through stems to leaves and seeds (Table 3).

Table 3. Root uptake (counts per minutes in thousands per gram of dry weight) versus vapour contamination of soybean foliage in chlorinated hydrocarbon (¹⁴C]DDT, [¹⁴C]dieldrin and [¹⁴C] heptachlor) treated soils (modified from Nash and Beall Jr, 1980).

Plant part	DDT	Dieldrin	Endrin	Heptachlor
Through roots				
Leaves	2.2	7.3	96.5	63.2
Stem	1.4	69.9	224.3	48.6
Pods	0.1	0.3	4.1	3.9
Seeds	0.1	0.2	1.3	1.8
Through vaporisation				
Leaves	13.7	26.8	19.3	14.5
Stem	1.5	1.5	1.5	2.1
Pods	1.1	1.2	1.1	1.2
Seeds	0.1	0.3	0.4	0.6

The plant availability of organic contaminants may be influenced by adsorption, longevity, diffusion to plant roots since these processes can affect the delivery of the organic contaminant to the plant-root interface. Root exudation and the presence of microbes in the root zone may also influence the uptake of contaminants since microbes

can degrade the contaminant molecule. These processes may in turn be influenced by processes that occur within the plant species.

The main soil factor controlling the phytoavailability of organic compounds, particularly non-polar organic chemicals, is organic matter due to the strong affinity of organic matter for non-polar organic chemicals. Soil organic matter controls the sequestration of non-polar organic chemicals by one or a combination of the following mechanisms: partitioning into soil organic matter; diffusion into and entrapment within soil nanopores; and physical sorption to soil surfaces. When soil organic matter is low mineral surfaces within soil become more important primarily through the encapsulation of the compounds that have sorbed to soil organic matter and clay particles. The clay fraction in soil can also result in the sorption of polar organic compounds due to the surface charge on clay particles.

6 RISKS TO AUSTRALIAN HOMEOWNERS

Only sections of the community that both live in an area that has contaminated soil or is subjected to aerial contamination and produce and consume large quantities of homegrown produce will be considered at risk. Although only a small proportion of the population would fit both categories there are some ethnic groups in Australia that may be deemed more at risk than other groups within the community because of their large rates of consumption of home-grown vegetable.

The recommended maximum dietary intake for As is 3 µg/kg bw/day (ANZFA 2002), for Cd 7 µg/kg bw/week (WHO 1989), for Cu 200 µg/kg bw/day (WHO 1996) and for Pb 25 µg/kg bw/week (WHO 1987). The recommended maximum dietary intake of As, Cd, Cu and Pb for a range of body weights is given in Table 4. Owing to the enormous range of metal concentrations in Australian soils the total dietary intake (TDI) has been determined for one specific case - vegetables grown in home gardens at Port Pirie. The calculation of TDI has been determined using the 95th percentile and median metal concentration in the vegetable (Table 5), the median adult (19 years and over) dietary intake and has assumed that 100% of produce consumed is homegrown. The latter is an extreme case and represents a 'worst-case' scenario.

Table 4 Recommended Maximum Dietary Intake ($\mu\text{g}/\text{day}$) for adults weighing between 50-100 kg.

Weight (kg)	As (3) ¹	Cd (1) ¹	Cu (200) ¹	Pb (3.6) ¹
50	150	50	10000	180
55	165	55	11000	198
60	180	60	12000	216
65	195	65	13000	234
70	210	70	14000	252
75	225	75	15000	270
80	240	80	16000	288
85	255	85	17000	306
90	270	90	18000	324
95	285	95	19000	342
100	300	100	20000	360

¹ Recommended maximum dietary intake ($\mu\text{g} / \text{kg}$ body weight/ day) obtained from ANZFA web site, anzfa.gov.au

Table 5 Metal concentrations in vegetables (median and 95th percentile) ($\mu\text{g}/\text{g}$ fresh weight) from Port Pirie from Tiller *et al.* (1976)

Vegetable	95 th percentile concentration		Median concentration	
	Pb	Cd	Pb	Cd
Tomatoes	0.056	0.06	0.01	0.03
Carrots	1.31	0.46	0.49	0.14
Silverbeet	1.83	0.45	0.59	0.10

Table 6 Total Dietary Intake ($\mu\text{g}/\text{day}$) determined using 95th percentile and median metal concentrations in homegrown vegetables from Port Pirie from Tiller *et al.* (1976).

Calculated using - Commodity	Median dietary intake adult ¹ (g/day)	95 th percentile concentrations		Median concentrations	
		Pb	Cd	Pb	Cd
Tomatoes, raw, fried and stewed	60.9	3.41	3.65	0.61	1.83
Tomatoes, canned or paste	100.1	5.61	6.01	1.00	3.00
Tomatoes, dried	10.7	0.60	0.64	0.11	0.32
Tomato sauce (pasta) homemade	134.4	7.53	8.06	1.34	4.03
Carrots	41.3	54.23	19.06	20.24	5.78
Lettuce and stalk vegetables	28.8	52.56	12.89	16.99	2.88

¹ Adult is defined as 19 years or over

Assuming an average male weighs 75 kg the maximum recommended dietary intake for Cd is 75 ($\mu\text{g}/\text{day}$) and for Pb is 270 ($\mu\text{g}/\text{day}$) (from Table 4). Similarly for an average female weighing 59 kg the maximum recommended dietary intake for Cd is 59 ($\mu\text{g}/\text{day}$) and for Pb is 212 ($\mu\text{g}/\text{day}$) (from Table 4). The percentage contribution of different vegetables sampled in the Port Pirie region to the maximum recommended dietary intake

is given in Table 7. Carrots and lettuce grown in the Port Pirie region with metal concentrations in the 95th percentile could potentially contribute 17-32% of an adult's maximum recommended dietary intake of Cd or Pb to residents who eat 100% homegrown produce (Table 7). Average body weights were the 50th percentile data for a 1983 National Heart Foundation study cited in NHMRC(1988).

There are many problems with attempting to assess contaminant exposure to residents including the following:

- Source and accuracy of dietary data
- Concentrations of contaminants in the vegetables collected from the area of concern will be limited by the sampling protocol (i.e. range of sampling and number of vegetables collected).
- In this case study median dietary intake for different commodities was used but this data was collected only over one day in 1995 and may not be representative of the behaviour of residents in the area of concern.
- Also the worst-case scenario has been assumed (i.e. 100% of produce consumed is homegrown). This is unlikely to occur in the majority of Australian households and again will vary with behaviour of residents in the area of concern, and seasonally with vegetable production etc.

In summary, to determine more accurately exposures to contaminants from consumption of home-produce each case must be assessed independently and the production and consumption of vegetables must be assessed for residents in the area of concern. Also it would be necessary to consider all possible exposure pathways for the residents.

Table 7 Percentage of maximum dietary intake of an average male (85kg) and an average female (65 kg) from different vegetables from the Port Pirie region.

Calculated using -	95 th percentile metal concentrations				Median metal concentrations			
	Male		Female		Male		Female	
	Pb	Cd	Pb	Cd	Pb	Cd	Pb	Cd
Tomatoes, raw, fried and stewed	1.1	4.3	1.5	5.6	0.2	2.2	0.3	2.8
Tomatoes, canned or paste	1.8	7.1	2.4	9.2	0.3	3.5	0.4	4.6
Tomatoes, dried	0.2	0.8	0.3	1.0	0.04	0.4	0.05	0.5
Tomato sauce (pasta) homemade	2.5	9.5	3.2	12.4	0.4	4.7	0.6	6.2
Carrots	17.7	22.4	23.2	29.3	6.6	6.8	8.6	8.9
Lettuce and stalk vegetables	17.2	15.2	22.5	19.8	5.6	3.4	7.3	4.4

¹ Adult is defined as 19 years or over

7 MANAGEMENT PRACTICES THAT MAY MINIMISE CONTAMINANT UPTAKE

- **Adjusting soil pH.**
Soil pH can be raised by liming and this will generally minimise the availability of contaminants that form cations (eg. Cu, Pb, Zn, Cd) but it will increase the

availability of contaminants that form anions (eg As). One problem with raising soil pH is that the phytoavailability of essential micronutrients (eg Zn, Mn, Cu) as well as contaminants will be decreased. Also the proposed pH may be unsuitable for plant growth or may be difficult to achieve.

- **Growing vegetables in raised garden beds with clean soil.**

This is a suitable management strategy only for shallow rooted vegetables and is unlikely to be effective for root vegetables. Root growth could be encouraged in the zone of clean soil by regular watering to minimise root movement to deeper soil layers in search of water.

- **Washing and peeling vegetables.**

Contaminants on soil may adhere to the vegetable particularly for root crops. Washing vegetables will remove any surface contamination. Contaminants as well as micronutrients generally accumulate in the outer skin layer (peel) of vegetables. For example Helgesen and Larsen (1998) found total As and total Cu in carrot peel was approximately 2x and 2.5x, respectively greater than in the core of the carrot. Higher Cd concentrations were found in potato peel than in the potato tuber (M J McLaughlin *pers. comm.*).

- **Restricting vegetables that are grown.**

Contaminant uptake can also be managed by restricting the vegetables that are grown. Generally higher metal concentrations are found in leafy vegetables and root crops than in other vegetables (Tiller *et al.* 1976; Spittler and Feder 1979), particularly fruiting vegetables such as tomatoes and peas.

8 SUMMARY

There are limited data about concentrations of Cu, Pb, Cd, As and DDT in Australian urban soils and in vegetables grown in urban soils. There are also very limited data about production and consumption of vegetables by Australians. As suggested by Cross and Taylor (1996) it is more appropriate to make site-specific assessments of risk to incorporate the wide site-specific differences in both environment and human behaviour. Sampling of existing or specially grown crops on the site will provide the most accurate data for risk assessments. Following the assessment of risk an information service would need to be put in place to advise on methods that could be utilised by the homeowners to minimise risk of ingesting unacceptably high levels of contaminants through the consumption of homegrown vegetables. Where management options are not likely to decrease the uptake of the contaminants by the vegetables to acceptable levels, remediation strategies would need to be considered.

ACKNOWLEDGEMENTS

The information provided by Ms Sally Record, CSIRO Health Sciences and Nutrition, and Dr Ray Correll, CSIRO Mathematics and Information Sciences are gratefully acknowledged.

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