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UPTAKE OF THALLIUM BY VEGETABLES: ITS SIGNIFICANCE FOR HUMAN HEALTH, PHYTOREMEDIATION, AND PHYTOMINING

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ABSTRACT

Eleven common vegetables (green bean, beetroot, green cabbage, lettuce, onion, pea, radish, spinach, tomato, turnip, and watercress) as well as the thallium hyperaccumulator *Iberis intermedia*, were grown in pot trials containing 0.7 and 3.7 mg/kg thallium added to a silt loam soil. The aims of the experiments were threefold: to estimate risks to human health of vegetables grown in thallium-rich soils, to demonstrate the potential of crops of these plants to remove thallium from polluted soils (phytoremediation), and finally to establish the degree to which part of the costs of remediation could be recouped by selling the extracted thallium (phytomining). Maximum thallium levels ranged from nearly 400 mg/kg (d.m.) in *Iberis* down to just over 1 mg/kg in green bean. The four vegetables with the highest

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thallium levels (watercress, radish, turnip and green cabbage) were all Brassicaceous plants, followed by the Chenopods beet and spinach. At a thallium concentration of 0.7 mg/kg in the soil only green bean, tomato, onion, pea and lettuce would be safe for human consumption. At 3.7 mg/kg thallium, only green bean and tomato could be eaten. The *Iberis* had by far the best potential for phytoremediation of thallium-contaminated land and would need 5 sequential croppings to reduce 1 mg/kg thallium to 0.1 mg/kg in the soil. By contrast rape would take 9 years and green cabbage over 30 years. Some of the costs of phytoremediation might be recouped by selling the thallium which currently has a world price of $US300/kg. It was concluded that phytoremediation of thallium-contaminated soils containing >1 mg/kg thallium will never be feasible by use of common vegetables. For soils containing 1 mg/kg thallium or less, use would have to be made of *Iberis intermedia* or *Brassica napus* (rape) rather than common vegetables.

Key words: Thallium; *Iberis intermedia*; Phytoremediation; Phytomining; Green bean; Beetroot; Green cabbage; Lettuce; Onion; Pea; Radish; Spinach; Tomato; Turnip; Watercress.

INTRODUCTION

Thallium is a highly toxic metal with a crustal abundance of about 0.7 mg/kg (ppm). Its toxicity is comparable to that of cyanide and it is used for rat poison and for the control of ants. It is also used in the electronics industry for semiconductors, switches and fuses. Thallium minerals are quite rare and are found almost exclusively in the realgar (arsenic) deposits of Allchar (Alsar) in Macedonia. High concentrations of thallium have been found in plants and domestic animals in the Allchar region (1). The current world price of thallium is about $US300/kg.

Anomalous levels of thallium in soils are derived from three main sources. The first of these is natural and is related to the geochemistry of the local rocks (2,3). The second source is anthropogenic as a result of mining of sulphide liberalization that usually contains a significant level of thallium. An example of this is the mining district of Les Malines along the southern margin of the Massif Central near Montpellier in France (4). A third significant source is the thallium derived from burning of coal at cement works in various countries and most significantly in Germany. It is probable that the contamination is derived from the raw materials used to make cement, rather than from the burning of coal in itself, otherwise significant thallium contamination would be found at all coal-burning sites.

Elevated concentrations of up to 20 mg/g (dry weight) thallium have been found in French vegetables (particularly in green cabbage) and up to 40 mg/g in rape seed (*Brassica napus* L.) (2,3,5).

Thallium has been found to be accumulated by food crops, particularly Brassicaceous plants (6,7) where up to 24 mg/kg thallium (d.m. = dry mass) was found in kale growing on contaminated soils containing only 1.4 mg/kg of this metal. It was, however, discovered that different cultivars of green cabbage have thallium levels (d.w.) ranging downwards from 24 mg/kg to as little as 1 mg/kg (6). Sequential cropping by use of cultivars with the highest thallium concentrations might be used to reduce the thallium burden of the contaminated soils to an acceptable level within 10 years.

Inordinately high accumulation (*hyperaccumulation*) of thallium was discovered in two plants from Les Malines (4,8); *Iberis intermedia* and *Biscutella laevigata,* that could contain respectively 0.4% and 1.94% thallium d.m. (8). Either or both of these species could rapidly remove the thallium burden of contaminated soils. Because of the high value of thallium, there should be some potential to recoup some of the costs of phytoremediation by recovery of thallium from the biomass.

Thallium concentrations in plants have excited the most interest from scientists concerned with potential harmful effects on animals and humans. In France, inordinately high uptake of thallium by plants of the Brassicaceae such as *Iberis intermedia, Biscutella laevigata* and *Brassica napus* has been reported (2,3,4,5,8). This high uptake clearly poses a risk to human and animal health in specific regions of Europe and possibly elsewhere.

Thallium is a relatively obscure metal that, until recently, has received little attention for its potential toxicity in animal or human nutrition. It is difficult to find specific recommendations by health authorities concerning allowable levels in foods and feedstuffs. However, a permissible range of 0.45–2.28 mg/kg thallium (d.m.) for animal feed has been proposed for Germany (9). A value of 0.25–0.5 mg/kg (f.m. = fresh mass) has been proposed (10) for human foods. Assuming a 12% conversion factor for f.m. to d.m., the recommended value is therefore about 2–4 mg/kg d.m. for humans. It is our opinion that this level is too high in comparison with levels set for animal feeds and that the former standard should also apply to human nutrition. It is clear from the published data that most crops grown in thallium-contaminated soils will have unacceptably high concentrations of this toxic metal.

The aim of the work presented below was to obtain independent data for thallium levels in common New Zealand vegetables intended solely for human consumption and to assess their risk for human consumption. Two further aims were to examine the possibility of using thallium-uptake by crops of these plants for phytoremediation (removal of pollutants by plants) and phytomining (growing a crop of a metal).

Since naturally occurring high-thallium soils were not available to us, we prepared mixtures of thallium added to a local silt loam. We also grew *Iberis intermedia* in these mixtures in order to serve as a control because we had already studied wild plants of this species growing under natural conditions at Les Malines in southern France and could thereby obtain a factor to relate apparent concentrations of thallium in pot-grown plants to those to be expected in vegetables grown under similar wild conditions.

MATERIALS AND METHODS

The names of plants used in the present study are shown in Table 1. They include *Iberis intermedia* and 11 vegetables commonly grown in New Zealand.

Plants were grown in the local *Manawatu Silt Loam* to which was added approximately 1.0 and 5.0 mg/kg (ppm) thallium as the sulphate. Thallium-free controls were also used. The top 15 cm of soil had a pH of 5.7, total organic carbon 6.3%, and exchange capacity of 13.4 meq/100 g. The P concentration was 30 mg/kg. The soils contained 3–4 month Osmocote slow release fertilizer granules at a rate recommended by the manufacturer. The soils were placed in 1 L plastic pots and allowed to stand on damp felt for two weeks with constant watering and reanalyzed before planting to give the equilibrium concentration of thallium left bound to the soil (largely in the humus) after the unbound thallium

Table 1. Species and Cultivars Used in Experiments

Common Name	Species	Family	Cultivar/Trade Name
Green bean	*Phaseolus vulgaris*	Fabaceae	Tendergreen
Beetroot	*Beta vulgaris*	Chenopodaceae	Derwent Globe
Green cabbage	*Brassica oleracea*	Brassicaceae	Early Ball
Iberis	*Iberis intermedia*	Brassicaceae	Iberis
Lettuce	*Lactuca sativa*	Asteraceae	Great Lakes
Onion	*Allium cepa*	Liliaceae	Pukekohe Longkeeper
Pea	*Pisum sativum*	Fabaceae	Greenleaf
Radish	*Raphanus sativus*	Brassicaceae	Salad Crunch
Spinach	*Spinaceae oleracea*	Chenopodaceae	Winter Queen
Tomato	*Lycopersicon esculentum*	Solanaceae	Groslisse
Turnip	*Brassica rapa*	Brassicaceae	Purple Top
Watercress	*Nasturtium officinale*	Brassicaceae	—

had been leached from these substrates. Seedlings of all 12 species were raised separately in commercial seed-raising mixture and then transplanted into the large pots. The plants were allowed to grow for 12 weeks in a greenhouse maintained at 15°–25°C with no humidity control. Pots were watered as required (usually every second day) and the pots switched around in a random manner to allow for equalization of light intensity. Five replicates for each treatment and for each species (180 in total) were prepared. After the 12-week period, the plants were harvested and dried at 60°C before analysis. At harvest time, composite samples of the substrate were collected and dried before analysis.

Vegetation and soil samples were digested with concentrated nitric acid on a hotplate (0.5 g samples + 10 mL of acid) and evaporated to low volume (ca. 1 mL). The samples were then diluted to about 15 mL with distilled water and analyzed for thallium either by flame atomic absorption spectrometry (FAAS) for samples with higher thallium concentrations, or by graphite furnace atomic absorption spectrometry (GFAAS) for lower concentrations of thallium. Care was taken to avoid the use of hydrochloric acid because the volatility of thallium in the presence of chloride would have precluded the use of GFAAS for analysis.

RESULTS AND DISCUSSION

Experimental Data

Thallium concentrations in whole plants of *Iberis intermedia, Brassica juncea* and in edible parts (roots or tops depending on plant) of 11 common vegetables are shown in Figure 1. The biological absorption coefficients (BAC = plant/substrate concentration quotient) for thallium in all the test plants are shown in Table 2 as their maximum values. It is noteworthy that whereas *Iberis intermedia* in its natural environment over thallium-rich tailings in southern France (Les Malines) had a mean of 1190 mg/kg thallium (d.m.), maximum of 3070 mg/kg, and mean BAC of 74 (4), the respective values for the pot-grown plants were 281, 408, and 76 for a potting medium with 3.7 mg/kg thallium. There is close agreement between the two BAC values. The higher means and maxima for the French plants result from the much higher thallium concentration (16 mg/kg) in the soil.

It will be observed from Fig. 1 that the four vegetables with the highest maximum or mean thallium levels, watercress, radish, turnip and green cabbage are all Brassicaceous. Analogous experiments have been carried out (6) with various vegetables including green cabbage for which a mean of 13.7 mg/kg thallium was reported when grown in a soil containing 1.4 mg/kg thallium.

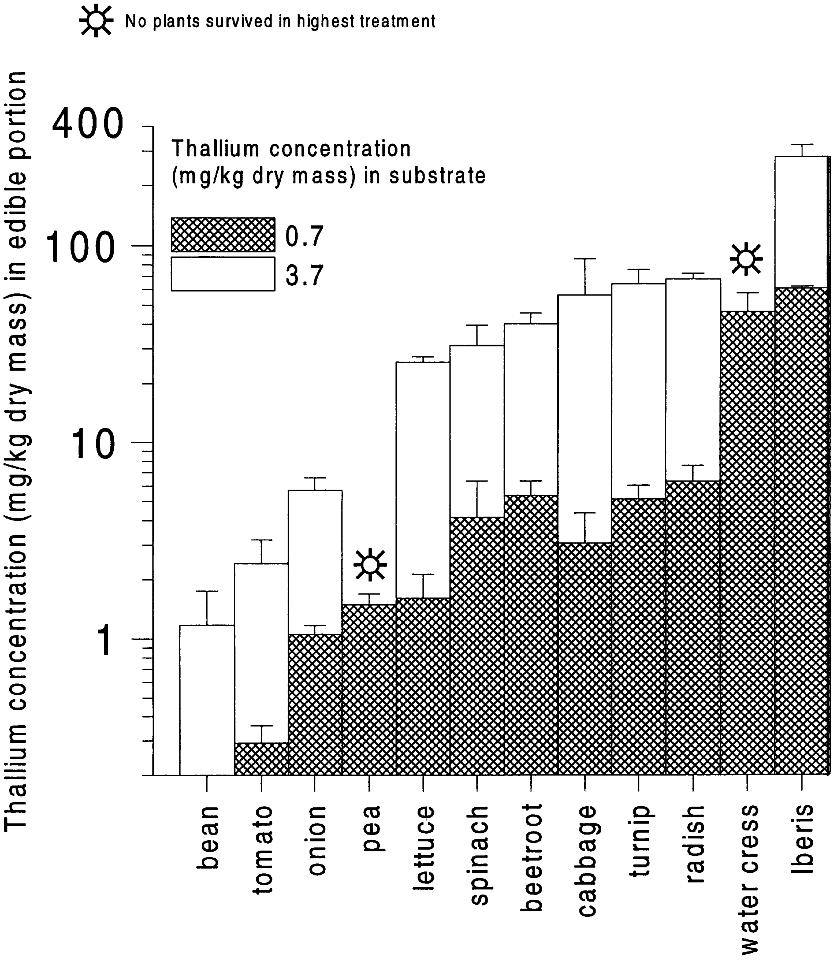

Figure 1. Thallium concentrations in vegetables and in the thallium-hyperaccumulator *Iberis intermedia* when grown in loam with added thallium.

We found 3 mg/kg thallium in green cabbage growing in 0.7 mg/kg thallium and 58 mg/kg in 3.7 mg/kg thallium. The results are therefore somewhat comparable.

Watercress when grown in a substrate with 0.7 mg/kg thallium, accumulated almost as much thallium (48 mg/kg) as the *Iberis,* but did not survive at the higher metal concentration. A similar behavior has been observed (11) for the

Table 2. Maximum Values of Biological Absorption Coefficients (BAC)* for Thallium in Plants

	0.7 mg/kg Thallium in Substrate		3.7 mg/kg Thallium in Substrate	
Plant	Max. Thallium**	BAC	Max. Thallium**	BAC
Green bean	n.d.	n.d.	3.49	0.94
Beetroot	8.75	12.5	52.0	14.0
Green cabbage	7.85	11.2	129	34.9
Iberis intermedia	60.9	87.0	408	110
Lettuce	3.1	4.42	30.8	8.32
Onion	1.42	2.02	8.44	2.28
Pea	1.82	2.60	n.s.	n.s.
Radish	10.4	14.9	70.0	18.9
Spinach	15.9	22.7	48.7	13.2
Tomato	0.53	0.76	4.95	1.33
Turnip	7.8	11.1	139	37.6
Watercress	84.4	121	n.s.	n.s.

*Plant/soil thallium concentration quotient.
**Thallium concentration in plant (mg/kg dry mass).
n.d. —not determined; n.s. —no survival.

uptake of arsenic by this plant. Over 300 mg/kg d.m. was found in plants growing near a geothermal source in the Waikato River, New Zealand.

The next highest thallium levels were found in the Chenopods beet and spinach. There were low levels of thallium in pea and green bean (Fabaceae) and in the tomato (Solanaceae). In the pea and tomato the edible parts are further distant physiologically from the rhizosphere than are leaves, so that these lower thallium levels may be a reflection of this greater isolation whereby translocation of thallium to the fruits was less than to the leaves.

Health Hazards of Ingestion of Thallium-Rich Vegetables

The health hazards inherent in consuming vegetables growing in thallium-rich soils are difficult to establish because of the lack of data on long-term effects of thallium ingestion by humans. There are many reports on the clinical effects of poisoning of humans by exposure to, or ingestion of, thallium (12). There are, however, few sources that pinpoint the acceptable daily intake (ADI) of thallium by humans, a toxic metal described as a new environmental contaminant (10). The toxic effects of thallium on humans are linked partly to the readiness whereby Tl^+ substitutes for K^+ as an activator of some reactions catalyzed by enzymes. The monovalent thallium ion has an especial affinity for SH- groups and can cause disorders of metabolic or energy processes in cells of animals.

In Europe (9) a maximum thallium level of 0.46–2.24 mg/kg d.m. for animal forage has been proposed. If we adopt the same maximum level of about 2 mg/kg d.m. thallium in human food as being acceptable for human health, it is clear from our data (Fig. 1) that only green beans, tomato, onion, pea and lettuce would be safe to eat if grown in soils containing 0.7 mg/kg thallium. However, it has been shown (7) that under field conditions, levels of thallium in vegetables were usually only one third of those found in pot-grown plants due to a lower root density in the field. Therefore, if the thallium levels in Fig. 1 are reduced to a third to make the conversion from pot-grown to field-grown plants, all of the vegetables except for radish and watercress could be added to the list that are safe to eat. For highly contaminated soils with 3.7 mg/kg thallium, only green bean, and tomato would have permissible concentrations of thallium. Although we do not have values for vegetables grown at various thallium concentrations between the limits of 0.7 and 3.7 mg/kg in the soil, extrapolation of the data indicates that even at a thallium concentration of 1 mg/kg, most of the vegetables and certainly spinach, beetroot, green cabbage and turnip would be unfit for human consumption.

Phytoremediation of Thallium-Contaminated Soils

The use of vegetables and other Brassicaceous plants as a means of phytoremediation of thallium-contaminated soils has already been proposed (4,6,7,8,13,14). It has been shown (6) that he cropping of rape (*Brassica napus*) and green cabbage in one growing season together, removed 6–8% of the thallium burden from the top 30 cm of contaminated soil. Using the high biomass *Iberis intermedia* growing over thallium-contaminated mine tailings at Les Malines in southern France, it was shown (4) that this plant with its very high thallium concentration (1190 mg/kg d.m.) could remove most of the thallium from the substrate in only three croppings. This does however presuppose that thallium yields were the same after each cropping. During three sequential croppings with green cabbage, thallium removal decreased from 50 mg/kg of soil in the first year to 20 mg/kg in the third (6).

From our own data (Table 3) it was possible to calculate annual removal of thallium from field crops of the plants used in our experiments. These experiments had been carried out as pot trials and it had been shown that field crops usually gave only one third of the annual removal of thallium compared with pot trials with their greater density of root development (6). Our calculated value of 81 g/ha of thallium removed by green cabbage (i.e., one third of the mean for the two sets of experiments) is within the range of 59–133 g/ha reported for

Table 3. The Potential of Plants for Phytoremediation of Thallium-Contaminated Soils

	3.7 mg/kg Thallium in Substrate			0.7 mg/kg Thallium in Substrate			
Species	A	B	C	D	E	F	G
Iberis	10.0	300	3000	60	600	1800	600
Radish	0.9	68	61	6.0	5.4	33	11
Turnip	3.6	64	230	5.3	19	125	42
Green cabbage	8.4	55	462	3.0	25	244	81*
Beet	6.7	42	281	5.5	37	159	53
Spinach	5.4	33	178	4.2	23	100	33
Lettuce	6.0	25	150	1.8	11	80	27
Onion	5.8	33	1.2	1.2	7	20	7
Tomato	9.0	2.5	22	0.1	0.9	12	4
Green bean	1.6	1.2	2.0	—	—	2.0	0.7
Pea	1.1	—	—	1.6	1.8	1.8	0.6

*Literature value (6) was about 60 g/ha.

A = biomass (tonnes/ha) (9), B = mean thallium concentration in mg/kg d.m., C = thallium removed (A × B) in g/ha, D = as B, E = as C, F = mean of C + D, G = F divided by 3 to make allowance for lower yields under field conditions (Kurz et al. 1997).

the same plant (6). Among the plants studied by us, only green cabbage, turnip, red beet and spinach would have potential for phytoremediation of thallium-contaminated soils because of their combination of high biomass and higher thallium concentrations. The *Iberis* has by far the best potential for phytoremediation (9). Our value of 600 g/ha is over double the 298 g/ha reported for *Brassica napus* (rape) (6).

A hectare of soil with a depth of 15 cm and density of 1.3 contains about 2000 tons of material. If the concentration of thallium is 1 mg/kg, the soil would contain 2000 g of thallium. A crop of *Iberis* would remove 1800 g of thallium over three years: leaving only 0.1 mg/kg thallium. A crop of green cabbage would take 22 years to achieve the same degree of remediation and a crop of spring rape (6) would achieve the same goal in six years. These calculations have assumed that there is no loss of thallium yield with successive crops. Allowing a further 50% of croppings to allow for such a loss, the *Iberis* would require up to 5 years, the rape 9 years and the green cabbage over 30 years. The following conclusions can be made from these calculations: 1) phytoremediation of thallium-contaminated soil is impracticable by use of common vegetables;

2) phytoremediation will only be economically feasible by use of plants such as *Iberis* or possibly rape.

Phytomining for Thallium

The fundamental difference between phytomining and phytoremediation is that the former operation (14) involves growing a crop of a metal for monetary return. The world price of thallium is currently around $US300/kg with world production almost entirely from one mine in Macedonia. It has been proposed (4) that it should be possible to produce 8 kg of thallium (worth $US2400) from a hectare of land containing 10 mg/kg thallium and planted with *Iberis intermedia*. If phytomining were to be combined with phytoremediation, it is obvious that much lower thallium concentrations would be treated: i.e., of the order of 1 mg/kg rather than 10 mg/kg. Using this scenario the hectare of *Iberis* yielding 600 g/ha thallium (Table 3) would produce a crop worth about $US180 (about half the return to an American farmer of a crop of wheat), whereas green cabbage would yield only $US40. The operation would obviously be uneconomic *per se,* particularly as it does not include the cost of removing the thallium from the biomass. It would however, serve to recoup some of the costs of phytoremediation. It must also be remembered however, that although the world price of thallium is very high, there is a very low demand worldwide for this metal and any sudden increase of supply from phytomining is likely to have a significant effect on the price and thereby reduce the attractiveness of the phytomining option.

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