CRITERIA FOR THE EVALUATION OF DIOXINS IN VEGETABLE PLANTS AND SOILS

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ABSTRACT

In FRG and other countries unequivocal criteria for the limitation of dioxins (PCDD's/PCDF's) in food like vegetables and fruits are lacking. These have to be directly associated with the limitation of dioxins in the soil and the deposition of particulate matter as the two main pathways for plant contamination. Based on recent investigations in the vicinity of cable-waste incinerators in Northrhine-Westfalia with comperatively high contents of dioxins in garden plants and soils, considerations are given for the establishment of the criteria urgently needed.

KEYWORDS

Chlorinated dibenzodioxins and dibenzofurans, plant uptake, soil content, transfer-factor soil/plant, particulate matter, bioindicator, theshold values.

INTRODUCTION

The contamination of soils and garden-grown vegetables by chlorinated dibenzodioxins and dibenzofuranes in the vicinity of cable-waste incinerators was investigated during 1989 by the Landesanstalt für Immissionsschutz (LIS), Essen, FRG. According to *screening investigations* the situation in Dortmund was especially critical, since the allotment gardens *Hobertsburg*, *Westerholz* and *Hafenwiese* were closely neighboured to the incinerator plant. In order to differentiate the two main pathways of plant contamination *soil/root* and ambient air/leaf organ, samples of potatoes, carrots, silver beet, and salat were taken from allotment gardens together with corresponding soil samples on July 4.,1989 and analysed for PCDD/PCDF respectively. Sampling depth ranged between 0 and 20 to 30 cm. To test the representativeness of the sampling procedure and the analytical quality additional samples of each plant-species as well as soil sample were taken from each allotment, mixed and and analysed. Thus analytical results of the unified samples could be compared with the calculated average of the three allotment samples. Reference samples of soil and the same plant species were taken from a farm operating with biologic-dynamic cultivation methods in a "clean air region" near Münster, 80 km northeast of the Ruhr area. All analyses were carried out by the Gesellschaft für Arbeitsplatz- und Umweltanalytik (GFA) in Münster-Roxel.

Together with the environmental agency of the city of Dortmund additional plant species like cucumber, beans, endive, plum, strawberry, apple, kohlrabi, red beet, savoy cabbage, and kale were sampled in the same allotments on August 9. and October 8. 1989. During this time samples from the allotment garden areas *Fredenbaum*, *Hansa*, and *Nord*, located 1.5 km north of the cable-waste incinerator were included.

In oder to evaluate the question of the contaminating source (soil or ambient air) for the analysed PCDD/PCDF-contents in plants, standardized grass cultures were exposed from August to October 1989 in the allotment garden areas of *Hobertsburg*, *Westerholz*, and *Hafenwiese* as well as on four other selected sites at the corner points of a 1 km square with the incinerator as centre. According to the method by Scholl (1971), gras of the species Lolium multiflorum is cultivated under standardized procedure

carrots to have a specific affinity for the accumulation of lipophilic organic substances.

CONSIDERATIONS ON REGULATORY CRITERIA

Based on the report of the Enmvironmentral Agency of the Federal Republic of Germany (Umweltbundesamt=UBA, 1984) an acceptale daily intake value (ADI) of 1 to 10 pg TE (kg body weight)$^{-1}$ is recommended. This value is derived from a NOAEL-value (no observed adverse effect level) of 1 ng TE (kg body weight)$^{-1}$ d^{-1} in animal experiments. This includes a safety factor of 100 to 1000. This procedure was recently confirmed by the Bundesgesundheitsamt in two consultancy reports (BGA, 1988; BGA, 1989). Due to the ubiquity of PCDD/PCDF in the environment, its accumulation in fatty tissues of animal nutrients as well as in human fatty tissue, including the breast milk, a major contamination source for sucklings, it seems to be advisable to limit the uptake of dioxins, which preferentially is via ingestion and only to a very limited extent via inhalation (Beck et al., 1989), to the lower value of the ADI rate quoted above.

Taking this value, the maximum tolerable content of PCDD/PCDF in vegetables and fruits grown in house gardens can be derived. Under the assumption of an average adult body weight of 75 kg and an average daily vegetable/fruit consumption of 250 g fresh weight with 10 % dry substance, the maximum tolerable PCDD/PCDF-content is 3 pg (g DS)$^{-1}$ calculated as toxicity equivalents (75 kg : 250 g x 1 pg = 3 pg g^{-1} DS). Taking this value, a transfer factor of 0.1 for the uptake soil/plant according to our results and a 50 % contribution to the maxium tolerable value in plants via this uptake mode, 15 ng TE kg^{-1} DS can be considered as a prophylactic guidevalue for limiting PCDD/PCDF in soils. This value should generally not be exceeded. As value asking for further action in the sence of limiting agricultural and horticultural use 30 ng kg^{-1} kg DS are proposed.

This prophylactic guidevalue of 15 ng TE kg^{-1} DS should also be taken as basis for deriving a maximum deposition value of PCDD/PCDF as particulate matter. So far no exact data are available on the half life time of PCDD/PCDF in soils. However, persistency is considered great and therefore complete accumulation is assumed for an exposure period of 100 years. Assuming a soil density of 1.3 t m^{-3} and a mixture depth for soils in garden areas of 25 cm, the maximum deposition value for particulate matter would calculate to 0,1 ng m^{-2} d^{-1}.

However, sofar no data are exsisting between total deposition and plant uptake. From earlier investigations at the LIS, concerning heavy metals, a ratio of 0.1 ng g^{-1} DS in leaves corresponds with 1 ng m^{-2}d^{-1} deposition rate. But uptake mechanisms for heavy metals (only soluble salts penetrate the leaf cuticle) are probably different from those for organic molecules such as PCDD/PCDF (high affinity to cuticular wax layer) and thus the ratio for PCDD/PCDF can be higher or lower than stated above. However, preliminary results from analysis of PCDD's/PCDF's in the leaf and deposited particulate matter indicate a range between 0.03 and 0,10 ng m^{-2}d^{-1} as safe value for limiting the deposition of dioxins in particulate matter. However, further investigations should elucidate this very point more exactly.

ACKNOWLEGEMENT

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Figure 1 PCDD/PCDF concentration [ng kg^{-1} DS] in vegetable plants and soils from three allotment garden areas in the vicinity (a,b) and at further distance (c) of a cable-waste incinerator plant in Dortmund, FRG as well as reference site near Münster (c).

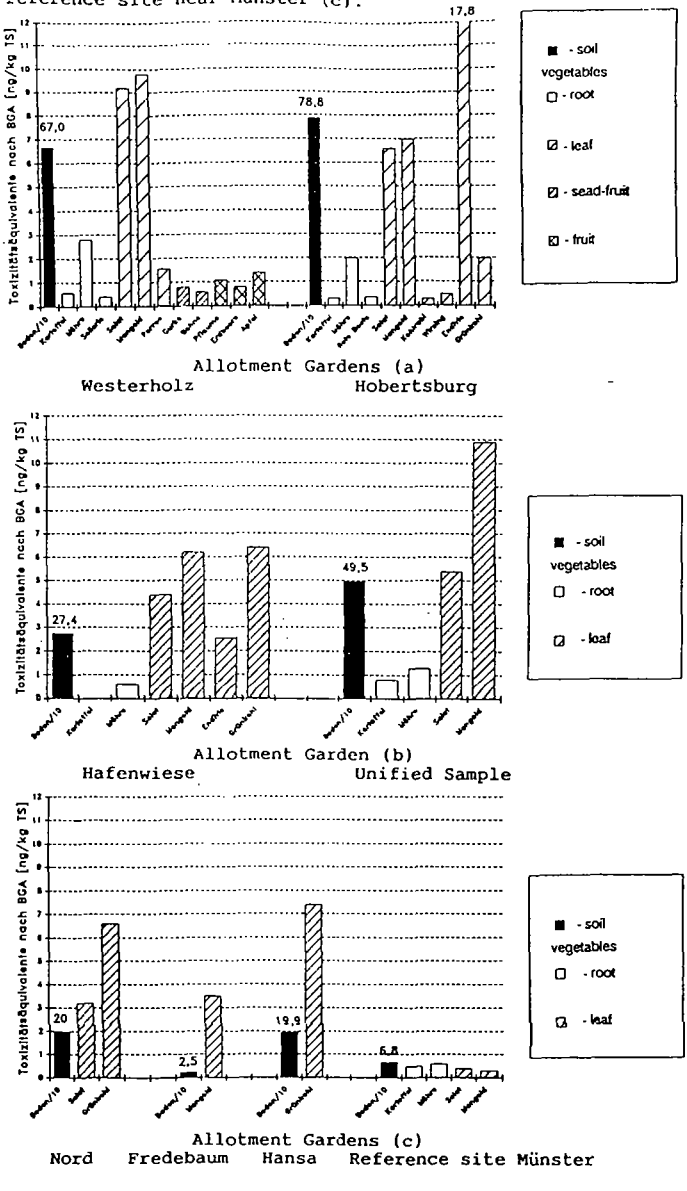
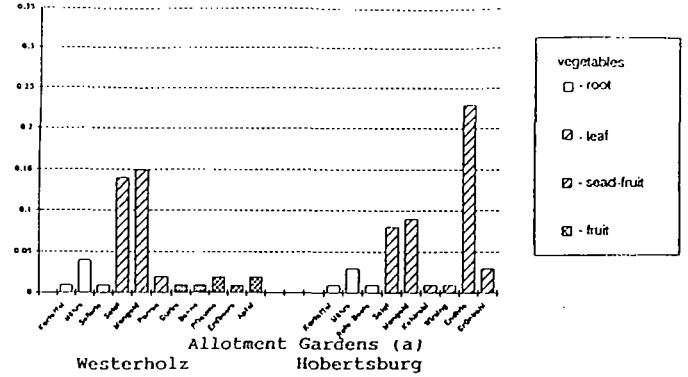
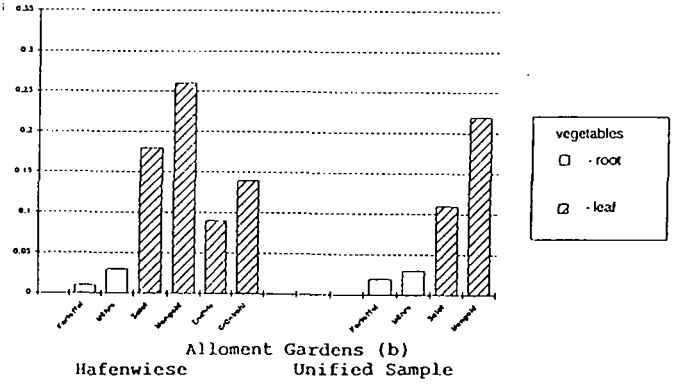
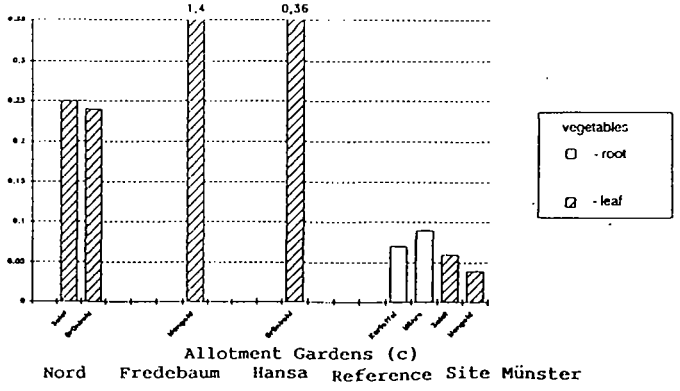


Figure 2 PCDD/PCDF transfer factors (uptake path soil/plant), expressed in TE according to the Bundesgesundheitsamt, for three allotment garden areas in the vicinity (a, b) and at further distance (c) of a cable-waste incinerator plant in Dortmund, FRG as well as reference site, near Münster (c).







analysed. Analyses were carried out by the GFA, Münster. Using this bioindicator method, PCDD/PCDF uptake by plants from soil can be excluded with highest probability.

ANALYTICAL RESULTS

In Fig 1a, b, c the analytical results of the various plant and soil samples are summarized. In order to represent all values within the same figure, the soil data were divided by a factor of 10. *Leaf* vegetables like salat and silver beet show much higher PCDD/PCDF-contents than the *root* vegetables potato and carrot. Highst accumulation was found in green kale and endive. These results were in a way contratictory to the exspectation that the soil functions as major source for plant uptake of PCDD/PCDF. Since the uptake of organic molecules via roots and systemic transport to leaf organs is generally very low (Harms, 1989), contamination risks for leaf vegetables were considered as more or less unimportant in comparison to root vegetables. However, PCDD/PCDF levels in cucumbers, beans and the fruit species plum, strawberry, and apple are very low and in so far in good agreement with uptake and distribution phenomena of heavy metals in plants.

Interestingly, relative high contents of PCDD/PCDF in soil and plants were not only found in the allotment garden areas close to the cable-waste incinerator but also in those areas located in the north of Dortmund, an overall highly industrialized zone. However, even at the reference sampling point near Münster, PCDD/PCDF content of the soil was 6 ng TE kg^{-1} DS (TE=toxicity equivalents; DS=dry substance), thus exceeding the recommended prophylactic value by the Health Agency of the Federal Republic of Germany (Bundesgesundheitsamt=BGA) for unrestricted agricultural and horticultural use of 5 ng kg^{-1} DS (BGA, 1988; BGA, 1989). The results of PCDD/PCDF contents in grass cultures are presented in Table 1, confirming that the northern parts of Dortmund show a relative high degree of contamination. PCDD/PCDF contents are in a similar order of magnitude as leaf vegetables from these locations. This can be taken as a further proof that the uptake pathway via ambient air/plant is of greater importance than via soil/plant.

Table 1 PCDD/PCDF content in leafs of standardized grass culture [ng TE kg^{-1} DS]. Mean from 14 day exposure periods (10.08. - 19.10.1989)

Allotment garden areas	Hafenwiese	7.2
	Westerholz	5.8
	Westerholz	7.5
	Hobertsburg	9.7
Corner points of 1 km^2 square with incinerator as center	Southwest	7.7
	Southeast	8.4
	Northwest	6.6
	Northeast	8.6

In Figure 2 a, b, c1 the ratios of PCDD/PCDF-content in plants are related to PCDD/PCDF-content in corresponding soils. It can be concluded - at least for the samples taken in the northern parts of Dortmund - that PCDD/PCDF contamination in leaf vegetables is much higher than could be expected from the soil contents under consideration of the results from root and bulb vegetables. Similar ratios were also found at the reference site near Münster. Summarizing the data, it is important to note, that the transfer factor for the pathway soil/plant is in any case equal or below 0.1. König and Hembrock (in preparation) could show simililar transfer rates for corn over a relative wide range of soil contents (0 to 10 ng TE kg^{-1} DS). Although within this relationship, carrots seem to show a somewhat higher affinity to accumulate PCDD's/PCDF's than other root vegetables. This is in agreement with results from Geyer et al. (1987) and Harms (1989) finding

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Explanations to the Figures:

German	English
Apfel	apple
Boden/10	soil/10
Bohne	bean
Endivie	endive
Erdbeere	strawberry
Grünkohl	kale
Gurke	cucumber
Kartoffel	potato
Kohlrabi	kohlrabi
Mangold	chart
Möhre	carrot
Pflaume	plum
Porree	leek
Rote Beete	red-beet
Salat	salad
Wirsing	savoy
Toxizitätsäquivalent	toxicity equivalents
Bundes Gesundheits Amt (BGA)	Federal Health Office of F.R.G.
trocken Substanz (TS)	dry substance (DS)