Sewage sludge land application study

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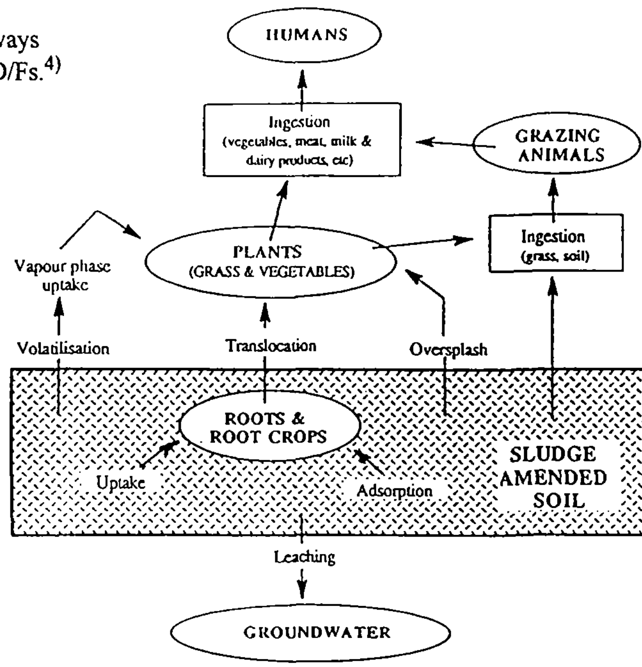
Abstract
On the request of the US Conference of Mayors, concentrations and time trends of polychlorinated dibenso-*p*-dioxins/furanes (PCDD/Fs) in US sewage sludge were studied. Also, the transfer of PCDD/Fs to plants was discussed, together with the transfer of PCDD/Fs to cattle. Incremental human exposure to sewage sludge-derived PCDD/Fs was calculated. Based on available data, concentrations of PCDD/Fs in sewage sludge over time are decreasing. The uptake of PCDDFs from soil into plants is considered negligible. Transfer to grazing cattle can occur through soil ingestion, but the main potential exposure route is through ingestion of sewage sludge stuck to the grass, following spraying of sewage sludge onto pasture. It is concluded that tilling of sewage sludge into soil results in only very minor exposure to PCDD/Fs, while spraying of sewage sludge onto pasture, whithout a prolonged waiting time before cattle are allowed to graze, could result in substantial increases in exposure. To minimize human exposure to sewage sludge-derived PCDD/Fs most efficiently, any measures taken should focus on application method, rather than on application rate.

Introduction
Sewage sludge, the residue obtained when waste water is treated, is rich in nutrients and organic matter and can be used for soil improvement and fertilization. However, sewage sludge also contain environmental pollutants such as PCDD/Fs. This paper is based on a report commissioned by the U.S. Conference of Mayors [1)], where sewage sludge concentrations, incremental soil concentrations, transfer to plants and animal tissues, and subsequent human exposure to PCDD/Fs were investigated.

Environmental media as well as biological samples contain varying mixtures of PCDD and PCDF congeners of varying toxicity. To facilitate comparisons of different sources or environmental media in which the congener mixtures vary, it is important that such mixtures of dioxins and dibenzofurans be assessed in a scientifically sound and standardized manner. In an attempt to achieve these objectives, the TEF/TEQ approach has been adopted internationally. Numerical Toxic Equivalent Factors (TEFs) have been developed, which enables the conversion from the concentrations of mixtures of 2,3,7,8-substituted congeners to toxic equivalent (TEQ) concentrations of the most potent congener, 2,3,7,8-tetrachlorodibenso-*p*-dioxin.[2)] The TEF approach can be used to transform analytical results of total sum of all PCDDs and PCDFs into toxic equivalents (TEQs).

For sewage sludge-derived PCDD/Fs to reach humans, the PCDD/Fs must be transfered from the sewage sludge to plant or animal tissues ingested by humans. As evident by Figure 1, there are many possible pathways. These were all investigated, using data from published studies, and/or estimations of transfer potentials based on physicochemical properties of PCDD/Fs. As a basis for all calculations, a representative mean TEQ sewage sludge concentration was derived from studies on PCDD/F sewage sludge concentrations in the US.[3]

Figure 1. Possible transfer pathways for sewage sludge-derived PCDD/Fs.[4]



Results and discussion

Concentrations of PCDD/Fs in samples of US sewage sludge

The National Sewage Sludge Survey (NSSS), conducted by the US Environmental Protection Agency between July 1988 and March 1989, is by far the most extensive study on PCDD/Fs in US sewage sludge. In a recalculation of these data, the median TEQ value in the NSSS was found to be 13.9 ng/kg d.m.[3] This value has been used in the calculations in the present study. It might represent the high end of the spectrum, because the samples analyzed in the NSSS are almost ten years old, and during the last decade, a series of studies has identified a downward time trend in the concentrations of PCDDs and PCDFs in several environmental compartments.[5,6]

Increased soil concentrations of PCDDs and PCDFs due to spreading of sewage sludge

Following sewage sludge application, the resulting increased soil concentration of PCDD/Fs will depend on the concentration of PCDD/Fs in the sewage sludge, the amount of sewage sludge spread per surface unit, and if (and how) the sewage sludge are incorporated into the soil. The resulting increases of soil PCDD/F concentrations was calculated using four different scenarios (see Table 1).

Persistence (t½) of PCDDs and PCDFs from sewage sludge in soil

Although the fate and behavior of PCDD/Fs in soil is still not adequately understood, it is generally agreed that PCDDFs have half-lives in soil of 10 years or more. Waiting periods before grazing or harvesting of animal fodder crops are usually 30 days or less (Jane B. Forste, Wheelabrator Bio Gro Division, pers. comm.), which will have only minor effects on the soil concentrations of sewage sludge-derived PCDD/Fs. Subsequent calculations for transfer to plants and livestock therefore assume that there is no breakdown of the PCDD/Fs.

It should be emphasized that any TEQ value given to sewage sludge, or to soil after sewage sludge application, will be incorrect following a period of time in soil, since the different congeners behave differently in soil.

Table 1. Possible sewage sludge application scenarios, and the resulting increase in PCDD/F soil concentration. The calculations are based on a TEQ concentration of 14 ng/kg d.m. sewage sludge[3]. It is assumed that the application rate is 7 metric tonnes d.m. sewage sludge/ha (Jane B. Forste, Wheelabrator Bio Gro Division, pers. comm.), that tilling and spraying lead to mixing of sewage sludge into 15 and 1 cm of soil, respectively, and that soil density is 1 g/cm^3. [7] Mean background TEQ values for US urban and rural soil are from the US EPA Dioxin Reassessment.

Application/ soil depth	Pasture location	Background soil TEQ conc. (ng/ton d.m. soil)	Addition from sewage sludge (ng/ton d.m. soil)	Resulting increase (%)
tilling/15 cm	urban	19 600	65	0.3
"	rural	4 000	"	1.6
spraying/1 cm	urban	19 600	980	5
"	rural	4 000	"	24

Transfer to plants
Uptake from soil into below- and above-ground plant parts seems to be very low or negligible.[7] Atmospheric deposition of TEQ is a much more important contributor to above-ground plant parts.[7] Direct contamination of above-ground plant structures is considered under the section below.

Transfer to livestock (dairy and beef cattle)
Since transfer of PCDD/Fs from soil to above-ground plant parts is assumed to be negligible, only soil ingestion and ingestion of grass/food crops sprayed with sewage sludge could result in exposure to PCDD/Fs originating from sewage sludge.
Stabled cattle could be exposed to PCDD/Fs originating from sewage sludge if fed harvested fodder crops to which sprayed sewage sludge still adhered. However, grazing cattle are probably more likely to be exposed to PCDD/Fs originating from sewage sludge if PCDD/Fs from sprayed sewage sludge still adhered to the pasture, or if the soil that grazing cattle ingest as part of their diet contains PCDD/Fs. The resulting grazing cattle TEQ intake was calculated using different scenarios (see Table 2). Also, since a substantial amount of soil is consumed by grazing cattle as part of their diet,[8,9] the TEQ intake via soil using different scenarios was calculated (see Table 3).

Transfer to milk and beef
For absorption of PCDD/Fs from the gastrointestinal tract and accumulation in tissue and milk fat, the degree of chlorination of the congeners is of critical importance. Increased chlorine substitution leads to lower uptake in the intestine[10] and reduces the likelihood that the substance will be secreted with the milk.[8] BCFs (calculated or measured) for transfer from fodder to both meat and milk of all 2,3,7,8-substituted dioxins and furans have been published.[8] Ideally, one should calculate values for each congener separately. However, for practical reasons, we used the same carryover rate/biotransfer factor for all congeners (see Table 4).

Table 2. Grazing cattle TEQ ingestion resulting from sewage sludge adhesion to plant parts, following spraying of sewage sludge onto pasture. Additional assumptions are that lactating and nonlactating cows eat 15 and 8 kg d.m. diet per day, respectively [8], that grazing cattle get 70% of their diet as herbage[7] and that plant material was 5% sewage sludge by weight thirty days following spraying of sewage sludge (studied by Buttigieg, reviewed in [7]). An additional scenario where plant material was 1% sewage sludge was also considered. See also the previous table.

Type of cattle	Total diet kg d.m. /day	% w/w sewage sludge on herbage	resulting % sewage sludge in diet	amount sewage sludge in diet (g)	cattle TEQ ingestion (ng/day)
Lactating	15	5	3.5	525	7.4
"	"	1	0.7	105	1.5
Non-lactating	8	5	3.5	280	3.9
"	"	1	0.7	56	0.8

Table 3. Grazing cattle TEQ ingestion resulting from soil ingestion, and the relative increase originating from sewage sludge application. The following additional assumption is made: An average of about 6% of the diet by weight may consist of soil.[8] A scenario where 1% of the diet consists of soil is also considered. For a nonlactationg cow, the sewage sludge-derived relative increases are the same as for lactating cows, even though the actual ingestion levels are lower (data not shown). See also previous tables.

Type of cattle	% Soil ingest.	Pasture location	Ingestion of background TEQ (ng/day)	Sewage sludge applic. method	Ingestion of sludge-derived TEQ (ng/day)	Sludge-derived increase (%)
Lactating	6	urban	17.6	tilling	0.06	0.3
				spraying	0.88	5
		rural	3.6	tilling	0.06	2
				spraying	0.88	24
	1	urban	2.9	tilling	0.01	0.3
				spraying	0.15	5
		rural	0.6	tilling	0.01	2
				spraying	0.15	24

Table 4. Calculations of sewage sludge-derived milk and beef TEQ levels. For milk, a PCDD/F carryover rate of 0.15, and a lactation rate of 25 kg/day was used, while for beef, a PCDD/F biotransfer factor of 0.2, and a dry matter content of 35% was used.[7] See also previous tables.

Type of cattle	% sewage sludge on herbage and % soil ingestion	Sewage sludge applic.	Total intake of sewage sludge-derived TEQ (ng/day)	Resulting milk TEQ level (pg/kg fresh wt)	Resulting beef TEQ level (pg/kg fresh wt)
Lactating	"worse case"	tilling	0.06	0.3	(4)
		spraying	8.23	49	(576)
	"better case"	tilling	0.01	<0.1	(1)
		spraying	1.62	10	(113)
Non-lactating	"worse case"	tilling	0.03	-	2
		spraying	4.39	-	307
	"better case"	tilling	0.005	-	0.4
		spraying	0.86	-	60

Background exposure of PCDDs and PCDFs to humans via food

Using a recent American food survey[11] as a basis for further calculations, it can be (roughly) estimated that beef contributes with approximately 70% of the TEQ intake for adults in the US, while milk and dairy products contribute approximately 24%.

For calculations of increased human exposure of PCDDs and PCDFs due to the use of sewage sludge, two different studies of existing PCDD/F levels in US foodstuff have been used.
In an American food survey[11] various beef products contained an average of 0.73 ppt (parts per trillion; equal to 0.73 ng/kg) TEQ, wet weight. Heavy cream contained 0.4 ppt TEQ, wet weight. Assuming that the fat content of heavy cream is 40%, this equals 1 ng TEQ/kg fat. Standard milk (with a 3% fat content) can be calculated to contain 0.030 ng TEQ/kg wet weight.
In a survey of food samples from southern Mississippi[12] ground beef contained an average of 0.75 ng TEQ/kg fat. Based on the known fat content of the ground beef samples, the average TEQ level in wet weight ground beef was 0.20 ng TEQ/kg. Dairy products contained an average of 0.77 ng TEQ/kg fat. These data are comparable to the Schecter study.[11]

Calculated increased exposure of PCDDs and PCDFs to humans due to use of sewage sludge

Transfer of sewage sludge-derived PCDD/Fs to food crops is very low or negligible. Thus, human exposure to PCDD/Fs via food crops will most likely not increase from the use of sewage sludge. Human exposure to PCDD/F via meat and dairy products may increase due to use of sewage sludge. The calculations of the increases in total human exposure to PCDD/Fs are based on the conservative assumption that all consumed milk and beef originates from cattle exposed to sewage sludge.

Milk and beef PCDD/F levels originating from sewage sludge have been calculated in Table 4. These data, together with the Schecter data[11] show that the "better case" spraying scenario (where grazing cattle consume 1% of their diet as soil, and herbage with 1% w/w sewage sludge) could increase background TEQ milk levels by 33% (0.030 ng TEQ/kg wet weight increased by 10 pg TEQ/kg fresh weight). The "worse case" tilling scenario (where grazing cattle consume 6% of their diet as soil) would only cause a 1% increase in milk levels (an increase of the background level by 0.3 pg TEQ/kg fresh weight).
If milk and dairy products contribute approximately 24% of the TEQ intake for adults in the US, sewage sludge could increase total human PCDD/F exposure by 8% or less than 0.3%, in the "better case" spraying scenario and the "worse case" tilling scenario, respectively.
For beef levels, the "better case" spraying scenario could increase background TEQ beef levels by 8% (0.73 ng TEQ/kg wet weight increased by 60 pg TEQ/kg fresh weight), while the "worse case" tilling scenario could cause a 0.3% increase (background levels increased by 2.2 pg/kg fresh weight). If beef contributes approximately 70% of the TEQ intake for adults in the US, sewage sludge could increase total human PCDD/F exposure by 6% or 0.2%, in the "better case" spraying scenario and the "worse case" tilling scenario, respectively.

Summary and conclusions

* Sewage sludge-derived PCDD/Fs can reach humans mainly through consumption of meat and/or dairy products from cattle. Human food crops seem to contribute only negligible amounts of sewage sludge-derived PCDD/Fs.
* Spraying of sewage sludge on pasture, where cattle will be allowed to graze a few months after spraying, is a situation that can lead to increased human TEQ exposure. Tilling, on the other hand, seems to entail only very slight increases in human TEQ exposure.

References

(1) Rappe C, Öberg L, Nilsson C, Håkansson H ("Biosolids land application study": unpublished report commissioned by the US Conference of Mayors).
(2) Ahlborg UG, Brouwer A, Fingerhut MA, Jacobson JL, Jacobson SW, Kennedy SW, Kettrup AAF, Koeman JH, Poiger H, Rappe C, Safe SH, Seegal RF, Tuomisto J, Van den Berg M *Eur J Pharmacol - Environ Toxicol Pharmacol Sect* **1992**, 228, 179-199.
(3) Rappe C and Öberg L ("The US EPA National Sewage Sludge Survey (NSSS) - a recalculation": abstract submitted to Dioxin '97).
(4) Duarte-Davidson R, Wilson SC, Alcock RE, Jones KC *UK Department of Energy* **1995**, Reference EPG 1/9/13
(5) Rappe C, Andersson R, Studer C, Karlaganis G ("Decrease in the concentrations of PCDDs and PCDFs in sewage sludge samples from Switzerland": abstract submitted to Dioxin '97).
(6) Kjeller L-O, Jones KC, Johnston AE, Rappe C *Environ Sci Technol* **1996**, 30, 1398-1403.
(7) Jones KC and Sewart AP *UK Department of Energy* **1995**, Reference EPG 1/9/13: 1-104.
(8) Wild SR, Harrad SJ, Jones KC *Environ. Pollut.* **1994**, 83, 357-369.
(9) Fries G, Marrow G, Snow PA *J Dairy Sci* **1982**, 65, 611-618.
(10) Fries GF *J Anim Sci* **1995**, 73, 1639-1650.
(11) Schecter A, Startin J, Wright C, Kelly M, Päpke O, Lis A, Ball M, Olson J *Chemosphere* **1994**, 29 Nos 9-11, 2261-2265.
(12) Fiedler H, Cooper KR, Bergek S, Hjelt M, Rappe C *Chemosphere* **1997**, 34 Nos 5-7, 1411-1419.