Uptake and Transfer of PCDD/Fs by Cattle Fed Naturally Contaminated Feedstuffs and Feed Contaminated as a Result of Sewage Sludge Application. 1. Lactating Cows

Michael McLachlan*,† and Wolfgang Richter‡

Ecological Chemistry and Geochemistry, University of Bayreuth, D-95440 Bayreuth, Germany, and Bayerische Landesanstalt für Tierzucht (Bavarian Center for Animal Husbandry), Professor-Dürrwaechter-Platz 1, D-85586 Poing, Germany

The uptake and transfer of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) was investigated in four Simmental cows with various lactation rates and body fat levels. During phase 1 a contaminant mass balance was conducted over 7 days using feed containing background levels of PCDD/Fs that were primarily of atmospheric origin. In phase 2 the same cows were fed grass silage from a field that had a history of repeated sewage sludge applications, and after 3 weeks a second mass balance was conducted over 7 days. The uptake of several of the PCDD/F congeners was up to 50 times higher during phase 2 than during phase 1. However, the transfer of these congeners to the milk was only ~2 times lower than during phase 1. Taking into consideration the fact that the phase 2 mass balance was not conducted at steady state, it was concluded that the carry-over of PCDD/Fs entering the feed as a result of sewage sludge fertilization is not significantly different from that for feed containing background levels of PCDD/Fs of atmospheric origin.

Keywords: *Cows; milk; carry-over; PCDD/F; sewage sludge*

INTRODUCTION

Dairy products are responsible for approximately one-third of human exposure to polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) (Birmingham et al., 1989; Fürst et al., 1990; Theelen et al., 1993). This makes the uptake and transfer of PCDD/Fs by lactating cows of critical importance in determining the human risk arising from the ubiquitous contamination of the environment with these compounds.

Feed is the primary source of PCDD/Fs to cattle (McLachlan et al., 1990). The carry-over of several PCDD/F congeners has been investigated in a number of feeding studies in which the compounds were dissolved in artificial carriers and fed to cows over periods of up to several months (Firestone et al., 1979; Jensen and Hummel, 1982; Olling et al., 1991; Heeschen et al., 1994). The carry-over rates (the quotient of the daily excretion of the compound in the milk and the daily ingestion in the feed) estimated from these studies ranged from <1% for the octachlorinated congeners to 35−60% for 2,3,7,8-Cl_4DD. It was also found that most of the 2,3,7,8-substituted PCDD/F congeners are very persistent in cows, with clearance half-lives in the order of 1−2 months.

One limitation of the feeding studies is that the chemical is generally ingested in a readily digestible solvent and not in the form in which it is present in the feed. This is a potentially critical weakness, since it has been shown that the bioavailability of PCDD/Fs is strongly influenced by the form in which the chemicals are offered in the feed (Slob et al., 1995). To be able to measure contaminant uptake and transfer from "naturally" contaminated feed, a novel method was developed, a contaminant mass balance in which uptake and excretion are measured at steady state (McLachlan et al., 1990; McLachlan, 1993). The carry-over rates obtained with this method using a single cow agreed quite well with those measured in classical feeding studies (McLachlan et al., 1990).

Application of sewage sludge to agricultural land has the potential to increase uptake of PCDD/Fs by cows, since sewage sludge is contaminated with these compounds (McLachlan et al., 1996). One of the important factors determining the extent of PCDD/F transfer into the food chain is the bioavailability of the PCDD/Fs in sewage sludge. It is conceivable that it is reduced due to binding of these compounds in the sludge. This could significantly influence the risk assessment of sewage sludge fertilization. However, we are not aware of information on the bioavailability of PCDD/Fs in sewage sludge to cows.

One purpose of the work presented in this paper was to gather further data on the uptake and transfer of PCDD/Fs from naturally contaminated feed to supplement the single controlled experiment on this subject in the literature. To this end a contaminant mass balance of four lactating cows was conducted. Thereafter, the same cows were fed feed harvested from a field that had received repeated applications of sewage sludge. After 3 weeks, the mass balance was repeated and the uptake and transfer from sewage sludge were compared with those from the naturally contaminated feed.

EXPERIMENTAL PROCEDURES

Experimental Design and Sampling. The experiment was conducted at the metabolism station of the Bavarian Center for Animal Husbandry. Four Simmental cows were

* Author to whom correspondence should be addressed.
† University of Bayreuth.
‡ Bavarian Center for Animal Husbandry.

S0021-8561(97)00922-9 CCC: $15.00
Published on Web 02/12/1998

Table 1. Characteristics of the Four Cows

	animal no.			
	713	742	783	799
birthdate	April 1988	April 1989	Feb 1990	Jan 1990
last calf	Jan 5	Feb 4	Jan 11	Feb 9
liveweight (kg)				
start	674	741	588	600
end	656	766	571	581
milk production (kg/day)				
mass balance 1	28.3	18.4	30.7	20.3
mass balance 2	26.8	16.4	27.7	17.9

selected from the herd at the State Experimental Farm to give a range in body fat weight and milk production (see Table 1).

Phase 1 of the study began in February 1994 following calving. The animals were placed on rations of feed that had been grown primarily on the experimental station and contained low levels of PCDD/Fs. After 12 weeks, the first mass balance was conducted. The daily ingestion of each feed type was monitored by weighing the feed offered and the feed remaining at the end of the feeding period. A representative sample of grass silage was collected daily, while the other feed components were sampled once. The feces were quantitatively collected with pails taped to the animals such that contamination with urine was prevented [due to the hydrophic nature of these compounds excretion via the urine is not significant in the mass balance (McLachlan et al., 1990)]. They were weighed on a daily basis and a representative aliquot was taken for later analysis. The daily milk production was measured, and toward the end of the 10 day period 1 L samples of the morning and evening milks were taken for later analysis.

For phase 2 of the study the "uncontaminated" grass silage was replaced with grass silage harvested from a meadow that had received repeated applications of sewage sludge. All other components of the feed remained the same. After 17 days on this diet, the second mass balance commenced. Grass silage and feed samples were collected for 7 days. Milk samples (morning and evening) were collected on days 3, 10, and 17−23.

The feed and feces samples were placed in aluminum boxes and sealed with aluminum. Aluminum bottles were used for the milk samples. All samples were stored at −18 °C until analysis.

Analytical Methods. For phase 1 the feces samples for 2 consecutive days were pooled, as were the morning and evening milk samples for a given day. All other samples were analyzed individually. The feed and feces samples were freeze-dried and—in the case of grass and corn silage—pulverized in a blender. The milk samples were centrifuged at 3000 rpm and 4 °C for 20 min. The cream layer was scooped off and mixed with Na_2SO_4 (1:5 w/w). All samples were Soxhlet extracted for 16 h, the feed samples in toluene, the cream and feces samples in *n*-hexane/acetone (2:1 v/v). An internal standard cocktail containing 12 $^{13}C_{12}$-labeled 2,3,7,8-substituted PCDD/F congeners was added to the solvent prior to the beginning of the extraction.

The extracts were first cleaned up on a mixed column of silica gel/H_2SO_4, silica gel, and silica gel/NaOH, which was eluted with *n*-hexane. The PCDD/Fs were then separated from other interfering contaminants on an alox column. $^{37}Cl_4$-Labeled 2,3,7,8-Cl_4DD was added as a recovery standard, and the volume was reduced to 15 μL for analysis. The cleanup method is described in detail in Horstmann and McLachlan (1997). For several samples it proved to be inadequate, and further cleanup steps involving florisil or gel permeation chromatography were employed.

The samples were analyzed using HRGC/HRMS on a VG-Autospec Ultima at a mass resolution of 8000−10000. The PCDD/F were separated on a 60 m Rtx2330 column (0.25 mm i.d., 0.1 μm film thickness) and quantified using the labeled internal standards. All 2,3,7,8-substituted congeners with the exception of 1,2,3,7,8,9-Cl_6DF were determined. A detailed description of the analytical parameters is found in Horstmann et al. (1995).

Table 2. Feed Consumption (Kilograms of Dry Weight per Day)

	animal no.			
	713	742	783	799
corn silage	1.63	1.63	1.63	1.63
sugar beet pulp[a]	1.82	1.82	1.82	1.82
barley	1.74	1.74	1.74	1.74
minerals	0.1	0.1	0.1	0.1
concentrate (mass balance 1)	6.8	1.5	7.5	2.6
concentrate (mass balance 2)	6.2	1.3	6.6	1.8
grass silage (mass balance 1)	7.5	7.3	7.0	7.8
grass silage (mass balance 2)	8.1	6.1	6.8	7.2

[a] Dried, molassed, pelleted.

RESULTS AND DISCUSSION

Quality of the Analytical Data. The recovery of 2,3,7,8-Cl_4DD was >90% for the milk samples and 80−90% for the feed and feces samples. With every batch of nine samples a method blank was analyzed. The quantities in the blanks were in almost all cases at least an order of magnitude lower than the quantities in the samples, with the exception of 2,3,7,8-Cl_4DF, 1,2,3,7,8-Cl_5DF, 1,2,3,4,7,8,9-Cl_7DF, and Cl_8DF in many of the milk samples. If the quantity in the sample was <3 times the quantity in the blank, the value was not included in the mass balance calculation. No consistent evidence of interferences was found in the chromatograms with the exception of 2,3,7,8-Cl_4DD in grass silage. This interference was estimated to account for 20% of the signal, and the data were corrected accordingly.

Uptake of PCDD/Fs. The average consumption of the different feed types is given in Table 2. There was a temporary drop in the grass silage consumption of several cows at the beginning of phase 1 following transfer to the metabolism station. As a result, only the last 7 days of phase 1 were included in the mass balance calculations.

The average concentrations in the feed are given in Table 3. The coefficient of variation for the 10 "uncontaminated" grass silage samples was <0.15 for 9 of the 16 congeners analyzed, indicating that these PCDD/Fs were homogeneously distributed in the grass silage and demonstrating the reproducibility of the analytical method. Higher variability was observed for 2,3,7,8-Cl_4DD, likely due to the proximity of the values to the limit of quantification, and for the hepta- and octachlorinated congeners, in particular 1,2,3,4,6,7,8-Cl_7DD (CV = 0.54) and Cl_8DD (CV 0.52). This variability is thought to be due to heterogeneous distribution of particle-bound material in the silage [the contamination of grass with higher chlorinated PCDD/Fs is thought to be primarily associated with particles (Welsch-Pausch and McLachlan, 1997)]. With the exception of the contaminated grass silage the concentrations in the different feeds were all very low, and the isomer distributions were consistent with atmospheric deposition as the source of contamination.

During phase 1 grass silage was the dominant source of PCDD/Fs in the cows' diet. For the animals with a low concentrate ration grass silage contributed >85% of the total uptake of most congeners, while for animals with a high concentrate diet grass silage still accounted for ~75%. This is a further illustration of the preeminent importance of grass as a source of PCDD/Fs in dairy products (McLachlan et al., 1990).

Table 3. PCDD/F Concentrations in the Feed (Picograms per Kilogram of Dry Weight)

	grass silage				
	phase 1	phase 2	corn silage + minerals	barley + sugar beet pulp[a]	concentrate
n	10	7	2	2	2
2,3,7,8-Cl_4DD	20	11	1.9	0.50	4.9
1,2,3,7,8-Cl_5DD	36	114	4.8	0.95	10.2
1,2,3,4,7,8-Cl_6DD	35	191	3.8	1.35	5.4
1,2,3,6,7,8-Cl_6DD	73	4000	8.5	2.9	20
1,2,3,7,8,9-Cl_6DD	63	1850	8.0	1.55	11.3
1,2,3,4,6,7,8-Cl_7DD	920	77000	103	29	470
Cl_8DD	5600	160000	400	126	4300
2,3,7,8-Cl_4DF	123	54	18.6	5.7	30
1,2,3,4/7,8-Cl_5DF	85	42	12.5	3.2	10.3
2,3,4,7,8-Cl_5DF	87	37	11.7	3.8	15.9
1,2,3,4,7,8/9-Cl_6DF	86	149	11.9	4.4	19.4
1,2,3,6,7,8-Cl_6DF	83	65	11.5	3.6	10.6
2,3,4,6,7,8-Cl_6DF	80	90	9.9	2.6	9.9
1,2,3,4,6,7,8-Cl_7DF	400	3900	26	7.3	85
1,2,3,4,7,8,9-Cl_7DF	35	370	5.2	2.5	17.4
Cl_8DF	460	17700	53	28	350

[a] Dried, molassed, pelleted.

The contaminated grass silage contained somewhat lower concentrations of 2,3,7,8-Cl_4DD, and many of the lower chlorinated furans. This is not unusual, since the grass grew at different locations and was harvested at different times. In contrast, the concentrations of the higher chlorinated dioxins and furans were elevated by as much as a factor of 50 compared to the uncontaminated grass silage. The particularly large increases in 1,2,3,6,7,8-Cl_6DD, 1,2,3,7,8,9-Cl_6DD, 1,2,3,4,6,7,8-Cl_7DD, Cl_8DD, and Cl_8DF as well as the presence of other non-2,3,7,8-substituted congeners are convincing evidence that the contamination originated from sewage sludge (Rappe et al., 1989; Hagenmaier et al., 1986). Since no sewage sludge was applied during the year in which the grass was harvested, the contamination must have been associated with soil particles that had adhered to the grass or that were scooped up during harvesting. This is reflected in a much more heterogeneous distribution of these congeners in the silage. The average CV for the seven samples of contaminated silage was 0.68 for the five congeners listed above, considerably higher than the CV for the uncontaminated silage.

The uptake of most congeners during phase 2 was completely dominated by grass silage, the exceptions being 2,3,7,8-Cl_4DD, 2,3,7,8-Cl_4DF, 1,2,3,7,8-Cl_5DF, 2,3,4,7,8-Cl_5DF, and 1,2,3,6,7,8-Cl_6DF, for which the concentrations and hence the contributions from grass silage were lower than during phase 1.

Mass Balance for Phase 1 ("Uncontaminated" Feed). The contaminant mass balance was conducted by multiplying the concentrations in the different matrices by their respective rates of ingestion and excretion to obtain daily fluxes. In Figure 1 the average daily excretion via milk and feces is expressed as a percentage of the average total daily uptake. For a substance that is not metabolized and which is at steady state (no net storage or mobilization of the substance in the cow), the sum of the excretion via milk and feces should add up to 100 % of the uptake. For most of the compounds this is approximately the case.

Two notable exceptions are 2,3,7,8-Cl_4DF and 1,2,3,7,8-Cl_5DF, for which the total excretion is much less than the uptake. These compounds have very low levels in milk, which indicates that they do not accumulate in the cow. Hence, the deficit in the mass balance must be the result of metabolism (McLachlan et al., 1990). This interpretation is consistent with the rapid clearance half-lives that have been reported for these congeners (Olling et al., 1991; Heeschen et al. 1994).

In Figure 1 the percentage excretion via the feces is seen to rise with increasing degree of chlorination for both the PCDDs and the PCDFs. Since the difference between uptake and feces excretion is equal to the net gastrointestinal absorption in the animal, this is equivalent to a decrease in absorption with increasing degree of chlorination. This is in agreement with previous observations (McLachlan et al., 1990) and with a mathematical model formulated to describe the absorption process (McLachlan, 1994). The model hypothesizes that the absorption of very hydrophobic compounds is limited by an aqueous resistance. Transport through this aqueous resistance is proportional to the water solubility of the compound, and since the water solubility of the PCDDs and PCDFs decreases with increasing degree of chlorination, the model predicts that the absorption will also decrease.

Mass Balance for Phase 2 (Contaminated Feed). The contaminant mass balance for phase 2 was conducted using the feed, feces, and milk data collected during days 17−23. The average daily fluxes were calculated as described above. The results are plotted in Figure 2 along with the uptake in phase 1 to give an indication of how the uptake changed with the feeding of the contaminated grass silage.

For most of the persistent congeners for which the uptake changed by a factor of <2 (2,3,7,8-Cl_4DD, 1,2,3,4,7,8-Cl_6DF, 1,2,3,6,7,8-Cl_6DF, and 2,3,4,6,7,8-Cl_6DF), the sum of the excretion through feces and milk approximately equaled the uptake. 2,3,4,7,8-Cl_5DF is an exception, with a mass balance surplus due primarily to a high excretion via milk. Apparently the cows were clearing this congener following the marked drop in uptake going from phase 1 to phase 2.

The other congeners generally show mass balance deficits. In the case of 2,3,7,8-Cl_4DF and 1,2,3,7,8-Cl_5DF this can again be attributed to metabolism. However, the other congeners are very persistent, so there must be another explanation. Common to all of these compounds is the increase in levels in feed in phase 2. The cows will react to this disturbance of the steady state by storing more contaminant in their tissue until

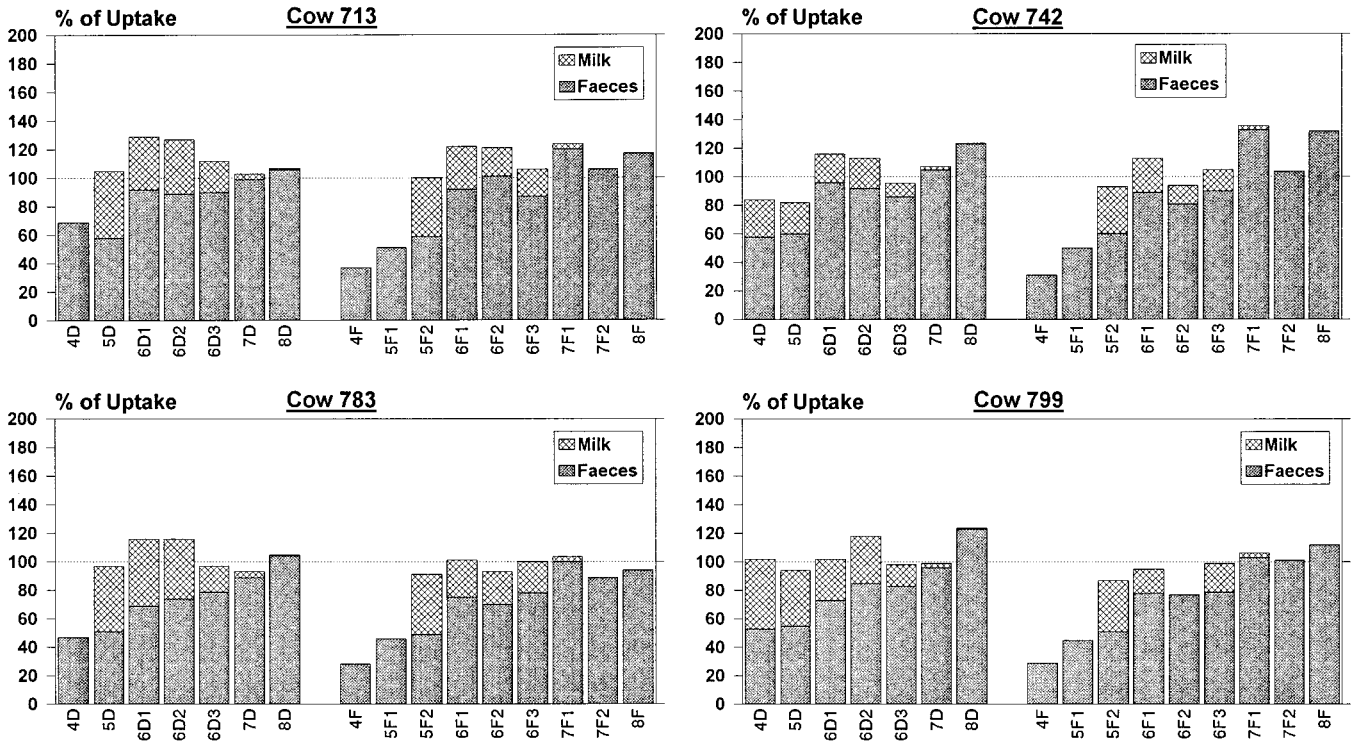

Figure 1. Results of the mass balance for each of the cows during phase 1. The average daily excretions via milk and feces are plotted as a percentage of the average daily uptake. Abbreviations: 4D, 2,3,7,8-Cl_4DD; 5D, 1,2,3,7,8-Cl_5DD; 6D1, 1,2,3,4,7,8-Cl_6-DD; 6D2, 1,2,3,6,7,8-Cl_6DD; 6D3, 1,2,3,7,8,9-Cl_6DD; 7D, 1,2,3,4,6,7,8-Cl_7DD; 8D, Cl_8DD; 4F, 2,3,7,8-Cl_4DF; 5F1, 1,2,3,4/7,8-Cl_5-DF; 5F2, 2,3,4,7,8-Cl_5DF; 6F1, 1,2,3,4,7,8/9-Cl_6DF; 6F2, 1,2,3,6,7,8-Cl_6DF; 6F3, 2,3,4,6,7,8-Cl_6DF; 7F1, 1,2,3,4,6,7,8-Cl_7DF; 7F2, 1,2,3,4,7,8,9-Cl_7DF; 8F, Cl_8DF.

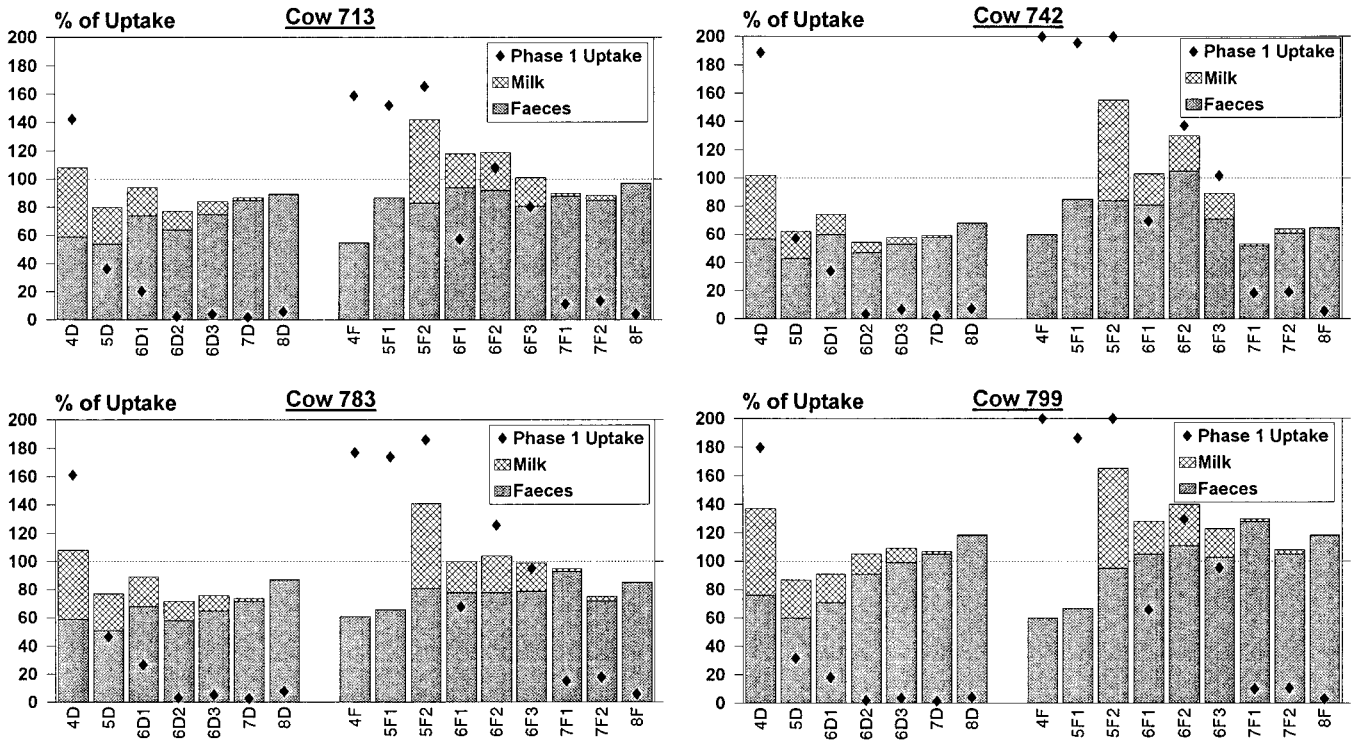

Figure 2. Results of the mass balance for each of the cows during phase 2. The average daily excretions via milk and feces as well as the average daily uptake during phase 1 are plotted as a percentage of the average daily uptake during phase 2.

a new steady state is reached for the higher level of uptake. Hence, the mass balance deficits could conceivably be attributable to storage. However, using Cl_8DD in cow 742 as an example, this would imply that 32% of the ingested compound was stored in the animal, whereas only 0.1% was excreted in the milk. This distribution between milk and body storage 3 weeks after the increase in Cl_8DD uptake is not plausible.

Instead, it is believed that the mass balance deficits are due to an overestimation of contaminant uptake. This could have arisen from the difficulty in preparing representative grass silage samples, since the com-

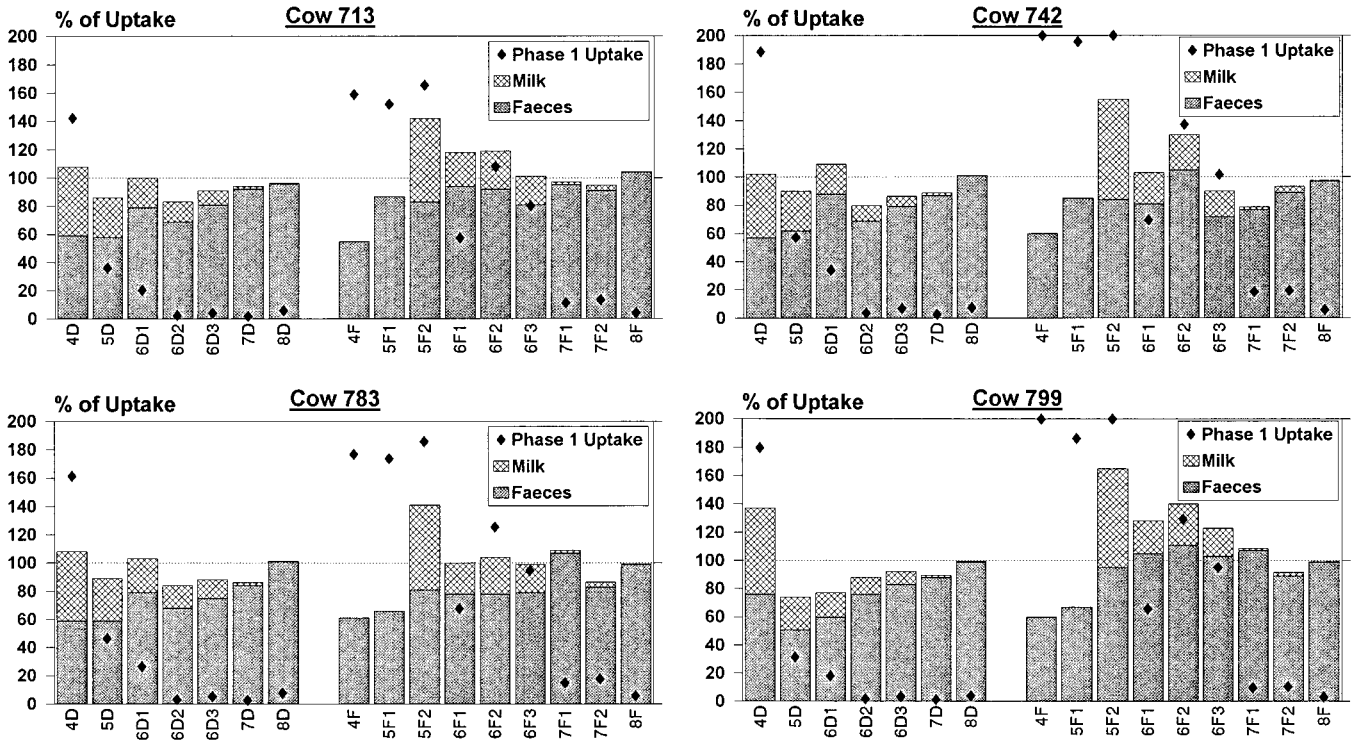

Figure 3. Results of the corrected mass balance for each of the cows during phase 2. The average daily excretions via milk and feces as well as the average daily uptake during phase 1 are plotted as a percentage of the average daily uptake during phase 2. See text for details of the correction.

pounds showing higher levels in the contaminated silage were associated with soil particles (see above) and the soil particles were not homogeneously distributed in the silage. It may also have been due to selective feeding behavior of the cattle resulting in a lower relative consumption of the soil present in the silage than of the silage itself. It was decided to correct for this error by assuming that the sum of feces and milk excretion was equal to the uptake for Cl_8DD and Cl_8DF. These compounds were chosen because their very low rates of excretion via milk indicate that digestive tract absorption is very low and hence that storage to tissue must also be small compared to excretion via feces. To perform the correction, the consumption of grass silage was increased until the uptake and excretion of each of these two compounds were equal. This modified grass silage consumption rate was used to recalculate the mass balances for the other congeners for which the concentrations in contaminated silage were at least a factor 2 higher than in uncontaminated silage. The results are shown in Figure 3. Most of the compounds consistently show a small mass balance deficit that is of similar magnitude to the milk excretion flux and that can plausibly be attributed to storage in the animal.

Carry-over. The carry-over rates, defined as the fraction of the daily intake of a contaminant that is excreted via the milk, are plotted in Figure 4. There is considerable variability among the different animals during phase 1. This may be due to the fact that the values are based on the analysis of only one milk sample. The differences among the animals measured during phase 2 on the basis of seven milk samples were much smaller, in particular for the three Cl_6DF congeners that were taken up in similar quantities in phases 1 and 2. This suggests that the carry-over rate is not strongly influenced by the lactation rate, body fat weight, or feed mix of the animal. This makes the carry-over rate an ideal parameter to describe contaminant transfer in lactating cows, since it is a constant that is insensitive to the characteristics of the individual animal. Other parameters such as the biotransfer factor (Travis and Arms, 1988) and the bioconcentration factor (Connett and Webster, 1987) are directly dependent on the lactation rate and/or the quantity of food ingested and are thus different for each individual animal.

The carry-over rates decrease with increasing degree of chlorination, from around 40% for 2,3,7,8-Cl_4DD to 0.5% for Cl_8DD. This is due to the decrease in digestive tract absorption with increasing degree of chlorination mentioned above. This range of a factor of 100 has a dramatic effect on the bioaccumulation of the different PCDD/Fs through the agricultural food chain, effectively filtering out the higher chlorinated congeners.

The average carry-over rates for phases 1 and 2 are listed in Table 4. There is very good agreement between the values for those congeners which were taken up in similar quantities during both phases (1,2,3,4,7,8/9-Cl_6-DF and 2,3,4,6,7,8-Cl_6DF). The carry-over rates of 2,3,7,8-Cl_4DD, 2,3,4,7,8-Cl_5DF, and 1,2,3,6,7,8-Cl_6DF were considerably higher during phase 2 than during phase 1. This can be explained by the lower uptake during phase 2 leading to a clearance of these compounds from the cows.

The carry-over rates for the remaining congeners were a factor of ~2 lower during phase 2. These are the compounds of sewage sludge origin for which the uptake was much higher during phase 2. The lower carry-over rates are likely largely attributable to the fact that the cows had probably not reached steady state at the time the sampling was done. Feeding studies have shown that although there is a rapid increase in milk levels immediately following the start of a feeding experiment, the steady state milk concentration is generally not reached for 9−11 weeks (Heeschen et al., 1994). The milk levels measured on days 17−23 of feeding may well

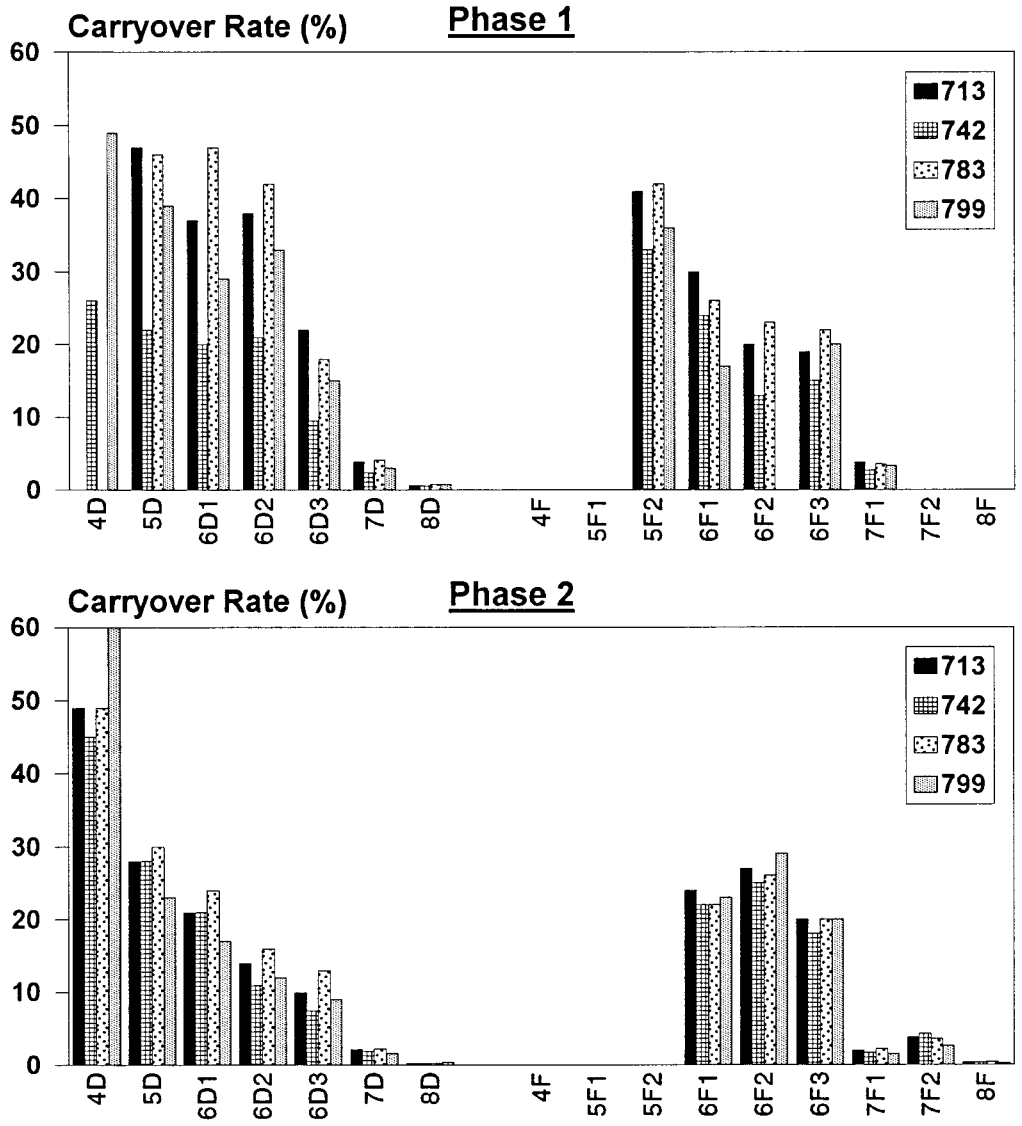

Figure 4. Plot of the carry-over rates for each cow during phases 1 and 2.

Table 4. Carry-over Rates (in Percent; Average for the Four Cows)

	phase 1	phase 2
2,3,7,8-Cl_4DD	38	51[a]
1,2,3,7,8-Cl_5DD	39	27[b]
1,2,3,4,7,8-Cl_6DD	33	21[b]
1,2,3,6,7,8-Cl_6DD	33	13.2[b]
1,2,3,7,8,9-Cl_6DD	16.0	9.9[b]
1,2,3,4,6,7,8-Cl_7DD	3.4	1.96[b]
Cl_8DD	0.68	0.28[b]
2,3,7,8-Cl_4DF	*c*	*c*
1,2,3,4/7,8-Cl_5DF	*c*	*c*
2,3,4,7,8-Cl_5DF	40	65[a]
1,2,3,4,7,8/9-Cl_6DF	24	23
1,2,3,6,7,8-Cl_6DF	18.7	27[a]
2,3,4,6,7,8-Cl_6DF	18.9	19.5
1,2,3,4,6,7,8-Cl_7DF	3.4	1.85[b]
1,2,3,4,7,8,9-Cl_7DF	*c*	3.6[b]
Cl_8DF	*c*	0.31[b]

[a] Steady-state carry-over rate overestimated due to clearance of chemical from body fat during phase 2. [b] Steady-state carry-over rate underestimated due to storage of chemical in body fat during phase 2. [c] Carry-over not estimated because the milk samples were below the limit of quantification.

represent only half of the steady state concentrations, which would explain the 50% lower carry-over rates. This interpretation is supported by the results of the corrected mass balance (Figure 3), according to which only about half of the absorbed chemical was excreted in the milk, while the other half was stored in the cows (see above). Even if the cows were considerably closer to a steady state, the carry-over rates for PCDD/Fs of sewage sludge origin (phase 2) were not substantially lower than the carry-over rates for PCDD/Fs from feed contaminated through atmospheric deposition (phase 1). Hence, we conclude that the digestive tract absorption and carry-over of PCDD/Fs are not significantly inhibited by the sewage sludge matrix.

The carry-over rates measured in this study are generally consistent with those determined in the original mass balance experiment with a single cow (McLachlan et al., 1990). A notable exception is the lower transfer of Cl_8DD and Cl_8DF reported here. The transfer rates of <1% measured for these congeners are in agreement with theoretical expectations (McLachlan, 1994) and with the one feeding study involving this congener: Firestone et al. (1979) reported an average excretion of Cl_8DD in the milk equal to 0.6% of the daily intake during days 40−70 of the feeding period. It is believed that the high values in the earlier study were due to sample contamination.

ACKNOWLEDGMENT

We thank Balthasar Spann and Anton Obermaier for their assistance in conducting the feeding trial and preparing the samples and Sandra Zeitler and Stefan Will for working up and helping to analyze the samples.

LITERATURE CITED

Birmingham, B.; Gilman, A.; Grant, D.; Salminen, J.; Boddington, M.; Thorpe, B.; Wile, I.; Toft, P.; Armstrong, V. PCDD/PCDF Multimedia Exposure Analysis for the Canadian Population: Detailed Exposure Estimation. *Chemosphere* **1989**, *19*, 637−642.

Connett, P.; Webster, T. An Estimation of the Relative Human Exposure to 2,3,7,8-TCDD Emissions via Inhalation and Ingestion of Cow's Milk. *Chemosphere* **1987**, *16*, 2079−2084.

Firestone, D.; Clower, M., Jr.; Borsetti, A. P.; Teske, R. H.; Long, P. E. Polychlorodibenzo-*p*-dioxin and Pentachlorophenol Residues in Milk and Blood of Cows Fed Technical Pentachlorophenol. *J. Agric. Food Chem.* **1979**, *27*, 1171−1177.

Fürst, P.; Fürst, C.; Groebel, W. Levels of PCDDs and PCDFs in Food-Stuffs from the Federal Republic of Germany. *Chemosphere* **1990**, *20*, 787−792.

Hagenmaier, H.; Brunner, H.; Haag, R.; Bechthold, A. PCDDs and PCDFs in Sewage Sludge, River and Lake Sediments from South West Germany. *Chemosphere* **1986**, *15*, 1421−1428.

Heeschen W., Blüthgen A.; Ruoff, U. *Untersuchungen zum Übergang ausgewählter polychlorierter Dibenzo-para-dioxine und -Furane nach oraler Supplementierung in die Milch laktierender Kühe*; Bundesanstalt für Milchforschung: Kiel, 1994.

Horstmann, M.; McLachlan, M. S. Results of an Initial Survey of Polychorinated Dibenzo-*p*-dioxins (PCDD) and Dibenzofurans (PCDF) in Textiles. *Chemosphere* **1995**, *31*, 2579−2589.

Horstmann, M.; McLachlan, M. S. Sampling Bulk Deposition of Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans. *Atmos. Environ.* **1997**, *31*, 2977−2982.

Jensen, D. J.; Hummel, R. A. Secretion of TCDD in Milk and Cream Following the Feeding of TCDD to Lactating Dairy Cows. *Bull. Environ. Contam. Toxicol.* **1982**, *29*, 440−446.

McLachlan, M. S. Mass Balance of Polychlorinated Biphenyls and Other Organochlorine Compounds in a Lactating Cow. *J. Agric. Food Chem.* **1993**, *41*, 474−480.

McLachlan, M. S. Model of the Fate of Hydrophobic Contaminants in Cows. *Environ. Sci. Technol.* **1994**, *28*, 2407−2414.

McLachlan, M. S.; Thoma, H.; Reissinger, M.; Hutzinger, O. PCDD/F in an Agricultural Food Chain. Part 1: PCDD/F Mass Balance of a Lactating Cow. *Chemosphere* **1990**, *20*, 1013−1020.

McLachlan, M. S.; Horstmann, M.; Hinkel, M. Polychlorinated dibenzo-*p*-dioxins and dibenzofurans in sewage sludge: sources and fate following sludge application to land. *Sci. Total Environ.* **1996**, *185*, 109−123.

Olling, M.; Derks, H. J. G. M.; Berende, P. L. M.; Liem, A. K. D.; de Jong, A. P. J. M. Toxicokinetics of Eight ^{13}C-Labeled Polychlorinated Dibenzo-*p*-dioxins and -furans in Lactating Cows. *Chemosphere* **1991**, *23*, 1377−1385.

Rappe, C.; Kjeller, L.-O.; Andersson, R. Analyses of PCDDs and PCDFs in Sludge and Water Samples. *Chemosphere* **1989**, *19*, 13−20.

Slob, W.; Olling, M.; Derks, H. J. G. M.; de Jong, A. P. J. M. Congener-specific Bioavailability of PCDD/Fs and Coplanar PCBs in Cows: Laboratory and Field Measurements. *Chemosphere* **1995**, *31*, 3827−3838.

Theelen, R. M. C.; Liem, A. K. D.; Slob, W.; van Wijnen, J. H. Intake of 2,3,7,8 Chlorine Substituted Dioxins, Furans, and Planar PCBs from Food in The Netherlands: Median and Distribution. *Chemosphere* **1993**, *27*, 1625−1635.

Travis, C. C.; Arms, A. D. Bioconcentration of Organics in Beef, Milk, and Vegetation. *Environ. Sci. Technol.* **1988**, *22*, 271−274.

Welsch-Pausch, K.; McLachlan, M. S. Pathways of Atmospheric PCDD/F to a Native Grassland Culture: The Importance of Particle-bound Deposition. *Organohalogen Compds* **1997**, *28*, 72−75.

Received for review October 28, 1997. Revised manuscript received December 8, 1997. Accepted December 15, 1997. We are grateful to the Bavarian State Ministry for Food, Agriculture and Forestry for financially supporting the analysis of the milk samples.

JF970922U