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Derivation and Field Testing of Air−Milk and Feed−Milk Transfer Factors for PCBs

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Detailed field experimental data on the air to herbage transfer of PCBs was combined with data on feed to milk transfers from a detailed feeding trial with lactating cows to derive congener-specific air to milk and feed to milk transfer factors ($TF_{A:M}$ and $TF_{F:M}$). The variability and uncertainties in these factors are discussed largely with reference to UK field conditions. $TF_{A:M}$ values were 2.4, 54, and 650 m^3 of air g^{-1} of milk fat for congeners 18, 74, and 170, respectively. The usefulness of the transfer factors as predictive tools was tested on (i) data from two milk and feed surveys (in late spring 1996 and winter 1997) of farms in Northwest England; (ii) data from a long-term monitoring study conducted throughout the 1996 growing season; and (iii) data from the literature. $TF_{A:M}$ and $TF_{F:M}$ gave excellent predictions of the milk PCB concentrations for all tested data sets, with milk concentrations of the persistent congeners predicted to within a factor of ∼2−3 at the local level and to well within an order of magnitude at the regional level. The main requirements of using $TF_{A:M}$ are that (i) pasture is the dominant feed; (ii) winter-fed silage is grown locally, and (iii) there is no local intermittent source. Survey results showed that levels of persistent PCB congeners in silage are directly correlated with milk output fluxes. Bioconcentration factors (BCFs) and carry-over rates (CORs) calculated for both study approaches were very similar to those found in "uncontaminated" feeding studies. Although CORs are theoretically preferable to BCFs the variability found for each showed that there is likely to be little practical advantage in collecting the extra data required for the calculation of CORs.

Introduction

PCBs and other semivolatile persistent, bioaccumulatory organic contaminants reach human tissue primarily via dietary intake, with ingestion of meat, dairy products, and fish generally dominating dietary exposure to PCBs for the average consumer (*1*−*3*). Grazing animals therefore supply PCBs to humans from the terrestrial environment and they, in turn, receive PCBs primarily through ingestion of grass, silage, and concentrate feed (*4*). PCBs and similar compounds reach grass and other vegetation principally via atmospheric deposition (*5*). Hence, there is a clear link along the pathway air−vegetation−grazing animals−meat/dairy products which results in human exposure to PCBs and a range of other persistent organic pollutants (POPs). As a consequence atmospheric emissions which affect air concentrations of POPs will ultimately exert a strong influence on human tissue concentrations.

Given the importance of the air−grazing animal pathway, researchers—including ourselves—have been studying the range of processes which essentially "control" the partitioning kinetics of POPs transfer between air−vegetation and grazing animal feed−milk/body fat (*4*−*11*). These processes can be related to key physical−chemical properties and molecular structure features of the POPs, and it therefore becomes possible to derive certain general relationships for well-characterized compounds, which facilitate the prediction of pasture grass concentrations from air concentrations (*11*) and milk concentrations from grass/silage/livestock feed (*4*). The purpose of this paper is therefore to go to the final step of deriving direct air−milk transfer factors from carefully conducted controlled field and feeding studies and to consider the usefulness of these transfer factors as a predictive/management tool. Variables which could potentially influence the congener-specific air−milk transfer factors ($TF_{A:M}$) are therefore systematically considered, and an attempt is made to quantify them. These were considered to include the spatial and temporal variation of atmospheric PCB concentrations, the variation in air to herbage transfers due to species differences or seasonal effects, dietary intake variability (both seasonally and through husbandry differences on grass or silage based diets, soil ingestion, and feed supplements), and variability in transfers to milk between cows and due to the stage of lactation, body weight, age etc. (*4*,*11*).

In the second part of the paper, the derived $TF_{A:M}$ values are tested for their applicability on data from two surveys of milk and feed from farms throughout Northwest England, on a monitoring study from a farm adjacent to the field site used for detailed air-grass transfer studies (*11*), and on previously published data from the United Kingdom and other countries. Carry-over rates (CORs, calculated as the total daily output flux in milk divided by the total daily input flux in feed) and bioconcentration factors (BCFs, calculated as the ratio of the concentrations of contaminants in the human foodstuff—milk—and the animal foodstuff—grass/silage/concentrate) are also tested on the data from the farm surveys and the long-term monitoring study.

Derivation of Transfer Factors and Factors Affecting Their Variability and Uncertainty

The purpose of this section is to derive congener-specific transfer factors $TF_{A:M}$ and $TF_{F:M}$ (feed−milk transfer factors, or BCF) values from the detailed air−grass field studies and the controlled feed−milk feeding trial referred to above (*4*, *11*) and then to consider the factors which might result in their variability.

Derivation of Air−Milk Transfer Factors. By multiplying BCFs and grass scavenging coefficients from the studies previously described $TF_{A:M}$ values were obtained, shown in Table 1. Only congeners which were reliably detected in all three matrices (milk fat, grass, and air) are shown. The RSDs for the $TF_{A:M}$ values were estimated by combining the RSDs of the BCFs (*4*) and scavenging coefficients (*11*) (for expressions of the form $x = ab$; $(RSD_x)^2 = (RSD_a)^2 + (RSD_b)^2$, assuming that the errors are random), and are around 50% for most of the congeners (for the persistent congeners the scavenging coefficients RSDs are about twice those for the BCFs). The differences in $TF_{A:M}$ between metabolized and unmetabolized congeners is very obvious, with the metabo-

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TABLE 1. Derivation of Air−Milk Transfer Factors

PCB	BCF[a] (g DW g^{-1} fat)	air−grass scavenging coefficient (m^3 air g^{-1} DW)	air−milk transfer factor ($TF_{A:M}$) (m^3 air g^{-1} fat)	estimated RSD on $TF_{A:M}$[b] (%)
18	0.4	6.9	2.4	290
28	0.5	6.4	3.2	120
66	2.6	11	29	49
74	6.2	8.7	54	39
101	0.8	14	11	49
110	0.5	13	6.5	51
118[c]	16	25	400	46
138[c]	12	33	380	53
141	1.2	18	21	110
149	1	16	16	67
153[c]	13	21	260	46
170[c]	10	63	650	56
180[c]	11	51	540	39
183[c]	10	41	430	65
187	1.6	35	54	54

[a] Averaged for five cows throughout a four-month period of lactation (see ref 4). [b] Calculated from the standard deviations of BCFs and Scavenging Coefficients. [c] Persistent (largely unmetabolized) congeners in cows (see ref *12*).

lized congener $TF_{A:M}$ values mostly having very high standard deviations. $TF_{A:M}$ values ranged between 2.4 m^3 g^{-1} of fat for PCB-18 through 260−400 m^3 g^{-1} for PCBs 153, 138, and 118, up to over 500 m^3 g^{-1} for PCBs 180 and 170 (Table 1).

Variability in Air Concentrations. PCB air concentrations vary spatially and temporally, which will affect the amounts available for transfer to grass and ultimately milk. If the United Kingdom is considered as a study area, representative of an industrial country where PCBs have been widely used in the past, rural air concentrations are generally lower than city center ones (*13, 14*). Concentrations in large city centers, such as Manchester or London, average ∼300−1400 pg of ∑PCB m^{-3} of air, for example, while semiurban/semirural sites average ∼100−300 pg of ∑PCB m^{-3} of air (*11, 15*) with remote sites typically up to about a factor of ∼5 lower again (*16*). Grazing livestock are predominantly reared for the human food chain in remote/rural/semiurban regions; it is therefore reasonable to expect atmospheric PCB concentrations varying spatially over about 1 order of magnitude will "supply" the vast majority of agroecosystems in the United Kingdom (*16, 17*).

Temporal variation in air concentrations in typical rural areas can vary annually, seasonally, and in the short-term (e.g., over hours). There is often a strong link between increased temperature and increased PCB air concentrations (*18*), suggestive of a temperature controlled air-surface exchange of PCBs. Having said this, annual PCB concentrations at any one place are now quite stable, varying only by a factor of 1.3 (between 1.09 and 1.45 ng of ∑PCB m^{-3}) in London city center over 5 years, for example (*14*). Similarly, weekly/biweekly air concentrations varied by less than a factor of 2 (summer > winter) in an urban center (*13*) and daily air concentrations only varied by a factor of ∼7 over nine months at a semiurban coastal site near Lancaster, despite receiving regional air masses with widely different back trajectories/source areas (*15*). Indeed, diurnal temperature-driven variations in air concentrations were greater than these seasonal or daily variations at Lancaster (*19*). These observations can be taken as evidence that PCBs have become well-mixed throughout the environment over time by environmental recycling (air−surface exchange). In summary, as we shall see below, this results in relatively little variability in pasture concentrations to supply PCBs to grazing animals (see also ref *17*). Importantly, however, spatial and temporal variability in air concentrations should just be viewed as influencing the "supply" of PCBs to grass/feed and—ultimately—milk, not the values of $TF_{A:F}$ (the air−feed transfer factors, or "scavenging coefficients") or $TF_{A:M}$ themselves.

Variability in Herbage Concentrations. In the United Kingdom, grazing livestock typically graze pasture herbage in the summer (normally ∼April−October) and silaged pasture collected from the field when they are kept indoors during the winter. Consequently, the bulk of their diet is made up of fresh pasture or silaged pasture exposed to ambient air during the periods of active grass growth in March/April−September/October. Air−pasture transfer factors are therefore of key importance in the link between air and milk, and it is appropriate to consider how variable air−pasture transfer is likely to be and factors which could influence $TF_{A:F}$. It is considered that $TF_{A:F}$ could potentially vary with mode of deposition, plant species, exposure (growth) period and the kinetics of uptake, and factors influencing the retention of PCBs by the pasture (i.e., temperature affecting the octanol:air partition coefficient (K_{OA}), shedding of cuticle etc.).

In the detailed field study (*11*), it was noted that total PCB pasture concentrations varied by a factor of 3 between the highest and lowest measurements between late April and late September, with winter concentrations typically about 2-fold and 5-fold higher than summer ones for PCBs 28 and 170, respectively. Air−herbage transfer was congener-specific and a strong function of compound K_{OA}. Temperature influences K_{OA} but during the growth of pasture *average* (weekly/fortnightly) air temperatures will vary rather little spatially throughout the United Kingdom (<∼5 °C) and through the growth period (<∼3 °C). The relatively consistent spatial and temporal variations in air concentrations (noted above) and pasture temperatures may therefore be expected to yield quite constant pasture concentrations. This assertion is supported by field observations, with pasture harvested from different UK sites during 1995 varying with a standard deviation of 47% (*20*) (see also data presented below).

An important issue of air−pasture transfer of POPs concerns the kinetics of uptake and whether it is valid to assume that the pasture (i.e., cuticle) has attained equilibrium with atmospheric gas phase POPs. Field data for the air−pasture study has shown that, for PCBs, this equilibrium is attained rapidly—in the order of a few days or less (*20*). This is in contrast to work on the air−grass transfer of PCDD/Fs, published by McLachlan et al. (*21*). This is of significance for the derivation of $TF_{A:F}$ and $TF_{A:M}$, because it indicates that grazing livestock will always be grazing/feeding on pasture/silage which has had time to equilibrate with the air while it grows in the field. Differences in husbandry techniques (e.g., silage versus free-range grazing) affecting the length of time the grass consumed by cows has been exposed should therefore have relatively little effect upon the measured grass scavenging coefficient. The short time required for air−grass equilibrium to be attained (*20*) means that PCB levels in grass will reflect recently prevalent air levels and environmental conditions, so PCB levels in grass will be expected to "reflect" underlying changes in air concentrations and temperature through its growth/exposure time. The actual time required for air-grass equilibrium to be attained is still not known precisely from our studies but, as noted above, it is a few days or less.

Little variability in $TF_{A:F}$ values between different species of grass is expected (*20*). Although other species of plant present in the pastureland sward could give different $TF_{A:F}$ values to grass (*8*), grass species generally dominate pasture and therefore the effect of the presence of other species on the $TF_{A:F}$ values for the whole sward composition is expected to be slight.

Dry particulate deposition and the particulate bound fraction of wet deposition are expected to be variable because the particulate loading of air is affected by seasonal and other temporal factors and the direction of the prevailing air mass, dry deposition varies widely with wind speed and micrometeorological characteristics, and wet deposition is dependent on the size, intensity, and type (i.e., snow, heavy rain, drizzle, fog, etc.) of precipitation events (*22*). However, it is estimated that, even in the winter in the United Kingdom, over 90% of, even heavier, PCBs are contained in the vapor phase of air (*4, 13, 23*), and these deposition routes are therefore expected to be relatively unimportant for PCBs. It has been reported that, even for PCDD/Fs with <6 chlorine atoms, dry gaseous deposition is the dominant uptake route for grasses (*5, 9*).

Variability in the Cows' Dietary Composition and Feed Intake Rates. As noted above, cows in the United Kingdom are expected to be exposed to relatively consistent PCB concentrations throughout the year as they predominantly eat fresh pasture only in the field and silage prepared from that pasture, generally cut in two, or sometimes three, sessions during the summer. Two factors which might therefore lead to variability in silage PCB composition are the silaging process itself and the timing of cutting. The silaging process could *potentially* alter the PCB concentrations found in grass by degradation/volatilisation and silage produced from different cuts might be expected to contain different PCB levels and mixtures because of the seasonality in the air−grass transfer of PCBs. However, in practice no evidence for either losses of PCBs during silage production or differences in PCB concentrations from silage produced at different times was noted in the feeding trial (*4*). This increases our confidence in applying the $TF_{A:M}$ values derived here more widely.

Feed consumption changes throughout the lactation cycle (described in ref *4*), and as cows become heavier with age their feed consumption increases correspondingly. Concentrate feeds are generally fed in fixed daily amounts from large homogeneous batches, so variations in total daily PCB intake in a herd are controlled by variations in the voluntary intake of herbage and other forage crops (roots for example). The intake of other potential sources of PCBs, such as soil, are heavily dependent on husbandry techniques and pasture quality and are therefore potentially highly variable. In particular, pasture quality declines at the height of summer, which is likely to cause an increase in soil intake (*24, 25*).

Given the relative consistency of the bulk dietary components—pasture and silage—noted above, variability of PCB milk concentrations between different animals/herds is expected to depend to a large extent on the source of any concentrate feed or "nonpasture" (i.e., root crops, grains) they receive. Certain ingredients (notably fishmeal) can contain extremely variable levels of PCBs (*4*). Certain sources of the ingredients used in concentrate feeds are often particular to certain regions (for example, fishmeal from the North Sea is mostly used in the eastern half of the United Kingdom, whereas in the western United Kingdom fishmeal usually comes from South American sources (*26*)), so it is expected that most farms within a particular region may have quite similar PCB levels in their concentrate feeds. However, in the absence of "special" sources of PCB intake such as soil ingestion and contaminated feed, voluntary herbage intake is the main source of variability in PCB intake for cows. The maximum intake rate (i.e., nanograms per day) during the lactation cycle occurs approximately 4 months after parturition and averages only about 1.5 times the intake shortly after parturition (*4*). Again, therefore, variability is rather low, particularly when averaged over an entire herd or over the composition of milk supplied to the large commercial dairies which can dominate the supply of dairy products to the human foodchain.

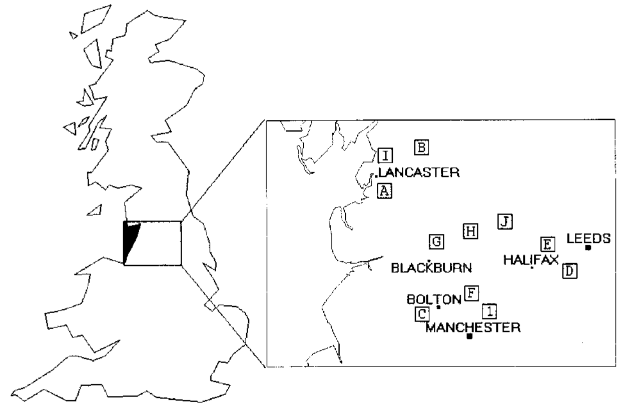

FIGURE 1. Map showing the locations of farms in the northwest of England selected for two surveys.

Variability in PCB Transfers to Milk. Feed to milk transfer factors (CORs and BCFs) are also subject to variability, but vary with a standard deviation of up to 30% both between cows at any time and within one cow through the lactation cycle (*4*) as a function of absorption efficiency through the gut, metabolism, and the stage of lactation (as discussed in ref *12*). This process supplying PCBs to human foodstuffs is therefore also rather uniform but, importantly, varies between congeners.

Overview Comments on Variability in the Air−Milk Transfer Factors. In summary, variability in $TF_{A:M}$ values can be due to a host of factors, but the most important is likely to be the composition of the cows' diet, particularly concentrate feeds. Other factors leading to variability in air−grass scavenging coefficients and feed to cow transfer factors combine to give an overall standard deviation for $TF_{A:M}$ values, excluding the role of "contaminated" feeds, of about 50%.

Testing Air to Milk Transfer Factors

Northwest England Farm Surveys. Eleven farms in a mixture of rural and semirural areas in the northwest of England were selected, located as shown in Figure 1. Samples of silage and milk were taken in May 1996 and samples of silage, milk, and complementary feeds were taken in February 1997. Milk samples (1 L) were taken directly from the farms' bulk tanks into hexane-cleaned glass bottles, centrifuged on the day of collection and the milk fat stored at −25 °C until analysis. Feed samples were taken into sealable polythene bags and stored at −25 °C until analysis. Farmers were issued a questionnaire on husbandry practices, herd composition, and milk yields, most of which were completed. In 1996 one farm was unavailable for sampling, and in 1997 one farmer refused to give samples of feed.

On both sampling occasions cattle at all of the farms were being kept indoors, under winter husbandry methods. The following statistics are for the farms with completed questionnaires. All cows in all of the herds were English Holstein/Friesian. At each sampling date each of the herds was comprised of cows at varying stages of the lactation cycle, although in all but two of the herds the cows were, on average, past the 20th week of lactation. All silage was produced for home use, and only one farmer had changed supplier of concentrate feed between surveys (less than three weeks before the second survey). One farmer prepared his own concentrate but was not prepared to supply samples of this, or of silage. The average milk fat yield was 0.74 kg per cow per day.

Long-Term Monitoring of One Farm. Throughout the growing season (approximately April−October) of 1996 milk samples were taken every two weeks from the bulk tank of a farm adjacent to the field site concurrently used for the air

TABLE 2. Summary of Concentrations of Semipersistent and Persistent PCBs Found in Surveys of Farms in Northwest England in 1996 and 1997[a]

	1996		1997		
PCB	silage (pg/g of DM)	milk (pg/g of fat)	silage (pg/g of DM)	concentrate (pg/g of DM)	milk (pg/g of fat)
47[c]	17 ± 15 (0−51)	400 ± 880 (51−2890)	45 ± 57 (15−200)	17 ± 7.9 (7−29)	230 ± 310 (54−1140)
66[c]	49 ± 40 (13−150)	100 ± 49 (32−200)	89 ± 120 (25−410)	25 ± 18 (10−53)	90 ± 45 (30−180)
74[c]	31 ± 17 (17−74)	220 ± 92 (100−350)	67 ± 120 (18−400)	22 ± 12 (9−37)	270 ± 140 (130−550)
105[c]	26 ± 18 (6−70)	180 ± 71 (73−270)	26 ± 22 (6−73)	11 ± 6.7 (1−24)	170 ± 81 (83−380)
128[c]	na[b]	na[b]	9.5 ± 7.9 (2−24)	7.2 ± 3.2 (2−12)	130 ± 60 (76−290)
187[c]	24 ± 17 (9−65)	59 ± 63 (20−230)	18 ± 12 (8−44)	12 ± 6.7 (3−25)	25 ± 8.9 (11−41)
118[d]	76 ± 57 (27−230)	1220 ± 510 (580−1950)	78 ± 61 (23−190)	38 ± 19 (9−74)	970 ± 480 (550−2250)
138[d]	120 ± 77 (64−330)	1800 ± 950 (960−3950)	84 ± 61 (27−190)	55 ± 27 (17−98)	950 ± 410 (590−2030)
153[d]	80 ± 63 (15−240)	1910 ± 1040 (1060−4420)	82 ± 57 (27−190)	59 ± 31 (13−100)	1070 ± 440 (650−2200)
170[d]	20 ± 16 (6−59)	290 ± 230 (140−910)	14 ± 11 (5−37)	8.8 ± 6.3 (2−24)	210 ± 90 (130−440)
180[d]	39 ± 33 (11−120)	690 ± 530 (300−2100)	29 ± 21 (11−75)	18 ± 11 (6−43)	450 ± 200 (280−960)
183[d]	10 ± 7.3 (3−28)	140 ± 88 (75−370)	7.5 ± 5.6 (3−18)	4.0 ± 3.1 (0−10)	120 ± 49 (75−250)

[a] The means ± SDs and the ranges are shown. [b] Not analyzed. [c] Semipersistent congener. [d] Persistent congener.

and grass measurements detailed above (*11*). The samples were treated as described above. Samples were taken from the pasture on one occasion to ensure its comparability with samples taken at the adjacent field site. A questionnaire on husbandry practices, herd composition, and milk yields was completed by the farmer.

The herd studied was composed of thirty English Holstein/Friesian cattle which all calved in late March/early April 1996. The average weight of the cows throughout the monitoring period was 450 kg, and average milk fat yields were 0.63 kg per day. Silage was grown on the farm, and concentrate feed was purchased and fed to the cows at a rate of 3.5 kg per cow per day. The cows were grazing pasture outdoors from late April until late October, when lactation ceased and the cows were brought indoors.

Chemical Analysis. The analytical methods for PCBs in the matrixes sampled in this study have been presented elsewhere (*4, 27*).

Other Datasets from the Literature. Two published surveys of PCB levels in milk in the UK (*28, 29*) were used to test the $TF_{A:M}$ in addition to our own field studies. To test the transfer factors on data from other regions PCB levels in air and milk from roughly the same area and at roughly the same time were sought from the literature. Due to the lack of suitable, congener-specific, background level analyses of cows' milk only three sets of data on PCB levels in milk could be coupled with relevant reports of air analyses. First, a survey of PCB levels in air in the Czech Republic (*30*) was linked with a retail milk survey performed in Slovakia (*31*). Obviously, it should be borne in mind that these data sets come from different laboratories and different places, and that the studies were not designed for the purpose of investigating the air to milk transfers of PCBs. Two studies performed in southern Germany (*32, 33*) which provided PCB levels in milk were coupled with (vapor phase) air data collected from the same area (*32*).

Results and Discussion

Northwest England Farm Surveys. The ranges and average concentrations of persistent and semipersistent PCBs found in the 10 feed and 11 milk samples from the two surveys are shown in Table 2. It can be seen that for all congeners on both sampling occasions the standard deviation for each matrix, particularly the silage and feed samples, was higher (50−100%) than for milk. This may be due to silage being cut and stored under different conditions at each farm, and different sources of concentrate feed ingredients being used. In particular, variation in PCB concentrations in concentrate feed was expected due to the inclusion of fishmeal as a source of protein, which can originate from various parts of the world, and have widely different levels of PCB contamination (*4*). In the 1997 survey two of the farms (both within Greater Manchester) had higher (more than double) concentrations of PCBs in their silage than the others, and this was mirrored in higher PCB milk output fluxes. Only one of these farms was sampled in the 1996 survey, but it did not have noticeably elevated PCB levels in either silage or milk fat at that time. Of particular note is that one of the farms with elevated PCB levels in silage did not have noticeably elevated levels of PCBs in milk, but reported almost double the average daily milk fat output of other farms, illustrating a benefit of using CORs rather than BCFs.

Concentrations of persistent PCB congeners (i.e. PCBs-118, 138, 153, 170, 180, and 183) found in milk fat in both surveys are within the ranges found in United Kingdom retail milk in 1990 (*28*) and in raw milk in Northwest England in 1993/4 (*29*). Average levels of persistent congeners found in 1996 were between 35% lower and 25% higher than those reported in the two surveys previously published by other workers, and in 1997 on average 30−40% lower than those previously reported (see Table 5 later). These observations support the earlier comments that PCB concentrations in milk are quite stable spatially and temporally in the UK environment.

A summary of the BCFs and CORs calculated for all of the farms for which data was available for each survey is shown in Table 3. For the purpose of calculating CORs it was assumed that the total dry matter (DM) intake averaged 18 kg per day, and that concentrate feed constituted 6 kg of DM per day. For farms where milk fat yields were not available the average found in this study (0.74 kg per day) was used. BTFs (the ratio of the human foodstuff concentration to the total daily input flux) were not calculated because without individual feed intake data for each farm these would not give any more information than BCFs. It can be seen that the BCFs and CORs are comparable for each of the surveys and for both persistent and semipersistent congeners, with the exception of PCB 138. However the relative standard deviation for 1996 BCFs and CORs were up to twice as high as those for the 1997 survey. It can also be seen that both BCFs and CORs are relatively constant from less to more chlorinated congeners within the persistent congeners.

Principal components analysis of the BCF and COR values for the persistent congeners from each of the surveys did not show any consistent patterns to differentiate between any of the farms. There did not appear to be any correlation between PCB levels in silage and the output flux in milk for the first survey (all correlation coefficients—r^2—below 0.3),

TABLE 3. CORs and BCFs for the Northwest England Farm Surveys

PCB	1996 COR average	1996 COR RSD (%)	1996 BCF average	1996 BCF RSD (%)	1997 COR average	1997 COR RSD (%)	1997 BCF average	1997 BCF RSD (%)
47[b]	0.48	110	10	97	0.37	129	11	152
66[b]	0.39	81	8.5	61	0.07	29	1.7	34
74[b]	0.13	92	2.8	67	0.31	38	8	36
105[b]	0.37	61	8.4	47	0.37	23	10	31
128[b]	na[a]	na[a]	na[a]	na[a]	0.66	26	17	33
187[b]	0.12	85	2.8	77	0.07	51	1.8	57
118[c]	0.88	69	20	50	0.66	27	17	33
138[c]	1.40	65	33	64	0.57	31	15	37
153[c]	0.84	78	18	61	0.63	31	16	36
170[c]	0.74	56	17	51	0.78	35	20	38
180[c]	0.93	57	21	54	0.80	36	20	39
183[c]	0.76	58	18	52	0.92	37	23	43
average[d]	0.93		21		0.73		19	
RSD[d] %	26		27		17		17	

[a] Not analyzed. [b] Semipersistent congener. [c] Persistent congener. [d] Of persistent congeners only.

or between PCB levels in concentrate feeds and milk in the second survey. However, strong correlations were found for all persistent PCBs between levels of PCBs in silage and the output milk flux for the second survey (the correlation coefficients—r^2—ranged from 0.72 to 0.90).

It is strange that no correlation was found between PCB levels in silage and milk in the first survey, considering the strength of the correlation in the second survey. This may be due to the late sampling date (May) when most farms are ready to turn cows outside for the summer grazing. This would have meant that very little silage would have been left in the clamps of any of the farms, and some farms may have purchased extra silage from other sources to fill any shortfall in their own supply. The higher variability found in samples from the first survey than from the second may also be explained by this possibility. The good correlation between PCB levels in silage and the PCB fluxes in milk in the second study indicate that PCB intake from silage dominates over that from concentrate feed; this is consistent with detailed input flux calculations performed from the cow mass balance study (*4*).

The CORs measured in these surveys are very similar to those measured in the controlled feeding study (*12*) and by other workers (e.g., ref *34*). For example, in the 1997 survey the COR for PCB 138 was 0.57 ± 0.18, the average value derived from the controlled study on 5 lactating cows over 4 months was 0.74 while McLachlan (*34*) reported a value of 0.63 for one cow in a feeding trial. Other studies (e.g., refs *35* and *36*) have produced BCFs which are considerably lower than those measured here, but these studies utilized spiked feeds which, as mentioned previously, may not have the same bioavailability as naturally contaminated feeds, and must be fed for extended periods before steady-state will be achieved.

Long-Term Monitoring of One Farm. Pasture PCB concentrations at the farm were in good agreement with levels found at the adjacent field station. Milk fat PCB concentrations averaged 3.7 ng of $\sum$PCB g^{-1} of fat over the monitoring period (late April to middle October) dropping slightly throughout lactation, as noted in another study (*4*).

No significant correlations were found over time (13 separate samples) for levels of persistent PCBs in milk fat with the levels in either grass or air. Because no measurements of feed intake or fluctuations in milk fat output are available average values of 18 kg of DM intake and 0.63 kg of milk fat output (reported by the farmer) were used to calculate CORs. A summary of the CORs and BCFs calculated

TABLE 4. A Summary of CORs and BCFs Derived from the Long-Term Monitoring of One Farm

PCB	COR average	COR RSD (%)	BCF average (g of feed g of fat)	BCF RSD (%)
18	0.004	169	0.3	40
28	0.005	91	0.2	31
66	0.08	26	2	26
74	0.34	33	9.8	33
101	0.015	33	0.4	33
110	0.004	96	0.2	57
118[a]	0.74	66	21.2	66
138[a]	0.48	54	13.8	54
149	0.02	47	0.5	47
153[a]	0.46	48	13	48
170[a]	0.31	40	8.9	40
180[a]	0.27	27	7.6	27
183[a]	0.41	37	11.7	37
187	0.02	76	0.6	57
average[b]	0.45	45	13	45
RSD[b]	38	30	38	30

[a] Persistent congener. [b] Of persistent congeners only.

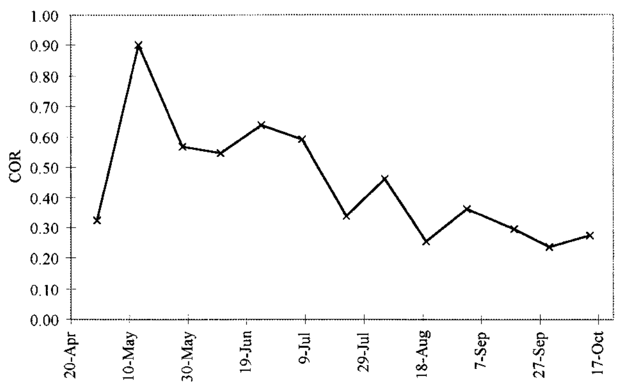

FIGURE 2. Average CORs for the persistent PCBs throughout the growing season.

using data on PCB levels in grass samples taken at the same time as the milk samples throughout the study period (from ref *11*) is shown in Table 4. Trends in the COR values throughout the study are shown in Figure 2; they are directly paralleled by the BCFs since there are no input or output data for particular sampling dates. It can be seen that because of the reduction in PCB concentrations in the milk throughout the growing season the COR values decrease by approximately 50% between the end of May and mid-October. The erratic first two measurements (April/May) probably reflect changes in the cows PCB balance as they go from winter husbandry to summer grazing conditions at this time. In April the cows had recently been put out to grass and would therefore be changing to a diet of fresh grass which will have been ungrazed since the autumn. At the time of the second sample (May) the cows would also have been reaching the highest weight loss period of the lactation cycle, and thus be mobilizing relatively large amounts of body fat, and therefore PCBs (*4*).

From Table 4 it can be seen that the RSDs for the CORs of each of the persistent PCB congeners over time are comparable to those found in the survey of farms at one time. However, the COR values range from approximately the same (for PCB 118) to approximately half (PCBs 170, 180, and 183) of the average CORs found for the farm survey and are approximately 0.3 lower than CORs calculated from the previous controlled feeding study (*12*).

Testing $TF_{A:M}$ on UK Data. The $TF_{A:M}$ values derived in Table 1 were used to predict milk concentrations for the

TABLE 5. Predicted Concentrations of Persistent PCBs in Milk Compared to Measured English Field Data (See Text for Details)

PCB	predicted (range) (pg/g of fat)	measured survey averages[a] 1996 (pg/g of fat)	1997 (pg/g of fat)	long-term[b] average (pg/g of fat)	1990 UK retail milk[c] (pg/g of fat)	1993 NW England[d] (pg/g of fat)
18	40 (0−170)	na[e]	55	41	na	na
28	60 (0−140)	na	45	32	280 ± 150	145 ± 96
66	190 (100−280)	104	90	na	na	230 ± 140
74	150 (90−210)	222	270	220	na	340 ± 150
101	50 (20−70)	na	22	23	150 ± 80	70 ± 39
110	20 (10−40)	na	7	8	na	65 ± 59
118	770 (410−1120)	1220	966	870	1540 ± 180	1240 ± 400
138	820 (380−1250)	1798	954	830	1640 ± 250	1430 ± 410
141	10 (0−30)	na	6	5	na	na
149	50 (20−90)	na	23	23	na	na
153	870 (470−1280)	1910	1070	780	1970 ± 290	1670 ± 430
170	210 (90−330)	289	213	150	na	320 ± 140
180	450 (280−630)	688	450	290	850 ± 180	720 ± 320
183	90 (30−150)	141	119	74	na	220 ± 79
187	30 (20−50)	na	25	13	na	150 ± 130

[a] From the Northwest England surveys. [b] From the local farm monitoring program. [c] From ref *28*, assumes 3.9% average milk fat content of whole milk. [d] From ref *29*. [e] Not available.

TABLE 6. Testing of Air to Milk Transfer Factors on Air and Milk Data Sets from Different Countries

matrix	place and year	units	PCB 138	PCB 153	PCB 180
air	Czech Rep. 1990−91[a]	pg m^{-3}	70	69	30
milk	*Czech Rep. predicted*	*ng (kg of fat)$^{-1}$*	*26 ± 14*	*18 ± 8*	*16 ± 6*
milk	Slovakia <1995[b]	ng (kg of fat)$^{-1}$	195	27	37
air	Bayreuth 1989[c]	pg m^{-3}	14	20	4.2
milk	*S. Germany predicted*	*pg (g of fat)$^{-1}$*	*5200 ± 2800*	*5200 ± 2400*	*2300 ± 880*
milk	Bayreuth 1990[c]	pg (g of fat)$^{-1}$	4400	5900	1900
milk	Augsburg 1992−93[d]	pg (g of fat)$^{-1}$	1900−5600	2300−6600	890−2300

[a] From ref *30*. [b] From ref *31*. [c] From ref *32*. [d] From ref *33*.

1996 and 1997 data sets from air concentrations measured at the semirural/urban Lancaster site in 1996/7 and tested against the data obtained from the two farm surveys (Table 2) and the long-term monitoring study at the local farm reported earlier. The results of this comparison are shown in Table 5; the predicted range is the predicted mean ± the estimated standard deviation of $TF_{A:M}$ (see earlier explanation). It can be seen that there is very good agreement between predicted and measured data for the 1997 survey and especially for the long-term monitoring study (probably illustrating the benefit of using PCB data from air samples taken during the correct growing season). The 1996 survey data for the persistent congeners 138, 153, and 180 was predicted less well, at approximately half of the measured values found in the 1996 survey. However, as mentioned earlier, the time of this survey could have caused the silage samples taken to be unrepresentative of the cows' normal diet. Persistent congener levels for both of the UK milk surveys taken from the literature are underpredicted by up to 50%, comparable to the 1996 survey, but obviously the spatial resolution in air data was necessarily compromised by the need to use air concentrations from the Lancaster site.

Testing $TF_{A:M}$ on Data Sets from Other Countries. The $TF_{A:M}$ were applied to data from other workers where levels of PCBs in both air and milk fat were found in the literature for the same region (Table 6). It was only possible to find relevant (matched) data for the persistent congeners 138, 153, and 180. PCB levels in air in various regions of the Czech Republic (*30*) were found to underpredict PCB levels in a milk survey in Slovakia (*31*) by up to 50% for congeners 153 and 180, but by a factor of ~7 for PCB 138. It is possible that this reflects analytical problems associated with PCB 138 in the milk survey, as it is generally reported in the literature that PCBs 138 and 153 are present at roughly the same levels in milk. PCB levels in milk predicted from an air data set in southern Germany (*32*) gave very good agreement with two German studies of PCB levels in milk (*32, 33*). The PCB levels in milk reported in the earlier study fitted the average predicted values very closely, while ranges of levels found in the later study showed considerable overlap with the predicted ranges. The lower limit of the range of PCB levels found in the later study was up to 35% lower than the lower end of the predicted range.

In summary, the predictive capabilities of the derived $TF_{A:M}$ values was considered to be excellent for the persistent PCB congeners, when appropriate air data and analytically matched milk data sets were available. This appears to be the case even when the cows may have been under different husbandry regimes from those used to derive $TF_{A:M}$ values, and where the air data were not closely matched in time.

Final Remarks

Several points of importance to foodstuff monitoring programs and future considerations for predicting the transfer of POPs to humans should be made. First, it was found that persistent PCB levels in silage directly correlate with milk fluxes, implying that steady state is approached for these chemicals, and therefore that persistent chemicals with similar physicochemical properties could be expected to behave similarly.

An important finding for the design of monitoring programs was that variability in BCFs measured over time and between different herds was comparable. Only one instance of a clear advantage of using CORs was evident, when silage levels were elevated and milk fat flux was twice the average.

Finally, and importantly, $TF_{A:M}$ values have been derived and shown to be an effective tool to predict milk concentrations from average air concentrations at the regional scale, to well within an order of magnitude. Indeed, if air data are available more locally, $TF_{A:M}$ can be used to predict the milk concentrations of persistent congeners to within a factor of ~2−3. The main requirements for this approach to monitoring and predicting PCB levels in milk fat are that (i) pasture is the dominant feed; (ii) silage fed in winter is grown locally to the site of interest; and (iii) there is no local intermittent source of PCBs (i.e., seasonally steady-state conditions of air−pasture transfer apply).

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