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ORIGINAL RESEARCH

A national survey of persistent, bioaccumulative, and toxic (PBT) pollutants in the United States milk supply

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This study measured 21 persistent, bioaccumulative, and toxic (PBT) pollutants in the US milk supply. Since milk fat is likely to be among the highest dietary sources of exposure to PBTs, it is important to understand their levels in this food. Nationwide samples were collected from 45 dairy plants in July of 2000 and again in January 2001. The levels of all chemicals in the chlorobenzene, pesticide and other halogenated organic groups were determined to be below their detection limits in all samples. National averages were computed for 11 chemicals or chemical groups found above the detection limits. The national average CDD/CDF and PCB TEQ concentrations were 14.30 and 8.64 pg/l, respectively, for a total of 22.94 pg/l. These levels are about half the values found in a similar study conducted in 1996. If this difference is in fact indicative of declining milk levels and assuming exposure levels from nondairy pathways have remained the same over this time period, this would result in an overall decrease in adult background dioxin exposure of 14%. Six PAHs were detected with national averages ranging from 40 to 777 ng/l. Cadmium concentrations ranged from 150 to 870 ng/l with a national average of 360 ng/l. Lead concentrations were consistently higher than those of cadmium, ranging from 630 to 1950 ng/l with a national average of 830 ng/l. PAHs showed the strongest seasonal/geographic differences, with higher levels in winter than summer, north than south and east than west. Average adult daily intakes from total milk fat ingestion were computed for all detected compounds and compared to total intakes from all pathways: CDD/CDF/PCB TEQs: 8 vs. 55 pg/day, PAHs: 0.6 vs. 3 μg/day, lead: 0.14 vs. 4–6 μg/day, and cadmium: 0.06 vs. 30 μg/day.
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Introduction

About 12% of the total fat ingested by Americans is from milk (USEPA, 1997). EPA estimates that on average the portion of total daily intake of dioxin (based on toxic equivalents (TEQ) — see Results for further details) from all dairy products is about 30% for adults and 50% for children (USEPA, 2000). Like dioxin, the other persistent, bioaccumulative, and toxic pollutants (PBTs) tend to be widely dispersed in the environment, bioaccumulated through the food chain and ultimately result in low-level contamination in most animal fats. Since milk fat is likely to be among the highest dietary sources of exposure to PBTs, it is important to understand their levels in this food. A second reason to study milk is that it offers the opportunity to examine geographic variability. Other animal fats are nationally distributed and difficult to trace back to a specific region. Milk, however, is produced and distributed regionally. Understanding regional variability may offer clues to sources which release these compounds and processes by which they enter our food supply.

Milk samples for this study were obtained through the EPA Environmental Radiation Ambient Monitoring System (ERAMS), an existing monitoring program that collected additional milk samples for the purposes of this study during two of its already established sampling intervals (USEPA, 1988). Although established and operating for a purpose different from this study's objective, ERAMS afforded an ideal collection network for milk samples for this study. The ERAMS program comprises a national network of monitoring stations that, since 1973, have regularly collected air, water, precipitation, and milk samples from which environmental radiation levels are derived. The major emphasis for

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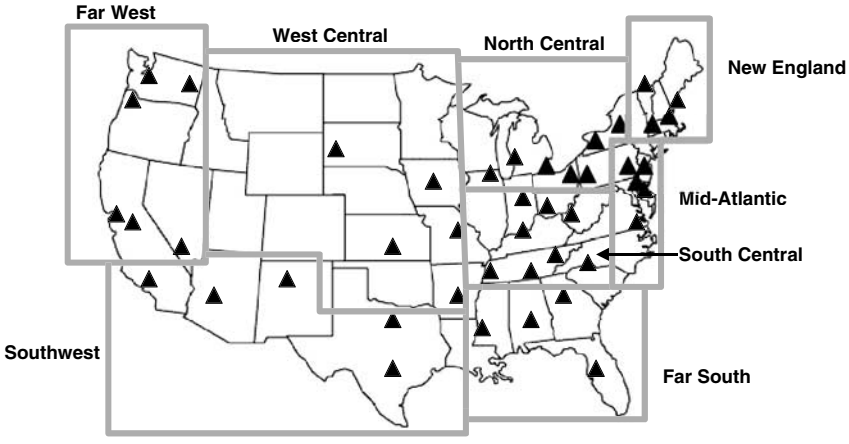
ERAMS is upon identifying trends in the accumulation of long-lived radionuclides in these matrices. For its milk collection effort, ERAMS has 46 sampling stations located within the major population centers of 41 (of 50) US states, and Puerto Rico (Figure 1). Individual stations collect milk four times per year, and send it to a central EPA ERAMS facility located in Montgomery, AL, USA. The milk sample from each ERAMS station is a proportional composite from large dairy plants supplying the population centers; that is, the amount of milk obtained from each of the dairy plants contributing to the sample is roughly proportional to the amount of milk the dairy plant supplies to the region. It is estimated that the ERAMS milk samples represent roughly 20% of the US milk supply.

The primary purpose of this study was to estimate the average concentrations of PBTs in the general pasteurized milk supply of the United States. Secondary objectives were to make preliminary evaluations of the seasonal and geographic differences of the levels of these compounds in milk. The PBTs selected for this study were ones that are currently regulated or targeted for regulation (either in the US or internationally); identified as a PBT of concern in the current scientific literature; and/or were not already included in the USDA Pesticide Data Program's milk survey (USDA, 1998). Originally, 37 chemicals were identified that met these criteria; however, a number of these were eliminated because of variable recoveries in the chemical analyses. The 21 chemicals addressed in this study are listed in Table 1 and include the chlorinated dibenzo-*p*-dioxins (CDDs), chlorinated dibenzofurans (CDFs), dioxin-like polychlorinated biphenyls (dioxin-like PCBs), polyaromatic hydrocarbons (PAHs), lead and cadmium. The chemicals that were excluded were: several polychlorinated naphthalenes and brominated diphenyl ethers, chlordecone, pendimethalin, 2,4,5-trichlorophenol, hexachlorobutadiene, benzo(g,h,i) perylene, and mercury.

Lorber et al. (1998) conducted a similar study in 1996. This study has essentially the same purpose as the earlier study, but has been expanded to address additional PBTs (besides CDDs, CDFs, and PCBs) and to provide a more current assessment.

Methods

The existing ERAMS milk collection network was used to collect samples in July 2000 and again in January 2001. The sampling equipment described below was mailed from the National Air and Radiation Environmental Laboratory in Montgomery, AL to each ERAMS station. Three liters (l) of milk were collected by each ERAMS station using 1-l high-density polyethylene jars precleaned to meet EPA's "Specification and Guidance for Contaminant- Free Sample Containers" OSWER Directive #9240.0-05A, December



Individual ERAMS Stations within the eight regional composites

New England: Hartford, Boston, Portland, Montpelier

Mid-Atlantic: Trenton, Dover, Baltimore, Philadelphia, Norfolk

South Central: Charleston, Cincinnati, Louisville, Charlotte, Chattanooga, Knoxville, Memphis, Indianapolis

North Central: Buffalo, Syracuse, Pittsburgh, Detroit, Grand Rapids, Cleveland, Chicago

West Central: Des Moines, Wichita, Jefferson City, Rapid City, Little Rock

Southwest: Phoenix, Albuquerque, San Antonio, Ft. Worth, Los Angeles

Far South: Montgomery, Tampa, Atlanta, Jackson, San Juan Puerto Rico (not shown)

Far West: Portland, Tacoma, Spokane, Sacramento, San Francisco, Las Vegas, Honolulu (not shown)

Figure 1. ERAMS milk sample collection points.

Table 1. Analytical methods and detection limits

Chemical name	Analytical method	LOD[a]	Percent recovery (LFM[b], MS[c])	RPD[d] (duplicate samples)
CDDs/CDFs/PCBs:		pg/l		
Chlorinated dibenzo-*p*-dioxins (CDDs)	Analytical method for the determination of individual CDD/CDF and co-planar PCB congeners in milk samples	1–70	72	22
Chlorinated dibenzofurans (CDFs)		1–70	75	20
Polychlorinated biphenyls (PCBs)		5–1000	82	10
CDD/CDF/PCB Total TEQ				5–16
Chlorobenzenes		ng/l		
Tetrachlorobenzene, 1,2,4,5-	Battelle method for miscellaneous organics extraction and clean-up procedure[e]	50	42, 48	
Trichlorobenzene, 1,2,4-		80	52, 60	26
Other halogenated organics		ng/l		
Bromophenyl phenyl ether, 4-Octachlorostyrene	Battelle method for miscellaneous organics extraction and clean-up procedure[e]	140	59, 52	
		80	52, 60	
Pesticides		ng/l		
Endrin	Battelle method for miscellaneous organics extraction and clean-up procedure[e]	300	72, 89	
Mirex		190	54, 66	
Pentachloronitrobenzene		1020	39, 53	
Pentachlorophenol		40	74, 117	
PAHs		ng/l		
Acenaphthene	Battelle method for miscellaneous organics extraction and clean-up procedure[e]	180	56, 108	
Acenapthylene		40	0, 72	
Anthracene		100	106, 69	
Benzo-a-pyrene		1000	40, 39	
Fluorene		70	45, 58	48
Naphthalene		70[g]	29, 61	49
Phenanthrene		60[f]	50, 65	40
Pyrene		40	91, 79	12
Metals		ng/l		
Cadmium	USEPA SW 846 test methods for evaluation of solid waste, method 3050B	10	96	5
Lead-alkyl		10	96	6

[a]LOD — limit of detection is in pg/l for CDDs/CDFs/PCBs and ng/l for all other compounds. Values were adjusted if recovery from LFM was <70%.
[b]LFM — laboratory fortified matrix (store-bought milk).
[c]MS — matrix spike (field sample). Acceptance critiera for recovery was 40–120% for ogranics and 50–200% for metals.
[d]RPD — relative percent difference, blanks indicate that it could not be calculated. Acceptance critieria was <30%.
[e]Based on mass spectra response.
[f]GC/MS conditions: column: Rtx- 5 Sil, 30 m × 0.25 mm ID; 0.5 μm film thickness; carrier Gas: He at 1 ml/min; oven temperature program: 50 C for 4 min, 50–140°C at 12°C/min, 140–310 at 8C/min, 310–330 at 20°C/min; hold 330 for 6.25 min; inlet temperature: 330°C; injection: 2 *ul* splitless for 1 min; transfer line temperature: 330°C.

1992. The earlier survey (Lorber et al., 1998) used formaldehyde to preserve collected milk samples, and a number of the samples contained mold and other evidence of degradation. Therefore, it was decided to try cooling as an alternative method of sample preservation for this study. The collectors put the jars into insulated bioshippers along with six refrigerant packs ($0.75 \times 4 \times 6\,in^3$) and sent them by overnight mail to the Environmental Chemistry Laboratory in Stennis Space Center, MS, USA, where one jar was removed from each bioshipper and stored in a freezer. The

remaining samples were shipped by overnight mail to Battelle Memorial Institute in Columbus, OH. Battelle stored all remaining samples in their freezer.

A total of 45 ERAMS stations provided milk samples that were collected in the first sampling interval and 44 in the second interval. Eight composite samples were then prepared by geographic region for each sampling interval, as shown in Figure 1. The eight geographic regions were: New England, Mid-Atlantic, South Central, North Central, West Central, Southwest, Far South, and Far West. The regional composites were prepared by combining equal amounts of the four to eight individual milk samples making up the geographic region, to total approximately 1 l. The purpose of these regional composites was to look for geographic variability. One grand composite was also prepared for each sampling interval. The grand composites were created by combining amounts from each sample adjusted on the basis of relative milk production represented by each ERAMS station. The purpose of the grand composites was to provide estimates of the national average.

The Environmental Chemistry Laboratory in Stennis Space Center, MS conducted the analysis for the CDDs/CDFs and PCBs. Battelle Memorial Institute analyzed the milk samples for the other PBTs listed in Table 1.

The analytical methods are summarized below for each of the chemical groups. Selected information, including detection limits and recoveries, are shown in Table 1. The relative percent difference (RPD) is listed for all chemicals measured above the detection limit in the duplicate samples, and was calculated as the difference between concentrations in duplicate samples divided by the mean of the concentrations ($\times 100$ to express as a percentage). The RPD is useful in judging whether a difference in measurements reflect real differences or simply the limits in the precision of the analytical method.

CDDs/CDFs/PCBs

Sample analyses for the CDDs/CDFs and PCBs were based on methods described previously (Ferrario et al., 1997; Lorber et al., 1998). Milk samples were stored at 4°C and extracted within 4 h of removal from storage. Briefly, 300 ml subsamples were liquid–liquid extracted with hexane after being basified and denatured by the addition of potassium hydroxide and ethyl alcohol. The hexane extracts were combined, dried over anhydrous sodium sulfate and the lipid removed by stirring the crude extract with acidified silica gel. The samples were further cleaned up using combined acid/base silica gel columns and neutral alumina columns. The PCBs were separated from the CDDs/CDFs using PX-21 graphitized carbon columns. Following extraction, sample extracts were stored at 4°C until analysis. Samples were analyzed using high-resolution gas chromatography/high-resolution mass spectrometry (GC/MS). Details of the chromatographic conditions and mass spectrometer acquisition parameters are provided in Ferrario et al. (1996).

Chlorobenzenes, other halogenated organics, pesticides, and PAHs

The extraction procedure for these miscellaneous organic analytes was developed previously at Battelle for the analysis of pesticides and PAHs in food matrices (Chuang et al., 1999). The method, as applied here, entailed homogenizing a 25-ml aliquot of milk sample with dichloromethane. The sample was centrifuged and the liquid layers were transferred to a separatory funnel. The dichloromethane was drained through a funnel containing sodium sulfate to remove water from the sample, and then concentrated. The residue weight was determined on a 100 μl aliquot to calculate the amount of fat in the sample. The dichloromethane sample extract was passed through a gel-permeation chromatography column; the eluent in the first 30 min, containing fats, was discarded. The remainder of the eluent was collected and concentrated. Both neutral and acidic analytes were contained in the extract; however, at this point the sample extract was split into two equal aliquots for separate analyses. The sample split was necessary because excess derivatizing agent in the sample interfered with the chromatography of some neutral analytes. One aliquot for the neutral analytes was applied directly to a Florisil column. The aliquot for the acidic analytes was derivatized with diazomethane first and then applied to a Florisil column. The acids were derivatized prior to application to the Florisil column because they would have irreversibly adsorbed to the Florisil column in their acidic state. The two separate extracts were concentrated after Florisil clean-up, spiked with internal standard, and analyzed using low-resolution gas chromatography/low-resolution mass spectrometry (GC/MS) equipped with a mass selective detector in the multiple ion detection mode.

This extraction procedure included spiking a surrogate recovery standard into the sample prior to extraction. The recovery of the surrogate standard was used as an indicator of the extraction efficiency and cleanup recovery of the analytical method. The compound used for the surrogate was fenchlorphos, a moderately polar organophosphate compound that is structurally similar to many pesticides currently on the market, but no longer approved for use in the US. Fenchlorphos was used as the surrogate standard when this method was originally developed at Battelle for detection of diverse semivolatile organic compounds (including PAHs, PCBs, phthalate esters, and organophosphate insecticides) in food. Owing its stability and robustness as an indicator of method performance, it was used here as the surrogate recovery standard for the miscellaneous organic extraction procedure.

Lead and cadmium

Inorganic lead and cadmium were digested according to EPA SW-846 Method 3050B (USEPA, 1996, Rev. 2). The method consisted of digesting 100-ml whole milk in acid, filtering, and analyzing the sample using inductively coupled

plasma mass spectrometry (ICP/MS), which is EPA SW-846 Method 6020 (USEPA, 1994).

Results

The results for each group of chemicals are discussed below in terms of the national average (as represented by the mean of the seasonal grand composites), range of results, geographic comparisons, and seasonal comparisons. Geographic comparisons were made by lumping the regions into north–south and east–west divisions as follows:

Northern division — New England, Mid-Atlantic, North Central, West Central, and Far West

Southern division — South Central, Southwest, and Far South

Eastern division — New England, Mid-Atlantic, South Central, North Central, and Far South

Western division — West Central, Southwest, and Far West

The levels of all chemicals in the chlorobenzene, pesticide, and other halogenated organic groups were determined to be below their detection limits in all samples (detection limits are listed in Table 1). The results for all other chemicals are presented below. All contaminant concentrations are reported in units of mass of contaminant per liter of milk as collected.

CDDs/CDFs and PCBs

Results from the survey are shown in Tables 2 and 3. All CDD/CDF and PCB concentrations were converted to the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) TEQ using the 1998 WHO toxic equivalency factors (TEFs) for PCBs, CDDs, and CDFs (Van den Berg et al., 1998). Since EPA and others believe that dioxins share a common mechanism of toxicity, scientists have developed an approach that adds together the toxicity of individual dioxins, accounting for the differing toxicity of each dioxin through the use of TEFs. Given these TEFs, the toxicity of a mixture of dioxins can be expressed in terms of its TEQ, which is the amount of 2,3,7,8-TCDD it would take to equal the combined toxic effect of all the dioxin-related compounds found in that mixture. This paper uses the convention of listing CDD, CDF, and PCBs with slashes in between them, to show the combination of chemicals that were considered in computing the TEQ of the mixture. Most TEQ estimates presented in this paper consider contributions from all three sets of compounds (designated CDD/CDF/PCB). In some cases, though, the paper reports CDD/CDF values separately from PCB values since the two sets are associated with different sources and their relative contributions to total TEQ are of interest to many researchers.

Results were calculated by assigning nondetectable (ND) amounts a value of one-half the detection limit. The TEQ estimates for total CDDs/CDFs assuming nondetects equal to zero were 15% lower than the estimate assuming nondetects equal to half the detection limit. For the PCBs, the nondetects had no impact on the TEQ estimates.

Based on the grand composites from both seasons, the national average CDD/CDF and PCB TEQ concentrations were 14.30 and 8.64 pg/l, respectively, for a total of 22.94 pg/l (Table 3). The national average concentrations of the individual congeners found in the milk samples are shown in Table 3. Levels of four of these congeners were clearly higher than the rest, all of them at TEQ concentrations above 2 pg/l and making up 70% of the total TEQ. Their relative

Table 2. TEQ concentrations of CDD/CDFs and PCBs in Milk (pg/l)

Composite location	CDD/CDF			PCB			Total[a]
	Jul 2000	Jan 2001	Mean [b]	Jul 2000	Jan 2001	Mean[b]	Mean[b]
New England	12.85	8.89	10.87	9.53	8.29	8.91	19.78
Mid-Atlantic	11.67	14.21	12.94	14.37	8.30	11.34	24.28
South Central	17.53	19.14	18.34	6.93	9.57	8.24	26.59
North Central	20.48	10.35	15.42	11.36	7.13	9.25	24.66
West Central	17.94	18.59	18.27	5.31	5.16	5.24	23.50
Southwest	13.27	6.21	9.74	6.71	6.29	6.50	16.24
Far South	18.13	35.82	26.98	8.07	7.59	7.83	34.81
Far West	22.50	13.20	17.85	8.80	8.33	8.57	26.42
Regional mean[c]	16.80	15.80	16.30	8.89	7.58	8.23	24.53
Grand composite mean[d]	18.70	8.89	14.30	8.70	8.58	8.64	22.94

[a]This is the sum of the CDD/CDF seasonal mean and the PCB seasonal mean.

[b]Average of summer and winter concentrations.

[c]The regional mean is the average concentration in the eight regional composites; regional composites were made up by combining equal amounts of milk from the ERAMS stations located in the specified region.

[d]The grand composite mean is the average concentration from duplicate analyses; grand composites were created by combining amounts from each sample adjusted on the basis of relative milk production represented by each ERAMS station.

Table 3. Average congener concentrations in milk (pg/l)[a]

	WHO 98 TEFs	Concentration (pg/l)	Concentration (pg TEQ/l)
2,3,7,8-TCDD	1	0.61	0.61
1,2,3,7,8-PeCDD	1	4.25	4.25
1,2,3,4,7,8-HxCDD	0.1	4.13	0.41
1,2,3,6,7,8-HxCDD	0.1	29.25	2.93
1,2,3,7,8,9-HxCDD	0.1	7.69	0.77
1,2,3,4,6,7,8-HpCDD	0.01	80.13	0.80
OCDD	0.0001	37.28	0.00
2,3,7,8-TCDF	0.1	0.41	0.04
1,2,3,7,8-PeCDF	0.05	2.00	0.10
2,3,4,7,8-PeCDF	0.5	4.68	2.34
1,2,3,4,7,8-HxCDF	0.1	7.78	0.78
1,2,3,6,7,8-HxCDF	0.1	4.76	0.48
1,2,3,7,8,9-HxCDF	0.1	4.23	0.42
2,3,4,6,7,8-HxCDF	0.1	2.00	0.20
1,2,3,4,6,7,8-HpCDF	0.01	14.42	0.14
1,2,3,4,7,8,9-HpCDF	0.01	2.00	0.02
OCDF	0.001	3.50	0.00
PCB 77	0.0001	71.11	0.01
PCB 118	0.0001	9642.88	0.96
PCB 105	0.0001	2813.71	0.28
PCB 126	0.1	65.41	6.54
PCB 156	0.0005	1233.11	0.62
PCB 157	0.0005	298.81	0.15
PCB 169	0.01	8.21	0.08
Total CDD/CDFs			14.30
Total PCBs			8.64
Total CDD/CDF/PCBs			22.94

[a]Average of summer and winter grand composite concentrations.

contributions to total TEQs are as follows: 1,2,3,7,8-PeCDD (18.5%); 1,2,3,6,7,8-HxCDD (12.8%); 2,3,4,7,8-PeCDF (10.2%); and PCB 126 (28.5%). With the exception of 2,3,7,8-TCDD, the dominant congeners found in these milk samples are the same congeners that are expected to contribute the most to background human exposure, in terms of both body burden and TEQ dose (USEPA, 2000).

Comparing these concentrations to those found in the earlier national survey of dioxin-like compounds in the US milk supply conducted in 1996 (Lorber et al. (1998); results adjusted to 3.2% lipid and using WHO 1998 TEFs), suggests that CDD/CDF/PCB TEQs have declined by about 50%. In the earlier study, the national average TEQ concentration of CDD/CDFs was 31 pg/l, slightly more than double the national average of 14.30 pg/l found in this study. Similarly, for the PCBs the earlier study's national average TEQ concentration of 16 pg/l is almost double the 8.64 pg/l national average TEQ concentration found in the current study. While this 50% decline is uncertain because of difficulty in establishing lipid levels in both studies, it is based on a difference that exceeds the RPD for the TEQs of 5–16% (Table 1). The comparability of the two studies was also evaluated on the basis of individual congeners. No significant changes were noted in the percent contribution of individual congeners to total TEQ.

Figure 2 illustrates the seasonal and broad regional averages along with the percent differences for both the CDD/CDFs and PCBs. The data in this study did not show large seasonal difference for either the CDD/CDFs or PCBs which matched the findings by Lorber et al. (1998).

No large geographic differences were observed for the CDD/CDFs, although the southern division was 20% higher than the northern division (which exceeds the CDD/CDF RPD). The highest CDD/CDF levels were observed in the Far South and South Central and the lowest levels in the Southwest. Lorber et al. (1998) made similar observations stating that the highest levels were in the Southeast (comparable to the area designated as Far South in this study) and the lowest in the Southwest.

No large geographic differences were observed for the PCBs, although the eastern division was 25% higher than the western division (which exceeds the CDD/CDF RPD) The highest PCB level was observed in the Mid-Atlantic composite and the lowest level was found in the West Central composite. The comparison across broad geographic divisions showed no evidence of significant differences (Figure 2). Lorber et al. (1998) also found weak evidence of geographic differences for PCBs, with levels highest in the Northwest and lowest in the Southeast. This pattern appears to be somewhat different than the one seen here, but differences in regional compositing between the two studies make these comparisons difficult.

PAHs

The milk samples were analyzed for eight PAHs as shown in Table 4. Based on the grand composite means across the seasons, the national average PAH levels ranged from nondetect (40 ng/l) to 777 ng/l. Naphthalene and phenanthrene had the highest grand composite values in both winter and summer. Acenaphthene and benzo(a)pyrene were not detected in any samples. Observations for the six detected PAHs are discussed below.

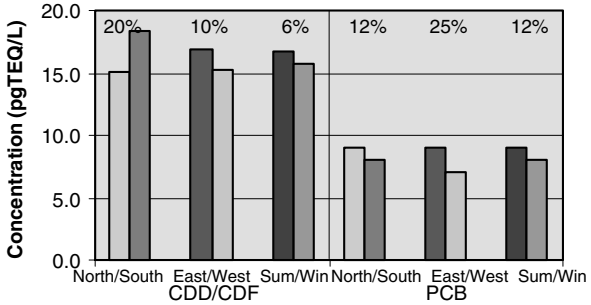


Figure 2. Geographic and seasonal differences for CDD/CDFs and PCBs (relative percent differences listed above data pairs).

Table 4. Concentrations of PAHs in milk (ng/L)

	Acenaph-thene	Anenaph-thylene	Anthracene	Benzo (a) pyrene	Fluorene	Naphthalene	Phenan-threne	Pyrene
July 2000								
New England	ND[a]	ND	ND	ND	[37.44]	545.86	165.60	69.60
Mid-Atlantic	ND	40.32	[45.60][b]	ND	182.40	1049.86	645.56	120.00
South Central	ND	ND	[37.92]	ND	[50.88]	546.34	240.95	76.32
North Central[c]	ND	ND	ND	ND	[36.00]	597.22	147.84	58.08
West Central	ND	58.08	ND	ND	[52.80]	871.30	220.80	79.20
Southwest	ND	ND	[62.40]	ND	534.72	550.66	3246.72	202.08
Far South	ND	ND	ND	ND	ND	749.86	208.80	70.08
Far West	ND	44.64	ND	ND	[57.60]	947.14	431.04	98.88
Regional mean	ND	30.38	49.49	ND	123.36	732.28	663.54	96.78
Grand composite[d]	ND	ND	[49.92]	ND	237.12	551.62	1124.16	174.24
January 2001								
New England	ND[a]	828.00	1404.00	ND	2529.60	3095.14	35943.84	630.72
Mid-Atlantic	ND	68.64	[63.84]	ND	436.32	1099.30	3826.24	295.68
South Central	ND	40.32	ND	ND	153.60	855.94	745.44	150.24
North Central[c]	ND	46.08	[42.24]	ND	141.60	991.78	594.72	133.92
West Central	ND	58.56	[60.48]	ND	468.00	1032.10	2895.84	306.24
Southwest	ND	57.12	[51.84]	ND	341.28	828.58	1909.44	126.24
Far South	ND	BLOQ[e]	[39.36]	ND	97.44	582.82	581.76	110.88
Far West	ND	39.84	[40.80]	ND	127.20	882.34	640.80	151.20
Regional mean	ND	144.82	219.07	ND	536.88	1171.00	5767.26	238.14
Grand composite[d]	ND	BLOQ	ND	ND	119.52	644.26	430.08	107.52
Grand composite mean[f]	ND	ND	34.96	ND	178.32	597.94	777.12	140.88

[a]Not detected; detection limits in ng/l milk: acenaphthene — 180, acenaphthylene — 40, anthracene — 100, benzo(a)pyrene — 1000, benzo(g,h,i)perylene — 630, fluorene — 70, naphthalene — 70, phenanthrene — 60, pyrene — 40. Half these values were used in calculating regional means.
[b]Values in brackets indicate that the concentration is below the limit of detection, but that the peak could be identified and estimated.
[c]Average of duplicate analyses.
[d]The grand composites were created by combining amounts from each sample adjusted on the basis of relative milk production represented by each ERAMS station.
[e]BLOQ = below the limit of quantitation, indicates that peak seen but below limit of detection and could not be estimated.
[f]The grand composite mean is the average of the two seasons.

The New England winter sample was clearly elevated above the others with values two to five times the winter regional means. In summer, however, the New England sample was similar to the others.

Consistent evidence of higher levels in the Winter than Summer was observed for all six PAHs (Figure 3a). Similarly, consistent geographic patterns were observed for all 6 PAHs with higher levels in east than west and north than south (Figure 3b).

Metals

Concentrations of cadmium and lead in milk are shown in Table 5. Figure 4 illustrates the seasonal and broad regional averages along with the percent differences for both cadmium and lead. Cadmium concentrations ranged from 150 to 870 ng/l with a grand composite mean of 360 ng/l. Higher levels were observed in the south than north, west than east and summer than winter.

Lead concentrations were consistently higher than those of cadmium for both sampling intervals, ranging from 630 to 1950 ng/l with a grand composite mean of 830 ng/l. Higher levels were observed in the south than north. The seasonal difference was relatively small.

Discussion

In conclusion, this study measured 21 persistent, bio-accumulative, and toxic (PBT) pollutants in the US milk supply and estimated national averages for 11 chemicals or chemical groups found above the detection limits. The discussion below addresses sampling issues and provides

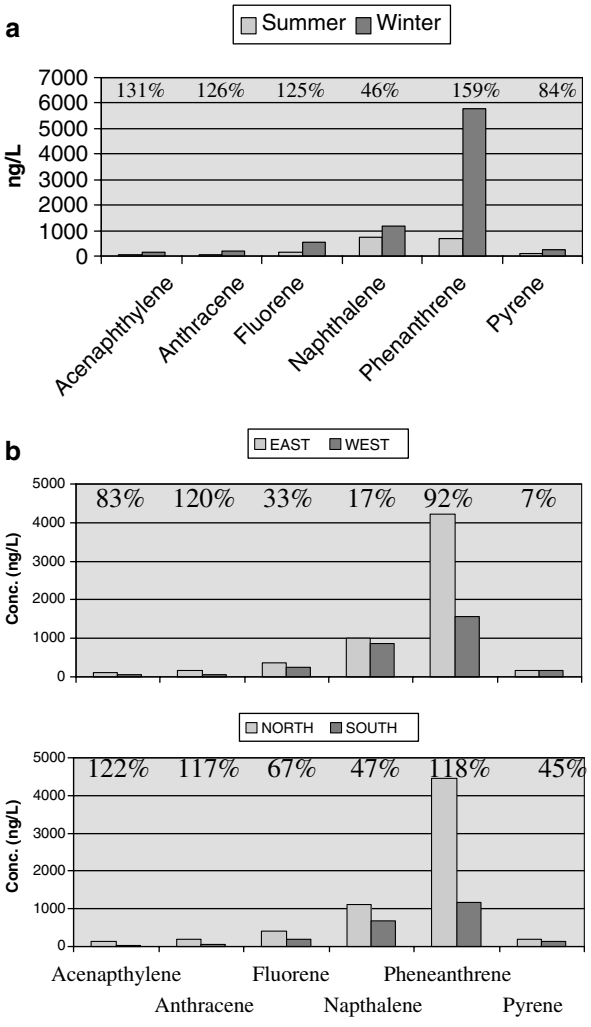


Figure 3. (a) Seasonal differences for PAHs in ng/l (relative percent differences listed above data pairs). (b) Geographic differences for PAHs in ng/l (relative percent differences listed above data pairs).

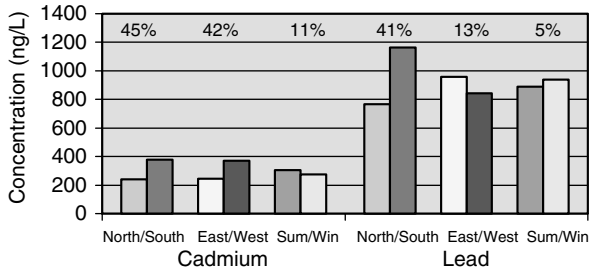


Figure 4. Geographic and seasonal differences for cadmium and lead (relative percent differences listed above data pairs).

some perspective on the levels found in milk by comparing the intake of these chemicals from milk to total exposure.

Sampling

This study used cooling to preserve samples rather than formaldehyde, which was used in the previous study. While this solved the problems of mold formation and possible chemical degradation, the freezing and thawing of the whole milk samples left the samples in a nonhomogenous state. It is recommended that in any future studies, the milk samples be kept refrigerated (not frozen) and that analyses be conducted as soon as possible after sample collection.

Exposure perspectives

The potential exposures resulting from the detected PBTs in milk were estimated using the means of the grand composites and assuming long-term ingestion at these levels. The lipophilic compounds (i.e. all the chemicals in this study except the metals) were assumed to partition completely into the milk fat. The concentrations of these chemicals in milk fat were computed using the estimated fat content of 3% by weight. This concentration was multiplied by the average

Table 5. Concentrations of metals in milk (ng/l)

Composite location	Cadmium			Lead		
	Jul 2000	Jan 2001	Mean	Jul 2000	Jan 2001	Mean
New England	195[a]	290[a]	243	665[a]	870[a]	768
Mid-Atlantic	170	280	225	660	1060	860
South Central	210	280	245	690	670	680
North Central	170	280	225	790	680	735
West Central	440	190	315	880	680	780
Southwest	870	350	610	750	1370	1060
Far south	250	300	275	1950	1540	1745
Far west	150	220	185	730	640	685
Regional mean	307	274		889	939	
Grand composite[b]	250	470	360	680	980	830

[a]Average of duplicate analyses.

[b]The grand composites were created by combining amounts from each sample adjusted on the basis of relative milk production represented by each ERAMS station.

adult daily ingestion rate for all milk fats (includes milk, cheese, milk desserts, and yogurt) which has been estimated as 9.8 g/day (USEPA, 2000). For the nonlipophilic compounds, the daily intake was estimated as the whole milk concentration multiplied by the average adult milk ingestion rate of 0.17 L/day (USEPA, 1997). The other milk products are also likely to contain some of these PBTs, but no reliable method was available to estimate these levels. The discussion below compares the intake of these chemicals from milk to total intake.

Dioxin Using the above procedure, the daily intake of CDD/CDF/PCB TEQs from ingestion of all milk fats was estimated to be 8 pg/day. The average background adult intake for typical exposures to CDD/CDF/PCBs from all pathways is estimated to be 65 pg TEQ_{WHO98}/day (USEPA, 2000). This estimate is based on food data collected primarily in the mid-1990s. The Draft Reassessment estimates that ingestion of milk and dairy products constitutes approximately 28% of the total adult daily intake of CDD/CDF/PCB TEQs. This study found CDD/CDF/PCB TEQ levels in milk to be about 50% lower than those reported in a similar study conducted in 1996 Lorber et al., 1998). If this difference is truly indicative of declining milk levels and assuming exposure levels from nondairy pathways have remained the same over this time period, this would result in an overall decrease in adult background CDD/CDF/PCB TEQ exposure of 15%. Several factors could account for these apparent changes in dioxin levels in milk including uncertainties in the approach as discussed earlier, reduced emissions, and changes in agricultural practices.

PAHs Using the above procedure, the combined daily intake of the detected PAHs from ingestion of all milk fats was estimated to be 0.6 μg/day. Exposure to PAHs occurs via inhalation of the compounds in tobacco smoke, wood smoke, and contaminated air, and ingestion of the compounds in foodstuffs. The total potential exposure to PAHs for adult males in the United States was estimated to be 3 μg/day (ATSDR, 1995).

Lead Using the above procedure, the daily intake of lead from milk ingestion was estimated to be 0.14 μg/day. The general population is exposed to lead in ambient air, in many foods, in drinking water, in soil, and in dust. Total dietary lead intake ranges from 1.8 to 4.2 μg/day and inhalation rates are typically about 2 μg/day (ATSDR, 1999a).

Cadmium Using the above procedure, the daily intake of cadmium from milk ingestion was estimated to be 0.06 μg/day. Human exposure to cadmium can result from consumption of food, drinking water or incidental ingestion of soil or dust contaminated with cadmium; from inhalation of cadmium-containing particles from ambient air; from inhalation of cigarette smoke. For nonsmokers, ingestion of food is the largest source of cadmium exposures. In the United States, adult intake of cadmium from food has recently been estimated to be about 30 μg/day based on the Total Diet Study, with the largest contribution from grain, cereal products, potatoes, and other vegetables (ATSDR, 1999b).

Disclaimer: This paper reflects the views of the authors and does not necessarily reflect the views of the Environmental Protection Agency and no official endorsement should be inferred. The mention of trade names, or commercial products constitute neither endorsement nor recommendation of use.

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