

Evaluation of Dioxin in U.S. Cow's Milk

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Introduction

Milk fat is likely to be among the highest dietary sources of exposure to persistent, bioaccumulative, and toxic (PBT) contaminants, thus it is important to understand PBT levels in milk. Schaum et al. has previously reported on concentrations of 21 PBTs in the United States (U.S.) milk supply¹. In that study, nationwide samples were collected from dairy plants in 45 different locations, estimated to represent 20% of the U.S. milk supply, 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 detection limits, including polychlorinated biphenyls (PCBs), chlorinated dibenzo-*p*-dioxins and furans (CDD/CDFs), polyaromatic hydrocarbons (PAHs), cadmium and lead.

This study is a follow-up to Schaum et al.¹. Its purpose is twofold: to refine some of the measurements reported earlier by recalculating them on a lipid basis, and to report on limited follow-up analyses conducted after the first study aimed at better understanding the source of a high measurement from the first study.

Materials and Methods

Milk samples for both studies were obtained through EPA's Environmental Radiation Ambient Monitoring System (ERAMS), as described in Schaum et al.¹. All milk samples for the first follow-up portion of this study were collected in January and February of 2001; milk samples for the second follow-up portion of this study were collected in January of 2003. All milk samples were analyzed by EPA's Environmental Chemistry Laboratory at Stennis Space Center, as described previously^{2,3,4}.

All CDD/CDF concentrations in both studies have been 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 CDDs and CDFs⁵. Results were calculated by assigning non-detectable (ND) amounts a value of one-half the detection limit. The TEQ estimates for total CDDs/CDFs assuming non-detects equal to zero were not appreciably different.

Generally, concentrations of lipophilic contaminants such as dioxins are reported on a lipid adjusted basis to normalize across milk samples which can have a wide range of fat content. Freezing and thawing of the whole milk samples collected in the first study resulted in non-

homogenous mixtures. This made measuring lipid content difficult, and it was decided to report all CDD/CDF concentrations in units of picograms per liter of whole milk as collected, approximately equal to parts per quadrillion. Subsequently, the EPA lab developed a new procedure which produced very homogenous samples from thawed milk and allowed accurate measurement of lipid content (see description below). Results from the earlier study have been recalculated based on these lipid measurements, and are presented in the results section.

Milk samples were removed from the freezer and warmed slightly in a water bath. A probe from a Polytron homogenizer was then used to re-mix the milk. The milk was homogenized at a low setting for approximately two minutes before a sub-sample was removed for analysis.

Lipid determinations, done in duplicate, demonstrated that this procedure was effective in producing homogeneous sub-samples.

Results and Discussion

Lipid Adjusted Values: Using the new procedure for warming and re-homogenizing the frozen whole milk samples, lipid concentrations for samples from the earlier study ranged from 1.56-3.07%. Whole milk samples from the first and second follow-up portions of this study had lipid concentrations ranging from 2.48-3.87% and 2.42-3.55%, respectively.

Based on these lipid measurements, the CDD/CDF TEQ concentrations in milk samples from the previous study were recalculated on a lipid basis. These concentrations are shown in Table 1 along with the CDD/CDF TEQ levels which had been calculated earlier on a whole milk basis.

Regional means recalculated on a lipid basis for the summer and winter milk samples were in close agreement at 0.68 and 0.64 pg TEQ/g lipid, respectively, indicating no seasonal difference. The national average CDD/CDF concentration (based on the grand composites from both seasons) was recalculated on a lipid basis as 0.71 pg TEQ/g. The national averages for each congener are shown in Table 2.

Table 1. TEQ_{DF}-WHO₉₈ concentrations of CDD/CDFs in milk (whole milk vs. lipid basis).

Composite location	CDD/CDFs (pg TEQ/L whole milk)		CDD/CDFs (pg TEQ/g lipid)	
	July 2000	Jan 2001	July 2000	Jan 2001
New England	12.85	8.89	0.46	0.35
Mid-Atlantic	11.67	14.21	0.45	0.67
South Central	17.53	19.14	0.76	0.77
North Central	20.48	10.35	0.81	0.66
West Central	17.94	18.59	0.67	0.72
Southwest	13.27	6.21	0.50	0.21
Far South	18.13	35.82	0.69	1.28
Far West	22.50	13.20	1.07	0.46
Regional mean ^a	16.80	15.80	0.68	0.64
Grand composite mean ^b	18.70	9.91 ^c	0.88	0.54
National average ^d	14.30		0.71	

^a 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 (n=45) located in the specified region.

^b The grand composite mean is a weighted 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 to give a national average.

^c Correction from earlier study; previous grand composite mean reported as 8.89 pg/L.

^d The national average value is the mean of the grand composites from both seasons.

Table 2. Average congener concentrations in milk (pg/g lipid).^a

	Concentration (pg/g lipid)	Concentration (pg TEQ/g lipid)
2,3,7,8-TCDD	0.03	0.03
1,2,3,7,8-PeCDD	0.21	0.21
1,2,3,4,7,8-HxCDD	0.21	0.02
1,2,3,6,7,8-HxCDD	1.44	0.14
1,2,3,7,8,9-HxCDD	0.38	0.04
1,2,3,4,6,7,8-HpCDD	3.94	0.04
OCDD	2.31	0.00
2,3,7,8-TCDF	0.02	0.00
1,2,3,7,8-PeCDF	0.10	0.01
2,3,4,7,8-PeCDF	0.23	0.12
1,2,3,4,7,8-HxCDF	0.39	0.04
1,2,3,6,7,8-HxCDF	0.23	0.02
2,3,4,6,7,8-HxCDF	0.22	0.02
1,2,3,7,8,9-HxCDF	0.10	0.01
1,2,3,4,6,7,8-HpCDF	0.71	0.01
1,2,3,4,7,8,9-HpCDF	0.10	0.00
OCDF	0.18	0.00
Total CDDs/CDFs	10.79	0.71

^aAverage of summer and winter grand composite concentrations.

Follow-up Analyses: *2001 Follow-up* – In the previous study, CDD/CDF TEQ levels were appreciably higher in the Far South winter composite than in any other composite, summer or winter (Table 1). In order to determine which ERAMS stations were contributing to the elevation in CDD/CDF levels seen in the 2001 Far South winter composite, the four individual samples which made up this composite were analyzed separately. These individual ERAMS stations were: Montgomery, AL; Tampa, FL; Atlanta, GA; and Jackson, MS. As shown in Table 3, CDD/CDF levels in the Atlanta, GA sample were considerably higher than those in the Montgomery, AL, Tampa, FL or Jackson, MS samples.

Table 3. TEQ concentrations of CDD/CDF's in milk samples from individual stations making up the Far South winter 2001 composite.

Individual Far South Stations	CDD/CDFs (pg TEQ/g lipid)
Montgomery, AL	0.68
Tampa, FL	0.69
Atlanta, GA	4.02
Jackson, MS	0.79

In addition, an Atlanta, GA sample collected approximately one month earlier than the sample used in the Far South winter composite was also analyzed. This sample contained CDD/CDF concentrations of 0.82 pg TEQ/g lipid, comparable to those observed in the remaining three Far South stations for this time period.

2003 Follow-up – To further investigate the high CDD/CDF levels in the Atlanta, GA winter sample described above, a second follow-up analysis was conducted. In this portion of the study, an additional Atlanta, GA milk sample was collected two years later, in January, 2003. CDD/CDF levels in this sample were 0.85 pg TEQ/g lipid, very close to the lower of the two values obtained for Atlanta, GA in 2001.

Discussion: This study converted the data reported by Schaum et al. on CDD/CDF levels in cow's milk to a lipid adjusted basis which should be a stronger basis for comparative analysis¹. The trends reported earlier and how they are affected are discussed below:

§ Schaum et al. observed that the CDD/CDF TEQ levels in milk collected in 2000 were about 50% lower than those reported in a similar study conducted in 1996^{1,4}. After converting these data to a lipid-adjusted basis, the difference in the two studies is much less (0.71 pg TEQ/g lipid in 2000 compared to 0.82 pg/g lipid in 1996).

§ Schaum et al. reported that no large seasonal (winter vs. summer) or regional differences were observed¹. These observations are unchanged when using lipid adjusted data.

Follow-up sampling determined that the elevated CDD/F levels seen in the 2001 Far South composite in the previous study were due to the Atlanta portion of this composite. The congener profile of this elevated sample was similar to the other milk samples analyzed, providing no additional information regarding possible sources. Additional sampling of Atlanta milk showed no elevations in samples collected later in 2001 and again in 2003.

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References

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