

High Body Burdens of 2,2',4,4'-Tetrabromodiphenyl Ether (BDE-47) in California Women

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Following our first report on elevated polybrominated diphenyl ether (PBDE) concentrations in California women, we expanded our investigation to include diverse groups of local women. We analyzed additional adipose and serum samples collected in the late 1990s from San Francisco Bay Area women participating in a breast cancer study and in a reproductive study, respectively. Adipose samples ($n = 32$) were analyzed by low-resolution mass spectrometry in negative-ion chemical ionization mode, whereas serum samples ($n = 50$) were analyzed by dual-column gas chromatography with electron capture detection. The results confirmed our earlier findings. Concentrations of 2,2',4,4'-tetrabromodiphenyl ether (BDE-47) in contemporary California women ranged between 5 and 510 ng/g lipid, with a median (16.5 ng/g lipid) 3–10 times higher than those reported from Europe. In contrast, PBDEs were not measurable in any of 420 archived serum samples collected in the 1960s from San Francisco Bay Area women participating in a study of child development. BDE-47 concentrations did not increase with age or with concentrations of a polychlorinated biphenyl (PCB-153), suggesting other routes of exposure in addition to diet. Rising body burdens of endocrine-disrupting chemicals such as PBDEs may pose a potential public health threat. **Key words:** adipose tissue, BDE-47, body burdens, California, PBDEs, persistent organic pollutants, polybrominated diphenyl ethers, serum, time trends. *Environ Health Perspect* 111:1175–1179 (2003). doi:10.1289/ehp.6220 available via <http://dx.doi.org/> [Online 10 March 2003]

Persistent organic pollutants (POPs) enter the natural environment via a multitude of pathways. Body burdens reflect cumulative exposures to such chemicals and can be used to assess temporal and spatial trends. Body burdens of organochlorine compounds [polychlorinated dibenzo-*p*-dioxins, polychlorinated biphenyls (PCBs), pesticides] are declining in most of the industrialized countries (Liem et al. 1995; Noren and Meironyte 1998, 2000; Smith 1999) as a result of source reduction measures. Polybrominated diphenyl ethers (PBDEs), on the other hand, show increasing trends worldwide (de Wit 2002). Three industrial formulations of PBDEs are used widely as flame retardants [Bromine Science and Environmental Forum (BSEF) 2001]. Deca-BDE (consisting almost completely of BDE-209) is used mainly in thermoplastics and textiles. In 1999, its use in the United States was estimated at 25,000 metric tons, or 44% of its global use (Hale et al. 2002). Octa-BDE (a mixture of hexa- to octa-BDE congeners) is used in acrylonitrile/butadiene/styrene (ABS) plastics. Its use in the United States was estimated at 1,400 metric tons in 1999, corresponding to about 36% of its global use (Hale et al. 2002). Penta-BDE (a mixture of tetra- and penta-BDE congeners) is used mainly in polyurethane foam. The U.S. market used about 8,000 metric tons in 1999, which is approximately 98% of the global production of penta-BDE (Hale et al. 2002). Although the more brominated formulations are used more

extensively worldwide than is penta-BDE, the tetra- and penta-congeners bioaccumulate to a greater degree than do the higher homologues (World Health Organization 1994). In fact, the congener pattern found in biota closely matches the pattern of the penta-BDE formulation (Hale et al. 2002), with 2,2',4,4'-tetrabromodiphenyl ether (BDE-47) as a dominant congener. Production of all PBDEs has increased over the last 20 years, accompanied by their emergence in environmental and biologic samples (de Boer et al. 1998; de Wit 2002). Hepatotoxicity, embryotoxicity, thyroid, and behavioral effects have been reported in animal studies (Darnerud et al. 2001; McDonald 2002). Of particular concern is the ability of PBDEs to disrupt thyroid hormone balance and to cause behavioral and learning deficits in rodents exposed *in utero* or postnatally (Eriksson et al. 2001). Elevated PBDE body burdens in women of childbearing age could therefore be an important public health issue.

Concentrations (mean, 86 ng/g lipid) of Σ PBDEs (sum of BDE-47, BDE-99, BDE-100, BDE-153, and BDE-154) in adipose tissues from a group of 23 California women (She et al. 2002) appear to be 3–10 times higher than concentrations reported from other parts of the world (Darnerud et al. 1998; de Wit 2002; Noren and Meironyte 2000; Ohta et al. 2002; Ryan and Patry 2000; Schroeter-Kermani et al. 2000). In addition, concentrations in archived blubber from San

Francisco Bay harbor seals demonstrate a 100-fold increase over the last decade (She et al. 2002). In this article, we report on additional PBDE measurements in adipose and serum from diverse groups of California women, in an effort to better understand sources and pathways leading to the observed high levels.

Materials and Methods

Subjects. The PBDE analyses were performed on adipose and serum samples collected in the course of three separate epidemiologic studies. All three studies involved women living in the San Francisco Bay Area of California.

Adipose from the late 1990s. Breast adipose samples from a group of 32 women, residents of the San Francisco Bay Area, were analyzed for PBDEs. This group was a random subsample from a case-control study on breast cancer and organochlorine exposures (Petreas et al. 2000), and it included women with malignancies, ductal carcinoma *in situ*, and benign breast disease. Participants of the original case-control study were recruited among women undergoing biopsies or lumpectomies at Stanford University Hospital or Kaiser-Oakland Hospital (both in the general San Francisco Bay Area). Eligibility criteria included age between 25 and 65 years, no prior cancer, and not taking tamoxifen or undergoing chemotherapy. Breast adipose tissue (~1 g) was collected during biopsy or

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