HIGH PBDE CONCENTRATIONS IN CALIFORNIA HUMAN AND WILDLIFE POPULATIONS

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Introduction

Persistent organic pollutants enter the natural environment via a multitude of pathways. Polybrominated diphenyl ethers (PBDEs) currently used as flame retardants show increasing trends in biota, including humans. Concentrations of PBDEs in tissues from California populations appear to be very high[1,2] and concentrations in archived harbour seal blubber demonstrated a 100-fold increase over the last decade[2]. In this paper we report on additional PBDE measurements in human adipose and serum, as well as in edible fish, in an effort to understand sources and pathways leading to high levels.

Materials and Methods

Fish were collected in 2000 from the San Francisco Bay and from Pacific coastal waters. Fish were selected based on human consumption patterns and only edible parts (filets with or without skin) were analysed. Each sample consisted of several individual fish of the same species within a certain size collected from distinct geographic areas. Species analysed included both bottom feeders and surface fish (white croaker, jacksmelt, California halibut, diamond turbot, surf perch, shiner perch, striped bass, etc.).

Adipose tissue was collected from a group of 53 women (aged between 28-68), residents of the San Francisco Bay Area, and participating as controls (cancer-free) in a breast cancer study[1]. Serum was collected from a group of 50 women (aged 19-40) participating in a reproductive outcome study[3]. The latter were born in SE Asia (primarily Laos) and had immigrated to the Bay Area within the previous 2-20 years (mean=14.4 yrs, sd=4.5 yrs). Participants of both human studies were recruited and sampled in the late 1990's. Serum archived in the 1960s from a group of over 400 San Francisco Bay Area women was also used for temporal comparison. These women were pregnant during the late 1960s and the serum samples had been collected and archived during their pregnancies.

Samples were kept frozen at -20 ^{o}C until analysis. Fish and adipose samples were homogenized, spiked with ^{13}C-PBDE #77 and extracted with 1:1 hexane:dichloromethane. The extracts were cleaned up with GPC followed by acid silica gel (fish) or Florisil (adipose), concentrated and analysed by LRMS in ECNI mode (Finnigan 4510), with a DB5ms column (60m, 0.25 mm ID, 0.25 um film thickness). Methane was used as the reagent gas; the ion source pressure was 0.6 Torr and the ion source temperature was 100 ^{o}C. The electron energy was typically 70eV and the electron current was kept at 0.3 mA. Lipid content of the fish and adipose samples was determined gravimetrically in an aliquot of the extract and the PBDE results were reported as ng/g lipid.

Serum was thawed, surrogate standards were added, proteins denatured with acetic acid and samples were extracted with hexane/dichloromethane, cleaned up through a glass column custom-packed with Florisil, eluted with hexane/dichloromethane, concentrated, and recovery standards were

Fig. 1 Concentration of Major Organohalogens in California Women

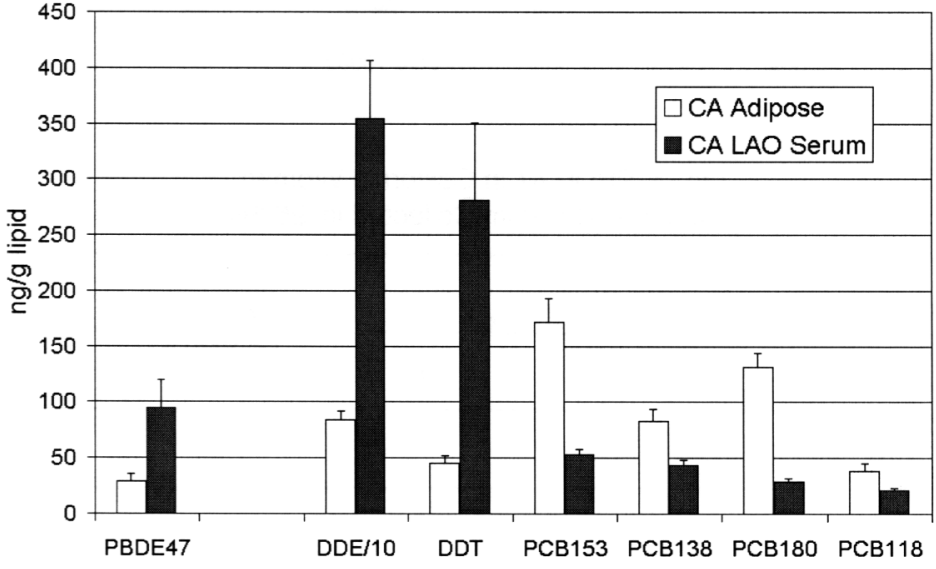
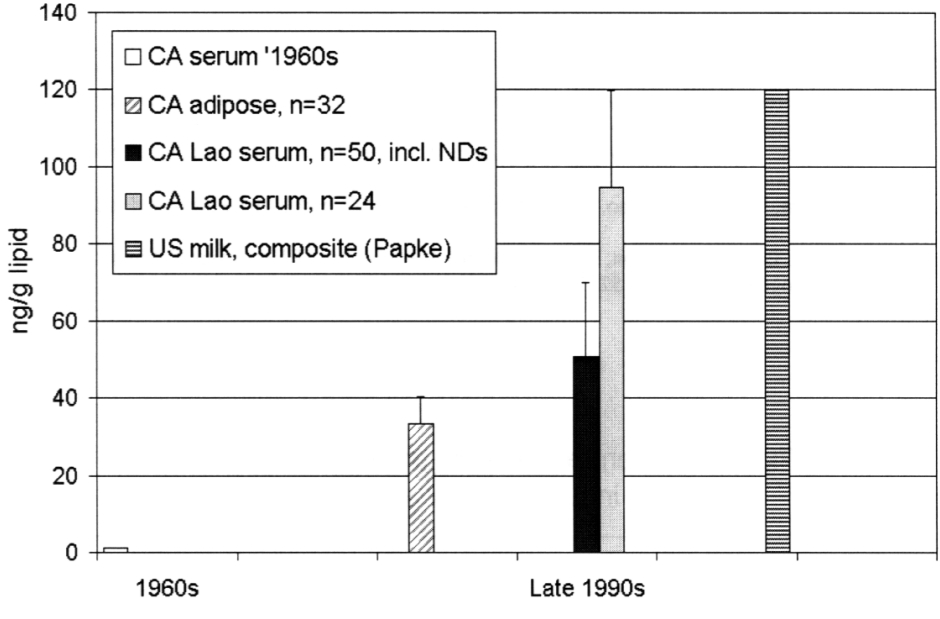
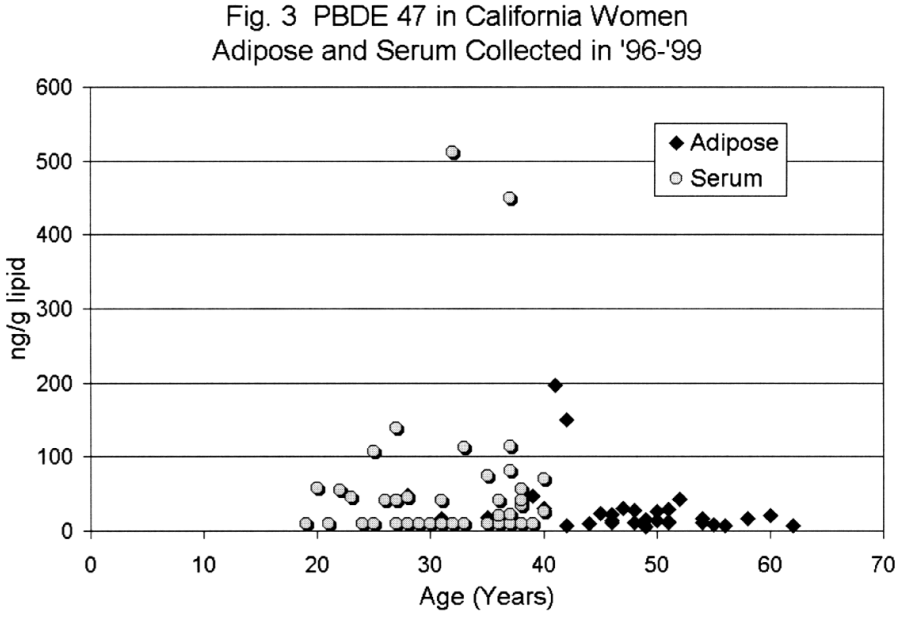


Fig. 2 PBDE 47 in California women





added. Analysis was performed by dual GC-ECD equipped with DB-XLB and Rtx-5ms columns. Elution orders and analyte identification were confirmed by GC/MS. Total lipids were calculated from total cholesterol and triglycerides as described by Phillips[4]. Serum results were reported as ng/g lipid.

Results and Discussion

Serum samples were originally scheduled for PCB and OCP analyses[3], with the PBDE analysis added on as a secondary objective. Only 1 mL of serum was available for analysis, limiting our ability to detect low levels and, therefore, only PBDE-47 could be measured above the blank. The reporting limit for PBDE #47 in serum, however, was much higher (10 ng/g fat) than in adipose (<0.5 ng/g fat). With this limitation, PBDE #47 could be measured in the serum of only 24 of the 50 Laotian immigrants. PBDE #47 was not measurable in any of the archived serum from the 1960s[5]. It is noteworthy that the immigrant women acquired PBDE body burdens equivalent to (or even greater than) those of the contemporary US-born (Fig. 1), while their PCB body burdens remained quite lower and their DDE and DDT levels quite higher[5]. This may reflect differences in exposure patterns and uptake and elimination rates. PBDE-47 was the major congener in all other matrices too, followed by PBDE 99, 100, 153, 154.

Fig. 2 shows concentrations of PBDE-47 in the two groups of contemporary California women (adipose from US-born and serum from Laotian immigrants) in relation to data from a composite sample of human milk from the US[6]. The adipose samples were divided into a “younger” and “older” group based on the median age (48 years). The difference between these two groups is statistically significant[2]. Fig. 3 also shows that younger women have higher and more variable levels of PBDE-47 than older women. Although no demographic information was provided, the composite milk sample was presumably obtained from young (lactating) women, consistent with the observation of higher levels in younger women[2,5].

The California data clearly demonstrate that PBDE concentrations in human and wildlife tissues are very high, compared to other parts of the world. This observation may be consistent with California regulations mandating that all polyurethane foam and textiles used in furnishings pass a flammability test (not necessarily requiring use of PBDEs).

References

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