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Research Article

Brominated flame retardants in US food

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We and others recently began studying brominated flame retardant levels in various matrices in the US including human milk and other food. This paper reviews the food studies. In our studies, ten to thirteen polybrominated diphenyl ether (PBDE) congeners were measured, usually including BDE 209. All US women's milk samples were contaminated with PBDEs from 6 to 419 ng/g, lipid, orders of magnitude higher than levels reported in European studies, and are the highest reported worldwide. We compared our market basket studies of meat, fish and dairy products with other US food studies of meat and fish. US studies showed somewhat higher levels of PBDEs than reported elsewhere. Fish were most highly contaminated (median 616 pg/g), then meat (median190 pg/g) and dairy products (median 32.2 pg/g). However, unlike some European countries where fish predominates, dietary intake of PBDEs in the US is mostly from meat, then fish and then dairy products. Broiling can decrease the amount of PBDEs per serving. We also measured levels of hexabromocyclododecane (HBCD), another brominated flame retardant, in human milk. The levels are lower than PBDEs, 0.16–1.2 ng/g, similar to European levels, unlike PBDEs where US levels are much higher than European levels.

Keywords: Brominated flame retardants / Dietary intake / Food / Hexabromocyclododecane / Polybrominated diphenyl ethers

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1 Introduction

Until recently, persistent organic pollutants (POPs) attracting the most interest included the dioxins, dibenzofurans, polychlorinated biphenyls (PCBs), and dichloro-diphenyltrichloroethane. Relatively recently, it was reported from Sweden [1, 2] that a type of lipophilic brominated flame retardant chemically and toxicologically similar to PCBs, polybrominated diphenyl ethers (PBDEs), was increasing exponentially over time in Swedish nursing mothers' milk whereas dioxins and PCBs were decreasing.

PBDEs are used as additive flame retardants in plastics for electronics, linings under carpets, drapes, computer and television casings and foam used in chairs, mattresses and sofas. There are three PBDE mixtures that have been commonly manufactured: Deca, Penta, and Octa. Octa and Penta are being phased out in Europe and in the US but Deca, which contains predominantly the ten brominated congeners BDE 209, continues to be manufactured [3]. In animal studies PBDE show adverse effects including neurotoxicity, developmental and reproductive toxicity, endocrine disturbances and cancers [3].

After the Swedish milk studies [1, 2], one of us [4] found high levels of PBDEs in human milk collected in Austin, Texas, where the corporate headquarters for a large American computer company is located. Concerned that PBDEs used as flame retardants in computer manufacture might be coming from this computer company, we began a series of studies on PBDE levels in human milk, food and environmental samples from Austin and also Dallas, Texas. Ryan and colleagues at Health Canada, and Päpke and colleagues at Eurofins ERGO analyzed the human milk. Päpke also analyzed food, blood and other samples. We concluded there was no association of the Texas computer facility with PBDEs found in the Austin, Texas milk. However, findings from these analyses led to a series of studies some of which are summarized in this paper which focuses on food PBDE

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Abbreviations: HBCD, hexabromocyclododecane; **PBDEs**, polybrominated diphenyl ethers; **PCBs**, polychlorinated biphenyls; **POPs**, persistent organic pollutants levels and the role of food as one major source of PBDEs in human tissue. We focus especially on our market basket survey of US meat, fish and dairy products because it is the largest and most inclusive of its type.

2 Materials and methods

2.1 Sample collection

Convenience samples of commonly consumed food were purchased from three large national chain supermarkets in Dallas, Texas. Meat, fish and dairy products were selected because previous POPs studies with dioxins and PCBs found these almost exclusively in animal fats with little to none in vegetables and fruits.

2.2 PBDE analysis – Eurofins-ERGO, laboratory

All analyses were performed following the isotope dilution method. Twelve native standards (BDE Nos. 17, 28, 47, 66, 77, 85, 99, 100, 138, 153 154, 183 were obtained from Cambridge Isotope Laboratories, Andover, USA. One native standard, BDE 209, was from Wellington Laboratories, Guelph Canada. Six internal ^{13}C labeled standards, BDE Nos. 28, 47, 99, 153, 154 and 183, were delivered by Wellington, one, BDE 209, was from Cambridge. Solvents were delivered by Merck (n-pentane), Promochem (cyclohexane, hexane, dichloromethane), Baker (diethyl ether), and Mallinckrodt (ethanol, toluene). Silica gel, alumina oxide, sodium sulfate and potassium oxalate were obtained from Merck. For quality control reasons, for each block of samples (6 – 10 samples) a qualtiy control pool and a laboratory blank were analyzed in parallel.

A total of 5 – 200 g tissue was homogenized and mixed with sodium sulfate. Before column extraction a mixture of seven internal BDE standards was added to the sample (100 pg/sample for each congener). For column extraction a mixture of cyclohexane and dichloromethane (1:1 v/v) was applied. The extract was washed with water and dried over sodium sulfate. After solvent evaporation gravimetric lipid determination was performed.

Clean up of all lipid extracts was performed by acid treated and activated silica gel and alumina oxide column. The final extract was reduced in volume by a stream of nitrogen, the final volume was 50 µL containing ^{13}C labeled BDE 139 for recovery standard.

The measurements were performed using high-resolution GC/MS (HP 5890 coupled with VG Autospec) at RP = 10 000 using a DB 5 (30 m, 0.25 mm id, 0.1 µm film) column for gas chromatographic separation. The two most abundant masses were used for measurement (M^+ for Tri- and Tetra-BDE, and $M-2BR^+$ for Penta- to Hepta-BDE). The identification of BDEs was based on retention time and isotope ratio. The quantification was performed by using a five point calibration curve.

Reduction of solvents and control of blank data is important in quality control when analyzing PBDEs at ultra trace levels. Solvents and reagents were tested before the laboratory procedures. All glassware was rinsed by solvents prior to use. Silica gel and sodium sulfate were pre-washed. Rotary evaporators were not used to reduce the risk of contamination. No plastic equipment was used. Quantification was only done if sample data was at least twice the blank value.

To the best of our knowledge, the analytic methods used by other US laboratories were comparable to those used by Eurofins ERGO.

3 Results

Figure 1 shows levels in US human milk in a study of 71 samples from Dallas and Austin, Texas where ten to thirteen PBDE congeners were measured. Usually we measured congeners BDE 17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183 and frequently BDE 209. We found PBDE contamination in all samples, with levels ranging from 6.2 – 419 ng/g, with a mean of 74 ng/g and a median of 34 ng/g [5, 6]. Figure 2 shows that congener 47 predominated in our milk samples in the US and also in samples from several other countries [5], usually followed by congener 99. While these US levels are orders of magnitude higher than reported from Europe and the highest reported worldwide [1, 2, 7 – 9], they are consistent with other recent US human data [10 – 14].

To document changes over time, presumably partly due to changes in food PBDE levels, we analyzed US human blood archived in Dallas, Texas since 1973 and found almost no PBDEs, unlike currently measured (2002 – 2004) blood samples [6]. This is consistent with the time period when PBDEs came into use as flame retardants in the US. In our studies, blood and milk had very similar PBDE levels as well as similar congener profiles. We assume these increases in humans reflect increases primarily in food and possibly also dust and air PBDE levels. Others have

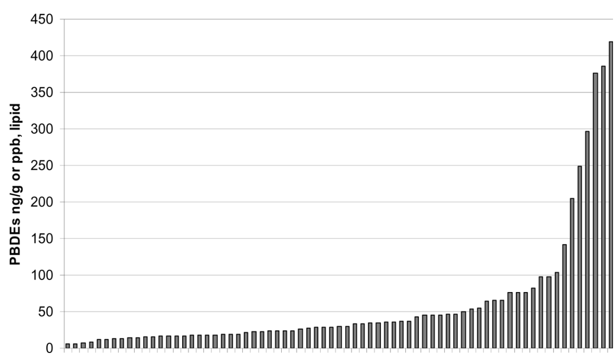

Figure 1. US human milk PBDE levels, 2002 – 2005, [5, 6] and Schecter *et al.*, unpublished data.

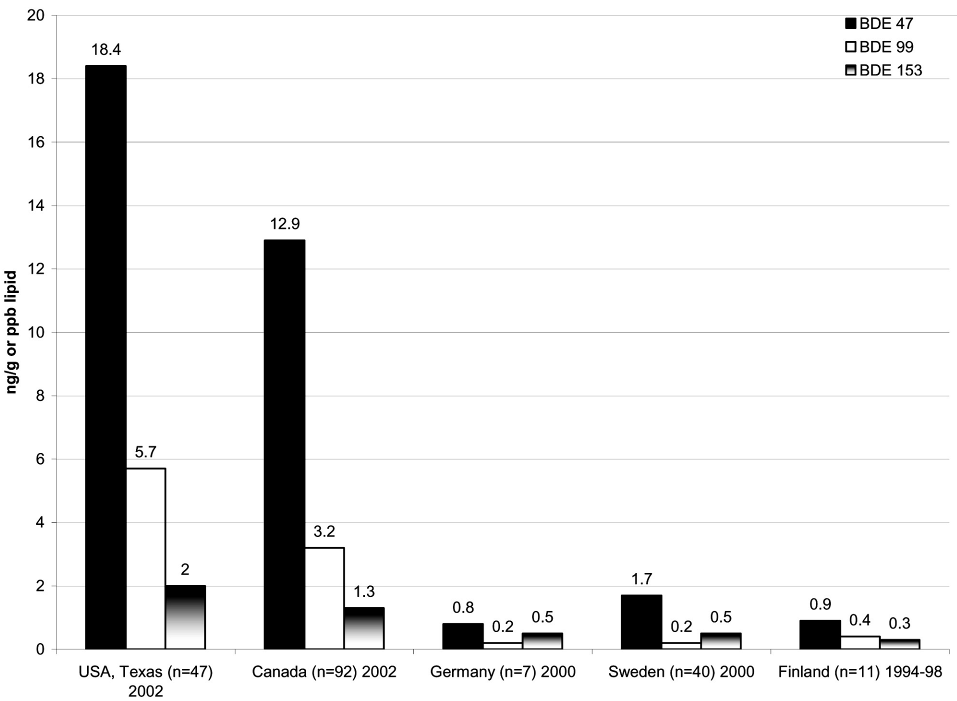

Figure 2. Median levels of PBDE 47, 99, 153 in human milk from different countries (ng/g, lipid) [5].

Table 1. Market basket survey of PBDE Levels in US Food [19], 2002–2005

	Sum of PBDE congeners[a)] (pg/g, ww)[b)]		
	Sum PBDEs Median	Mean	Range
Fish: N=24	616	1120	11–3726
Meat: N=18	190	383	39–1378
Dairy: N=15	32.2	116	7.9–683
Miscellaneous: N=5	–	–	84–2835

a) Congeners measured, BDE 17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209
b) ww, wet weight or whole weight

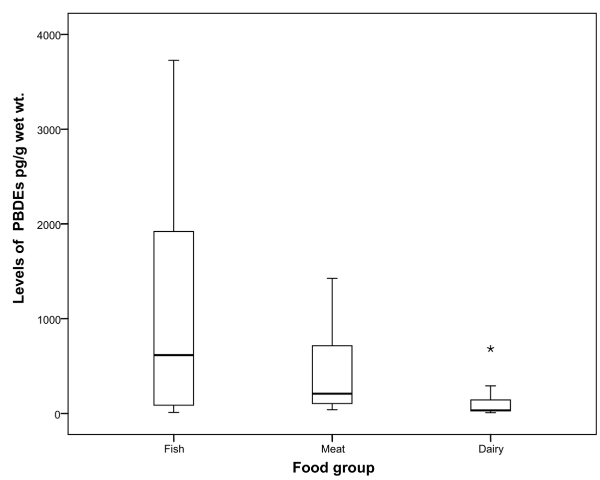

Figure 3. Levels of PBDEs in three food categories in the USA, 2002–2005 [19]; * outlier.

reported similar increases in PBDE levels in Americans [12, 15].

Since dioxins' and similar POPs' route of entry into the general population is almost exclusively from food of animal origin [16, 17], we performed two market basket surveys from Dallas, Texas supermarkets focusing on food of animal origin [18, 19]. Table 1 shows levels of PBDE in the edible portion of 62 food samples from these surveys. Fish had the highest levels followed by meat and then dairy although there was great sample-to-sample variation within food types. Fish (N=24) had a range of 11–3726 pg/g, with a mean 1120 pg/g and a median of 616 pg/g on whole weight basis. Meat (N=18) had levels ranging from 39–1378 pg/g, with a mean of 383 pg/g and a median of 190 pg/g. Dairy products (N=15) had a range of 7.9–683 pg/g, with a mean of 116 pg/g and a median of 32.2 pg/g. Figure 3 is a box plot showing the distribution of total PBDEs in these food categories. These values were higher than reported from some other countries including Japan, Spain and the UK [20, 21, 22] but not orders of magnitude higher as was the case for human milk.

Figure 4 shows that the amount of PBDEs can decrease with cooking [23], here when the food was broiled and the fat dripped away. This effect was similar to previously reported reductions in dioxin levels after broiling food [24].

Table 2. PBDE concentrations (pg/g usually wet weight) in US fish.

Study	Sample Size	Mean	Median	Range
[19][a)]	24[b)]	1120	616	11–3726
[28][c)]	60[d)]	36600	18000	1300–126000[e)]
[27][f)]	20[g)]	1068	489	88–4955
[29][h)]	21[i)]	80100	77800	44600–149000
[30][j)]	18[l)]	132000	26000	1400–1250000[k)]
[31][m)]	16[n)]	429000 Lipid Basis	414000 Lipid Basis	257000–752000 Lipid Basis
[31][m)]	8[o)]	394000 Lipid Basis	275000 Lipid Basis	19000–1144000 Lipid Basis

a) PBDEs 17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209
b) Canned tuna, catfish, catfish fillet, halibut, herring, mahi mahi, salmon, salmon fillet, sardines, shark, shrimp, tilapia, trout, tuna, wild perch
c) PBDEs 47, 99, 100, 153, 154, 183
d) Lake trout, salmon, burbot, whitefish, deeperwate sculpin, bloater chub, alewife, rainbow smelt, benthic amphipods, mysid shrimp bulk zooplankton
e) Outlier 126000, Range without outlier 1300–95000
f) 31 PBDE congeners
g) Wild fish – swordfish, pacific swordfish steak, yellowfin tuna ahi, salmon, coho salmon, wild sockeye fillet, Alaskan halibut fillet, tilapia, scallop, can tuna, petralesole and farm raised fish – salmon, Atlantic salmon fillet, catfish fillet, catfish nugget
h) PBDEs 47, 66, 99, 100, 153, 154
i) Salmon only
j) Tetra BDEs, penta BDEs, hexa BDEs
k) Outlier 1250000, Range without outlier 1400–297000
l) Rainbow trout whole, mountain whitefish fillet, channel catfish fillet, carp fillet, largescale sucker whole, rainbow trout fillet, largescale sucker fillet, mountain whitefish whole, starry flounder whole
m) PBDEs 47, 99, 100, 153, 154
n) Bottom feeders and surface fish including white croaker, California halibut, diamond turbot, surf perch, shiner perch, striped bass from the San Francisco Bay
o) Bottom feeders and surface fish including white croaker, California halibut, diamond turbot, surf perch, shiner perch, striped bass from the Pacific coastal waters

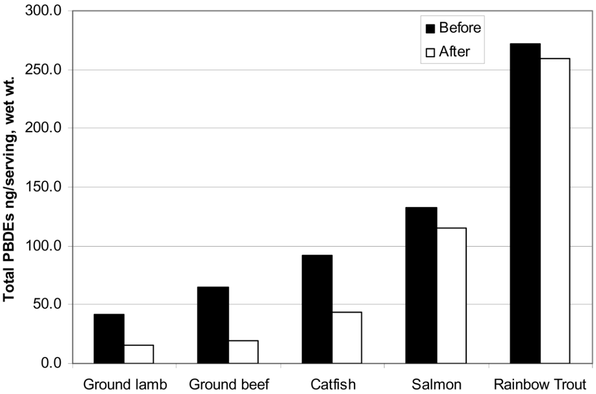

Figure 4. Decrease in PBDE levels after cooking [23].

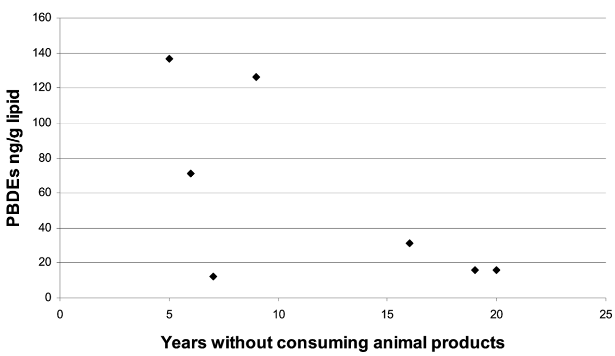

Figure 5. Vegan PBDE blood levels, 2002–2005 [25].

The type of food one eats can determine PBDE body burdens. Vegetables and fruits are low in dioxins, PCBs and presumably other fat soluble POPs [16]. Vegans had lower PBDE levels than the general population, and the longer the time from eating animal products (including fish and dairy), the lower the blood PBDE levels as shown in Fig. 5 [25]. This was similar to our previous findings of low dioxin levels in vegans [26].

Table 2 shows concentration of PBDEs in US fish from various studies. Our fish levels (mean 1120 pg/g, median 616 pg/g, and range 11–3,726 pg/g) [19] are comparable to other food market basket study done in northern California with a mean of 1068 pg/g and a median of 489 pg/g [27] for meat and fish only but lower than other two studies done with Lake Michigan fish [28, 29] with mean of 36600 pg/g and 80100 pg/g respectively and median of 18000 pg/g and 77800 pg/g respectively and range of 1300–95000 (with a outlier of 126 000) and 44600–149000 respectively. Our results are also lower than one study done in Washington State [30] on fish with a mean of 132000 pg/g, a median of 26000 pg/g and a range of 1400–297000 (with a outlier of 1250000). There is one other US study [31] which measured PBDE levels in fish and reported these on a lipid basis. They reported much higher levels of PBDE in fish than we

Table 3. PBDE concentrations (pg/g usually wet weight) in US meat

Study	Sample Size	Mean	Median	Range
[27][a]	15[b]	452	196	85–2516
[34][c]	22[d]	755 Lipid Basis	400 Lipid Basis	100–4620 Lipid Basis
[34]	43[e]	1943 Lipid Basis	890 Lipid Basis	190–16620 Lipid Basis
[33][f]	13[g]	13340	7210	1760–39430
[19][h]	18[i]	383	190	39–1426

a) 31 PBDE congeners
b) Ground beef, ground deer and fowl products including chicken free range, chicken thighs, chicken thighs free range, duck, goose, ground turkey, ground turkey free range, pheasant
c) PBDEs 28, 33, 47, 85, 99, 100, 153, 154, 183
d) Hamburger (11), Bacon (11)
e) Meat trimmings (Chicken Fat, Pork fat, Beef fat)
f) PBDEs 17, 25, 33, 47, 66, 85, 99, 100, 153, 154, 140, 138, 183
g) Chicken fat samples from different places
h) PBDEs 17, 28, 47, 66, 77, 85, 99, 100, 138, 153, 154, 183, 209
i) Bacon, beef ground, beef tenderloin, chicken breast, duck, ground chicken, ground lamb, ground pork, ground turkey, pork, pork sausage, sausage, wieners

Table 4. PBDE and HBCD levels in human milk (ng/g, lipid) from Austin, Texas USA [35], 2002 and 2004

Chemical	Sample Size	Year	Median	Mean	Range
Total PBDEs[a]	24	2002	44	82	7.7–402
Total PBDEs[a]	25	2004	43	56	8.9–246
α-HBCD	20	2002	0.40	0.46	0.16–0.90
α-HBCD	20	2004	0.40	0.49	0.16–1.2

a) PBDEs, BDE 28, 47, 99, 100, 153, 154, 183

found in our study [19] even when our data are reported on a lipid basis. Of these US studies, ours was the only one which measured BDE 209, which is still in production and in use, and therefore total PBDE levels are not entirely comparable between studies.

Concentrations of PBDEs are have been shown to be frequently higher in farm raised salmon than wild salmon, and farm raised salmon from Europe have higher levels of PBDEs than farm raised salmon raised in US [32]. Thus, concentration of PBDEs in fish varies by location, the type of fish as well as how the fish are raised.

Table 3 shows comparative studies of concentration of PBDEs in US meat. Our whole weight meat levels (mean 383 pg/g, median 190 pg/g, and range 39–1426 pg/g) [19] are comparable to one study done in northern California with a mean of 452 pg/g, a median of 196 pg/g and a range of 85–2516 pg/g [27] but lower than another US study where PBDEs concentration in chicken fat is measured with a mean of 13340 pg/g, a median of 7210 pg/g and a range of 1760–39430 pg/g [33]. We note one other US study which measured concentration of PBDEs in hamburger, bacon, chicken fat, pork fat and beef fat [34] and reported these on lipid basis where the results are similar to what we found in our study [19] when our data is reported on a lipid basis.

Although PBDEs in milk from the US general population were far higher than in Europe, levels of hexabromocyclododecane (HBCD), another brominated flame retardant, in US milk were similar to the lower European levels with a median of 0.5 ng/g and a range of 0.16–1.2 ng/g, where only α-HBCD was detected, as shown in Table 4. In the commercial HBCD, the γ-HBCD predominates [35]. There was essentially no change in levels of PBDEs in our milk samples from Austin, Texas, from 2002 to 2004 and there was also no significant change found in HBCD concentration [35]. No other data on HBCD in US food is available, to the best of our knowledge.

4 Discussion

In this series of ongoing studies, we documented very high PBDE levels, the highest worldwide, in US human milk, the first food consumed, as well as blood, which had similar levels. PBDE levels in US blood increased from almost not detectable in 1973 to very high levels in 2003. We presume the same was the case for human milk. All persons studied were contaminated with PBDEs, with levels ranging from approximately 6–419 ng/g, lipid, in blood or milk from adults. We also found all human fetal tissues measured contaminated with PBDEs but at lower levels than in adducts indicating transfer from mother to fetus before birth through blood, which serves as nourishment to the fetus [36]. The exposure of PBDEs during organogenesis as well as during nursing is of concern.

US Market basket surveys showed contamination highest in fish, then meat and least in dairy products, but the average American has most dietary PBDE intake from meat, then fish and dairy. Cooking can reduce the amount of fat and of PBDEs per portion of meat or fish. Vegans appear to have lower levels of PBDEs than the general US population and this appears to be related to the length of time they were vegans and especially, in our study, stopped eating meat.

Although health data are available only from rodent studies at this time the structural and toxicological similarity of PBDEs to PCBs along with these elevated levels in humans is worrisome. The rapid increase in human levels of these persistent, toxic and bioaccumulative chemicals is cause for concern, especially when seen in fetal tissue and when calculating very high intake from nursing (320 ng/kg/day) as well as the relatively high intake in adults (1.2 ng/kg/day) [37].

At present decaBDE alone remains in use in western countries. DecaBDE, consisting almost exclusively of BDE 209, with ten bromine atoms, has been shown to debrominate to less brominated BDEs in carp although the significance of this in humans is unknown [38]. As is the case with POPs such as dioxins and PCBs, depot sources of PBDEs are expected to remain in the environment for decades or longer.

Further research is needed to establish levels of exposure from food and other sources of the general population as well as workers such as fishermen and electronic recycling workers, in developed and developing countries and to determine levels of concern with respect to human health. Future study should help to provide representative levels in food and nursing mothers' milk from different locations, from different ethnic groups and from persons with differing dietary consumption habits. "Safe" levels of various brominated flame retardants in food have not yet been determined.

Whatever the degree of PBDE toxicity to humans, it seems reasonable for governmental agencies to work with industry and environmental groups to lower the amount of these chemicals in the environment. Dioxins, dibenzofurans and PCB levels in the environment, in food and in humans have declined considerably since government regulations were written and put into effect. Hopefully, the same will occur with the brominated flame retardants.

We wish to thank all mothers who donated milk as well as the Mothers' Milk Bank at Austin where some specimens were collected, Dr. Jake Ryan of Health Canada and Dr. Robin Rosen and Barbara Moses, RN, both of UT Southwestern Medical Center, the CS Fund, and others without whose generous support and hard work these studies could not have been performed.

The authors have declared no conflict of interest.

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