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INFLOME

Human Exposure to Organic Flame Retardants

Stuart Harrad

WHY DO WE WANT TO MEASURE EXPOSURE & WHAT DO WE MEAN BY IT?

- **Why do we want to measure it?**
- **Essentially as an input into models predicting potential adverse effects that may arise from exposure to chemicals (dose-response relationship)**
- **How would you define what is meant by exposure to chemicals?**

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WHAT WE MEAN BY EXPOSURE

- **Consensus is that "exposure" means *contact* with chemical**
- **Visualise the body as possessing an outer boundary separating the inside of the body from the external environment**
- **Exposure considered as chemical *contact* with that outer boundary and a possible working definition is "the product of chemical concentration in external environment and extent of contact with external environment " - for inhalation, multiple of chemical mass per unit air volume (chemical concentration) in a given location, and volume of air breathed whilst in that location (this in turn is a multiple of the daily air inhalation rate (e.g. m^3/d), & the number of hours spent in that location).**
- **For diet, exposure is a multiple of chemical mass per unit food mass (concentration), and the amount of that food ingested per day.**

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CALCULATING EXTERNAL EXPOSURE

- **Basic principle of calculating exposure is no different to calculating how much alcohol you consume. For example, a 150 mL glass of red wine contains 13% alcohol by volume (= 0.13 mL mL^{-1}). Hence, drinking 1 glass exposes you to 0.13 x 150 mL = 19.5 mL alcohol.**
- **If the concentration of BDE-47 in this room right now is 700 pg m^{-3}, during the next hour, how much BDE-47 will you be exposed to given that a typical adult inhales 20 m^3 air per day?**
- **If your lunch today contains 100 ng kg^{-1} of HBCD and you eat 100 g of this; how much HBCD will you be exposed to from eating your lunch?**

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DIFFERENT APPROACHES TO EXPOSURE ASSESSMENT (I)

- **2 basic approaches to exposure monitoring**
- ***External exposure* monitoring involves measurement or modelling of concentrations of chemical in external media combined with knowledge of transfer to within the body (e.g. ingestion/inhalation rates) (*environmental monitoring*)**
- **Monitoring of *internal exposure* involves measurement of chemical (or metabolite(s)) in internal tissue (or excretion/secretion) (*biomonitoring*). Knowing how much is in the body, means we can estimate how much must enter it daily to support that amount**

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DIFFERENT APPROACHES TO EXPOSURE ASSESSMENT (II)

- What are the advantages and disadvantages of:
- External, and…
- Internal exposure monitoring?

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DIFFERENT APPROACHES TO EXPOSURE ASSESSMENT (III)

- Advantage of external exposure monitoring is that it permits evaluation of relative significance of different exposure pathways - useful in that if exposure via inhalation is negligible, no need to monitor it
- Conversely, internal exposure methods allow for processes affecting transfer from external environment and, by giving a single measure of aggregate or overall exposure, can reduce monitoring effort
- Ethical and participant recruitment/compliance issues
- The two approaches are complementary

RELATING EXTERNAL AND INTERNAL EXPOSURE

- **External and internal exposure related at *steady state* by $E = I/\tau$**
- **i.e. intake into a system equals the amount of the substance in the system divided by the lifetime of the substance from that system**
- **This is how internal exposure measurements of body burden (I), may be converted into estimates of external exposure (E), provided we know what τ is**
- **This is the basis of the "simple one-compartment PBPK models" appearing currently in the literature**
- **To illustrate, if your body fat contains 10 μg/kg of PBDE, you have 15 kg body fat, the residence time of PBDE in humans is put at 2 years, and your body is assumed to be at steady state with respect to PBDEs; how much PBDE are you exposed to per day?**

Abdallah & Harrad, Environ Int (2011) 37, 443-448
Lorber, J Exp Sci Environ Epi (2008) 18, 2-19

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METHODS OF MEASURING DIETARY EXPOSURE

- **Duplicate diet monitoring (*direct*) - excellent reflection of individual (but not population) exposure over the sampling period, but expensive and volunteer compliance hard to maintain for extended periods. Also, doesn't identify source of elevated exposures**
- **Combination of measurements of contamination in representative sub-samples of different food types and data on food consumption patterns by different population groups (*indirect*) - addresses above problems, but is not necessarily an accurate reflection of individual dietary exposure, or even of population-level exposure. Are food type sub-samples really representative?**

Harrad et al Journal of Environmental Monitoring, 5, 224–228, (2003).

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MEASURING INHALATION EXPOSURE

- For an agoraphobic, relatively simple. Measure pollutant level in residence and multiply by subject's inhalation rate
- For others, more difficult. Spend varying amounts of time in a variety of microenvironments, all of which may exhibit different levels of pollution.
- Also inhalation rate will vary according to activity

VARIATIONS IN INDOOR AIR QUALITY WITHIN A SINGLE BUILDING

Room #	Concentration of PBDE #47 (ng m^{-3})
207	0.62
229	1.3
109	7.0
240	4.6

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METHODS OF MEASURING INHALATION EXPOSURE

- 3 main methods, that can be categorised as either direct or indirect methods of measuring an individual's inhalation exposure:
- Single point monitors - most widely used, but questionable how well they reflect individual exposure
- Personal exposure monitors - ideal for providing measurements of personal exposure, but don't yield information about levels in individual microenvironments
- Combination of point monitors in individual microenvironments and personal "activity diaries"- solves above problem, but not necessarily an accurate reflection of individual personal exposure
- Problem with both 2nd and 3rd methods is cost and difficulty in maintaining volunteer compliance over extended periods
- Of the 3 methods, which are direct and which are indirect?

MEASURING EXPOSURE VIA DUST INGESTION

- In effect, a combination of “point monitors” in individual microenvironments and personal “activity diaries”
- This provides measurements of BFR concentrations in dust
- This then multiplied by the “contact rate”
- For dust ingestion, this is a mass per day (akin to dietary ingestion)
- Dust contamination can be highly spatially heterogeneous, so where, when and how dust is sampled is crucial
- Concentrations can vary between microenvironments (MEs), within rooms within same building (like air) and also within same room
- Unclear as to whether detailed sampling in different MEs provides most “biologically relevant” measure of exposure, or vacuum cleaner bag contents

Harrad et al EST (2010) 44, 4198-4202

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ISSUES RELATED TO WITHIN-ROOM SPATIAL & TEMPORAL VARIABILITY

- How does within-room spatial and temporal variability:
  - Influence human exposure via dust ingestion (biological relevance of single "grab" dust samples)?
  - Provide insights into sources of contamination?

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SOME EXPERIMENTAL DATA

- **Spatial variability studied by:**
  - **Sampling five separate (non-overlapping) 1 m^2 areas in same room at same time (5 rooms - 2 for BDE-209)**
- **Temporal variability examined by:**
  - **Sampling same 1 m^2 area in the same room every month for 9-10 months (3 rooms)**
- **Assessed against:**
  - **Analytical variability (replicate analyses of SRMs 2584/2585).**
  - **Combined variability due to sample processing (homogenisation & sieving) & analysis (replicate analyses of a single sample derived from the same 1 m^2 area). RSDs for both <10%**

Harrad et al, Environ Int 34, 1170-1175 (2008)

Harrad et al, Environ Int 35, 573-579 (2009)

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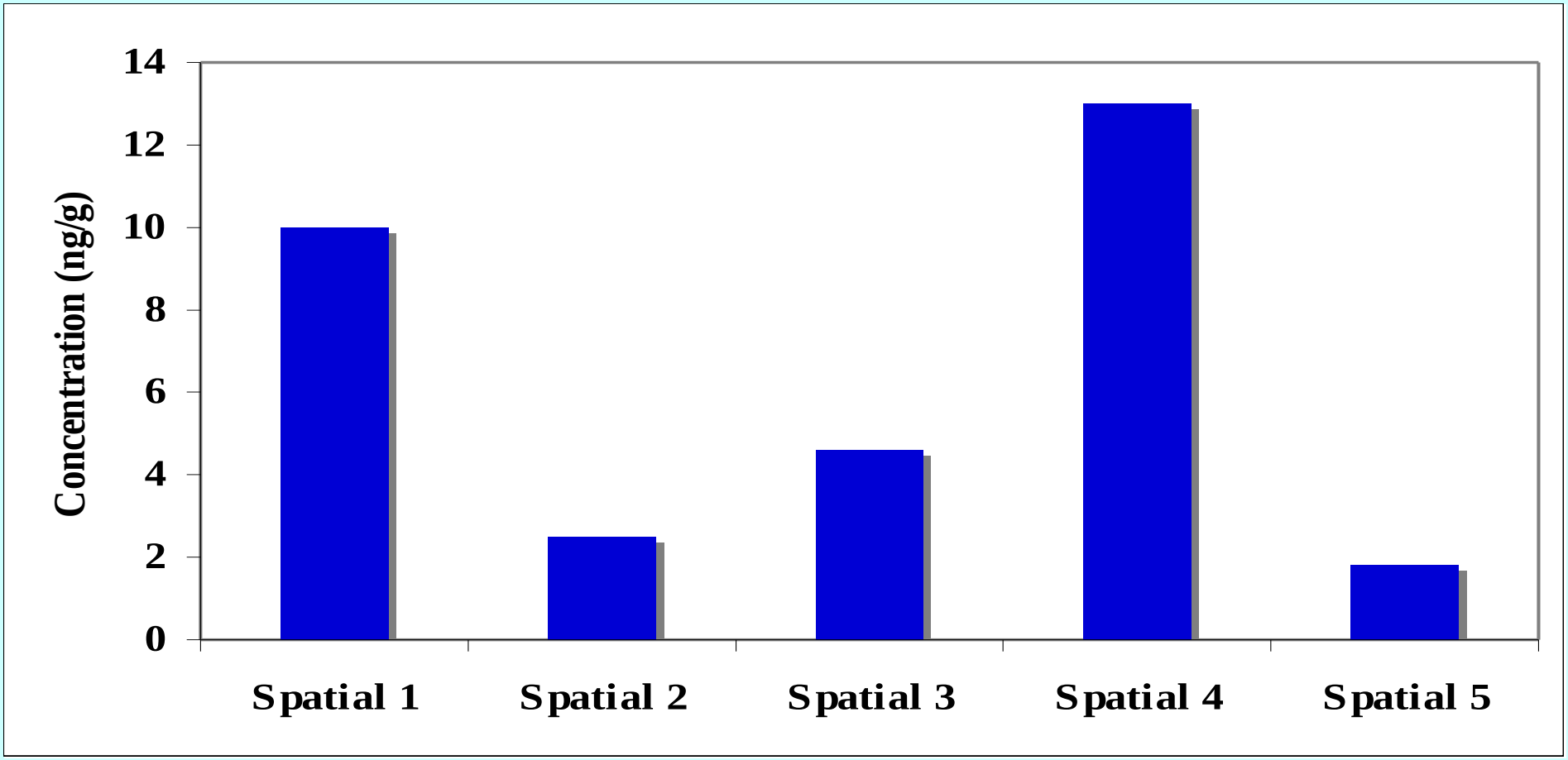
SPATIAL VARIABILITY IN BDE-99



RSD= 77%

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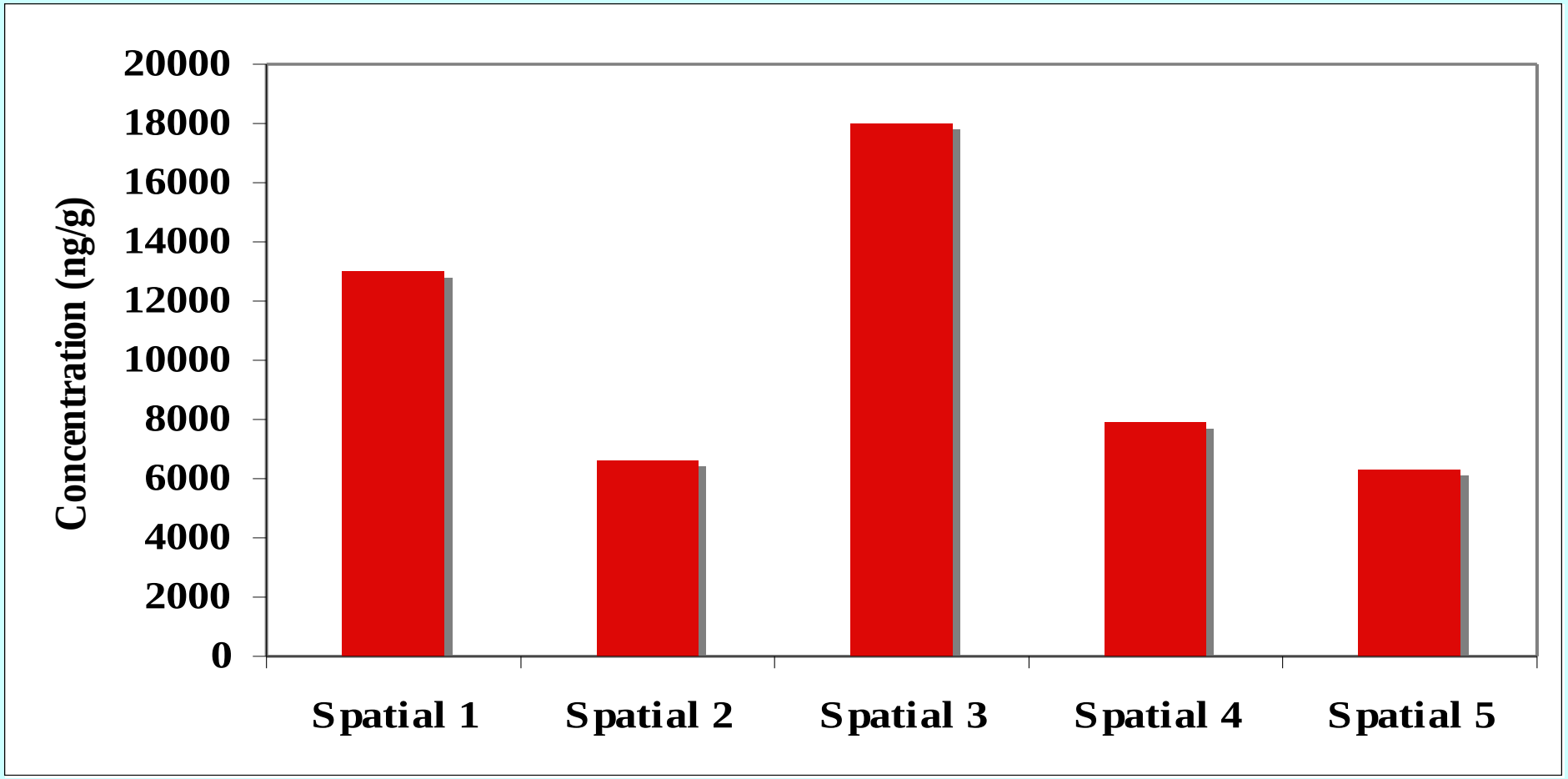
SPATIAL VARIABILITY IN BDE-209



RSD= 49%

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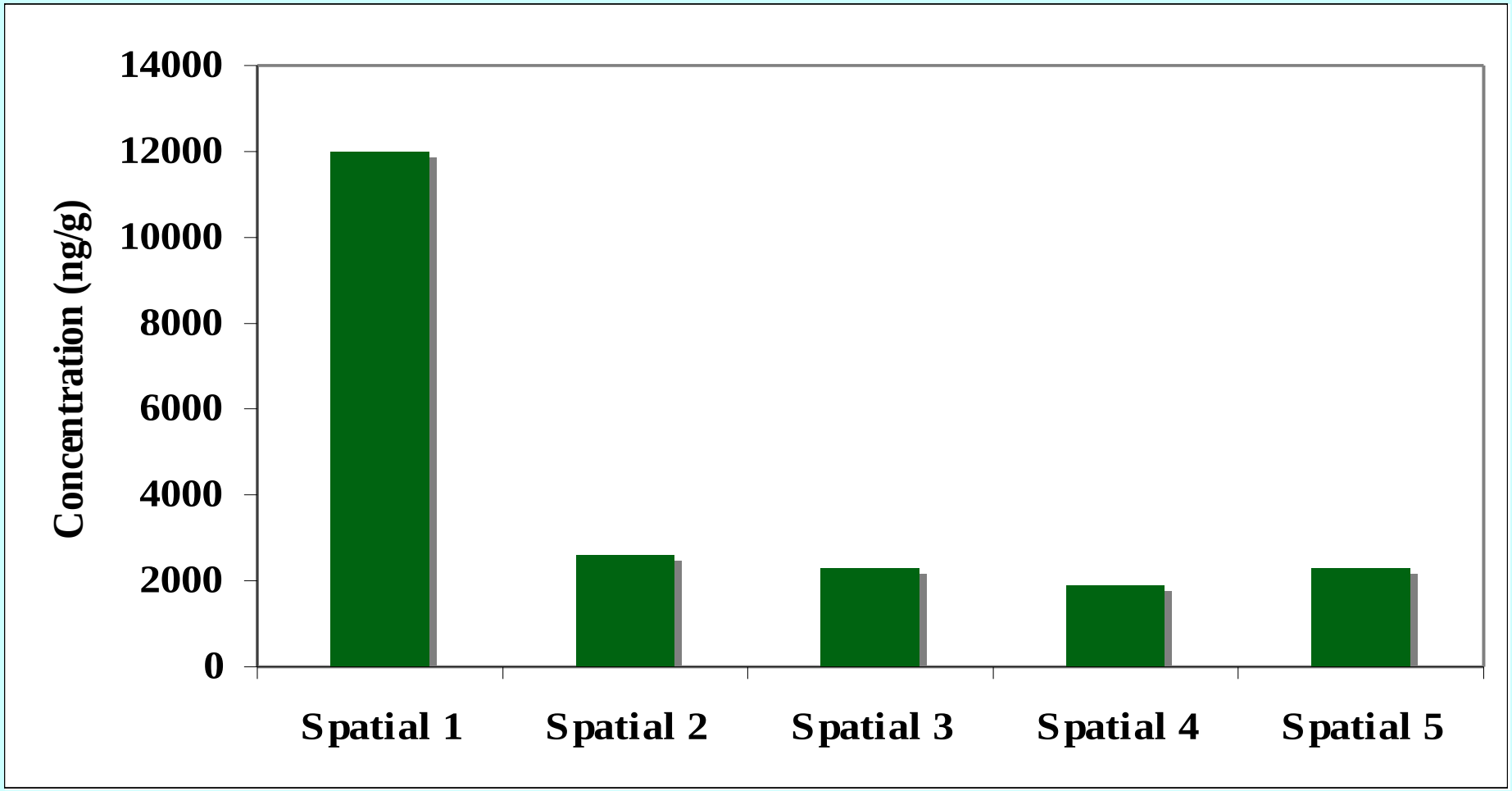
SPATIAL VARIABILITY IN ΣHBCDs



RSD= 100%

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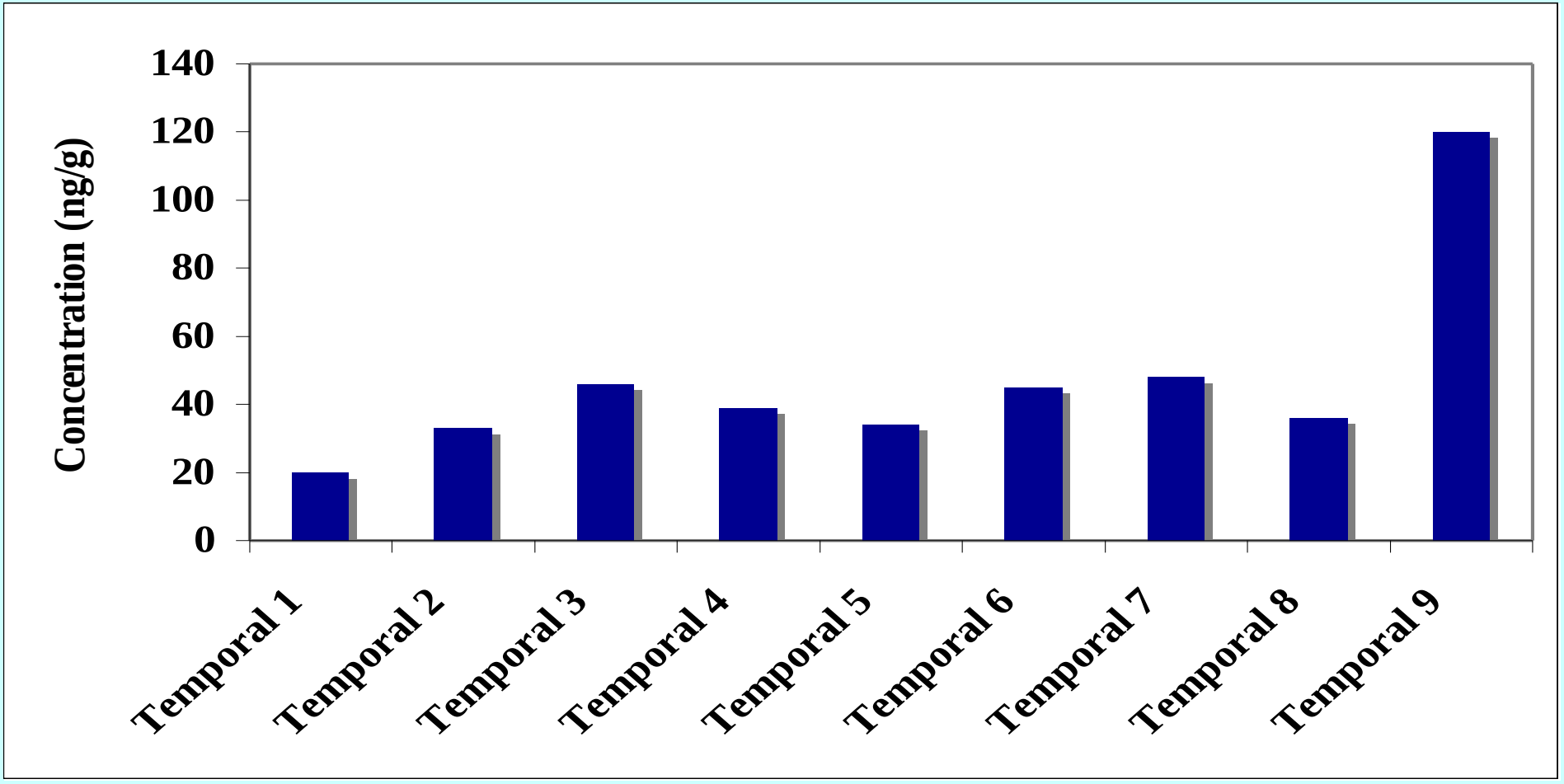
TEMPORAL VARIATION IN BDE-99



RSD= 57%

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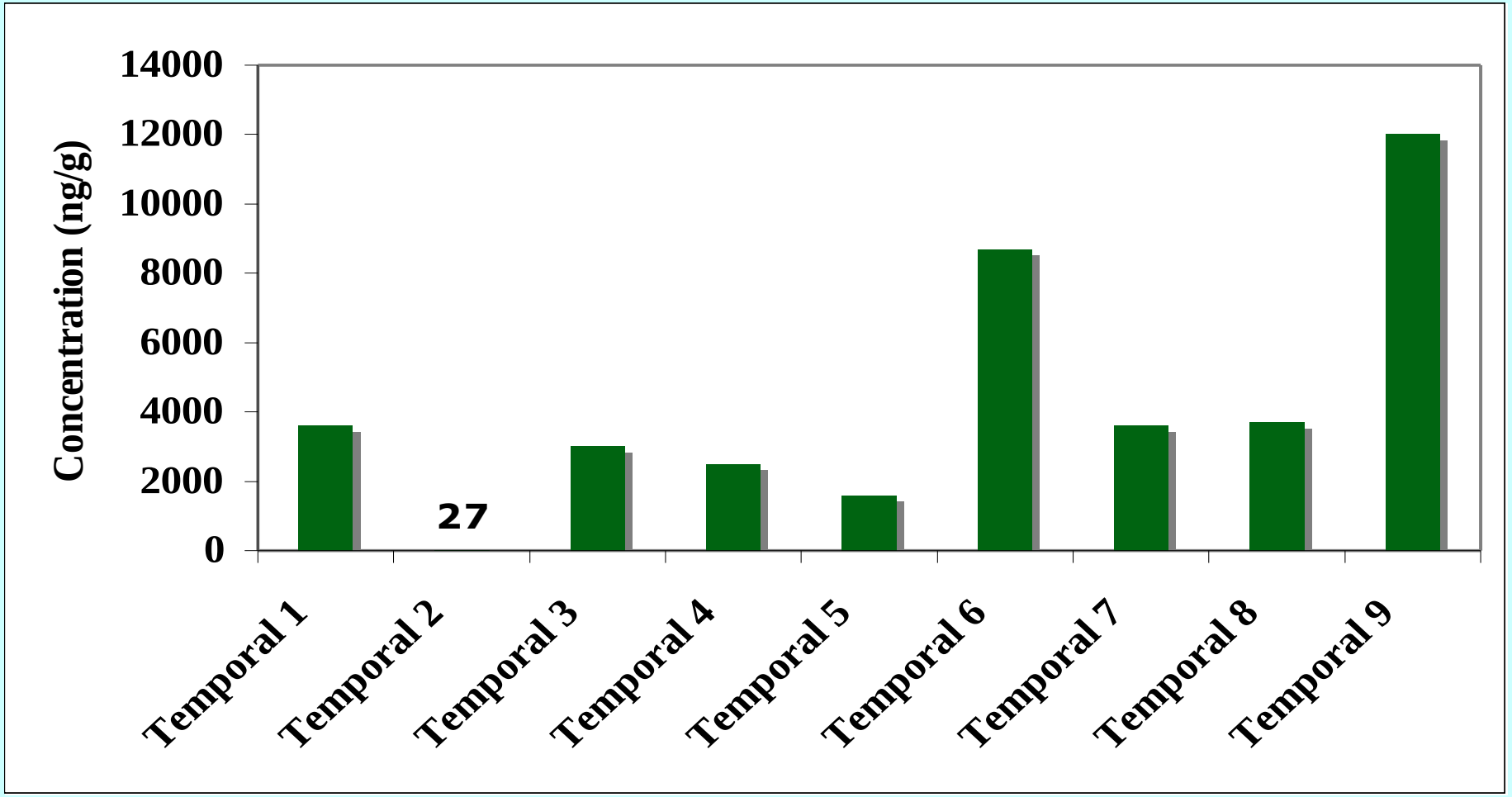
TEMPORAL VARIATION IN BDE-209



RSD= 82%

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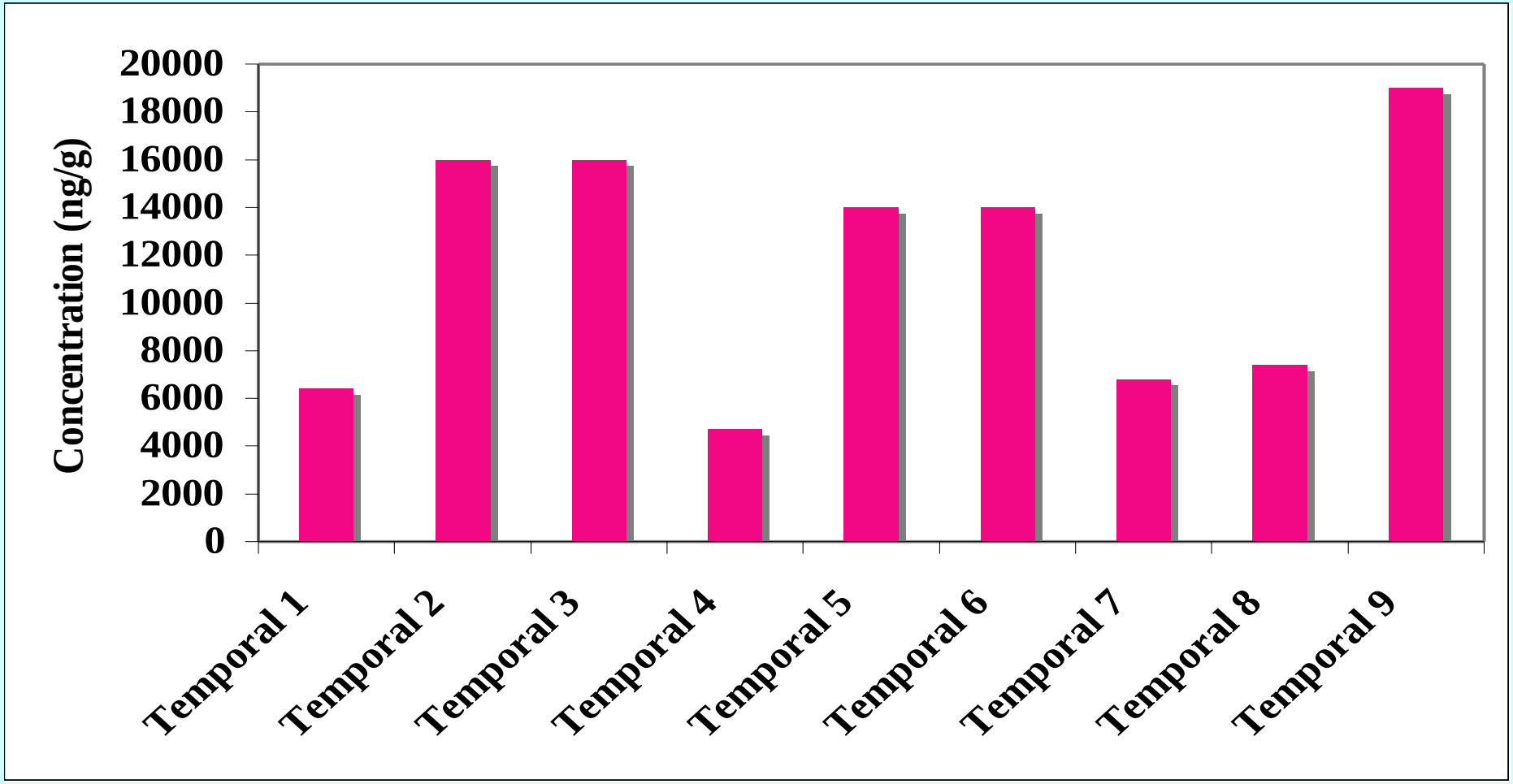
TEMPORAL VARIATION IN ΣHBCDs



RSD= 42%

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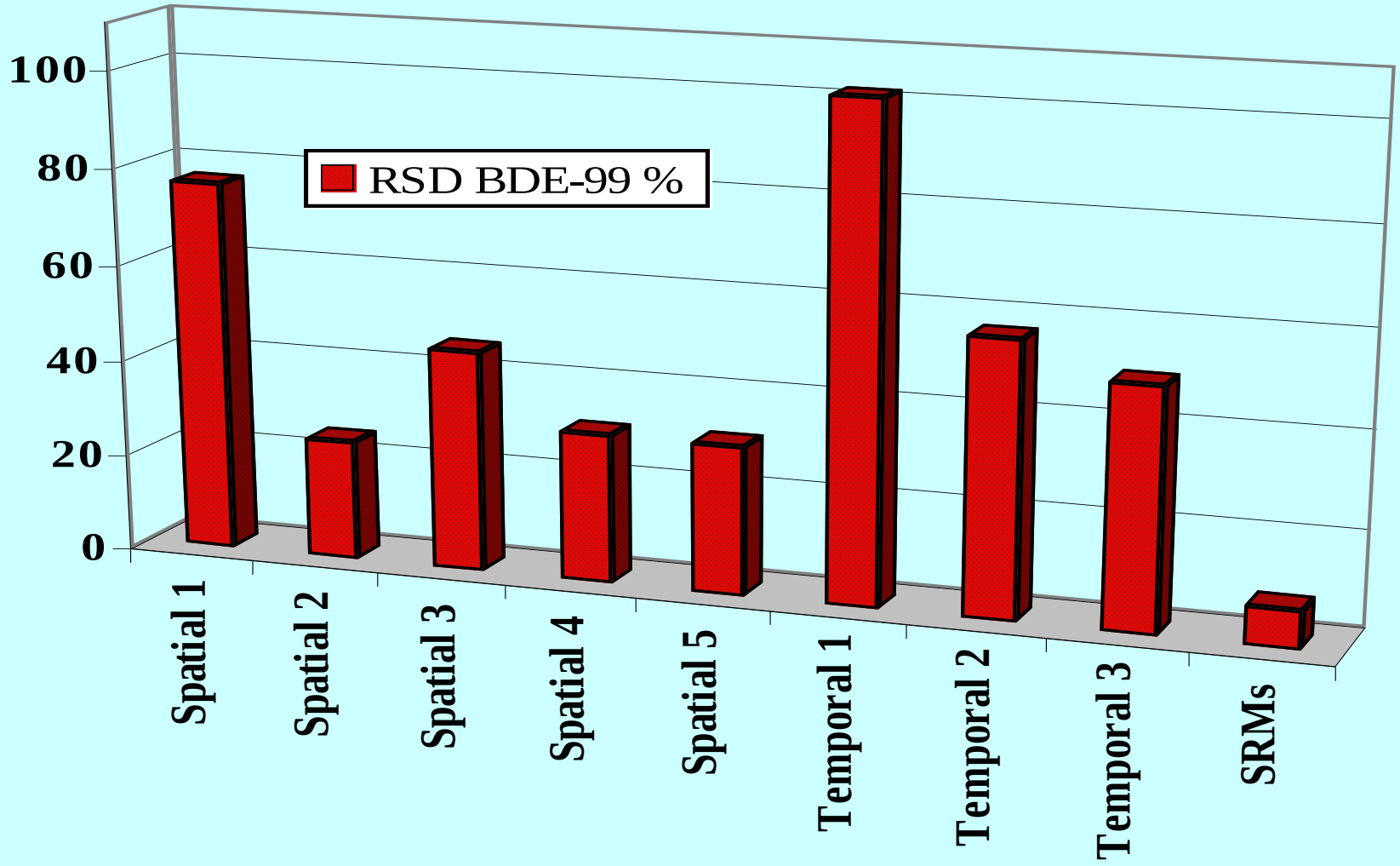
SUMMARY FOR BDE-99



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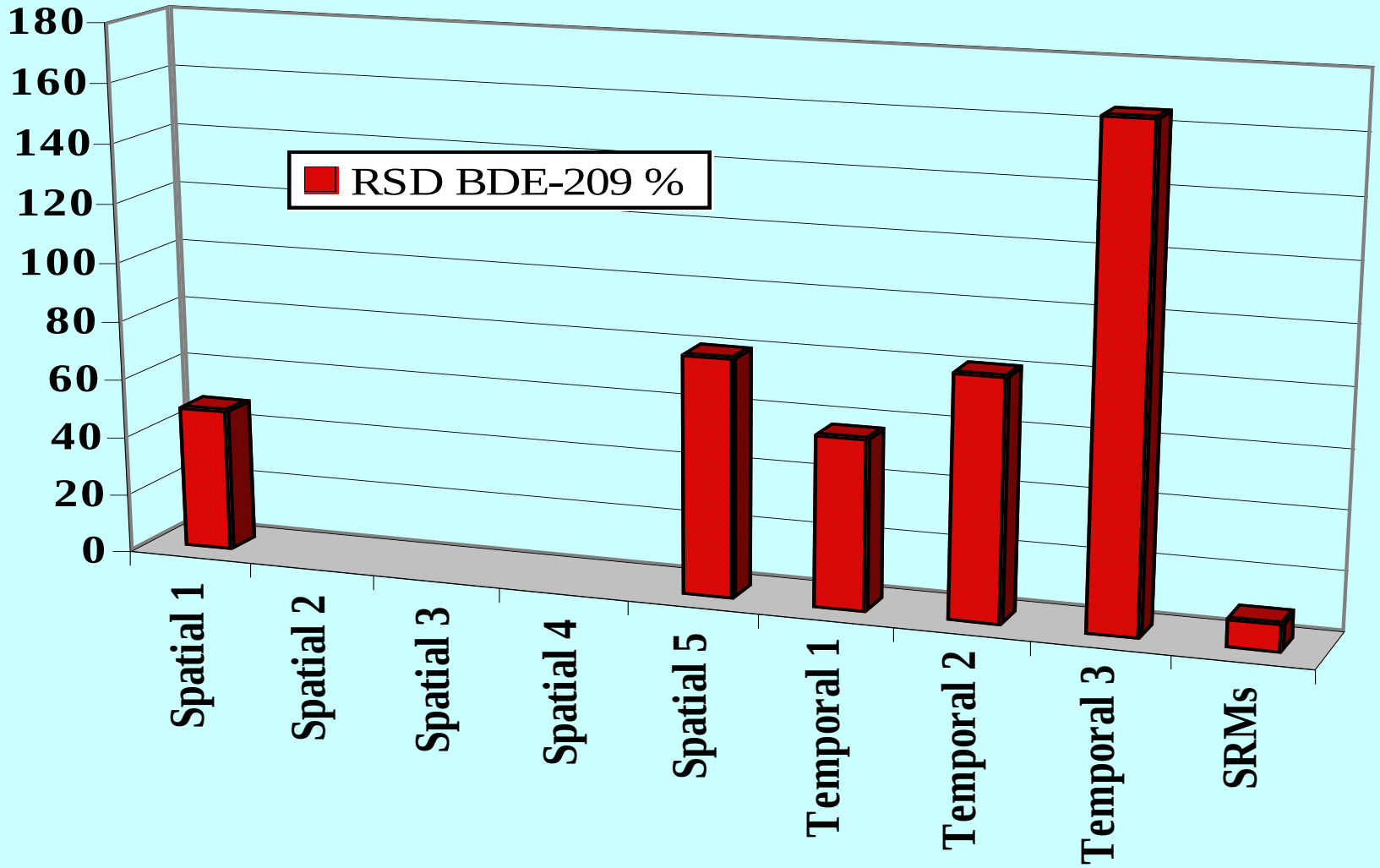
SUMMARY FOR BDE-209



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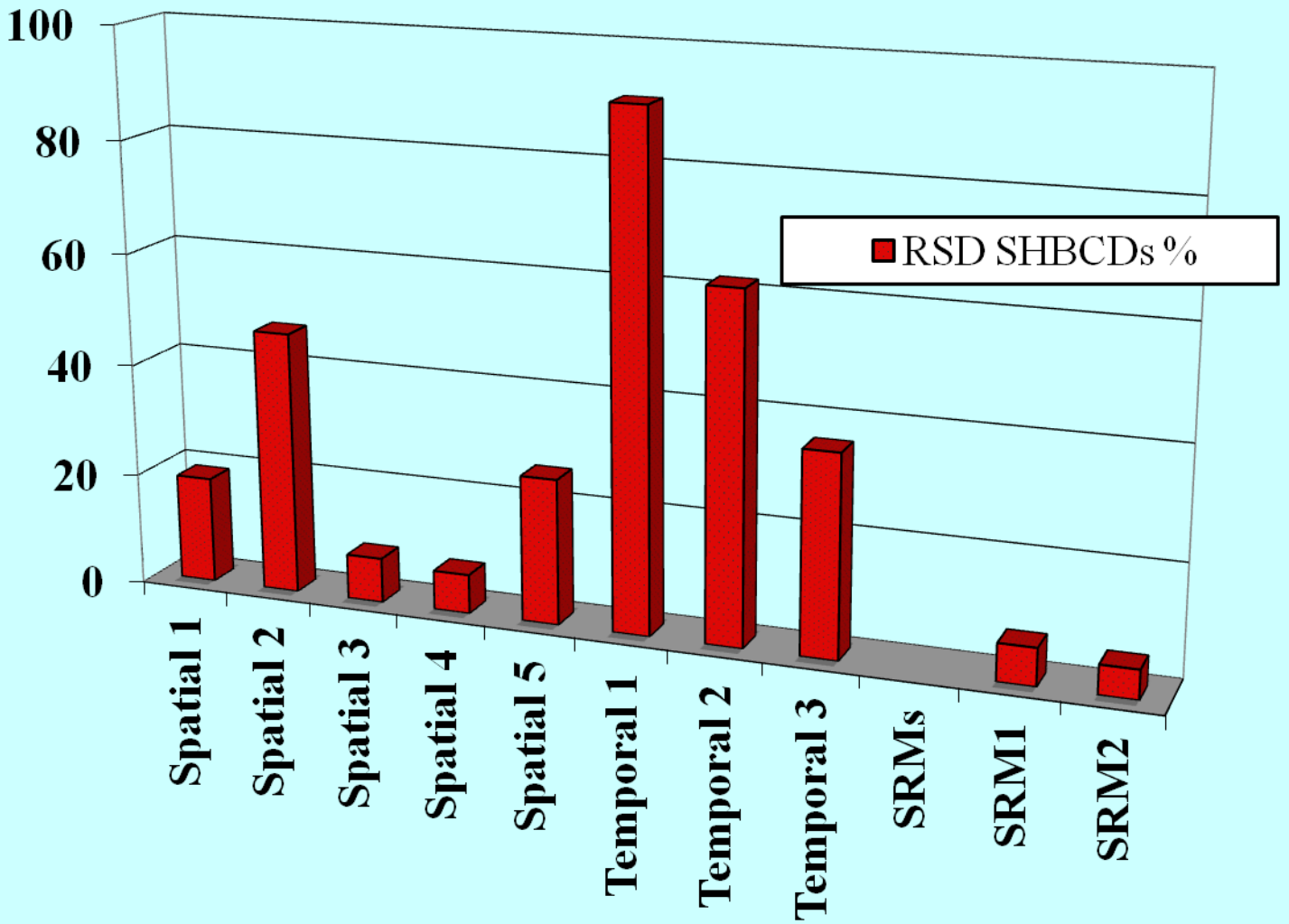
SUMMARY FOR ΣHBCDs



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EXPOSURE IMPLICATIONS OF SPATIAL VARIABILITY

- Variations that exceed the variability in sample processing and analysis (i.e. in most rooms) may suggest best approach is to sample entire room surface. However…
- May complicate the procurement of biologically-relevant samples where for example concentrations in more-frequented areas of room are substantially different to those elsewhere in room

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EXPOSURE IMPLICATIONS OF TEMPORAL VARIABILITY

- In many cases, indications are that such variability is not so great as to compromise seriously the biological-relevance of a single "grab" sample at one point in time. However…
- Highest and lowest concentrations of BDE-209 in one room over a 9 month period differ 400-fold
- Contamination within-room influenced by presence/absence and exact location of point source(s)
- For contaminants where human half-life is short (e.g. BDE-209), may be wise to integrate more than one dust sample over a period of time to reflect better human exposure
- Spatial & temporal variations may confound relationships between dust and human contamination

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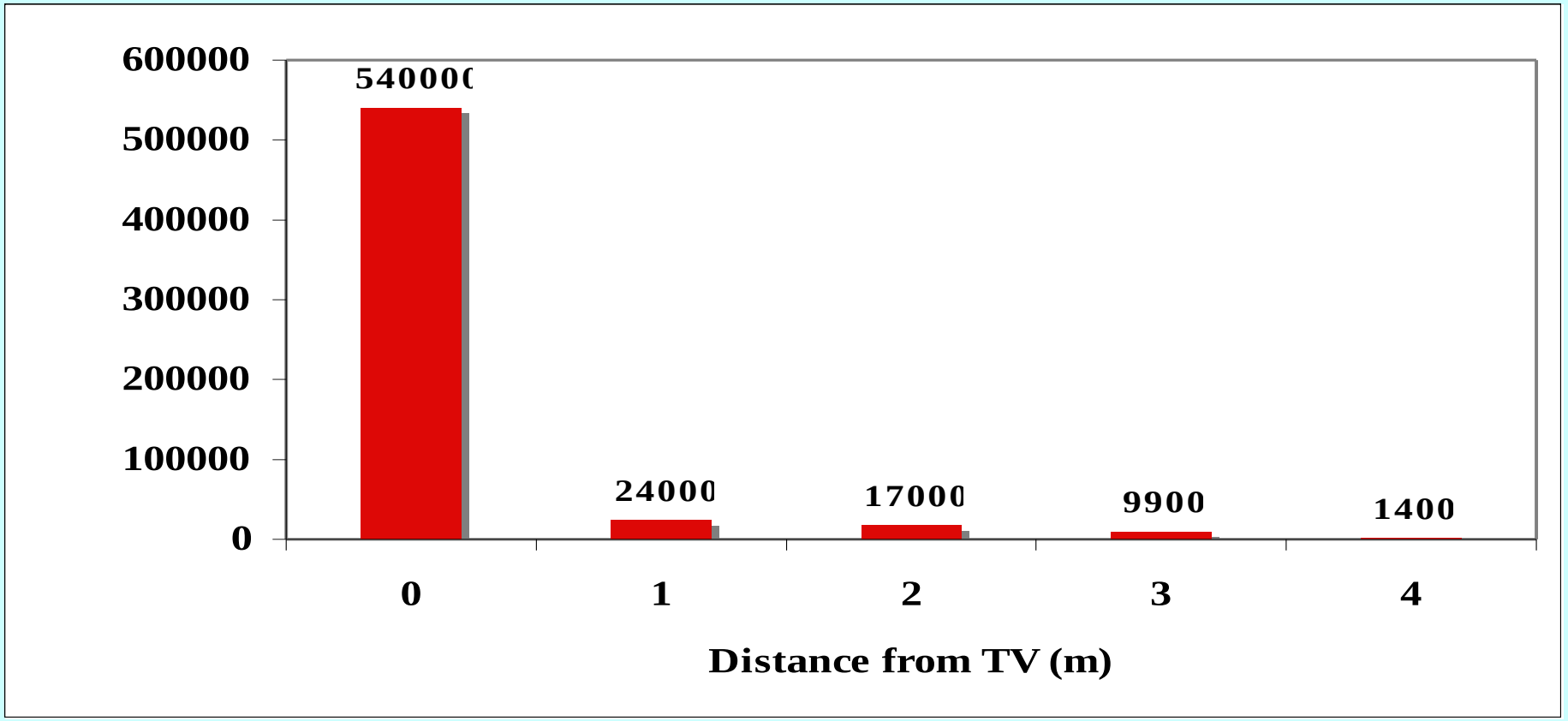
ATTENUATION OF ΣHBCDs IN DUST WITH DISTANCE FROM A TV



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WHAT FACTORS INFLUENCE HUMAN EXPOSURE TO FRs?

- Physicochemical properties
- Use pattern & lifestyle
- Age

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PHYSICOCHEMICAL PROPERTIES RELEVANT TO HUMAN EXPOSURE

- Essentially…
- K_{ow}
- Vapour pressure
- Water solubility, and
- Persistence/resistance to metabolism

OCTANOL-WATER PARTITION COEFFICIENT (K_{OW})

- As it's frequently very large, more conveniently expressed as Log K_{ow} - e.g. K_{ow} = 100 000; Log K_{ow} = 5 (100 000 = 10^5)
- Represents the solubility at equilibrium of a chemical in octanol/solubility in water - e.g. if Log K_{ow} = 6, K_{ow} = 1 000 000, and at equilibrium the compound is 1 000 000 times more soluble in octanol than water
- K_{ow} is important because octanol is considered a surrogate lipid, thus K_{ow} approximates to K_{lw}, the lipid-water partition coefficient, and high K_{ow} means that the chemical is likely to be stored in fatty tissues of biota rather than excreted in urine, and that in aquatic environments it will partition into fish in preference to water.

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IN VIVO PERSISTENCE

- Commonly expressed as lifetime or residence time (τ) - time taken for concentration/amount to decay to 37% of original value - i.e. longer than half-life
- Lifetime = 1/k where k is a rate constant describing how quickly decay process occurs - larger the value of k, the quicker it occurs
- Long lifetimes increase chances of pollutants emissions (intake) exceeding rate of removal (excretion), and amount present in an animal or environment increasing
- Combined with high K_{ow}, results in a marked tendency to accumulate in fatty tissues. Thus diet (particularly consumption of animal fats) is main exposure route

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VAPOUR PRESSURE (P)

- Represents the tendency of a pollutant to exist as a vapour. Put another way, its tendency to evaporate.
- Called a pressure (and thus has units of Pa), because it is the pressure (or concentration) of a chemical's vapour phase in equilibrium with its solid or liquid phase
- Lower vapour pressure means less in air, and less exposure via inhalation

WATER SOLUBILITY (W_s)

- Inversely related to K_{ow}

- Higher water solubility enhances significance of water consumption as an exposure pathway

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GENERAL TRENDS IN VALUES OF PHYSICOCHEMICAL PROPERTIES

- Environmental persistence and K_{ow} all increase with increasing molecular weight (M_w) for a given class of compounds (e.g. PBDEs)
- P and W_s decrease with increasing M_w for a given compound class
- Overall effect is that higher M_w compounds are often more persistent, often (but not always) more likely to biomagnify, & diet is a more significant exposure route
- Lower M_w compounds are less persistent, more water-soluble, have a greater proportion of their total burden present in air, & water consumption & air inhalation become more significant exposure pathways

PROBLEM: FOR THE FOLLOWING POLLUTANTS, WHAT WILL BE THE MAIN ROUTE OF HUMAN EXPOSURE…DIET, WATER CONSUMPTION, OR INHALATION?

Pollutant	W_s (mol/m^3)	P (Pa)	Log K_{ow}
A	22.8	12700	2.1
B	872	70.6	1.5
C	6×10^{-8}	1×10^{-7}	6.8

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HOW USE PATTERN & LIFESTYLE CAN INFLUENCE EXPOSURE PATHWAY

- In the mid-1990s thought exposure to persistent organic chemicals like dioxins and PCBs occurred primarily through diet
- True for dioxins, but on-going presence of PCBs in indoor environments shown to lead to high indoor air concentrations that in the UK contribute substantially to overall human exposure (on average 25% ΣPCB)

Currado and Harrad (1998), EST, 32, 3043-3047

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- ***What about exposure to FRs?***
- **Example of PCBs shows exposure to bioaccumulative chemicals not necessarily restricted to diet**

PBDE EXPOSURE ASSESSMENT

- Physicochemical properties of PBDEs are broadly similar to those of PCBs
- Also extensive indoor use – primarily as Penta-BDE and Deca-BDE (also Octa-BDE)
- Combined, these led early research (2004) to investigate both diet and indoor air inhalation as exposure pathways
- UK study attributed 93% of Penta-BDE congener exposure to arise from diet; 7% from indoor air
- Similar findings for Canadian population (4% from indoor air)

Harrad et al EST (2004) 38, 2345-2350.
Wilford et al EST (2004) 38, 5312-5318.

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PBDE EXPOSURE ASSESSMENT

- This early work did not consider exposure via indoor dust
- This of importance due to the lower vapour pressures of PBDEs c.f. PCBs
- 2005 saw seminal papers emerge that considered the importance of dust ingestion to ALL PBDEs (including Deca-BDE) to North Americans
- Also highlighted the potential for especially elevated exposure of young children (dust representing 80-90% of overall exposure)

Jones-Otazo et al, EST (2005) 39, 5121-5130
Stapleton et al, EST (2005) 39, 925-931
Wilford et al, EST (2005) 39, 7027-7035

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PBDE EXPOSURE ASSESSMENT

- **European work followed, suggesting a lower but still substantial contribution from dust for Penta-BDE**
- **Interestingly, due to UK fire regulations for furniture, use of Deca-BDE and hence contamination by BDE-209 is disportionately higher than elsewhere in Europe and more akin to that in the USA**
- **Hence, dust likely the most important *external* exposure pathway for BDE-209 in the UK**

Hazrati et al, EST (2006) 40, 4633-4638
Harrad et al, Environ Int (2008) 34, 1170-1175
Harrad et al, Environ Int (2008) 34, 232-238

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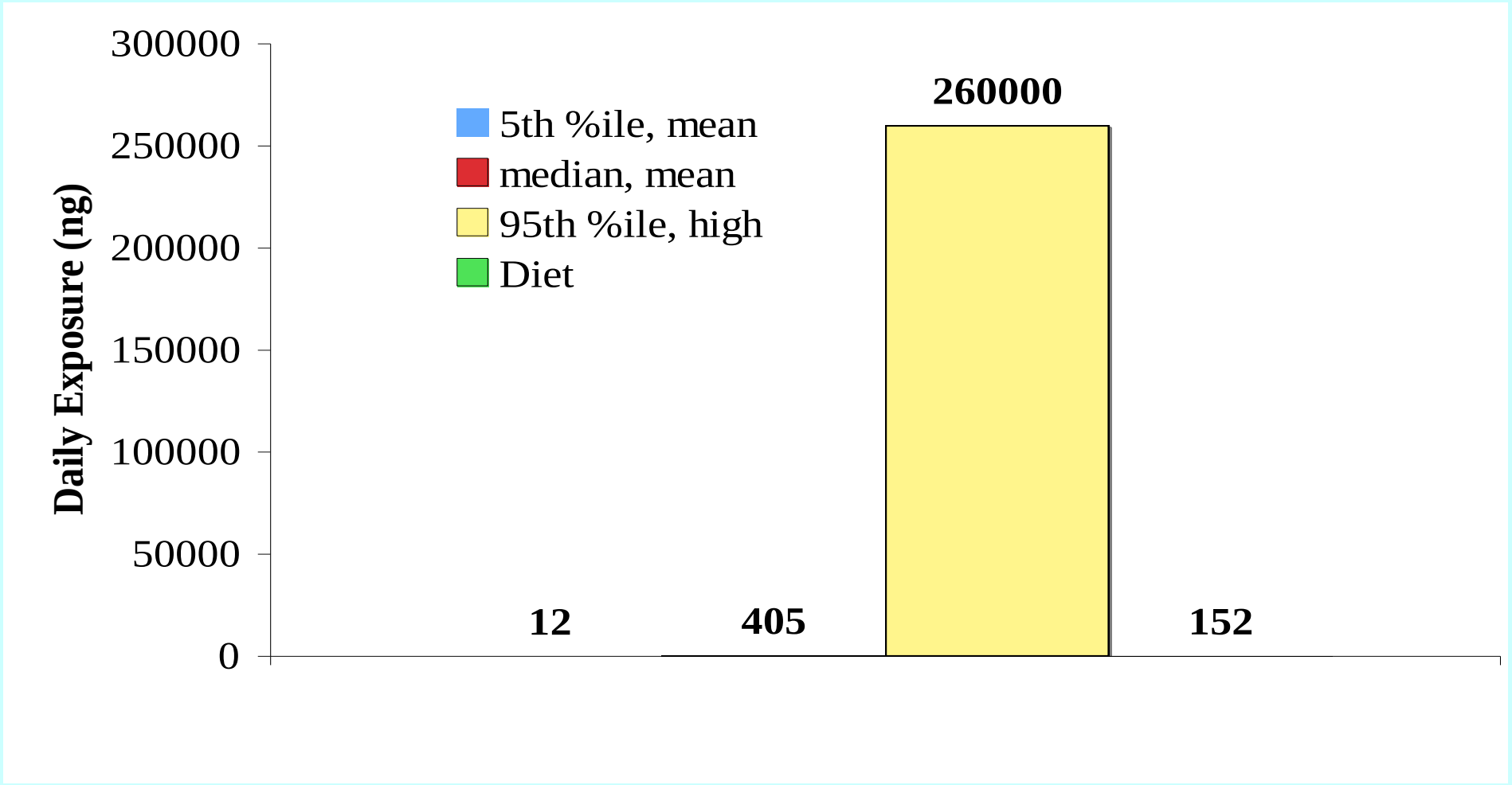
ESTIMATED UK TODDLER EXPOSURE (NG/DAY)TO BDE-209 VIA DUST INGESTION & DIET



- Mean dust ingestion 50 mg/d;
- High dust ingestion 200 mg/d

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NOT “JUST” PBDEs

- Burgeoning evidence of PBDEs in dust turned attention to other high volume BFRs
- In Europe, Hexabromocyclododecane (HBCD) used substantially (more than in the Americas)
- Concentrations in UK dust in line with those in North America
- In UK, HBCD<10xBDE-209 and 10-50x>Penta-BDE
- Dust thus an important exposure pathway of exposure to HBCD

Abdallah et al, EST (2008) 42, 459-464

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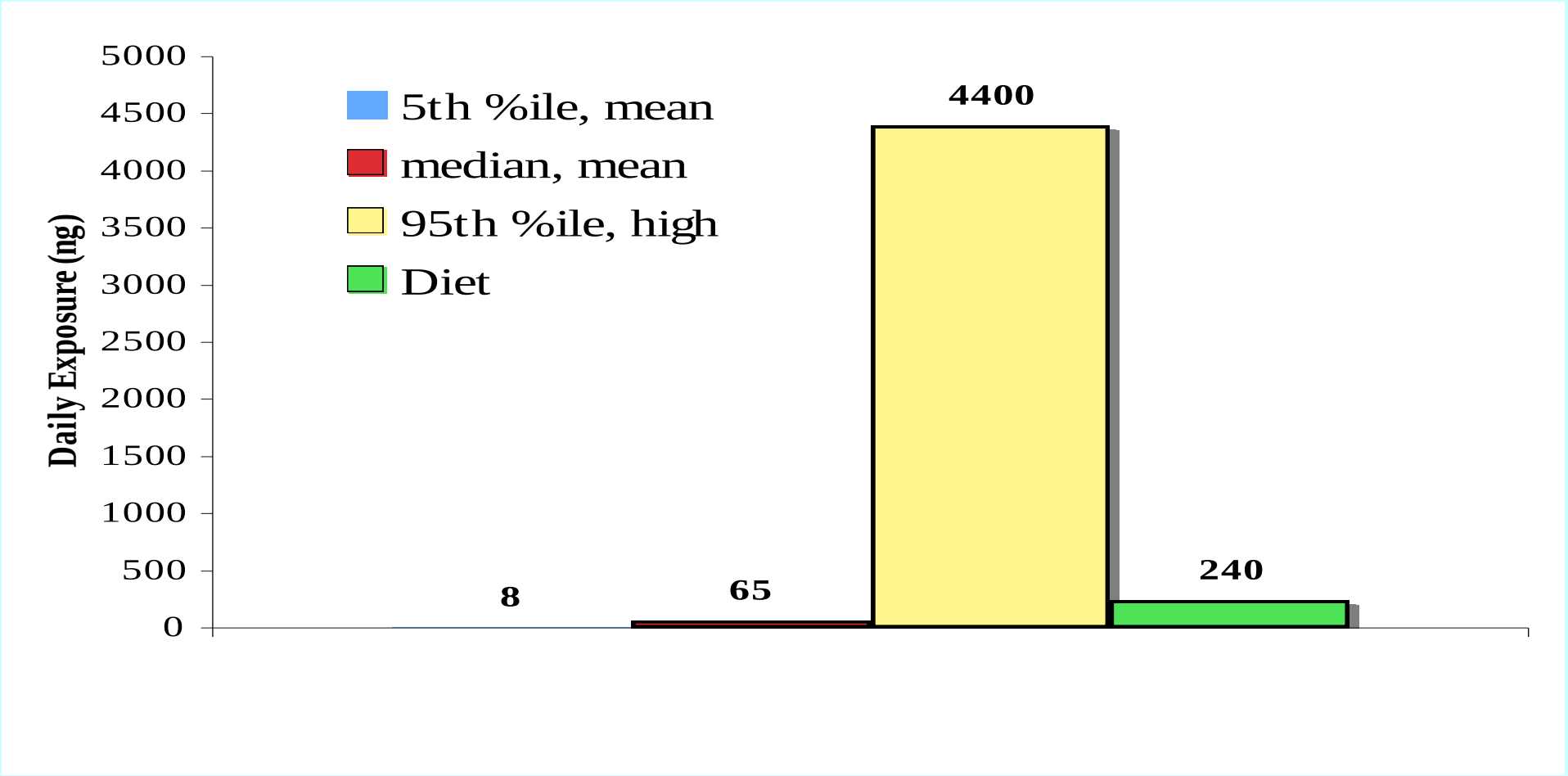
ESTIMATED UK TODDLER EXPOSURE (NG/DAY)TO ΣHBCD VIA DUST INGESTION & DIET



Mean dust ingestion 50 mg/d;
High dust ingestion 200 mg/d

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WHAT EXPOSURE PATHWAYS INFLUENCE BODY BURDENS?

- **Clearly, BFRs in dust constitute an exposure hazard, but how does it translate into body burdens?**
- **While correlation NOT reported in some studies, several studies have shown positive correlation between PentaBDE (n=4) and HBCD (n=1) in indoor dust and in human tissues of dust donors**
- **No such correlation reported for BDE-209 – only found in ~20% of humans; points to low bioavailability and half-life**
- **Important to bear in mind that dietary exposure is also substantial (in some studies correlates with body burden)**

Johnson et al, EST (2010) 44, 5627-5632

Roosens et al, EHP (2009) 117, 1707-1712

Stapleton et al, DX2011

Vorkamp et al, Environ Int (2011) 37, 1-10

Wu et al, EST (2007), 41, 1584-1589

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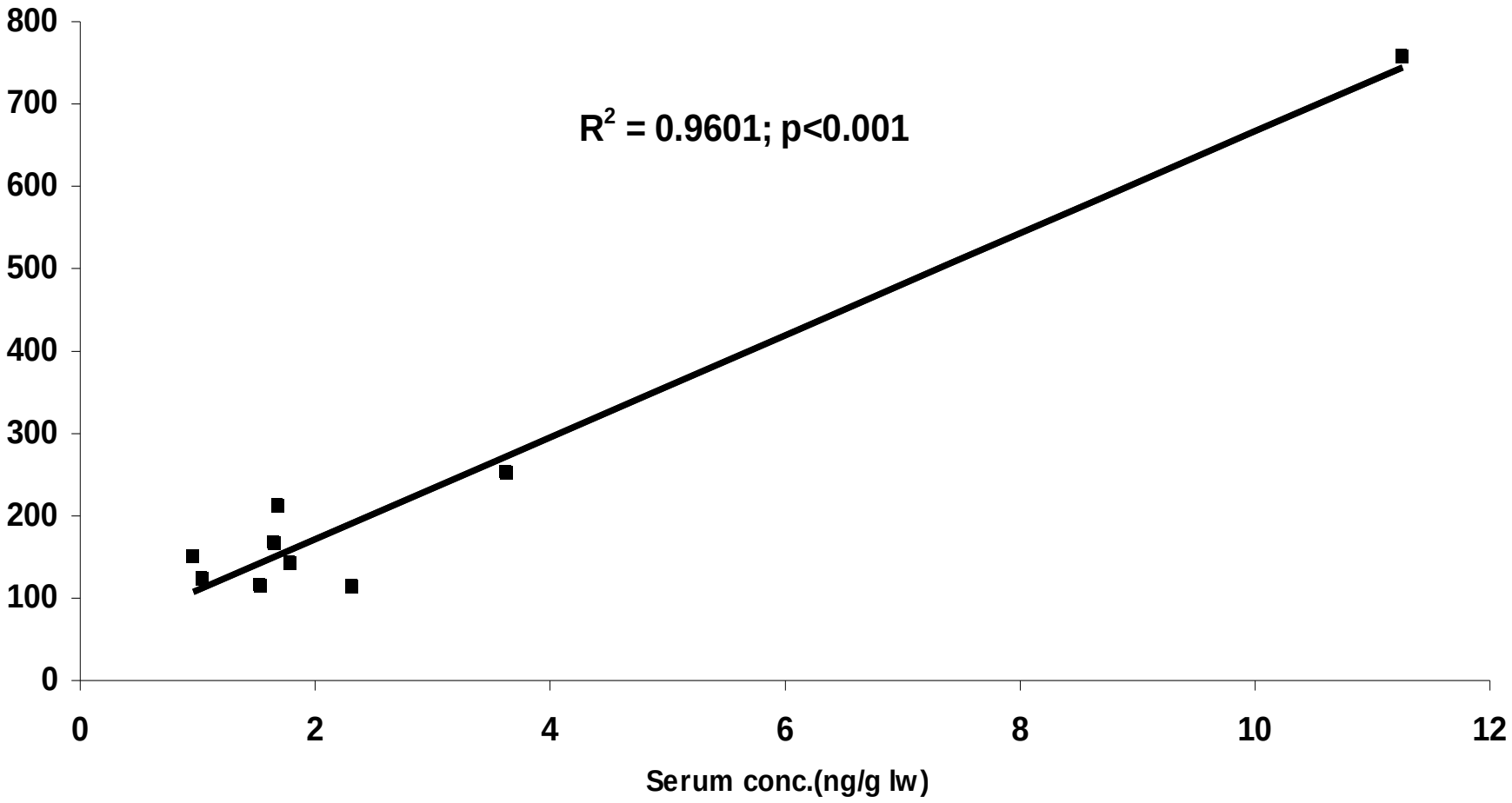
CORRELATION BETWEEN ΣHBCDs IN DUST AND BLOOD SERUM



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CAN WE EXPLAIN PBDE DISTRIBUTION IN HUMAN POPULATIONS?

- **Biomonitoring of human populations suggests a highly positively skewed distribution of Penta-BDE concentrations; with 5-10% of individuals substantially more contaminated than the rest**
- **This consistent with the distribution in dust, less so for diet**
- **Simple, one-compartment pharmacokinetic models that attempt to relate external and internal exposures, suggest dust ingestion to be the major exposure pathway of Americans to PBDEs and to contribute ~25% of UK adult body burdens of HBCDs**

Abdallah and Harrad, Environ Int (2011) 37, 443-448
Lorber, J Exp Sci Environ Epi (2008) 18, 2-19
Sjödin et al, EST (2008) 42, 1377–1384
van Bavel et al Organohalogen Compd. 2002, 58, 161-164.

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HOW DO FRs IN DUST TRANSFER TO HUMANS?

- Current thinking suggests it occurs via hand-to-mouth behaviour leading to contact with dust, with exposure occurring either via incidental ingestion or dermal absorption; or direct contact with FR-treated items
- Hand-wipes show higher PBDE levels on palms c.f. backs of hands for US adults and children
- Congener patterns in hand-wipes and dust similar
- For US office-workers, correlation between dust & handwipes ($p=0.06$ for Penta; 0.07 for BDE-209)
- Handwipes and serum also correlated ($p=0.03$) for Penta, not for BDE-209
- Frequent hand washing (>4 times/day) likely reduced exposure - Penta-BDE in serum of high handwashers 3x less
- NO dust-serum correlation for office workers, but YES for kids

Stapleton et al, EST (2008) 42, 3329-3334
Watkins et al, EHP in press
Stapleton et al Dx2011

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AGE - ARE CHILDREN MORE EXPOSED?

- Intuitively, observation suggests young children indulge more frequently in hand-to-mouth behaviour than adults
- "Hard" data on this is scarce though
- However, although derived from studies in the early 1990s designed to quantify soil ingestion; figures used for dust ingestion imply 2-4 times greater dust ingestion for toddlers

- Children (0-4 yrs) reported to have on average 16-18 hand-to-mouth contact events/hour
- No data for adults, but for older children this is much less (2/hour)

Stapleton et al, EST (2008) 42, 3329-3334
Watkins et al, EHP in press

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EXPOSURE OF CHILDREN – BODY BURDEN PICTURE

- **FRs in dust transfer to hands and children indulge in far more frequent hand-to-mouth behaviour, but what are implications for body burdens?**
- **Limited data available are consistent with higher exposures of young children**
- **Higher Penta-BDE in 0-4 year olds than adults in Norway**
- **Highest Penta-BDE in 2.6-3 year olds in Australian population**
- **Californian children have three times higher Penta-BDE than their mothers & levels of BDE 47 peak in US 4-6 year olds**
- **Evidence suggests that elevated exposure is post natal and not related to breast feeding (though BDE-153 different)**

Thomsen et al, J. Chrom. B (2007) 846, 252–263.

Toms et al, EHP (2009) 117, 1461–1465.

Eskenazi et al EHP (in press)

Sjödin et al, Dx2011

Webster et al, Dx2011

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HOW, WHEN & WHERE ARE CHILDREN EXPOSED?

- **Via breast milk** Abdallah and Harrad, Dx2011

	This study (UK)	Norway	France	Spain	Sweden	USA	Canada	Australia	China
Penta-BDE	5.95	2.34	2.51	2.14	3.57	34.00	42.80	7.6	2.53
BDE-209	0.31	0.61	1.62	2.9		0.92	0.43	0.31	3.00
∑HBCDs	5.95	1.7	2.2	47	0.45	0.5	3.8		2.4

Concentrations (ng/g lipid weight)

Lower BDE-209 body burdens in UK ≠ high UK levels in dust – suggests dust not crucial driver for adults, but toddlers??

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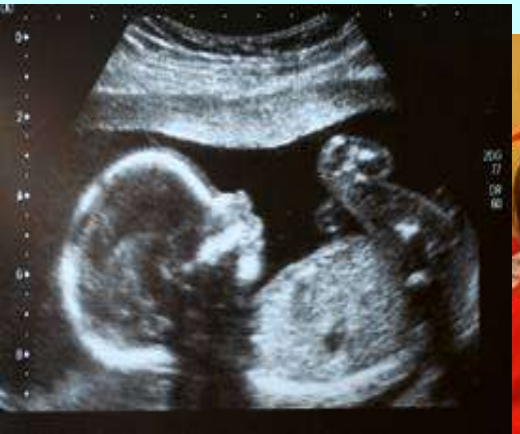
HOW, WHEN & WHERE ARE CHILDREN EXPOSED?

- ***In utero*** **– reports of PBDEs in human cord blood, placenta & fetal liver**
- **Via diet – note that children ingest more food than adults normalised to body weight**
- **Via inhalation**
- **Via contact with dust**
- **Indoor exposures shown to occur in homes, cars, and schools/kindergartens**
- **Data to date do not suggest *major* differences in levels of BFR contamination between different microenvironment categories**

Harrad et al EST (2010) 44, 4198-4202

Rawn et al, Dx2011



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SHOULD WE BE CONCERNED?

- **For HBCD - recent EFSA statement concluded "that current dietary exposure in the EU does not raise a health concern. Also additional exposure, particularly of young children, from house dust is unlikely to raise a health concern."** ***Degradation products??***
- **For BDE-99, high-end exposure estimate for UK children exceeds 10-fold a recently published HBLV of 0.23-0.30 ng/kg bw/d. "Typical" exposure estimate is 20-25% of the HBLV.** ***North American exposure?***
- **For BDE-209, high-end exposure estimate for UK children is double USEPA RfD of 7 μg/kg bw/d.**

Bakker et al Mol. Nutr. Food Res. 2008, 52, 204–216.

Harrad et al EST (2010) 44, 4198-4202

www.efsa.europa.eu/en/efsajournal/pub/2296.htm?WT.mc_id=EFSAHL01&emt=1

USEPA cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid)190307

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WHAT ABOUT CHILDREN IN DEVELOPING COUNTRIES?

- Likely lower exposure than in developed world via pathways discussed earlier
- However, some evidence that elevated exposure occurs due to unregulated recovery of materials from waste dumps
- Nicaraguan children working as waste scavengers and living at a municipal waste site (little e-waste), have Penta-BDE levels 20-50 times those in co-located urban children, and exceed those in the developed world
- Concern must be for populations elsewhere where emissions from waste dumps are uncontrolled – especially where e-waste is involved (also fabrics/furniture)

Anathasiadou et al EHP (2008) 116, 400-408

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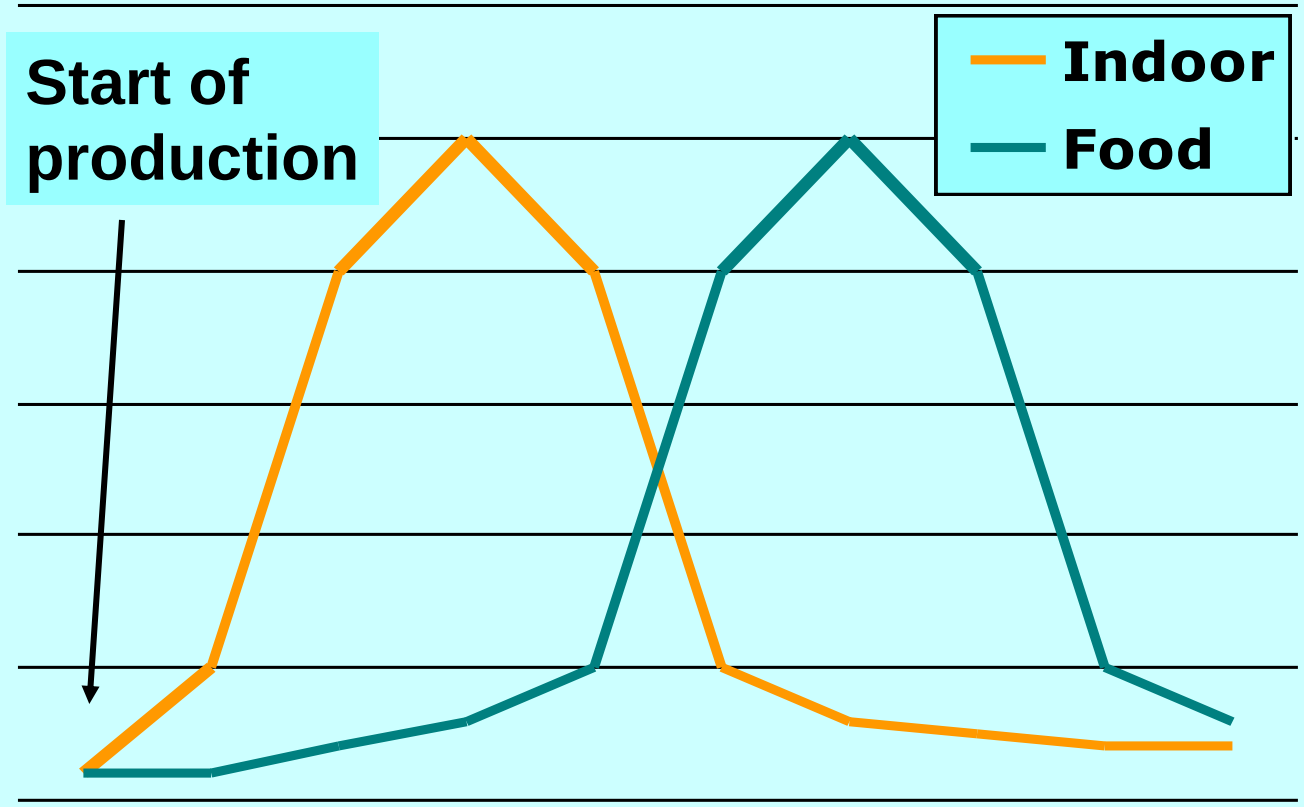
FUTURE EXPOSURE SCENARIOS



Harrad & Diamond (2006) *Atmos Environ*, 40: 1187-1188

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HAVE WE LEARNT FROM PCBs?

- I contend not as much as we should
- PCBs in UK indoor air did not fall between 1997 and 2003-04
- Likewise, although UK dietary exposure to dioxin-like PCBs (for which combustion is an important source) fell between 1992 and 2001, exposure to other PCBs has not
- “Just” banning new manufacture and use is unlikely enough

Currado and Harrad EST, (1998) 32, 3043-3047
Harrad et al, EST (2006) 40, 4633-4638
Defra, UK NIP for Stockholm Convention, pp 37-38

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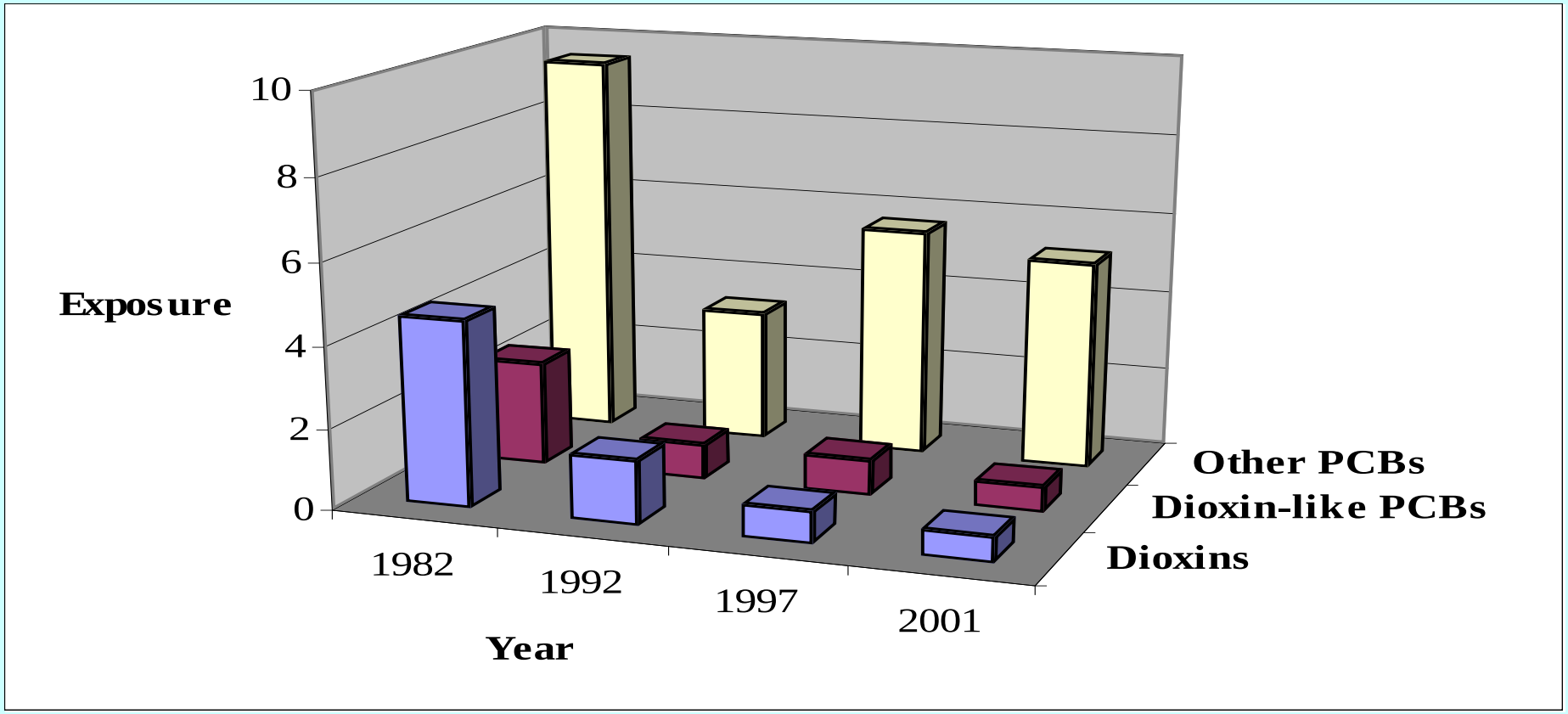
TEMPORAL TREND IN UK DIETARY EXPOSURE TO DIOXINS AND DIOXIN-LIKE PCBs (pg/kg bw/d) + NON-DIOXIN-LIKE PCBs (ng/kg bw/d)



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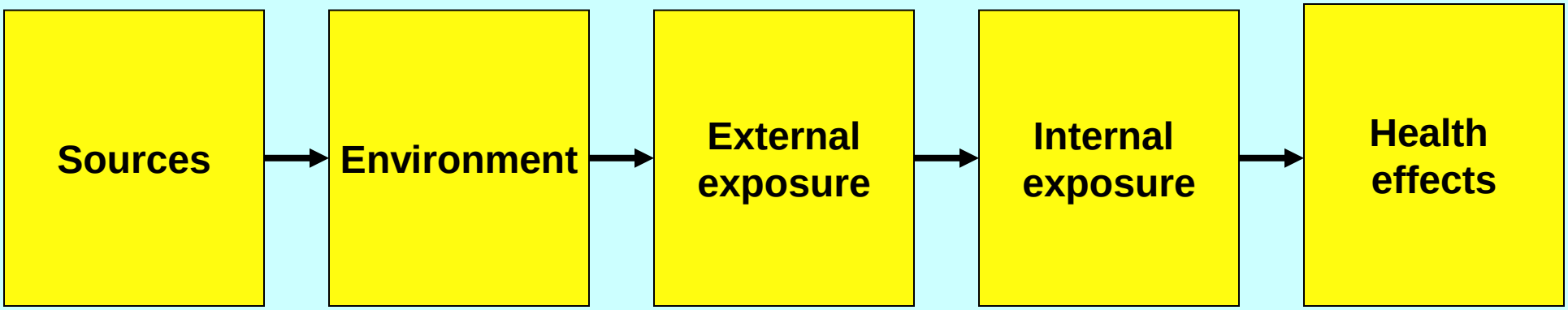
FUTURE RESEARCH DIRECTIONS



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FUTURE RESEARCH DIRECTIONS

- Health impacts of exposure – especially related to early-life
- How and what rate do FRs migrate from treated goods/materials?
- We need to monitor BOTH external AND internal exposure
- Improve understanding of how external exposure relates to body burden for BDE-209
- More data on human contact/ingestion rates with dust
- Exactly how are we exposed to FRs in dust? Ingestion or dermal?
- If via ingestion, how effective is absorption?
- Develop effective non- or minimally invasive ways of biomonitoring children

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FUTURE RESEARCH DIRECTIONS

- Investigate releases from end-of-life treatment of flame-retarded waste
- Use findings to inform sustainable waste management
- Investigate long-term environmental fate and impact of FRs – degradation products?
- Restrictions on PBDEs and HBCDs, mean we need to evaluate fully exposure and health impacts of replacements (NBFRs, OPFRs)
- Sustainable approaches to fire safety required

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INFLAME

DON’T TRY THIS AT HOME ☺

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