



# Consuming Chemicals

Hazardous chemicals in house dust as an  
indicator of chemical exposure in the home

**GREENPEACE**

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# Introduction

*"Manufactured chemicals are widespread in the air, soil, water sediments and biota of Europe's environment following the marketing of up to 100 000 chemicals in the EU, their use and disposal and degradation. There is a serious lack of monitoring and information on these chemicals...widespread exposures to low doses of chemicals may be causing harm, possibly irreversibly, particularly to sensitive groups such as children and pregnant women..."*

*European Environment Agency (1998).*

## **Chemicals in Europe**

Current regulation of chemical production and use in Europe is weak, cumbersome and ineffective. This has led to a situation in which there are very few data on the great majority of the thousands of chemicals currently used by industry, and next to nothing is known about their potential environmental and human health impacts. According to the European Commission;

*"The lack of knowledge about the impacts of many chemicals on human health and the environment is a cause for concern...understandably the public is worried when hearing about the exposure of their children to certain phthalates released from toys and about increasing amounts of the flame retardant pentabromo diphenyl ether in human breast milk...legislative action takes too long before yielding results.*

*European Commission (2001)*

The data presented in this report shows just how serious and widespread the problem of chemical contamination is. This contamination is not just of the environment "out there". It affects our homes, our offices, our daily lives. Moreover the pollutants we have targetted for investigation are not coming from traffic fumes, industrial chimneys or pesticides. They are brought into our homes as unseen and unlabeled chemical additives in everyday consumer products. It may seem surprising that the sort of chemicals that we tested for are used in everyday consumer products at all. They are the same chemicals currently causing great concern among scientists, governments and environmental groups because we know they can interfere with reproductive and immune systems, imitate hormones and cause cancer in a variety of living organisms. It is still more surprising that they are appearing in house dusts with such frequency because one of the arguments of manufacturers has been that most of these chemicals are bound into products and do not therefore represent an exposure threat.

It is important to recognise that we cannot be certain the chemicals in question are actually having adverse effects on human beings. There is simply no way of doing a controlled experiment on human subjects to find out. As the European Commission, the European Environment Agency, the United Nations and others have made clear, we just do not know. It is because we do not know that we must take action.

What this report shows is that chemicals that may present a long-term hazard to human health are present in significant

amounts in virtually every one of over 100 homes we visited. Here then is a clue as to why levels are increasing, exponentially in some cases, in human breast milk, blood and other body tissues. We cannot assume that there will be no adverse effects from this. We expect government to act to end this state of affairs.

The action required is simple. The EU has proposed new laws that will enable the chemicals of highest concern, the sort of chemicals we have studied in this report, to be identified. An "authorisation" will be required to continue production of these substances. Greenpeace supports this approach. But without the next step it will mean nothing. The second step must be to clearly state that where a viable, safer alternative exists, an authorisation will not be granted. If a viable, safer alternative does not exist and the chemical in question has a socially useful function, production can continue for a limited time period only, while a viable alternative is developed. This is the principle of mandatory substitution. If this principle is enshrined into EU law we will have taken a giant step towards ridding our environment, our homes and our lives, of chemicals that enter our bodies and linger there, threatening cancer, genetic damage or any of the other effects we know they are capable of in some species.

For too long the public in Europe have faced what sometimes seems like an onslaught of alarming facts detailing their daily exposure to toxic chemicals. For too long they have felt helpless to prevent this chemical assault on themselves and their children. The pending EU chemicals legislation is an unprecedented opportunity to change that. It is, for the ordinary citizen, a glimpse of light at the end of a long tunnel. That light represents an environment free of intentionally produced hazardous man-made chemical contaminants. Europe can lead the way towards that goal and in the process revitalise its chemical industry, ensuring it has a healthy future in the manufacture of more sustainable products.

Greenpeace is not opposed to the manufacture and use of synthetic chemicals, but we do insist that it is unacceptable for a child to be born already contaminated by industrial pollutants. Put another way, we contend that the chemical industry, and downstream users of its products, have no right to subject the population at large to involuntary exposure to industrial chemicals, many of which have unknown characteristics. But that is exactly what they currently do. National and European governments have a duty to protect their citizens from such exposure. But that is exactly what they currently do not do.

Greenpeace has two objectives in publishing this report. One is to make sure there is no doubt about how seriously and ubiquitously our environment, including our homes, is being contaminated. The second is to ensure the public know that their elected representatives have, right now, an opportunity to change that.

## **References**

*European Environment Agency (1998). Chemicals in the European Environment: Low Doses, High Stakes?*

*European Commission (2001). White Paper: Strategy for a Future Chemicals Policy*

# Executive Summary

Although the widespread presence of hazardous man-made chemicals in the environment is becoming increasingly well documented, few people are aware that many of these same chemicals are used as additives in consumer goods we buy and use in the home everyday. From carpets and curtains to toys and televisions, computers and printers to cosmetics and perfumes, chemical additives are a hidden fact of modern life. They are rarely labelled and never seen, but they are nearly always there.

Of course, they are generally there for a reason: to make plastics soft or stop them breaking down; to carry perfumes; to protect against fire; to kill dust-mites or mould. The problem is that, as a consequence of their use in consumer goods, we are constantly exposed to these chemicals and the hazards they pose. They can escape from products during normal use, or through wear and tear over time, contaminating the indoor environment of our homes.

Surprisingly, the extent of this indoor contamination and its significance in terms of overall chemical exposure has rarely been studied. Where such studies have been conducted, evidence points to widespread contamination of the home environment with a variety of man-made chemicals. Some come predominantly from outside sources, such as lead from traffic pollution. Others result from deliberate use of chemicals (e.g. insecticides) in the home. However, by far the majority arise from their use in consumer goods commonly used in the home. These include hazardous chemicals such as:-

- hormone disrupting alkylphenols, used in cosmetics and other personal care products
- phthalate esters toxic to reproduction, used mainly to soften PVC (vinyl)
- immunotoxic organotin compounds used to stabilise PVC or to kill dust-mites
- brominated chemicals which mimic thyroid hormones, used as fire retardants in furniture and electronic goods
- chlorinated paraffins which may be carcinogenic, used in plastics, paints and rubbers

This study has been conducted in order to describe in more detail the chemical environment of the home, using samples of dust collected from 100 volunteer households representing a total of 10 regions across the UK from Scotland to the South West. Working with laboratories in the UK, the Netherlands and Germany, these samples were analysed (either singly or after “pooling” samples from individual regions) for each of the five target groups of hazardous chemicals listed above. In addition, some samples from each region were investigated for the presence of other chemical contaminants. Finally, a small number of dust samples from other European countries were included for comparative purposes (three each from Finland and Denmark, two from Sweden and one each from France and Spain).

All dust samples from UK households contained phthalates, brominated flame retardants and organotin compounds. More than three quarters also contained nonylphenol and short-chain chlorinated paraffins. With just one exception, all UK

dusts investigated also contained a range of other man-made chemicals, including solvents, pesticides and plastic additives. On average, each gramme of dust contained a total of around half a milligramme (1 part in 2000) of the five hazardous chemical groups specifically quantified. The identification of a diversity of other man-made chemicals in the qualitative screening analysis suggests that the overall chemical content of house dust may be substantially higher.

UK dusts contained from 1.6 to over 1000 parts per million (ppm) of phthalates (average 430 ppm, or 0.43 milligrammes per gramme), with the reproductive toxin DEHP the most abundant. Alkylphenols (primarily the estrogen mimic nonylphenol) were found at up to 36 ppm (average 10.9), short-chain chlorinated paraffins up to 13 ppm (average 4.3) and organotins up to 5 ppm (average 2.7). Although banned from some uses because of the hazards they pose, chlorinated paraffins are still allowed to be used as additives in plastics, rubbers and other materials.

Of the brominated flame retardants present, the most abundant was decabromodiphenyl ether (deca-BDE or BDE-209, at 3.8 to 19.9 ppm), used widely to flame-proof plastics and textiles. While our exposure to this persistent chemical through other routes is generally considered low, intake through exposure to contaminated dusts in the indoor environment may well be significant. Other more bioaccumulative brominated flame retardants, for which exposure is more commonly linked to intake through foodstuffs, were also present at substantial concentrations in the dusts. For example, penta-BDE, shortly to be banned from sale and use in Europe as it accumulates in breast milk, was found in all dust samples at between 0.018 and 2.1 ppm. Hexabromocyclododecane (HBCD), a common component of textiles and expanded polystyrene, was found at between 0.94 and 6.9 ppm across all regions. Tetrabromobisphenol-A (TBBP-A) was detected in 4 out of the 10 regional samples at levels up to 0.34 ppm, despite the assumption that it is generally tightly bound to the products in which it is used.

Concentrations of these hazardous substances varied from sample to sample, although there was no clear trend for higher contamination overall in any one region. For example, the highest levels of organotin compounds, used as stabilisers in PVC (vinyl) products, were found in the North East, North West and Scotland, whereas the highest levels of BDE-209 occurred in the South West, South East and East Midlands. The pooled sample from London contained the lowest BDE-209 levels but the highest levels of short-chain chlorinated paraffins, while the sample from the East Midlands showed the reverse.

Among the other man-made chemicals most commonly identified in the house dusts were styrene (a component of polystyrene), the pesticide permethrin, a number of non-phthalate plastic softeners (plasticisers) and a chlorinated organophosphorus chemical which may be a commonly-used flame retardant. Permethrin, a hazardous pyrethroid insecticide, was found in just under one quarter of houses

sampled, possibly resulting from its incorporation into certain brands of carpet as a treatment against dust-mites.

Patterns of chemical contamination in the non-UK households sampled were similar to those in the UK. One of three samples from Finland contained the highest levels of both the phthalate softener DEHP and the organotin stabiliser DOT recorded in this study, almost certainly resulting from an abundance of PVC products. The three samples from Denmark were relatively uncontaminated with phthalates and organotins while those from Sweden, France and Spain contained intermediate levels.

Levels of BDE-209 in single samples analysed from Finland and Denmark were between 10 and 100 times lower than those found in UK samples in this study, though concentrations of other brominated flame retardants were more similar. At the same time, the single Danish sample analysed for TBBP-A yielded the highest level recorded in this study (0.4 ppm). Levels of short-chain chlorinated paraffins were in the same range as for UK samples, confirming the widespread distribution of these persistent chemicals in the home.

Taken together, these data show that the ongoing use of hazardous chemicals in consumer products is leading to ubiquitous and complex contamination of the home environment. Patterns of contamination in any one household, or even in regional samples pooled from several homes, will depend greatly on the types of products present in those homes sampled. Clearly these results cannot be taken as fully representative of dust contamination levels in the 10 regions sampled. Nevertheless, these data as a whole do provide a snap-shot of chemical contamination in the home across the UK, and in other parts of Europe. In short, they confirm that we are all living with the chemical consequences of the widespread use of hazardous additives in consumer goods.

At the same time as these data illustrate the pervasive nature of hazardous chemicals, they also indicate that exposure to dusts in the home is potentially a significant route of direct human exposure to these chemicals.

This may be of particular concern with respect to children, as other studies have shown that they have the greatest exposures to dust-related contaminants through inhalation, ingestion and direct skin contact. Of course, we can never be certain that such exposure is causing adverse health effects, but given the hazards associated with the chemicals in question, there is no reason for complacency. To date, the issue of chemical exposure in the home has generally been poorly investigated and improperly assessed.

It is vital that consumer products should be safe to use and this must also include freedom from hazardous chemicals. Requirements for fire safety, commonly conferred through the use of hazardous brominated or chlorinated flame retardants or chlorinated paraffins, can already be met through the use of less hazardous alternatives, including through the use of different materials or designs which make products inherently less flammable. Moreover, many of the indoor chemical hazards identified in this study could be significantly reduced by the use of less hazardous and more sustainable alternatives to the plastic PVC, a source of phthalates, organotins and other hazardous additives. Such alternatives are already available for all PVC products used in the home.

During 2003, the opportunity exists for the UK, along with other European governments, to take decisive and effective action to tackle the problem of chemical use and exposure, in the home and elsewhere. Recognising the scale of the problem, the lack of knowledge about chemical impacts on human health and the poor progress to date in developing protective measures, the European Commission is currently preparing new regulations to control hazardous chemicals. The intention is that these new laws should provide for a high level of protection for the environment and human health. If they are to do so, however, it will be vital that they effectively address and ultimately prevent the use of hazardous chemicals in consumer goods through their substitution with less hazardous alternatives. This is the only way in which the chemical safety of our home environment can be ensured.

### Summary of analytical results for key chemical groups in UK dust samples

Compound	UK average value	UK lowest value	UK highest value
Phthalates (ppm)	431.7	1.6	1019.1
Alkylphenols (ppm)	10.9	nd	36.1
Deca-BDE (ppb)	9820	3800	19900
HBCD (ppb)	3158	940	6900
TBBP-A (ppb)	116	<10	340
Organotins (ppb)	2669	1581	5047
Chlorinated Paraffins (ppm)	4.34	<0.12	13.0

### **Chemicals in the home**

Many of the common consumer products we use or come into contact with every day, in the home or in the office, contain chemical additives. These additives are present to give the products we buy certain properties. For example, some additives are included to make plastics flexible or textiles fire retardant, others to kill dust mites or mould or to carry perfumes.

We are generally quite oblivious to the chemical consequences of the products we buy, use and ultimately dispose of, and understandably so as information on chemical composition of consumer products is rarely provided. At the same time, it could be argued that as long as those chemicals are serving useful functions, this is all that matters. So should we be concerned about chemical additives in consumer goods?

The problem is that some of the chemicals commonly incorporated into a wide range of consumer goods are inherently hazardous, i.e. they are toxic to mammals and/or other animals, do not readily degrade into harmless by-products and can enter the food chain. Moreover, they do not stay locked away inside the plastics, textiles or other materials in which they are used and may even leach out as a result of normal everyday use of the products. So, at the same time as they perform the functions for which they were intended, many additives can also present us with a significant but unseen chemical hazard.

For example, some phthalate esters (phthalates) widely used as softeners in flexible PVC (vinyl) flooring, toys or other products are known to be toxic to reproductive system development in mammals. Alkylphenols, reportedly used in some shampoos and other "personal care products", and a range of brominated chemicals used as fire retardants, can interfere with hormone systems vital to growth and development. Organic compounds of the metal tin (organotins), used as fungicides in some carpets or as stabilizers in PVC products, are harmful to the immune system in mammals, including humans.

We are therefore exposed to these chemical hazards on a daily basis, through use of products containing them, through contact with dusts in which these chemicals can accumulate and to some extent through the air we breathe in the indoor environment.

As well as being inherently toxic, widely used chemical additives such as organotins, brominated flame retardants and chlorinated paraffins are also very persistent, i.e. once released into the indoor or outdoor environment, they do not readily break down into harmless by-products. Rather, they may simply become ever more widely dispersed through the environment, carried on air currents or in water. Moreover, because of their chemical nature, some have a high affinity for fatty tissues and may therefore accumulate over time in the bodies of animals, through the food chain and in humans.

Chemicals possessing such a combination of properties are commonly known as Persistent Organic Pollutants, or POPs. The nature and extent of the threats presented by POPs, now on a global scale, are increasingly being recognized and efforts are being made to bring them under control (for example the 2001 Stockholm Convention on POPs). At present, however, only a handful of chemicals fall under such controls. Many chemicals with similar POP-like properties remain in widespread production and use, both within Europe and further afield.

As a consequence, over many years hazardous chemical additives have become ever more widespread as environmental contaminants. Chemical releases may occur at any stage in the lifecycle of a product, during its manufacture, during use or after it enters the waste stream. Some of the most commonly used chemical additives are now even detectable in remote areas of the planet, such as the high Arctic and the deep oceans. And as a further consequence, we are therefore exposed to them through our food.

We all carry the consequences in our bodies, in the form of residues of these toxic and persistent chemicals in our blood and body fat. For those chemicals which have the highest propensity to accumulate in fatty tissues, food may be the most significant source of our daily exposure. For others, exposure through the use of consumer goods, or through contact with contaminated air or dusts may be equally or even more significant. However, because so little information is available concerning the presence and quantities of chemical additives in household or other consumer products, nobody really knows.

This study is an attempt to describe in more detail the chemical environment of the home, through the collection and analysis of house dusts. This is only a small part of the picture, and it will not answer the question of how much of our body burden of hazardous chemicals results from exposure in the home. Nevertheless, it should help to improve our level of knowledge and understanding of the chemical environment in which we live.

### **House dust as a chemical indicator in the home**

House dust itself is not a simple physical or chemical substance, but a highly heterogeneous mixture of organic and inorganic particles and chemicals. Its precise make-up in any particular building, or even room, will depend on a large number of factors including the location and construction of the building, the use of the room, types of decorating and furnishing materials used, heating and ventilation systems, how well and often the area is cleaned, even the time of year (Edwards *et al.* 1998, Butte and Heinzow 2002).

The human health hazards of dust which stem purely from its physical nature, especially from the presence of very small particle sizes, have been well recognized and documented for many years. The significance of dusts as "sinks" and reservoirs of chemicals in the home, and therefore as potential sources of chemical exposure, are much less well studied.

We may be exposed to dust, and any chemicals it may contain, through a combination of inhalation, ingestion from contaminated food, toys or other surfaces and even direct absorption of chemicals through the skin (Lewis *et al.* 1994). Dusts, both indoor and outdoor, may be a particularly significant source of chemical exposure for children (Butte and Heinzow 2002). For example, in terms of outdoor exposure, Yin *et al.* (2000) highlight the substantial contribution to summertime lead exposure in children from contaminated street dusts. In the indoor environment also, dust exposure is increasingly being taken into account in assessment of chemical exposure in children (Wilson *et al.* 2001).

As well as the significance of direct exposure, the resuspension of contaminated dusts in the atmosphere may contribute to the more widespread distribution of dust-bound chemicals in the environment. Regular disposal of house dusts collected in vacuum cleaners undoubtedly also acts as a potential source of more pervasive contamination, in the same way that run-off of outdoor dusts (especially roadside dusts) to sewers and storm-drains can lead to substantial secondary inputs to rivers (Irvine and Loganathan 1998).

Butte and Heinzow (2002) provide the most extensive review to date of investigations into chemical contaminants in house dust. Although they summarise the numerous surveys conducted into the consequences of household pesticide application, particularly in the USA, Butte and Heinzow's review also serves to highlight the paucity of available data relating to other chemical contaminants. This is especially true for those chemicals which are not deliberately or knowingly used in the home but which occur, as noted above, simply as a consequence of their widespread use in consumer products. Thus, although it is well known and documented that phthalates, organotins and brominated flame retardants migrate out of products during use and through normal wear and tear, very few data exist to describe their prevalence in house dust.

Rudel *et al.* (2001) reported the presence of phthalates, pesticide residues and polycyclic aromatic hydrocarbons (PAHs) in office and household air and dusts, with phthalates present at concentrations up to 0.5 g per kg of dust (i.e. 500 mg/kg or parts per million, ppm). Nonylphenol compounds were also present, reaching levels of up to 14 ppm. Moreover, a wide array of other compounds, many of which are suspected endocrine disruptors i.e. (capable of interfering with hormone systems) were also identified in the majority of the houses and offices studied. Lagesson *et al.* (2000) similarly reported a variety of man-made chemicals as common constituents of indoor dusts.

Both brominated and chlorinated fire retardants have also previously been reported as contaminants of indoor air and dust. Bergman *et al.* (1997) identified a range of such chemicals in dust suspended in the air of a number of computerized offices in Stockholm. More recently, Sjödin *et al.* (2001) reported similar findings at an electronics recycling plant, as well as in other work environments, with some of

the highest concentrations being those of the largest molecules, chemicals for which exposure from other sources is often thought to be insignificant. Ingerowski *et al.* (2001) described the presence of chlorinated organophosphate compounds, used as flame retardants in foams, paints, varnishes and wallpapers, in indoor air and dust (at levels up to 375 ppm in dust).

In the year 2000, Greenpeace International in conjunction with Greenpeace national offices collected samples of dust from parliament buildings in a number of European countries. All samples were found to contain substantial levels of brominated flame retardants and organotin compounds (Santillo *et al.* 2001, Leonards *et al.* 2001). Once again, the heavier (larger molecular size) bromine chemicals (especially decabromodiphenyl ether, or deca-BDE) were present at the highest concentrations, although the lighter and more bioaccumulative compounds were also detected in all samples. As noted above, this is particularly significant as it suggests that for compounds such as deca-BDE, for which exposure through food is likely to be less significant, a combination of ingestion, inhalation and skin contact with dust residues may contribute substantially to overall exposure. Deca-BDE was found at between 0.26 and 6.9 ppm in the Parliament dusts. Organotin compounds were also prevalent, with total concentrations ranging from 0.49 to 3.5 ppm, dominated by those forms (mono- and dibutyl tin, or MBT and DBT) used as stabilizers in PVC.

The Parliament dust study, along with the limited array of other studies published to date, illustrate the utility of dust analysis as one way to characterize further the indoor chemical environments to which we are most often exposed, namely the workplace and home. The current study, reported below, aims to extend the existing knowledge base by applying similar techniques over a wider area and to a greater number of potential chemical contaminants.

Samples of dust have therefore been collected for analysis from 100 households or other buildings across the UK, split in to 10 regions in order to provide representative data on as broad a geographical spread as possible. Together the data will provide the most extensive UK survey so far of the chemical environment in the home. The regional approach may also allow the determination of any consistent spatial trends in contaminant distributions, such as may be expected if there were significant external sources or drivers of contaminant levels, or perhaps substantial regional differences in lifestyle, though at the outset, no regional trends are expected.

As a supplementary investigation, a small number of samples have been included from households in other European countries, namely 3 from Denmark, 3 from Finland, 2 from Sweden and 1 each from France and Spain. The intention is that these will provide some comparative data to those for the UK.



### **Chemicals targeted for investigation**

The main focus of this current study is the presence in dusts of hazardous chemicals which arise as a consequence of their widespread (though poorly documented) use in everyday consumer products in the home. We therefore decided to target the analyses towards five main compound groups, based on their reported high volume use in common household furnishings and other products and on their intrinsic hazardous properties. However, though these five groups are not the only hazardous chemicals used widely in consumer products, they are representative of a much wider problem:-

- *Alkylphenols* (nonylphenol, octylphenol and their derivatives) – primarily used as non-ionic surfactants in industrial detergents, though also used in textile and leather finishing treatments, water based paints and as components of some personal care products;
- *Brominated flame retardants* (polybrominated diphenyl ethers or PBDEs, hexabromocyclododecane or HBCD and tetrabromobisphenol-A or TBBP-A) – applied to textiles and/or incorporated into plastics, foams and components of electrical goods to prevent or retard the spread of fire;
- *Organotin compounds* (butyltins, octyltins) – including mono- and di- butyl and octyl tins, used as stabilizers in plastics, especially PVC, and tri-butyltin (TBT) used as a treatment against dust mites and mould in some carpets and PVC floorings;
- *Phthalate esters* used as softeners in flexible PVC products, including floors, wallpapers, furnishings, clothing and toys, as well as ingredients in cosmetics and perfumes;
- *Short-chain chlorinated paraffins* (SCCPs) – now less widely manufactured and used than before, but still used in some plastics, rubbers, paints and sealants and still a major contaminant from the past.

Each of these groups is chemically distinct and exhibit markedly different properties, in some cases even within individual groups. Nevertheless, they do all share a number of common characteristics which justify the established and increasing concerns surrounding their use:-

- they are all toxic to one or more organisms, though they are effective through a diversity of different mechanisms;
- they are not readily broken down to harmless by-products, i.e. they tend to persist in both the outdoor and indoor environment;
- they are all able to leach out of, or otherwise be lost from, consumer products during normal use and/or wear and tear;
- they have all been reported as contaminants in the human body, in many cases as widespread contaminants, though at a wide range of concentrations.

The hazards presented by these chemicals or chemical groups are firmly established. For example:-

- Short-chain chlorinated paraffins are classified under EU law as being “very toxic to aquatic organisms” and as presenting a “possible risk of irreversible effects” as a consequence of their carcinogenic properties;
- The organotin compound TBT is classified as “harmful in contact with skin, toxic if swallowed, irritating to the eyes and skin” and as presenting “danger of serious damage to health by prolonged exposure through inhalation or if swallowed”;
- The phthalates DEHP and DBP (dibutyl phthalate) are classified as “toxic to reproduction”.

# Sampling programmes and analytical methods

## UK samples

### Sample collection

During October 2002, Greenpeace UK issued a number of appeals through the national and local media for volunteers to participate in this study by allowing their houses or business addresses to be sampled. Ultimately, 100 volunteers were selected from the responses received, representing a wide geographical spread across the UK mainland. The 100 addresses chosen were primarily private households, though some business addresses (e.g. local radio stations) were also among those selected. In total, the final sample set comprised 10 individual samples from each of 10 regions of the UK mainland:-

All samples were collected between the dates of 30th October and 8th November 2002 by prior arrangement with the selected volunteers. Volunteers were asked to avoid vacuum cleaning their homes for at least one week prior to the samples being collected in order that the quantity of dust collected would be sufficient for analysis.

All samples were collected using the same make and model of vacuum cleaner (AEG Vampyr 1700 Watt), using a new AEG dust filter bag for each address sampled. The number of rooms sampled varied from one location to another (minimum of one complete room) depending on the quantity of dust present, again in order to obtain sufficient dust to allow analysis. Thus, while the dusts obtained may be considered representative of the household in question, they do not necessarily represent dusts from any one particular room.

After each sampling, the dust filter bag was removed from the vacuum cleaner, sealed with tape and sealed again inside a strong polyethylene (PE) bag. All samples were then returned in region batches to the Greenpeace Research Laboratories at the University of Exeter for processing.

### Sample processing

On receipt at the Greenpeace Laboratories, all samples were immediately sieved through a pre-cleaned, solvent-rinsed<sup>1</sup> 2mm gauge sieve to remove any large and recognizable particles and debris which might otherwise have disproportionately affected (biased) the sample results. All residue retained by the 2mm sieves was immediately disposed of. The fraction passing through the sieves in each case was collected on fresh, solvent-rinsed aluminium foil. Contact with the dust during this operation was avoided and gloves used to handle the filter bags and sieves were pre-cleaned with analytical grade pentane in order to remove any organic residues from the manufacture of the gloves. Sieving was carried out in a draught-free environment in order to avoid sample loss and cross-contamination.

Of the 10 sieved samples from each region, three pre-designated samples were set aside for individual chemical analysis.<sup>2</sup> The remaining seven samples in each case were separately homogenized and then combined in equal quantities (equal weights) to form a single representative composite or pooled sample for each of the 10 regions.

All individual and pooled samples were immediately repackaged into two layers of solvent-rinsed aluminium foil, enclosed in lightweight polyethylene bags and sealed in brown paper envelopes in order to minimise the potential for cross-contamination or contamination from other materials in the laboratory. All samples were then cooled at 4°C pending analysis.

All 29 individual samples (see footnote 2) were subsequently forwarded to the laboratories of LGC (Teddington) Ltd (UK) for quantitative analysis of a range of phthalate esters and alkylphenol compounds. LGC also performed a qualitative analysis on each of the samples in order to identify (as far as possible) any other organic contaminants present in significant quantities.

Each of the 10 pooled samples was divided in two at the Greenpeace Laboratories, with one half of each then being forwarded to the laboratories of GALAB (Geestacht, Germany) for quantitative determination of a range of organotin compounds. The remaining halves of each of the 10 pooled samples were then sent to the laboratories of the Netherlands Institute for Fisheries Research (RIVO, IJmuiden, Netherlands) for quantitative analysis of a range of brominated flame retardants and short-chain chlorinated paraffins.

Region	Region code
Scotland	SC
North East	NE
North West	NW
East Midlands	EM
West Midlands	WM
East Anglia	EA
Wales	WL
London	LD
South East	SE
South West	SW

Table 1: regions represented by UK dust samples (10 samples per region)

<sup>1</sup> analytical grade pentane

<sup>2</sup> Only two samples in the case of the East Midlands as one of the three designated samples contained too little material for further processing and analysis

## Non-UK samples

### Sample collection

In addition to the 100 UK samples, a smaller number of dust samples were collected from other countries in Europe for purposes of comparison:-

In each case, the samples were full or partially filled dust filter bags donated by individual volunteers rather than purposefully collected samples. They therefore represented more integrated samples of dust collected from the specific addresses over time. The dusts were not specifically collected for scientific analysis but had simply been collected in the filter bags during routine cleaning. Furthermore, different makes and models of vacuum cleaner were used in each case. These factors must be taken into account when considering the analytical results for these samples.

All dust filter bags were sealed and packaged as for the UK samples and were transported to the Greenpeace Research Laboratories for processing.

### Sample processing

As with the UK samples, all non-UK samples were sieved through solvent-rinsed 2mm gauge sieves on to pre-cleaned aluminium foil and separately homogenized.

The single samples from France and Spain were then divided in two, with one half forwarded to the LGC laboratories (for phthalate, alkylphenol and qualitative screen analyses) and the other half forwarded to GALAB (for organotin analysis).

Of the samples from the Nordic countries, two samples from Finland, two from Denmark and the two samples from Sweden were split in two and forwarded to LGC and GALAB as above. The remaining single samples from Finland and Denmark were split into three equal portions, with one portion sent to LGC, a second to GALAB and the third to RIVO (for analysis of brominated flame retardants and short-chain chlorinated paraffins).

### Sample analysis

Brief descriptions of the analytical methods employed are given below. More detailed descriptions are included in Annex 3.

#### Alkylphenol compounds and phthalate esters (LGC)

Approximately 10g of each dust sample were extracted in hot dichloromethane for 2½ hours, the extract concentrated up to 50ml and stored at 4°C until analysis. A blank sample (acid-washed sand) was extracted along with each batch of 10 samples to check for laboratory contamination. Standard solutions of the target compounds were analyzed alongside the samples in order to calibrate the instruments. A deuterated internal standard (i.e. labeled with deuterium, the non-radioactive isotope of hydrogen) was added to each sample prior to extraction to allow estimation of recovery (extraction efficiency) of the target compounds.

Extracts were analysed by gas chromatography/mass spectrometry (GC-MS) with the following specific target compounds being quantified:-

- Phthalate esters - di-methylphthalate (DMP), di-ethylphthalate (DEP), di-n-propylphthalate (DPP), di-isobutylphthalate (DiBP), di-n-butylphthalate (DnBP), Butylbenzylphthalate (BBP), di-2-ethylhexylphthalate (DEHP), di-isononylphthalate (DiNP) and di-isodecylphthalate (DiDP).
- Alkylphenol compounds - 4-n-octylphenol (4OP), 4-nonylphenol (4NP) and 4-(1,1,3,3-tert-methylbutyl)phenol (4TMBP).

Limits of detection varied from compound to compound and depended on sample size. For purposes of reporting quantitative data, however, limits of quantification were 0.1 ppm in each case.

#### Qualitative screen for other organic contaminants (LGC)

In addition to the quantitation of phthalates and alkylphenols described above, these same extracts were further subjected to a qualitative GC-MS screen analysis (in accordance with BS6920). The supplementary procedure was performed in order to identify any other organic contaminants present in the dust in significant quantities (i.e. yielding an instrument response significantly above background). These additional, non-target compounds have been identified, where possible, using a combination of computer library search matching and expert interpretation of mass spectra. All identities must therefore be considered tentative (i.e. not 100% certain) as they have not been confirmed against standard solutions for each of the additional compounds identified; indeed, in many cases such standards are simply not available. Nevertheless, this analysis does yield useful supplementary information regarding other contaminants which may be subject to verification and quantitative analysis in the future.

#### Brominated flame retardants and short-chain chlorinated paraffins (RIVO)

Dust samples were extracted with hot hexane:acetone (3:1) mixture for 12 hours and, following addition of internal standards (PCB 112 and labeled BDE-209), the extract was concentrated on a rotary evaporator, acidified and the organic layer collected. The water layer was extracted two further times with iso-octane before all organic extracts were combined and concentrated in 2 ml of dichloromethane. Each extract was cleaned by gel permeation chromatography (GPC), concentrated under nitrogen, dissolved in iso-octane and further purified by shaking with sulphuric acid. Finally, the extracts were concentrated under nitrogen to 2 ml, eluted through a silica gel column and concentrated to 1 ml for analysis.

Analysis was conducted by GC-MS, using electron capture negative ionisation (ECNI). Concentrations of the following compounds/congeners were determined in each sample:-

- Polybrominated diphenylethers (PBDEs) – tri- (BDE-28), tetra- (BDE-47, 66, 71, 75, 77), penta- (BDE-85, 99, 100, 119), hexa- (BDE-138, 153, 154), hepta- (BDE-190) and deca- (BDE-209).

- Polybrominated biphenyls (PBBs) – di- (BB-15), tetra- (BB-49, 52), penta- (BB-101), hexa- (BB-153, 155) and deca- (BB-209).
- Hexabromocyclododecane (HBCD)
- Tetrabromobisphenol-A (TBBPA) – plus its methyl derivative.

Limits of detection (dry weight basis) were as follows:- PBDEs, 0.12-0.62 ppb (ng/g); PBBs, 0.18-2.8 ppb, HBCD, 2.5-12.8 ppb, methyl-TBBPA, 0.1-0.5 ppb; TBBPA, 0.5-3 ppb. As they are highly complex mixtures, analysis for SCCPs was semi-quantitative only.

#### **Organotin compounds (GALAB)**

All samples were further sieved through a 0.065 mm sieve before extraction using a methanol:hexane mixture and analysis by gas chromatography/atomic emission detection (GC/AED) according to accredited methods. Concentrations of the following compounds were determined in each sample:-

- Butyltins - mono-, di-, tri- and tetrabutyltin (MBT, DBT, TBT and TeBT respectively)
- Octyltins - mono- and di-octyltin (MOT and DOT respectively)
- Tricyclohexyltin (TCHT)
- Triphenyltin (TPT)

Limits of detection for all organotin compounds were 1 ng tin cation/g dry weight of sample (ppb) in each case.

Country	No. samples
Denmark	3
Finland	3
France	1
Spain	1
Sweden	2

Table 2: summary of non-UK dust samples included in the study

# Results and Discussion

## Target compounds

Concentrations of the five target groups of compounds analysed quantitatively in the current study are summarized for the UK samples in Table 3. Along with the frequency with which each compound or group was found (i.e. the number of individual or pooled samples out of the total), Table 3 also gives mean (average) and median (middle) values for concentrations across all regions. These means and medians have been calculated using results from all samples analysed (i.e. 29 individual samples for the phthalates and alkyphenols, 10 pooled samples for the brominated flame retardants, chlorinated paraffins and organotins), taking all values below limits of detection as zero. Ranges of concentrations for each compound or group are also given, in the form of the maximum and minimum (highest and lowest) values recorded for all UK samples.

Detailed results for each of the 10 regions are given in Annex 1A, in each case comparing the regional values with the summary statistics (mean, median, etc.) for all UK samples analysed in this study (i.e. those statistics summarized in Table 3).

## Phthalate esters

Of the 9 individual phthalate esters specifically quantified, 4 (DEP, DiBP, DnBP and DEHP) were found above limits of detection (LOD) in all 29 UK samples; BBP was found in all but one sample. DPP did not appear in any of the 29 individual samples. The isomeric phthalates DiNP and DiDP were found in roughly a third of samples.

DEHP comprised between 24 and 79% of the total concentration of phthalates quantified, and was the most abundant phthalate in the majority of samples. This is as might be expected from its reported common and widespread use in soft PVC (vinyl) products in the home (e.g. flooring, some wall-coverings, shower curtains, furnishings, toys, clothing). Concentrations ranged from 0.5 parts per million (ppm,  $\mu\text{g/g}$  dust) to over 400 ppm (0.4 mg/g) across the 29 individual samples. DEHP is a known developmental toxin, classified in Europe as “toxic to reproduction”, and yet it remains in such high volume use that our exposure to it is continuous and substantial.

Of the other phthalates, DiBP, DnBP and BBP were relatively abundant in most samples. BBP was the most abundant phthalate in two of the samples, and DiBP in two also. Although found in only a fraction of the total sample set, the isomeric phthalates DiNP and DiDP tended to be present in substantial concentrations where they were found (12.7-337 ppm and 4.3-157 ppm respectively). Indeed, DiNP was the most abundant phthalate in two samples. Although it is not possible to deduce specific source products in any one case, these differences in total and relative abundances of phthalates commonly used as PVC additives probably reflect differences in the type and number of PVC products in the rooms sampled in each case.

It is interesting that Allsopp *et al.* (2000) found only DiNP in five samples of new PVC flooring purchased in the UK, suggestive of a market shift away from DEHP for this application in recent years. At the same time, household dusts may be expected to reflect accumulation from a wider range of product sources than flooring alone, and might also be influenced by relatively old PVC products with a different balance of phthalate plasticizers. Another phthalate, DEP, was a common component of all 29 samples, probably resulting from its widespread use in perfumes, cosmetics and other personal care products, although (with a few notable exceptions) it was generally present at lower concentrations than other phthalates (0.6-115 ppm). Very recent research suggests that this phthalate may be capable of interfering with sperm development in humans (Duty *et al.* 2003).

Despite the propensity of phthalate-plasticised products in the common home environment, few other published data regarding levels in household dusts are available. Certainly, however, median concentrations found in the current study for BBP (24.5 ppm), DiBP (43.2 ppm), DnBP (52.8 ppm) and DEHP (195 ppm), are of a similar order to those summarised from the handful of studies which do exist by Butte and Heinzow (2002). Total phthalate concentrations determined for the 29 UK samples in the current study (i.e. with a maximum of over 1000 ppm, i.e. more than 1mg/g of dust) are also in a similar range to that reported by Rudel *et al.* (2001), though perhaps slightly lower than other values reported for households in Germany (Butte and Heinzow 2002). Clearly, however, phthalates are abundant contaminants common to the indoor household environment.

Further information on the common uses and hazards of a range of common phthalates is provided in Annex 2.

## Alkyphenols

Of the three alkyphenol compounds quantified, nonylphenol (actually a mixture of 7 related isomers) was by far the most commonly found and most abundant, suggesting that this substance still has quite widespread use and/or occurrence in household products. Nonylphenol (4OP) was found above limits of detection in 22 of the 29 samples at concentrations ranging from 0.4 to 36 ppm. Mean and median values (calculated including zero values for non-detects) were similar at 10.9 and 9.8 ppm respectively. In turn, these values are of the same order as those for technical nonylphenol reported by Butte and Heinzow (2002), and for nonylphenol and its ethoxylates by Rudel *et al.* (2001). Nonylphenol is widely recognised as a hormone (endocrine) disruptor, particularly due to its estrogenic properties, and is also suspected of exerting direct effects on sperm function in mammals (e.g. Adeoya-Osiguwa *et al.* 2003).

4-tert-methylbutylphenol (4TMBP) was found in only four of the 29 samples (0.1-2.4 ppm), and 4-n-octylphenol (4OP) in only one (8.6 ppm). The uses of these substances are clearly not as widespread as those of nonylphenol, perhaps limited to a small array of more specialist products.

Further information on the uses and properties of the most common alkylphenols (especially nonylphenol) is provided in Annex 2.

### **Organotin compounds**

Of the eight organotin compounds monitored in the current study, five were found in all pooled regional samples (MBT, DBT, TBT, MOT and DOT). TPT was found in only one pooled sample (that for Scotland), whereas neither TeBT nor TCHT were found in any of the samples at above limits of detection.

Although there was some variation from sample to sample, the pattern of relative abundance of the different organotin compounds was relatively consistent, with MBT the most abundant (0.81–2.8 ppm), followed by DBT and MOT (0.157–1.3 ppm and 0.083–1.3 ppm respectively). This mirrors the pattern we reported previously in dusts from Parliament buildings across Europe (Santillo *et al.* 2001) and may reflect the relative frequencies with which these substances are present as stabilizer additives in plastics, especially PVC, in the home. In contrast, however, Allsopp *et al.* (2000) reported consistently higher levels of DBT than MBT in five samples of new PVC flooring purchased in the UK (37.7–569 ppm and 0.33–48.8 ppm respectively). While it is not known whether this is reflective of the PVC market in general, it does raise the possibility that the predominance of MBT in the dust samples may result in part from the partial degradation of DBT, or perhaps simply from the greater mobility of MBT compared to DBT. Nevertheless, DBT, toxic to the developing immune and nervous systems in mammals (Kergosien and Rice 1998), was present at significant levels in all samples.

TBT was surprisingly abundant in all pooled dust samples, ranging from 0.02 to 0.76 ppm. Although TBT can arise as a contaminant in formulations of DBT and other organotin compounds, it is probable that the use of TBT, as a fungicide or treatment against dust-mites in carpets, textiles and PVC also contributed to the levels found. Allsopp *et al.* (2000) reported levels of TBT in five new PVC flooring samples in the range 0.13–17.9 ppm. Although most of the eight new carpet samples tested by Allsopp *et al.* (2001) contained only low ppb concentrations of TBT, two products contained TBT levels well in excess of the concentrations of other organotins (2.7 and 47.5 ppm TBT), indicative of deliberate treatment of the carpet fibres with this chemical. Although most notorious because of its effects on sexual development in marine snails, TBT is also reported to be toxic to the immune system in mammals (Belfroid *et al.* 2000).

The presence of TPT in one pooled sample was unexpected, given that there are no known domestic uses of this hazardous chemical (which has been most commonly used as an agricultural fungicide, especially on potato crops). It is possible that its presence in the pooled sample for Scotland resulted from its presence in just one of the seven individual samples combined to prepare the pooled sample, perhaps in turn a consequence of its local application to farmland. This clearly requires further investigation to elucidate.

Total organotin concentrations ranged from 1.58 to 5.05 ppm, slightly higher than the range we reported previously for the Parliament dust samples (0.49–3.48 ppm, Santillo *et al.* 2001). TBT concentrations in particular were noticeably higher in the housedust samples than in Parliament dusts, perhaps a reflection of its more widespread use in PVC flooring, carpets and other products more closely associated with private households.

## **Brominated flame retardants**

### **Decabromodiphenyl ether (BDE-209)**

This brominated flame retardant was found in all ten regional pooled samples at between 3.8 and 19.9 ppm. This is considerably higher than those concentrations recorded in Parliament dusts in 2001 (0.29–6.9 ppm, Santillo *et al.* 2001). BDE-209 is most commonly used as an additive flame retardant in a range of plastics and textiles, especially in high impact polystyrene (HIPS), in electrical components and in styrene rubbers used in carpet backing or in furniture (Lassen *et al.* 1999). Sjödin *et al.* (2001) reported it to be among the most predominant brominated flame retardants found associated with airborne particles inside an electronics recycling plant in Sweden, and even to be detectable in air from normal computerized office environments.

The presence of substantial ppm concentrations of BDE-209 in housedust suggest that exposure to such dusts, through inhalation, ingestion or direct skin contact, may represent a significant additional route of human exposure to this chemical in the home. This may be particularly important given that exposure to BDE-209 through other common routes, especially through food, is generally considered much less significant than for other, more bioaccumulative, lower-brominated BDE congeners (e.g. tetra- and penta-BDE). Jakobsson *et al.* (2002) reported that BDE-209 was detectable alongside other PBDEs at higher levels in the blood of computer technicians than in other workers, presumably arising from direct exposure in the workplace. Half-lives for BDE-209 in humans are thought to be relatively short compared to other PBDEs. Nevertheless, these data suggest that our exposure to this highly persistent chemical may be continuously “topped up” through its presence in air and dusts in the indoor environment. Although having relatively low acute (short-term exposure) toxicity, prenatal exposure to BDE-209 has been shown to effect bone development in rats (Olsson *et al.* 1998), through mechanisms with possible relevance to humans.

### **Other brominated diphenyl ethers**

Although BDE-209 was by far the most abundant PBDE, other lower brominated congeners were also present in all samples, albeit generally at low to mid part per billion levels. The more bioaccumulative tetra- and penta-BDEs (represented in Table 3 by BDE-47 and BDE-99 respectively) were present in all 10 pooled samples from the UK. Excluding the single sample from Scotland (see below), concentrations ranged from 10 to 76 ppb and 18 to 370 ppb for BDE-47 and BDE-99 respectively, similar to levels found in Parliament dusts from around Europe in 2001 (Leonards *et al.* 2001).

The pooled sample from Scotland was unusually heavily contaminated with tetra- and penta-, and even hexa-, BDE congeners, with concentrations of BDE-47 and BDE-99 of 1980 and 2100 ppb (1.98 and 2.1 ppm) respectively. This is indicative of the presence in one or more of the households included in preparing the Scottish pooled sample of products flame retarded with commercial mixes of penta- or perhaps octa-, BDE. Penta-BDE has been most widely used in epoxy resins, textiles, polyesters and polyurethanes while octa-BDE is primarily used in ABS (e.g. in computer housings).

It is these BDEs (especially penta-BDE), which were found to be increasing in concentration in breast milk in Europe and the US over the past few decades (Meironyte *et al.* 1999, Darnerud *et al.* 2001). Although it is commonly assumed that food represents the most significant intake route for these bioaccumulative BDEs, their presence at significant (and sometimes substantial) levels in housedust suggests that more direct exposure routes might also be important in the home. Exposure to penta-BDE in the womb has been found to have permanent effects on brain development in rats (Eriksson *et al.* 1999). Moreover, metabolites of this and other lower brominated BDEs may be even more toxic than the parent compounds themselves (de Boer *et al.* 2000).

Under new EU legislation, neither penta- nor octa-BDE will be permitted for continued use within Europe although, as these data indicate, their presence in and loss from older products in the home will remain a problem for some time to come. Restrictions on deca-BDE (BDE-209) are less certain, despite its known toxicity and ability to degrade to lower-brominated BDE congeners once released into the environment.

#### **Hexabromocyclododecane (HBCD)**

Like BDE-209, HBCD was a prominent component of all UK dust samples, with concentrations ranging from 0.94 to 6.9 ppm (mean and median 3.2 ppm). These data appear to confirm the widespread use of this brominated flame retardant in products likely to be found in the home. HBCD is reportedly particularly widely used in textiles and expanded polystyrene products. Just as for BDE-209, HBCD is a highly persistent chemical, but unlike BDE-209, is also highly bioaccumulative. Once again, direct exposure in the home may represent a significant additional exposure route for humans. In common with some lower-brominated PBDEs, HBCD is capable of interfering with genetic material in human cell lines (Helleday *et al.* 1999), a possible indicator of carcinogenic (cancer-causing) potential.

Levels recorded in the UK samples were slightly higher than those recorded in the Parliament dusts in 2001 (<0.0025 to 3.7 ppm). The highest level recorded in that study was for one of two dusts from the UK Parliament, and the consistently high levels in the current survey of UK housedusts may indicate its particularly widespread use in the UK (though data remains very limited for other countries).

#### **Tetrabromobisphenol-A (TBBP-A)**

TBBP-A was found in 4 of the 10 pooled samples, at

concentrations between 0.19 and 0.34 ppm, substantially higher than in the Parliament dusts (found in 7 of 16 samples at between 0.005 and 0.047 ppm). Although less abundant than other brominated flame retardants like BDE-209 and HBCD, its presence in dust confirms that it is released from products in the home during use and/or normal wear and tear. TBBP-A is widely used in printed circuit boards, motor housings and other electrical and electronic components, as well as more generally in plastics and resins.

This is particularly interesting as it is generally thought that TBBP-A in its most commonly used reactive form (especially in printed circuit boards) is very tightly bound to the plastics or resins in which it is used and, therefore, unlikely to be lost to the environment. Its presence in dusts could result from its less frequent use as a simple additive flame retardant (especially in ABS, polystyrene and PET), though it is not possible to speculate further on sources on the basis of existing data. What is clear, however, is that even for this chemical, exposure in the home through contact with dusts could be a significant and, as yet, underestimated exposure route.

TBBP-A is reported as a common and relatively abundant contaminant in the office environment (Bergman *et al.* 1997, Sjodin *et al.* 2001) and is also detectable in the blood of computer technicians (Jakobsson *et al.* 2002). In common with the PBDEs, the acute toxicity of TBBP-A is thought to be low, though it does exhibit toxic effects in mammals following longer-term exposures. It is particularly noted for its ability to interfere with the binding of thyroid hormones, responsible for many aspects of growth and development in mammals (Meerts *et al.* 1998).

#### **Short-chain chlorinated paraffins (SCCPs)**

Short-chain chlorinated paraffins were a prominent component in the majority of dusts analysed in the current study, appearing in 8 of the 10 UK pooled samples at concentrations ranging from approximately 1.9 to 13.0 ppm. Although these data are semi-quantitative, as SCCPs remain particularly difficult to analyse in a quantitative manner, they confirm the ubiquitous presence of SCCPs in the home environment. This presence presumably arises from their ongoing and/or previous widespread use as additives in plastics (especially PVC cables), rubbers, paints etc.

Recent EU legislation has banned the use of SCCPs in metal working and leather processing applications, which were deemed to cause significant environmental releases and exposures, but did not address uses as flame-retardants or other additives in consumer products. Although from a limited number of samples, these data on presence in housedust confirm the importance of measures to address these other uses as they clearly present the potential for direct and continuous exposure in the home. Although information on the consequences of long-term exposure in mammals remains very limited, SCCPs are recognised as "Category 3" carcinogens in Europe, presenting "possible risks of irreversible effects".

### **Regional trends in concentrations of target compounds**

In addition to presenting detailed results for individual and pooled samples for each region, Annex 1 also includes ranked tables for a number of the target compounds, listing the samples in order of decreasing concentration (Annex 1B). It is clear from these tables that no consistent and reliable regional patterns can be discerned, as may be expected from the small number of representative samples in each case. For total phthalates, the two samples from the East Midlands were the most contaminated, while those from London tended to be the least. For organotin compounds, a decreasing trend from north to south is apparent by eye. However, neither of these apparent trends has any strong statistical basis.

More than many other types of sample, housedusts may be expected to be influenced greatly by the very specific circumstance in each of the homes sampled, i.e. the types of products present, size of rooms, how (and how frequently) the rooms are cleaned, etc. In other words, variation in contaminant levels from home to home may be reasonably expected to be substantially greater than any underlying regional trend. So, while the sample set provide a reasonable statistical basis for the analysis of ranges and averages over the whole of the UK, they cannot be expected to give detailed information on regional variations, if such variations indeed exist.

### **Other organic compounds (non-target compounds)**

In addition to the quantitative analyses for the five target compound groups, the non-target screening analysis of the 29 individual samples revealed the presence of a diversity of other organic compounds in house dust. In total, more than 140 other chemicals were detected, of which 127 could be at least tentatively identified. Only one sample, one of three from London, contained no other identifiable organic compounds (and this sample was also among the least contaminated with the target compounds). The remaining 28 contained between 7 and 27 additional chemicals. The key chemicals, based on the frequency with which they were found and/or on their environmental/toxicological significance, are summarized in Table 4.

A total of seven different pesticide residues were found in at least one sample, plus one pesticide synergist (a compound which enhances the activity of a pesticide). All those found were insecticides. The most abundant group were the synthetic pyrethroids, especially permethrin (7 samples), which are still available for non-professional pest-control use in the home. The synergist piperonyl butoxide, found in two samples, is also commonly associated with pyrethroid insecticides and has been used as an indicator of pyrethroid exposure in the home (Whyatt *et al.* 2002).

Although all pyrethroids in house dust may arise from deliberate application for pest control by the householder, the frequency with which permethrin was found in the current study (almost a quarter of all individual samples), suggests that its presence may be more closely related to its inclusion

as a treatment against dust mites in certain brands of carpet. For example, Allsopp *et al.* (2001) reported finding permethrin in six out of eight new carpet samples purchased in the UK.

Concerns have existed for many years regarding the damage that permethrin exposure can cause to the immune system and nervous system in mammals, with possible relevance for humans (Institoris *et al.* 1999, Punareewattana *et al.* 2001, Prater *et al.* 2003). Chen *et al.* (2002) highlight the ability of certain pyrethroids (including permethrin), to mimic estrogen hormones, an effect which may be even greater for compounds formed as the pesticides themselves start to degrade (Tyler *et al.* 2000), although the significance of these findings to whole organisms has recently been challenged by industry (Kunimatsu *et al.* 2002). In addition, some pyrethroids, especially in combination with piperonyl butoxide, can induce allergic responses in sensitive individuals (Diel *et al.* 1999).

The ability of permethrin to adhere to surfaces and dusts in the home and, thereby, to lead to significant inhalation and ingestion exposure, has been recognized for some time (e.g. IEH 1999). At the same time, studies have indicated that permethrin impregnation of carpets may be entirely ineffective in controlling dust mite populations (Brown 1996), the very reason for which it is included.

The carbamate insecticide bendiocarb and the organochlorine lindane (each found in one sample) may also be present as a result of deliberate use in the home (e.g. in proprietary home pest control products). In contrast, the organochlorine pesticide DDT (and its breakdown product DDE), again found in one home, more probably reflect contamination of dust from the wider environment since DDT has not been permitted for use in the UK for many years. If there are particularly contaminated sites surrounding this sample location, DDT contamination could have resulted from the settling of wind-blown dust in the home or its carriage into the home on shoes or clothing.



Compound	Found in...	UK mean (average) value	UK median (middle) value	UK minimum (lowest) value	UK maximum (highest) value
<b>Phthalate esters</b>		<b>µg/g dust (parts per million, ppm)</b>			
DMP	11/29	0.12	nd	nd	1.1
DEP	29/29	12.2	3.5	0.6	114.8
DPP	0/29	-	-	-	-
DiBP	29/29	52	43.2	0.2	157.4
DnBP	29/29	50.2	52.8	0.1	106.4
BBP	28/29	56.5	24.5	nd	238.9
DEHP	29/29	191.5	195.4	0.5	416.4
DiNP	11/29	48.5	nd	nd	337.2
DiDP	11/29	20.8	nd	nd	156.6
<b>Total phthalates</b>	<b>-</b>	<b>431.7</b>	<b>354.3</b>	<b>1.6</b>	<b>1019.1</b>
<b>Alkylphenol compounds</b>		<b>µg/g dust (parts per million, ppm)</b>			
4TMBP	4/29	0.12*	nd	nd	2.4
4OP	1/29	0.3*	nd	nd	8.6
4NP	22/29	10.5	9.8	nd	35.2
<b>Total alkylphenols</b>	<b>-</b>	<b>10.9</b>	<b>9.8</b>	<b>nd</b>	<b>36.1</b>
<b>Brominated flame retardants</b>		<b>ng/g dust (parts per billion, ppb)</b>			
BDE-28 (tri-)	7/10	4.14	0.35	<0.1	33
BDE-47 (tetra-)	10/10	222.8	24.8	10	1980
BDE-99 (penta-)	10/10	286.5	44	18	2100
BDE-153 (hexa-)	9/10	33.8	23	<0.1	170
BDE-183 (hepta-)	7/10	19.2	9.5	<0.1	87
BDE-209 (deca-)	10/10	9820	7100	3800	19900
HBCD	10/10	3158	3250	940	6900
TBBP-A	4/10	116*	<10	<10	340
<b>Organotin compounds</b>		<b>ng/g dust (parts per billion, ppb)</b>			
MBT	10/10	1375	1350	810	2800
DBT	10/10	563	519	157	1300
TBT	10/10	144.5	49.9	21.6	759
TeBT	0/10	-	-	-	-
MOT	10/10	450.6	349	82.5	1300
DOT	10/10	129.2	62.7	17.6	545
TCHT	0/10	-	-	-	-
TPT	1/10	6.9*	<1	<1	68.9
<b>Total</b>	<b>-</b>	<b>2669</b>	<b>2432</b>	<b>1581</b>	<b>5047</b>
<b>Short-chain chlorinated paraffins</b>		<b>µg/g dust (parts per million, ppm)</b>			
<b>Total</b>	<b>8/10</b>	<b>4.34</b>	<b>3.7</b>	<b>&lt;0.12</b>	<b>13.0</b>

Table 3: summary of analytical results for key chemicals in the five target compound groups for the UK dust samples

\*as these compounds were found in a small number of samples only, the mean values cannot be considered representative

#### Abbreviations

- *Phthalate esters*: DMP - di-methylphthalate, DEP - di-ethylphthalate, DPP - di-propylphthalate, DiBP - di-isobutylphthalate, DnBP - di-n-butylphthalate, BBP - butylbenzylphthalate, DEHP - di-2-ethylhexylphthalate, DiNP - di-isononylphthalate, DiDP - di-isodecylphthalate.
- *Alkylphenol compounds*: 4TMBP - 4-(1,1,3,3-tert-methylbutyl)phenol, 4OP - 4-n-octylphenol, 4NP - 4-nonylphenol.
- *Brominated Flame Retardants*: BDE - brominated diphenylethers (tribromo- to decabromo-), HBCD - hexabromocyclododecane, TBBP-A - tetrabromobisphenol-A.
- *Organotin compounds*: MBT - monobutyltin, DBT - dibutyltin, TBT - tributyltin, TeBT - tetrabutyltin, MOT - monoocetyl tin, DOT - dioctyltin, TCHT - tricyclohexyltin, TPT - triphenyltin.

Compound	Notes	Frequency
<b>Insecticides</b>		
Bendiocarb	Carbamate	1
Cypermethrin	Pyrethroid	1
Lindane	Organochlorine	1
p,p'-DDE	Organochlorine	1
p,p'-DDT	Organochlorine	1
Permethrin	Pyrethroid	7
Piperonyl butoxide	Pesticide synergist, esp. in pyrethroids	2
Tetramethrin	Pyrethroid	1
<b>Plasticisers</b>		
Bis-(2-ethylhexyl) adipate		1
Di-(2-ethylhexyl)-isophthalate		1
2-ethylhexyl-dibenzylphosphonate	Also used as flame retardant	7
Tri-[2-Butoxyethanol]phosphonate	Also used as flame retardant	7
Tris(2-Ethylhexyl)trimellitate		3
<b>Plastic/resin components</b>		
Bisphenol A		1
Butyl methacrylate		1
Ethyl methacrylate		2
Methyl methacrylate	Perspex and other resins	7
Nonanoic acid	Component of some laquers/plastics	1
Phthalic anhydride	Curing agent for some resins	3
Styrene	Monomer of polystyrene/resins	24
Toluene-2,4-diisocyanate	Polyurethane foams and varnishes	1
<b>Flame retardants</b>		
Triphenylphosphate	Also used as varnish plasticiser	1
<b>Fragrance chemicals</b>		
1,8-Cineole (Eucalyptol)	Mainly natural extracts or their synthetic counterparts, possibly from perfumes, detergents, essential oils, etc.	2
Acetophenone		2
Limonene		6
Linalool		2
Methyl dihydrojasmonate		1
p-Cymene		2
α-Hexylcinnamaldehyde		1
α-Pinene		6
β-Phellandrene		1
α-Terpineol		1
γ-Terpinene		1
Totarol		5
Vitamin E/ Vitamin E acetate		1

Table 4: summary of other key compounds found in the 29 individually analysed UK dust samples, with an indication of the number of samples in which they were found.

Compound	Notes	Frequency
<b>Other additives</b>		
Benzaldehyde	Solvent in perfumes/flavour additive	3
Benzyl salicylate	Fixing agent in perfumes and sunscreens	2
Butylated hydroxy toluene	Antioxidant used in foods	1
Cyclohexane	Solvent	3
Dodecan-1-ol	Possible component of detergents	6
Heptanal	Flavour additive	2
Hexanal	Flavour additive	5
N,N,N',N'-Tetraacetylenediamine	EDTA - chelating agent in detergents, etc.	9
N,N-Tetradecanamine	Possible component of detergents	3
Nonanal	Flavour additive	25
Octanal	Flavour additive	1
Pentaethylene glycol	Possible component of surfactants	2
Polyethylene glycol	PEG – cosmetics and toiletries	1
<i>p</i> -Toluenesulfonamide	Preservative in some paints	2
Tributyl acetyl citrate	Plasticiser/food additive/adhesives	1
Triethylene glycol	Plasticiser/solvent	1
Triphenylphosphonate	Unknown – possibly flame retardant?	1
Tris(2-chloroethyl) phosphonate	Unknown – possibly flame retardant?	1
Tris(3-chloropropyl) phosphonate	Unknown – possibly flame retardant?	14
<b>Polycyclic aromatic hydrocarbons (PAHs)</b>		
Benzo(b)fluoranthrene		2
Chrysene	Most commonly formed as products of incomplete combustion. May result from general environmental contamination (outdoor sources) or from open heating systems (indoor sources)	1
Fluoranthrene		2
Perylene		1
Phenanthrene		3
Pyrene		1
<b>Other environmental contaminants</b>		
Benzene	Petrol	1
Butan-2-one	Petrol additive (methyl ethyl ketone)	4
<b>Natural fats/oils</b>		
Cholest-4-en-3-one	Most probably from human and/or animal skin and/or hair	4
Cholesta-3,5-dien-7-one		2
Cholesta-3,5-diene		5
Cholesta-4,6-dien-3-ol		8
Cholesterol		20
Sesquiterpene		2
Squalane		1
Stigmast-4-en-3-one		1

Table 4 (continued)

Among the man-made chemicals most commonly found in the housedusts in the current study were non-phthalate plasticizers (phosphonates and trimellitates, found in 7 and 3 samples respectively), the irritant resin monomer methyl methacrylate (7 samples) and the resin curing agent phthalic anhydride (in 3 samples). Styrene, the chemical building block of polystyrene, was found in 24 samples, one of the most widely found contaminants. Its widespread presence may result from the presence of small quantities of unpolymerised (unreacted) styrene which are generally found in polystyrene, small fragments of which may have been present in the sieved dust samples. However, contributions from other uses, such as certain styrene-containing resins, cannot be ruled out.

A wide array of other chemicals used as solvents, fixatives, flavour additives and components (or breakdown products) of detergents were also found (see Table 4), as were numerous chemicals used as fragrance additives, though also occurring as natural components in many essential oils and plant extracts. Together these results indicate the wide range of chemical constituents in house dust which are likely to arise from everyday presence and use of consumer products in the home. While it is known that some of these are hazardous when encountered alone, the possible effects of combined and continuous exposure to such complex mixtures of natural and man-made chemicals are simply not known.

In addition to those compounds identified above, a number of chemicals tentatively identified as alkyl or chloroalkyl phosphonates were also found. One of these compounds, identified by the organic screen as tris(3-chloropropyl) phosphonate, was found in almost half of all the individual samples analysed. Although it is possible that these residues arise from some common, but poorly documented use of phosphonates in the home environment, it seems more likely that these are actually residues of the closely related tris(2-chloroethyl) phosphate (TCEP) and tris(3-chloropropyl) phosphate (TCPP), used widely as flame retardants in plastics (especially polyurethanes and some polyesters). Indeed, TCEP has previously been reported as a common contaminant of indoor air and dust (Butte and Heinzow 2002). Both TCEP and TCPP were detectable at substantial levels in air particulates from an electronics recycling plant in Sweden (Sjödin *et al.* 2001), though even in the normal office environment they may be present at significant levels (Bergman *et al.* 1997). If these are indeed the compounds identified in the current study, they represent a long-recognised and persistent hazard to humans and the environment (Huse 1995, Lassen *et al.* 1999).

Aside from those chemicals likely to have arisen from the presence and/or use of products or preparations in the home, a number of other important chemical groups were represented. The most commonly found chemicals were cholesterol and its derivatives, almost certainly arising from particles of human skin and hair (or those of pets), which form a substantial component of dust in all inhabited indoor environments. Also prominent, however, were a group of chemicals called polycyclic aromatic hydrocarbons (PAHs),

widespread environmental contaminants which arise in the urban environment primarily as a result of the incomplete combustion of fuels (i.e. from traffic, heating systems, etc.). In the home environment, these may be present through a combination of the settling of windblown dust from outside sources and from open heating systems in the home (e.g. open fires) where these are present. Finally, four samples were found to contain the common petrol additive methyl ethyl ketone (MEK), undoubtedly transported in to the home from outside sources on wind-blown dust or contaminated shoes or clothing.

## Non-UK samples

Analytical results for the 10 non-UK samples are presented in Annex 1C. All 10 samples were subjected to individual analysis for phthalate esters, alkylphenol compounds, organotin compounds and a non-target GC-MS screen. Only two of the 10 non-UK samples were subjected to analysis of brominated flame retardants and short-chain chlorinated paraffins, samples HD02105 from Finland and HD02110 from Denmark.

Although there are many similarities with results for the UK samples, there are also some notable differences. Of course, given the small sample sizes, it is not possible to state that these are in any way representative of dust contamination levels in households in these countries more generally.

### Phthalate esters

Patterns of phthalate distribution varied from sample to sample. DEHP was the most abundant phthalate found in 9 out of 10 samples (with DiNP predominating in the single sample from France), at concentrations ranging from 45.5 to 579 ppm. This higher figure, found in one of three samples from Finland (HD02107), was the highest concentration recorded for DEHP in any of the samples (compared to UK maximum of 416 ppm, see Table 3). A second sample from Finland (HD02105) contained the highest recorded level of DEP (136 ppm), commonly used in perfumes and cosmetics. As a result, two of the three samples from Finland contained total phthalate levels (907 and 765 ppm) close to the maximum recorded for all UK samples (1019 ppm).

In contrast, the three samples from Denmark were among the least contaminated with phthalates (155–291 ppm). None of these three samples contained residues of the isomeric phthalates DiNP or DiDP, whereas DiNP was a common characteristic of most other non-UK samples. Total phthalate levels in the two samples from Sweden and in the single samples from France and Spain were more or less intermediate (from 411 to 641 ppm).

### Alkylphenol compounds

Of the three alkylphenol compounds specifically analysed for in the current study, neither 4-tert-methylbutyl phenol (4TMBP) nor 4-octylphenol (4OP) nonylphenol were found in any of the non-UK samples. 4-nonylphenol (4NP) was found in 6 out of the 10 non-UK samples, at concentrations from 3.3 to 13.1 ppm, around the average levels recorded for the 29 individual UK samples.

### **Organotin compounds**

As for the phthalate esters, the pattern of organotin compound contamination across the non-UK samples bore some resemblance to the UK samples, specifically with MBT predominating, followed by DBT, MOT and DOT. TBT was also present at significant levels in all non-UK samples, while TPT (triphenyl tin) was found in one of three samples from Finland (HD02107) and one of three from Denmark (HD02109). Sample HD02107 from Finland contained the highest recorded levels of the PVC stabiliser additive DOT (3.6 ppm, giving a total of 5.8 ppm for all organotins), coinciding also with the highest recorded levels of DEHP in the current study.

Organotin concentrations in the three Danish samples were comparatively low (0.139 to 0.894 ppm), below the range recorded for UK samples (1.58 to 5.05 ppm). The appearance of TPT in sample HD02109 is interesting and deserves further investigation to elucidate potential sources.

Organotin concentrations in samples from Sweden, France and Spain were at the lower end of the range recorded for the UK samples (1.169 to 1.59 ppm).

### **Brominated flame retardants**

#### ***Decabromodiphenyl ether (BDE-209)***

One of the most striking differences between the UK and non-UK samples (a single sample each from Finland and Denmark) was in relative concentrations of the flame retardant BDE-209. Levels in the dust from Finland (0.1 ppm) and Denmark (0.26 ppm) were between 10 and 100 times lower than those recorded for the 10 regional pooled samples from the UK (range 3.8 to 19.9 ppm). Moreover, the levels in these two individual non-UK samples were lower than previously recorded in Parliament dusts from the same two countries (Finland 1.1 ppm, Denmark 0.33 and 0.47 ppm, Leonards et al. 2001). Of course, it must be stressed that these latest results cannot be considered representative of BDE-209 concentrations in dusts from Finnish and Danish households more generally. Nevertheless, the scale of the differences between these and the UK samples included in the current study deserve further investigation as they may well reflect existing regional differences within Europe regarding the extent of use of this particular brominated diphenyl ether.

#### ***Other brominated flame retardants***

Concentrations of other PBDEs in the single dust samples from Finland and Denmark were at the lower end of the range for the UK dusts and generally lower than those data reported previously for the Finnish and Danish Parliament building samples (Leonards et al. 2001). HBCD was present in both non-UK samples in the current investigation, again at levels (0.79 and 1.00 ppm) at the lower end of the UK range. TBBP-A was found in both non-UK samples, with the level of 0.4 ppm in the Danish samples being the highest recorded in this study. Of course these data cannot be considered representative of these countries, from which only single samples were analysed, though they do add to the overall data set with regard to brominated chemicals in the home environment.

### **Short-chain chlorinated paraffins (SCCPs)**

As for the UK samples, both the Finnish and Danish dusts contained substantial levels of SCCPs (9.6 and 5.1 ppm respectively), indicative of their widespread presence in the home as a result of their continued use and presence in household products.

### **Other organic compounds (non-target compounds)**

Complete listings of other (non-target) organic compounds identified in the non-UK dust samples are also provided in Annex 1C. Individual samples contained between 5 and 15 additional compounds which were tentatively identified, the majority similar to those reported for the UK samples (Annex 1A and summarised in Table 4 above). Again the polystyrene building block styrene, the flavour additive nonanal and the chelating agent EDTA were prominent in many samples. Plasticisers, solvents, non-brominated flame retardants and other additives were also common contaminants, highlighting again the diverse array of chemicals to which we are consequently exposed in the home.

# Conclusions

The results of this study demonstrate the widespread contamination of household dusts with a variety of hazardous chemicals, including brominated flame retardants, organotin compounds, phthalates, alkylphenols and short chain chlorinated paraffins. This provides further evidence that our exposure to these and other hazardous chemicals is continuous and ubiquitous, even in the home environment.

Although we cannot use these data to identify from which specific products these chemicals arise, they undoubtedly enter the dusts as a result of losses from a wide variety of furnishings and other household goods present in the rooms from which the samples were collected. Such losses may occur through volatilisation or leaching to air, followed by adsorption to dust particles or more directly attached to fine particles lost through abrasion during normal wear and tear. Irrespective of the mechanism, however, these data provide strong and direct evidence that the ongoing use of hazardous chemicals in consumer products is leading to ubiquitous and complex contamination of the home environment.

Patterns of contamination in any one household, or even in regional samples pooled from several homes, will depend greatly on the types of products present in those homes sampled. Clearly these results cannot be taken as fully representative of dust contamination levels in the 10 regions sampled. Nevertheless, these data as a whole do provide a snap-shot of chemical contamination in the home across the UK, and in other parts of Europe. In short, they confirm that we are all living with the chemical consequences of the widespread use of hazardous additives in consumer goods.

In addition, although this study does not provide (and indeed was not intended to provide), data from which human exposure could be estimated, the results clearly demonstrate the possibility for continuous exposure to these compounds through inhalation, ingestion or direct contact of the skin with dusts. This may be of particular concern with respect to children, as other studies have shown that they have the greatest exposures to dust-related contaminants through inhalation, ingestion and direct skin contact (Butte and Heinzow 2002). Of course, we can never be certain that such exposure is causing adverse health effects, but given the hazards associated with the chemicals in question, there is no reason for complacency. To date, the issue of chemical exposure in the home has generally been poorly investigated and improperly assessed.

For substances which are known to accumulate in the body, such as penta-BDE, HBCD, chlorinated paraffins and some of the organotins, such exposure may contribute further to an overall body burden otherwise dominated by intake from food. Moreover, for substances thought to be less bioaccumulative, such as deca-BDE, their presence in dusts at ppm levels may well help to explain why they are nevertheless detectable in a significant proportion of the general population as background contaminants. It may also explain the somewhat wider environmental distribution of the brominated diphenyl ethers in general, than may be predicted on the basis of chemical mobility.

The effects which may result from such continuous exposure are not known, but the presence of deca-BDE in dusts, for example, may mean that everyone, not just workers in electronics manufacturing and/or recycling plants, will carry some levels of these highly persistent chemicals around in their bodies. Furthermore, irrespective of the potential for exposure to these hazardous substances through contact with dusts in the home, the ultimate disposal of dusts from vacuum cleaners and other sources may represent a significant input of these and other hazardous substances into waste repositories and, ultimately, the surrounding environment.

It is vital that consumer products should be safe to use and this must also include freedom from hazardous chemicals. Requirements for fire safety, commonly conferred through the use of hazardous brominated or chlorinated flame retardants or chlorinated paraffins, can already be met through the use of less hazardous alternatives (see e.g. Lassen *et al.* 1999), including through the use of different materials or designs which make products inherently less flammable. Moreover, many of the indoor chemical hazards identified in this study could be significantly reduced by the use of less hazardous and more sustainable alternatives to the plastic PVC, a source of phthalates, organotins and other hazardous additives. Such alternatives are already available for all PVC products used in the home.

All five of the chemical groups selected for quantitative analysis in this current study have already been identified as priority hazardous substances by the UK and other European governments under the 1992 OSPAR Convention. In 1998, this Convention, (which aims to protect the marine environment of the North East Atlantic region), agreed to stop releases of hazardous substances to the environment within one generation (by 2020). OSPAR included brominated flame retardants, alkylphenols, short-chain chlorinated paraffins, organotin compounds and certain phthalates (DEHP and DBP) on the first list of chemicals requiring action to meet this cessation target (OSPAR 1998). This study makes clear that, until such time as action is taken to replace these chemicals in consumer goods, their release to the indoor environment and the potential thereafter for dusts to contaminate the wider environment will remain a problem.

During 2003, the opportunity exists for the UK, along with other European governments, to take decisive and effective action to tackle the problem of chemical use and exposure, in the home and elsewhere. Recognising the scale of the problem, the lack of knowledge about chemical impacts on human health and the poor progress to date in developing protective measures (EC 2001), the European Commission is currently preparing new regulations to control hazardous chemicals. Their stated intention is that these new laws should provide for a high level of protection for the environment and human health.

If this new legislation is to be effective, however, it will be vital that European governments take action to prevent the use of hazardous chemicals in consumer goods through their

substitution with less hazardous, or preferably non-hazardous, alternatives. The results of this current study provide further evidence that this is the only way in which the chemical safety of our home environment can ultimately be ensured.

# References

- Adeoya-Osiguwa, S.A., Markoulaki, S., Pocock, V., Milligan, S.R. & Fraser, L.R. (2003) 17-beta-estradiol and environmental estrogens significantly effect mammalian sperm function. *Human Reproduction* 18(1): 100-107
- Adgate, J.L., Rhoads, G.G. & Lioy, P.J. (1998) The use of isotope ratios to apportion sources of lead in Jersey City, NJ, house dust wipe samples. *The Science of the Total Environment* 221: 171-180
- Allsopp, M., Santillo, D. & Johnston, P. (2000). Hazardous chemicals in PVC flooring. Greenpeace Research Laboratories Technical Note 14/00, November 2000: 10 pp. [published under cover title "Poison Underfoot: Hazardous Chemicals in PVC Flooring and Hazardous Chemicals in Carpets", ISBN 90-73361-68-0]
- Allsopp, A., Santillo, D. & Johnston, P. (2001) Hazardous chemicals in carpets. Greenpeace Research Laboratories Technical Note 01/2001, January 2001: 14 pp. [published under cover title "Poison Underfoot: Hazardous Chemicals in PVC Flooring and Hazardous Chemicals in Carpets", ISBN 90-73361-68-0]
- Belfroid, A.C., Purperhart, M. & Ariese, F. (2000) Organotin levels in seafood. *Marine Pollution Bulletin* 40(3): 226-232
- Bergman, Å., Ostman, C., Nyborn, R., Sjödin, A., Carlsson, H., Nilsson, U. & Wachtmeister, C.A. (1997) Flame retardants and plasticisers on particulates in the modern computerized indoor environment. *Organohalogen Compounds* 33: 414-419
- Brown S.K. (1996). A lack of influence of permethrin treatments of wool carpet on habitation by house dust mites. *Experimental and Applied Acarology* 20: 355-357
- Butte, W. & Heinzow, B. (2002) Pollutants in house dust as indicators of indoor contamination. *Reviews in Environmental Contamination and Toxicology* 175: 1-46
- Chen, H.Y., Xiao, J.G., Hu, G., Zhou, J.W., Xiao, H. & Wang, X.R. (2002) Estrogenicity of organophosphorus and pyrethroid pesticides. *Journal of Toxicology and Environmental Health-Part A* 65(19): 1419-1435
- Curl, C.L., Fenske, R.A., Kissel, J.C., Shirai, J.H., Moate, T.F., Griffith, W., Coronado, G. & Thompson, B. (2002) Evaluation of take-home organophosphorus pesticide exposure among agricultural workers and their children. *Environmental Health Perspectives* 110(12): A787-A792
- Darnerud, P.O., Eriksen, G.S., Johannesson, T., Larsen, P.B. and Viluksela, M. (2001) Polybrominated diphenyl ethers: Occurrence, dietary exposure, and toxicology. *Environmental Health Perspectives* 109(Suppl. 1): 49-68
- de Boer, J., de Boer, K. & Boon, J.P. (2000) Polybrominated Biphenyls and Diphenylethers. Chapter 4 in: Paasivirta, J. [Ed], *The Handbook of Environmental Chemistry* Vol. 3, Part K: New Types of Persistent Halogenated Compounds, Springer-Verlag, Berlin: 61-95
- Diel, F., Horr, B., Borck, H., Savtchenko, H., Mitsche, T. & Diel, E. (1999) Pyrethroids and piperonyl-butoxide affect human T-lymphocytes in vitro. *Toxicology Letters* 107(1-3): 65-74
- Duty, S.M., Singh, N.P., Silva, M.J., Barr, D.B., Brock, J.W., Ryan, L., Herrick, R.F., Christiani, D.C. & Hauser, R. (2003) The relationship between environmental exposures to phthalates and DNA damage in human sperm using the neutral comet assay. *Environmental Health Perspectives* (in press)
- EC (2001) Commission White Paper: Strategy for a future Chemicals Policy (COM(2001) 88 final), Commission of the European Communities, Brussels, 27.2.2001: 32 pp.
- Edwards, R.D., Yurkow, E.J. & Lioy, P.J. (1998) Seasonal deposition of house dusts onto household surfaces. *The Science of the Total Environment* 224: 69-80
- Eriksson, P., Våberg, H., Jakobsson, E., Örn, U. & Fredriksson, A. (1999) PBDE, 2,2',4,4',5-pentabromodiphenyl ether, causes permanent neurotoxic effects during a defined period of neonatal brain development. *Organohalogen Compounds* 40: 333-336
- Gurunathan, S., Robson, M., Freeman, N., Buckley, B., Roy, A., Meyer, R., Bukowski, J. & Lioy, P.J. (1998) Accumulation of chlorpyrifos on residential surfaces and toys accessible to children. *Environmental Health Perspectives* 106 (1): 9-16
- Helleday, T., Tuominen, K.L., Bergman, Å. & Jenssen, D. (1999) Brominated flame retardants induce transgenic recombination in mammalian cells. *Mutation Research – Genetic Toxicology and Environmental Mutagenesis* 439(2): 137-147
- Huse, A. (1995) Chloro-organics used as flame retardants. *TemaNord* 1995: 39, Nordic Council of Ministers, Copenhagen.
- IEH (1999). Institute for Environment and Health, Indoor Air Quality: Pesticides in the home, IEH report for DETR, UK, October 1999
- Ingerowski, G., Friedle, A. & Thumulla, J. (2001) Chlorinated ethyl and isopropyl phosphoric acid trimesters in the indoor environment – an inter-laboratory exposure study. *Indoor Air – International Journal of Indoor Air Quality and Climate* 11(3): 145-149
- Institoris, L., Undeger, U., Siroki, O., Nehez, M. & Desi, I. (1999) Comparison of detection sensitivity of immuno- and genotoxicological effects of subacute cypermethrin and permethrin exposure in rats. *Toxicology* 137(1): 47-55
- Irvine, K.N. & Loganathan, G.G. (1998) Localized enrichment of PCB levels in street dust due to redistribution by wind. *Water Air and Soil Pollution* 105(3-4): 605-615
- Jakobsson, K., Thuresson, K., Rylander, L., Sjödin, A., Hagmar, L. & Bergman, Å. (2002) Exposure to polybrominated diphenyl ethers and tetrabromobisphenol A among computer technicians. *Chemosphere* 46(5): 709-716
- Kergosien D.H. and Rice C.D. (1998). Macrophage secretory function is enhanced by low doses of tributyltin-oxide (TBTO), but not tributyltin-chloride (TBTC). *Arc. Environ. Contam. Toxicol.* 34: 223-228
- Kunimatsu, T., Yamada, T., Ose, K., Sunami, O., Kamita, Y., Okuno, Y., Seki, T. & Nakatsuka, I. (2002) Lack of (anti-) androgenic or estrogenic effects of three pyrethroids (esfenvalerate, fenvalerate, and permethrin) in the Hershberger and uterotrophic assays. *Regulatory Toxicology and Pharmacology* 35(2): 227-237
- Lagesson, H.V., Nilsson, A. & Tagesson, C. (2000) Qualitative determination of compounds adsorbed on indoor dust particles using GC-UV and GC-MS after thermal desorption. *Chromatographia* 52(9-10): 621-630
- Lassen, C., Lokke, S. & Hansen, L.I. (1999) Brominated Flame Retardants: substance flow analysis and substitution feasibility study. Danish Environmental Protection Agency Environmental Project No. 494, Copenhagen, ISBN 87-7909-415-5: 240 pp.
- Leonards, P.E.G., Santillo, D., Brigden, K., van der Ween, I., Hesseltingen, J.v., de Boer, J. & Johnston, P. (2001) Brominated flame retardants in office dust samples. In: Asplund, L.; Bergman, Å.; de Wit, C., *et al.* eds. *Proceedings of the Second International Workshop on Brominated Flame Retardants, BFR 2001, Stockholm, May 14-16 2001*: 299-302
- Lewis, R.G., Fortmann, R.C. & Camann, D.E. (1994) Evaluation of methods for monitoring the potential exposure of small children to pesticides in the residential environment. *Archives of Environmental Contamination and Toxicology* 26: 37-46
- Meerts, I.A.T.M., Marsh, G., van Leeuwen-Bol, I., Luijckx, E.A.C., Jakobsson, E., Bergman, A. & Brouwer, A. (1998) Interaction of polybrominated diphenyl ether metabolites (PBDE-OH) with human transthyretin in vitro. *Organohalogen Compounds* 37: 309-312
- Meironyte, D., Noren, K. & Bergman, Å. (1999) Analysis of polybrominated diphenyl ethers in Swedish human milk. A time-related trend study, 1972-1997. *Journal of Toxicology and Environmental Health - Part A* 58(6): 329-341
- Meyer, I., Heinrich, J. & Lippold, U. (1999) Factors affecting lead and cadmium levels in house dust in industrial areas of eastern Germany. *The Science of the Total Environment* 234: 25-36
- Olsson, P.-E., Borg, B., Brunstrom, B., Hakansson, H. & Klasson-Wehler, E. (1998). Endocrine disrupting substances - Impairment of reproduction and development. *Swedish Environmental Protection Agency Report 4859*. ISBN 91-620-4859-7: 150 pp.
- OSPAR (1998) OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34
- Prater, M.R., Gogal, R.M., Blaylock, B.L. & Holladay, S.D. (2003) Cis-urocanic acid increases immunotoxicity and lethality of dermally administered permethrin in C57BL/6N mice. *International Journal of Toxicology* 22(1): 35-42
- Punareewattana, K., Smith, B.J., Blaylock, B.L., Longstreth, J., Snodgrass, H.L., Gogal, R.M., Prater, R.M. & Holladay, S.D. (2001) Topical permethrin exposure inhibits antibody production and macrophage function in C57BL/6N mice. *Food and Chemical Toxicology* 39(2): 133-139
- Rudel, R.A., Brody, J.G., Spengler, J.C., Vallarino, J., Geno, P.W., Sun, G. & Yau, A. (2001) Identification of selected hormonally active agents and animal mammary carcinogens in commercial and residential air and dust samples. *Journal of the Air & Waste Management Association* 51(4): 499-513
- Santillo, D., Johnston, P. & Brigden, K. (2001) The presence of brominated flame retardants and organotin compounds in dusts collected from Parliament buildings from eight countries. Greenpeace Research Laboratories Technical Note 03/2001, March 2001: 24 pp.
- Sjödin, A., Carlsson, H., Thuresson, K., Sjölin, S., Bergman, Å. & Ostman, C. (2001) Flame retardants in indoor air at an electronics recycling plant and at other work environments. *Environmental Science and Technology* 35(3): 448-454
- Tong, S.T.Y. & Lam, K.C. (2000) Home sweet home? A case study of household dust contamination in Hong Kong. *The Science of the Total Environment* 256: 115-123
- Tyler, C.R., Beresford, N., van der Woning, M., Sumpster, J.P. & Thorpe, K. (2000) Metabolism and environmental degradation of pyrethroid insecticides produce compounds with endocrine activities. *Environmental Toxicology and Chemistry* 19(4): 801-809
- Vojta, P.J., Friedman, W., Marker, D.A., Clickner, R., Rogers, J.W., Viet, S.M., Mullenberg, M.L., Thorne, P.S., Arbes, S.J. & Zeldin, D.C. (2002) First national survey of lead and allergens in housing: Survey design and methods for the allergen and endotoxin components. *Environmental Health Perspectives* 110 (5): 527-532
- Whyatt, R.M., Camann, D.E., Kinney, P.L., Reyes, A., Ramirez, J., Dietrich, J., Diaz, D., Holmes, D. & Perera, F.P. (2002) Residential pesticide use during pregnancy among a cohort of urban minority women. *Environmental Health Perspectives* 110(5): 507-514
- Wilson, N.K., Chuang, J.C. & Lyu, C. (2001) Levels of persistent organic pollutants in several child day care centers. *Journal of Exposure Analysis and Environmental Epidemiology* 11(6): 449-458
- Yiin, L.M., Rhoads, G.G. & Lioy, P.J. (2000) Seasonal influences on childhood lead exposure. *Environmental Health Perspectives* 108 (2): 177-182



# **Annex 1A: detailed UK regional results for target and non-target compounds in individual and pooled samples**

# Region: Scotland

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02015	nr Stirling	nd	39.3	103.3	67.6	7.4	102.9	nd	nd	320.5	
HD02016	Aberdeen	0.1	7.3	27.3	24.4	8.8	145.9	221.7	63.4	498.9	
HD02022	Edinburgh	0.3	3.8	48.6	71.6	14.8	215.2	nd	nd	354.3	
UK mean (average) value											
UK median (middle) value											
UK minimum (lowest) value											
UK maximum (highest) value											

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02015	nr Stirling	0.2	nd	0.4	0.6	
HD02016	Aberdeen	nd	nd	9.8	9.8	
HD02022	Edinburgh	2.4	nd	25.9	28.3	
UK mean (average) value						
UK median (middle) value						
UK minimum (lowest) value						
UK maximum (highest) value						

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
SC	Scotland	<0.12
UK mean (average)		
UK median (middle)		
UK minimum (lowest)		
UK maximum (highest)		

## Brominated flame retardants – pooled sample analysis Brominated diphenylethers (PBDEs)

Sample code	Region	Concentration of individual brominated diphenylether congeners (ng/g dust, parts per billion, ppb)															
		Tri-	Tetra-	Penta-			Hexa-			Hepta-	Deca-						
		28	47	66	71	75	77	85	99	100	119	138	153	154 *	183	190	209
SC	Scotland	33	1980	59	85	44	<0.1	88	2100	230	<0.1	41	170	110	87	5.4	5500
UK mean (average)																	
UK median (middle)																	
UK minimum (lowest)																	
UK maximum (highest)																	

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD	TBBP-A	methyl-TBBP-A		
SC	Scotland	<0.3	<0.3	<0.3	<0.3	-	<3	3800	340	-			
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158	116	-			
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250	<10	-			
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940	<10	-			
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900	340	-			

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
SC	Scotland	1500	716	759	<1	384	42.6	<1	<1	3402	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02015: nr Stirling	HD02016: Aberdeen	HD02022: Edinburgh
Cyclohexane	Cyclohexane	StyreneButan-1-ol
Methyl methacrylate	Styrene	Methyl methacrylate
Ethyl methacrylate	$\alpha$ -Pinene	Ethyl methacrylate
Styrene	1,8-Cineole (Eucalyptol)	Hexanal
Nonanal	Nonanal	Styrene
1,2,3,4,4a,9,10,10a-octahydrophenanthrene	N,N,N',N' - Tetraacetylenediamine (EDTA)	Nonanal
Totanol	N-(2-Hydroxyethyl)-decanamide	N,N,N',N' - Tetraacetylenediamine (EDTA)
Piperine (plus unidentified alkane)	Tris(3-chloropropyl) phosphonate	N,N-Tetradecanamine
Freidelin	Hexadecyl benzoate	Butyl octadecanoate
Unidentified triglyceride (similar mass spectrum to that of trilaurin)	4b,5,6,7,8,8a,9,10-Octahydrophenanthren-2-ol	Tri-[2-Butoxyethanol]phosphonate
	Piperine (plus unidentified alkane)	Permethylin
	Stigmast-4-en-3-one	Piperine
	Unidentified triglyceride	Cholesta-3,5-dien-7-one
	Sesquiterpene (plus unidentified high molecular weight compound)	Chole-4-en-3-one
		Unidentified triglyceride

# Region: North East

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02086	Sheffield	nd	6.6	29.3	64.7	189.1	204.3	nd	nd	494	
HD02091	Newcastle	nd	3.5	20.1	73.6	11.4	231.2	nd	nd	339.8	
HD02094	Darlington	nd	2.5	70.6	76.7	103.1	152.4	103.5	nd	508.8	
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UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum (lowest) value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum (highest) value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02086	Sheffield	nd	nd	6.1	6.1	
HD02091	Newcastle	nd	nd	nd	nd	
HD02094	Darlington	nd	nd	16.6	16.6	
<hr/>						
UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
NE	North East	2.4
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UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)															
		Tri-	Tetra-		Penta-			Hexa-			Hepta-	Deca-					
		28	47	66	71	75	77	85	99	100	119	138	153	154 *	183	190	209
NE	North East	<0.1	15	1.8	15	9.3	<0.1	2.7	28	6.4	<0.1	0.8	10	3.2	<0.1	<0.1	12100
<hr/>																	
UK mean (average)		4.14	223	7.8	49	30.1	<0.1	12.2	287	33	2.55	5.06	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	3.5	44	8.5	0.33	0.3	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	<0.1	2.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	59	110	67	<0.1	88	2100	230	17	41	170	110	87	5.4	19900

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD	TBBP-A	methyl-TBBP-A		
NE	North East	<0.3	<0.3	<0.3	<0.3	-	<3	940	<10	-			
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158	116	-			
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250	<10	-			
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940	<10	-			
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900	340	-			

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
NE	North East	2800	1300	24.6	<1	703	219	<1	<1	5047	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02086: Sheffield	HD02091: Newcastle-upon-Tyne	HD02094: Darlington
Styrene Nonanal Nicotine (and Nicotine stereoisomer) Dodecan-1-ol Ibuprofen N,N,N',N'-Tetraacetylenediamine (EDTA) p-Toluenesulfonamide Unidentified branched aldehyde (x2) 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Di-(2-ethylhexyl)-isophthalate Cholesta-4,6-dien-3-ol Cholesterol Vitamin E acetate	Butan-2-one Styrene α-Pinene Limonene Triethylene glycol Nonanal Pentaethylene glycol Tris(3-chloropropyl) phosphonate Unidentified organonitrogen compound (possibly N-Propylbenzamide) Piperine Cholesterol	Styrene Heptanal β-Phellandrene Limonene Nonanal N,N,N',N'-Tetraacetylenediamine (EDTA) Tris(3-chloropropyl) phosphonate Phenanthrene Fluoranthrene Pyrene 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Chrysene Benzo(b)fluoranthrene Permethrin Piperine (and Piperine stereoisomer) Perylene Cholesterol

# Region: North West

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02100	Macclesfield	nd	1.2	50.7	59.1	238.9	228.1	12.7	nd	590.7	
HD02102	Manchester	nd	1.8	34	23.7	52.5	256.5	nd	nd	368.5	
HD02104	Oldham	nd	0.6	32.6	15.3	25	159.3	nd	nd	232.8	
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UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum (lowest) value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum (highest) value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02100	Macclesfield	nd	nd	22	22	
HD02102	Manchester	nd	nd	25.7	25.7	
HD02104	Oldham	nd	nd	nd	nd	
<hr/>						
UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
NW	North West	4.7
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UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)															
		Tri-	Tetra-	Penta-	Hexa-	Hepta-	Deca-										
NW	North West	28	47	66	71	75	77	85	99	100	119	138	153	154 *	183	190	209
		0.9	15	1.3	21	16	<0.1	2.4	37	5.6	1.1	<0.1	23	3.1	<0.1	<0.1	6300
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UK mean (average)		4.14	223	7.8	49	30.1	<0.1	12.2	287	33	2.55	5.06	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	3.5	44	8.5	0.33	0.3	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	<0.1	2.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	59	110	67	<0.1	88	2100	230	17	41	170	110	87	5.4	19900

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
NW	North West	<0.3	<0.3	<0.3	<0.3	-	<3	1400			300	-	
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158			116	-	
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250			<10	-	
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940			<10	-	
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900			340	-	

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)										Total organotins
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT			
NW	North West	1300	478	43.9	<1	1300	199	<1	<1	<1	3321	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669		
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432		
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581		
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047		

Other compounds tentatively identified by GC-MS screen

HD02100: Macclesfield	HD02102: Manchester	HD02104: Oldham
Styrene	Methyl methacrylate	Styrene
Nonanal	Butan-2-one	Tris(3-chloropropyl) phosphonate
Tris(3-chloropropyl) phosphonate	Styrene	1,1'-[(Methylthio)ethylene]bis-benzene
3-(4-Methoxyphenyl)-2-ethylhexylpropanoate	Levulinic acid	Tri-[2-Butoxyethanol]phosphonate
Tri-[2-Butoxyethanol]phosphonate	Nonanal	Permethylin (and Permethylin stereoisomer)
Benzo(b)fluoranthrene	Undecylinic acid	Cholesta-4,6-dien-3-ol/Phthalate
Permethylin	Pentaethylene glycol	Cholesta-3,5-diene
Cholesta-4,6-dien-3-ol/Phthalate	Tris(3-chloropropyl) phosphonate	Cholesterol
Cholesta-3,5-diene	Permethylin (and Permethylin stereoisomer)	
Cholesterol	Cholesta-4,6-dien-3-ol/Phthalate	
	Cholesta-3,5-diene	
	Cholesterol	

# Region: East Midlands

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02009	Peterborough	0.7	87.1	95	87.4	74.9	362.6	118.8	156.6	983.1	
HD02010	Leicester	1.1	28.1	100.7	106.4	204.7	416.4	88	73.7	1019.1	
UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum (lowest) value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum (highest) value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)					
		4TMBP	4OP	4NP	Total		
HD02009	Peterborough	nd	nd	29.7	29.7		
HD02010	Leicester	nd	8.6	nd	8.6		
UK mean (average) value		0.12	0.3	10.5	10.9		
UK median (middle) value		nd	nd	9.8	9.8		
UK minimum (lowest) value		nd	nd	nd	nd		
UK maximum (highest) value		2.4	8.6	35.2	36.1		

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
EM	East Midlands	<0.12
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)															
		Tri-	Tetra-	Penta-	Hexa-	Hepta-	Deca-										
EM	East Midlands	28	47	66	71	75	77	85	99	100	119	138	153	154 *	183	190	209
East Midlands		0.4	10	1.2	14	7.7	<0.1	1.5	18	4.3	<0.1	<0.1	6.5	2.2	6.4	0.2	16600
UK mean (average)		4.14	223	7.8	49	30.1	<0.1	12.2	287	33	2.55	5.06	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	3.5	44	8.5	0.33	0.3	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	<0.1	2.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	59	110	67	<0.1	88	2100	230	17	41	170	110	87	5.4	19900



Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
EM	East Midlands	<0.3	<0.3	<0.3	<0.3	-	-	<3	1000			<10	-
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3158			116	-
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3250			<10	-
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	940			<10	-
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	6900			340	-

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)										Total organotins
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT			
EM	East Midlands	1500	560	30.5	<1	430	545	<1	<1	<1	3066	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669		
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432		
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581		
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047		

Other compounds tentatively identified by GC-MS screen

HD02009: Peterborough	HD02010: Leicester
Butan-2-one	N-t-butylethanamine
Styrene	2,4,4-Trimethylpent-1-ene
Acetophenone	3,4,4-Trimethylpent-2-ene
Nonanal	2,4,4-Trimethylpent-2-ene
2,3-Isopropylidene-dioxyphenol (suspected breakdown product of Bendiocarb)	2-Ethyl-3-methyloxazolidine
Toluene-2,4-difluorinate	Styrene
Bendiocarb	α-Pinene
Bis-(2-ethylhexyl) adipate	N-(1-Hydroxymethyl-2-methylpropyl)-formamide
Triphenylphosphate	2-Pentylfuran
Octicizer (2-ethylhexyl-dibenzylphosphonate)	2-Chlorotoluene or Benzyl chloride
Cypermethrin	2-Ethylhexan-1-ol
	Limonene
	Benzyl alcohol
	Linalool
	3-Ethylhexa-3-amine
	2-(t-Butylamino)ethanol
	5-Formyl-1H-pyrrolicarboxylic acid, methyl ester
	Linalyl acetate
	Butylated hydroxy toluene (plus an unidentified amine)
	1-Isobutyl-2-methyl-1,3-dipropandyl-2-methylpropanoate
	Methyl dihydrojasmonate
	p-Toluenesulfonamide
	Benzyl salicylate
	1,1'-(Methylthio)ethylenedibis(benzene)
	Tri-[2-butoxyethanol]phosphonate
	Octicizer (2-ethylhexyl-dibenzylphosphonate)

# Region: West Midlands

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02079	Oxford	nd	0.9	38.1	22.7	24.5	230	nd	nd	316.2	
HD02080	Oxford	nd	3.9	15.4	21.9	16.9	64.1	nd	nd	122.2	
HD02081	Twyford	0.3	5.1	41	56.9	84	195.4	337.2	79.9	799.8	
UK mean (average) value											
UK median (middle) value											
UK minimum (lowest) value											
UK maximum (highest) value											

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)					
		4TMBP	4OP	4NP	Total		
HD02079	Oxford	nd	nd	11.5	11.5		
HD02080	Oxford	nd	nd	2.1	2.1		
HD02081	Twyford	0.1	nd	13.8	13.9		
UK mean (average) value							
UK median (middle) value							
UK minimum (lowest) value							
UK maximum (highest) value							

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
WM	West Midlands	1.9
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)																	
		Tri-	Tetra-	Penta-	Hexa-	Hepta-	Deca-												
WM	West Midlands	28	47	15	66	71	75	77	85	99	100	100	119	138	153	154 *	183	190	209
UK mean (average)		4.14	223	7.8	49	30.1	<0.1	12.2	287	33	2.55	5.06	16.8	19.2	0.75	9820			
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	3.5	44	8.5	0.33	0.3	4.7	9.5	<0.1	7100			
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	2.1	<0.1	<0.1	3800			
UK maximum (highest)		33	1980	59	110	67	<0.1	88	2100	230	17	41	110	87	5.4	19900			

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
WM	West Midlands	<0.3	<0.3	<0.3	<0.3	-	-	<3	1640			<10	-
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3158			116	-
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3250			<10	-
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	940			<10	-
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	6900			340	-

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
WM	West Midlands	810	157	21.6	<1	749	82.8	<1	68.9	1889	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02079: Oxford	HD02080: Oxford	HD02081: Twyford
Styrene	Cyclohexane	Methyl methacrylate
Phthalic anhydride	1-Methylpyrrolidin-2-one	2,4,4-Trimethylpent-1-ene
Lindane	Nonanal	Pentan-2,4-dione
Phenanthrene	Nonanoic acid	Styrene
Fluoranthrene	Dodecanoic acid	Nonanal
Cholesterol	Tetradecanoic acid	α-Hexylcinnamaldehyde
Cholest-4-en-3-one	Pentadecanoic acid	Tris(2-chloroethyl) phosphonate
Tris(2-Ethylhexyl)trimellitate	Hexadecanoic acid	Tris(3-chloropropyl) phosphonate
	Octadec-(9Z)-enoic acid	Totarol
	Octadecanoic acid	Tri-[2-Butoxyethanol]phosphonate
	Bisphenol A	Cholesterol
	n-Tricosane	
	3-(4-Methoxyphenyl)-2-ethylhexylpropenoate	
	n-Tetracosane	
	n-Pentacosane	
	Unidentified alkane or phthalate (x2)	

# Region: East Anglia

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02038	Harleston	nd	5.5	30.8	14.1	8.8	224.7	nd	nd	283.9	
HD02042	Norwich	0.3	2.3	20.4	32.3	31.6	279.6	96.8	48.4	511.7	
HD02043	Harlow	0.2	3.8	46.1	52.8	174.2	211	nd	nd	488.1	
UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum (lowest) value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum (highest) value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02038	Harleston	nd	nd	nd	nd	
HD02042	Norwich	nd	nd	11.3	11.3	
HD02043	Harlow	nd	nd	11.9	11.9	
UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
EA	East Anglia	4.5
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)									
		Tri-28	Tetra-47	Penta-85	Hexa-138	Hepta-183	Deca-209				
EA	East Anglia	0.3	29	4.1	2.3	6.1	23	<0.1	<0.1	5900	
UK mean (average)		4.14	223	12.2	5.06	16.8	19.2	33.8	0.75	9820	
UK median (middle)		0.35	24.8	3.5	0.33	4.7	9.5	23	<0.1	7100	
UK minimum (lowest)		<0.1	10	1.5	<0.1	2.1	<0.1	<0.1	<0.1	3800	
UK maximum (highest)		33	1980	88	41	110	87	170	5.4	19900	

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
EA	East Anglia	<0.3	<0.3	<0.3	<0.3	-	-	<3	4700			<10	-
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3158			116	-
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3250			<10	-
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	940			<10	-
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	6900			340	-

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)										Total organotins
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT			
EA	East Anglia	1400	621	55.9	<1	314	89.2	<1	<1	<1	2480	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669		
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432		
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581		
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047		

Other compounds tentatively identified by GC-MS screen

HD02038: Harleston	HD02042: Norwich	HD02043: Harlow
Hexanal	Hexanal	Benzaldehyde
Styrene	Styrene	Nonanal
Nonanal	Nonanal	N,N,N',N'-Tetraacetylenediamine (EDTA)
1-(2-Methoxy-1-methylethoxy)-propan-2-ol	Nicotine	Tris(3-chloropropyl) phosphonate
p,p'-DDT	Tris(3-chloropropyl) phosphonate	Octicizer (2-ethylhexyl-dibenzylphosphonate)
p,p'-DDE	Phenanthrene	Cholesta-4,6-dien-3-ol/phthalate
Cholesterol	Octicizer (2-ethylhexyl-dibenzylphosphonate)	Cholesterol
	Unidentified triglyceride (x2)	
	Cholesterol	
	Tri-(2-Ethylhexyl)-trimellitate	

# Region: Wales

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02066	Newport	nd	4	58.9	44.8	9.8	154.7	50.5	24.1	346.8	
HD02068	Cardiff	0.2	114.8	157.4	102	6.8	138.9	nd	39.3	559.4	
HD02069	Swansea	nd	1.6	34.1	24.9	22.8	173.1	nd	4.3	260.8	
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UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02066	Newport	nd	nd	10.7	10.7	
HD02068	Cardiff	nd	nd	5.9	5.9	
HD02069	Swansea	nd	nd	22.6	22.6	
<hr/>						
UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
WL	Wales	9.5
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UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)													
		Tri-28	Tetra-47	Penta-77	Hexa-100	Hepta-183	Octa-209								
WL	Wales	0.4	43	<0.1	81	75	58	<0.1	83	13	26	<0.1	<0.1	<0.1	7900
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UK mean (average)		4.14	223	<0.1	49	30.1	<0.1	12.2	287	33	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	<0.1	45	24	<0.1	3.5	44	8.5	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	<0.1	110	67	<0.1	88	2100	230	170	110	87	5.4	19900

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
WL	Wales	<0.3	<0.3	<0.3	<0.3	-	<3	4700			<10	-	
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158			116	-	
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250			<10	-	
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940			<10	-	
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900			340	-	

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)										Total organotins
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT			
WL	Wales	841	570	79.7	<1	126	27.2	<1	<1	<1	1644	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669		
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432		
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581		
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047		

Other compounds tentatively identified by GC-MS screen

HD02066: Newport	HD02068: Cardiff	HD02069: Swansea
Styrene	Styrene	Styrene
Benzaldehyde	Octanal	Butyl methacrylate
Acetophenone	<i>p</i> -Cymene	Nonanal
Nonanal	Limonene	Phthalic anhydride
Dodecan-1-ol	Nonanal	Nicotine (and Nicotine stereoisomer)
Tris(3-chloropropyl) phosphonate	Nicotine (and Nicotine stereoisomer)	Dodecan-1-ol
Totanol	Dodecan-1-ol	N,N-Dodecanamine
Triphenylphosphonate	N,N,N',N'-Tetraacetylenediamine (EDTA)	N,N-Tetradecanamine
Octicizer (2-ethylhexyl-dibenzylphosphonate)	N,N-Tetradecanamine	Permethrin (and Permethrin stereoisomer)
Unidentified phthalate (not common form)	Benzyl salicylate	Cholesta-3,5-diene
Piperine	3-(4-Methoxyphenyl)-2-ethylhexylpropanoate	Cholesterol
Cholesterol	Cholesterol	
Vitamin E acetate	Vitamin E acetate	

# Region: London

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DiNP	DiDP	Total	
HD02029	London SE4	nd	2.9	4.2	28.8	6.6	74.2	nd	nd	116.7	
HD02030	London E5	nd	0.8	0.2	0.1	nd	0.5	nd	nd	1.6	
HD02031	London N4	nd	7.3	95.5	22.9	62.9	123.6	nd	nd	312.2	
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UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7	
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3	
UK minimum (lowest) value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6	
UK maximum (highest) value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1	

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02029	London SE4	nd	nd	7.4	7.4	
HD02030	London E5	nd	nd	nd	nd	
HD02031	London N4	nd	nd	10.9	10.9	
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UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
LD	London	13.0
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UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)											
		Tri-28	Tetra-47	Penta-77	Hexa-100	Hepta-183	Octa-209						
LD	London	6.2	76	<0.1	8.6	130	<0.1	36	49	21	46	1.9	3800
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UK mean (average)		4.14	223	<0.1	12.2	287	5.06	33	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	<0.1	3.5	44	0.33	8.5	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	1.5	18	<0.1	3.9	<0.1	2.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	<0.1	88	2100	41	230	170	110	87	5.4	19900



Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)										
		Brominated biphenyls					HBCD			TBBP-A	methyl-	TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209				
LD	London	<0.3	<0.3	<0.3	<0.3	-	<3			2700	<10	-
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3			3158	116	-
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3			3250	<10	-
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3			940	<10	-
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3			6900	340	-

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
LD	London	1400	465	264	<1	228	27.5	<1	<1	2385	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02029: London SE4	HD02030: London E5	HD02031: London N4
Hexanal	none	Hexanal
Styrene		Butan-2-one
Benzaldehyde		Styrene
Nonanal		Nonanal
Polyethylene glycol		Dodecan-1-ol
Tris(3-chloropropyl) phosphonate		Piperonyl butoxide
Octicizer (2-ethylhexyl-dibenzylphosphonate)		Tetramethrin
Unidentified triglyceride		Cholesta-3,5-diene
		Cholesterol
		Cholest-4-en-3-one

# Region: South East

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02047	Canterbury	nd	1.0	68.1	36.7	107.4	183.8	70.7	18.8	486.5	
HD02052	Crawley	0.1	1.9	77.3	93.5	72.1	260.8	242.8	66.7	815.2	
HD02054	Arundel	nd	1.7	71.5	59.1	5.7	204.7	nd	nd	342.7	
UK mean (average) value											
UK median (middle) value											
UK minimum (lowest) value											
UK maximum (highest) value											

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02047	Canterbury	nd	nd	17.1	17.1	
HD02052	Crawley	nd	nd	2.5	2.5	
HD02054	Arundel	nd	nd	nd	nd	
UK mean (average) value					10.9	
UK median (middle) value					9.8	
UK minimum (lowest) value					nd	
UK maximum (highest) value					36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
SE	South East	4.1
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)																	
		Tri-28	Tetra-47	Penta-77	Hexa-100	Hepta-183	Octa-209												
SE	South East	0.15	23.5	2.95	51	71	75	77	85	99	100	100	119	138	153	154 *	183	190	209
UK mean (average)		4.14	223	7.8	49	30.1	<0.1	<0.1	12.2	287	33	2.55	5.06	16.8	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	<0.1	3.5	44	8.5	0.33	0.3	4.7	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	<0.1	1.5	18	3.9	<0.1	<0.1	2.1	<0.1	<0.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	59	110	67	<0.1	<0.1	88	2100	230	17	41	110	170	110	87	5.4	19900

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD			TBBP-A	methyl-TBBP-A
SE	South East	<0.3	<0.3	<0.3	<0.3	-	<3	3800			330	-	
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158			116	-	
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250			<10	-	
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940			<10	-	
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900			340	-	

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
SE	South East	1000	458	23.2	<1	82.5	17.6	<1	<1	1581	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02047: Canterbury	HD02052: Crawley	HD02053: Arundel
<ul style="list-style-type: none"> <li>α-Pinene</li> <li>p-Cymene</li> <li>Limonene</li> <li>Eucalyptol</li> <li>γ-Terpinene</li> <li>Linalool</li> <li>Nonanal</li> <li>Terpin-4-ol</li> <li>α-Terpineol</li> <li>Sesquiterpene</li> <li>N,N,N',N'-Tetraacetylenediamine (EDTA)</li> <li>Unidentified PAH</li> <li>2,2-Diphenyl-2H-1-benzopyran</li> <li>Piperine</li> <li>Vitamin E</li> <li>Cholesterol</li> </ul>	<ul style="list-style-type: none"> <li>Methyl methacrylate</li> <li>Styrene</li> <li>α-Pinene</li> <li>Nonanal</li> <li>Octicizer (2-ethylhexyl-dibenzylphosphonate)</li> <li>Cholesta-4,6-dien-3-ol/phthalate</li> <li>Cholesterol</li> <li>Cholest-4-en-3-one</li> </ul>	<ul style="list-style-type: none"> <li>Styrene</li> <li>α-Pinene</li> <li>Nonanal</li> <li>Phthalic anhydride</li> <li>1-Methyldodecylbenzene</li> <li>Tributyl acetyl citrate</li> <li>Cholesterol</li> <li>Cholesta-3,5-dien-7-one</li> </ul>

# Region: South West

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)									
		DMP	DEP	DiBP	DnBP	BBP	DEHP	DINP	DiDP	Total	
HD02059	Cannington	0.2	2.5	43.2	43.2	14.1	271.2	62.9	27.4	464.7	
HD02062	Plymouth	0.1	7.6	27	57.9	53.6	158.4	nd	nd	304.6	
HD02064	Romsey	nd	3.4	66.5	70	6.1	129.2	nd	nd	275.2	
UK mean (average) value											
UK median (middle) value											
UK minimum (lowest) value											
UK maximum (highest) value											

## Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMBP	4OP	4NP	Total	
HD02059	Cannington	nd	nd	5.6	5.6	
HD02062	Plymouth	0.9	nd	35.2	36.1	
HD02064	Romsey	nd	nd	nd	nd	
UK mean (average) value					10.9	
UK median (middle) value					9.8	
UK minimum (lowest) value					nd	
UK maximum (highest) value					36.1	

## Short-chain chlorinated paraffins (SCCPs) – pooled sample analysis

Sample code	Region	Concentration of CPs (ug/g, parts per million, ppm)
SW	South West	3.3
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Brominated flame retardants – pooled sample analysis

Sample code	Region	Concentration of individual brominated diphenylethers (PBDEs) (ng/g dust, parts per billion, ppb)																	
		Tri-28	Tetra-47	Penta-85	Hexa-138	Hepta-183	Deca-209												
SW	South West	<0.1	26	<0.1	64	71	75	77	85	99	100	119	138	153	154 *	183	190	<0.1	19900
UK mean (average)		4.14	223	<0.1	49	30.1	<0.1	12.2	287	33	2.55	5.06	16.8	33.8	16.8	19.2	0.75	9820	
UK median (middle)		0.35	24.8	<0.1	45	24	<0.1	3.5	44	8.5	0.33	0.3	4.7	23	4.7	9.5	<0.1	7100	
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	2.1	<0.1	2.1	<0.1	<0.1	3800	
UK maximum (highest)		33	1980	<0.1	110	67	<0.1	88	2100	230	17	41	110	170	110	87	5.4	19900	

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)											
		Brominated biphenyls					HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209	HBCD	TBBP-A	methyl-TBBP-A		
SW	South West	<0.3	<0.3	<0.3	<0.3	-	<3	6900	190	-			
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	<3	3158	116	-			
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	<3	3250	<10	-			
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	<3	940	<10	-			
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	<3	6900	340	-			

Organotin compounds – pooled sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)									
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT	Total organotins	
SW	South West	1200	306	143	<1	189	42.1	<1	<1	1880	
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669	
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432	
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581	
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047	

Other compounds tentatively identified by GC-MS screen

HD02059: Cannington (Somerset)	HD02062: Plymouth	HD02064: Romsey
Methyl methacrylate Limonene Nonanal N,N-Dimethyl(dodecamine N,N,N',N'-Tetraacetythylenediamine (EDTA) Tris(3-chloropropyl) phosphonate 2,6-Di-tert-butyl-1,4-benzoquinone Totarol Tri-[2-Butoxyethanol]phosphonate Tris(2-Ethylhexyl)trimellitate	Benzene Methyl methacrylate Styrene Nonanal Dodecan-1-ol N,N,N',N'-Tetraacetythylenediamine (EDTA) Tris(3-chloropropyl) phosphonate Piperonyl butoxide Cholesterol	Styrene Heptanal 1,2-Dimethoxypropane Nonanal Tris(3-chloropropyl) phosphonate Totarol Tri-[2-butoxyethanol]phosphonate Unidentified amine Permethrin Cholesta-4,6-dien-3-ol/Phthalate Cholesterol

# **Annex 1B: ranked tables of UK regional results for target and non-target compounds in individual and pooled samples**

**1. Total Phthalates (29 individual samples)**  
(sum of MBP, DEP, DiBP, DIBP, DnBP, BBP, DEHP, DnBP and DiDP)

Rank	Region	Concentration (ppm)
1	East Midlands	1019.1
2	East Midlands	983.1
3	South East	815.2
4	West Midlands	799.8
5	North West	590.7
6	Wales	559.4
7	East Anglia	511.7
8	North East	508.8
9	Scotland	498.9
10	North East	494
11	East Anglia	488.1
12	South East	486.5
13	South West	464.7
14	North West	368.5
15	Scotland	354.3
16	Wales	346.8
17	South East	342.7
18	North East	339.8
19	Scotland	320.5
20	West Midlands	316.2
21	London	312.2
22	South West	304.6
23	East Anglia	283.9
24	South West	275.2
25	Wales	260.8
26	North West	232.8
27	West Midlands	122.2
28	London	116.7
29	London	1.6

**2. Nonylphenol (29 individual phthalates)**

Rank	Region	Concentration (ppm)
1	Wales	35.2
2	East Midlands	29.7
3	North West	25.9
4	London	25.7
5	West Midlands	22.6
6	West Midlands	22
7	South East	17.1
8	North West	16.6
9	East Anglia	13.8
10	East Anglia	11.9
11	London	11.5
12	North East	11.3
13	North East	10.9
14	North East	10.7
15	West Midlands	9.8
16	Wales	7.4
17	South West	6.1
18	Scotland	5.9
19	Scotland	5.6
20	South West	2.5
21	South West	2.1
22	South East	0.4
23	East Midlands	<0.1
24	East Anglia	<0.1
25	Scotland	<0.1
26	North West	<0.1
27	South East	<0.1
28	Wales	<0.1
29	London	<0.1

**3.1 Decabromodiphenyl ether (BDE-209)  
(10 pooled samples)**

Rank	Region	Concentration (ppm)
1	South West	19.9
2	East Midlands	16.6
3	South East	14.4
4	North East	12.1
5	Wales	7.9
6	North West	6.3
7	East Anglia	5.9
8	West Midlands	5.8
9	Scotland	5.5
10	London	3.8

**3.2 Hexabromocyclododecane (HBCD)  
(10 pooled samples)**

Rank	Region	Concentration (ppm)
1	South West	6.9
2	East Anglia	4.7
3	Wales	4.7
4	Scotland	3.8
5	South East	3.8
6	London	2.7
7	West Midlands	1.64
8	North West	1.4
9	East Midlands	1.0
10	North East	0.94

**4. Total organotin compounds  
(10 pooled samples)  
(sum of MBT, DBT, TBT, MOT, DOT and TPT)**

Rank	Region	Concentration (ppm)
1	North East	5.05
2	Scotland	3.4
3	North West	3.32
4	East Midlands	3.07
5	East Anglia	2.48
6	London	2.38
7	West Midlands	1.89
8	South West	1.88
9	Wales	1.64
10	South East	1.58

**5. Short-chain chlorinated paraffins  
(SCCPs) (10 pooled samples)**

Rank	Region	Concentration (ppm)
1	London	13.0
2	Wales	9.5
3	North West	4.7
4	East Anglia	4.5
5	South East	4.1
6	South West	3.3
7	North East	2.4
8	West Midlands	1.9
9	East Midlands	<0.12
10	Scotland	<0.12



# **Annex 1C: detailed non-UK results for target and non-target compounds**

# Region: non UK samples

## Phthalates – individual sample analyses

Sample code	Location	Concentration of phthalate esters (ug/g dust, parts per million, ppm)										
		DMP	DEP	DIBP	DnBP	BBP	DEHP	DnBP	DIBP	DEHP	DnBP	Total
HD02105	Finland	nd	136.6	25.3	49	27	353.5	248.2	67.4	907		
HD02106	Finland	0.4	2	6.1	140.9	32.2	148	nd	nd	329.6		
HD02107	Finland	2.2	84.7	18.7	37.8	38.5	579.3	4.3	nd	765.5		
HD02108	Denmark	nd	0.7	8.8	33.5	67.1	45.5	nd	nd	155.6		
HD02109	Denmark	nd	0.7	6.1	79	26.1	179.3	nd	nd	291.2		
HD02110	Denmark	nd	1.5	13.2	8.5	13.6	183.6	nd	nd	220.4		
HD02111	Sweden	nd	2.5	31.2	21.9	60.2	207	88.9	nd	411.7		
HD02112	Sweden	nd	20.3	10.8	101.6	97.4	239.2	71.1	nd	540.4		
HD02113	France	nd	43.6	68.4	22.1	9.3	185.4	312.4	nd	641.2		
HD02114	Spain	0.4	9.5	37.6	119.9	141.8	194.4	117.8	nd	621.4		
UK mean (average) value		0.12	12.2	52	50.2	56.5	191.5	48.5	20.8	431.7		
UK median (middle) value		nd	3.5	43.2	52.8	24.5	195.4	nd	nd	354.3		
UK minimum value		nd	0.6	0.2	0.1	nd	0.5	nd	nd	1.6		
UK maximum value		1.1	114.8	157.4	106.4	238.9	416.4	337.2	156.6	1019.1		

### Alkylphenols – individual sample analyses

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)				
		4TMPBP	4OP	4NP	Total	
HD02105	Finland	nd	nd	13.1	13.1	
HD02106	Finland	nd	nd	nd	nd	
HD02107	Finland	nd	nd	11.1	11.1	
HD02108	Denmark	nd	nd	nd	nd	
HD02109	Denmark	nd	nd	nd	nd	
HD02110	Denmark	nd	nd	9.3	9.3	
HD02111	Sweden	nd	nd	nd	nd	
HD02112	Sweden	nd	nd	3.3	3.3	
HD02113	France	nd	nd	10.9	10.9	
HD02114	Spain	nd	nd	7.3	7.3	
UK mean (average) value		0.12	0.3	10.5	10.9	
UK median (middle) value		nd	nd	9.8	9.8	
UK minimum (lowest) value		nd	nd	nd	nd	
UK maximum (highest) value		2.4	8.6	35.2	36.1	

### Brominated flame retardants – individual sample analysis for two samples only

Brominated diphenylethers (PBDEs)

Sample code	Region	Concentration of individual brominated diphenylether congeners (ng/g dust, parts per billion, ppb)															
		Tri-	Tetra-		Penta-			Hexa-		Hepta-		Deca-					
HD02105	Finland	28	47	66	71	75	77	85	99	100	119	138	153	154 *	183	190	209
HD02110	Denmark	0.1	9.9	0.7	<0.1	<0.1	<0.1	1.8	8.8	3.5	<0.1	<0.1	<0.1	3.8	<0.1	<0.1	100
		3	66	3.6	13	10	<0.1	1.9	<0.1	11	<0.1	0.2	23	1.8	11	0.6	260
UK mean (average)		4.14	223	7.8	49	30.1	<0.1	12.2	287	33	2.55	5.06	33.8	16.8	19.2	0.75	9820
UK median (middle)		0.35	24.8	1.55	45	24	<0.1	3.5	44	8.5	0.33	0.3	23	4.7	9.5	<0.1	7100
UK minimum (lowest)		<0.1	10	<0.1	9.8	4.7	<0.1	1.5	18	3.9	<0.1	<0.1	<0.1	2.1	<0.1	<0.1	3800
UK maximum (highest)		33	1980	59	110	67	<0.1	88	2100	230	17	41	170	110	87	5.4	19900

# Region: non UK samples (continued)

Brominated biphenyls (PBBs), hexabromocyclododecane (HBCD) and tetrabromobisphenol-A (TBBP-A)

Sample code	Region	Concentration of additional brominated flame retardant compounds (ng/g, ppb)												
		Brominated biphenyls						HBCD					TBBP-A	methyl-TBBP-A
		BB-15	BB-49	BB-52	BB-101	BB-153	BB-155	BB-209						
HD02105	Finland	<0.3	<0.3	<0.3	<0.3	-	-	<3	790	25	-	-		
HD02110	Denmark	<0.3	<0.3	<0.3	<0.3	-	-	<3	1000	400	-	-		
UK mean (average)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3158	116	-	-		
UK median (middle)		<0.3	<0.3	<0.3	<0.3	-	-	<3	3250	<10	-	-		
UK minimum (lowest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	940	<10	-	-		
UK maximum (highest)		<0.3	<0.3	<0.3	<0.3	-	-	<3	6900	340	-	-		

## Organotin compounds – individual sample analysis

Sample code	Region	Concentration of organotin compounds (ng/g, parts per billion, ppb)											Total organotins
		MBT	DBT	TBT	TeBT	MOT	DOT	TCHT	TPT				
HD02105	Finland	764	225	7.8	<1	154	32.8	<1	<1	1184			
HD02106	Finland	200	73.7	6.4	<1	240	47.3	<1	<1	567			
HD02107	Finland	918	330	21.5	<1	908	3600	<1	31.1	5809			
HD02108	Denmark	80.1	25.1	12.4	<1	15.9	5.6	<1	<1	139			
HD02109	Denmark	267	86.5	155	<1	60.4	2.8	<1	39.2	611			
HD02110	Denmark	462	265	20.1	<1	93.7	53.3	<1	<1	894			
HD02111	Sweden	754	311	20.4	<1	383	102	<1	<1	1570			
HD02112	Sweden	1200	169	81.4	<1	90.9	12.5	<1	<1	1554			
HD02113	France	632	269	49	<1	178	41.3	<1	<1	1169			
HD02114	Spain	662	158	3.5	<1	671	96	<1	<1	1591			
UK mean (average)		1375	563	144.5	<1	450.6	129.2	<1	6.9	2669			
UK median (middle)		1350	519	49.9	<1	349	62.7	<1	<1	2432			
UK minimum (lowest)		810	157	21.6	<1	82.5	17.6	<1	<1	1581			
UK maximum (highest)		2800	1300	759	<1	1300	545	<1	68.9	5047			

## Short-chain chlorinated paraffins (SCCPs) - individual sample analysis for two samples only

Sample code	Region	Concentration of SCCPs (ng/g, parts per billion, ppb)
HD02105	Finland	9.6
HD02110	Denmark	5.1
UK mean (average)		4.3
UK median (middle)		3.7
UK minimum (lowest)		<0.12
UK maximum (highest)		13.0

## Finland

### Other compounds tentatively identified by GC-MS screen

HD02105: Finland	HD02106: Finland	HD02107: Finland
1-Methoxypropan-2-ol Styrene 2-Ethylhexan-1-ol Nonanal Phthalic anhydride Decanoic acid p-tert-Butylbenzoic acid N,N-Dimethyl(dodecamine N,N,N',N'-Tetraacetylenediamine (EDTA) Hexyl salicylate Tributyl acetyl citrate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Cholesterol	Nonanal DDT 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Tri-[2-Butoxyethano]lphosphonate Triphenylphosphonate 2-Methyl-1H-indole Trimethylphenylphosphonate (several isomers) Cholesterol Vitamin E acetate Sesquiterpene	1-Methoxypropan-2-ol $\alpha$ -Pinene p-Cymene Nonanal 1-Methoxy-4(prop-2-enyl)benzene Thymol Phthalic anhydride N,N,N',N'-Tetraacetylenediamine (EDTA) Cholesterol

## Denmark

### Other compounds tentatively identified by GC-MS screen

HD02108: Denmark	HD02109: Denmark	HD02110: Denmark
Hexanal 5-Isocyanato-1-(isocyanatomethyl)-1,3,3-trimethylcyclohexane Fluoranthrene Tributyl acetyl citrate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate 2-Methyl-1H-indole	Hexanal Nonanal 2-Methyl-1H-indole N-Propylbenzamide Cholesterol	Nonanal Triacetin Theobromine Bis-(2-ethylhexyl) adipate 2-Methyl-1H-indole 2-ethylhexan-1-ol, ester Piperine Cholesterol

## Sweden

### Other compounds tentatively identified by GC-MS screen

HD02111: Sweden	HD02112: Sweden
Butan-2-one Nonanal Dodecan-1-ol Tri-[2-Butoxyethano]lphosphonate Piperine Cholesterol	Hexanal Styrene Phthalic anhydride N,N,N',N'-Tetraacetylenediamine (EDTA) Tributyl acetyl citrate Piperine Cholesterol Vitamin E acetate

# Region: non UK samples (continued)

## France and Spain Other compounds tentatively identified by GC-MS screen

HD02113: France	HD02114: Spain
Styrene	Methyl methacrylate
2,2'-Oxybis-ethanol	Styrene
Butyl methacrylate	Nonanal
Octanal	Ibuprofen
1,8-Cineole (Eucalyptol)	1-Methyldodecylbenzene
Nonanal	3-(4-Methoxyphenyl)-2-ethylhexylpropenoate
Decanal	Tri-[2-Butoxyethanol]phosphonate
Triethylene glycol	Hexadecyl 2-ethylhexanoate
N,N,N',N'-Tetraacetylenediamine (EDTA)	Cholesta-4,6-dien-3b-ol/Phthalate
N,N-Dimethyltetradecamine	Cholesterol
Hexadecyl 2-ethylhexanoate	
Octadecyl 2-ethylhexanoate	
Cholesterol	
Vitamin E acetate	
Cholesta-3,5-dien-7-one	

## **Annex 2: use, distribution, hazard and regulatory profiles for the five key target groups of chemical contaminants investigated**

# Alkylphenols and their derivatives (APs, APEs)

Alkylphenols (APs), are non-halogenated chemicals manufactured almost exclusively to produce alkylphenol ethoxylates (APEs), a group of non-ionic surfactants. The most widely used APEs are ethoxylates of nonylphenol (NPEs) and, to a lesser extent, octylphenol (OPEs). Once released to the environment, APEs can degrade back to APs, which are persistent, bioaccumulative and toxic to aquatic life.

## Uses

NPEs have been used as surfactants, emulsifiers, dispersants and/or wetting agents in a variety of industrial and consumer applications. Of the 77 000 tonnes used in Western Europe in 1997, the largest share (almost 30%) was used in industrial and institutional cleaning products (detergents), although uses such as emulsifiers (11%), textile finishers (10%), leather finishers (7%) and as components of pesticides and other agricultural products (6%) and water-based paints (5%) were also significant (OSPAR 2001). Moreover, a substantial proportion (16%, or over 12 000 tonnes) was reportedly used in “other niche markets” (including as ingredients in cosmetics, shampoos and other personal care products), or were simply “unaccounted for”. This latter category is thought to include uses in glues and sealants, though information is extremely limited. NP derivatives are reportedly also used as antioxidants in some plastics (Guenther et al. 2002).

OPEs are reported to have a similar range of uses to NPEs, although fewer reliable data are available for this group (OSPAR 2001). For both groups, the extent to which use patterns may have changed over the last 5 years is not well documented.

## Environmental distribution

Both APEs and APs (especially nonylphenol and its derivatives), are widely distributed in fresh and marine waters and in particular, in sediments, in which these persistent compounds accumulate. Because of their releases to water, APEs and APs are also common components of sewage sludge, including that applied to land. Research into levels in wildlife remains very limited, although there have been reports of significant levels in fish and aquatic birds downstream from sites of manufacture and/or use of APEs. Both NP and OP are known to accumulate in the tissues of fish and other organisms, and to biomagnify through the food chain (OSPAR 2001).

Recent research demonstrated the widespread presence of NP in a variety of foods in Germany (Guenther et al. 2002), although the consequences for human exposure have yet to be fully evaluated. The extent and consequences of direct exposure from use in consumer products are also poorly described, although both NP and OP residues have recently been reported as contaminants in house dust (Butte and Heinzow 2002).

## Hazards

The main hazards associated with APEs result from their partial degradation to shorter-chain ethoxylates and to the parent APs themselves (i.e. NP and OP), both of which are

toxic to aquatic organisms. The EU risk assessment for nonylphenol identified significant risks through current uses of NPEs to the aquatic environment, to the soil and to higher organisms through secondary poisoning (i.e. resulting from the accumulation of NP through the food chain, OSPAR 2001). With respect to human exposure through use in consumer products, the EU’s Scientific Committee on Toxicity, Ecotoxicity and the Environment (CSTEE 2001) concluded inter alia that the:-

*“serious lack of measured data for NP in connection with production and use of this compound and its derivatives makes the assessment of both occupational and consumer exposure uncertain”.*

The most widely recognised hazard associated with APs (both NP and OP), is undoubtedly their oestrogenic activity, i.e. their ability to mimic natural oestrogen hormones. This can lead to altered sexual development in some organisms, most notably the feminisation of fish (Jobling et al. 1995, 1996), a factor thought to have contributed significantly to the widespread changes in fish sexual development and fertility in UK rivers (Jobling et al. 2002). Atienzar et al. (2002) recently described direct effects of NP on DNA structure and function in barnacle larvae, a mechanism which may be responsible for the hormone disruption effects seen in whole organisms.

Hazards to human health remain unclear, although recent studies have highlighted concerns directly relevant to humans. For example, Chitra et al. (2002), and Adeoya-Osiguwa et al. (2003), describe effects on mammalian sperm function, while DNA damage in human lymphocytes has also recently been documented (Harreus et al. 2002).

## Existing controls

In 1998, the Ministerial Meeting of OSPAR agreed on the target of cessation of discharges, emissions and losses of all hazardous substances to the marine environment by 2020 (the “one generation” cessation target) and included NP/NPEs on the first list of chemicals for priority action towards this target (OSPAR 1998). Since then, NP has been included as a “priority hazardous substance” under the EU Water Framework Directive, such that action to prevent releases to water within 20 years will be required throughout Europe (EU 2001). A decision on the prioritisation of OP/OPEs under the Directive remains under consideration.

Already, however, the widely recognised environmental hazards presented by AP/APEs have led to some restrictions on use. Of particular note in the European context is the Recommendation agreed by the Paris Commission (now part of the OSPAR Commission) in 1992, which required the phase-out of NPEs from domestic cleaning agents by 1995, and industrial cleaning agents by the year 2000 (PARCOM 1992). However, the precise extent to which this measure has been effective is unclear.



As noted above, the risk assessment conducted under the EU system has concluded that, for NP, there is a need for further risk reduction in some areas, although proposals for restrictions on marketing and use of NP and its derivatives remain under discussion. At the same time, very little information exists regarding the ongoing uses of NP, OP and their derivatives in consumer products and, as a consequence, our direct exposure to them.

- Adeoya-Osiguwa, S.A., Markoulaki, S., Pocock, V., Milligan, S.R. & Fraser, L.R. (2003) 17-beta-estradiol and environmental estrogens significantly effect mammalian sperm function. *Human Reproduction* 18(1): 100-107
- Atienzar, F.A., Billinghurst, Z. & Depledge, M.H. (2002) 4-n-nonylphenol and 17-beta-estradiol may induce common DNA effects in developing barnacle larvae. *Environmental Pollution* 120(3) 735-738
- Butte, W. & Heinzow, B. (2002) Pollutants in house dust as indicators of indoor contamination. *Reviews in Environmental Contamination and Toxicology* 175: 1-46
- Chitra, K.C., Latchoumycandane, C. & Mathur, P.P. (2002) Effect of nonylphenol on the antioxidant system in epididymal sperm of rats. *Archives of Toxicology* 76(9): 545-551
- CSTEE (2001) EC Scientific Committee on Toxicity, Ecotoxicity and the Environment, Opinion on the results of the Risk Assessment of: 4-NONYLPHENOL (Branched) AND NONYLPHENOL - Report version (Human Health effects) : November 2000. Opinion expressed at the 22nd CSTEE plenary meeting, Brussels, 6/7 March 2001: [http://europa.eu.int/comm/food/fs/sc/sct/ou t91\\_en.html](http://europa.eu.int/comm/food/fs/sc/sct/ou t91_en.html)
- EU (2001) Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC, *Official Journal L* 249 , 17/09/2002: 27-30
- Guenther, K., Heinke, V., Thiele, B., Kleist, E., Prast, H. & Raecker, T. (2002) Endocrine disrupting nonylphenols are ubiquitous in food. *Environmental Science and Technology* 36(8): 1676-1680
- Harreus, U.A., Wallner, B.C., Kastenbauer, E.R. & Kleinsasser, N.H. (2002) Genotoxicity and cytotoxicity of 4-nonylphenol ethoxylate on lymphocytes as assessed by the COMET assay. *International Journal of Environmental Analytical Chemistry* 82(6): 395-401
- Jobling, S., Coey, S., Whitmore, J.G., Kime, D.E., van Look, K.J.W., McAllister, B.G., Beresford, N., Henshaw, A.C., Brighty, G., Tyler, C.R. & Sumpter, J.P. (2002) Wild intersex roach (*Rutilus rutilus*) have reduced fertility. *Biology of Reproduction* 67(2): 515-524
- Jobling, S., Reynolds, T., White, R., Parker, M.G. & Sumpter, J.P. (1995) A variety of environmentally persistent chemicals, including some phthalate plasticizers, are weakly estrogenic. *Environmental Health Perspectives* 103(6): 582-587
- Jobling, S., Sheahan, D., Osborne, J.A., Matthiessen, P. & Sumpter, J.P. (1996) Inhibition of testicular growth in rainbow trout (*Oncorhynchus mykiss*) exposed to estrogenic alkylphenolic chemicals. *Environmental Toxicology and Chemistry* 15(2): 194-202
- OSPAR (1998) OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34
- OSPAR (2001) Nonylphenol/nonylphenolethoxylates, OSPAR Priority Substances Series, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London, ISBN 0-946956-79-0: 18 pp.
- PARCOM (1992) PARCOM Recommendation 92/8 on nonylphenol-ethoxylates, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London: 1 p.

# Brominated flame retardants

Brominated flame retardants are a diverse group of organobromine compounds which are used to prevent combustion and/or retard the spread of flames in a variety of plastics, textiles and other materials. Although more than 70 brominated compounds or groups are reportedly in use as flame retardants (Lassen et al. 1999), three chemical groups dominate current usage; the polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) and brominated bisphenols (especially TBBP-A).

## Uses

Brominated flame retardants are used in a wide array of industrial and consumer products including electrical and electronic appliances, vehicles, lighting and wiring, textiles (including carpets and other furnishings), and packaging and insulating materials (especially polystyrene) (Lassen et al. 1999). PBDEs and HBCD are used as additives, whereas TBBP-A is more commonly used as a reactive component, becoming more tightly bound to the polymers in which it is incorporated. Nevertheless, some additive uses do exist for TBBP-A.

Three PBDEs remain in use within the EU; penta-BDE, octa-BDE and deca-BDE. European usage for these additives in 1999 have been estimated at 210 tonnes, 450 tonnes and 7500 tonnes respectively (OSPAR 2001), with deca-BDE (also known as BDE-209) receiving by far the greatest and most diverse use. In the same year, use of HBCD in the EU stood at 9 200 tonnes, around 85% of which was used in rigid polystyrene panels for building insulation (OSPAR 2001). Production of TBBP-A is increasing worldwide; within the EU, estimated uses for 1999 amounted to 13 800 tonnes (BSEF 2000). A further group, the polybrominated biphenyls (PBBs), are no longer produced within Europe, though undoubtedly substantial quantities remain in existing and imported products and in wastes.

## Environmental distribution

The majority of brominated flame retardants are environmentally persistent chemicals. Some, particularly penta-BDE, are highly bioaccumulative but all those listed above are bioavailable and can be measured in the tissues of wildlife and humans. Indeed, their manufacture has led to their widespread and, in some cases, growing presence in the environment.

Although the first reports of their presence in wildlife stem from the early 1980s, the widespread nature of PBDE contamination was only recognised in the early 1990s (Sellström et al. 1993, Jansson et al. 1993). Since then, PBDEs have been reported in almost all environmental compartments, including sediments (Allchin et al. 1999), freshwater and marine fish (Asplund et al. 1999a, b) and even whales from the deep oceans and the Arctic (de Boer et al. 1998, Stern and Ikonomou 2000). Fewer data exist for the other brominated flame retardants in common use, partially because of analytical difficulties, although recent research suggests that HBCD contamination might also be a widespread phenomenon (Allchin and Morris 2002).

PBDEs have also been reported as common contaminants in humans, including reports from Sweden, Spain, Finland and North America (Lindstrom et al. 1997, Meneses et al. 1999, Strandman et al. 1999, She et al. 2000). Concentrations of PBDEs in human breast milk and blood have shown increasing trends over the last two decades (Meironyte et al. 1999, Thomsen et al 2002), and there is some evidence for an upward trend also for TBBP-A. The presence of deca-BDE in human serum, despite its large molecular size, demonstrates its bioavailability.

Although the primary route of exposure is likely to be through foods (especially for the more bioaccumulative PBDEs), other sources of exposure are also likely to be significant, including direct contact with flame-retarded products. PBDEs, HBCD and TBBP-A have all been detected in indoor air and/or dusts in the workplace (Sjödin et al. 2001, Jakobsson et al. 2002) and, to some extent, concentrations in the blood correlate with e.g. contact with computers in the office environment (Hagmar 2000). In our previous study of contaminant levels in dusts from Parliament buildings across Europe, we reported the presence of PBDEs, HBCD and TBBP-A, with deca-BDE and HBCD generally present at the highest concentrations (up to several parts per million, Leonards et al. 2001).

## Hazards

As noted above, brominated flame retardants are generally highly persistent chemicals, some of which are also highly bioaccumulative but all of which are bioavailable. Although their mechanisms of toxicity are gradually being elucidated, their long-term, low-dose toxicity generally remains poorly described.

While their acute toxicity is considered to be low, chronic exposure (especially in the womb) has been shown to interfere with brain and skeletal development in rats (Eriksson et al. 1999), which may in turn lead to permanent neurological effects (Eriksson et al. 2001). Common metabolites of the PBDEs, as well as TBBP-A, are reported to interfere with the binding of thyroid hormones (Meerts et al. 1998, 2001), raising the potential for diverse effects on growth and development. Helleday et al. (1999), report genotoxic effects for both PBDEs and HBCD in mammalian cell lines.

Irrespective of the chemical form of the brominated flame retardant used, incineration of wastes containing these compounds contributes to the formation of brominated dioxins and furans, which exhibit equivalent toxicities to their chlorinated counterparts (IPCS 1998).

## Existing controls

The environmental and human health hazards of brominated flame retardants have been recognised for some time. In 1998, the Ministerial Meeting of OSPAR agreed on the target of cessation of discharges, emissions and losses of all hazardous substances to the marine environment by 2020 (the "one generation" cessation target) and included brominated flame retardants as a group on the first list of

chemicals for priority action towards this target (OSPAR 1998). OSPAR has since reviewed opportunities for action for the PBDEs and HBCD, but is awaiting the outcome of assessments within the EU before developing specific measures (OSPAR 2001). Work on TBBP-A within OSPAR remains ongoing.

Under the EU Existing Substances programme, risk assessments are now complete for two of three PBDEs in common use, penta- and octa- BDE (see e.g. EC 2001) and Europe-wide bans on marketing and use have been agreed for both (EU 2003). While substantial data gaps remain in order to complete the assessment for deca-BDE, EU Member States have nevertheless agreed that risk reduction measures should be “considered without delay” and developed in parallel (EC 2002a).

Even prior to completion of these assessments, the phase out of PBDEs from electrical and electronic equipment by 2006 had already been agreed under the Waste Electrical and Electronic Equipment/Restrictions on Hazardous Substances (WEEE/ROHS) Directive (EC 2002b), which entered into force this year. Their presence in older equipment will, however, remain a problem for waste management for some considerable time to come.

Because of its high persistence and propensity to bioaccumulate, penta-BDE has been proposed for classification as a “priority hazardous substance” under the EU Water Framework Directive (EU 2001), although this remains under discussion. At the same time, penta-BDE is being considered as a case study (Peltola and Yla-Mononen 2001) for addition to the list of persistent organic pollutants (POPs) subject to global control under the 2001 Stockholm Convention developed under the auspices of UNEP (REF), in recognition of its “POP-like” properties.

At a national level, Sweden has proposed for several years the phase-out of PBBs and PBDEs from all applications (KEMI 1999). Very recently, the Norwegian government has adopted an action plan to address brominated flame retardants which includes inter alia proposals for prohibitions of penta-, octa- and deca-BDE and close monitoring of HBCD and TBBP-A (SFT 2003). Even when national and/or regional bans take effect, however, a substantial legacy of all brominated flame retardants will remain in products still in use and/or in the waste stream.

- Allchin, C. & Morris, S. (2002) The determination and occurrence of three groups of brominated flame retardants (polybrominated diphenyl ethers, tetrabromobisphenol A and hexabromocyclododecane) in samples of aquatic origin from the UK. In: Readman, J.; Worsfold, P., eds. Proceedings of ISEAC 32, International Symposium on the Environment and Analytical Chemistry, Plymouth, 17-20 June 2002: 15
- Allchin, C.R., Law, R.J. & Morris, S. (1999) Polybrominated diphenylethers in sediments and biota downstream of potential sources in the UK. *Environmental Pollution* 105: 197-207
- Asplund, L., Athanasiadou, M., Sjödin, A., Bergman, Å. & Borjeson, H. (1999b) Organohalogen substances in muscle, egg and blood from healthy Baltic salmon (*Salmo salar*) and Baltic salmon that produced offspring with the M74 syndrome. *Ambio* 28(1): 67-76
- Asplund, L., Hornung, M., Peterson, R.E., Turesson, K. & Bergman, Å. (1999a) Levels of polybrominated diphenyl ethers (PBDEs) in fish from the Great Lakes and Baltic Sea. *Organohalogen Compounds* 40:351-354
- BSEF (2000) An introduction to brominated flame retardants, Bromine Science and Environment Forum, Brussels, July 2000: 29 pp. <http://www.ebfrp.org/download/weeeqa.pdf>
- de Boer, J., Wester P.G., Klamer H.J.C., Lewis, W.E. & Boon J.P. (1998) Do flame retardants threaten ocean life? *Nature* 394 (2 July): 28-29
- EC (2001) European Union Risk Assessment Report, diphenyl ether, pentabromo derivative ether, 1st Priority List, Volume 5, EUR 19730 EN: 293 pp.
- EC (2002a) European Union Risk Assessment Report, bis(pentabromophenyl) ether, 1st Priority List, Volume 17, EUR 20402 EN: 294 pp.
- EC (2002b) European Community Common Position (EC) No 19/2002 of 4 December 2001 adopted by the Council, acting in accordance with the procedure referred to in Article 251 of the Treaty establishing the European Community, with a view to adopting a Directive of the European Parliament and of the Council on the restrictions of the use of certain hazardous substances in electrical and electronic equipment (RoHS). *Official Journal of the European Communities*, 2002 /C 90/E, Vol. 45: 12-18
- Eriksson, P., Viberg, H., Ankarberg, E., Jakobsson, E., Örn, U. & Fredriksson, A. (2001) Polybrominated diphenylethers (PBDEs): a novel class of environmental neurotoxicants in our environment. In: Asplund, L.; Bergman, Å.; de Wit, C., *et al.* eds. Proceedings of the Second International Workshop on Brominated Flame Retardants, BFR 2001, Stockholm, May 14-16 2001: 71-73
- Eriksson, P., Viberg, H., Jakobsson, E., Örn, U. & Fredriksson, A. (1999) PBDE, 2,2',4,4',5-pentabromodiphenyl ether, causes permanent neurotoxic effects during a defined period of neonatal brain development. *Organohalogen Compounds* 40: 333-336
- EU (2001) Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC, *Official Journal L* 249 , 17/09/2002: 27-30
- EU (2003) Directive 2003/11/EC of the European Parliament and of the Council of 6 February 2003 amending for the 24th time Council Directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (pentabromodiphenyl ether, octabromodiphenyl ether), *Official Journal L* 42, 15.02.2003: 45-46
- Hagmar, L., Jakobsson, K., Thuresson, K., Rylander, L., Sjödin, A. & Bergman, Å. (2000) Computer technicians are occupationally exposed to polybrominated diphenyl ethers and tetrabromobisphenol A. *Organohalogen Compounds* 47: 202-205
- Helleday, T., Tuominen, K.L., Bergman, Å. & Jenssen, D. (1999) Brominated flame retardants induce transgenic recombination in mammalian cells. *Mutation Research – Genetic Toxicology and Environmental Mutagenesis* 439(2): 137-147
- Ikonomou, M.G., Rayne, S. & Addison, R.F. (2002) Exponential increases of the brominated flame retardants, polybrominated diphenyl ethers, in the Canadian Arctic from 1981 to 2000. *Environmental Science and Technology* 36(9): 1886-1892
- IPCS (1998) Polybrominated dibenzo-p-dioxins and dibenzofurans, *Environmental Health Criteria*, No. 205, International Programme on Chemical Safety, UNEP/ILO/WHO, ISBN 92 4 157205 1: 303 pp.
- Jakobsson, K., Thuresson, K., Rylander, L., Sjödin, A., Hagmar, L. & Bergman, Å. Exposure to polybrominated diphenyl ethers and tetrabromobisphenol A among computer technicians. *Chemosphere* 46(5): 709-716
- Jansson, B., Andersson, R., Asplund, L., Litzen, K., Nylund, K., Sellström, U., Uvemo, U.-B., Wahlberg, C., Wideqvist, U., Odsjo, T. & Olsson, M. (1993) Chlorinated and brominated persistent organic compounds in biological samples from the environment. *Environmental Toxicology and Chemistry* 12(7): 1163-1174
- KEMI (1999) Phase-out of PBDEs and PBBs: Report on a Governmental Commission, The Swedish National Chemicals Inspectorate, 15th March 1999: 34 pp.
- Lassen, C., Lokke, S. & Hansen, L.I. (1999) Brominated Flame Retardants: substance flow analysis and substitution feasibility study. Danish Environmental Protection Agency Environmental Project No. 494, Copenhagen, ISBN 87-7909-415-5: 240 pp.
- Leonards, P.E.G., Santillo, D., Bridgen, K., van der Ween, I., Hesseltingen, J.v., de Boer, J. & Johnston, P. (2001) Brominated flame retardants in office dust samples. In: Asplund, L.; Bergman, Å.; de Wit, C., *et al.* eds. Proceedings of the Second International Workshop on Brominated Flame Retardants, BFR 2001, Stockholm, May 14-16 2001: 299-302
- Lindstrom, G., van Bavel, B., Hardell, L. & Liljegren, G. (1997) Identification of the flame retardants polybrominated diphenyl ethers in adipose tissue from patients with non-Hodgkin's lymphoma in Sweden. *Oncology Reports* 4(5): 999-1000
- Meerts, I.A.T.M., Letcher, R.J., Hoving, S., Marsh, G., Bergman, Å., Lemmen, J.G., van der Burg, B. & Brouwer, A. (2001) In vitro estrogenicity of polybrominated diphenyl ethers, hydroxylated PBDEs and polybrominated bisphenol A compounds. *Environmental Health Perspectives* 109(4): 399-407
- Meerts, I.A.T.M., Marsh, G., van Leeuwen-Bol, I., Luijckx, E.A.C., Jakobsson, E., Bergman, Å. & Brouwer, A. (1998) Interaction of polybrominated diphenyl ether metabolites (PBDE-OH) with human transthyretin in vitro. *Organohalogen Compounds* 37: 309-312
- Meironyte, D., Noren, K. & Bergman, Å. (1999) Analysis of polybrominated diphenyl ethers in Swedish human milk. A time-related trend study, 1972-1997. *Journal of Toxicology and Environmental Health - Part A* 58(6): 329-341
- Meneses, M., Wingfors, H., Schuhmacher, M., Domingo, J.L., Lindstrom, G. & van Bavel, B. (1999) Polybrominated diphenyl ethers detected in human adipose tissue from Spain. *Chemosphere* 39(13): 2271-2278
- OSPAR (1998) OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34
- OSPAR (2001) Certain Brominated Flame Retardants – Polybrominated Diphenylethers, Polybrominated Biphenyls, Hexabromocyclododecane, OSPAR Priority Substances Series, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London: 25pp.
- Peltola, J. & Yla-Mononen, L. (2001) Pentabromodiphenyl ether as a global POP. *TemaNord* 2001:579, Nordic Council of Ministers, Copenhagen, ISBN 92-893-0690-4: 78 pp.
- Sellström, U., Jansson, B., Kierkegaard, A., de Wit, C., Odsjo, T. & Olsson, M. (1993) Polybrominated diphenyl ethers (PBDE) in biological samples from the Swedish environment. *Chemosphere* 26(9): 1703-1718
- SFT (2003) Norwegian Pollution Control Authority Press Release, <http://www.sft.no/english/news/dbafile8520.html>.
- She, J., Winkler, J., Visita, P., McKinney, M. & Petreas, M. (2000) Analysis of PBDEs in seal blubber and human breast adipose tissue samples. *Organohalogen Compounds* 47: 53-56
- Sjödin, A., Carlsson, H., Thuresson, K., Sjölin, S., Bergman, Å. & Ostman, C. (2001) Flame retardants in indoor air at an electronics recycling plant and at other work environments. *Environmental Science and Technology* 35(3): 448-454
- Strandman, T., Koistinen, J., Kiviranta, H., Vuorinen, P.J., Tuomisto, J. & Vartiainen, T. (1999) Levels of some polybrominated diphenyl ethers (PBDEs) in fish and human adipose tissue in Finland. *Organohalogen Compounds* 40:355-358
- Thomsen, C., Lundanes, E. & Becher, G. (2002) Brominated flame retardants in archived serum samples from Norway: A study on temporal trends and the role of age. *Environmental Science and Technology* 36(7): 1414-1418
- UNEP (2001) United Nations Environment Programme, Final Act of the Conference of Plenipotentiaries on the Stockholm Convention on Persistent Organic Pollutants, UNEP/POPS/CONF/4, 5 June 2001: 44 pp.

# Organotin compounds

Organotins are organic compounds containing at least one bond between carbon and the metal tin. By far the best known is tributyltin (TBT) which, as a result of its widespread use in antifouling paints on ships and boats, has led to widespread changes in sexual development in marine snails. However, several other organotin compounds are in common use, most notably mono- and dibutyltin (MBT, DBT), octyltins (MOT, DOT) and triphenyltins (TPT).

## Uses

As noted above, TBT has been used for many years as an antifouling agent for ship paints. Its use on small vessels (<25m) has been banned in many countries for more than 10 years, following the devastating impacts on populations of oysters and other marine molluscs (Santillo *et al.* 2001a). Its use is still currently permitted on larger vessels, although this is now subject to phase-out (see below).

Although antifouling paints have accounted for the majority of TBT used, this compound is also used as an antifungal agent in some consumer products, including certain carpets, textiles and PVC (vinyl) flooring (Allsopp *et al.* 2000, 2001). Most abundant in consumer products, however, are MBT and DBT, used as heat stabilisers in rigid (pipes, panels) and soft (wall-coverings, furnishings, flooring, toys) PVC products and in certain glass coating applications (Matthews 1996). PVC represents about two-thirds of the global consumption of these compounds (Sadiki and Williams 1999), which can comprise up to 2% by weight of the finished product. Mono- and dioctyl tins (MOT, DOT) are also used as PVC stabilisers, primarily in food contact applications. Kawamura *et al.* (2000) reported levels up to the g/kg range for MOT in PVC containers. According to industry figures ([www.ortepa.org](http://www.ortepa.org)), approximately 15 000 tonnes of organotins were used as PVC in Europe in 1995.

## Environmental distribution

Much of the research describing the environmental distribution of organotin compounds has, understandably, focused on the spread of TBT and its break-down products (including DBT) in the marine environment. The global use of TBT antifouling paints has resulted in contamination on a global scale. The relative persistence of butyl tins, combined with their affinity for biological tissues, has led to their widespread occurrence in fish, seals, whales and dolphins in all major sea areas (Iwata *et al.* 1995, Kannan *et al.* 1996, Ariese *et al.* 1998).

Much less information is available concerning the distribution of organotins in other environmental compartments. In one of the few studies which have been conducted, Takahashi *et al.* (1999) reported the presence of butyltin residues in the livers of monkeys and other mammals in Japan, as well as in human livers, and suggested that uses in consumer products may represent an important exposure route. The presence of organotin compounds in a wide range of construction and consumer products, especially PVC products, has been highlighted above. It has also been recognised for some time that butyltin stabilisers can migrate from such products during normal use (Sadiki and Williams 1999).

A recent study in Germany raised concern about the presence of comparatively high levels of TBT and other organotins in PVC flooring (Oeko-Test 2000). The data of Allsopp *et al.* (2000, 2001) for both PVC flooring and carpets available for retail in the UK confirm the ongoing use of these compounds in floor coverings, occasionally at very high concentrations (up to 0.57 g/kg DBT in PVC, 0.047 g/kg TBT in treated carpet fibre). Such uses undoubtedly contribute to the widespread presence of organotin compounds in dusts from the indoor environment (see e.g. Santillo *et al.* 2001b).

## Hazards

Organotins are known to be toxic at relatively low levels of exposure not only to marine invertebrates but also in mammals. In marine invertebrates, TBT is generally more toxic than DBT, which is in turn more toxic than MBT (Cima *et al.* 1996). However, this is by no means always the case, as DBT is more toxic than TBT to certain enzyme systems (Bouchard *et al.* 1999, Al-Ghais *et al.* 2000). In fish, DBT is frequently a more potent toxin than TBT (O'Halloran *et al.* 1998), with the immune system the primary target.

Organotins have been demonstrated to have immunotoxic and teratogenic (developmental) properties also in mammalian systems (Kergosien and Rice 1998), with DBT again frequently appearing more toxic than TBT (Ema *et al.* 1995, De Santiago and Aguilar-Santelises 1999). DBT is neurotoxic to mammalian brain cells (Eskes *et al.* 1999). Ema *et al.* (1996, 1997), demonstrated the importance of the precise timing of exposure to DBT in induction of defects in developing rat embryos. Very recently, Kumasaka *et al.* (2002) have described toxic effects on testes development in mice.

Estimates of the significance of human exposure to organotins from consumption of contaminated seafood have taken the potential immunotoxicity of these compounds to humans as an effect parameter (Belfroid *et al.* 2000). While seafood probably remains the predominant source of organotin exposure for many consumers, exposure to consumer products which contain them or to dusts in the home may also be significant.

## Existing controls

To date, legislative controls on organotin compounds have focused primarily on TBT in antifouling paints. A series of national bans on the use on small vessels, starting in France and the UK, was followed by an EU wide ban on vessels less than 25m in length in 1991 (Evans 2000). More recently, the International Maritime Organisation (IMO) agreed on a global phase-out of all TBT applications (from January 2003) and TBT presence on ships (from 2008) under its Convention on Harmful Anti-fouling Systems (see [www.imo.org](http://www.imo.org)). The first of these deadlines has recently been transposed into EU law (EU 2002a).

At the same time, and despite the toxicity to mammals noted above, TBT continues to be used as an additive in some consumer products, as do uses of other butyltins and octyltins. Organotin compounds must not be used for certain

textiles to qualify for an “eco-label” within the EU (EU 2002b), but there are otherwise no restrictions on use unless the treated materials or products are used in contact with water. This is despite the fact that TBT is classified under the EU's labelling Directive as “harmful in contact with skin, toxic if swallowed, irritating to the eyes and skin” and as presenting a “danger of serious damage to health by prolonged exposure through inhalation or if swallowed”.

In 2001, Germany notified the European Commission of its intention to introduce stricter controls for organotins, including controls on use in consumer products. However, such controls were rejected by the Commission as “inadmissible” (EC 2002).

In 1998, the Ministerial Meeting of OSPAR agreed on the target of cessation of discharges, emissions and losses of all hazardous substances to the marine environment by 2020 (the “one generation” cessation target) and included organotin compounds on the first list of chemicals for priority action towards this target (OSPAR 1998). Initially, OSPAR's action focused on the achievement of the IMO Convention on Harmful Anti-fouling Systems (OSPAR 2000). In 2001, OSPAR began to consider the scope for action on other uses and organotin compounds, including the widespread use of butyltin stabilisers, though so far, no further measures are proposed.

- Al-Ghais, S.M. & Ahmad, A.B. (2000) Differential inhibition of xenobiotic-metabolizing carboxylesterases by organotins in marine fish. *Ecotoxicology and Environmental Safety* 46(3): 258-264
- Allsopp, A., Santillo, D. & Johnston, P. (2001) Hazardous chemicals in carpets. Greenpeace Research Laboratories Technical Note 01/2001, January 2001: 14 pp. [published under cover title "Poison Underfoot: Hazardous Chemicals in PVC Flooring and Hazardous Chemicals in Carpets, ISBN 90-73361-68-0]
- Allsopp, M., Santillo, D. & Johnston, P. (2000). Hazardous chemicals in PVC flooring. Greenpeace Research Laboratories Technical Note 14/00, November 2000: 10 pp. [published under cover title "Poison Underfoot: Hazardous Chemicals in PVC Flooring and Hazardous Chemicals in Carpets, ISBN 90-73361-68-0]
- Ariese, F., van Hattum, B., Hopman, G., Boon, J. & ten Hallers-Tjabbes, C. (1998) Butyltin and phenyltin compounds in liver and blubber samples of sperm whales (*Physeter macrocephalus*) stranded in the Netherlands and Denmark. Institute for Environmental Studies, Vrije Universiteit, Amsterdam, Report W98-04, March 1998
- Belfroid, A.C., Purperhart, M. & Ariese, F. (2000) Organotin levels in seafood. *Marine Pollution Bulletin* 40(3): 226-232
- Bouchard, N., Pelletier, E. & Fournier, M. (1999) Effects of butyltin compounds on phagocytic activity of hemocytes from three marine bivalves. *Environmental Toxicology and Chemistry* 18(3): 519-522
- Cima, F., Ballarin, L., Bressa, G., Martinucci, G. & Burighel, P. (1996) Toxicity of organotin compounds on embryos of a marine invertebrate (*Styela plicata*; Tunicata). *Ecotoxicology and Environmental Safety* 35(2): 174-182
- de Santiago, A. & Aguilar-Santelises, M. (1999) Organotin compounds decrease *in vitro* survival, proliferation and differentiation of normal human B lymphocytes. *Human and Experimental Toxicology* 18(10): 619-624
- EC (2002) Commission Decision 2001/570/EC of 13 July 2001 on draft national provisions notified by the Federal Republic of Germany on limitations on the marketing and use of organostannic compounds. *Official Journal L* 202, 27/07/2001: 37-45
- Ema, M., Harazono, A., Miyawakai, E. & Ogawa, Y. (1997) Effect of the day of administration on the developmental toxicity of tributyltin chloride in rats. *Archives of Environmental Contamination and Toxicology* 33(1): 90-96
- Ema, M., Iwase, T., Iwase, Y., Ohyama, N. & Ogawa, Y. (1996) Change of embryotoxic susceptibility to di-n-butyltin chloride in cultured rat embryos. *Archives of Toxicology* 70(11): 742-748
- Ema, M., Kurosaka, R., Amano, H. & Ogawa, Y. (1995) Comparative developmental toxicity of butyltin trichloride, dibutyltin dichloride and tributyltin chloride in rats. *Journal of Applied Toxicology* 15(4): 297-302
- Eskes, C., Honegger, P., Jones-Lepp, T., Varner, K., Matthieu, J.M. & Monnet-Tschudi, F. (1999) Neurotoxicity of dibutyltin in aggregating brain cell cultures. *Toxicology In Vitro* 13(4-5): 555-560
- EU (2002a) Commission Directive 2002/62/EC of 9th July 2002 adapting to technical progress for the ninth time Annex 1 to Council Directive 76/769/EEC on the approximations of the laws, regulations and administrative provisions of the member States relating to restrictions on the marketing and use of certain dangerous substances and preparations (organostannic compounds). *Official Journal L* 183, 12.7.2002: 58-59
- EU (2002b) Commission Decision 2002/371/EC of 15 May 2002 establishing the ecological criteria for the award of the Community eco-label to textile products and amending Decision 1999/178/EC. *Official Journal L* 133, 18/05/2002: 29-41
- Evans, S.M. (2000) Marine antifoulants. In: C. Sheppard [Ed.], *Seas at the Millennium: An Environmental Evaluation, Volume III: Global Issues and Processes*, Elsevier Science Ltd, Oxford, ISBN: 0-08-043207-7, Chapter 124: 247-256
- Iwata, H., Tanabe, S., Mizuno, T. and Tatsukawa, R. (1995) High accumulation of toxic butyltins in marine mammals from Japanese coastal waters. *Environmental Science and Technology* 29: 2959-2962
- Kannan, K., Corsolini, S., Focardi, S., Tanabe, S. & Tatsukawa, R. (1996) Accumulation pattern of butyltin compounds in dolphin, tuna and shark collected from Italian coastal waters. *Archives of Environmental Contamination and Toxicology* 31: 19-23
- Kawamura, Y., Machara, T., Suzuki, T. & Yamada, T. (2000) Determination of organotin compounds in kitchen utensils, food packages and toys by gas chromatography/atomic emission detection method. *Journal of the Food Hygienic Society of Japan* 41(4): 246-253
- Kergosien D.H. and Rice C.D. (1998). Macrophage secretory function is enhanced by low doses of tributyltin-oxide (TBTO), but not tributyltin-chloride (TBTC). *Arc. Environ. Contam. Toxicol.* 34: 223-228
- Kumasaka, K., Miyazawa, M., Fujimaka, T., Tao, H., Ramaswamy, B.R., Nakazawa, H., Makino, T. & Satoh, S. (2002) Toxicity of the tributyltin compound on the testis in premature mice. *Journal of Reproduction and Development* 48(6): 591-597
- Matthews, G. (1996) *PVC: Production, Properties and Uses*. The Institute of Materials, London: 379 pp.
- O'Halloran, K., Ahokas, J.T. & Wright, P.F.A. (1998) Response of fish immune cells to *in vitro* organotin exposures. *Aquatic Toxicology* 40(2-3): 141-156
- Oeko-Test (2000). *Sondermüll im Haus. öko-test magazine* 5/2000: 74-79
- OSPAR (1998) *OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34*
- OSPAR (2000) *OSPAR Background Document on Organic Tin Compounds, OSPAR Priority Substances Series, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London, ISBN 0-946956-56-1: 16pp.*
- Sadiki A-I. and Williams D.T. (1999). A study on organotin levels in Canadian drinking water distributed through PVC pipes. *Chemosphere* 38 (7): 1541-1548
- Santillo, D., Johnston, P. & Langston, W.J. (2001a) Tributyltin (TBT) antifoulants: a tale of ships, snails and imposex. Chapter 13 in: P. Harremoës, D. Gee, M. MacGarvin, A. Stirling, J. Keys, B. Wynne, S.G. Vaz [eds], *Late Lessons from Early Warnings: the precautionary principle 1896-2000*, European Environment Agency Environmental Issue Report No 22, Office for Official Publications of the European Communities, Luxembourg, ISBN 92-9167-323-4: 135-148
- Santillo, D., Johnston, P. & Brigden, K. (2001b) The presence of brominated flame retardants and organotin compounds in dusts collected from Parliament buildings from eight countries. Greenpeace Research Laboratories Technical Note 03/2001, March 2001: 24 pp.
- Takahashi, S., Mukai, H., Tanabe, S., Sakayama, K., Miyazaki, T. & Masuno, H. (1999) Butyltin residues in livers of humans and wild terrestrial mammals and in plastic products. *Environmental Pollution* 106: 213-218



# Phthalates (phthalate esters)

Phthalates are non-halogenated ester derivatives of phthalic acid which are widely used in a range of industrial and consumer applications. Some are marketed as discreet chemical products (e.g. the well-known di(ethylhexyl) phthalate or DEHP), while others are complex isomeric mixtures comprising many individual compounds with similar chemical structures (e.g. di-iso-nonyl phthalate, DINP, and di-iso-decyl phthalate, DIDP). As a result of their high volume uses in open applications, they are now among the most ubiquitous man-made chemicals found in the environment.

## Uses

Phthalates have a range of applications, dependent on the precise chemical form, although by far their greatest use is as plasticising (softening) additives in flexible plastics, especially PVC. They are produced in very large quantities in Europe, almost 1 million tonnes per year, primarily for use within the EU. For example, estimated production volumes in the mid-1990s were 595 000 tonnes DEHP, 185 000 tonnes DINP and around 200 000 tonnes for DIDP (CSTEE 2001a, b, 2002).

Of these three main phthalates, over 90% of use is in PVC applications, including toys, flooring and other building/furnishing materials, car interiors, cables and medical equipment (see e.g. <http://www.ecpi.org/plasticisers/index.html>). Minor applications include use as components of inks, adhesives, paints, sealants and surface coatings. Other phthalates, including di(butyl) phthalate (DBP) and di(ethyl) phthalate (DEP), have also been used as PVC additives, but are also used as solvents and fixatives in perfumes and as ingredients in other cosmetics (Koo et al. 2002).

## Environmental distribution

All uses of phthalates, especially the major use as PVC plasticisers, result in large-scale losses to the environment (both indoors and outdoors) during the lifetime of products, and again following disposal (amounting to thousands of tonnes per year across the EU, CSTEE 2001a). As a consequence, phthalates have long been recognised as one of the most abundant and widespread man-made environmental contaminants (Mayer et al. 1972) and our exposure to phthalates is therefore widespread and continuous.

Although some degradation is possible, phthalates are considered to be relatively persistent, especially in soils and sediments. They also have the inherent ability to accumulate in biological tissues, although continuous exposure undoubtedly also contributes to tissue levels. Risk assessments conducted under the EU system have documented the widespread distribution of phthalates in all environmental compartments (e.g. see CSTEE 2001c, d). A number of recent studies have reported the presence of phthalates and their primary metabolites in the human body (Colon et al. 2000, Blount et al. 2000).

Because of their extensive use in building materials and household products, phthalates are common contaminants in indoor air (Otake et al. 2001, Wilson et al. 2001). They have

also been reported as substantial components of house dust, in some cases at more than 1 part per thousand (1g/kg) of the total mass of dust (Butte and Heinzow 2002).

## Hazards

As noted above, phthalates are relatively persistent in the environment and can bioaccumulate. Substantial concerns also exist with regard to their toxicity to wildlife and to humans, although the precise mechanisms and levels of toxicity vary from one compound to another. In many cases, it is the metabolites of the phthalates which are responsible for the greatest toxicity (e.g. Dalgaard et al. 2001).

EU risk assessments for DEHP, DINP and DIDP concluded that there were no significant risks to aquatic or terrestrial organisms. However, the EU's Scientific Committee on Toxicity, Ecotoxicity and the Environment (CSTEE 2001c, d) has disagreed with this conclusion for the terrestrial environment, noting that there is very little evidence to justify such a conclusion. The CSTEE has also highlighted concerns relating to secondary poisoning, i.e. the build up of phthalates through the food chain.

With respect to humans, although substantial exposure can occur through contaminated food, direct exposure to phthalates from consumer products and/or medical devices is likely to be very significant. Perhaps the best known example is the exposure of children to phthalates used in soft PVC teething toys (see e.g. Stringer et al. 2000), now subject to emergency controls within Europe (see below).

DEHP, still the most widely used phthalate in Europe, is a known reproductive toxin, interfering with testes development in mammals, and is classified in the EU as "toxic to reproduction". Indeed, its toxicity to the developing male reproductive system has been recognised for more than 50 years (Park et al. 2002). Observed toxicity is due mainly to the compound MEHP, formed in the body as a metabolite of DEHP, and appears to impact on many aspects of development and liver function, including hormone metabolism and immune function (Dalgaard et al. 2001, Wong and Gill 2002). Other recent studies have reaffirmed the reproductive toxicity of several other commonly used phthalates, including butylbenzyl phthalate (BBP) and dibutyl phthalate (DBP) (Ema and Miyawaki 2002, Mylchreest et al. 2002). As for DEHP, DBP is classified in the EU as "toxic to reproduction".

Reproductive toxicity is generally thought to be of lower concern for the other widely used phthalates DINP and DIDP, although Gray et al. (2000) did report evidence for abnormal sexual development in rats exposed to DINP. Prior to this, Harris et al. (1997) reported the weak oestrogenicity of several phthalates, including DINP. Other concerns for DINP and DIDP relate primarily to toxic effects on the liver and kidney. Very recent research suggests possible effects on human sperm development for DEP (Duty et al. 2003), a phthalate widely used in cosmetics and perfumes and, until now, considered to be of relatively little toxicological significance.

In the indoor environment, correlations have been reported between incidence of bronchial obstruction (asthma) in children and the abundance of phthalate-containing materials in the home (Oie et al 1997).

### **Existing controls**

At present, there are few controls on the marketing and use of phthalates, despite their toxicity, the volumes used and their propensity to leach out of products throughout their lifetime. Of the controls which do exist, probably the best known is the EU-wide emergency ban on the use of six phthalates in children's toys designed to be chewed (first agreed in 1999 and recently renewed for the 13th time, EU 2003). While this ban addressed one important exposure route, exposures through other toys and, indeed, other consumer products, as well as through PVC medical devices, remain unaddressed.

Following the conclusion of the EU risk assessment for DEHP, proposals have now been made for a ban on uses in certain medical devices and tight restrictions on other uses, though these remain under discussion at EU level. No formal proposals have yet been made for the other phthalates undergoing assessment within the EU.

In 1998, the Ministerial Meeting of OSPAR agreed on the target of cessation of discharges, emissions and losses of all hazardous substances to the marine environment by 2020 (the "one generation" cessation target) and included the phthalates DBP and DEHP on the first list of chemicals for priority action towards this target (OSPAR 1998). DEHP is also proposed as a "priority hazardous substance" under the EU Water Framework Directive (EU 2001), such that action to prevent releases to water within 20 years will be required throughout Europe, though a decision on this classification remains under consideration.

- Blount, B.C., Silva, M.J., Caudill, S.P., Needham, L.L., Pirkle, J.L., Sampson, E.J., Lucier, G.W., Jackson, R.J. & Brock, J.W. (2000) Levels of seven urinary phthalate metabolites in a human reference population. *Environmental Health Perspectives* 108(10): 979-982
- Butte, W. & Heinzow, B. (2002) Pollutants in house dust as indicators of indoor contamination. *Reviews in Environmental Contamination and Toxicology* 175: 1-46.
- Colon, I., Caro, D., Bourdony, C.J. & Rosario, O. (2000) Identification of phthalate esters in the serum of young Puerto Rican girls with premature breast development. *Environmental Health Perspectives* 108(9): 895-900
- CSTEE (2001a) EC Scientific Committee on Toxicity, Ecotoxicity and the Environment, Opinion on the results of the Risk Assessment of: 1,2-Benzenedicarboxylic acid, di-C8-10-branched alkyl esters, C9-rich and di-"isononyl" phthalate - Report version (Human Health Effects): Final report, May 2001. Opinion expressed at the 27th CSTEE plenary meeting, Brussels, 30 October 2001: 7 pp. [http://europa.eu.int/comm/food/fs/sc/sct/out120\\_en.pdf](http://europa.eu.int/comm/food/fs/sc/sct/out120_en.pdf)
- CSTEE (2001b) Scientific Committee on Toxicity, Ecotoxicity and the Environment (European Commission), Opinion on the results of the Risk Assessment of: 1,2-Benzenedicarboxylic acid di-C9-11-branched alkyl esters, C10-rich and di-"isodecyl"phthalate - Report version (Human health effects): Final report, May 2001. Opinion expressed at the 24th CSTEE plenary meeting, Brussels, 12 June 2001, [http://europa.eu.int/comm/food/fs/sc/sct/out103\\_en.html](http://europa.eu.int/comm/food/fs/sc/sct/out103_en.html)
- CSTEE (2001c) EC Scientific Committee on Toxicity, Ecotoxicity and the Environment, Opinion on the results of the Risk Assessment of: 1,2-Benzenedicarboxylic acid, di-C8-10-branched alkyl esters, C9-rich and di-"isononyl" phthalate - Report version (Environment): Final report, May 2001. Opinion expressed at the 27th CSTEE plenary meeting, Brussels, 30 October 2001: 5 pp. [http://europa.eu.int/comm/food/fs/sc/sct/out122\\_en.pdf](http://europa.eu.int/comm/food/fs/sc/sct/out122_en.pdf)
- CSTEE (2001d) Scientific Committee on Toxicity, Ecotoxicity and the Environment (European Commission), Opinion on the results of the Risk Assessment of: 1,2-Benzenedicarboxylic acid di-C9-11-branched alkyl esters, C10-rich and di-"isodecyl"phthalate - Report version (Environment): Final report, May 2001. Opinion expressed at the 24th CSTEE plenary meeting, Brussels, 12 June 2001, 5 pp. [http://europa.eu.int/comm/food/fs/sc/sct/out121\\_en.pdf](http://europa.eu.int/comm/food/fs/sc/sct/out121_en.pdf)
- CSTEE (2002) EC Scientific Committee on Toxicity, Ecotoxicity and the Environment, Opinion on the results of the Risk Assessment of Bis (2-ethylhexyl) phthalate (DEHP). Report version: Human Health, September 2001. Opinion expressed at the 29th CSTEE plenary meeting, Brussels, 09 January 2002: 8 pp. [http://europa.eu.int/comm/food/fs/sc/sct/out141\\_en.pdf](http://europa.eu.int/comm/food/fs/sc/sct/out141_en.pdf)
- Dalgaard, M., Nellemann, C., Lam, H.R., Sorensen, I.K. & Ladefoged, O. (2001) The acute effects of mono(2-ethylhexyl)phthalate (MEHP) on testes of prepubertal Wistar rats. *Toxicology Letters* 122: 69-79
- Duty, S.M., Singh, N.P., Silva, M.J., Barr, D.B., Brock, J.W., Ryan, L., Herrick, R.F., Christiani, D.C. & Hauser, R. (2003) The relationship between environmental exposures to phthalates and DNA damage in human sperm using the neutral comet assay. *Environmental Health Perspectives* (in press)
- Ema, M. & Miyawaki, E. (2002) Effects on development of the reproductive system in male offspring of rats given butyl benzyl phthalate during late pregnancy. *Reproductive Toxicology* 16: 71-76
- EU (2001) Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC, Official Journal L 249 , 17/09/2002: 27-30
- EU (2003) Commission Decision 2003/113/EC amending Decision 1999/815/EC concerning measures prohibiting the placing on the market of toys and childcare articles intended to be placed in the mouth by children under three years of age made of soft PVC containing certain phthalates. Official Journal L 46, 20.2.2003: 27-28
- Gray, L.E., Ostby, J., Furr, J., Price, M., Veeramachaneni, D.N.R. & Parks, L. (2000) Perinatal exposure to the phthalates DEHP, BBP and DINP, but not DEP, DMP or DOTP, alters sexual differentiation of the male rat. *Toxicological Sciences* 58(2): 350-365
- Harris C.A., Henttu, P., Parker, M.G. & Sumpter, J.P. (1997) The estrogenic activity of phthalate esters in vitro *Environmental Health Perspectives* 105 (8): 802-811
- Koo J-W, Parham F, Kohn MC, Masten SA, Brock JW, Needham LL, *et al.* 2002. The association between biomarker-based exposure estimates for phthalates and demographic factors in a human reference population. *Environmental Health Perspectives* 110:405-410
- Mayer, F.L., Stalling, D.L. & Johnson, J.L. (1972) Phthalate esters as environmental contaminants. *Nature* 238: 411-413
- Mylchreest, E., Sar, M., Wallace, D.G. & Foster, P.M.D. (2002) Fetal testosterone insufficiency and abnormal proliferation of Leydig cells and gonocytes in rats exposed to di(n-butyl) phthalate. *Reproductive Toxicology* 16: 19-28
- Oie, L., Hersoug, L.G. & Madsen, J.O. (1997) Residential exposure to plasticizers and its possible role in the pathogenesis of asthma. *Environmental Health Perspectives* 105 (9): 972-978
- OSPAR (1998) OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34
- Otake, T., Yoshinaga, J. & Yanagisawa, Y. (2001) Analysis of organic esters of plasticizer in indoor air by GC-MS and GC-FPD. *Environmental Science and Technology* 35(15): 3099-3102
- Park, J.D., Habeebu, S.S.M. & Klaassen, C.D. (2002) Testicular toxicity of di-(2-ethylhexyl)phthalate in young Sprague-Dawley rats. *Toxicology* 171: 105-115
- Stringer, R., Labunska, I, Santillo, D., Johnston, P., Siddorn, J. & Stephenson, A. (2000) Concentrations of phthalate esters and identification of other additives in PVC children's toys. *Environmental Science and Pollution Research* 7(1): 27-36
- Wilson, N.K., Chuang, J.C. & Lyu, C. (2001) Levelsof persistent organic pollutants in several child day care centres. *Journal of Exposure Analysis and Environmental Epidemiology* 11(6): 449-458
- Wong, J.S. & Gill, S.S. (2002) Gene expression changes induced in mouse liver by di(2-ethylhexyl) phthalate. *Toxicology and Applied Pharmacology* 185(3): 180-196

# Short-Chain Chlorinated Paraffins (SCCPs)

Chlorinated paraffins are organochlorine chemicals simply produced by reacting chlorine gas with paraffins (hydrocarbons). Short-chain chlorinated paraffins, or SCCPs, are those which have a carbon backbone of between 10 and 13 carbon atoms (C10-C13).

## Uses

SCCPs have been used in a wide range of industrial and consumer applications, including use as components of industrial cutting oils for metal working, as flame retardants or other additives in rubbers, paints and sealants and as finishing agents for leather goods and certain textiles (OSPAR 2001). To some extent, SCCPs were used as replacements for PCBs (polychlorinated biphenyls) when these were phased out.

In 1994, it was estimated that of a total of 13 200 tonnes of SCCPs used in the EU, more than 70% were used in metal working applications. By 1998, the total had declined to just over 4 000 tonnes, mainly as a result of reductions in this main use (OSPAR 2001). In 1994, there were two production facilities within the EU, Hoechst in Germany and ICI in the UK. Hoechst has since ceased production of SCCPs (Koh *et al.* 2001).

However, uses in paints, coatings and sealants (726 tonnes) and as flame retardants in rubbers (638 tonnes) had declined to a lesser extent. Moreover, quantities used for a range of other unspecified sectors increased from 100 tonnes in 1994 to 648 tonnes in 1998 (OSPAR 2001). At the same time, quantities imported to the EU as additives in finished products are simply not known, though they are likely to be substantial. There is also likely to be a large reservoir of SCCPs in existing consumer products and in the waste-stream within the EU, though again there is very little information on this. The recent work of Koh *et al.* (2002), which identified SCCPs in some window and door seals in office buildings in Germany, is one of very few studies available.

## Environmental distribution

SCCPs are persistent organic pollutants with a high potential to accumulate in biological tissues. Because of the way in which they are produced, SCCPs are complex mixtures of individual chemicals, varying in chain length and degree of chlorination. This makes their study very difficult, such that data on distribution and effects still remain limited.

Nevertheless, SCCPs have been detected in a range of freshwater (mussels, fish), marine (fish, seals, whales) and terrestrial (rabbits, moose, osprey) organisms and in humans (Stern and Tomy 2000). As a result of their persistence and ability to be carried on air-currents, they are now widespread environmental contaminants, even appearing in remote areas of the Arctic (Tomy *et al.* 1999). Recent research has found that SCCPs are also widespread contaminants in the air in the UK (Peters *et al.* 2000), despite earlier assumptions used in risk assessments that any concentrations in the atmosphere would be "very small". No published levels could be found for household dusts.

## Hazards

SCCPs are very toxic to fish and other aquatic organisms, and have been shown to cause damage to the liver, kidney and thyroid in rats following long-term exposure in the laboratory (Farrar 2000). Information on impacts of long-term low level exposure remains very limited (Fisk *et al.* 1999). Because of the known hazards, however, SCCPs have been classified as "Category 3" carcinogens ("possible risk of irreversible effects") and as "Dangerous for the Environment" ("very toxic to aquatic organisms, may cause long-term adverse effects in the aquatic environment"), under the EC's Classification and Labelling Directive. The primary exposure route for humans is likely to be through food, although the significance of other routes (including contact with products, inhalation in the indoor environment and contact with contaminated dusts), has never been properly evaluated.

## Existing controls

Because of the hazards they pose to the marine and freshwater environment, SCCPs have long been recognised as priorities for regulatory action. In 1998, the Ministerial Meeting of OSPAR agreed on the target of cessation of discharges, emissions and losses of all hazardous substances to the marine environment by 2020 (the "one generation" cessation target) and included SCCPs on the first list of chemicals for priority action towards this target (OSPAR 1998). More recently, SCCPs have been included on the list of "priority hazardous substances" under the Water Framework Directive, such that action to prevent releases to water within 20 years will be required throughout Europe (EU 2001).

In terms of more specific measures, the Paris Commission (now part of the OSPAR Commission) agreed in 1995 on a prohibition of the use of SCCPs in a wide range of uses within the North-East Atlantic region (PARCOM 1995), including in metal working fluids, as additives in paints and sealants and as flame retardants in rubbers and plastics. This decision still remains to be fully implemented.

In the mean time, the EU has completed a risk assessment for SCCPs (EC 2000) and agreed upon restrictions only for use in metal working and leather processing (EU 2002). This leaves almost half of current uses within the EU, mainly uses in consumer products, unregulated. Inevitably, the risk assessment was based on very limited data in some areas, especially regarding toxicity to sediment and soil-dwelling animals and to humans.

During 2003, the EU will consider extending the prohibition on marketing and use to cover these other uses. So far, however, its Scientific Committee on Toxicity, Ecotoxicity and the Environment (CSTEE 2002) has advised against further controls, despite the hazards which SCCPs present and despite the CSTEE's recognition that some uses of SCCPs could continue to increase and that imports as components of products could be high. It is clear that current EU restrictions will not only fail to ensure that OSPAR's cessation target for SCCPs will be met in full, but will also permit continued exposure to, and environmental releases of, SCCPs from a diversity of products containing them.

- CSTEE (2002) EC Scientific Committee on Toxicity, Ecotoxicity and the Environment, Opinion on "Short-Chain Chlorinated Paraffins (SCCPs)", Follow-up of Directive 2002/45/EC (Opinion expressed at the 35th CSTEE plenary meeting, Brussels, 17 December 2002): 8 pp. [http://europa.eu.int/comm/food/fs/sc/sct/out174\\_en.pdf](http://europa.eu.int/comm/food/fs/sc/sct/out174_en.pdf)
- EC (2000) European Union Risk Assessment Report, alkanes, C10-13, chloro, 1st Priority List, Volume 4, EUR 19010 EN: 176 pp.
- EU (2001) Decision No 2455/2001/EC of the European Parliament and of the Council of 20 November 2001 establishing the list of priority substances in the field of water policy and amending Directive 2000/60/EC, Official Journal L 249 , 17/09/2002: 27-30
- EU (2002) Directive 2002/45/EC of the European Parliament and of the Council of 25 June 2002 amending for the twentieth time Council Directive 76/769/EEC relating to restrictions on the marketing and use of certain dangerous substances and preparations (short-chain chlorinated paraffins), Official Journal L 177, 06/07/2002: 21-22
- Farrar, D.G. (2000) Chlorinated paraffins – their toxicology and environmental effects and regulatory implications. *Organohalogen Compounds* 47: 129-130
- Fisk, A.T., Tomy, G.T. & Muir, D.C.G. (1999) Toxicity of C-10-, C-11-, C-12- and C-14-polychlorinated alkanes to Japanese medaka (*Oryzias latipes*) embryos. *Environmental Toxicology and Chemistry* 18(12): 2894-2902
- Koh, I.-O., Rotard, W. & Thiemann, W.H.-P. (2002) Analysis of chlorinated paraffins in cutting fluids and sealing materials by carbon skeleton reaction gas chromatography. *Chemosphere* 47: 219-227
- OSPAR (1998) OSPAR Strategy with Regard to Hazardous Substances, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR 98/14/1 Annex 34
- OSPAR (2001) Short Chain Chlorinated Paraffins, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London, ISBN 0-946956-77-4: 16 pp.
- PARCOM (1995) PARCOM Decision 95/1 on the Phasing Out of Short Chained Chlorinated Paraffins, OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic, OSPAR Commission, London: 3 pp.
- Peters, A.J., Tomy, G.T., Jones, K.C., Coleman, P. & Stern, G.A. (2000) Occurrence of C10-C13 polychlorinated *n*-alkanes in the atmosphere of the United Kingdom. *Atmospheric Environment* 34: 3085-3090
- Stern, G.A. & Tomy, G. (2000) An overview of the environmental levels and distribution of polychlorinated paraffins. *Organohalogen Compounds* 47: 135-138
- Tomy, G.T., Stern, G.A., Lockhart, W.L. & Muir, D.C.G. (1999) Occurrence of C-10-C-13 polychlorinated *n*-alkanes in Canadian mid-latitude and arctic lake sediments. *Environmental Science and Technology* 33(17): 2858-2863

# **Annex 3: details of analytical methodologies employed**

This Annex provides more detailed descriptions of the analytical methods and instrumentation employed by the three participating laboratories.

#### **Quantitative analysis for phthalate esters and alkylphenol compounds and qualitative GC-MS screen for non-target compounds**

These analyses were conducted by the laboratories of LGC Ltd, located in Teddington, UK.

Approximately 10g of dust sample were Soxhlet extracted with 200ml of dichloromethane for 2½ hours. The dust was spiked with a deuterated internal standard mix to facilitate quantitation. In each batch of 10 samples a blank and standard recovery solution were also extracted. 10g of acid-washed sand was used as the matrix simulant. At the end of the heating period, the heat was removed and the dichloromethane concentrated to below 50ml under a stream of dry nitrogen at 30°C. The extract was quantitatively transferred to a 50ml volumetric flask and made up to volume. All extracts were stored at 4°C until analysis.

5 standard mixtures containing the internal standards were analysed, bracketed around the samples. Response factors were calculated for each of the specific determinands. The efficiency of the extraction procedure was monitored by calculating the percentage recovery for each analyte of interest against the internal standard used for quantitation (phenanthrene-d10).

Recovery efficiencies are displayed in Table A3.1

#### **Quantitative analysis for brominated flame retardants and short-chain chlorinated paraffins**

These analyses were conducted by laboratories of the Netherlands Institute for Fisheries Research (RIVO) located in IJmuiden, The Netherlands.

Dust samples were Soxhlet extracted for 12 h with hexane:acetone (3:1, v/v, 70 °C). After addition of internal standards (2,3,5,6,3'-pentachlorobiphenyl (CB112) and 13C BDE-209), the extract was concentrated on a rotary evaporator, demi-water (pH=2) was added and the organic layer collected. The water was extracted two further times with iso-octane. Organic extracts were combined and concentrated in 2 ml of dichloromethane.

Each extract was cleaned by gel permeation chromatography (GPC) through two Polymer Laboratories (PL) gel columns (100 x 25 mm, pore size 10 µm), using dichloromethane at 10 ml/min. The collected fraction was that eluting between 18 and 23 minutes. This fraction was concentrated under nitrogen, dissolved in iso-octane and further purified by shaking with sulphuric acid. Finally, the pentane/iso-octane mixture was concentrated under nitrogen to 2 ml (iso-octane) and eluted through a silica gel column (2% water) with 11 ml iso-octane and 10 ml 20% diethylether in iso-octane. Both fractions were concentrated to 1 ml (iso-octane).

The final analysis was carried out by GC-MS, using electron capture negative ionisation (ECNI) as the ionisation technique, with methane as a reagent gas. A 50m CP Sil 8 column (i.d. 0.25 mm, film thickness 0.25 µm) was used for the determination of all brominated flame retardant target compounds (with one exception) and short-chain chlorinated paraffins (SCCPs). The flame retardant decabromodiphenyl ether (BDE-209) was analysed separately using a 15 m DB-5 column (i.d. 0.25 mm, film thickness 0.2µm). Peak identification was based for polybrominated diphenyl ethers (PBDEs, except BDE-209) on retention time and the recognition of the Br<sup>-</sup> (bromine) ion (m/z 79/81), and on specific target ions in the case of BDE-209, hexabromocyclododecane (HBCD) and the SCCPs.

Concentrations of the following compounds/congeners were determined in each sample:-

- Polybrominated diphenylethers (PBDEs) – tri- (BDE-28), tetra- (BDE-47, 66, 71, 75, 77), penta- (BDE-85, 99, 100, 119), hexa- (BDE-138, 153, 154), hepta- (BDE-190) and deca- (BDE-209).
- Polybrominated biphenyls (PBBs) – di- (BB-15), tetra- (BB-49, 52), penta- (BB-101), hexa- (BB-153, 155) and deca- (BB-209).
- Hexabromocyclododecane (HBCD)
- Tetrabromobisphenol-A (TBBPA) – plus its methyl derivative.

Limits of detection varied from sample to sample and from congener to congener, depending on sample size and method/instrument sensitivity respectively. For PBDEs, detection limits ranged from 0.12 to 0.62 µg/kg (ppb) on a dry weight basis. For PBBs, detection limits ranged from 0.18 to 2.8 ppb, for HBCD from 2.5 to 12.8 ppb, for methyl-TBBPA from 0.1 to 0.5 ppb and for TBBPA itself from 0.5 to 3 ppb.

The limit of determination was set by the lowest concentration of the multi-level (6 point) calibration curve in each case. Quantification of SCCPs is rather difficult due to very complex mixture of compounds and, therefore, is semi-quantitative.

#### **Quantitative analysis for organotin compounds**

These analyses were conducted by laboratories of GALAB, located in Geestacht, Germany.

All samples were further sieved to remove all particles with dimensions greater than 65 µm (0.065 mm) prior to analysis. Organotin compounds were extracted using a mixture of methanol and hexane (with NaBEt<sub>4</sub>) and quantified by gas chromatography/atomic emission detection (GC/AED), according to accredited methods after DIN EN 17025. Concentrations of the following compounds were determined in each sample:-

- Butyltins - mono-, di-, tri- and tetrabutyltin (MBT, DBT, TBT and TeBT respectively)
- Octyltins - mono- and di-octyltin (MOT and DOT respectively)

- Tricyclohexyltin (TCHT)
- Triphenyltin (TPT)

Limits of detection for all organotin compounds were 1 ug tin cation/kg dry weight of sample in each case.

#### Organotin compounds – individual sample analysis

Batch Date	10/12/02	10/12/02	11/12/02	11/12/02	12/12/02	12/12/02	13/12/02	13/12/02
Injection	(A)	(B)	(A) *	(B) *	(A)	(B)	(A)	(B)
<b>Analyte</b>								
Di-methylphthalate	115.2	105.4	110.5	106.4	97.5	100.0	93.4	99.8
Di-ethylphthalate	178.2	128.4	96.4	102.4	100.5	100.0	95.0	107.1
4-(1,1,3,3-tert-methylbutyl)phenol	115.3	111.2	60.9	69.8	102.0	105.2	98.9	104.2
4-Nonylphenol	178.5	166.3	68.3	87.1	105.1	128.7	55.4	90.6
4-n-Octylphenol	100.9	113.8	33.8	39.3	97.2	109.1	105.3	106.8
Di-n-propylphthalate	107.7	100.3	81.3	87.6	97.9	100.3	94.2	104.5
Di-isobutylphthalate	186.6	173.5	130.3	139.7	115.7	117.7	175.5	191.5
Di-n-butylphthalate	129.3	127.1	78.2	86.4	104.0	110.2	124.0	136.2
Butylbenzylphthalate	91.3	111.6	25.4	34.1	94.0	104.4	96.7	106.4
Di-2-ethylhexylphthalate	101.8	129.2	187.3	29.6	70.8	154.4	110.4	109.7
Di-isononylphthalate	67.1	110.0	25.2	26.1	87.8	91.2	94.7	80.5
Di-isodecylphthalate	68.9	98.8	24.7	26.8	84.6	95.0	97.2	87.2
Mean Recovery	<b>120.1</b>	<b>123.0</b>	<b>76.9</b>	<b>69.6</b>	<b>96.4</b>	<b>109.7</b>	<b>103.4</b>	<b>110.4</b>

The recoveries of the analytes for the 11/12/02 are poorer than the other days as it was discovered after the analysis sequence had been run that the level in the original vial analysed was either below the syringe needle or had evaporated prior to injection. The results above are from a re-analysis of the same solution a few weeks later (after the Christmas shutdown) when the contents had presumably started to degrade.





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