Consuming Chemicals #2

Hazardous chemicals in house dusts as indicators of chemical exposure in the home

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Authors: David Santillo, Iryna Labunska, Maureen Fairley & Paul Johnston

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Greenpeace Research Laboratories, Department of Biological Sciences, University of Exeter, Exeter EX4 4PS, UK

Lay-out : French actress Marion Cotillard hoovering her own flat as a participation in the Greenpeace Housedusts sampling programme.

Executive Summary

Although society is becoming increasingly aware of the widespread presence of hazardous man-made chemicals in the environment, 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, be it to make plastics soft or stop them breaking down, to carry perfumes, to protect against fire or even 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
- chlorinated paraffins which may be carcinogenic, used in plastics, paints and rubbers

In a study published by Greenpeace in May 2003, samples of household dust collected from a total of 100 households from 10 regions across the UK were analysed for the hazardous chemicals listed above (Santillo *et al.* 2003). This research confirmed the widespread presence of phthalate esters, brominated flame retardants and organotin compounds in household dusts, with the phthalate diethylhexyl phthalate (DEHP) and the flame retardants decabromodiphenyl ether (BDE-209) and hexabromocyclododecane (HBCD) in particular abundance. Nonyphenol and short-chain chlorinated paraffins (SCCPs) were also commonly found, as well as a wide range of other man-made chemicals including pesticides and solvents.

The current study was conducted in order to extend our earlier research to encompass a larger number of samples from a broader range of European countries, namely Germany, Spain, France, Italy and Slovakia. A total of 58 samples were collected from France, principally representing five major cities, while 22 samples were collected from Spain, representing four regions. Five samples each were collected from different cities in Germany and from Roma in Italy, and eight samples were collected in Slovakia, representing two geographical regions.

In very broad terms, these wider European samples yielded somewhat higher levels of phthalate esters and short-chain chlorinated paraffins, similar levels of organotins and somewhat lower levels of alkylphenols and brominated flame retardants compared to the UK samples analysed in the previous study.

All samples analysed contained phthalates, brominated flame retardants, organotin compounds and short-chain chlorianted paraffins (SCCPs). In contrast to the UK, relatively few samples contained detectable residues of nonylphenol. On average, each gramme of dust contained in the order of 1 mg (i.e. 1 part in every 1000) in total of the five hazardous chemical groups specifically quantified, although the ranges of concentrations of individual chemicals underlying this average were, as expected, fairly broad.

Phthalate esters, especially the reproductive toxicant DEHP, accounted for the major part of these contaminant levels in all samples. DEHP was found at concentrations ranging from 14.9 to 3289 ppm (3.3 mg/g, or parts per thousand). The highest concentration of phthalates overall, 7237 ppm (7.2 mg/g), was found in a single sample from France, containing roughly equal proportions of DEHP and butylbenzyl phthalate (BBP).

As for the UK samples, the most abundant brominated flame retardants identified in samples from Spain, France, Germany and Italy were decabromodiphenyl ether (BDE-209, 92-3400 ppb) and hexabromocyclododecane (HBCD, 77-1600 ppb). Both are known to be in widespread use as flame retardant additives in plastics, textiles and other applications across Europe. However, absolute concentrations for both chemicals were generally below the range recorded in the UK (3800-19900 ppb and 940-6900 ppb respectively). These data reaffirm the presence of lower levels in non-UK samples suggested by our previous study.

In contrast, concentrations of short-chain chlorinated paraffins (SCCPs) in samples from Germany, France, Spain and Italy were consistently higher than those previously reported for the UK samples (17-95 ppm, compared to <0.12-13 ppm for UK). These high levels of SCCPs were unexpected, but do serve to demonstrate the scale of ongoing releases of these hazardous chemicals from materials commonly found in the home environment. It may be hypothesised that the relatively high levels of SCCPs in samples from mainland Europe compared to the UK, coupled with relatively lower levels of brominated flame retardants, reflect historic and, possibly, ongoing differences in use of flame retardant chemicals in different parts of Europe. Alternatively such differences could result from differing preferences in furnishings and/or interior finishes between countries, such as differences in the frequency of rooms fitted with carpets or PVC (vinyl) flooring.

Despite the scale of the sampling programmes involved in this and in our previous study, the numbers of composite samples subjected to brominated flame retardant and SCCP analyses were necessarily limited (10 samples from UK compared to 14 samples from mainland Europe). Clearly, further sampling and analysis would be required to confirm any such differences and to begin to describe underlying causes. Nevertheless, these data do serve to raise interesting questions regarding the distribution of hazardous chemicals in the indoor environment across Europe.

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 complacence. 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, but 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 avoided altogether 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.

The opportunity is now here for all 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 has prepared 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.

Introduction -Chemicals in Europe

"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 Sept 1998

Current regulation of chemical production and use is weak, cumbersome and ineffective. This, the EU has stated, has led to a situation in which there is very little toxicity data on the great majority of the 100 000 plus chemicals currently used by industry, and next to nothing is known about their potential environmental and human health impacts.

This situation has led the European Commission to propose new legislation that offers greater protection to human health and the environment. In its White Paper "Strategy for a Future Chemicals Policy" the Commission states:

"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.

These examples expose the weakness of current EU chemicals policy…there is a general lack of knowledge about the properties and uses of existing substances. The...process is slow and resource intensive and does not allow the system to work…effectively. Information on uses of substances is difficult to obtain and information about exposure…is generally scarce. Decisions on further testing of substances can only be taken via a lengthy committee procedure and can only be requested from industry after authorities have proven a substance may present a serious risk. Without test results however it is almost impossible to provide such proof. Final risk assessments have therefore only been completed for a small number of substances.

European Commission White Paper: Strategy for a Future Chemicals Policy 27.2.2001

There is then, no doubt that a complete overhaul of EU chemicals policy and new primary legislation governing the manufacture, marketing and use of all intentionally manufactured chemicals in the European Union is necessary.

Following months of detailed drafting and consultation, the European Commission is now about to adopt new legislation governing the use of chemicals within Europe. The new system, known as REACH (Registration, Evaluation, Authorisation of Chemicals), aims firstly to gather vital information on the uses and hazards of a vast array of chemicals on the market in Europe, chemicals which have, in many cases, been used for decades without full knowledge even of such basic properties as toxicity and persistence in the environment. Moreover, for chemicals which are judged, on the basis of these hazardous properties, to be of very high concern (such as carcinogens, persistent and bioaccumulative chemicals and endocrine disruptors), REACH introduces a requirement for positive authorization if their continued use is to be permitted. It is envisaged that authorisation will only be granted if the uses are essential and no safer alternatives are available, or if the risks can be "adequately controlled". For chemicals which are carcinogenic, toxic to reproduction, persistent and bioaccumulative or capable of endocrine disruption, it is difficult to see how any continued use could be authorized under the concept of "adequate control" of risks. This is especially true if the uses in question result, for example, in widespread contamination of the indoor environment and, therefore, the possibility of direct and continuous chemical exposure in the home.

It is to be hoped that the new EU regulations will create a Europe-wide and, ultimately, global process to bring to an end the current unsustainable situation with regard to chemical use and exposure. So far we have seen action to control some long-recognised chemical hazards (asbestos, PCB's, DDT) only after years of widespread use. For many of these, environmental levels are now declining, but not before serious and irreversible damage has already been done. As the European Environment Agency has noted, we have all too often learnt "*late lessons from early warnings*". At the same time, we have seen some of these old problems simply replaced with a new generation of persistent, man-made chemicals, which are also, quite predictably, now accumulating in the environment, in our foods, even in our bodies. Moreover this is happening in an insidious way as chemicals are unwittingly brought into our homes as unseen and unlabeled chemical additives in everyday consumer products. It seems we are not even learning the lessons late.

In May 2003, Greenpeace published its first study into chemical contamination of house dusts, using a total of 100 samples drawn from 10 different regions of the UK to confirm the widespread presence of a broad range of hazardous chemicals (Santillo *et al.*2003). In this second study, we have explored further the extent to which our unwitting reliance on a range of hazardous chemicals present in household and other consumer goods is leading to chemical contamination of our home environment, this time using samples of dust collected from households in five other European countries. From a scientific point of view, this report should help throw light on one route of exposure to long-term hazardous chemicals - and all of the chemicals studied here are intentionally produced for use in consumer goods. But it also has another purpose, to demonstrate why our national politicians also need to recognise the problem and commit to supporting and implementing strong and effective, EU legislation.

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 new EU chemicals legislation (REACH) presents 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 is an environment free of man-made chemical contaminants. Europe can lead the way to 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, often with unknown characteristics. But that is exactly what they currently do. The 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 contaminated. The second is to ensure the public know that their elected representatives have, right now, an opportunity to change that.

Greenpeace expects those representatives to take that opportunity. We believe the public will expect that too.

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, be they carpets, curtains or cosmetics, televisions or toys, certain properties. For example, some additives are included to make plastics flexible or textiles fire retardant, others to guard against dust mites or mould or to carry perfumes. Of course, we don't see these chemicals. Nor, in most cases, do we even know they are there.

In fact, 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 many of the chemicals commonly incorporated in 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, etc. 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. And organic compounds of the metal tin (organotins), used as fungicides or as stabilizers in hard and soft PVC products, are harmful to the immune system in mammals, including humans.

Although we are not aware of it, we are therefore exposed to these chemical hazards on a daily basis, though use of products containing them, through contact with dusts in which these chemicals can accumulate, even 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 to the indoor or outdoor environment, they do not readily break down into harmless byproducts. Rather they may simply become ever more wide dispersed through the environment, carried on air currents or in water. Moreover, because of their very chemical nature, some have a high affinity for fatty tissues and may therefore accumulate over time in the bodies of animals, including through the food chain.

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. 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 the many years for which we, as consumers, have unknowingly relied upon hazardous chemical additives, these 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 of our chemical society 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 themselves, or through contact with contaminated air and/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, in which the chemicals of greatest concern are expected to accumulate. This is only a small part of the picture, of course, 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.

As noted above, this study represents the second phase of Greenpeace's work on the chemical contamination of house dusts, following on from that published in May 2003 (Santillo *et al.* 2003) which focused primarily on households in the UK. The current study extends this research to a larger number of countries across Europe, including France, Spain, Germany, Italy and Slovakia. One of the principal objectives is to determine whether the pattern and scale of contamination recorded in UK households are typical of dusts from European households more generally, or whether any consistent and significant differences can be discerned. At the same time, it is hoped that the results of this investigation, combined with those from our previous work, will contribute to the body of scientific understanding regarding chemical distributions and exposures in the home.

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 summer time 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).

Until recently, the chemical characterization of dusts, and evaluation of the significance as contributors to overall exposure, were largely limited to a few toxic heavy metals, predominantly lead. Lead is an important developmental toxin and is also a major contaminant associated worldwide with traffic pollution, resulting from use of leaded petrol and of lead weights for wheel balancing (e.g. see Vojta *et al.* 2002). It is also released by a range of industrial sources, including mining, smelting and waste incineration.

Early studies of chemical contamination of indoor dust therefore focused on its propensity to accumulate lead and other metals arising primarily from outside sources. For example, higher dust lead levels were clearly detectable in households located in close proximity to certain industrial point sources in Germany (Meyer *et al.* 1999) and in areas of dense traffic in Hong Kong (Tong and Lam 2000). At the same time, in households more remote from such obvious external sources, levels of lead and other metals in dust were found to be more reflective of indoor sources. Thus, Tong and Lam (2000) found some correlation between lead in house dust and the colour of paints used on internal walls and fixtures, possibly reflecting the different lead composition of the paints. Certainly, in houses in which lead paints have been used or remain from historical applications, resulting levels in dust are thought likely to be at least as significant as outdoor sources, if not more so (Adgate *et al.* 1998).

For other chemical groups, the situation is likely to be similar. Where there are substantial localized external sources of pesticide residues, for example in some agricultural residences, such sources may make the primary contribution to overall indoor dust contamination as a result of residents bringing the contaminants in on footwear or clothing (e.g. Curl *et al.* 2002).

By contrast, in the urban environment, substantial pesticide residues in dusts are much more likely to result from direct use of these chemicals for pest control in the home. For example, Gurunathan *et al.* (1998) reported the accumulation of residues of the organophosphorus pesticide chlorpyrifos on a number of surfaces in homes in the USA. Although this particular issue is perhaps likely to be a lesser concern in most parts of Europe, as household use of pesticides tends to be less prevalent, it nevertheless serves to illustrate that indoor sources of contamination must not be overlooked. This may well be the case for a diverse array of other chemicals which are present as components of consumer products.

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 suggest 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.

In May 2003, Greenpeace published an investigation into the chemical contamination of dusts collected from a total of 100 households from 10 regions across the UK (Santillo *et al.* 2003). This research confirmed the widespread presence of phthalate esters, brominated flame retardants and organotin compounds in household dusts, with the phthalate DEHP and the flame retardants decabromodiphenyl ether (BDE-209) and hexabromocyclododecane (HBCD) in particular abundance. The common presence of nonylphenol and of short-chain chlorinated paraffins (SCCPs) was also noted, as well as a wide range of other man-made chemicals including pesticides and other common chemical additives used in consumer products. This earlier study also included a small number of samples drawn from other countries in Europe, revealing similar ranges and patterns of contamination, albeit with some indication of lower levels of brominated flame retardants.

The current study, reported below, aimed to extend this work over a much wider geographical area, by applying similar techniques to dust samples collected from houses and other buildings in five European countries; Germany, Spain, France, Italy and Slovakia.

A total of 58 samples were collected from France, principally representing five major cities, though also including 7 additional samples from specific locations of interesest. 22 samples were collected from Spain, representing four regions. Five samples each were collected in Germany, from five different cities, and in Italy, in this case all from Roma. Eight samples were collected in Slovakia, four from each of two geographical regions.

For logistical reasons, not all of the samples from each country could be analysed for the full range of target chemicals identified below. Wherever possible, analyses were performed on individual samples to maintain statistical power. However, some analyses were performed using composite (pooled) samples, prepared by combining equal quantities of individual samples after sieving and homogenization. This inevitably reduced statistical power, but provided a way of extending the range of target chemical groups and yielded some idea of average concentrations for the region from which the pooled samples were drawn, while keeping within the limited time and resources available. Details of the sampling programme and the preparation of individual and composite samples for analysis are given in the methods section below.

Chemicals targeted for investigation

As explained above, 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 the same five main compound groups covered in the UK study, based on their reported high volume use in common household furnishings and other products and on their intrinsic hazardous properties:-

- *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 compojnents of electrical and electronic 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* (including the well known di(2-ethylhexyl) phthalate, or DEHP, among others) – used as softeners in flexible PVC products, including floors, wallpapers, furnishings, clothing and toys, as well as 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 byproducts , i.e. they tend to persist in both the outdoor and indoor environment;
- – they are all able to leach out of, or other wise 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”.

A further illustration of the long-recognized and widely accepted concerns surrounding the hazardous nature of these chemical groups is provided by the fact that all five groups have been prioritized for action under the OSPAR Convention's strategy to protect the marine environment. The OSPAR Convention, signed in 1992 and entering in to force in 1998, is a legal agreement between 15 European countries, all of which discharge waters to the North East Atlantic region, and the European Community. OSPAR's mission is to protect the marine environment of the North East Atlantic from a diversity of threats, including pollution from hazardous chemicals. In 1998, at Ministerial level, OSPAR agreed on its Strategy with Regard to Hazardous Substances (OSPAR 1998), including the target of cessation of all releases of hazardous substances to the marine environment by 2020 (the "one generation" cessation target). As a first step towards this target, OSPAR drew up a list of chemicals requiring priority action, including *inter alia* alkylphenols, brominated flame retardants, organotins, certain phthalates and short-chain chlorinated paraffins.

Some uses of some of these chemicals are already, or will shortly be, subject to regulatory control at EU level (such as for the short-chain chlorinated paraffins, TBT and, very recently, some of the brominated flame retardants). However, to a large extent their use and presence in consumer products continues unabated.

More detailed profiles for each of these five key groups, outlining the uses, environmental distributions, hazards and current regulatory status, are provided in Annex 2. In addition to these targeted quantitative analyses, we also subjected a number of the dusts collected to a broader qualitative screen analysis in order to provide some preliminary information on the presence of other indicative man-made chemicals.

Overall, it is hoped that this current study will contribute to knowledge and understanding of the chemical environment in which we live, and of the types and levels of chemicals to which we are continuously exposed through this route. It must be stressed at the outset, however, that this study will not provide (and is not intended to provide) any estimates of intake of chemicals from exposure to dusts in the home, either in absolute terms or relative to other potential sources. This would be an extremely complex, uncertain and subjective task at this stage. Nevertheless, the baseline information it provides may ultimately be useful as additional input to any such assessments which may be conducted in the future.

Sampling programmes and analytical methods

Sample collection

Samples of dust were collected from all five participating countries during the second quarter of 2003, through direct participation of Greenpeace staff and volunteers from the respective Greenpeace national or regional offices. In all cases, samples were collected with prior informed consent from the householder or other person(s) responsible for the premises sampled. While precise details of site selection inevitably varied from country to country, equipment and procedures were harmonized as far as practicable in order to avoid cross-contamination and the introduction of systematic bias between countries.

All samples were therefore 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

A total of 98 samples of dust were received by our laboratories from the five countries participating in this study, as summarised below.

Country	Sample codes	Total number of samples
Germany	HD03001 – 03005	5
Spain	HD03006 – 03027	22
Slovakia	HD03028 - 03035	8
Italy	HD03036-03040	5
France	HD03041 - 03098	58

On receipt at the Greenpeace Laboratories, all samples were immediately sieved through a pre-cleaned, solvent-rinsed[1] 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.

Germany

Five samples of dust from locations in Germany were received by our laboratory (sample codes HD03001-03005). The

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[1] analytical grade pentane

locations from which these samples were collected are summarised in table below.

Sample codes	Location
HD03001	Hambourg
HD03002	Regensburg
HD03003	Leipzig
HD03004	Berlin
HD03005	unknown

Following sieving and homogenisation, sub-samples of each individual sample were immediately repackaged into two layers of solvent-rinsed aluminium foil, enclosed in lightweight polyethylene bags and sealed in brown paper envelopes. These sub-samples 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 these samples in order to identify (as far as possible) any other organic contaminants present in significant quantities.

A single composite or pooled sample was prepared by combining equal quantities (equal weights) of sieved, homogenised material from all three samples for which sufficient material remained, namely HD03002, 03003 and 03004. Quantities of HD03001 and HD03005 remaining after sieving and sub-sampling for analysis by LGC were insufficient to use as contributions to the pooled sample and so were excluded. The pooled sample is therefore a representative “average” of three of the five samples received from Germany.

The single pooled sample (coded GERMANY) was homogenised and divided into two equal portions, one half being forwarded to the laboratories of GALAB (Geestacht, Germany) for quantitative determination of a range of organotin compounds. The remaining half was 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.

Spain

A total of 22 samples were received from Spain, representing four different regions as summarised below:-

Sample codes	Region
HD03006 – 03010 (5 samples)	Madrid
HD03011 – 03016 (6 samples)	Granada
HD03017 – 03022 (6 samples)	Valencia
HD03023 – 03027 (5 samples)	Asturias/Leon

Sub-samples were taken from each individual sample, as described above for the samples from Germany, and forwarded to LGC for quantitative analysis of phthalates and alkylphenols and qualitative analysis of other organic compounds present.

Preparation of pooled (composite) samples representing each of the four regions for analysis of other target compounds was made difficult by the very limited total quantities of dust present in many of the samples after sieving. In most cases, quantities remaining after sub-sampling were insufficient to use as contributions to pooled samples. For this reason, pooled samples for each region were again prepared from a sub-set of the total number of samples from that region as follows:-

Region	Samples used to prepare pooled sample
Madrid	HD03007 and 03010 only
Granada	HD03013, 03014 and 03016
Valencia	HD03018, 03021 and 03022
Asturias/Leon	HD03024 and 03026 only

These pooled samples were coded according to the region they represented. As was the case for the single pooled sample from Germany, the four regional pooled samples from Spain were subsequently divided into two, with one half forwarded for analysis of organotins by GALAB and the other half for analysis for brominated flame retardants and chlorinated paraffins by RIVO.

Italy

Five samples of dust were received from Italy, all collected from locations within Roma itself (HD03036-03040). Once again, sub-samples of sieved material from each sample were forwarded individually for analysis by LGC. As was the case for Germany, a single pooled sample was prepared from the three out of the five individual samples for which sufficient sieved material remained (namely HD03036, 03038 and 03039). This pooled sample, denoted ITALY, was divided and forwarded for analysis by GALAB and RIVO as above.

France

A total of 58 samples of dust were received from France. 51 of these were from households located in five different cities as summarised below.

The remaining seven samples were collected from other selected locations around France, including a school, a country house and the houses of politicians and other prominent people. These samples are coded HD03058 and HD03062 – 03067.

Sample codes	City
HD03041 – 03050 (10 samples)	Lille
HD03051 – 03057 and 03059 – 03061 (10 samples)	Toulouse
HD03068 – 03077 (10 samples)	Lyon
HD03078 – 03088 (11 samples)	Nantes
HD03089 – 03098 (10 samples)	Paris

From each set of regional (city) samples, a subset of five samples was selected to go forward for individual analysis of phthalates, alkylphenols and other organic contaminants by LGC. The five samples selected for each city were those yielding the largest quantities of material after sieving.

In addition, a single pooled sample was prepared to represent each of the five cities, combining equal quantities of sieved, homogenised material from seven of the total of ten or eleven samples in each city set (summarised below). Unfortunately, as for the sample sets representing other countries, insufficient dust was available from some of the samples from each city for them to form part of the overall pooled sample. Once again, the selection of a sub-set for pooling was considered a reasonable and defensible approach to overcome this limitation.

City	Samples used to prepare pooled sample
Lille	HD03041, 03042, 03043, 03045, 03046, 03047 and 03049
Toulouse	HD03054, 03055, 03056, 03057, 03059, 03060 and 03061
Lyon	HD03069, 03071, 03072, 03073, 03074, 03076 and 03077
Nantes	HD03078, 03080, 03081, 03082, 03083, 03084 and 03088
Paris	HD03078, 03080, 03081, 03082, 03083, 03084 and 03088

As for all other pooled samples described above, these five pooled samples from France were each divided into two and forwarded to GALAB and RIVO for analysis.

Of the remaining seven samples, representing other specific locations around France, one sample (HD03067) was judged unsuitable for analysis because the manner in which the sample had been packaged for shipment to our laboratory could have introduced the possibility of contamination from packing materials. This sample was excluded from all further analysis.

In the case of the other six samples, sieved material was divided into three equal portions in each case, which were subsequently forwarded for analysis by LGC, GALAB and RIVO respectively.

Slovakia

Eight samples were received from Slovakia, having been collected from public buildings (mother care centres and youth centres) located in two regions of Slovakia, HD03028 to 03031 from the South West of the country and HD03032 to 03035 from the North East.

All samples were sieved and homogenised separately as for samples from all other countries, but none of the samples from Slovakia were forwarded for analysis individually. Instead, a single pooled sample was prepared for each of the two regions described above (i.e. combining equal quantities of four samples in each case). These pooled samples were then divided in two, with one half forwarded to LGC for quantitative analysis of phthalates and alkylphenols and qualitative analysis of other organic contaminants and the other half forwarded to RIVO for analysis of chlorinated paraffins only.

Samples from Slovakia were not analysed for either organotin compounds or brominated flame retardants.

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 isooctane 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) varied from compound to compound, depending on detector sensitivity, and were affected in some cases by interference from co-eluting chemicals. 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 organic tin/g dry weight of sample (ppb) in each case.

Results and Discussion

Target compounds

Concentrations of the five target groups of compounds analysed quantitatively in the current study are summarised for all countries in Tables 1 to 5. Wherever possible, three statistics are shown for each compound in each country:-

- **median** (middle) values as a measure of centrality;
- **range** (lowest and highest values recorded), as an indication of the spread of data; and
- **frequency of detection** (number of individual or pooled samples out of the total number analysed in which each compound or group was found).

Given the highly skewed nature of some of the data sets (i.e. with a small number of very high values having a disproportionate effect on the average), the median values were considered to be more representative measures of centrality than the mean (average) values. Hence only the medians, calculated as the middle value of the concentrations when placed in numerical order, are reported here. In each case, medians have been calculated assuming that all values below limits of detection/quantification were zero.

In some cases, where only single pooled (composite) samples have been analysed for a particular country, these single values are reported in the tables.

Detailed results for each compound group, including data for individual samples and/or pooled (composite) national or regional samples, are provided in Annex 1.

Tables 1 to 5 also include data from our previous study of UK housedust samples (Santillo *et al.* 2003) for purposes of comparison. In very broad terms, the samples analysed in the current study, representing five countries in mainland Europe, yielded somewhat higher levels of phthalate esters and short-chain chlorinated paraffins, similar levels of organotins and somewhat lower levels of alkylphenols and brominated flame retardants than in UK samples analysed in the previous study.

Phthalate esters

Of the 9 individual phthalate esters specifically quantified, 3 (DiBP, DnBP and DEHP) were found at levels above limits of detection (LOD) in all samples analysed from each country (Table 1). A fourth, BBP, was found in all but one sample (from France), while DEP was found in all but two samples (again both from France).

The isomeric phthalates DiNP and DiDP were found in roughly half to a third of all samples, with slightly higher frequencies in France than in other countries. DPP was detected in only 3 samples analysed in this study (all from France), and then at relatively low levels ranging from 1.22 to 3.0 parts per million (ppm); these data are not included in the summary table 1.

In almost all samples, the most abundant phthalate ester was the commonly used DEHP, representing between 22 and 89% by weight of total phthalate concentrations, most commonly in the range of 40 to 60%. A similar predominance was noted in our previous study of UK dusts (Santillo *et al.* 2003). Concentrations of DEHP in dusts in the current study varied over a fairly wide range, from 15 to 3289 ppm, but were generally higher than those recorded in the UK dusts. The highest median value was that for the five samples from Germany (996 ppm, or almost 1mg DEHP per gramme of dust), although both composite samples from Slovakia contained higher levels (1290 and 2124 ppm). Dusts from Italy, France and Spain yielded lower median DEHP values (434, 504 and 317 ppm respectively), although the spread of concentrations for individual dust samples was high in all cases and some individual samples from France and Spain contained among the highest DEHP levels recorded in this study. For example, one sample each from Lille, Toulouse and Nantes in France, and from the Asturias region of Spain, contained more than 2000 ppm (2 mg/g dust) DEHP (see Annex 1). One additional sample from France (HD03062, or SP01) contained more than 3000 ppm DEHP.

DiBP was the most abundant phthalate in 7 of the 65 samples, whereas DnBP and BBP predominated in only one sample in each case. Nevertheless, all three compounds were found in the vast majority of samples, with median and maximum values in a similar range to those found in our UK study (Santillo *et al.* 2003) and those reported previously for house dusts from Germany by Butte and Heinzow (2002). DEP, widely used in perfumes, cosmetics and other personal care products, was also a compound common to almost all samples, again at concentrations similar to those previously reported for UK dusts.

It is evident from Table 1 that the summed concentrations of all individual phthalates analysed (denoted “total phthalates”) from all five countries included in the current study were generally higher than those we previously reported for comparable dusts from the UK (median total phthalates 354 ppm, range 1.6-1019 ppm). For example, almost half of the 65 individual samples analysed in the current study contained greater than the maximum recorded in the UK study. In the cases of Germany, Italy and Slovakia, statistical power is limited by the relatively small number of individual samples analysed. However, statistical comparison of total phthalate concentrations in dusts from France, Spain and UK indicated clearly that concentrations in UK samples were significantly lower. Moreover, although the difference is smaller, similar data analysis confirmed that dusts from France were, on average, significantly more contaminated than those from Spain.

Because of the small number of samples involved, coupled with the fairly wide spread of individual concentrations, it is difficult to draw any firm conclusions regarding regional differences in phthalate contamination within countries. Nevertheless, certain individual samples do stand out. Of particular note is sample HD03062 (SP01) from France, which contained more than 7000 ppm (7 mg/g dust) of phthalates, including more than 3000 ppm each of BBP and DEHP. The possibility cannot be ruled out that these, and perhaps some other, very high levels of phthalates in dust arise from the presence of small fragments of PVC passing through the sieve in the homogenised dust fraction, although every effort was taken to ensure that none of the prepared samples contained any recognisable fibres or fragments. Whatever the case, the results do confirm that, as a result of their widespread use in products found in the home, phthalate esters are ubiquitous and very abundant chemical components of household dusts throughout Europe.

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

Alkylphenols

In contrast to our findings for the UK house dusts, nonylphenol was found only infrequently in the current study (Table 2). This compound was found in 22 of the 29 UK samples, at concentrations up to 35.2 ppm (median 9.8 ppm). In comparison, nonylphenol was detected in only 5 of 22 samples from Spain, in 1 of the 31 samples from France and in 1 of 5 samples from Italy. Where it was found, concentrations were in the low ppm range, and generally below 1ppm. None of the 5 German samples contained detectable levels. These results also indicate a lower abundance in other parts of Europe than those summarised by Butte and Heinzow (2002), who reported a median concentration for German house dusts of 6.2 ppm.

Both composite samples from Slovakia contained nonylphenol, at concentrations again in the low ppm range. Clearly it is not possible to determine if these levels reflect general contamination across all individual samples from which the two regional composites were produced or if they resulted from dilution of a single, more contaminated sample when preparing these composites.

Neither octylphenol nor 4-(1,1,3,3-tert-methylbutyl)phenol (4TMBP) were detected in any of the 65 samples analysed in the current study. 4TMBP had been detected in 4 of 29 samples, and octylphenol in 1 of 29 samples, from the UK (Santillo *et al.* 2003), although these other compounds made only a minor contribution to total alkylphenol concentrations in all cases.

Despite the relatively low frequency with which nonylphenol was detected in the current study, given the hazards associated with this endocrine disrupting chemical, its presence even in a subset of the dusts is clearly of concern. Obviously it is not possible to determine likely sources of this chemical in any particular case, although a number of uses of this substance are known (see Annex 2 for further details of hazards and uses). Recent findings of this compound, and of the closely related nonylphenol ethoxylates, in some items of children's clothing purchased in the UK (Peters 2003) provides an illustration of one possible source to the indoor environment.

Organotin compounds

Of the eight organotin compounds monitored in the current study, four were found in all samples (MBT, DBT, TBT and MOT), while one was found in all but one sample (DOT). None of the samples contained detectable residues of TeBT, TCHT or TPT (Table 3).

The relative abundances of the different compounds were similar in the majority of samples from France and in single composite samples for Germany and Italy, and similar to those we previously reported for the UK, with a predominance of MBT, slightly lower levels of MOT and DBT and lower concentrations again of DOT and TBT. The four composite samples from Spain appeared to show a more equitable contribution from MBT and MOT, although this could simply be an artefact arising from the small number of samples analysed.

Overall, the sums of organotin compounds were highest for France (median 2234 ppb, 8 samples) and lowest for Spain (1495 ppb, 4 samples), with the single composite samples for Germany and Italy yielding intermediate values (1956 and 1790 ppb respectively). Together, these values are similar to, if slightly lower than, the median of 2432 ppb determined previously for 10 composite samples from the UK. Concentrations of each organotin compound in each individual or composite sample are tabulated in Annex 1.

Whereas total organotin concentrations for composite samples for all four regions in Spain fell within a fairly narrow range (1125-1958 ppb), concentrations in the five regional composites from France were more widely spread. Three samples yielded organotin concentrations in excess of 5000 ppb (or 5 ppm), with the composite for Lille containing 5399ppb, that for Toulouse, 8806 ppb, while the composite representing Nantes contained over 18000 ppb (18 ppm), by far the highest value we have recorded in house dust to date. In composites from both Lille and Nantes, the bulk of these totals was comprised of MOT, although the Nantes sample also contained more than 1000 ppb (over 1 ppm) of DBT, known to be toxic to the developing immune and nervous systems in mammals (see Annex 2). Of course, whether these differences between composites represent true regional variation (perhaps even arising from local external sources), or simply result from one or more very highly contaminated individual samples included in the composites, cannot be determined from these data. The particularly high levels of organotins in these samples do, however, provide justification for further, more detailed investigation of individual samples.

It is interesting that the individual sample HD03062 (SP01) from France, which contained the highest overall levels of phthalate esters, contained among the lower concentrations of organotin compounds (at 928 ppb, the second lowest value recorded in the current study). Given that organotins are likely to occur most commonly in the home environment as a result of their use as stabilisers in plastics such as PVC, a closer correlation with phthalate esters might be expected. One explanation for the disparity could be that the high phthalate levels in sample HD03062 arose primarily from soft PVC products which are not normally stabilised with organotins, such as children’s toys, for example. Once again further investigation of this specific sample location would be required if firmer conclusions were to be drawn.

Although a relatively minor contribution to total organotin concentrations, TBT was a conspicuous contaminant in samples from all countries. Concentrations, ranging from 11.6 ppb for the Italian composite to a median of 16.2 ppb for the 8 samples from France, were lower than the median of 49.9 ppb recorded for 10 UK composite samples, but not insignificant. Indeed, the composite sample prepared for Toulouse contained more than 500 ppb (0.5 ppm) of TBT, a recognised immunotoxin in mammals (see Annex 2). Its origins in the household environment are poorly described, although it is known to be used as an active ingredient in certain textile finishes and can be present as a low-level contaminant in preparations of other butyltins.

Brominated flame retardants

Decabromodiphenyl ether (BDE-209)

Of all the brominated diphenyl ethers detected in dust samples in the UK study, BDE-209 was by far the most abundant (median 7100 ppb, or 7.1 ppm), as may be expected from its continued widespread use in the face of restrictions and voluntary phase-outs of other PBDE formulations. BDE-209 was also the most abundant congener (of those quantified) in all samples from Spain and France and in the single composites from Germany and Italy, in the current study, albeit at concentrations consistently lower than those recorded for UK dusts (Table 4). Indeed, the highest value recorded in the current study, 3400 ppb or 3.4 ppm for the composite sample from Lyon in France, fell below the lowest value of 3.8 ppm for the UK samples.

Although such differences could be an artefact arising from limited sample sizes, this should have been limited to some degree by the preparation of composite samples from a larger number of individuals. It seems reasonable to suggest, therefore, that these values reflect real differences in the propensity of use of this chemical, or of products containing it, and in its consequent abundance in the indoor environment between the UK and other countries. This hypothesis is further supported by the substantially lower levels of BDE-209 noted for the two non-UK samples included in our previous study (Santillo *et al.* 2003). Although none of the individual or composite samples we have analysed could be considered to provide a representative sample of all households in the countries from which they were drawn, the difference apparent between the 10 UK samples and the 16 non-UK samples analysed to date is statistically highly significant.

Notwithstanding such differences, the levels of BDE-209 in the samples from Germany, France, Spain and Italy are still of considerable interest and could clearly contribute to ongoing human exposure to this chemical within the home environment. Although the contribution to overall exposure arising from dust is not yet known, these results highlight once again the potential significance of this additional exposure route.

Other brominated diphenyl ethers

Aside from BDE-209, a number of other PBDE congeners were commonly found in dust samples from Germany, Spain, France and Italy (Table 4), albeit at concentrations one or two orders of magnitude (10-100 times) lower than for BDE-209. Both the tetrabrominated congener BDE-47 and the pentabrominated congener BDE-99 were found in all samples analysed in the current study in the low to medium ppb concentration range. For both these congeners, concentrations in the four Spanish composites (11-16 ppb and 14-21 ppb for BDE-47 and BDE-99 respectively) were notably lower than for the five French composites (11-73 ppb and 16-78 ppb respectively). However, the highest values recorded in the current study for both congeners were found in sample HD03058 (SP05), also from France,which contained 260 ppb BDE-47 and 720 ppb BDE-99 (see Annex 1 for details).

Unlike BDE-209, lower-brominated congeners such as BDE-47 and BDE-99 have a high propensity to bioaccumulate, a factor which led to their rapid rise in concentration in body tissues and breast milk across Europe and North America (see Annex 2) and which, ultimately, has also contributed to restrictions on marketing and use within the EU. One again, however, their presence in house dust, probably arising from a variety of different flame-retarded products, illustrates the fact that exposure in the home from existing products will remain a problem for many years to come even after use in new products is prohibited.

Unfortunately, one of the other more abundant PBDE congeners in the UK samples, BDE-153, could not be quantified in the current study because of analytical interferences (which incidentally also prevented the quantification in this study of tetrabromobisphenol-A).

Hexabromocyclododecane (HBCD)

Like BDE-209, HBCD was detected as a prominent contaminant in all samples analysed in the current study, although concentrations were generally slightly lower than for BDE-209 (Table 4). Once again, levels in samples from Spain, France, Germany and Italy were almost exclusively lower than those recorded for the UK dusts, with only one composite sample from France (Lille, 1600 ppp) and the composite from Germany (1200 ppb) exceeding the lowest concentration recorded for the 10 UK composites (940 ppb) in our previous study. Nevertheless, these data also confirm the widespread presence of this persistent flame retardant in dusts from the indoor environment across Europe. Given its suspected genotoxicity and carcinogenicity, coupled with its potential to bioaccumulate, the presence of HBCD from the high ppb to low ppm range in housedust gives substantial cause for concern.

Tetrabromobisphenol-A

As noted above, as a result of co-elution of this compound with BDE-153, tetrabromobisphenol-A could not be quantified in the current study. The reasons for this interference are not known. It is important to note, however, that this does not imply that these compounds were not present in the samples, simply that their presence could not be verified nor quantified. Given the apparent differences in abundance of other brominated flame retardants noted above, this limitation is unfortunate but unavoidable. Further sampling and analysis, perhaps with some methods development, would be necessary to resolve this issue.

Short-chain chlorinated paraffins (SCCPs)

In contrast to the lower concentrations of brominated compounds identified in dusts from mainland Europe compared to the UK, levels of SCCPs were consistently and quite markedly higher than in UK dusts (Table 5). Concentrations ranged from 17-41 ppm for Spain to 30-95 ppm for France, with the single composites from Italy and Germany, and for the two composites from Slovakia, yielding intermediate levels. In all cases, concentrations exceeded the highest concentration recorded for the UK composites (13 ppm).

The region for this striking difference between the UK and mainland Europe, and for these alarmingly high levels, is not clear. Given that one application for SCCPs is as flame retardant additives, in textiles, rubbers and certain plastics, it could be hypothesised that the higher SCCP concentrations in samples from Germany, Spain, France, Italy and Slovakia, coupled with lower concentrations of brominated flame retardants, reflects differences in historic (or even ongoing) patterns of use of flame retardant additives between the UK and other parts of Europe. Alternatively, such differences could conceivably arise from differences in the patterns of use of different indoor finishes and/or furnishings, such as, for example, the relative abundance of carpets compared to other flooring types in different parts of Europe. Of course, both hypotheses are highly speculative, especially given the small number of samples on which the relative distributions can be judged, but is certainly an interesting anomaly deserving of further investigation.

On the basis of the risk assessments conducted on SCCPs within the EU in the late 1990s, restrictions were introduced on the marketing and use of these chemicals (EU 2002). However, these restrictions ban the use of SCCPs only for metal working and leather processing applications, based on a judgement that other uses of SCCPs, including as plasticizing and flame retarding additives in polymers and textiles, did not give rise to significant concerns regarding environmental releases or human health. The presence of SCCPs at concentrations up to 13 ppm in the UK samples highlighted the urgent need to re-evaluate the limited scope of these legal restrictions and extend them to cover other uses of these hazardous chemicals. Their presence at concentrations consistently in excess of this level, and at up to 95 ppm, in dusts from other parts of Europe, serves to reinforce this conclusion and the urgency with which additional restrictions must be introduced.

Further information on the uses and hazards of SCCPs are provided in Annex 2.

Other organic compounds (non-target compounds)

In addition to the target compounds discussed above, qualitative analysis by GC/MS revealed a diversity of other organic compounds in the dust samples, including other plastic additives, solvents, pesticides and various chemicals which most probably arise as residues of detergents and personal care products. These findings are summarised in Table 6 below. Details of the additional (non-target) compounds identified in individual samples are included in Annex 1. The numbers of additional compounds isolated varied from only 1 to a maximum of 49 (in one sample from Germany). Not all of these additional compounds could be identified in all cases.

Pesticides

Of the pesticides identified, the synthetic pyrethroid permethrin was the most frequently encountered (appearing in 17 of the total of 65 samples subjected to this analysis). This frequency of detection (approx. 26% of samples) is very similar to that reported in our earlier study of UK dusts (approx. 24%, Santillo *et al.* 2003), in which permethrin was also the most frequently detected pesticide. The pesticide synergist piperonyl butoxide (PBO), commonly used in synthetic pyrethroid formulations, was found in 14 of the 65 samples analysed in the current study, in the majority of cases in association with pyrethroid residues. Another synthetic pyrethroid, tetramethrin, was found in 3 of the 22 dust samples analysed from Spanish households. The high frequency with which permethrin and PBO, in particular, can be found in indoor dust has been noted in a number of other studies (see Butte and Heinzow 2002 for a useful review). Most recently, Rudel *et al.* (2003) recorded permethrin residues in approximately half of 119 homes sampled in the Cape Cod region of Massachusetts (USA), and at the highest concentrations of all pesticides measured.

It is not possible to determine from these results whether these residues arise from deliberate application of pyrethroid insecticide products for pest control in the home or whether they arise primarily from their presence in pre-treated fabrics such as carpets. As for the UK study, the frequency with which permethrin was found in the current study (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. However, without further details of the carpeting, other furnishings and pesticide use in the homes sampled, it is not possible to speculate further on likely sources in individual cases.

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.

Other pesticide residues identified included the organophosphate insecticide chlorpyrifos in one sample (HD03001) from Germany, *p,p'*-DDD in one sample (HD03038) from Italy, the organophosphate pesticide malathion in sample HD03052 from France and pentachlorophenol in sample HD03070, also from France. Once again it is not possible with the information available to link these findings with any particular sources in the home. It is possible that, in every case, the primary source is contaminated soils, dusts and/or clothing brought in to the home from outside. For the organophosphates chlorpyrifos and malathion, this is particularly possible if the households concerned are situated in an agricultural area or are occupied by one or more people who are occupationally exposed to these agents (Mukerjee *et al.* 1997, Sexton *et al.* 2003). However, the possibility that these residues arise from direct application of pesticide formulations used in the home cannot be ruled out.

Chlorpyrifos appears as a relatively common contaminant in household dust samples collected in the USA (Fenske *et al.* 2002, Pang *et al.* 2002), perhaps reflecting more widespread use for pest control in and around the home. Indeed, Gurunathan *et al*. (1998) highlighted the contribution to daily intake of this pesticide by infants resulting from the adherence of chlorpyrifos residues to toys and other surfaces in the home. More recently, Rudel *et al.* (2003) reported chlorpyrifos in a significant proportion (18%) of all the homes sampled in the Cape Cod area, though in this case it was identified less frequently than residues of permethrin. The appearance of chlorpyrifos in only one of the samples analysed in our study implies that it is less widely used within Europe.

The findings of the organochlorines *p,p'*-DDD (a persistent breakdown product of the banned pesticide DDT) and pentachlorophenol (PCP) in household dusts, although only in one of the households sampled in each case, are of particular concern given the very high persistence and toxicity of these compounds. The appearance of these chemicals warrants further detailed investigations into possible sources of contamination in both individual households. One possible source of PCP in dust is historically treated timber (Schnelle-Kreis *et al.* 2000). Clearly steps should be taken to eliminate any identified sources of these chemicals within the home.

Plasticisers and flame retardants

In addition to the phthalate esters quantified in this study, a number of other commonly used plastics additives were identified in the dust samples. Most prominent in terms of non-phthalate plasticisers were bis-(2-ethylhexyl) adipate (DEHA) and tris-(2-ethylhexyl) trimellitate, which were found in 13 and 9 of the 65 samples screened in this study respectively. DEHA, in particular, was identified with a higher frequency in the samples from Germany, Spain, Slovakia and Italy than in UK dusts. It is interesting to note, however, that none of the French dusts analysed in the current study contained detectable residues of DEHA, though the trimellitate was found in 5 of the 31 samples from France. Rudel *et al*. (2003) reported DEHA residues in dust from all 119 households sampled in the Cape Cod region of Massachusetts, in what appears to be the only other study to date to report the presence of this compound in household dust.

Given the increasing concerns relating to the reproductive toxicity of the commonly used phthalate bis-(2-ethylhexyl) phthalate (DEHP) and the toxicity of other phthalate formulations, use of DEHA as an alternative plasticiser has undoubtedly increased in certain applications in recent years. DEHA has long been used as a plasticiser in food contact PVC films (Petersen and Breindahl 1998). Nevertheless, concerns have also been raised regarding the toxicity of DEHA to foetal development in laboratory studies (Dalgaard *et al.* 2003); although endocrine disrupting effects may not be of relevance to DEHA, the developmental toxicity of this additive may be of significance. The importance of dust as a contributor to overall exposure route to this compound is not known, but clearly is a potential source which cannot be neglected.

Phthalic anhydride, an important precursor used in the manufacture of phthalate ester plasticisers, was identified in 14 of the 65 dust samples screened in the current study. Aside from this common use, phthalic anhydride is also used as a monomer in certain synthetic resins. Whether its appearance in house dusts arises from carry-over as a contaminant from phthalate ester formulations or from other uses cannot be determined.

The organophosphate compound tri-(2-butoxyethanol) phosphonate (tris-(2-butoxyethyl) phosphate, or TBEP) was identified in approximately one third of all samples analysed in the current study (22 or 65 samples screened). This compound, which is widely used as a plasticiser in plastics and rubbers, as a flame retardant and as an additive in water-based paints and floor polishes, also appeared relatively frequently in the UK dusts (7 of 29 samples). Already recognised as a significant contaminant in surface and groundwaters in parts of Europe (Fries and Puttmann 2003), TBEP has also recently been reported in indoor dusts collected in Sweden (Marklund *et al.* 2003).

Triphenyl phosphate (TPP), used as a flame retardant and as a plasticiser in some PVC articles, was also reported as a widespread contaminant of the indoor environment by Marklund *et al.* (2003), albeit at lower concentrations than TBEP. These relatively low concentrations might explain the relative infrequency with which TPP was identified in dusts in our previous (Santillo *et al.* 2003) and current studies. At the same time, it is interesting that the chlorinated organophosphate tris-(3-chloropropyl) phosphonate (possibly the flame retardant TCPP) was not identified in any of the 65 samples screened in the current study, despite being found in almost half of the house dust samples we analysed from the UK.

Other organic contaminants

Aside from the groups discussed above, numerous other organic contaminants were identified in the dust samples. Among the most frequently encountered were a group of fatty tertiary amines (N,N-dimethyldodecan-1-amine and closely related compounds). For example, around half the dust samples from Spain contained N,N-dimethyldodecan-1-amine and/or N,N-dimethyloctadec-1-amine. These compounds are known to be used as intermediates in the manufacture of cationic detergents, in fuel detergent formulations and as processing aids in the manufacture of certain types of rubber. Their widespread presence in this sample set could result from common use in one or more groups of household consumer products, though identification of specific sources would require more detailed research. Very little is known about the possible hazards, if any, of long-term household exposure to these chemicals in dust.

Other frequently identified chemicals included hexadecyl-2-ethylhexanoate and octadecyl-2-ethylhexanoate, probably constituents of emollient formulations (perhaps arising from personal care products), and 2-ethylhexyl dodecanoate and 2-ethylhexyl tetradecanoate, whicih are both used as surfactant ingredients. The cinnamate chemical 3-(4-methoxyphenyl)-2-ehtylhexylpropenoate (Parsol), also found relatively commonly in dust samples from Europe, could be entering house dust from its use in UV sun screen creams or other similar preparations.

Although Table X lists those organic contaminants which were detected relatively frequently in the dust samples from the five European countries, certain other findings in a small number or even in individual samples are worth noting. For example, the endocrine disrupting chemical bisphenol-A was identified in one of the samples from Italy (HD03036), along with the chemical diethyl toluamide (DEET) which is commonly used in insect repellent formulations. Four samples contained detectable residues of toluene-2,4-diisocyanate (TDI), a hazardous chemical intermediate used in polyurethane manufacture, while three contained the solvent butan-2-one (methyl ethyl ketone, or MEK).

Three of the five samples collected from households in Germany contained a variety of so far unidentified alkoxybenzene compounds. The precise nature and origin of these, and why they appeared only in these samples from Germany, is simply not known. In addition, one of the three German dust samples (HD03003) contained four organochlorine compounds, namely 1,1-dichloro-2-ethenylcyclopropane, 1,1,3-trichloro-2-methylprop-1-ene, 1,1,2-trichloroprop-1-ene and a tetrachloropyridine derivative. Once again, the origins of these rather unusual chemicals in house dust, and the significance of these residues in terms of chemical hazard and exposure, are not currently known.

Table 1: Summary of phthalate ester concentrations (medians, ranges and frequencies detected) in dust samples from five countries, compared to previous data for UK dusts

Compound	**Germany**	**Spain**	**France**	**Italy (Roma)**	**Slovakia**	**UK**
Phthalate esters	ug/g dust (parts per million, ppm) [median values, (range), frequency detected]					
DMP	1.42 (<0.1-2.83) 3/5	<0.1 (<0.1-0.92) 2/22	<0.1 (<0.1) 0/31	<0.1 (<0.1-1.5) 1/5	* (<0.1) 0/2	<0.1 (<0.1-1.1) 11/29
DEP	12.9 (1.86-368) 5/5	5.33 (1.09-64.6) 22/22	6.87 (<0.1-49.4) 29/31	6.78 (1.92-23.6) 5/5	* (1.22-4.84) 2/2	3.5 (0.6-114.8) 29/29
DiBP	36.5 (27.9-358) 5/5	148.9 (66.1-409) 22/22	118.8 (16.7-488) 31/31	180.1 (158.2-370.7) 5/5	* (137-149) 2/2	43.2 (0.2-157.4) 29/29
DnBP	44.1 (22.3-1511) 5/5	79.4 (48.6-201) 22/22	55.3 (11.6-624) 31/31	42.8 (22.8-46.8) 5/5	* (600-1029) 2/2	52.8 (0.1-106.4) 29/29
BBP	82.2 (4.4-218) 5/5	4.54 (0.81-153) 22/22	28.2 (<0.1-3551) 30/31	23.6 (9.0-308) 5/5	* (3.8-5.4) 2/2	24.5 (<0.1-238.9) 28/29
DEHP	996 (547-1586) 5/5	317.2 (113-2151) 22/22	504.6 (14.9-3289) 31/31	434.3 (314-933) 5/5	* (1290-2124) 2/2	195.4 (0.5-416.4) 29/29
DiNP	113 (<0.1-250) 4/5	<0.1 (<0.1-717) 6/22	115.3 (<0.1-466) 18/31	<0.1 (<0.1-532) 2/5	* (146-173) 2/2	<0.1 (<0.1-337.2) 11/29
DiDP	<0.1 (<0.1-67.7) 1/5	<0.1 (<0.1-98.8) 6/22	<0.1 (<0.1-170) 11/31	<0.1 (<0.1-380) 1/5	* (<0.1) 0/2	<0.1 (<0.1-156.6) 11/29
Total phthalates	**1287 (1121-3168)**	**706.2 (291-2644)**	**956.5 (43.2-7237)**	**808.1 (552-1992)**	*** (2208-3454)**	**354.3 (1.6-1019)**

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.
- * - two composite samples only – median not calculated

NB: UK data from Santillo *et al.* (2003)

Table 2: Summary of alkylphenol concentrations (medians, ranges and frequencies detected) in dust samples from five countries, compared to previous data for UK dusts

Compound	**Germany**	**Spain**	**France**	**Italy (Roma)**	**Slovakia**	**UK**
Alkylphenols	ug/g dust (parts per million, ppm) [median value, (range), frequency detected]					
4TMBP	<0.1 (<0.1) 0/5	<0.1 (<0.1) 0/22	<0.1 (<0.1) 0/31	<0.1 (<0.1) 0/5	* (<0.1) 0/2	<0.1 (<0.1-2.4) 4/29
4OP	<0.1 (<0.1) 0/5	<0.1 (<0.1) 0/22	<0.1 (<0.1) 0/31	<0.1 (<0.1) 0/5	* (<0.1) 0/2	<0.1 (<0.1-8.6) 1/29
4NP	<0.1 (<0.1) 0/5	<0.1 (<0.1-4.5) 5/22	<0.1 (<0.1-3.35) 1/31	<0.1 (<0.1-0.22) 1/5	* (1.27-2.53) 2/2	9.8 (<0.1-35.2) 22/29
Total alkylphenols	**<0.1** **(<0.1)**	**<0.1** **(<0.1-4.5)**	**<0.1** **(<0.1-3.35)**	**<0.1** **(<0.1-0.22)**	***** **(1.27-2.53)**	**9.8** **(<0.1-36.1)**

Abbreviations

- *Alkylphenol compounds:* 4TMBP - 4-(1,1,3,3-tert-methylbutyl)phenol, 4OP – 4-n-octylphenol, 4NP - 4-nonylphenol.
- * - two composite samples only – median not calculated

NB: UK data from Santillo *et al.* (2003)

Table 3: Summary of organotin compound concentrations (medians, ranges and frequencies detected) in dust samples from five countries, compared to previous data for UK dusts

Compound	**Germany**	**Spain**	**France**	**Italy (Roma)**	**Slovakia**	**UK**
Organotin compounds	ng/g dust (parts per billion, ppb)[median value, (range), frequency detected]					
MBT	(1400)	631 (449-978) 4/4	1300 (425-6950) 8/8	(917)	n/a	1350 (810-2800) 10/10
DBT	(255)	202.5 (67.7-342) 4/4	150 (49.8-1150) 8/8	(317)	n/a	519 (157-1300) 10/10
TBT	(13.4)	16.2 (5.3-32.3) 4/4	15 (3.1-521) 8/8	(11.6)	n/a	49.9 (21.6-759) 10/10
TeBT	(<1)	<1 (<1) 0/4	<1 (<1) 0/8	(<1)	n/a	<1 (<1) 0/10
MOT	(270)	554 (520-632) 4/4	367.5 (136-10700) 8/8	(481)	n/a	349 (82.5-1300) 10/10
DOT	(17.9)	61 (35.7-83.4) 4/4	14.5 (<1-2490) 7/8	(63.4)	n/a	62.7 (17.6-545) 10/10
TCHT	(<1)	<1 (<1) 0/4	<1 (<1) 0/8	(<1)	n/a	<1 (<1) 0/10
TPT	(<1)	<1 (<1) 0/4	<1 (<1) 0/8	(<1)	n/a	<1 (<1-68.9) 1/10
Total organotins	**1956**	**1495** **(1125-1958)**	**2234** **(657-18155)**	**1790**	n/a	**2432** **(1581-5047)**

Abbreviations:

- *Organotin compounds*: MBT – monobutyltin, DBT – dibutyltin, TBT – tributyltin, TeBT – tetrabutyltin, MOT – monooctyltin, DOT – dioctyltin, TCHT – tricyclohexyltin, TPT – triphenyltin.
- n/a – not analysed for these samples

NB: UK data from Santillo *et al.* (2003)

Table 4: Summary of brominated flame retardant concentrations (medians, ranges and frequencies detected) in dust samples from five countries, compared to previous data for UK dusts

Compound	**Germany**	**Spain**	**France**	**Italy (Roma)**	**Slovakia**	**UK**
Brominated flame retardants	ng/g dust (parts per billion, ppb) [median value, (range), frequency detected]					
BDE-28 (tri-)	(<0.3) 0/1	<0.3 (<0.1-0.7) 1/4	0.25 (0.1-4.8) 6/8	(<0.3) 0/1	n/a	0.35 (<0.1-33) 7/10
BDE-47 (tetra-)	(31) 1/1	13 (11-16) 4/4	24 (7.3-260) 8/8	(23) 1/1	n/a	24.8 (10-1980) 10/10
BDE-99 (penta-)	(37) 1/1	17.5 (14-21) 4/4	28.5 (15-720) 8/8	(36) 1/1	n/a	44 (18-2100) 10/10
BDE-153 (hexa-)	*	*	*	*	n/a	23 (<0.1-170) 9/10
BDE-190 (hepta)	(1.8) 1/1	4.1 (1.7-39) 4/4	8.0 (2.6-44) 8/8	(62) 1/1	n/a	9.5 (<0.1-87) 7/10
BDE-209 (deca-)	(2800)	425 (92-1700) 4/4	420 (69-3400) 8/8	(1600)	n/a	7100 (3800-19900) 10/10
HBCD	(1200) 1/1	225 (190-850) 4/4	485 (77-1600) 8/8	(250) 1/1	n/a	3250 (940-6900) 10/10
TBBP-A	*	*	*	*	n/a	<10 (<10-340) 4/10

Abbreviations:

- *Brominated Flame Retardants*: BDE - brominated diphenylethers (tribromo- to decabromo-), HBCD – hexabromocyclododecane, TBBP-A – tetrabromobisphenol-A.
- n/a – not analysed for these samples
- * - quantification not possible due to co-elution interference

NB: UK data from Santillo et al. (2003)

Table 5: Summary of short-chain chlorinated paraffin concentrations (medians, ranges and frequencies detected) in dust samples from five countries, compared to previous data for UK dusts

Compound	**Germany**	**Spain**	**France**	**Italy (Roma)**	**Slovakia**	**UK**
Short-chain chlorinated paraffins	ug/g dust (parts per million, ppm)[median value, (range), frequency detected]					
SCCPs	**(48)**	**25 (17-41) 4/4**	**45 (30-95) 8/8**	**(34)**	**(24-66) 2/2**	**3.7 (<0.12-13.0) 8/10**

NB: UK data from Santillo *et al.* (2003)

Table 6: summary of other (non-target) organic compounds identified in the 65 samples of dust analysed from across Europe, with an indication of the frequency with which they were found.

	Germany	**Spain**	**Slovakia**	**Italy**	**France**
Pesticides					
Permethrin	1/5	8/22	1/2	1/5	6/31
Tetramethrin	0	3/22	0	0	0
Piperonyl butoxide	1/5	2/22	0	2/5	9/31
Plasticisers and flame retardants					
Diheptyl phthalate	1/5	0	0	0	2/31
Phthalic anhydride	2/5	3/22	0	1/5	8/31
Bis-(2-ethylhexyl) adipate	2/5	7/22	2/2	2/5	0
Tris(2-Ethylhexyl)trimellitate	0	2/22	0	2/5	5/31
Tributyl citrate	2/5	3/22	0	0	3/31
Tri-(2-butoxyethanol)phosphonate	2/5	5/22	1/2	3/5	11/31
Triphenyl phosphate	0	0	0	1/5	2/31
Other organic contaminants					
2-Ethylhexyl dodecanoate	1/5	4/22	0	3/5	3/31
2-Ethylhexyl tetradecanoate	1/5	4/22	0	0	3/31
N,N-Dimethyldodecan-1-amine	1/5	11/22	0	5/5	13/31
N,N-Dimethyltetradec-1-amine	0	5/22	0	4/5	10/31
N,N-Dimethylhexadec-1-amine	0	6/22	0	0	3/31
N,N-Dimethyloctadec-1-amine	1/5	10/22	0	0	5/31
N,N,N',N'-Tetraacetylethylenediamine (EDTA)	1/5	10/22	0	1/5	8/31
3-(4-Methoxyphenyl)-2-ethylhexylpropenoate	2/5	9/22	0	2/5	5/31
Hexadecyl 2-ethylhexanoate	2/5	16/22	0	4/5	3/31
Octadecyl 2-ethylhexanoate	1/5	12/22	0	2/5	2/31
PAHs	3/5	0	0	0	3/31

Conclusions

The results of this most recent study, together with those previously reported for dusts from the UK, reaffirm the widespread contamination of household dusts with a variety of hazardous chemicals, including brominated flame retardants, organotin compounds, phthalates, alkyphenols 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 data from either study 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 across Europe.

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 countries, cities or regions sampled. Nevertheless, these data as a whole do provide a snap-shot of chemical contamination in the indoor environment in a number of countries across 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 these studies do not provide (and indeed were 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 complacence. 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 (BDE-209), 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, but 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 avoided altogether 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.

A very recent study published by the Greenpeace Environmental Trust (Greenpeace 2003) lists a diversity of examples of how the substitution of a number of hazardous chemicals and materials with safer alternatives has already been achieved in practice.

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.

Now that the REACH legislation for control of chemical use is about to be finalized by the European Commission, we can only hope that this new system will provide for the high level of protection for the environment and health to which we all have a right. 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 is the only way in which the chemical safety of our home environment can ultimately be ensured.

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Annex 1 : detailed results for target and non-target compounds in individual and pooled samples

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 – monooctyltin, DOT – dioctyltin, TCHT – tricyclohexyltin, TPT – triphenyltin.

NB**:** * - indicates co-elution interference prevented quantitation of BDE-153 and TBBP-A

Phthalate esters – individual sample analyses

COUNTRY: GERMANY

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
		DMP	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03001	**Hamburg**	2.34	13.4	358.1	44.1	4.4	546.8	249.7	67.7	**1286.6**
HD03002	**Regensberg**	1.42	12.9	27.9	1510.9	218.4	1306.5	89.7	<0.1	**3167.7**
HD03003	**Leipzig**	2.83	1.86	30.8	308.8	82.2	695.7	115.5	<0.1	**1237.6**
HD03004	**Berlin**	<0.1	7.48	82.6	22.3	13.4	995.7	<0.1	<0.1	**1121.4**
HD03005		<0.1	367.7	36.5	33.4	110.8	1586.1	112.7	<0.1	**2247.2**
Germany mean (average) value		1.32	80.7	107.1	383.9	85.8	1026.2	113.5	13.5	**1812.1**
Germany median (middle) value		1.42	12.9	36.5	44.1	82.2	995.7	112.7	<0.1	**1286.6**
Germany minimum (lowest) value		<0.1	1.86	27.9	22.3	4.4	546.8	<0.1	<0.1	**1121.4**
Germany maximum (highest) value		2.83	367.7	358.1	1510.9	218.4	1586.1	249.7	67.7	**3167.7**

Phthalate esters – individual sample analyses

COUNTRY: SPAIN

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Madrid		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03006		<0.1	7.88	336.7	78.8	8.3	370.1	<0.1	<0.1	**801.8**
HD03007		<0.1	13.2	201.0	144.8	11.1	583.8	289.3	<0.1	**1243.3**
HD03008		<0.1	5.13	409.1	149.1	4.5	286.4	<0.1	<0.1	**854.3**
HD03009		<0.1	3.41	197.9	201.6	3.5	293.2	<0.1	51.0	**750.6**
HD03010		0.70	40.6	179.5	80.8	5.3	785.9	58.0	<0.1	**1150.7**
Madrid mean (average) value		0.14	14.0	264.8	131.0	6.55	463.9	69.5	10.2	**960.1**
Madrid median (middle) value		<0.1	7.88	201.0	144.8	5.31	370.1	<0.1	<0.1	**854.3**
Granada										
HD03011		<0.1	4.92	257.1	81.7	4.1	267.4	<0.1	<0.1	**615.2**
HD03012		<0.1	1.09	174.4	72.3	<0.1	159.7	<0.1	<0.1	**407.4**
HD03013		<0.1	2.95	137.8	76.6	2.0	211.6	<0.1	<0.1	**431.0**
HD03014		<0.1	1.87	173.9	64.0	1.9	112.6	<0.1	<0.1	**354.2**
HD03015		<0.1	13.0	121.7	52.8	0.8	137.0	<0.1	<0.1	**325.2**
HD03016		<0.1	5.30	200.3	99.3	153.4	1917	112.8	72.2	**2560**
Granada mean (average) value		<0.1	4.85	177.5	74.4	32.4	467.6	18.8	12.0	**782.3**
Granada median (middle) value		<0.1	3.94	174.1	74.4	2.00	185.6	<0.1	<0.1	**419.2**

Phthalate esters – individual sample analyses

COUNTRY: SPAIN (CONTINUED)

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Valencia		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03017		<0.1	1.54	142.4	62.2	1.8	1064	<0.1	<0.1	**1273**
HD03018		<0.1	5.37	127.9	70.2	9.1	315.0	<0.1	<0.1	**527.6**
HD03019		<0.1	2.58	189.7	90.6	2.4	272.4	<0.1	6.03	**563.8**
HD03020		<0.1	4.40	131.8	64.6	10.9	189.8	<0.1	<0.1	**401.4**
HD03021		<0.1	64.6	76.7	53.9	7.3	319.4	83.7	<0.1	**605.7**
HD03022		<0.1	34.9	113.0	60.6	6.6	507.0	<0.1	<0.1	**722.1**
Valencia mean (average) value		<0.1	18.9	130.3	67.0	6.36	444.7	14.0	1.01	**682.2**
Valencia median (middle) value		<0.1	4.89	129.8	63.4	6.94	317.2	<0.1	<0.1	**584.7**
Asturias/Leon										
HD03023		<0.1	9.52	140.7	80.1	2.9	358.3	<0.1	98.8	**690.3**
HD03024		<0.1	6.57	82.2	135.5	9.7	2151	231.3	27.2	**2644**
HD03025		0.92	5.82	155.4	131.7	4.2	818.2	<0.1	39.3	**1156**
HD03026		<0.1	3.67	66.1	48.6	4.1	169.0	<0.1	<0.1	**291.4**
HD03027		<0.1	16.2	107.1	96.0	5.4	407.0	717.0	<0.1	**1349**
Asturias mean (average) value		0.18	8.35	110.3	98.4	5.27	780.7	189.7	33.1	**1226**
Asturias median (middle) value		<0.1	6.57	107.1	96.0	4.20	407.0	<0.1	27.2	**1156**

Phthalate esters – individual sample analyses

COUNTRY: FRANCE

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Lille		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03043		<0.1	2.23	126.7	64.2	16.4	298.2	<0.1	<0.1	**507.8**
HD03045		<0.1	9.53	64.8	66.1	11.2	456.2	18.0	<0.1	**626.0**
HD03047		<0.1	<0.1	65.8	51.3	5.50	372.5	213.2	62.2	**770.5**
HD03049		<0.1	13.0	125.2	55.3	734.3	2639	186.6	104.7	**3858**
HD03050		<0.1	3.81	201.5	102.7	10.4	266.8	<0.1	<0.1	**585.2**
Lille mean (average) value		<0.1	5.71	116.8	67.9	155.6	806.6	83.6	33.4	**1270**
Lille median (middle) value		<0.1	3.81	125.2	64.2	11.2	372.5	18.0	<0.1	**626.0**
Toulouse										
HD03052		<0.1	10.0	67.8	27.9	9.33	941.4	301.8	115.3	**1474**
HD03053		<0.1	12.6	106.1	18.4	4.27	246.0	<0.1	<0.1	**387.5**
HD03054		<0.1	2.73	118.8	120.6	48.6	427.5	155.6	85.0	**958.8**
HD03055		<0.1	2.69	139.6	72.6	315.9	2172	<0.1	<0.1	**2702**
HD03061		<0.1	32.9	112.2	99.7	48.4	1065.1	183.9	<0.1	**1542**
Toulouse mean (average) value		<0.1	12.2	108.9	67.8	85.3	970.3	128.3	40.1	**1413**
Toulouse median (middle) value		<0.1	10.0	112.2	72.6	48.4	941.4	155.6	<0.1	**1474**

Phthalate esters – individual sample analyses

COUNTRY: FRANCE (CONTINUED)

Sample code **Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Lyon	**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03070	<0.1	6.87	136.5	35.1	28.2	1444	160.7	<0.1	**1812**
HD03071	<0.1	5.57	75.1	15.1	17.9	275.7	<0.1	<0.1	**389.4**
HD03075	<0.1	4.67	159.0	168.0	332.7	753.2	140.3	170.2	**1728**
HD03076	<0.1	15.2	40.6	29.7	6.65	155.4	<0.1	75.4	**322.9**
HD03077	<0.1	3.43	154.4	18.0	3.26	118.3	<0.1	<0.1	**297.4**
Lyon mean (average) value	<0.1	7.14	113.1	53.2	77.7	549.5	60.2	49.1	**910.0**
Lyon median (middle) value	<0.1	5.57	136.5	29.7	17.9	275.7	<0.1	<0.1	**389.4**
Nantes									
HD03081	<0.1	<0.1	16.7	11.6	<0.1	14.9	<0.1	<0.1	**43.2**
HD03082	<0.1	5.28	134.7	58.6	1006	2507	221.9	<0.1	**3933**
HD03084	<0.1	2.91	49.0	127.4	28.2	596.9	105.3	<0.1	**909.8**
HD03087	<0.1	7.10	165.4	37.9	18.1	504.6	136.9	<0.1	**870.0**
HD03088	<0.1	4.18	487.6	33.9	10.6	989.9	<0.1	<0.1	**1526**
Nantes mean (average) value	<0.1	3.90	170.7	53.9	212.6	923.0	92.8	<0.1	**1456**
Nantes median (middle) value	<0.1	4.18	134.7	37.9	18.1	596.9	105.3	<0.1	**909.7**

Phthalate esters – individual sample analyses

COUNTRY: FRANCE (CONTINUED)

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Paris		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03091		<0.1	12.4	94.6	33.2	481.5	2750	<0.1	<0.1	**3371**
HD03093		<0.1	8.72	73.8	65.8	11.9	322.5	298.4	122.3	**903.4**
HD03095		<0.1	6.93	133.9	624.2	419.0	1792.7	197.3	<0.1	**3174**
HD03097		<0.1	6.91	86.1	104.7	23.8	332.0	115.3	118.7	**787.5**
HD03098		<0.1	14.4	68.7	43.9	200.1	356.2	<0.1	<0.1	**683.3**
Paris mean (average) value		<0.1	9.88	91.4	174.4	227.3	1111	122.2	48.2	**1784**
Paris median (middle) value		<0.1	8.72	86.1	65.8	200.1	356.2	115.3	<0.1	**903.4**

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Individual samples		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03062	**SP01**	<0.1	5.30	151.7	119.2	3551	3289	<0.1	118.8	**7235**
HD03063	**SP02**	<0.1	4.89	220.5	168.9	45.8	159.1	<0.1	<0.1	**599.2**
HD03064	**SP03**	<0.1	49.4	88.8	44.4	161.2	1062	326.4	166.7	**1899**
HD03065	**SP04**	<0.1	19.7	250.3	27.4	370.7	343.1	466.2	83.4	**1561**
HD03058	**SP05**	<0.1	10.1	48.1	43.2	7.77	713.5	133.8	<0.1	**956.5**
HD03066	**SP06**	<0.1	3.47	127.3	144.7	28.3	2650	154.4	<0.1	**3109**

Phthalate esters – individual sample analyses

COUNTRY: ITALY (ROMA)

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Italy (Roma)		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
HD03036		<0.1	6.78	138.3	45.2	308.4	933.4	180.0	380.0	**1992**
HD03037		1.50	11.4	370.7	42.8	14.9	366.8	<0.1	<0.1	**808.1**
HD03038		<0.1	1.92	180.1	46.8	9.0	314.2	<0.1	<0.1	**552.0**
HD03039		<0.1	23.6	158.2	25.3	89.2	467.0	531.7	<0.1	**1295**
HD03040		<0.1	4.31	257.6	22.8	23.6	434.3	<0.1	<0.1	**742.6**
Italy mean (average) value		0.30	9.60	221.0	36.6	89.0	503.1	142.3	76.0	**1078**
Italy median (middle) value		<0.1	6.78	180.1	42.8	23.6	434.3	<0.1	<0.1	**808.1**
Italy minimum (lowest) value		<0.1	1.92	158.2	22.8	9.0	314.2	<0.1	<0.1	**552.0**
Italy maximum (highest) value		1.50	23.6	370.7	46.8	308.4	933.4	531.7	380.0	**1992**

COUNTRY: SLOVAKIA

Sample code	**Location**	**Concentration of phthalate esters (ug/g dust, parts per million, ppm)**								
Slovakia		**DMP**	**DEP**	**DiBP**	**DnBP**	**BBP**	**DEHP**	**DiNP**	**DiDP**	**Total**
Slovakia I	**South West**	<0.1	1.22	149.0	1028.6	5.4	2123.8	145.8	<0.1	**3454**
Slovakia II	**North East**	<0.1	4.84	137.0	599.8	3.8	1289.6	172.5	<0.1	**2208**

Alkylphenols – individual sample analyses

COUNTRY: GERMANY

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)			
		4TMBP	4OP	4NP	Total
HD03001	**Hamburg**	<0.1	<0.1	<0.1	<0.1
HD03002	**Regensberg**	<0.1	<0.1	<0.1	<0.1
HD03003	**Leipzig**	<0.1	<0.1	<0.1	<0.1
HD03004	**Berlin**	<0.1	<0.1	<0.1	<0.1
HD03005		<0.1	<0.1	<0.1	<0.1

COUNTRY: SPAIN

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)			
Madrid		4TMBP	4OP	4NP	Total
HD03006		<0.1	<0.1	<0.1	<0.1
HD03007		<0.1	<0.1	<0.1	<0.1
HD03008		<0.1	<0.1	<0.1	<0.1
HD03009		<0.1	<0.1	<0.1	<0.1
HD03010		<0.1	<0.1	<0.1	<0.1

Alkylphenols – individual sample analyses

COUNTRY: SPAIN (CONTINUED)

Sample code	**Location**	**Concentration of alkylphenols (ug/g dust, parts per million, ppm)**			
Granada		**4TMBP**	**4OP**	**4NP**	**Total**
HD03011		<0.1	<0.1	<0.1	<0.1
HD03012		<0.1	<0.1	<0.1	<0.1
HD03013		<0.1	<0.1	<0.1	<0.1
HD03014		<0.1	<0.1	<0.1	<0.1
HD03015		<0.1	<0.1	<0.1	<0.1
HD03016		<0.1	4.49	<0.1	4.49
Valencia					
HD03017		<0.1	0.75	<0.1	0.75
HD03018		<0.1	<0.1	<0.1	<0.1
HD03019		<0.1	<0.1	<0.1	<0.1
HD03020		<0.1	<0.1	<0.1	<0.1
HD03021		<0.1	<0.1	<0.1	<0.1
HD03022		<0.1	<0.1	<0.1	<0.1
Asturias/Leon					
HD03023		<0.1	<0.1	<0.1	<0.1
HD03024		<0.1	0.68	<0.1	0.68
HD03025		<0.1	<0.1	<0.1	<0.1
HD03026		<0.1	0.67	<0.1	0.67
HD03027		<0.1	<0.1	<0.1	<0.1

Alkylphenols – individual sample analyses

COUNTRY: FRANCE

Sample code	Location	Concentration of alkylphenols (ug/g dust, parts per million, ppm)			
Lille		**4TMBP**	**4OP**	**4NP**	**Total**
HD03043		<0.1	<0.1	<0.1	<0.1
HD03045		<0.1	<0.1	<0.1	<0.1
HD03047		<0.1	<0.1	<0.1	<0.1
HD03049		<0.1	<0.1	<0.1	<0.1
HD03050		<0.1	<0.1	<0.1	<0.1
Toulouse					
HD03052		<0.1	<0.1	<0.1	<0.1
HD03053		<0.1	<0.1	<0.1	<0.1
HD03054		<0.1	<0.1	<0.1	<0.1
HD03055		<0.1	<0.1	<0.1	<0.1
HD03061		<0.1	<0.1	<0.1	<0.1
Lyon					
HD03070		<0.1	<0.1	<0.1	<0.1
HD03071		<0.1	<0.1	<0.1	<0.1
HD03075		<0.1	<0.1	<0.1	<0.1
HD03076		<0.1	<0.1	<0.1	<0.1
HD03077		<0.1	<0.1	<0.1	<0.1

Alkylphenols – individual sample analyses

COUNTRY: FRANCE (CONTINUED)

Sample code	**Location**	**Concentration of alkylphenols (ug/g dust, parts per million, ppm)**			
Nantes		**4TMBP**	**4OP**	**4NP**	**Total**
HD03081		<0.1	<0.1	<0.1	<0.1
HD03082		<0.1	<0.1	<0.1	<0.1
HD03084		<0.1	<0.1	<0.1	<0.1
HD03087		<0.1	<0.1	<0.1	<0.1
HD03088		<0.1	<0.1	<0.1	<0.1
Paris					
HD03091		<0.1	<0.1	<0.1	<0.1
HD03093		<0.1	<0.1	<0.1	<0.1
HD03095		<0.1	<0.1	<0.1	<0.1
HD03097		<0.1	<0.1	<0.1	<0.1
HD03098		<0.1	<0.1	<0.1	<0.1
Individual samples					
HD03062	**SP01**	<0.1	<0.1	<0.1	<0.1
HD03063	**SP02**	<0.1	<0.1	<0.1	<0.1
HD03064	**SP03**	<0.1	<0.1	<0.1	<0.1
HD03065	**SP04**	<0.1	<0.1	3.35	3.35
HD03058	**SP05**	<0.1	<0.1	<0.1	<0.1
HD03066	**SP06**	<0.1	<0.1	<0.1	<0.1

Alkylphenols – individual sample analyses

COUNTRY: ITALY (ROMA)

Sample code	**Location**	**Concentration of alkylphenols (ug/g dust, parts per million, ppm)**			
Italy (Roma)		**4TMBP**	**4OP**	**4NP**	**Total**
HD30036		<0.1	<0.1	0.22	0.22
HD30037		<0.1	<0.1	<0.1	<0.1
HD30038		<0.1	<0.1	<0.1	<0.1
HD30039		<0.1	<0.1	<0.1	<0.1
HD30040		<0.1	<0.1	<0.1	<0.1

COUNTRY: SLOVAKIA

Sample code	**Location**	**Concentration of alkylphenols (ug/g dust, parts per million, ppm)**			
Slovakia		**4TMBP**	**4OP**	**4NP**	**Total**
Slovakia I	**South West**	<0.1	<0.1	2.53	2.53
Slovakia II	**North East**	<0.1	<0.1	1.27	1.27

Organotins – pooled and individual sample analyses

Country/region	**Concentration of organotin compounds (ng/g dust, parts per billion, ppb)**								
	MBT	**DBT**	**TBT**	**TeBT**	**MOT**	**DOT**	**TCHT**	**TPT**	**Total organotins**
Germany									
Single composite	1400	255	13.4	<1	270	17.9	<1	<1	**1956**
Spain									
Madrid	563	273	7	<1	530	81.8	<1	<1	**1455**
Granada	449	67.7	5.31	<1	520	83.4	<1	<1	**1125**
Valencia	978	342	25.4	<1	577	35.7	<1	<1	**1958**
Asturias/Leon	699	132	32.3	<1	632	40.2	<1	<1	**1536**
Mean (average) value	*672*	*204*	*17.5*	*<1*	*565*	*60.3*	*<1*	*<1*	***1518***
Median (middle) value	*631*	*202.5*	*16.2*	*<1*	*553.5*	*61*	*<1*	*<1*	***1495***
France									
Lille	1290	149	49.8	<1	2550	1360	<1	<1	**5399**
Toulouse	6950	992	521	<1	322	21	<1	<1	**8806**
Lyon	1040	135	15.6	<1	141	<1	<1	<1	**1332**
Nantes	3800	1150	15.1	<1	10700	2490	<1	<1	**18155**
Paris	1910	292	11.6	<1	413	14.7	<1	<1	**2641**
HD03058 (SP05)	425	72.7	14.9	<1	136	7.9	<1	<1	**657**
HD03062 (SP01)	610	151	12.8	<1	140	14.2	<1	<1	**928**
HD03064 (SP03)	1310	49.8	3.1	<1	452	12.1	<1	<1	**1827**
Mean (average) value	*2167*	*373.9*	*80.5*	*<1*	*1857*	*490*	*<1*	*<1*	***4968***
Median (middle) value	*1300*	*150*	*15*	*<1*	*367.5*	*14.5*	*<1*	*<1*	***2234***
Italy (Roma)									
Single composite	917	317	11.6	<1	481	63.4	<1	<1	**1790**

Brominated flame retardants – pooled and individual sample analyses

Country/region	**Concentration of individual brominated diphenyl ether congeners (ng/g dust, parts per billion, ppb)**																
	Tri-	**Tetra-**						**Penta-**				**Hexa-**			**Hepta-**		**Deca-**
	28	**47**	**49**	**66**	**71**	**75**	**77**	**85**	**99**	**100**	**119**	**138**	**153***	**154**	**183**	**190**	**209**
Germany																	
Single composite	<0.3	31	<0.1	<0.3	15	<0.3	<0.3	2.4	37	8.0	<0.3	<0.5	*	4.5	1.8	<2.3	2800
Spain																	
Madrid	0.7	13	0.4	0.9	12	<0.3	<0.3	<0.4	14	2.4	<0.3	<0.4	*	3.0	39	<2.1	430
Granada	<0.1	16	0.4	0.6	3.5	<0.3	<0.3	1.1	21	4.1	<0.3	<0.5	*	1.8	3.3	<2.3	420
Valencia	<0.3	13	<0.1	<0.3	4.8	<0.3	<0.3	1.1	16	3.3	<0.3	<0.4	*	1.5	4.8	<2.0	1700
Asturias/Leon	<0.3	11	0.1	<0.3	2.2	<0.3	<0.3	1.1	19	4.2	0.9	<0.4	*	1.8	1.7	<2.2	92
Mean (average) value	<0.3	13.3	0.23	0.38	5.63	<0.3	<0.3	0.83	17.5	3.5	<0.3	<0.4	*	1.7	12.2	<2.2	660
Median (middle) value	<0.3	13	0.25	0.3	4.15	<0.3	<0.3	1.1	17.5	3.7	<0.3	<0.4	*	.8	4.1	<2.2	425
France																	
Lille	0.3	41	<0.1	0.4	42	<0.3	<0.3	2.4	48	10	<0.3	<0.4	*	3.4	3.5	<2.1	500
Toulouse	0.3	73	0.3	0.5	16	<0.3	<0.3	4.3	78	17	0.7	<0.4	*	1.0	9.2	<2.2	1800
Lyon	<0.3	11	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	20	4.5	<0.3	<0.3	*	2.6	2.6	<0.3	3400
Nantes	0.1	32	0.3	<0.3	7.4	<0.3	<0.3	2.0	37	7.9	<0.3	<0.5	*	4.3	9.8	<2.4	300
Paris	0.2	13	0.2	<0.3	2.4	<0.3	<0.3	0.9	16	3.7	<0.3	<0.4	*	2.1	6.8	<2.1	340
HD03058 (SP05)	4.8	260	3.7	7.3	<0.4	<0.3	<0.3	43	720	92	<0.3	11	*	64	4.1	<2.2	69
HD03062 (SP01)	<0.3	7.3	<0.1	<0.3	12	<0.3	<0.3	<0.4	7.5	1.4	<0.3	<0.5	*	1.1	44	<2.3	140
HD03064 (SP03)	1.9	16	2.3	1.2	<0.5	<0.3	<0.3	<0.4	15	2.6	<0.3	<0.5	*	2.8	29	<2.4	800
Mean (average) value	0.95	56.7	0.85	1.18	9.98	<0.3	<0.3	7.23	117.7	17.4	<0.3	1.38	*	11.5	13.6	<2.2	919
Median (middle) value	0.25	24	0.25	<0.3	4.9	<0.3	<0.3	0.9	28.5	6.2	<0.3	<0.4	*	2.8	8.0	<2.2	420
Italy (Roma)																	
Single composite	<0.3	23	<0.1	<0.3	29	<0.3	<0.3	1.9	36	6.5	<0.3	2.3	*	6.5	62	<2.5	1600

Brominated flame retardants – pooled and individual sample analyses (continued)

Country/region	**Concentration of additional brominated flame retardants (ng/g, ppb)**							
	Brominated biphenyls (PBBs)					**HBCD**	**TBBP-A**	**methyl-TBBP-A**
	BB-15	**BB-49**	**BB-52**	**BB-101**	**BB-209**			
Germany								
Single composite	<0.3	<0.3	<0.3	<0.3	<3	1200	*	<0.1
Spain								
Madrid	<0.3	<0.3	<0.3	<0.3	<3	850	*	<0.1
Granada	<0.3	<0.3	<0.3	<0.3	<3	190	*	<0.1
Valencia	<0.3	<0.3	<0.3	<0.3	<3	250	*	<0.1
Asturias/Leon	<0.3	<0.3	<0.3	<0.3	<3	200	*	<0.1
Mean (average) value	<0.3	<0.3	<0.3	<0.3	<3	373	*	<0.1
Median (middle) value	<0.3	<0.3	<0.3	<0.3	<3	225	*	<0.1
France								
Lille	<0.3	<0.3	<0.3	<0.3	<3	1600	*	<0.1
Toulouse	<0.3	<0.3	<0.3	<0.3	<3	830	*	<0.1
Lyon	<0.3	<0.3	<0.3	<0.3	<3	470	*	<0.3
Nantes	<0.3	<0.3	<0.3	<0.3	<3	660	*	<0.2
Paris	<0.3	<0.3	<0.3	<0.3	<3	240	*	<0.1
HD03058 (SP05)	<0.3	<0.3	<0.3	<0.3	<3	77	*	<0.1
HD03062 (SP01)	<0.3	<0.3	<0.3	<0.3	<3	500	*	<0.1
HD03064 (SP03)	<0.3	<0.3	<0.3	<0.3	<3	160	*	<0.2
Mean (average) value	<0.3	<0.3	<0.3	<0.3	<3	567	*	<0.1
Median (middle) value	<0.3	<0.3	<0.3	<0.3	<3	485	*	<0.1
Italy (Roma)								
Single composite	<0.3	<0.3	<0.3	<0.3	<3	250	*	<0.2

* - co-elution interference prevented quantitation of BDE-153 and TBBP-A

Short-chain chlorinated paraffins (SCCPs) - pooled and individual sample analyses (continued)

Country/region		**Concentration of short-chain chlorinated paraffins (ug/g dust, parts per million, ppm)**
Germany		
Single composite		48
Spain		
Madrid		41
Granada		22
Valencia		28
Asturia/Leon		17
Mean (average) value		*27*
Median (middle) value		*25*
France		
Lille		30
Toulouse		35
Lyon		95
Nantes		49
Paris		43
HD03058	(SP05)	52
HD03062	(SP01)	48
HD03064	(SP03)	41
Mean (average) value		*49*
Median (middle) value		*45*
Italy (Roma)		
Single composite		34
Slovakia		
Slovakia I	South West	24
Slovakia II	North East	66

Germany

HD03001	HD03002	HD03003	HD03004	HD03005
bis(1-Methylpropyl) butandioate Polyethylene Glycol type compound 1,1-oxybis-Octane 2-Dodecyloxyethanol Phenanthrene Chlorpyrifos 1,3-Dihydro-[2H]-benzimidazol-2-one Fluoranthrene Pyrene 11H-Benzo[a]fluorene Bis-(2-ethylhexyl) adipate Piperonyl butoxide N-Propylbenzamide Chrysene Benzo[a]anthracene Permethrin Unidentified PAH Tritriacontane Indeno[1,2,3-cd]pyrene	Phenanthrene N,N-Dimethyloctadec-1-amine Tributyl citrate 1,5-Diphenylpentan-1,5-dione 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Tri-[2-Butoxyethanol] phosphonate 2-Ethylhexyl dodecanoate 2-Ethylhexyl tetradecanoate Unidentified PAH N-Ethylbenzamide 4,4,6α,6β,8α,11,**11**,14β-Octamethyloctadecahydro-2H-picen-3-one α-Amyrin Freidelin	Methyl Methacrylate 1,1-Dichloro-2-ethenylcyclopropane Butan-2-one 1,1,3-Trichloro-2-methylprop-1-ene 1,1,2-Trichloroprop-1-ene Butyl Methacrylate Phthalic Anhydride 4-Hydroxybenzaldehyde Toluene-2,4-diisocyanate Tetrachloropyridine N,N,N',N'-Tetraacetylethyl enediamine (EDTA) Phenanthrene 4,4-Methylenebis-phenol Tributyl citrate Bis-(2-ethylhexyl) adipate Phenyl propanoate Hexadecyl 2-ethylhexanoate Various unidentified alkoxybenzene compounds	Alkyl amide Hexadecyl 2-ethylhexanoate Various unidentified alkoxybenzene compounds Octadecyl 2-ethylhexanoate	Pentan-2,4-dione 4,4-Dimethyl oxazolidine 1,1,2,2-Tetrachloroethane Phthalic Anhydride *p*-tert-Butylbenzoic acid N,N-Dimethyldodecan-1amine Azelaic acid 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Alkyl amide Various unidentified alkoxybenzene compounds 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Alkyl isocyanate Tri-[2-Butoxyethanol] phosphonate Diheptyl phthalate 2-Ethylhexyl dodecanoate Unidentified phthalate

Spain: Madrid

HD03006	HD03007	HD03008	HD03009	HD03010
Pyrazine derivative Phthalic Anhydride N,N-Dimethyldodecan-1amine 2-Dodecyloxyethanol 1-Isocyanato-octadecane N,N-Dimethylhexadec-1-amine Unidentified Amine 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate Permethrin Alkyl 2-ethylhexanoate Tris(2-Ethylhexyl)trimellitate	3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate Benzyl 2-ethylhexylphthalate Sesamin	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) 1,1'-[(Methylthio)ethenyl idene]bis-benzene Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate	2-(2-Butoxyethoxy)-ethanol Tributyl citrate Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate N-Propylbenzamide Tris(2-Ethylhexyl)trimellitate	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) 1,3-Dihydro-[2H]-benzimidazol-2-one N,N-Dimethyloctadec-1-amine Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Permethrin Unidentified Alkane

Spain: Granada

HD03011	HD03012	HD03013	HD03014	HD03015	HD03016
Ethylene glycol Toluene-2,4-diisocyanate N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyloctadec-1-amine Alkyl isocyanate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Unidentified branched alkane Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate 2-Ethylhexyl tetradecanoate Unidentified Amine Freidelin	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N-Propylbenzamide Unidentified alkane 2-Ethylhexyl tetradecanoate	N,N-Dimethyldodecan-1-amine N,N-Dimethyloctadec-1-amine Unidentifed Alkyl isocyanate Hexadecyl 2-ethylhexanoate Tetramethrin Unidentifed Amine Unidentified Alkene Octadecyl 2-ethylhexanoate Permethrin	Toluene-2,4-diisocyanate N,N-Dimethyldodecan-1-amine N,N-Dimethyltetradec-1-amine Hexadecyl 2-ethylhexanoate N,N'-Dimethyleicosamine N-Propylbenzamide	N,N-Dimethyldodecan-1-amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyloctadec-1-amine 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate Tetramethrin Octadecyl 2-ethylhexanoate	N,N-Dimethyldodecan-1-amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyltetradec-1-amine 3,7-Dihydro-3,7-dimethylpurine [1H]-2,6-dione N,N-Dimethylhexadec-1-amine N,N-Dimethyloctadec-1-amine N-Methyl-N-benzyldodec-1-amine 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate

Spain: Valencia

HD03017	HD03018	HD03019	HD03020	HD03021	HD03022
2,2-Dimethyl-1-(2-hydroxy-isopropyl) 2-methylpropanoate 3-Hydroxy-2,4,4-trimethyl 2-methylpropanoate N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) Tributyl citrate Hexadecyl 2-ethylhexanoate Unidentified alkane	N,N-Dimethyldodecan-1-amine N,N-Dimethyltetradec-1-amine N,N-Dimethylhexadec-1-amine Unidentified butylbenzyl compound 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate α-Amyrin	Triethylene glycol N,N-Dimethyldodecan-1-amine 5-Isocyanato-1-(isocyanatomethyl)-1,3,3-trimethylcyclohexane N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) 2-(Alkyloxy)-ethanol Pentaethylene glycol 3,7-Dihydro-3,7-dimethyl purine [1H]-2,6-dione 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate N-Propylbenzamide Octadecyl 2-ethylhexanoate Unidentified branched alkane Permethrin Piperine Unidentified phthalate	N,N-Dimethyloctadec-1-amine 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate Tetramethrin Octadecyl 2-ethylhexanoate	iso-Butyl Methacrylate N,N-Dimethyldecan-1-amine N,N-Dimethyldodecan-1-amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyltetradec-1-amine N,N-Dimethylhexadec-1-amine N,N-Dimethyloctadec-1-amine N-Methyl-N-benzyldodec-1-amine Unidentified branched alkane Permethrin	N,N-Dimethyldodecan-1-amine N,N-Dimethyltetradec-1-amine N,N-Dimethylhexadec-1-amine Unidentified butylbenzyl compound 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate α-Amyrin

Spain: Asturias/Leon

HD03023	HD03024	HD03025	HD03026	HD03027
N,N-Dimethyldodecan-1-amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethylhexadec-1-amine 1-Methyldodecylbenzene N,N-Dimethyloctadec-1-amine 2-Ethylhexyl tetradecanoate	3,7-Dihydro-3,7-dimethylpurine [1H]-2,6-dione 1-Methyldodecylbenzene Tributyl citrate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Unidentified branched alkane Tetramethrin	Benzoic acid Octanoic acid Phthalic Anhydride N,N-Dimethyldodecan-1amine 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate Octadecyl 2-ethylhexanoate	Polyethylene Glycol Benzophenone N,N-Dimethyloctadec-1-amine Bis-(2-ethylhexyl) adipate Unidentified branched alkane Piperonyl butoxide Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate Octadecyl 2-ethylhexanoate Permethrin	Metronidazole N,N-Dimethyloctadec-1-amine Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate Octadecyl 2-ethylhexanoate 2-Ethylhexyl tetradecanoate

Slovakia

Slovakia I	Slovakia II
Glycol-type compound	Unidentified branched amine
N,N-Dimethyldodecan-1-amine	Bis-(2-ethylhexyl) adipate
Benzenesulfonamide	Permethrin
N,N-Dimethyltetradec-1-amine	Unidentified alkyl compound
Unidentified quinoline-type compound	Unidentified alkane
Clorophene	
Unidentified branched amine	
Bis-(2-ethylhexyl) adipate	
Tri-[2-Butoxyethanol] phosphonate	
Unidentified branched amine	
Unidentified phthalate	
2-Ethylhexyl dodecanoate	

Italy: Roma

HD03036	HD03037	HD03038	HD03039	HD03040
N,N-Dimethyldodecan-1amine Diethyltoluamide N,N-Dimethyltetradec-1-amine Bisphenol A Unidentified benzoate Diethylene glycol dinonanoate Bis-(2-ethylhexyl) adipate Tri-[2-Butoxyethanol] phosphonate Unidentified phthalate Unidentified branched alkane Piperonyl butoxide Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate 2,4-bis-(1-Phenylethyl)-phenol Octadecyl 2-ethylhexanoate Permethrin Tris(2-Ethylhexyl)trimellitate	Toluene-2,4-diisocyanate N,N-Dimethyldodecan-1amine Unidentified benzoate Triphenyl phosphate Hexadecyl 2-ethylhexanoate Octadecyl 2-ethylhexanoate	Phthalic anhydride N,N-Dimethyldodecan-1amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyltetradec-1-amine 1-Methyltridecylbenzene Unidentified butylbenzyl compound p,p'-DDD Unidentified branched alkane 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Tri-[2-Butoxyethanol] phosphonate Tris(2-Ethylhexyl)trimellitate	Butan-2-one N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine 1-Methyldodecylbenzene Unidentified benzoate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Piperonyl Butoxide Hexadecyl 2-ethylhexanoate 2-Ethylhexyl dodecanoate	N,N-Dimethyldodecan-1amine 1-Methyldodecylbenzene Unidentified benzoate Bis-(2-ethylhexyl) adipate 2-Methyl-2-(4-methyl-3-pentenyl)-7-pentyl-2H-1-benzopyran-5-ol Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate 5a,6,7,8,9,9a-Hexahydro-6-methyldibenzofuran-1,6-diol 2-(3,7-Dimethylocta-2,6-dienyl)-5-pentylbenzen-1,3-diol

France: Lille

HD03043	**HD03045**	**HD03047**	**HD03049**	**HD03050**
Piperonyl butoxide Triphenyl Phosphate Permethrin	Totarol Piperonyl butoxide Unidentified substituted aromatic Permethrin	Phthalic Anhydride Pyrene 2-Ethylhexyl dodecanoate 2-Ethylhexyl tetradecanoate Permethrin Unidentified alkane Tris(2-Ethylhexyl) trimellitate	*p*-tert-Butylbenzoic acid N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine N,N-Dimethylhexadec-1-amine N,N-Dimethyloctadec-1-amine Unidentified butylbenzyl compound 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate 2-Ethylhexyl dodecanoate 2-Ethylhexyl tetradecanoate	Irgacure 184 (Polymerising agent for acrylates) Tri-[2-Butoxyethanol] phosphonate Piperonyl butoxide N,N'-Dimethyleicosamine N-Propylbenzamide Unidentified phthalate Permethrin Unidentified alkene Unidentified alkane Tris(2-Ethylhexyl)trimellitate

France: Toulouse

HD03052	**HD03053**	**HD03054**	**HD03055**	**HD03061**
N,N-Dimethyldodecan-1-amine 3,7-Dihydro-3,7-dimethylpurine [1H]-2,6-dione Unidentified alkene Malathion Methyl di-n-decylamine Totarol Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate Octabenzone Sesamin Tris(2-Ethylhexyl)trimellitate	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) Tri-[2-Butoxyethanol] phosphonate Piperine	Phthalic Anhydride N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) Tri-[2-Butoxyethanol] phosphonate 2-Ethylhexyl dodecanoate	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyloctadec-1-amine Unidentified 2-ethylhexyl ester	N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine N,N-Dimethyloctadec-1-amine 1,1'-oxybis Naphthalene 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Tri-[2-Butoxyethanol] phosphonate Piperonyl butoxide Unidentified phthalate Unidentified PAH Permethrin

France: Lyon

HD03070	HD03071	HD03075	HD03076	HD03077
N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) Pentachlorophenol Tributyl citrate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Hexadecyl 2-ethylhexanoate 2-(Methoxymethyl)-2-phenyl-1,3-dioxolane Unidentified amine Benzyl 2-ethylhexyl phthalate Benzyl n-octyl phthalate 2-Ethylhexyl tetradecanoate	N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine Benzylphthalate 5a,6,7,8,9,9a-Hexahydro-6-methyldibenzofuran-1,6-diol	Phthalic Anhydride N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine Fluoranthrene Pyrene 1-Methylpyrene Piperonyl butoxide Benzo[b]naphtho[2,1-d] thiophene Unidentified PAH Unidentified phthalate Unidentified branched alkane Benz(c)acridine Chrysene Bis-(2-ethylhexyl)-phthalate Methylchrysene Cholesta-3,5-diene Indeno[1,2,3-cd]pyrene Benzo[ghi]perylene Dibenzopyrene	N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyloctadec-1-amine Pyrene Tri-[2-Butoxyethanol] phosphonate Hexadecyl 2-ethylhexanoate Unidentified amine Unidentified phthalate Chrysene Octadecyl 2-ethylhexanoate	Glycerol N,N,N',N'-Tetraacetyl ethylenediamine (EDTA)

France: Nantes

HD03081	HD03082	HD03084	HD03087	HD03088
N,N,N',N'-Tetraacetyl ethylenediamine (EDTA)	Toluene-2,4-diisocyanate 1,6-Di-isocyanatohexane N,N-Dimethyldodecan-1amine Benzenesulfonic acid ester N,N-Dimethyltetradec-1-amine 1-Pentylheptylbenzene 1-Butyloctylbenzene Tetradecanoic Acid 1-Propylnonylbenzene 1-Ethyldecylbenzene 1-Methylundecylbenzene 1-Pentyloctylbenzene 1-Butylnonylbenzene 1-Propyldecylbenzene 1-Methyldodecylbenzene 1-Hexyloctylbenzene 1-Pentylnonylbenzene 1-Butyldecylbenzene 1-Ethyldodecylbenzene 1-Methyltridecylbenzene Unidentified branched alkane Unidentified phthalate Unidentified PAH	Phthalic Anhydride N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine Unidentified butylbenzyl compound Unidentified 2-ethylhexyl ester Tris(2-Ethylhexyl)trimellitate	N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine Tetradecanoic acid	Methyl di-n-decylamine

France: Paris

HD03091	**HD03093**	**HD03095**	**HD03097**	**HD03098**
N,N-Dimethyldodecan-1amine Piperonyl butoxide	Tri-[2-Butoxyethanol] phosphonate Piperonyl butoxide Unidentified substituted aromatic 5a,6,7,8,9,9a-Hexahydro-6-methyldibenzofuran-1,6-diol Unidentified phthalate Permethrin 4,4,6a,6b,8a,11,**12**,14b-Octamethyloctadecahydro-2H-picen-3-one 4,4,6a,6b,8a,11,**11**,14b-Octamethyloctadecahydro-2H-picen-3-one Methyl 3-oxo-Urs-12-en-24-oate	Methyl Methacrylate Phthalic Anhydride N,N-Dimethyldodecan-1amine N,N-Dimethyltetradec-1-amine N,N-Dimethylhexadec-1-amine Unidentified butylbenzyl compound 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate	Unidentified 2-ethylhexyl ester	N,N-Dimethyldodecan-1amine N,N,N',N'-Tetraacetyl ethylenediamine (EDTA) N,N-Dimethyltetradec-1-amine Acetaminophen N,N-Dimethylhexadec-1-amine 1,3-Dihydro-[2H]-benzimidazol-2-one 2-Ethylhexyl decanoate Tri-[2-Butoxyethanol] phosphonate 2,4-bis-(1-Phenylethyl)-phenol

France: individual samples

HD03058	HD03062	HD03063	HD03064	HD03065	HD03066
N,N-Dimethyldodecan-1-amine Benzyl Salicylate Tributyl citrate Piperonyl butoxide Triphenylphosphate Tris(2-Ethylhexyl) trimellitate	Phthalic Anhydride 2-(2-Butoxyethoxy)-ethyl acetate N,N-Dimethyldodecan-1-amine 2,2,4-Trimethyl-3-carboxyisopropyl..pentanoic acid N,N-Dimethyltetradec-1-amine Methyl di-n-decylamine Unidentified butylbenzyl compound Tributyl citrate 3-(4-Methoxyphenyl)-2-ethylhexylpropenoate Tri-[2-Butoxyethanol] phosphonate Dibenzyl phthalate	Tri-[2-Butoxyethanol] phosphonate Unidentified branched alkane Freidelin	N,N-Dimethyloctadec-1-amine Unidentified fatty acid Tri-[2-Butoxyethanol] phosphonate Piperonyl butoxide Unidentified 2-ethylhexyl ester N-Acetyl-N-benzyl nonylamine	Butan-2-one m-Xylene 1-Acetylpiperidine 2-Acetyl-5-methylfuran Glycocyanidine 5-Ethenyldihydro-5-methyl-2[3H]-Furanone Glycocyanidine 1-Methylheptan-2-one 2,2,3,4-Tetramethylhex-5-en-3-ol 2,4-Dimethylphenyl acetate Phthalic Anhydride 1,1'-Methylene-bis-pyrrolidine Hexanoic acid anhydride Benzophenone	Unidentified substituted aromatic 1,2-bis(1-Methylethyl)-benzene 1,4-bis(1-Methylethyl)-benzene 1,1-Dimethylbutylbenzene Phthalic Anhydride a,a,a',a'-Tetramethyl benzene-1,4-dimethanol Octadecyl 2-ethylhexanoate

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

Alkylphenols and their derivatives (APs, APEs)

Alkyphenols (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 industrial and institutional cleaning products (detergents), although uses 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 though 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, 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.

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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

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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/out91_en.html

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OSPAR (2001) Nonylphenol/nonylphenolethoxylates, OSPAR Priority Subtances 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 7 500 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.

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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), 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 toxicity 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 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). 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 Antifoulants (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.

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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 though 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.

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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 Europe, 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 Europe, 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 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

the Environment” (“very toxic to aquatic

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

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Annex 3: details of analytical methodologies employ

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).

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 ^{13}C 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 isooctane. 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^{-} (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. 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 um (0.065 mm) prior to analysis. Organotin compounds were extracted using a mixture of methanol and hexane (with NaBEt4) 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 ng organic tin/g dry weight of sample (ppb) in each case.

Greenpeace European Unit
Chaussée de Haecht 159
1030 Brussels/Belgium
Tel.: +32 2 274 1900
Fax: +32 2 274 1910
www.eu.greenpeace.org

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Greenpeace European Unit
Chaussée de Haecht/
Haachtsesteenweg 159
B -1030 Brussels
t +32 2 274 02 00
f +32 2 201 19 50
e info@be.greenpeace.org

Greenpeace Central & Eastern Europe
siebenbrunnengasse 44
A - 1050 Vienna
t +43 1 545 4580
f +43 1 545 4580-98
e office@greenpeace.at

Greenpeace France
22 rue des rasselins
75020 Paris
t +33 1 44 64 02 02
f +33 1 44 64 02 00
e contact@diala.greenpeace.org

Greenpeace Germany
Grosse Elbstrasse 39
D-22767 Hamburg
t +49 40 306 180
f +49 40 306 18100
e mail@greenpeace.de

Greenpeace Italy
Viale Manlio Gelsomini 28
00153 Rome
t +39 06 572 9991
f +39 06 578 3531
e staff@greenpeace.it

Greenpeace Spain
San Bernardo 107
28015 Madrid
t +34 91 444 14 00
f +34 91 447 15 98
e greenpeace.spain@diala.greenpeace.org

Greenpeace UK
Cannonbury Villas
London N1 2PN
t +44 207 865 8100
f +44 207 865 8200
e info@uk.greenpeace.org