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Research Articles

Concentrations of Phthalate Esters and Identification of Other Additives in PVC Children's Toys *

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Abstract. This study was intended to provide data on the composition of soft PVC toys, addressing the widest practicable range of chemical additives and including non-phthalate additives. The study also included toys from as many countries as possible, since for many, no data were available. A total of 72 toys were purchased in 17 countries. The majority (64) were PVC or had PVC sections. In almost all the soft PVC toys analysed, phthalates comprised a sizeable proportion (most frequently 10-40%) of the total weight of the toy.

The predominant phthalates detected were diisononyl phthalate (DINP) and di(2-ethylhexyl) phthalate (DEHP). Other phthalates identified in high concentrations in some toys include isomeric mixes of diisooctyl phthalate (DIOP) and diisodecyl phthalate (DIDP). The estrogenic chemical nonylphenol was isolated from 13 toys, while 2 toys were found to contain the fungicide Fungitrol 11 (Folpet). 78% of PVC toys contained one or more extractable organic compounds in addition to those reported above.

Keywords: DEHP; di(2-ethylhexyl) phthalate (DEHP); DIDP; diisodecyl phthalate (DIDP); diisononyl phthalate (DINP); diisooctyl phthalate (DIOP); DINP; DIOP; Folpet; fungicides; Fungitrol 11; Fungitrol; non-phthalate additives; nonylphenol; phthalates; PVC toys

1 Introduction

The intentions of this study were to identify and, where possible, quantify chemical additives and/or contaminants contained in a range of plastic children's toys purchased in a number of countries, with a particular focus on toys manufactured from plasticised polyvinyl chloride (PVC). The study was not intended to generate dose or risk data for any of the chemicals identified. Rather it was designed to generate empirical data on chemical composition influenced as little as possible by the choice of extraction method or other subjective elements, such as assumptions about the amount of time a child might spend interacting with a toy. Such considerations are routinely incorporated into leaching tests and can contribute to considerable variation between results obtained from different test regimes. It was our aim to identify components that might be further studied by other researchers or regulatory bodies, but not to conduct those studies ourselves.

Whilst manufacturers may hold information on the composition of their products, few data have been presented in the scientific literature. This study is broader both in geographical and chemical coverage than any other we have found in the scientific literature to date and for that reason would extend the knowledge base of those whose primary information source is scientifically based rather than commercially based.

PVC (polyvinyl chloride, or vinyl) is a widely used material, including extensive use in toys and other children's products. PVC is never formulated without a range of additives to enhance its properties (Ehrig, 1992). In order to soften PVC for the manufacture of toys such as chewy teethers, soft figures and inflatable toys, softeners or plasticisers are added. A number of chemicals are used, although by far the most common are the phthalates (phthalic acid esters or benzenedicarboxylic acid esters). They represent 69% of plasticiser use in the USA, 92% in Western Europe and 81% in Japan. Overall, they represent 82% of the 2.5 million tonnes (5400 million pounds) of plasticiser use in these three regions (Bizzari et al., 1996). They are moderately persistent and, as a consequence of their wide use, are the most abundant man-made chemicals in the environment (Jobling et al., 1995).

Phthalates in soft PVC products are not tightly bound to the plastic, but are present as mobile components of the plastic matrix. Loss of phthalates by volatilisation over time from soft PVC has long been recognised (Cadogan et al., 1993, UK Department of the Environment, 1991). Phthalates can also be released from soft PVC by surface contact, especially where mechanical pressure is applied (e.g. during chewing of a PVC teether). Releases of phthalates during manufacture, use and disposal of PVC products, in addition to their use as additives in inks, lubricants, perfumes and other open-ended

* Some of these data have been released previously by Greenpeace and made available on the internet. This report, however, which is intended for a technically expert audience, has been peer reviewed and contains further analytical data that have not been published before.

applications, has lead to their ubiquitous distribution and abundance in the global environment.

Exposure to phthalates is, therefore, twofold. Humans may be exposed to background levels of contamination in air, food and water, but may also be exposed through direct contact with soft PVC products, particularly those intended as high contact products, such as children's toys and teethers. Although less persistent in the body and environment than many other synthetic organic compounds, the continued release of large quantities of these chemicals to the open environment ensures that concentrations and, therefore, exposures remain substantial. Moreover, recent research suggests that phthalates may persist in human body tissues for longer periods than previously assumed (DIRVEN et al., 1993).

Given the known and potential toxicity of the phthalates, individually and as a chemical class, and the ability of the phthalates to leach out of the PVC matrix through contact or volatilisation, the continued use of soft PVC as a material for children's toys raises serious concerns. This is especially true since children will receive their greatest exposure to many of these products, particularly teethers, at an early and sensitive stage of their development. It has been known since the early eighties that DEHP was present in high concentrations in children's products and could leach out under simulated dermal and oral contact (HANSON, 1983).

However, leaching tests do not produce definitive results. Research carried out for the US Consumer Product Safety Commission (CPSC) found no consistent relationship between either the concentration of phthalate in the product or the duration of the experiments and the amount of DEHP that leached out (HANSON, 1983). Nevertheless, it was apparent that DEHP would continue to leach out of a product over an extended period. Moreover, if pacifiers ("dummies") were squeezed during the leaching experiments, to imitate the action of a child chewing, the rate of release of DEHP was increased.

More recently, both the Dutch and Danish governments initiated research into the leaching of phthalates from PVC teethers. These studies showed that, for some products, leaching rates were substantial and, according to McKenzie (1997), might lead to doses of phthalates which would exceed tolerable daily intakes agreed by the European Commission Scientific Committee for Food (SCF) (JANSSEN et al., 1998; VIKELSOE et al., 1997; SCF, 1995). Even these more sophisticated tests, however, were based on a number of critical assumptions. The fundamental criteria of appropriate test solution, sample manipulation via squeezing or sonication, and estimates of contact time undoubtedly have a substantial influence on the leaching rates recorded and subsequent interpretation of their significance. Other factors will also affect estimates and it should be recognised that leaching tests will retain an element of uncertainty.

In the light of these concerns, data on the composition of toys may be regarded as less subjective. While relationships between content and leaching rates will remain complex and variable, an overview of phthalate use in children's toys would give an indication of the scale of the problem. However, few data currently exist.

This study does not provide (and was not intended to provide) data for calculating leaching from the toys studied or potential doses which children might ingest through chewing or sucking them. Nevertheless, the finding that PVC toys routinely contain high percentage concentrations of phthalate esters is of concern. In addition, it is clear that DEHP has been replaced by the isomeric mixture DINP on an almost global basis. This, and the identification of a number of compounds which are not widely known to be incorporated into toys, indicates the complexity of the toxicological questions posed by these products.

2 Materials and Methods

A total of 72 toys were purchased off the shelf from retail outlets in 17 countries and forwarded to the Greenpeace Research Laboratories for analysis. The distribution of toys received according to country of purchase is summarised in **Fig. 1** and a description of each is given in **Table 1** (*see* Appendix, *p.* 8). Distribution according to country of manufacture is shown in **Fig. 2**, illustrating the key importance of China as a predominant source of the plasticised PVC toys retailed in other countries.

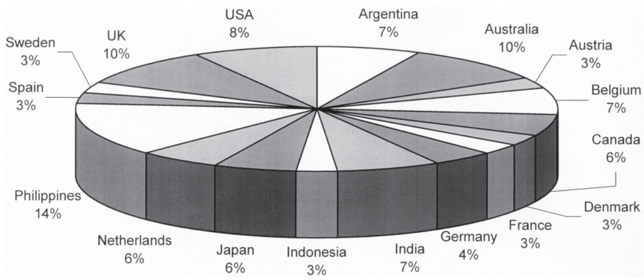

Fig. 1: Distribution of the toys analysed according to country of purchase

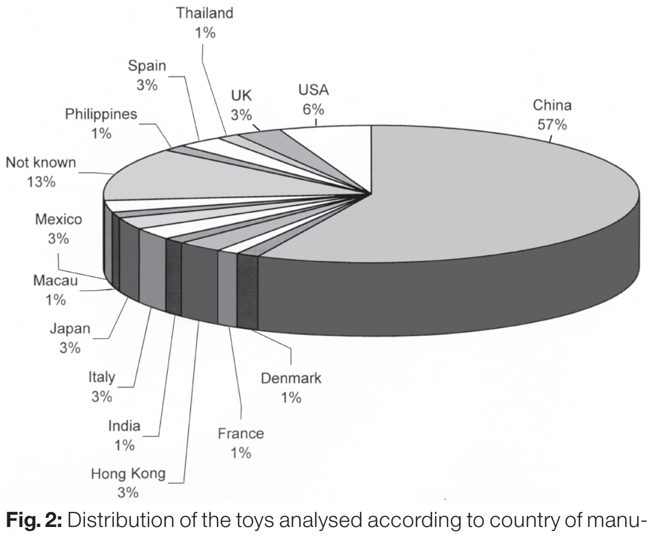

Fig. 2: Distribution of the toys analysed according to country of manufacture

On receipt at the laboratory, samples were taken of each of the toys (samples of each section where the toys consisted of several discrete materials or textures) for determination of chlorine content as an indicator of PVC (using X-ray microanalysis; results not shown). All PVC toys and a selection of non-PVC toys were subsequently analysed for phthalate plasticisers.

To ensure efficient extraction of the plasticisers, each toy, or section of toy, was frozen in liquid nitrogen for several seconds and then grated to produce a finely ground material (with a typical particle size of less than 2 mm). Discussion with other laboratories indicated that method had proved satisfactory in the past. Grating was not applicable to the baby books which consisted of a thin PVC covering over foam pages. The PVC component of these books was instead cut with scissors into pieces of less than 2 mm^2.

Approximately 1 g of each grated section (weighed accurately) was transferred to a glass bottle and shaken with a known volume of hexane, followed by sonication for 30 min to improve contact between solvent and sample. The solvent fraction was decanted off, fresh solvent added and the process repeated. The two solvent fractions were combined, evaporated or diluted to a known volume and analysed using a Hewlett Packard GC/MS (gas chromatograph/mass spectrometer), operated in scan mode. Phthalates were identified according to their retention times and mass spectra using standards obtained both from laboratory suppliers, and commercial producers/suppliers. Concentrations of individual phthalates in the toys or toy sections were determined using calibrations prepared using these standards.

Extraction and analytical procedures were carefully quality controlled. QC procedures and acceptance criteria were based on USEPA method 606 for the analysis of phthalates in water samples (USEPA, 1984). Extraction efficiency was tested by spiking blank matrix and by standard addition to phthalate-containing samples. QC data are given in **Table 2**. Further, each batch of samples was analysed in concert with reagent blanks, matrix blanks and spiked samples as appropriate. No phthalates or other analytes were detected either in reagent blanks or in matrix blanks, which were prepared using a phthalate-free plastic matrix. Results were not corrected for recovery.

During the initial phase of the survey the following phthalates were quantified: diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), diisobutyl phthalate (DIBP), butyl benzyl phthalate (BBP), di(ethylhexyl) phthalate (DEHP) and diisononyl phthalate (DINP). Other phthalates and alkyl phenols were identifiable through mass spectral matching and comparison of retention times with target analytes. However, they were not fully quantifiable because of the non-availability of analytical standards. They are therefore reported in **Table 3** (*see* Appendix, *p.* 8) as being detected but not quantified. For some samples, this precludes calculation of a figure for total phthalate concentration. Acquisition of further phthalate standards, development of methods and further quality control work allowed an extended range of compounds to be quantified during later analyses, although some phthalates remained unidentified and were not reported. Octyl and nonyl phenols were also quantified for later samples. Additional compounds extracted from the toys were identified as far as possible by mass spectral matching against the Wiley 138 spectral database, with each computer match being checked by experienced personnel.

Table 2: Recovery of standard analytes from blank matrix and spiked samples

Compound	% Recovery from blank matrix (% standard deviation)	% Recovery from spiked sample
DEP	82.4 (1.9)	81.6
DnBP	84.3 (2.3)	74
DIBP	83.0 (2.4)	64.2
BBP	84.0 (3.1)	77.4
DnOP	88.4 (4.0)	88.2
DEHP	84.4 (3.5)	87.9
DIOP	99.5 (1.0)	155
DINP	79.6 (0.9)	94.0
DIDP	95.5 (4.5)	133
nonyl phenol	80.9 (0.7)	55.7

Sample 6044 presented difficulties for other reasons. The phthalates isolated corresponded with DINP based on retention time. However, the peak profile of the analyte within these retention time boundaries differed markedly from that of the standard, indicating a variation of composition sufficient to add a degree of uncertainty to the quantitation.

3 Results

Almost all toys, or subsections of toys, identified as PVC contained phthalate esters, with the majority in the range of 10-40% by weight (→ *Table 3*, *see* Appendix, *p.* 8). Notable exceptions include samples 6068, 6078, 7008, 7009 and 7013b&c, which XRM analysis identified as PVC but which did not contain phthalates. Samples 6068, 7013b and 7013c were hard, and so absence of plasticisers is understandable. Sample 7001, which contained only 0.004% phthalate, was also hard PVC. Of the others, the identity of the plastic was uncertain in 6078, 7008 and 7009; XRM identified traces of chlorine at close to detection limits in these samples, yet their appearance and texture were more consistent with polyolefins (possibly polyethylene or polypropylene). Only one non-PVC toy contained phthalates (sample 6022). The trace concentrations of DEHP probably result from surface contamination from the packaging, a transparent, plasticised PVC pouch, in which it was sold.

DINP was the phthalate most frequently found in toy samples and tended to be present in the highest concentrations. DEHP was the next most frequently found. Where present, isomeric phthalates generally comprised 10-40% of the weight of the toy, though the maximum recorded was over 50% DINP in a teether. This is in line with other studies on the phthalate content of children's products (Hanson, 1985; 1983). Not infrequently, toys would contain high concentrations of DINP and traces of DEHP. This is most probably due to DEHP being present as an impurity in the DINP (Janssen et al., 1998).

The isomeric phthalates diisooctyl phthalate (DIOP) and diisodecyl phthalate (DIDP) were found at concentrations

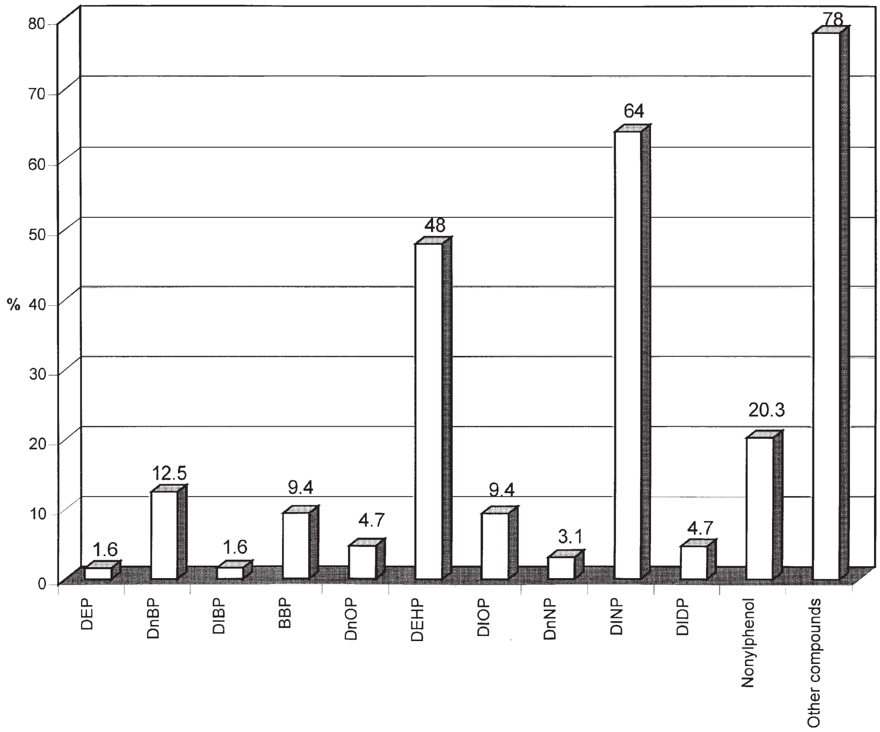

Fig. 3: Frequency distribution for phthalates and other organic compounds in PVC toys. % indicates proportion of all PVC toys which each compound was found

Table 4: Compounds identified through computer-based spectral matching in one or more toy samples

Compounds identified to >90% match quality	
1,3-Benzodioxole-5-carboxaldehyde	Dieldrin
1,3-Isobenzofurandione	Docosane
1,3-Propanedione, 1,3-diphenyl-	Eicosane
1-Dodecanol	Elaidinic acid, isopropy ester
1-Hexadecanol	Fungitrol 11 (Folpet)
1H-Indole, 2-phenyl-	Heptacosane
1H-Isoindole-1,3(2H)-dione	Heptadecane
1-Octadecanol	Heptadecene-(8)-carbonic acid-(1)
1-Octadecene	Hexadecane
1-Pentadecanol	Hexadecanoic acid
1-Phenanthrenecarboxylic acid	Hexanedioc acid, dioctyl ester
1-Tetradecanol	Hexatriacontane
2,2'-Paracyclophan	Methanone, diphenyl-
2-Cyclohexen-1-one, 3,5,5-trimethyl-	Naphthalene, 1,2,3,4-tetrahydro-1-phenyl-
2-Hydroxybenzoate, methyl ester	Naphthalene, 1,2-dihydro-4-phenyl-
3,3-(D2)Menth-1-ene	Nonacosane
3-Phenylindole	Nonadecane
9-Octadecanoic acid (Z)	Nonylphenol, isomeric
Benzaldehyde, 3-ethoxy-4-hydroxy-	Octadecane
Benzaldehyde, 4-hydroxy-3-methoxy-	Octadecanoic acid
Benzaldehyde, 4-methoxy-	Octadecanoic acid, 2-methylpropyl ester
Benzene, 1,1'-(1,2-cyclobutanediyl)bis-, trans-	Pentacosane
Benzene, 1,1'-(1,3-propanediyl)bis-	Pentadecane
Benzoic acid	Pentyl 2-ethylhexanoate
Benzoic acid, 4-hydroxy-, methyl ester	Phenol, 2,6-bis(1,1-dimethylethyl)-4-methyl-
Benzyl benzoate	Phenol, 4,4'-(1-methylethylidine)bis-
Butyl benzoate	Phosphonic acid, dioctadecyl ester
cis 1,1-(1,2-Cyclobutanediyl)bisbenzene	Phosphoric acid, triphenyl ester
Cyclodecane	Tetracosane
Cyclododecane	Tetradecane
Cyclooctane, 1,2-dimethyl-	Triacontane
Cyclotetradecane	Tricosane
Decanedioic acid, dibutyl ester	Tridecane
DEHA (Di-(2-ethylhexyl) ester of adipic acid)	

in excess of 10 percent in a few samples and were present, but not quantified, in several others. The following phthalate esters were found at comparatively low concentrations in the toys in this study: DEP (0.16%) DnBP (0.002-0.18%), DIBP (0.45%), BBP (0.001-0.02%), di-n-octyl phthalate (DnOP), di-n-nonyl phthalate (DnNP, not quantified). The distribution of phthalates and other compounds in the toys is shown in **Fig. 3**. Although the phthalate data are already presented on a sample-specific basis in **Table 3** (*see* Appendix, *p.* ♦), the information on the other compounds is presented in summary form only (→ *Table 4*), i.e. as a compiled list of those identified, but without frequency data. **Fig. 3** provides an indication of the relative frequency of the appearance of the full range of analytes detected in PVC toys, including the proportion in which one or more of the organic compounds listed in **Table 4** were also identified.

The following phthalates, for which standards were obtained, were not detected in any of the toys: bis(2-n-butoxyethyl) phthalate, bis(2-ethoxyethyl) phthalate, bis(2-methoxyethyl) phthalate, bis(4-methyl-2-pentyl) phthalate, diamyl phthalate, dicyclohexyl phthalate, di-n-hexylphthalate, dimethyl phthalate and hexyl 2-ethylhexyl phthalate.

Nonylphenol was detected in 13 toys (in both sections analysed for sample 7006), at concentrations, where quantified, of 0.009-0.36%. All the toys containing nonylphenol were PVC. Octylphenol was not detected in any of the toys. A number of other compounds were isolated from some of the toys. Those that could be identified to greater than 90% certainty through computer-based spectral matching are listed in **Table 4**. These included the fungicide Fungitrol 11 (found in samples 6069 and 6072), the antioxidant butylated hydroxytoluene (BHT, found in seven samples) and a range of long-chain paraffins (C10-C30, identified in 15 samples). More tentative identifications were obtained for esters of bis(2-ethylhexyl) adipate (DEHA, five samples), esters of phosphonic and propanoic acid, alkyl benzenes and derivatives of phenol and butyric acid. No further attempts were made to confirm identities or quantify these additional compounds.

4 Discussion

This study was targeted at plasticised PVC toys and so does not cover the entire range of toys on sale. Nevertheless, some trends were evident. Although the majority of toys received were made wholly or partly of PVC, the plastic was only infrequently identified on the packaging. All inflatable toys, bath toys, and baby books received were composed of phthalate-plasticised PVC. Squeeze toys, which appeared in a number of categories, were also consistently phthalate-plasticised PVC. Other categories were less consistent. One other notable fact is that the majority of the toys were manufactured in China. A breakdown of the country of manufacture of toys is given in **Fig. 2**.

Contrary to initial expectations, DEHP was not the most commonly used phthalate in the toys analysed. This contrasts with a report prepared for the US Consumer Product Safety Commission in 1983 (Hanson, 1983), in which DEHP was the only phthalate identified in eleven of fourteen children's products analysed. DEHP was also the focus of the study by Meek and Chan in 1994 (Meek & Chan, 1994). In the current study, DINP (diisononyl phthalate), a mixture of 30 or more isomers of dinonyl phthalate, was the most common form identified, although other, long-chain, isomeric mixtures were also isolated. This may reflect a market shift away from DEHP towards the use of the much more poorly characterised isomeric phthalates. This is especially true in China, the origin of the majority of the toys analysed.

Such a market shift to DINP may be a response to serious concerns relating to the suspected reproductive toxicity and other hazards associated with DEHP. Data currently available for DINP suggest that this isomeric phthalate may also impact similar target organs in chronic studies (especially the liver, kidney and reproductive tracts) as DEHP. DINP, for example, has been shown to exhibit a range of toxic effects in laboratory animals. There is, therefore, little reason to assume that isomeric phthalates present less of a potential hazard.

Much of the existing data on the toxicity of DINP has been collated by the Swedish National Chemicals Inspectorate (KEMI, 1994) and by the European Commission under the IUCLID initiative (European Chemical Bureau, 1996). Additional data is available from the US EPA (Myers, 1991; USEPA, 1991 & 1992) and from the Aristech Chemical Safety Information Database (ARISTECH, 1995). DEHP has been even more extensively studied, and recent papers continue to extend understanding of both effects and mechanisms of action. For example, Arcadi et al. (1998) report that perinatal exposure to DEHP significantly increased the time taken for female rats to perform a learned task. The significance of effects mediated through peroxisome proliferation has been an area of regulatory interest, since this mechanism, of particular importance in some animal models, is not thought to operate in humans. However Ward et al. (1998) has established that mice lacking the necessary receptor still exhibit renal and testicular toxicity, indicating that DEHP acts through more than one mechanism.

Variation between batches and the contamination of commercial and industrial mixes with other phthalates, or other compounds, is frequently noted (see e.g. Harris et al., 1997). Indeed, differences in DINP profiles from different samples were seen in this study. It is recognised that technical preparations of DINP may contain smaller quantities of DEHP as a contaminant of the manufacturing process (Janssen et al., 1998). This could account for the DEHP reported in some of the toys in this study in which DINP was the dominant phthalate. Although there is currently no regulatory necessity to further characterise DINP or other isomeric phthalates, it is of interest in purely scientific terms. Increasingly, the environmental toxicology and chemistry of materials supplied as technical mixtures are based on the analysis and testing of known components of particular interest. PCBs are perhaps the best known example. At present the analytical chemist would benefit from the availability of a wider range of standards both of technical mixes of phthalates and individual compounds. Provision of characterised mixes and components would further facilitate evaluation of the toxicity, pharmacokinetics and environmental behaviour of phthalate mixes as a whole. In this context, it would also be advantageous to have a recognised analytical procedure for the determination of constituents of plastics.

The data from this investigation do not allow estimation of the dose to which a child playing with any toy will be exposed. As discussed above, it is well recognised that DINP and other phthalates leach from PVC to the air, in to liquid solvents and on to the skin. The dose received by an individual child playing with a certain toy will depend on many factors, including phthalate composition and concentration, toy design and purpose, exposure time, method of play and degree of mouthing and chewing activity. A number of different methods are under development for the estimation of leaching rate and there may be limited comparability between methods (see e.g. Marin et al., 1998). That different leaching tests may yield significantly different results has been recognised for some time (Pindar et al., 1993). Any estimates of dose are, therefore, likely to remain highly subjective.

Some experts in this field are of the opinion that leaching tests are useful and could form a reliable basis for regulation. However, the validity of these tests has not been universally accepted. Researchers at this laboratory remain highly sceptical as to their utility and have made representations to this effect to the EC and in other fora. More importantly, however, it was never the purpose of this study to provide information of biological relevance; it is a chemical study of the components of these products and makes no attempt to quantify either dose or effect. Therefore, further discussion of the human volunteer study or the *in vitro* leaching simulations that have been developed were not thought relevant to this particular study.

Whilst attention naturally focuses on those components which are present in, and leachable from, toys in the largest quantities, some of the minor components are also worth noting.

Several phthalates were found in concentrations too low to have plasticising function. These were DEP (found in sample 6038 at a concentration of 0.16% by weight); DnBP (found in seven samples with a concentration range of 0.002-0.18%); DIBP (found in one sample at a concentration of 0.45%); BBP (found in six samples with a concentration range of 0.001-0.02%); DnOP (found in three samples, but only quantified in one, where it was present at a concentration of 0.012%) and DnNP (present but unquantified in two toys).

Three possible explanations for the presence of these phthalates are proposed: 1) as a constituent or contaminant of another phthalate; 2) constituent of an ink or paint used on the toy; 3) through use of the phthalate in the facility that manufactured the toy, either as a processing aid or in another product.

Both DEP and DBP were found at low concentrations in a study of phthalates in packaging (Balafas et al., 1999). They suggest inks or process contamination as the most likely sources of these compounds in their samples.

Several of the samples present profiles of phthalates which are suggestive of use of an impure primary plasticiser. DIBP is only present in one sample (6057), where both it and DnBP are minor constituents and other, unidentified phthalates are present in much higher concentrations. DnOP is present in sample 6029 at a concentration of 0.012% where the chemically similar DEHP was present at almost 20%. In two other samples, DnOP and DnNP were the only two identified components of a complex mix for which no suitable standard was obtainable.

Non-phthalate components of significance included nonylphenol, identified in thirteeen samples (6044, 6047, 6065, 6068, 6069, 6073, 6085, 6086, 7001, 7002, 7005, 7006 & 7013). Alkyl phenols are used as antioxidants in PVC and polystyrene (Olsson et al. 1998). Nonylphenol (NP) is a persistent chemical that has been found to mimic the hormone estrogen and impair sperm production in fish (Jobling et al., 1996). The toxicity of nonylphenol and related compounds led to a recommendation by the Paris Commission to phase out the use of NPEs in household and industrial detergent (PARCOM, 1992). Sweden is expected to call for a binding decision to phase out all uses of NPEs which result in contamination of the environment (ENDS, 1996), citing in particular the known estrogenicity of nonylphenol. With this in mind, applications that could lead to ingestion by children are of concern.

As noted above, a number of other compounds were isolated from some of the toys, many of which were identified only tentatively or not at all. These include phthalate esters for which standards were not obtainable and members of other chemical groups. Those compounds that were identified to a certainty of better than 90% by computer matching are listed in **Table 4** (*p.* ♦). The source of these compounds in toys is currently not known. As with some of the phthalates discussed above, some may have been added intentionally as colourants, processing aids or for other purposes. Still others may have been utilised in the manufacturing facility for purposes unrelated to the toys and have been acquired accidentally. Of these compounds, the fungicide Fungitrol 11 (samples 6069 and 6072) deserves further consideration. Fungitrol 11, also sold as Folpet, is a phthalimide fungicide. It is recognised as a skin irritant (Royal Society of Chemistry 1987) and is highly toxic to fish and aquatic invertebrates (Royal Society of Chemistry, 1987; USEPA, 1987b). It can also cause tumours in mice (USEPA, 1987a & b), but not rats (USEPA, 1987b). Teratogenicity has been reported for rats and rabbits (USEPA, 1986 & 1987b). Folpet is no longer sold in the United States (Thomson, 1990).

PVC resin itself is not usually vulnerable to microbiological attack. However, the phthalate plasticisers may be more susceptible. As the fungicide was found in toys with over 40% by weight phthalate content, it could have been added with the intention of preventing microbiological attack on the plasticisers or other additives.

As the identities of the majority of other compounds isolated could not be confirmed, their toxicological significance cannot be determined. Nevertheless, it is probable that a proportion of these compounds will also leach from the toys as a result of chewing or other contact and could, therefore, add to the overall burden of chemicals to which children playing with PVC toys will be exposed.

5 Conclusion

The current survey is the widest conducted to date on the presence of phthalate esters and other components in children's toys. Despite the fact that children have been known to be exposed to phthalates from PVC toys for some years, and the continuing concern surrounding the toxicity of phthalate esters, PVC toys still routinely contain a significant proportion of phthalates. The apparent market shift from DEHP to DINP and other high molecular weight isomeric phthalates is not reassuring. Recent research, particularly that carried out by the Danish and Dutch governments, has heightened concerns about the exposure of children to phthalates leaching from toys.

The results presented here indicate that the majority of soft PVC toys contain substantial proportions of phthalate esters that have the potential to leach out during use and therefore present an exposure risk to infants. The presence of non-phthalate additives is generally overlooked whereas this study indicates that the presence and extractability of multiple chemicals, some with known potential for harm, requires further attention. Given the vulnerability of the developing child to toxic insult, exposure to all toxic chemicals should be minimised. The results of this study indicate the potential for significant exposure to toxic chemicals from PVC toys and calls into question the appropriateness for continued use in this application.

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Appendix: Tables 1 and 3

Table 1: Description of toys analysed, with country of purchase and of manufacture

No:	Bought in:	Made in:	Description
6065	Argentina	China	Animal toy
6067	Argentina	China	Teether
6068	Argentina	Mexico	Teether
6069	Argentina	China	Animal toy
6071	Argentina	not known	Animal toy
6023	Australia	China	Teether
6024	Australia	not known	Teether
6025	Australia	China	Teether
6026	Australia	China	Teether
6027	Australia	China	Teether
6033	Australia	China	Teether
6034	Australia	China	Teether
6041	Austria	Italy	Teether
6042	Austria	Italy	Teether
6049	Belgium	China	Teether
6050	Belgium	China	Baby book
6051	Belgium	Spain	Nasal aspirator
6052	Belgium	Spain	Teether
6053	Belgium	China	Baby book
6063	Canada	China	Bracelet doll's house
6064	Canada	China	Inflatable toy
7013a	Canada	China	Key teether - soft section
7013b	Canada	China	Key teether - hard section
7013c	Canada	China	Key teether - hard ring
7014	Canada	China	Animal toy
6035	Denmark	Macau	Doll
6038	Denmark	Denmark	Teether
6032	France	France	Bath toy
6036	France	Mexico	Teether
6083	Germany	Hong Kong	Teether
6085	Germany	China	Bath toy
6086	Germany	China	Bath toy
6054	India	not known	Teether
6055	India	not known	Teether
6056	India	not known	Animal toy
6057	India	India	Doll
6061	India	not known	Inflatable toy
6078	Indonesia	China	Teether
6081	Indonesia	not known	Teether
6028	Japan	Hong Kong	Aeroplane
6029	Japan	Japan	Doll
6039	Japan	Japan	Doll
6040	Japan	China	Toy food
6006	Netherlands	China	Plastic duck
6008	Netherlands	China	Baby book
6072	Netherlands	China	Doll
6073	Netherlands	Thailand	Animal toy
7001	Philippines	China	Teether
7002	Philippines	USA	Pacifier
7005	Philippines	China	Toy food
7006a	Philippines	China	Bath toy - clam
7006b	Philippines	China	Bath toy - frog
7007	Philippines	USA	Teether
7008	Philippines	UK	Teether
7009	Philippines	not known	Teether
7010	Philippines	China	Teether
7011	Philippines	Philippines	Teether
7012	Philippines	China	Inflatable toy
6015	Spain	China	Animal toy
6020	Spain	China	Teether
6013	Sweden	China	Vinyl ball
6014	Sweden	China	Vinyl ball

Table 1: Description of toys analysed, with country of purchase and of manufacture (*continued*)

No:	Bought in:	Made in:	Description
6016	UK	China	Teether
6018	UK	USA	Teether
6022	UK	UK	Teether
6043	UK	not known	Rings on a stick
6044	UK	China	Animal toy
6045	UK	China	Animal toys
6046a	UK	China	Gift set- teether
6046b	UK	China	Gift set - head
6046c	UK	China	Gift set - squeeze bulb
6001	USA	China	Teether
6002	USA	China	Doll
6004	USA	China	Teether
6005	USA	USA	Teether
6047	USA	China	Teether
6048	USA	China	Baby book

Table 3: Quantities of phthalate esters and alkyl phenols detected in children's toys. Where a toy is recorded as being part PVC, a PVC subsection was analysed. n/d: not detected; present: detected but not quantified; *: uncertainty in quantification due to disparity between sample and analytical standards

Sample	PVC?	% DEP	% DnBP	% DIBP	% BBP	% DnOP	% DEHP	% DIOP	% DnNP	% DINP	% DIDP	Total % phthal.	% nonyl phenol
Toys bought in Argentina													
6065	Yes	n/d	n/d	n/d	n/d	n/d	**0.06**	n/d	n/d	**33.5**	n/d	**33.6**	**0.15**
6067	Part	n/d	n/d	n/d	n/d	n/d	**0.1**	n/d	n/d	n/d	**20.1**	**20.2**	n/d
6068	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**0.009**
6069	Yes	n/d	n/d	n/d	n/d	n/d	**0.04**	n/d	n/d	**43.9**	n/d	**44.0**	**0.34**
6071	Yes	n/d	n/d	n/d	n/d	n/d	**0.006**	n/d	n/d	**37.7**	n/d	**37.7**	n/d
Toys bought in Australia													
6023	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6024	Part	n/d	n/d	n/d	n/d	n/d	n/d	present	n/d	n/d	n/d	present	n/d
6025	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**31.7**	n/d	**31.7**	n/d
6026	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6027	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6033	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**27.7**	n/d	**27.7**	n/d
6034	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
Toys bought in Austria													
6041	Yes	n/d	**0.18**	n/d	n/d	n/d	n/d	n/d	n/d	**35.3**	n/d	**35.5**	n/d
6042	Yes	n/d	**0.04**	n/d	n/d	n/d	n/d	n/d	n/d	**34.6**	n/d	**34.6**	n/d
Toys bought in Belgium													
6049	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**29.2**	n/d	**29.2**	n/d
6050	Yes	n/d	n/d	n/d	n/d	n/d	**0.06**	n/d	n/d	**19.6**	n/d	**19.7**	n/d
6051	Yes	n/d	n/d	n/d	n/d	n/d	**25.7**	n/d	n/d	n/d	n/d	**25.7**	n/d
6052	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6053	Yes	n/d	n/d	n/d	n/d	n/d	**0.05**	n/d	n/d	**23.2**	n/d	**23.3**	n/d
Toys bought in Canada													
6063	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**24.3**	n/d	**24.3**	n/d
6064	Yes	n/d	n/d	n/d	n/d	n/d	**0.042**	n/d	n/d	**19.6**	n/d	**19.6**	n/d
7013A	Yes	n/d	n/d	n/d	**0.004**	n/d	**0.24**	n/d	n/d	**38.1**	n/d	**38.3**	n/d
7013B	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**0.021**
7013C	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
7014	Yes	n/d	n/d	n/d	**0.001**	n/d	**0.043**	n/d	n/d	**35.9**	n/d	**35.9**	n/d
Toys bought in Denmark													
6035	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6038	Yes	**0.16**	n/d	n/d	n/d	n/d	**1.62**	n/d	n/d	**30.6**	n/d	**32.3**	n/d
Toys bought in France													
6032	Yes	n/d	n/d	n/d	n/d	n/d	**35.5**	n/d	n/d	n/d	n/d	**35.5**	n/d
6036	Yes	n/d	n/d	n/d	n/d	n/d	n/d	present	n/d	n/d	n/d	present	n/d

Table 3: Quantities of phthalate esters and alkyl phenols detected in children's toys. Where a toy is recorded as being part PVC, a PVC subsection was analysed. n/d: not detected; present: detected but not quantified; *: uncertainty in quantification due to disparity between sample and analytical standards (*continued*)

Sample	PVC?	% DEP	% DnBP	% DIBP	% BBP	% DnOP	% DEHP	% DIOP	% DnNP	% DINP	% DIDP	Total % phthal.	% nonyl phenol
Toys bought in Germany													
6083	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**37.6**	n/d	**37.6**	n/d
6085	Yes	n/d	**0.004**	n/d	n/d	n/d	**0.02**	n/d	n/d	**25.5**	n/d	**25.5**	**0.07**
6086	Yes	n/d	**0.003**	n/d	n/d	n/d	**0.022**	n/d	n/d	**26.3**	n/d	**26.3**	**0.067**
Toys bought in India													
6054	Part	n/d	n/d	n/d	n/d	n/d	**0.78**	n/d	n/d	n/d	n/d	**0.78**	n/d
6055	Yes	n/d	n/d	n/d	n/d	n/d	**11.4**	n/d	n/d	n/d	n/d	**11.4**	n/d
6056	Yes	n/d	n/d	n/d	n/d	n/d	n/d	present	n/d	n/d	present	present	n/d
6057	Yes	n/d	**0.02**	**0.45**	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**0.46**	n/d
6061	Yes	n/d	n/d	n/d	n/d	n/d	**13.8**	n/d	n/d	n/d	n/d	**13.8**	n/d
Toys bought in Indonesia													
6078	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6081	Yes	n/d	**0.003**	n/d	**0.01**	n/d	**0.03**	**10.2**	n/d	n/d	n/d	**10.3**	n/d
Toys bought in Japan													
6028	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**33.1**	n/d	**33.1**	n/d
6029	Part	n/d	**0.006**	n/d	n/d	**0.012**	**19.1**	n/d	n/d	n/d	n/d	**19.1**	n/d
6039	Yes	n/d	n/d	n/d	n/d	n/d	**15.4**	n/d	n/d	n/d	n/d	**15.4**	n/d
6040	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**17.4**	n/d	**17.4**	n/d
Toys bought in the Netherlands													
6006	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**41.7**	n/d	**41.7**	n/d
6008	Yes	n/d	n/d	n/d	n/d	n/d	**0.16**	n/d	n/d	**16.4**	n/d	**16.5**	n/d
6072	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**46.3**	n/d	**46.3**	n/d
6073	Yes	n/d	n/d	n/d	n/d	n/d	**0.09**	n/d	n/d	**37.9**	n/d	**38.0**	**0.17**
Toys bought in the Philippines													
7001	Part	n/d	n/d	n/d	n/d	n/d	**0.004**	n/d	n/d	n/d	n/d	**0.004**	**0.024**
7002	Yes	n/d	n/d	n/d	**0.02**	n/d	**0.34**	**17.1**	n/d	n/d	n/d	**17.4**	**0.02**
7005	Part	n/d	n/d	n/d	n/d	n/d	**0.01**	n/d	n/d	**44.1**	n/d	**44.1**	**0.02**
7006A	Yes	n/d	n/d	n/d	n/d	n/d	**0.003**	n/d	n/d	**44.7**	n/d	**44.7**	**0.36**
7006B	Yes	n/d	n/d	n/d	n/d	n/d	**0.02**	n/d	n/d	**43.8**	n/d	**43.8**	**0.36**
7007	Yes	n/d	n/d	n/d	n/d	n/d	**0.005**	n/d	n/d	n/d	**15.7**	**15.7**	n/d
7008	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
7009	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
7010	Part	n/d	n/d	n/d	**0.004**	n/d	**0.05**	n/d	n/d	**51.0**	n/d	**51.0**	n/d
7011	Yes	n/d	**0.002**	n/d	**0.004**	n/d	**0.008**	n/d	n/d	**37.9**	n/d	**37.9**	n/d
7012	Yes	n/d	n/d	n/d	n/d	n/d	**16.7**	n/d	n/d	**0.4**	n/d	**17.1**	n/d
Toys bought in Spain													
6015	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**32.5**	n/d	**32.5**	n/d
6020	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**36.3**	n/d	**36.3**	n/d
Toys bought in Sweden													
6013	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**24.0**	n/d	**24.0**	n/d
6014	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**24.3**	n/d	**24.3**	n/d
Toys bought in the UK													
6016	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**37.2**	n/d	**37.2**	n/d
6018	Yes	n/d	n/d	n/d	n/d	present	n/d	n/d	present	n/d	n/d	present	n/d
6022	No	n/d	n/d	n/d	n/d	n/d	**0.01**	n/d	n/d	n/d	n/d	**0.01**	n/d
6043	No	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d
6044	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**31***	n/d	**31***	present
6045	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**30.4**	n/d	**30.4**	n/d
6046a	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**30.7**	n/d	**30.7**	n/d
6046b	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**23.4**	n/d	**23.4**	n/d
6046c	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**13.1**	n/d	**13.1**	n/d
Toys bought in the USA													
6001	Part	n/d	n/d	n/d	n/d	n/d	**0.06**	n/d	n/d	**37.2**	n/d	**37.3**	n/d
6002	Yes	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**8.80**	n/d	**8.80**	n/d
6004	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**35.7**	n/d	**35.7**	n/d
6005	Part	n/d	n/d	n/d	n/d	present	n/d	n/d	present	n/d	n/d	present	n/d
6047	Part	n/d	n/d	n/d	n/d	n/d	n/d	present	n/d	n/d	n/d	present	present
6048	Part	n/d	n/d	n/d	n/d	n/d	n/d	n/d	n/d	**14.7**	n/d	**14.7**	n/d