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Müller, Anne Kirstine; Nielsen, Elsa Ebbesen; Ladefoged, Ole

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Anne Kirstine Müller, Elsa Nielsen, Ole Ladefoged

Institute of Food Safety and Nutrition

Ministeriet for Fødevarer, Landbrug og Fiskeri
Fødevaredirektoratet

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National IT and Telecom Agency
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www.danmark.dk/netboghandel

The Danish Veterinary and Food Administration
Mørkhøj Bygade 19, DK-2860 Søborg
Tel. + 45 33 95 60 00, fax + 45 33 95 60 01
Web site: www.foedevaredirektoratet.dk

The Danish Veterinary and Food Administration is part of the Danish Ministry of Agriculture, Food and Fisheries. The Danish Veterinary and Food Administration is responsible for the administration, research and control within food and veterinary areas “from farm to fork”, as well as practical matters relating to animal protection (otherwise under the Ministry of Justice).

Making of regulations, co-ordination, research and development, take place in the Administrations center in Mørkhøj. The 11 Regional Authorities handle the practical inspection of food and veterinary matters, including import/export etc.

The central administration of The Danish Veterinary and Food Administration employ a staff of approx. 550 full-time employees, whilst the 11 regional authorities employ a further approx.1600 full-time employee.

Preface

Phthalates are widely used as plasticisers in various polymers (e.g., PVC) that are found in a wide range of consumer products including floor- and wall covering, furnishing, toys, car interior, clothing, hoses etc. Phthalates are also applied to paints and lacquers, adhesives and sealants, printing inks etc. As the phthalates are not chemically bound in the polymers, migration or emission of the phthalates from the product to the environment is likely to occur and the phthalates are thereby widespread in the environment.

The overall aim of this project, which is a part of the research activities of Centre for Environment and the Lung, financed by The Danish Environmental Research Programme 1998-2001, was to carry out exposure assessments for humans exposure to the five most used phthalates – di(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP) and benzyl butyl phthalate (BBP).

The report reflects the views and opinions of the authors, but not necessarily the views and opinion of the Danish Veterinary and Food Administration (FDIR).

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Content

Summary

Phthalates are widely used as plasticisers in the production of flexible plastic products (mainly PVC) and are thereby present in a wide range of consumer products including floor- and wall covering, furnishing, toys, car interior, clothing, hoses etc. , but they can also be applied to other products such as e.g., paints, lacquers, adhesives and printing inks.
The phthalates are not chemically bound in the polymers. Therefore, migration or emission of the phthalates from the product to water, air or other media in contact with the product is likely to occur. The emission of phthalates occurs in all life stages of a product (from production to disposal) and the phthalates are thereby widespread in the environment.

Humans may be exposed to phthalates at work (workplace exposure), by the use of phthalate containing consumer products (consumer exposure) and via the intake of food, air and water contaminated with phthalates (indirect exposure via the environment).

Within the EU, specific programs on risk assessment for new and existing chemical substances are ongoing. A Risk Assessment Report (EU-RAR) is prepared for each substance evaluated. The indirect exposure of man via the environment is generally estimated by the use of the European Union System for the Evaluation of Substances (EUSES) - a computer modelling program. For consumer exposure, a number of computer models have been developed; no particular model is recommended above any other.

EUSES is not specifically designed for site-specific assessments as defaults represent a standard EU region, but adjustment of parameters may allow for insight into specific local or regional situations. Therefore, the overall aim of this project was to refine EUSES to a “DK-USES” in order to carry out exposure assessments for human exposure in Denmark to five of the most widely used phthalates – di(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP), and benzyl butyl phthalate (BBP). The focus has been to assess human exposure via the environment, including food, and via consumer products, whereas exposure via the working environment has not been covered in this project. The main effort has been laid on the gathering of data for the five phthalates on measured concentrations and exposures in Denmark as well as on exposures estimated by EUSES according to the EU-RARs of these phthalates in order to evaluate if EUSES exposure estimates for the five phthalates seem reliable for the region Denmark.

In order to assess the indirect exposure of man via the environment in Denmark, a set of input parameters specific for the region Denmark was applied to EUSES. Based on the specific Danish profile, the Danish tonnages of the phthalates and specific usages, and substance specific information as given in the EU-RARs of each phthalate, predicted environmental concentrations (PECs) and exposures have been estimated. Measured concentrations in the Danish environment and in Danish food obtained by monitoring studies have been compared with the estimated concentrations and exposures. For a refinement of the EUSES exposure estimations, monitoring data from Denmark have also been used as input data in EUSES.

Consumer exposures have been estimated for several different exposure scenarios: toys, building materials etc., infant formulae and baby food, artificial leather and gloves, paints etc., and nail

polish. The estimation of the exposures are different in the various scenarios depending on the available data, but exposures have generally been estimated based on e.g., total amounts in the consumer products, emission data from products (e.g., toys, building materials) as well as measured concentrations (e.g., infant formulae and baby food, house dust). Furthermore, the computer exposure modelling program CONSEXPO has been used for estimations of dermal exposure to nail polish and paints. Also measured and/or estimated exposures from the EU-RARs have been included.

The use of a specific Danish profile in EUSES only changed the exposure assessment (DEHP as model substance) slightly when compared with the estimations based on EUSES with the standard profile.

DEHP and DBP were predicted to occur in highest concentrations in the environmental compartments and therefore also resulted in the highest estimated total daily intake. In general, the predicted environmental concentrations (PECs) of DEHP and DBP in many of the environmental compartments were within the range of available measured concentrations. However, the PECs were not always worst-case concentrations as e.g., in soil, the estimated concentration was at the lower end of the range of measured values. Only for DBP in surface water, the estimated concentration exceeded all measured concentrations.
Comparisons of estimated and measured concentrations of DIDP, DINP and BBP in environmental compartments were generally not possible. For DINP, measured data were only available for the soil compartment; for DIDP, no measured concentrations were available; and for BBP, no EUSES estimations have been performed.

Very few measurements have been made on the concentration of phthalates in food and drinking water in Denmark.
The estimated concentrations of DEHP in crops and milk were much lower than the few available measured concentrations of DEHP in Danish products, whereas the estimated concentration in fish was comparable to measured concentrations of fish in other European countries. Also, the total daily intake of DEHP and DBP estimated by EUSES appear to be underestimated when compared to measured values.
For drinking water, the measured concentration (only one measurement) of DBP was lower than the estimated concentration whereas for DEHP, the measured concentration was much higher than the estimated one.
Comparisons of estimated and measured concentrations of DINP, DIDP and BBP in food and drinking water have not been performed. For DINP and DIDP, no Danish measurements of the concentrations in food and drinking water are available. For BBP, no measurements of the concentrations in food are available, one measured concentration is available for drinking water, but no EUSES estimations have been performed.

Several reasons for the discrepancies between estimated and measured concentrations in environmental compartments and in food and drinking water can be suggested. EUSES might be an inappropriate model in order to estimate the exposure to the phthalates due to their special properties such as low water solubility, low vapour pressure etc. It could also be due to poor data input into the model as both the physico-chemical data and the data regarding the Danish tonnages of the substances and specific usages are subjected to uncertainties. Also, the parameters used to estimate the uptake of substances in crops, the bioconcentration factor in fish, and the bioaccumulation in meat and milk can be incorrect and thereby, EUSES predicts too low

concentrations in food. Furthermore, the measured concentrations in food also include phthalates entering the food during processing, by contact to packaging materials etc. Food packaging materials can lead to a substantial contamination of the food; however, this contribution is not included in the EUSES exposure estimations. On the other side, measured concentrations in the environmental compartments and especially in foods and drinking water are scarce and may not be representative for the actual situation.

When PECs were replaced by measured environmental concentrations in EUSES, the estimated total daily intake increased about three times for DEHP and about 16 times for DBP. Furthermore, when estimated concentrations of DEHP in food were replaced with the few available measured concentrations in food, this resulted in a further increase (around 30 times) in the estimated total daily intake.

In order to improve the exposure assessment of man via the Danish environment, the results of this project document a need for further data regarding concentrations of phthalates, especially in the various foodstuffs including crops that, according to the EUSES estimations, contribute significantly to the total daily intake via the environment, but also of phthalate concentrations in the various environmental compartments.

Phthalates occur as contaminants in infant formulae and ready-to-use baby food. The daily intake was estimated based on measured concentrations in infant formulae and baby food on the Danish market. In general, the daily intake of phthalates by infants via infant formulae and ready-to-use baby food is approximately the same as the estimated daily intake of phthalates by children via the environment (food in general).

Children are exposed to phthalates during their playing with toys. The potential exposure via mouthing phthalate-containing toys (worst case scenario) has been estimated to be relatively high. Furthermore, dermal exposure to phthalates also occurs during their handling of toys; however, this contribution was low compared to the oral exposure. In Denmark, the use of phthalates in toys and other childcare articles intended for children 0-3 years old are restricted (maximum concentration 0.05% (w/w)). However, investigations of toys on the Danish market, the lack of regulation of phthalates in toys intended to older children as well as the allowed use of phthalates in inflatable toys suggest that exposure to phthalates via toys still can occur.

The exposure via indoor air to phthalates emitted from building materials, furniture, cables etc. has been estimated based on measured vapour phase concentrations of the phthalates and the concentrations of the phthalates adsorbed to house dust in Danish homes. The present assessment indicated that the vapour phase and the house dust contributed equally to the exposure via indoor air. The contribution of airborne phthalates to the total exposure is low compared with other sources.

Consumers can also be exposed to phthalates via dermal contact to phthalate-containing products such as waterproof clothes, boots, gloves and artificial leather. The exposure seemed in some cases to be relatively high as e.g., for adults wearing DEHP-containing gloves.

The dermal and inhalatory exposures to phthalates via use of paints etc. and nail polish (DBP only) were estimated to be very low. DEHP and DBP have been classified as toxic to reproduction, which means that concentrations above 0.5 % are no longer allowed in chemical

products sold for private uses. It is therefore expected that exposure of consumers to these phthalates via chemical products like paints, lacquers, glue etc. will decrease in the future.

As for the exposure to phthalates via food and the environment, this project also documents that the estimations of consumer exposure would be improved considerably if additional measurements of phthalates in consumer products are carried out.

When combining all the exposure pathways, the most important route of exposure to the phthalates assessed in the present report is the oral route. Food is the dominant source, no matter how the EUSES estimations were made (input data), except for DEHP, DINP and DIDP in the young children, where also oral exposure through mouthing on toys potentially could play a significant role.

Introduction

Phthalates are widely used as plasticisers in a range of polymers (e.g., PVC) that are found in a wide range of consumer products including floor- and wall covering, furnishing, toys, car interior, clothing, hoses etc. Phthalates are also applied to paints and lacquers, adhesives and sealants, printing inks etc.
The phthalates are not chemically bound in the polymers. Therefore, migration or emission of the phthalates from the product to water, air or other media in contact with the product is likely to occur. The emission of phthalates occurs in all life stages of a product (from production to disposal) and the phthalates are thereby widespread in the environment.

Five of the most widely used phthalates are di-(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP) and benzyl butyl phthalate (BBP). These are the phthalates in focus in the present report.

Within the EU, specific programs on risk assessment for new and existing chemical substances, including the above-mentioned phthalates, are ongoing. A directive (EEC 1993a) provides the regulation on risk assessment of new notified chemical substances[1] and two Council regulations (EEC 1993b, EEC 1994) on risk assessment of existing substances.

The risk assessment process, in relation to both human health and the environment, entails a sequence of actions:
1) Effect assessment comprising a) hazard identification (identification of the adverse effects which a substance has, and b) dose (concentration) - response (effects) assessment (estimation of the relationship between dose, or level of exposure to a substance, and the incidence and severity of an effect.
2) Exposure assessment (estimation of the concentrations/doses to which human populations or environmental compartments are exposed).
3) Risk characterisation (estimation of the incidence and severity of the adverse effects likely to occur in a human population or environmental compartment due to actual or predicted exposure).

A Risk Assessment Report (EU-RAR) is prepared for each prioritised substance. The EU-RAR for DBP was published 18 March 2003 (http//www.ecb.jrc.it); however the final draft for decision on the conclusions by the EU Competent Authorities (DBP 2001) has been used in the present report**.** Regarding DEHP, DINP and DIDP, the EU-RARs have not yet been published and the final drafts for decision on the conclusions by the EU Competent Authorities have been used in the present report (DEHP, 2001; DINP, 2001, DIDP, 2001). For BBP, discussions are still ongoing in the EU Working Group and the draft report is not yet public available.

The exposure assessment and risk characterisation are carried out separately for three subgroups of the human population: workers, consumers, and man exposed indirectly via the environment.

Indirect exposure of humans via the environment may occur by consumption of food (fish, crops, meat and milk) and drinking water, inhalation of ambient air, and ingestion of soil. For existing substance, measured levels in various environmental compartments may be available; however,

[1] Substances not on the EU market in the 10 years prior to 18 September 1981 and therefore not appearing in the European INventory on Existing Commercial chemical Substances (EINECS).

for new substances, usually no relevant measured data are available and concentrations of a substance in the environment must be estimated. The indirect exposure is estimated by the use of the European Union System for the Evaluation of Substances (EUSES) - a computer modelling program. EUSES estimates concentrations in food and the total daily intake of a substance based on predicted environmental concentrations for (surface) water, groundwater, soil, sediment and ambient air. The indirect exposure is principally assessed on two spatial scales: locally near a point source of the substance, and regionally using averaged concentrations over a larger area. It should be noted that a third spatial scale, the continental scale, is also assessed by EUSES; however, this it not included in the estimations of the indirect exposure. EUSES can specifically be used in the initial (or screening) and intermediate (or refined) stages of assessment. On the basis of the screening, it can be decided if more data need to be generated and if a more refined assessment is necessary. EUSES can also be applied for refined assessments by allowing the replacement of default values, estimated parameter values, or intermediate results by more accurately estimated values or by measured data. EUSES is not specifically designed for site-specific assessments (defaults represent a standard region in EU), but adjustment of parameters may allow for insight into specific local or regional situations.

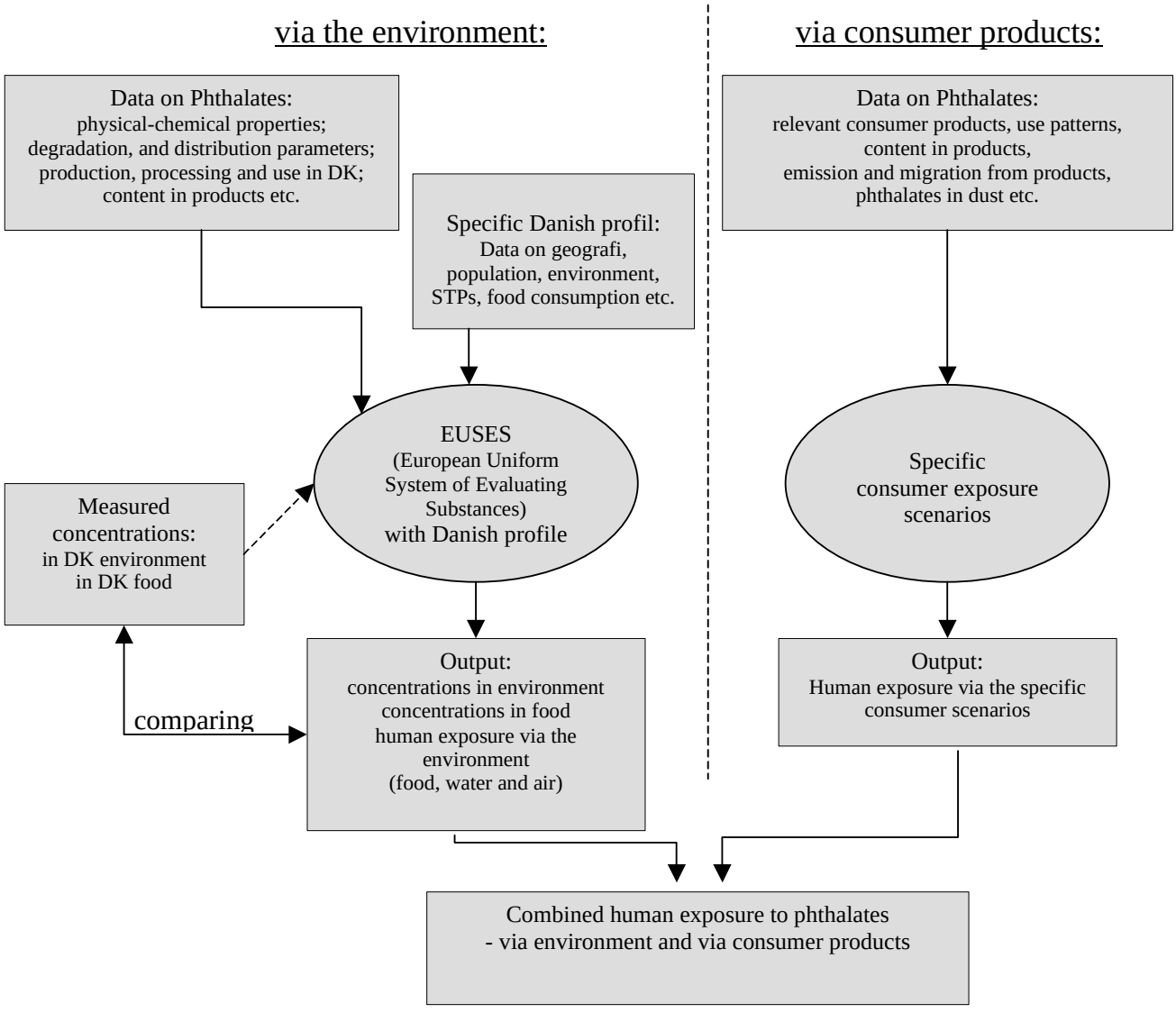
The consumer, i.e. a member of the general public who may be of any age, either sex, and in any stage of health may be exposed to a new or existing substance by using consumer products. A consumer product is one, which can be purchased from retail outlets by members of the general public and may be the substance itself, or a preparation, or an article containing the substance. To assess the exposure to substances present in consumer products, information is needed on two sets of parameters: 1) contact parameters (where, how long and how often contact with the consumer occurs) and 2) concentration parameters (the concentration of as substance in a medium that might contact the body. Measured data on external exposure for specific scenarios may be available for a number of substances; however, most often such data are not available. A number of computerised models have been developed; no particular model is recommended above any other. Whenever possible measured or estimated values should be used for each of the numerical parameters in a given model, but when this is not possible default values may be derived from available data sources. Within the EU available consumer products are more or less the same and the consumer exposure assessments in the EU-RARs are likely also to represent more site-specific assessments and thus, are representative for Danish consumers as well. However, in some cases, measured data from Denmark exist as e.g., in dust from Danish homes.

The overall aim of the project is to refine EUSES to a “DK-USES” in order to carry out exposure assessments for the human exposure in the region Denmark to the five phthalates – di(2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), di-isononyl phthalate (DINP), di-isodecyl phthalate (DIDP) and benzyl butyl phthalate (BBP). The focus will be on the human exposure via the environment, including food, and via consumer products; exposure via the working environment is not included in this project. The main effort will be laid on the gathering of data for the five phthalates on measured concentrations and exposures in Denmark as well as on exposures estimated by EUSES according to the EU Risk Assessment Reports (EU-RAR) of these phthalates in order to evaluate if EUSES exposure estimates for the five phthalates seem reliable for the region Denmark.

In the first part (Part 1) of the report, indirect exposure via the environment is assessed (see Fig.1). The indirect exposure via the environment is assessed by the use of EUSES. Instead of the standard default input parameters representing a standard region in EU and used in the EU-

RARs, a set of input parameters specific for Denmark will be used. Based on the specific Danish profile, the Danish tonnages of the substances and the specific usages, and the substance specific information as given in the EU-RARs of each phthalate, predicted environmental concentrations (PECs) and exposures are estimated. Measured concentrations in the Danish environment and in Danish food obtained by monitoring studies will be compared with the estimated concentrations and exposures. For a refinement of the EUSES exposure estimations, monitoring data from Denmark will also be used as input data in EUSES. Furthermore, it will be evaluated whether the exposure assessments in the EU-RARs of the five phthalates apply for Denmark as well.
In the estimations made by EUSES, all substance specific information will be used as reported in the EU-RARs of each phthalate and there will be no detailed discussion in this report on substance properties.

Fig 1. An overview of the assessment of human exposure to phthalates in Denmark



In the second part (Part 2) of the report, the consumer exposure is assessed (see Fig. 1). Consumer exposures have been estimated for several different exposure scenarios: toys, building materials etc., infant formulae and baby food, artificial leather and gloves, paints etc., and nail polish. The estimation of the exposures are different in the various scenarios depending on the available data, but exposures have generally been estimated based on total amounts in the consumer products, emission data from products (e.g., toys, building material) as well as measured concentrations (e.g., infant formulae and baby food, house dust). Furthermore, the computer exposure modelling program CONSEXPO has been used for estimations of dermal exposures to nail polish and paints. Measured and/or estimated exposures from the EU-RARs have been included.

In the third part (Part 3) of the report, the exposures from all the pathways and all the routes estimated in Part 1 and Part 2 will be combined in order to quantify the combined exposure via the environment and via consumer products.

PART 1: Indirect exposure via the environment

1 Introduction

The indirect exposure of humans to chemical substances via the environment may occur predominantly through ingestion of in food and water, and by inhalation of ambient air, but also via exposure to soil (ingestion and dermal contact). Phthalates are widely distributed in the environment. In Denmark, they are found in all environmental compartments.

The indirect exposure to the five selected phthalates via the environment in Denmark will be estimated by EUSES after entering of site-specific (Danish) defaults. First (section 2), the site specific input data will be presented. Thereafter, the predicted environmental concentrations (PECs) and estimated concentrations in food (section 3) and measured concentrations in the Danish environment and food (section 4) will be presented. Finally, the data are compared (section 5) and discussed (section 6).

2 Input parameters

The EUSES program has default values for most of the parameters in the model. The exception is a few substance specific parameters - physico-chemical properties and tonnage - that have to be entered by the user. The default values in the program are intended to represent a standard region in Europe. To make the prediction representative for Denmark, a Danish country profile has been applied in the present project, including e.g., geographical data, number of inhabitants, size of sewage treatment plants (STPs), and figures for food intake.

At the regional scale, diffuse, continuous emissions to a standard environment are considered. Steady-state partitioning between environmental compartments is assumed and the regional computation is performed by means of the SimpleBox model.

2.1 Specific Danish profile

In Denmark, much effort has already been made to investigate how Danish conditions differ from the original Uniform System for the Evaluation of Substances - USES 1.0 – which was developed in 1994 in The Netherlands and therefore based on Dutch conditions (RIVM et al., 1994).
These efforts resulted in different suggestions of how the various models – SimpleBox, SimpleTreat, OPS, Pestla and Slootbox - included in the USES could be changed to be more suitable for Danish conditions (Fredenslund et al., 1995a; Fredenslund et al., 1995b; Mikkelsen, 1995). Some of these suggestions have been included in the present version, EUSES 1.0, e.g., the option to estimate emission distribution in sewage treatment plants with or without a primary settler. The specific Danish geographical situation, with the ocean and marine sediment, was also taken into consideration by the development of a six-compartment model, which, however, is not yet included in EUSES (which has a four-compartment model).

Furthermore, a country file defining the Danish standard scenarios was established (Fredenslund et al., 1995a). The authors clearly pointed out that the country file should be used with great care since other parameters than those in the profile are also important. Although, the country profile did not include all relevant variables for Denmark, the parameters suggested by Fredenslund et al. (1995a) and Mikkelsen (1995) will, in the present project, be changed to values specific for Denmark to make the exposure estimation more representative for Denmark. Some of the values from Fredenslund et al. (1995a) and Mikkelsen (1995) are still suitable; others have to be updated.

2.1.1 Geography, population and environment

The specific values for the distribution of areas, the number of inhabitants, the weather conditions etc. that are used as specific Danish parameters are given in Table 2.1. together with the EUSES default values.

Table 2.1. The default standard regional profile in EUSES and the specific Danish country profile used in the present project.

Spatial scale	Parameter	EUSES[5]	Denmark	Ref
Regional	Size of regional STP[6]	2,000,000 PE[7]	8,500,000 PE	[1]
Regional	Area	40,000 km^2	43,098 km^2	[2]
Regional	Fraction $Area_{water}$	0.03	0.015	[2]
Regional	Fraction $Area_{natural}$	0.6	0.212	[2]
Regional	Fraction $Area_{agricultural}$	0.27	0.633	[2]
Regional	Fraction $Area_{urban}$	0.1	0.140	[2]
Regional	Fraction of water flow to the region	0.034	0.00	[3]
Regional	Water depth of system	3 m	3.2 m	[3]
Regional	Suspended solids concentration	15 mg l^{-1}	18 mg l^{-1}	[3]
Regional and cont.	Average annual precipitation	700 mm yr^{-1}	712 mm yr^{-1}	[2]
Regional and cont.	Fraction of rainwater infiltrating soil	0.25	0.36	[3]
Regional and cont.	Fraction of rainwater running off soil	0.25	0.10	[3]
Local	Average air temperature	12°C	7.7 °C	[2]
Local	Windspeed	3 m s^{-1}	5 m s^{-1}	[2]
Local	Dry sludge application rate, agri. soil	5,000 kg ha^{-1} yr^{-1}	3,000 kg ha^{-1} yr^{-1}	[4]

[1] MST (2001)
[2] Statistic Denmark (2001)
[3] Fredenslund et al. (1995a)
[4] MST (2002), personal communication
[5] EC (1996)
[6] STP: Sewage treatment plant
[7] PE: Personal equivalent

The ideal situation would be to perform an EUSES estimation, in which the Danish profile was only used for the regional scale estimations. However a few of the parameters that are specific for Denmark, are also used by EUSES for the exposure estimations on the continental scale. Nevertheless, as the regional scale estimations are more essential for this project, the specific Danish values will be applied and therefore also used for some of the continental scale estimations (see Table 2.1).

The forests in Denmark will, in the present area distribution, be considered as natural areas even though most of them are cultivated. In several ways, they are more comparable to the natural soil compartment than to the agricultural soil compartment in EUSES. The mixing depth of the forest soil equals the natural soil and not the agricultural soil mixing depth and furthermore, forests do not normally receive sludge.

According to Statistical Yearbook (Statistic Denmark, 2001), the fraction of urban areas in Denmark in 1995 was 19%. This high fraction also includes summer dwelling areas and scattered residences. These areas are not comparable to the industrial soil compartment in EUSES since many of these areas do not receive any direct emission. Therefore, some of the areas included in the urban areas in the Statistical Yearbook are more comparable to the natural soil compartment than to the industrial soil compartment in EUSES. In the present Danish country profile, the area fraction of industrial soil is set to 0.14 (5% less than the value in the Statistical Yearbook). On the other hand, the area fraction of natural soils (including forests) is set to 0.212, which is 5% higher than the values in the Statistical Yearbook.

The parameters concerning the Danish weather conditions are 30 years averages (1961-90). Denmark is almost entirely surrounded by marine waters and there are no streams crossing the boundaries. There is no freshwater flow into Denmark.

There are huge variations in soil types throughout Denmark and therefore, a representative fraction of water infiltrating or running off soils is difficult to determine. The chosen values are from Fredenslund et al. (1995a).

The sludge application rate of 3 tonnes/ha is chosen as a realistic worst case situation. Although 3 tonnes/ha is a commonly applied amount of sludge, this amount is normally only applied every 3 years because of the high content of phosphor in the sludge. Furthermore, only around 1% of the agricultural soils in Denmark receive sludge (municipal).

2.1.2 Sewage Treatment Plants (STPs)

Many of the values for the STPs in Denmark that will be used in the present project are from Mikkelsen (1995) with some more recent information from MST (2001). Mikkelsen (1995) selected specific Danish values to be used in the SimpleTreat model for simulation of the fate of organic chemicals in (STPs). SimpleTreat model is a part of EUSES. The values are primarily based on measurements in Danish STPs (Table 2.2).

According to MST (2001), 1363 STPs existed in Denmark in 2000, which received a total amount of wastewater of 8,500,000 population equivalents (PE). The size of the STPs varies from a capacity of approximately 30 PE to a capacity of more than 100,000 PE. The maximum

loading of a STP was around 500,000 PE. Sixty-one of the STPs had a capacity of more than 50,000 PE and they received 68% of the total amount of wastewater. Therefore, 50,000 PE seems to be a realistic worst-case size of a Danish STP. The average sewage flow is according to MST (2001) 235 litre/PE/day.

According to the Technical Guidance Document (TGD, 1996), the fraction of wastewater connected to STPs in Denmark is high (0.98) whereas the average fraction in EU is 0.7 (0.8 in the revised TGD, 2003). The fraction is important for the distribution of a substance in the environment. A value of 0.98 is used as the Danish (the regional) fraction of wastewater connected to sewage treatment plants, but for the estimations of the continental emission to the environment, the average European fraction of 0.7 is used.

Table 2.2. EUSES defaults and the specific Danish profile of the Sewage Treatment Plants (STPs).

Parameter	EUSES[5]	Denmark	Ref.
Sewage flow	200 l d^{-1} PE^{-1}	235 l d^{-1} PE^{-1}	[1]
Fraction connected to sewer system, Regional	0.7	0.98	[2]
Fraction connected to sewer system, Continental	0.7	0.7	[2]
Number of population equivalents (PE)/STP	10,000 PE	50,000 PE	[1]
Temperature of air	15°C	12°C	[3]
Temperature of water	15°C	12°C	[3]
Height of air column above STP	10 m	4 m	[3]
Depth of primary settler	4 m	3 m	[3]
Depth of solid-liquid separator	3 m	4 m	[3]
Dry weight solids produced pr. person	0.09 kg PE^{-1} d^{-1}	0.086 kg PE^{-1} d^{-1}	[3+4]

[1] MST (2001)
[2] TGD (1996)
[3] Mikkelsen (1995)
[4] Fredenslund et al. (1995a)
[5] EC (1996)

2.1.3 Food consumption

A nation-wide survey on dietary habits among 1,837 adults and 1,261 children in Denmark has been carried out (Andersen et al., 1995). The study population included both adults and children grouped in two age groups (1-6 and 7-14 years) who provided detailed data on their food consumption during a period of 7 days.
The consumption of various food groups given in ECETOC (2001), and also recommended for use in the TGD (1996), is based upon European Marketing Data and Statistics. The figures are average values and they do not include children. In the standard EUSES, the maximum consumption figures for each foodstuff from any country are used as defaults.

In Table 2.3, the food consumption values from both sources are presented together with the EUSES defaults. The rows printed in bold are the categories needed as defaults in EUSES. The 90-percentile from the survey of Andersen et al. (1995) is lower for meat and fish than the values from ECETOC (2001), but for all the other categories, it is higher.

In the present exposure assessment, the values from Andersen et al. (1995) for all three age groups will be used. Since it is important that the exposure assessment includes the majority of the Danish population, the 90-percentile will be used as a realistic worst-case scenario. It is important to remember that when the 90-percentile for all of the food categories is used, the total amount of food consumed in a day is high. The average food consumption from the Danish survey is presented in Appendix 1.
In EUSES, the intake of crops (fruit, vegetables and cereals) has to be separated into the intake of root and leaf crops (including fruit and cereals). The consumption of root vegetables by the adult participants in the Danish survey was approximately 65% of the total intake of vegetables (Larsen et al., 2002). The remaining vegetable consumption (35%) together with the consumption of fruit and cereals is used as leaf crops consumption.

Table 2.3. Food consumption rates (kg/day) from Denmark and the EUSES defaults.

Main category	EUSES category	Food intake (kg/day)				
		Adult			Child (7-14 yr)	Child (1-6 yr)
		Average[1]	90 percentile	EUSES [6]	90 percentile	90 percentile
Meat			0.199[2]		0.148[2]	0.085[2]
Poultry			0.049[2]		0.045[2]	0.021[2]
	Meat	**0.298**	**0.248[3]**	**0.301**	**0.193[3]**	**0.106[3]**
Fish	**Fish**	**0.115**	**0.052[2]**	**0.115**	**0.037[2]**	**0.026[2]**
Milk etc.			0.691[2]		0.909[2]	0.765[2]
Cheese			0.062[2]		0.042[2]	0.027[2]
	Dairy	**0.257**	**0.753[4]**	**0.561**	**0.951[4]**	**0.792[4]**
Fruit			0.327[2]		0.335[2]	0.297[2]
Vegetables			0.381[2]		0.306[2]	0.207[2]
Cereals			0.325[2]		0.312[2]	0.226[2]
	Leaf crops	**0.665**	**0.786[5]**	**1.200**	**0.754[5]**	**0.598[5]**
	Root crops	**0.172**	**0.247[5]**	**0.384**	**0.199[5]**	**0.135[5]**
Water etc.	**Water**	**2.000**	**3.127[2]**	**2.000**	**1.321[2]**	**0.764[2]**

[1] ECETOC (2001)
[2] Data for the main categories is from Andersen et al. (1995). Values for the EUSES categories are estimated based on the data from Andersen et al. (1995).
[3] Meat = meat + poultry
[4] Dairy = milk + cheese
[5] A fraction of 0.65 of the total consumption of vegetables is root vegetables (Larsen et al., 2002). The remaining together with the fruit and cereals are leaf vegetables.
[6] EC (1996)
bold: The parameters needed in EUSES

The consumption of water as given in the Danish survey includes - besides drinking water - also consumption of tea, coffee, fruit syrup etc. Since water is a main ingredient in these beverages, the value for total consumption of water will be used as the drinking water intake.

2.1.4 Body weight and inhalation rate

The body weight of a Danish adult is set to 70 kg and the inhalation rate (i.e., ventilation) to 20 m^3/day.
For the 2 groups of children (1-6 years and 7-14 years), it is difficult to choose a representative body weight and inhalation rate for the whole group since there are big differences between e.g., a 1-year and a 6-year old child. To be certain that the children with the highest relative inhalation rate are included, body weights and inhalation rates are chosen to represent the smallest children in each group. In the prediction of the human exposure via the environment, the body weight is used to estimate the total daily intake of the specific substance pr. kg body weight. The smallest children in a food-intake group (e.g., 1-6 years) also have the highest rate of food intake pr. kg body weight and therefore, have the highest intake of the specific substance. Therefore, the body weight of children between 1 and 6 years will be set to 8 kg and the inhalation rate to 9.3 m^3/day as suggested in the EU-RARs of the five selected phthalates (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001). The body weight of the children between 7 and 14 years will be set to 26 kg and the inhalation rate to 13 m^3/day. These are values, which have been published for 6-8 years old children (US-EPA, 1997).

2.1.5 Bioavailability

The exposure route dependent bioavailability values given in Table 2.4 are taken from the EU-RARs of each phthalate (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002). For a further discussion of the values, the reader is referred to the specific reports.

Table 2.4. Bioavailability depending on exposure route.

Route of exposure	Group	DBP	DEHP	DINP	DIDP	BBP
Oral	Adults	100%	50%	50%	50%	100%
	Infants	100%	100%	100%	100%	100%
Inhalation	Adults	75%	75%	75%	75%	100%
	Infants	75%	100%	100%	100%	100%
Dermal	Adults	100%	5%	0.5%[1]	0.5%[1]	5%
	Infants	100%	5%	0.5%[1]	0.5%[1]	5%

[1]By assuming a 10 times lower absorption than for DEHP

2.2 Substance identity and physico-chemical properties

As already mentioned, data on substance identity and physico-chemical properties are some of the parameters that need to be entered in EUSES. The substance identity and the physico-chemical data for the phthalates of interest are listed in Table 2.5. In general, there are wide ranges of values for some of the properties, but characteristic for the phthalates is the low vapour pressure and the high partition coefficient (log K_{ow}). Only those parameters that are obligatory to run EUSES are presented and for each parameter, the Table gives the value used in the EU-RAR of each phthalate (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002). For further details, the reader is referred to the specific reports.

For some of the parameters, the value is higher (or lower) than what is recommended as a maximum (or minimum) in EUSES. This is the case for the log K_{ow} of DEHP, DIDP and DINP, and for the water solubility of DIDP and DINP. In the present exposure assessment, the actually measured values will be used regardless of whether they are above (or below) the recommended value and consequently outside the validated range for the parameters used in the model. In order to evaluate the influence of these parameters for the resulting exposure estimates, the distribution to environmental compartments of DIDP will also be estimated with the maximum and minimum recommended values.

Table 2.5. The physico-chemical properties of the phthalates.

Property	DBP[1]	DEHP[1]	DINP[1]	DIDP[1]	BBP[1]
CAS No	84-74-2	117-81-7	68515-48-0 / 28553-12-0	68515-49-1 / 26761-40-0	85-68-7
EINECS No.	201-557-4	204-211-0	271-090-9 / 249-079-5	271-091-4 / 247-977-1	201-622-7
Molecular formula	$C_{16}H_{22}O_4$	$C_{24}H_{38}O_4$	$C_{22}H_{42}O_4$[2]	$C_{28}H_{46}O_4$[2]	$C_{19}H_{20}O_4$
Molecular weight (g/mol)	278.34	390.6	420.6[1]	446.68[1]	312.4
Melting point (°C)	-69	-55	-50	-45	-35
Boiling point (°C)	340	230	440	400	370
Vapour pressure (Pa)	0.0097 at 25°C	0.000034 at 20°C	0.00006 at 20°C	0.000051	0.00004 at 20°C
Log K_{ow}	4.57	7.5 (7.0)	8.8 (7.0)	8.8 (7.0)	4.84
Water solubility (mg/l)	10	0.003	0.00061 (0.001)	0.0002 (0.001)	2.8

[1] The physico-chemical properties are quoted from the EU-RARs (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002)
[2] Given as an average of the complex mixture of isomers
(): Values in brackets are the maximum or minimum recommended value in EUSES.

The term ‘DIDP’ actually covers two different di”isodecyl” phthalates with two different CAS numbers. Each of these DIDPs are mixtures of 1,2-benzenedicarboxylic acid, di-C9-11-branched alkyl esters (C10-rich). These two phthalates are prepared essentially identical processes and are considered to be fully interchangeable within their whole range of market end-uses.

There are 3 different DINPs covered by 2 different CAS numbers; one of the DINPs has not been produced since 1995. Each of these DINPs are 1,2-benzenedicarboxylic acid, di-C8-10-branched alkyl esters (C9-rich), but are manufactured by different processes and thus, these 3 DINPs could have different properties.
Therefore some of the properties for DIDP and DINP in Table 2.5 are averages. The human exposure to DINP and DIDP will be evaluated as if only one mixture of each exist.

2.3 Specific values for distribution and degradation of the phthalates

The amount of substance in an environmental compartment is not only the consequence of the direct emission to the compartment, but also of the transport, distribution and the degradation of the substance in the environment. EUSES is able to estimate all the necessary degradation and distribution parameters on the basis of the physico-chemical properties listed in the previous section. However, some of the values have been investigated experimentally and, in accordance with the EU-RARs (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001), the measured values will be used instead of the EUSES estimations (Table 2.6) in the present assessment. For detailed information on each value, the reader is referred to the specific EU-RARs.

The values in brackets are estimated by EUSES. Some parameters, e.g., the half-life in soil, are identical for all four substances even though the value has been set for DEHP, DINP and DIDP, and estimated by EUSES for DBP. If EUSES had estimated the half-lives in soil for DEHP, DINP and DIDP, the half-lives would have been longer (3000 days) than the half-life estimated for DBP because of the different physico-chemical properties of DBP compared to DEHP, DINP and DIDP. The differences in physico-chemical properties between the phthalates are therefore not reflected in the half-life in soil and sediment of the specific phthalates when using the values from the EU-RARs.

The values for the different parameters of DEHP, DINP and DIDP are very similar, but for DBP, especially the organic carbon-water partitioning coefficient (K_{oc}) and the bioaccumulation factor for fish are much lower than for the other substances.

In the present assessment, all other parameters than those listed in Table 2.6, which are used to estimate degradation and distribution are estimated by EUSES for all four phthalates.

2.4 The amount of phthalates in production and use

In order to carry out the assessment of emission to the environment in EUSES, detailed information on the tonnage and the use of the substance in Denmark is important.
EUSES estimates emissions on three different spatial scales: The continental, the regional and the local.

EU represents the continental scale. Information from the EU-RARs of each phthalate (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001) on the European tonnage of a specific substance will therefore be used as the continental tonnage. Since the region is Denmark, the amounts of a given substance used in specific product categories in Denmark will be used for the regional

scale. In EUSES, the local emission (i.e., near a point source) is estimated on basis of standard scenarios using the tonnage of the region.

Table 2.6. Specific degradation and distribution parameters for the four phthalates.

		DBP[1]	DEHP[1]	DINP[1]	DIDP[1]
Characterisation of biodegradability		Readily	Readily	Readily	Readily[6]
Rate constant, degradation in air (d^{-1})		-	-	0.99	0.6
Half-life (d)	air	1.8	1	-	-
	surface water	(15)	50	50[2]	50[2]
	soil	(300)	300	300[2]	300[2]
	sediment, aerobic	(300)	300	300[2]	300[2]
	sediment, anaerobic	-	Infinite	Infinite[2]	Infinite[2]
	sediment, total	(3,000)	3,000	3,000[2]	3,000[2]
Partitioning coefficient	organic C-water (K_{oc})	(6,340)	165,000	286,000[3]	286,000
	plant-water	(220)	1,940	3,432[4]	3,432[4]
Bioaccum. (l/kg)	aquatic biota	(1,530)	2,700	4,000[3]	4,000
Bioconc. Factor	fish	1.8	840	840[1]	840[1]
	earthworms	(13.3)	1	1[1]	1[1]
Bioaccum. Factor	meat	(0.00093)	0.0002[5]	(0.0794)	(0.0794)
	milk	(0.000295)	0.00006[5]	(0.0251)	(0.0251)
Fraction associated with aerosol particles(%)		-	-	100[5]	100[5]

[1] The parameters are quoted from the EU-RARs (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001)
[2] The value is assumed to be similar as for DEHP
[3] The value is assumed to be similar as for DIDP
[4] Estimated values
[5] No justifications in the EU-RARs for the value
[6] Failing the 10-day window criterion (EUSES parameter)
( ): The values in brackets are estimated by EUSES

2.4.1 EU – The continental scale

The production, import and export of phthalates

The amounts of production, import and export in EU are presented in Table 2.7. The production volumes of DINP and DIDP are from 1994, whereas the volumes of DEHP and of DBP are from 1997 and 1998, respectively. For DBP, there is a decreasing tendency in the production whereas for DINP and DIDP, the trend is an increase in the total consumption. The consumption of DEHP has been relatively constant for the last 20 years.

Table 2.7. The volumes of phthalates produced, imported and exported in EU.

Substance	Tons/year			
	Production	Import	Export	Consumption
DBP[1]	26,000	0	8,000	18,000
DEHP[2]	595,000	67,000	186,000	476,000
DINP[3]	185,200	5,400	83,400	107,200
DIDP[4]	279,000	(0)	(38,000)	200,000

[1] DBP (2001) – data from 1998
[2] DEHP (2001) – data from 1997
[3] DINP (2001) – data from 1994
[4] DIDP (2001) – data from 1994

Use patterns in EU

The use patterns of phthalates in EU, as they have been reported in the EU-RARs of each specific phthalate, are given in Table 2.8 (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001). In the present assessment, the distribution of tonnage for the specific use patterns is not only used for the continental scale, but also for the regional scale, if no other data from Denmark were available.

The use of phthalates in ceramics and grouting agents is not transferred to the regional level since there are no data on the use in ceramics in Denmark, and the grouting agents are pooled with the adhesives and sealant in one common product category. This is in concordance with the EU-RAR of DEHP, DINP and DIDP. The product categories (ceramics and grouting agents) are still relevant on the continental scale, but the regional emission is set to zero, see below.

Table 2.8. Total tonnage for the specific applications in EU.

Compound	Polymer plasticiser	Adhesive, sealants etc.	Printing Inks	Grouting Agents	Lacquers, paints	Ceramics
DBP[1]	76 %	14%	7%	3%		
DEHP[2]	97 %	2.35%	0.35%		0.3%	0.0061%
DINP[3]	97.5%	0.86%	0.86%		0.86%	
DIDP[4]	98.7%	0.135%	0.135%		0.27%	0.765%

[1] DBP (2001)
[2] DEHP (2001)
[3] DINP (2001)
[4] DIDP (2001)

2.4.2 Denmark – The regional scale

The most detailed analysis on the different product categories in Denmark for the specific phthalates is a mass-balance analysis made by Hoffmann (1996) where the major part of data is from 1992. The analysis is based on information obtained from the Danish Product Register (PROBAS), Statistic Denmark, and the Danish Trade. The analysis will be used even though data are not up-to-date.
In order to get an idea of whether the data from Hoffman (1996) are suitable to describe the present use of phthalates in Denmark, data from the mass-balance analysis on the import of pure phthalates and PVC compounds will be compared with recent data from The External Trade Statistic for year 2001 (Statistic Denmark, 2002). Also the more recent investigations on possible substitutes for the phthalates published by the Danish Environmental Protection Agency (MST, 1999; MST, 2000) will be discussed. Unfortunately, these more updated data are mainly on the entire group of phthalates with no specifications of substances and therefore, they can mainly be used to evaluate whether data from 1992 are still applicable.
Furthermore, the total amount of phthalates registered in chemical products in PROBAS in 1994 (Hoffman, 1996) will be compared with PROBAS data from 2001 (Flyvholm, 2001).

For several product categories, Hofmann (1996) did not estimate substance specific tonnages and data has to be obtained from elsewhere. Due to the limitations in the registration, it is not possible to generate these data from the Danish Product Register (PROBAS). The information in PROBAS is based on mandatory registrations of products, other data obtained by the authorities, and investigations or research projects concerning chemical products. The mandatory registration of products has mainly focused on chemical products meant for occupational uses and considered as hazardous according to the classification criteria in the EU, and products containing specific substances such as e.g., potential carcinogenic substances. Therefore, PROBAS mainly includes chemical products used in the industry besides a number of consumer products as e.g., cosmetics and cleaning agents. Several other phthalate containing products than those registered in PROBAS exist such as e.g., building materials and phthalate containing PVC products. These products contribute significantly to the total amount of phthalates on the Danish market, but they are not registered in PROBAS.
Another important limitation of PROBAS is that quantitative information is only registered for about half of the products registered containing phthalates (from 42% (DINP) to 73% (DIDP) of the products). Furthermore, the data in PROBAS are not regularly updated. These obvious limitations make the use of PROBAS-data less suitable for generating quantitative data on the Danish use-patterns.

In the EU-RARs of the specific phthalates, the fractions of substance used in specific product categories have been estimated. If it is assumed that the use-pattern of phthalates in Denmark is similar to the use-pattern of phthalates in EU (Table 2.8), these fractions can be used to estimate the Danish tonnage for several use-patterns. The information from the EU-RARs are not specific for Denmark and for several use-categories, the estimated tonnage based on these general fractions from the EU-RARs is lower than what has been registered in PROBAS (Flyvholm, 2001).
DBP in paints is not included as a use-pattern in the EU-RAR of DBP (2001), whereas DBP is the phthalate that is registered in the highest number of paints, lacquers etc. in Denmark (Flyvholm, 2001), and it is also mentioned as the dominating phthalate in paints and lacquers in

the mass-balance analysis (Hoffmann, 1996). Since no other data of DBP in paints exist, quantitative PROBAS-data will be used.

The data of most value for the exposure-assessments would be the amounts of phthalates processed, formulated and privately used in Denmark since emission from each life-cycle step is estimated separately in the EUSES. Imported products, which are not formulated or processed in Denmark, emit only phthalates during private use and waste lifecycle-steps, whereas products for export only emit phthalates during processing and/or formulation. Unfortunately, the available data from Denmark is not detailed enough to separate the yearly amounts of phthalates into amounts processed, formulated or privately used. In the present exposure assessment, it will be assumed that e.g., the amount of phthalates imported in chemical products are similar to the amounts exported in chemical products that have been processed in Denmark. Therefore, the available information on the quantity of phthalates for a specific product category will be used as the quantity for all life stages of the product.

Import of phthalates in Denmark

There is no production of phthalates in Denmark.

The import of phthalates has been estimated to be 11,500-14,000 tons/year. Of this imported tonnage, 6,500 tons is as pure phthalates, 3,000-5,000 tons phthalates in PVC compounds, and 2,000-2,500 tons phthalates in semi-manufactured products. The import of phthalates in products and also the re-export of phthalates via products manufactured in Denmark are unknown (Hoffmann, 1996).
The total amount of pure phthalates that was imported in 2001 (Statistic Denmark, 2002) is lower than in 1992 (Hoffmann, 1996) (see Appendix 2). In Statistic Denmark, the groups of substances are not identical in 1992 and 2002 and therefore, they are not easy comparable. However, the overall decrease in import covers a decrease in the imported amount of DBP to 20% of the amount imported in 1992 and a decrease in the imported amount of DEHP to 70% of the amount imported in 1992.
In 2001, the 1992-group “di-isononyl- and di-isodecyl orthophthalates” in the Danish External Trade Statistic does not exist. Di-isononyl- and di-isodecyl phthalates are now included in the group of “other phthalates than DBP, DEHP, dinonyl- and didecyl phthalates” (Told og Skat, personal communication, 2002). The import of the group “other phthalates than DBP, DEHP, dinonyl- and didecyl phthalates” has increased, but not to the level of the import in 1992 of the group “di-isononyl- and di-isodecyl orthophthalates”. In IUCLID (2000), the term “dinonyl phthalate” is mentioned for one of the DINPs (CAS nr. 28553-12-0). Didecyl phthalate does not exist in IUCLID. Therefore, it is likely that the data on import/export of the group “dinonyl- and didecyl orthophthalates” includes a part of the imported/exported di-isononyl phthalates. But still the increase in the import of “dinonyl- and didecyl orthophthalates” together with the increase in the import of “other phthalates than DBP, DEHP, dinonyl- and didecyl phthalates” is less than what was imported of the group “di-isononyl- and di-isodecyl phthalates” in 1992.

The import of PVC compounds in 2001 was 15,636 tons (Appendix 2). Assuming that the average phthalate content in a PVC compound is 30% (Hoffmann, 1996), the import of phthalates in PVC compounds is around 4,500 ton. This is similar to the import of phthalates in PVC compounds in 1992 (3,000-5,000 tons).

From the External Trade Statistic, the import of semi-manufactured phthalates and phthalates imported in products can only be estimated with large uncertainties since only the total import of a product group as e.g., cables is available and not the amount of e.g., soft PVC in the cables or the amount of phthalates in the soft PVC. In the present assessment, these estimations will not be made.

Based on the registration of chemical products in PROBAS, the amounts of phthalates in products on the Danish market in 1994 and 2001 are listed in Table 2.9 (Hofmann, 1996; Flyvholm, 2001). The total amount of DBP, BBP and DEHP in registered chemical products is higher in 1994 compared with 2001, whereas the amount of DIDP and DINP in registered products is higher in 2001 compared with 1994. These figures does not necessarily represent the actual trend, but could be explained by different ways of generating data from PROBAS in 1994 and 2001.
For 2001, the total number of products registered with phthalates and the number of products where the yearly amount of phthalate in the product has been registered is also included in Table 2.9. As can be seen, information on the amount is only given for about half of the products (from 42% (DINP) to 73% (DIDP) of the products).

Table 2.9. Amounts of phthalates in chemical products registered in the Danish Product Register.

Compound	Ton/year		No. of products (with data on amount)	Active products[3]/ Outdated products[4] 2001[2]	
	1994[1]	2001[2]	2001[2]	based on no. of products with data	based on amount of phthalate
DBP	3,200	2,031	904 (467)	1.4	3.5
BBP	730	364	511 (239)	3.3	0.6
DEHP	7,500	3,935	372 (171)	0.8	0.7
DINP	143	366	175 (58)	4.6	6.9
DIDP	76	243	105 (75)	1.9	6.2

[1] Hoffmann (1996)
[2] Flyvholm (2001)
[3] Active products are products registered or updated within the last five years
[4] Outdated products are products registered or updated more than five years ago
(): Number of products where information on the amount of phthalate in the product has been registered

The figures for 2001 are limited to products registered and updated within the last five years (active products). In order to get an idea of the trend of the use of the phthalates, the proportion of “active” products and “outdated” products (products registered or updated more than five years ago) is given (Table 2.9) (Flyvholm, 2001). The proportion is calculated based on the number of products with data on the yearly amount of the product.

The presently available information about the total use of phthalates in Denmark is fragmentary and especially the PROBAS data has to be used with the above-mentioned reservations. According to the latest inventory on the use of phthalates in Denmark from the Danish

Environmental Protection Agency the amount of phthalates used in Denmark is decreasing (MST, 2003). The overall decrease has been estimated to be approximately 15% since 1995 especially the use of DEHP, DBP and BBP seems to be decreasing. Therefore, consumption data from Hoffman (1996) for these substances is likely to represent a worst-case situation in year 2002. In EU, the consumption of DEHP has been relatively constant for the last 20 years (DEHP, 2001) and for DBP, there is a decreasing tendency in the production (DBP, 2001).
For DINP and DIDP, the increasing tendency of consumption on the European market (DINP, 2001; DIDP, 2001) is also reflected in PROBAS and mentioned by MST ("003), but not in the import of pure DINP and DIDP. This could be due to the fact that the products containing DINP and DIDP are not processed in Denmark, but instead DINP and DIDP are imported into Denmark in PVC compounds, semi-manufactured or imported products. The data for DINP and DIDP from Hofmann (1996) will be used in the present exposure assessment keeping in mind that they may be underestimates of the present use of DINP and DIDP.

Use patterns in Denmark

In general, 90% of the total amount of phthalates imported into Denmark is used as plasticisers in PVC products. The rest (10%) is used in other products than PVC. The proportion used in PVC products of the different phthalates is not the same, since DEHP is mainly used in PVC, whereas DBP more frequently has other applications than as plasticiser in PVC, e.g., in adhesives, paints and printing inks.

In Table 2.10, information from Denmark on the product categories containing phthalates is summarised together with the fractions taken from the EU-RARs. Despite of the mentioned limitations of the PROBAS-data, they are also presented in Table 2.10 in order to compare data, but as mentioned, these data will not be used as quantitative input data to EUSES (except for DBP in paints).
The information from Hofmann (1996) and MST (1999; 2000) are a combination of data obtained from the Danish Product Register, Statistic Denmark, and information from the trade. The amounts of phthalates used in non-PVC applications have been reduced mainly due to substitution of phthalates with other plasticisers (MST, 2000). On the other hand, in 1999 the use of phthalates in PVC seemed to be more constant and maybe even increasing (MST, 1999).

In a not yet published report from the Danish Environmental Protection Agency (MST, personal communication), the use of phthalates in PVC in 2000 and 2001 has been estimated to be approximately 10,000 tons/year. In PROBAS, the phthalates of interest were most frequently registered in printing inks, fillers and paint, lacquers etc. (Table 2.10) (Flyvholm, 2001). For DBP, DINP and DIDP, the maximum amount was also registered in one or more of these product categories, whereas for DEHP, the maximum amount was registered as plasticiser (not presented in Table 2.10). As mentioned, it is mainly chemical products that have been registered in PROBAS; PVC products are not included. That is the reason why DBP, DINP and DIDP are not registered in highest amount as plasticiser in polymers.

Based on the data in Table 2.10, EUSES input data on regional tonnage of phthalates have been generated and they are presented in Table 2.11.
Information from several different sources is not necessarily consistent and it is therefore necessary to make some priorities. First, any available information from Hofmann (1996) on the

amount of an individual phthalate applied for a specific use pattern will be used keeping in mind, as discussed above, that these data do not necessarily describe the present situation (yearly amount). If there is a range, the highest value will be chosen to ensure that EUSES does not underestimate the emissions. If there is no quantitative information from Hofmann (1996), the percentage of tonnage used for the applications will be taken from the EU-RAR of the particular substance. In one case (DBP in paint, lacquers etc.), quantitative data from PROBAS (Flyvholm, 2001) will be used since there is no information on this use-pattern in the EU-RAR of DBP (2001).

Table 2.10. The amount (tons/year) of substance in specific applications in Denmark estimated by Hofmann (1996), the amount (tons/year) registered in chemical products in PROBAS (Flyvholm, 2001), or the percentage (%) of substance in specific applications from the EU-RARs.

Compound	Polymer plasticisers		Adhesive[1]		Fillers (sealants, grouting agents)[1]		Adhesive & sealants[1]	Printing inks		Paints, lacquers, etc.	
	Hofmann (1996)[3]	EU-RAR[4]	Hofmann (1996)[3]	Probas[2]	Hofmann (1996)[3]	Probas[2]	EU-RAR[4]	Probas[2]	EU-RAR[4]	Probas[2]	EU-RAR[4]
DBP	-	76%	100-140	70	260	214.5	14%	857	7%	447	-
DEHP	6,000 - 9,540	97 %		3.6		5.6	2.35%	0.4	0.35%	19	0.3%
DINP	690 - 1,350	97.5%		248.4		7.3	0.86%	1.9	0.86%	0.1	0.86%
DIDP	400	98.7%		3.2	35	129	0.135%	-	0.135%	63	0.27%
Total (1992)	ca. 11,000[3]		160-220[3]		ca.400[3]			270[3]		225[3]	
Total - 1995[5]; 2000[6]	ca. 11.000[5]		220[6]		100[6]			50[6]		70[6]	

[1] In Hofmann (1996), adhesives and fillers are treated separately whereas in the EU-RARs, they are together in one category (adhesives and sealants). From PROBAS, adhesives consist of adhesives, paste, glues and hardeners. Fillers consist of grouting agents and sealants.
[2] Flyvholm (2001)
[3] Hoffmann (1996)
[4] Proportions from EU-RARs of each phthalate.
[5] Danish Plastic Industry (1996)
[6] MST (2000)

For several of the product categories, the sum of all phthalates used in the specific product category (row named “Sum” in Table 2.11) is similar to the reported values from Hofmann (1996) (row named “Total” in Table 2.11). The sum of one phthalate from all product categories (columns named “Sum”) is higher than the information from PROBAS (Flyvholm, 2001) (columns named “Registered in PROBAS”). That is also what was expected since not all relevant product categories are included in the PROBAS registration and only about half of the products are registered with information on volume used (see previously).

The estimated total sum of phthalates (14,877 tons/year) is very close to the highest value given for the total import of phthalates (11,500-14,000 tons/year) (Hoffmann, 1996). However, there are other important phthalates, which are not included, e.g., BBP.
The average content of phthalates in the different products as given in the EU-RARs (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001) is also listed in Table 2.11. For each phthalate and each type of product, there is a range of content, i.e., the content of DEHP and DIDP in PVC is usually between 20-35% and 25-50%, respectively (Hoffmann, 1996).

Table 2.11. The amounts and product categories of phthalates in Denmark. EUSES input data.

Compound	Tons/year					
	Polymer plasticisers (mainly PVC)	Adhesive & sealants	Printing inks	Lacquers, paints	Sum	Registered in PROBAS[3]
DBP	2,171[2]	400[1]	200[2]	447[3]	3,218	2,031
DEHP	9,540[1]	230[2]	34[2]	30[2]	9,835	3,935
DINP	1,350[1]	12[2]	12[2]	12[2]	1,385	366
DIDP	400[1]	36[2]	-	2[2]	439	243
Sum	13,700	678	268	491	14,877	
Total[1]	Ca. 11,000[1]	560-620[1]	270[1]	225[1]		
Average phthalate content in product[1]	30%	10%	5%	5%		

[1] Hoffmann (1996)
[2] Estimated based on the fractions from the EU-RAR of DEHP (2001), the EU-RAR of DBP (2001), the EU-RAR of DINP (2001) and the EU-RAR of DIDP (2001).
[3] Flyvholm (2001)

Below, the estimations of tonnage for each phthalate (Table 2.11) are explained in details.

DBP:
Information on the amount used as adhesives and fillers in Denmark exists. In Hoffmann (1996), DBP is not mentioned as a plasticiser in polymers, but in EU, 76% of the total use of DBP is as plasticiser in polymers. It seems unlikely that DBP should only be used as a plasticiser in polymers in the rest of Europe, but not in Denmark. There is also a discrepancy between the total amount registered in the Danish Product Register (2,031 tons/year) and the amounts that have been reported in use (140+260 tons/year). The total amount of DBP will be estimated based on the fractions from the EU-RAR of DBP (2001) and the information from Hofmann (1996) on the amount of DBP in adhesives, sealants etc. The total amount of DBP will therefore be 400 tons/year divided by 0.14 = 2,857 tons/year. Of this total amount, 76% are used as plasticiser (2,171 tons/year) and the amount used for printing inks is 0.07 multiplied by 2,857 tons/year = 200 tons/year. DBP is the phthalate that is registered in the highest number of paints, lacquers etc. in Denmark (Flyvholm, 2001) and it is also mentioned as the dominating phthalate in paints and lacquers in the mass-balance analysis (Hoffmann, 1996). In the EU-RAR of DBP (2001), the

use of DBP in paints and lacquers is not included and thus, no EU data are available. Therefore, the amount of DBP registered in PROBAS (Flyvholm, 2001) will be used instead. Information on amounts used is only given for 122 of the 174 registered paints, lacquers etc.

DEHP:
The most important use pattern for DEHP is as a polymer plasticiser and this is also the only use category of DEHP where data are available for Denmark.
In EU, the amount of DEHP in polymers is 97% of the total amount and the remaining 3% are used in non-polymer application, mainly in adhesives. The fractions of DEHP used for the non-polymer applications in Denmark are assumed to be equal to the fractions used in EU.
To be certain that the total consumption is included, the highest reported amount of DEHP used in polymers will be chosen.
The total amount of DEHP from the different use categories exceeds the amount of DEHP registered in the Danish Product Register. Also, the total amount of phthalates used as polymer plasticisers exceeds the reported amount (Hoffmann, 1996). The amount used in e.g., adhesives is (9540/0.97)*0.0235 = 230 tons/year.

DINP:
As for DEHP, the only specific use pattern of DINP, where Danish data are available, is as a polymer plasticiser. In EU, 97.5% of the total amount of DINP is used in polymer applications, whereas the remaining 2.5% is used for other applications. The fraction of DINP used in non-polymer applications in Denmark is assumed to be equal to the fraction used in EU. The amount of DINP in polymers is 1350 tons/year and therefore (1350/0.975)*0.025 = 36 tons/year are used in adhesives and printing inks and paints. The amount is equally distributed among these three groups.

DIDP:
DIDP is mainly used as a plasticiser in PVC. In Denmark, the remaining amount of DIDP is used in grouting agents, and a small amount also in paints and adhesives. The amount of DIDP used in non-polymer applications in EU is approximately 1% of the total. It is assumed that the amount is equally distributed between the two remaining use patterns. The content of DIDP in PVC is usually between 25-50% (EU).

2.4.3 The specific use patterns

EUSES distinguishes between three categories besides the life cycle step, i.e., the industrial category, the main category, and the use category. The main category gives an indication of how big the emission factor is. The industrial category describes “where” the chemical is used and the use category covers the function of the substances.

In the EU-RARs of the four phthalates DBP, DEHP, DINP and DIDP, the use patterns are categorised quite differently, even though some of the use patterns of the four phthalates are very similar. In the present project, the categories for each specific use have been made more uniform among substances. For each use, e.g. as adhesives, a decision has been made of which categories (including industrial category, use category and main category) that were suitable to describe that specific use. These specific categories have then been applied to all the phthalates that are used for the same purpose (Table 2.12).

Table 2.12. Specific use patterns of the different phthalates.

Use pattern	Industry Category	Use category	Life cycle step	Main cat.	Substances	Spatial scale
Polymer plasticiser	Polymers industry (11)	Softener (47)	Prod. Proc.	III II	All	Cont Cont/Reg
Paints, lacquers etc.	Paints, lacquers and varnishes industry (14)	Softener (47)	Form. Proc. Priv. use	III III	All	Cont/Reg Cont/Reg Cont/Reg
Adhesives (+ sealants)	Engineering Industry (16)	Softener (47)	Form. Proc.	III II	All	Cont/Reg Cont/Reg
Printing Inks	Pulp, paper and board industry (12)	Solvent (48)	Form. Proc.	III III	DEHP, DBP and DINP	Cont/Reg Cont/Reg
Grouting Agents	Others (15)	Others (55)	Form. Proc.	III III	DBP	Cont. Cont.
Printing inks, textile	Textile processing industry (13)	Softener (47)	form. proc.	III III	DIDP	Cont. Cont.
Ceramics	Others (15)	Softener (47)	Form. Proc.	III III	DEHP, DIDP DIDP	Cont. Cont.

The emission factors, fractions of the main local source, and the days of emission for each category used in the present assessment in EUSES can be seen in Appendix 3. When a use pattern is only relevant on one spatial scale, the emission from the other spatial scale is zero. The only industrial category, where local emission factors for the life cycle step "private-use" exist, is the paint, lacquers and varnishing industry. This is included in the exposure scenario. There are no industrial categories where the life cycle step "waste" are included. Therefore, EUSES does not estimate the emission during use (except for paints) and waste (or recycling) of end products. If emissions from these life cycle steps have to be taken into account, there is a need for specific data on the emission.
In the next section, the emission from the use of end products will be estimated. The emission during waste will not be included in the estimation due to the lack of data on the emission from this life cycle step.

2.4.4 Emission during use of end products

Phthalates are not only emitted during manufacturing of phthalate containing products, but also during use of the end products. It is evident for some soft PVC products that during time they loose their softness. This is due to the migration of the plasticisers – the phthalates. They have been emitted to either air or water in contact with the product.
There are two approaches for estimation of the emission to air and water during use of end products. In the first approach, a general emission factor is applied to an unspecified total volume of the phthalate. The second approach is more detailed. The emission from a product is more dependent on the surface area of the product than on the content of phthalates in the product. To account for this, a more detailed approach can be applied where the evaporation or

leaching rate/m^2 of the total area of specific articles is considered and the total emission is summarised. The more detailed second approach should be preferred. However, in the present assessment, data on the different uses is not always detailed enough to allow an estimation of the emission from each product group based on surface areas and therefore, the first approach, where more general emission factors are used, will also be applied.

The emission during use of end products has been included in the EU-RARs of the phthalates (DEHP, 2001; DINP, 2001; DIDP, 2001; DBP, 2001). The methods to estimate the emission of DEHP, DINP and DIDP are quite detailed and also quite similar for the three substances, whereas for DBP, only very general fractions of emission are used. In the present assessment, a more consistent estimation of the release of the individual phthalates during use in Denmark will be applied for all the phthalates, based on the EU-RARs of DEHP, DINP and DIDP. Emission rates to air and water per surface area will be applied for paints, lacquers, adhesives, sealants and printing inks. For PVC, a different approach will be applied due to lack of data.

Paints, lacquers etc. and adhesives, sealants etc.

The emission rate from paints, lacquers etc. and adhesives, sealants etc. to air used in both the EU-RAR of DEHP, DINP and DIDP is 9.5 mg/m^2/year as measured for DEHP (Environ Corporation, 1988 – cited in DEHP, 2001). In the present assessment, this value will also be used for DBP.

The emission rate of DINP and DIDP to water is estimated to be 1.05 g/m^2/year based on an outdoor experiment of leaching of a phthalate mix of C_8-C_{10} and iso-C_{10} from roofing material (Pastuska et al., 1988; 1990 - cited in DINP, 2001). Based on the same study and taking into account the shorter chain length of DEHP compared to DINP and DIDP, which is expected to increase the migration, the emission rate of DEHP is estimated to be 1.98 g/m^2/year in the EU-RAR of DEHP (2001). This value will also be used to estimate the emission of DBP to water.

Since these values are emission rates per surface area, it is necessary to estimate the total surface area for the different product groups. The total surface area can be estimated by the use of a surface correction factor (SCF). The SCF is defined as the relative change in emitting surface area compared to the roofing material that was used in the above-mentioned experiment of leaching from roofing material, e.g., if a material is expected to be 10 times thinner than the roofing material (1.5 mm), the SCF will be 10 and therefore, the surface area per ton will be ten times higher than the surface area per ton of the roofing material (532 m^2/ton). The SCFs for the different products are given in Appendix 4.A.

Furthermore, the emissions from end products are, in contrast to the emissions from industrial uses, often extended in time. For products with service lives longer than one year, the estimation of the annual emission must take into account the fact that products produced in year 1 also contribute to the emission in year 2 etc. By multiplying the surface area of the product group produced per year with the technical lifetime of the product, the products produced earlier but still in use will be included in the total surface area. It is assumed that the release rate is constant during the whole lifetime, that the present use pattern is not changed, and that the amount of phthalate used in a product group has reached steady state. For technical lifetimes of different product groups, see Appendix 4.A.

It will be assumed, as in the EU-RARs of DINP (2001), DIDP (200) and DEHP (2001), that 50% of the paints etc. and adhesives etc. will be used outdoor where the primary recipients of the leached amount is surface water (50%) and soil (50%), and 50% are used indoor, where the leached amount is directed 100% to wastewater. Emission to air will take place both during indoor and outdoor end-use. The estimated emitted amounts of individual phthalates to each compartment from the end-use of paints and adhesives are given in Appendix 4.B.

Printing inks

It will be assumed that only indoor end-use of printing inks will occur and therefore, the emission will be directed to air and to wastewater during the recycling of paper.
DEHP is the only phthalate, for which it is assumed in the EU-RAR that emission to air from printing inks occurs. In the present assessment, emission to air from printing inks will be assumed for all the phthalates and the emission rate to air will be the same (9.5 mg/m^2/year) as for paints, adhesives etc. The SCF and lifetime of printing inks is given in Appendix 4.A.

According to the TGD (1996), between 6% and 90% of ink can be leached to wastewater during paper recycling. Because of the hydrophobic nature of the phthalates, the lower value (6%) seems most reliable. Furthermore, during primary treatment of the wastewater, a removal rate of 90% of the phthalates is assumed. The recycling fraction of paper in Denmark is around 50% (Tønning, 2002).

Due to the tendency of the phthalates to remain in fibres and thereby to remain in the paper during recycling, the amount of phthalates present in the paper is expected to be higher than the amount added with inks. Therefore, a correction factor of 2 will be used in the estimations of emission to both air and wastewater. The estimated emitted amounts of individual phthalates to each compartment from the end-use of printing inks and recycling of paper are given in the Appendix 4.B.

PVC

The PVC product group is very diverse. Therefore, it is not realistic to use one Surface Correction Factor (SCF) for the whole product group. A total emitting surface area for the entire group of PVC products can only be estimated if detailed data are available on the amounts used for the different PVC products such as e.g., cables, floorings, car undercoating etc. combined with either specific emission rates or SCFs for each product group. The more detailed calculations have been made in the EU-RARs of DEHP (2001), DINP (2001) and DIDP (2001), but it is not possible to do in the present assessment due to lack of data.

Instead of the detailed estimations of the emission from several different PVC product groups made in the EU-RARs of DEHP (2001), DINP (2001) and DIDP (2001), the total emission from all PVC product groups has been used in the present assessment to estimate a general fraction of emission from end-use of PVC products (Appendix 4.C). If it is assumed in the present assessment, that the fraction of phthalates used in the different PVC product groups in Denmark is similar to the fractions in EU, and that the distribution of indoor end-use and outdoor end-use, and the direction of the emission indoor and outdoor (indoor: 100% wastewater; outdoor: 50%

soil, 50% surface water) are the same as in EU, these fractions can be used to calculate the amount emitted from PVC products in Denmark.

The fractions of emission obtained from the EU-RARs of the phthalates differ only slightly and average values for the fractions will be used to predict the amount emitted to air, wastewater, surface water and soil for DEHP, DINP, DIDP, and also for DBP (see Appendix 4.C). The estimated emitted amounts from the end-use of PVC products is also presented in Appendix 4.C.

Total

In Table 2.13, the total emission to air, water and soil from the use of products based on the above-mentioned assumptions is given. The values will be used as input to EUSES in private use emission scenarios of the individual phthalates. See Appendix 4 for more details on the estimated values.

Table 2.13. The emission of phthalates to air, soil and water from the use of end products.

Substance	Tons/year			
	Wastewater	Surface water	Soil	Air
DEHP	30.7	18.6	18.6	4.2
DINP	4.4	2.6	2.6	0.6
DIDP	1.4	0.8	0.8	0.2
DBP	74.0	36.9	36.9	3.3
Total	110.5	58.9	58.9	8.3
Total[1]	3-74		-	0.4 - 8

[1] Hoffmann (1996)

Hoffmann (1996) has also estimated the emission of phthalates during use of end products. The estimates are mainly based on general emission factors (see Appendix 4.D) and the uncertainty of the estimations is reflected in the broad ranges of the emission intervals.
The emission from PVC products to the air compartment has been estimated to be 0.4-5.5 tons/year. The emission to air from lacquers is low (0.01-0.05 tons/year) and the emission from the use of cosmetics (mainly DEP) is also low (<2 tons/year). This gives a total emission to air of 0.4-8 tons/year, which is similar to the value estimated in the present assessment based on the assumptions from the EU-RARs (DEHP, 2001; DINP, 2001; DIDP, 2001).

The emission to air during use of end products, and also during waste disposal, is considered to be negligible compared to the emission during the manufacturing process of PVC, while the emissions to the aquatic compartment during manufacturing processes are minimal compared to the emissions during use of plasticiser containing products (Hoffmann, 1996). Water is only rarely involved in the manufacturing processes, whereas the products are washed during use (cars, floors, textiles etc.). According to Hoffmann (1996), the emission of phthalates to water by the washing of PVC products is estimated to be between 0.1-16 tons/year. For washing of printed

textiles and lacquered floors, 1.3-13 tons/year and 1.5-15 tons/year has been estimated as the emission to the water, respectively. Cleaning of machines and other products containing adhesives results in the release of 1-40 tons/year and the emission from cosmetics is estimated to <1.3 tons/year (mainly diethyl phthalate (DEP)). This gives a total emission to water of 3-74 tons/year. The emission reported by Hoffmann (1996) is not separated into the amount directed to wastewater and the amount directed to surface water as in the present assessment. The range of emission to water is lower than the emission to wastewater plus surface water (95 tons/year) estimated on basis of the EU-RARs.

According to Hofmann (1996), emission to soil is not assumed to occur.

3 Estimated environmental concentrations and indirect exposure

On the basis of the specific Danish default values, the specific values for degradation and distribution of each phthalate, the amounts of phthalates used in EU and in Denmark, and the specific use patterns of each substance in EU and in Denmark, predicted environmental concentrations (PECs) and indirect human exposure via the environment have been estimated by the use of EUSES. The estimated values will be presented in this section. In section 4, measured concentrations from the Danish environment will be presented and in section 5, the measured and the estimated concentrations will be compared.

As already mentioned, the EUSES program estimates the exposure on different spatial scales: the continental (EU), the regional (in this case DK), and the local. Concentrations on the continental scale are used as boundary conditions for the region and therefore, they will not be mentioned in the present report. The regional assessment can be interpreted as the potential average exposure due to continuous, diffuse emissions. The local assessment can give indications of the exposure of a population or location at higher risk than the average. The intention was to make a relatively conservative scenario (EC, 1996).

3.1 The regional scale

In Table 3.1, the regional PECs for the different environmental compartments estimated by EUSES are presented.

DEHP and DBP are the substances that are predicted to occur in highest concentrations. This is not surprising in light of the higher tonnage of these two substances compared with DINP and DIDP. The concentration of DBP is highest in the different water compartments (surface water and agricultural soil pore water (groundwater)). The higher concentration of DBP in the water compartments is mainly due to the higher water solubility and the 100 times lower K_{oc} of DBP compared with the other substances (Table 2.6).

The higher concentration of DEHP, DINP and DIDP in natural soil compared with agricultural soil may seem odd since natural soil represents soil that only receives atmospheric deposition, whereas agricultural soil, in addition to the atmospheric deposition, also receives sludge from the

STPs. One of the reasons why the estimated concentration in natural soil is higher compared with agricultural soil is that the total emission to a certain compartment is divided with the volume of the compartment. The volume of e.g., the agricultural soil compartment is the product of the area of agricultural soil and the mixing depth. The mixing depth of the agricultural soil is higher (20 cm) than that of the natural and industrial soils (5 cm) and the area fraction of agricultural soil in Denmark (a parameter in the specific Danish profile Table 2.1) is high compared with the area fraction of natural and industrial soils. Because the total volume of the agricultural soil compartment in Denmark is a lot higher than the volume of the natural soil compartment, the emission to the agricultural soil compartment via sludge has to be high for the concentration of a substance in the agricultural soil to exceed the concentration in the natural soil. This is only the case for DBP. For DEHP, DIDP and DINP, the emission via sludge is not high enough to compensate for the increased volume of the agricultural soil compartment and therefore, the concentration in the agricultural soil compartment does not exceed the concentration in the natural soil.

Table 3.1. Regional predicted environmental concentrations (PECs).

Regional PEC	DEHP	DBP	DINP	DIDP
in surface water (total) (μg/m^3)	296	1,160	19.6	5.86
in surface water (dissolved) (μg/m^3)	228	1,150	12.9	3.86
in air (ng/m^3)	5.26	1.50	0.82	1.56
in agricultural soil (μg/kg dry weight (dw))	7.96	4.38	1.11	1.05
in pore water of agricultural soils (μg/m^3)	2.41	34.5	0.19	0.18
in natural soil (μg/kg dw)	10.8	0.19	1.39	2.78
in industrial soil (μg/kg dw)	75.7	112	10.4	5.85
in sediment (μg/kg dw)	3,480	634	366	102

The general physico-chemical properties can also be used to quantify the behaviour in the environment in a visual model, the EQC-model (EQC, 1997). It is assumed that thermodynamic equilibrium is achieved. In this model, the phthalates mainly tend to distribute to the soil (94.8-97.8%) and around 2% distribute to the sediment. For DBP and BBP, some will also be distributed to the water compartment (2.8% and 1.6%, respectively). See Appendix 5 for further details.

The fate of the substances in the STP is given in Table 3.2 as the estimated fraction of emission directed to air, water, sludge, and the estimated fraction that has been degraded. According to the estimations, DBP is mainly degraded in the STP whereas DEHP, DINP and DIDP are mainly distributed to the sludge. The fate of DEHP, DINP and DIDP in the STP is very similar.

The estimated concentration in the different groups of food is presented in Table 3.3.
In the drinking water, as in the surface and groundwater, DBP is present in highest concentrations compared with the other phthalates. But the differences in water concentrations are not reflected in the concentration in fish, where the concentration of DEHP is around 100 times higher than that of DBP. The bioconcentration factors (BCFs) for fish for the two

substances, which have been used in the EU-RARs, are very different: For DEHP, the BCF is 840 whereas for DBP, it is only 1.8 (Table 2.6). Both values are based on experiments. The BCF for DEHP was chosen from the higher end of a range of values to represent a realistic worst-case scenario, including also the bioconcentration of the major metabolite (MEHP), whereas the BCF for DBP was the lowest observed value and it only includes the parent compound and not any metabolites (DEHP, 2001; DBP, 2001).

Table 3.2. The estimated fate of phthalates in the STP.

Fraction of emission	DEHP	DBP	DINP	DIDP
directed to air	0.002	0.001	0.007	0.02
directed to water	0.08	0.11	0.08	0.10
directed to sludge	0.77	0.29	0.81	0.84
degraded	0.15	0.60	0.10	0.04

In meat and milk, the concentrations of DIDP, DINP and DBP are more than ten times higher than for DEHP. This is because the bioaccumulation factor (BAF) for DEHP has been set 100 times lower than what EUSES estimates on the basis of log K_{ow} for the other substances. So, even though the cattle get higher doses of DEHP through the air, soil, grass and water, the use of a low BAF results in a lower estimated concentration of DEHP in the meat and the milk, especially when compared to DINP and DIDP. No justification is given for using the selected BAF for meat and milk in the EU-RAR of DEHP (2001). If the BAF instead is estimated by EUSES, as for the other phthalates, the concentration of DEHP in meat and milk increases about 400 times each and the total daily intake for adults doubles up (e.g., regional: $2.55*10^{-4}$ mg/kg bw/day).

In general, the children between 1-6 years old have the highest estimated daily intake of phthalates and the adults have the lowest. The group of children between 7-14 years has a total daily intake in between those two groups. The total daily intake for adults and children are highest for DEHP and lowest for DIDP (20 times lower). The total daily intake of DBP and DINP are in between the two other phthalates.

3.2 The local scale

The determination of the releases from point sources on a local scale through local emission scenarios, as for the continental and regional emission, depends on use category and life cycle step. But what is different from the predictions on the regional and continental scale is that local environmental concentrations are estimated separately for every compartment and each relevant life cycle step. In the present report, the estimated minimum and maximum local concentrations are presented in Table 3.4. For the detailed data of the local PECs for each relevant life cycle step, see Appendix 6.

Table 3.3. Indirect human exposure via the regional environment.

Concentration in	DEHP	DBP	DINP	DIDP
fish (mg/kg wet weight (ww))	0.191	$2.07*10^{-3}$	0.0108	$3.24*10^{-3}$
root tissue of plant (mg/kg ww)	$6.69*10^{-3}$	0.0108	$9.49*10^{-4}$	$9.03*10^{-4}$
leaves of plant (mg/kg ww)	$1.67*10^{-3}$	$3.09*10^{-3}$	$8.23*10^{-5}$	$5.25*10^{-5}$
drinking water (µg/m^3)	56.9	574	3.23	0.48
meat (mg/kg ww)	$2.40*10^{-5}$	$2.26*10^{-4}$	$5.00*10^{-4}$	$3.33*10^{-4}$
milk (mg/kg ww)	$7.20*10^{-6}$	$7.15*10^{-5}$	$1.58*10^{-4}$	$1.05*10^{-4}$
Total daily intake[1], adult (mg/kg bw/d)	$1.9*10^{-4}$	$1.0*10^{-4}$	$1.6*10^{-5}$	$9.2*10^{-6}$
Total daily intake[1], child 1-6 years (mg/kg bw/d)	$8.7*10^{-4}$	$4.9*10^{-4}$	$8.0*10^{-5}$	$4.6*10^{-5}$
Total daily intake[1], child 7-14 years (mg/kg bw/d)	$3.8*10^{-4}$	$2.1*10^{-4}$	$3.5*10^{-5}$	$2.0*10^{-5}$

[1] The total daily intake also include inhalation of air.

The overall pattern of the distribution of substances locally is the same as regionally with DEHP and DBP being the phthalates present in highest concentrations.
For all substances and all compartments, the maximum local environmental concentration is much higher than the regional. This is also the case for the estimates concerning local indirect exposure of humans (Table 3.5). For the detailed data of the local indirect exposure for each relevant life cycle step, see Appendix 6.

Table 3.4. Range of local predicted environmental concentrations (local PECs).

PEC	Comment	DEHP	DBP	DINP	DIDP
Air (ng/m^3)	annual average	5.3-3,640	1.5-2,480	0.82-772	1.6-230
Deposition (µg/m^2/day)	annual average	$9.9*10^{-10}$-98.9	$9.9*10^{-9}$-4.4	$6.0*10^{-11}$-17.8	$4.3*10^{-4}$-5.6
Surface water (µg/m^3)	annual average	228-6,90	1,150-21,400	12.9-317	5.7-1,100
Sediment (µg/kg dry weight (dw[1]))	during emission	2,120-77,800	414-9,270	209-18,700	129-22,200
Agricultural soil (µg/kg dw)	average, 180 d	10.8-4,510	0.19-2,760	1.4-709	5.1-784
Natural soil (µg/kg dw)	average, 180 d	10.8-3,030	0.019-1,960	1.4-506	4.3-521
Groundwater (µg/m^3)	under agri. soil	3.26-1,370	1.5-21,700	0.24-124	0.90-137
Sewage sludge (mg/kg dw)		$1.6*10^{-5}$- 3,120	$2.3*10^{-3}$-2,070	$5.0*10^{-7}$-529	1.6-546
Influent (mg/l)		$6.65*10^{-9}$ - 1.3	$2.5*10^{-6}$ – 2.3	$2.0*10^{-10}$ – 0.2	$6.2*10^{-4}$ – 0.21
Effluent (mg/l)		$5.4*10^{-10}$ – 0.1	$2.7*10^{-7}$ – 0.25	$1.6*10^{-11}$ – 0.02	$6.2*10^{-5}$ – 0.021

For all four phthalates, the scenarios that leads to the highest local concentration in sludge, soils, sediment and surface water is either the formulation or the processing of adhesives, sealants etc. grouped together in the industrial category “Engineering Industry”. For DEHP, DIDP and DINP,

the highest local concentration in air and the highest local annual deposition is from the scenario of processing polymers, whereas for DBP the highest local concentration in air is estimated for the processing of printing inks.

In the exposure assessment of DINP, DBP and DEHP, the local scenario of the private use of paints, lacquers etc. gives the minimal local PECs in all compartments. The amount of DIDP used in paints etc. is so small that the fraction of the main local source, as given in the emission factor tables for different use categories (B-Tables), is zero. Therefore, emission from this scenario cannot be estimated and the minimal local PECs of DIDP is instead from the processing of paints, lacquers etc.

Since the local emission from the private use of paints, lacquers etc. is very small, the minimal local concentrations in the environment are identical to - or sometimes even lower than - the regional concentrations depending on how the regional background concentrations are estimated. For the air and surface water compartments, the regional PEC is simply added to the local PEC as background concentration (EC, 1996). In agricultural soil, the regional PEC in natural soil is used as a background level of the local agricultural soil (EC, 1996) and if the regional concentration in natural soil is lower than in the agricultural soil, the local PECs will also be lower (see e.g., DBP). It would, however, seem more realistic that the minimal local concentration at least is at a regional level.
Regarding the sediment, the local concentration is estimated on the basis of the local concentration in the surface water and no background concentration is added.

Table 3.5. Range of indirect human exposure via the local environment.

Concentration in	DEHP	DBP	DINP	DIDP
Drinking water (μg/m^3)	56.9-1,720	574-21,700	3.2-124	0.90-139
Fish (mg/kg wet weight (ww))	0.2-5.8	0.0021- 0.04	0.011-0.27	$4.8*10^{-3}$-0.93
Roots (mg/kg ww)	$9.9*10^{-3}$-3.8	$4.7*10^{-4}$-6.8	$1.2*10^{-3}$-0.61	$4.4*10^{-3}$-0.67
Leaves (mg/kg ww)	$1.7*10^{-3}$-1.2	$3.1*10^{-3}$-5.1	$8.2*10^{-5}$-0.078	$5.3*10^{-5}$-$2.4*10^{-3}$
Meat (mg/kg ww)	$2.4*10^{-5}$-0.02	$2.2*10^{-4}$-0.32	$5.1*10^{-4}$-0.43	$4.5*10^{-4}$-0.031
Milk (mg/kg ww)	$7.3*10^{-6}$ -$4.7*10^{-3}$	$7.1*10^{-5}$ –0.10	$1.6*10^{-4}$-0.14	$1.4*10^{-4}$-0.0097
Total daily intake, adult (mg/kg/day)[1]	$1.9*10^{-4}$-0.0195	$6.5*10^{-5}$-0.0602	$1.7*10^{-5}$-$5.1*10^{-3}$	$2.3*10^{-5}$-$2.9*10^{-3}$
Total daily intake, child 1-6 years (mg/kg/day)[1]	$9.1*10^{-4}$-0.10	$3.1*10^{-4}$-0.40	$8.6*10^{-5}$-0.030	$1.1*10^{-4}$-0.015
Total daily intake, child 7-14 years (mg/kg/day)[1]	$3.9*10^{-4}$-0.041	$1.3*10^{-4}$-0.16	$3.7*10^{-5}$-0.013	$5.0*10^{-5}$-$6.7*10^{-3}$

[1] The total daily intake also include inhalation.

As for the predicted environmental concentrations, EUSES estimates the highest local concentrations in different food groups in the formulation or processing scenarios for adhesives etc., or the processing scenario for PVC (DEHP, DINP and DIDP); or processing of printing inks

(DBP). The minimal local concentrations in different food groups are from the scenario “private use of paints etc.” except for DIDP where it is from “processing of paints etc.” (See Appendix 6).

For the estimation of the total daily intake, it is assumed that the entire food basket is sourced from the vicinity of the local point source (EC, 1996). This is less conservative than it might appear since for some substances, very few indirect exposure pathways dominate the total exposure (for DBP it is vegetables, Table 5.1) and it is not unlikely that one or two of the pathways could be almost entirely from the local contaminated site.

Ingestion of soil is not included in the exposure of man via the environment since the average ingestion rate of soil is relatively small (data from the US-EPA (1997): 0.2 g/day). Soil ingestion is only relevant if the soil is heavily contaminated, the substance is highly toxic, and if infants and children can come into contact with the soil; in these cases, a separate soil ingestion scenario would be necessary to evaluate.

3.3 Sensitive parameters

There is a need to supply the EUSES model with different parameters, but which of these parameters are actually those that really make a difference? The identification of the parameters with the largest impact on the model is important and, although it is out of the scope of this project to make a sensitivity analysis of the model, determinant parameters will be discussed in this section. It should be noticed that parameters having high impact on the PECs or total daily intake for one substance do not necessarily have a high impact on the same estimations for another substance. It depends on the physico-chemical properties of the substances (lipophilicity, volatility, biodegradability etc.).

Of the total input parameters in EUSES (around 500 parameters, including parameters of the sewage treatment plant, the regional distribution model, the exposure model and substance specific parameters), nearly 40% showed very little impact on the estimation of the total daily intake of 11 different substances, including DEHP (Schwartz, 2000). Taking all 11 substances together, approximately another 40% of the parameters showed high impact, with substance individual differences. In general, the models contributing to the main exposure pathway for the specific substance also showed the most sensitive parameters. For DEHP, the most sensitive parameters for the overall model estimating the total daily intake were the regional emission to air, the fraction connected to sewage treatment plants, the area of the regional system, the atmospheric mixing height, the volume fraction of water in the sediment, the daily intake of grass by cattle, the body weight, and some plant specific parameters (wet bulk density of plant tissue, correction exponent for differences between plant lipids and octanol) (Schwartz, 2000). None of the basic physical or chemical parameters were among the most sensitive parameters for DEHP. The reason why some of the sensitive parameters are from the plant model section is that in the calculations performed by Schwartz (2000), and also in the present assessment (Table 5.1), one of the important exposure pathways of DEHP for humans is through plants. The daily intake of grass by cattle, the body weight, and the plant specific parameters are all included in the modelling of the indirect exposure, but not in the modelling of PECs in the different environmental compartments.

Even though the sensitivity analysis was made for DEHP, it is not necessarily valid for the present DEHP exposure estimations in EUSES due to the fact that some of the DEHP specific physical and chemical properties used in Schwartz (2000) are different from those used in the present assessment. The vapour pressure was more than 100 times higher and the water solubility around 10 times higher than the values used in the present assessment.

Schwartz et al. (2000) carried out a sensitivity analysis of all parameters in EUSES with focus on the impact on the concentration of a polycyclic musk fragrance (HHCB) in surface water. For a readily biodegradable, lipophilic substance, the parameters with a strong impact on the regional concentration in surface water, beside the emission rate, were the fraction connected to the sewer system, the area of the region, and the area fraction of water. At the local level, besides the emission rate, the fraction connected to the sewer system, the degradation rate, the dilution rate, and the number of inhabitants feeding one sewage treatment plant were also important.

Berding et al. (2002) investigated the sensitivity of regional parameters only in the regional distribution model (SimpleBox). They concluded that the important parameters for prediction of environmental concentrations are the area fraction of water, the area fraction of agricultural soil, and the fraction connected to the sewer system, but other parameters can, in certain circumstances, also influence the predictions considerably. For example did Berding et al. (2002) not consider the area fraction of urban soil as a significant parameter not because it does not influence the predictions, but because the variation in the area fraction between different regions (countries) is low.

Although it is difficult to generalise on the basis of these studies, the fraction connected to the sewage system seems to be important whether it is in the calculation of PECs or total daily intake. Also, the total area of the region and parameters used to estimate the volume of compartments, where the main part of the emission is directed (the area fraction of the compartment, the mixing height etc.), and the emission to that compartment is mentioned several times as being important, both in the calculation of PECs and total daily intake. In the prediction of the total daily intake, the models that contribute to the estimation of the major intake pathway of the specific substance show the most sensitive parameters. Since the total intake of a substance is divided by the body weight to obtain the total daily intake per kilogram bodyweight it is obvious that the body weight has great impact on the total daily intake.

3.3.1 Physical and chemical properties

Even though the basic physical and chemical parameters did not show a high impact on the estimation of the total daily intake of DEHP compared with other parameters (Schwartz, 2000), these parameters often show a large variety of measured and extrapolated values and consequently contribute to the uncertainty of the estimations. In this report, these parameters will only be mentioned with regard to the fact that in some cases, the chosen values are beyond or below the values recommended for use by EUSES.

Water solubility

Water solubility is used in estimating the Henry's Law constant and the air-water partitioning coefficient.

Using DIDP as an example, changing the water solubility from the actual estimated value (0.0002 mg/l) to the recommended minimum value in EUSES (0.001 mg/l) decreases the $K_{air-water}$ around 5 times. This means that less of the emission will be directed to the air and more to the water compartment. Consequently, the PEC in the water and sediment increases, whereas the PEC in air and soil decreases when 0.001 mg/l is used instead of 0.0002 mg/l. At the same time, the daily intake, which combines many of the PECs, increases, from $9.2*10^{-6}$ to 1.8×10^{-5} mg/kg bw/day.

Log K_{ow}

The log K_{ow} is used to estimate the organic carbon-water partition coefficient (K_{oc}) and also the bioconcentration factor (BCF) for fish and the partition coefficient between water and plant tissue. Since, for three of the phthalates (DEHP, DIDP and DINP), these three parameters are set manually, the log K_{ow} is not important, but instead the manually set values are important for the further estimations. Therefore, in this case, it is not a problem that the recommended maximum value is lower than the actual values.

3.3.2 Standard default values

Instead of the specific Danish parameters, the default values for the standard region, as set by EUSES, have been used to estimate PECs for DEHP. There are differences, but they are not dramatically. The concentration in all compartments increases when using the EUSES standard region default values instead of the specific Danish parameters, but not even two times in any of the compartments. The total daily intake doubles up, from $1.9*10^{-4}$ to $4.4*10^{-4}$ mg/kg bw/day. This is in agreement with Fredenslund et al. (1995b) where only minor changes were found when evaluating the compartment model, Simplebox, comprised in EUSES with a Simplebox with the Danish default values. Seven different substances were evaluated. For six of them, no evident or only minor differences were seen. The seventh substance, LAS, showed significant changes due to some of its unusual surface properties. In most cases, if the concentration changed in the compartment, it decreased when applying the specific Danish parameters to the model compared with the standard scenario.

4 Measurements of phthalates in Denmark

4.1 Introduction

The focus of section 4 is the measured concentrations of the phthalates in the Danish environment in order both to compare the concentrations estimated by EUSES with what is actually measured in Denmark and to make a refined estimation of the exposure where measured concentrations are used as environmental concentrations in EUSES instead of the predicted environmental concentrations (PECs). The comparisons and refined estimations will be presented in the next section (section 5).

Unfortunately, there are only few data on the concentrations of DINP in the Danish environment and to our knowledge, no data on the concentrations of DIDP. Therefore, mainly data on DEHP and DBP are presented and used in the further assessments. We have also included some data on BBP.
The data come from several investigations and different environmental compartments. The compartments are soil, marine and freshwater sediment and water, atmosphere (as atmospheric deposition), sludge, and biota (presented as the concentration in certain food items and a daily intake through the food).
The extent of data available varies between compartments. For some compartments, more than one investigation have been carried out and the data therefore cover more than one region, but still the investigations have been restricted to certain areas in Denmark. It is uncertain whether the variations in the concentrations of phthalates within these areas cover the total spatial variation within Denmark. Furthermore, there could be temporal variations. For some compartments (e.g., the water), seasonal measurements have been included, but for other compartments, only one sampling time has been used.
The number of locations in each investigation differs a lot as well as the number of replicates from each location. When information on the sampling procedure, e.g., number of replicates, exists, it is included in the text.

It is a general problem when measuring the concentration of phthalates that there tends to be high background levels in blank samples as there is a risk of contamination from glassware and chemicals and because some of the phthalates are ubiquitous. The high blanks increase the detection limit, making it difficult to measure phthalates, particularly the low concentrations in some environmental compartments. This is also one of the reasons for the variation in the sensitivity between different investigations. There are sensitivity differences between compartments in different investigations, but also differences in the detection limit for the same compartment between investigations. Concentrations measured in one investigation can therefore be below the detection limit in another investigation.

4.2 The environment

4.2.1 Soil

The concentrations of phthalates in the soil compartment have been analysed in several studies. Data from both natural and agricultural soils have been found. To our knowledge, no data from industrial soils are available.

In one investigation (Vikelsøe et al., 1999), the aim was to investigate the effect of different fertilisers and/or cultivation methods, including sludge amendment, on the phthalate concentration in soils. Therefore, the selected soils were quite similar in humus content, texture etc. and with similar atmospheric deposition of phthalates, but included a broad range of fertilisation practices. The investigation was restricted to a limited geographical region near the city Roskilde on the island of Zealand. Soil samples were taken from five different soil depths (0-10 cm; 10-20 cm; 20-30cm; 30-40cm; and 40-50cm). Table 4.1 shows the lowest and highest concentrations measured in the five depths as an average of four replicate samples from each depth. In the more clayish soil, a concentration profile was observed with maximum concentration in a depth of 20-30 cm. In one soil, sampling was repeated two years later and the

maximal concentration was found in a depth of 40-50 cm indicating a downward movement of the substances. Furthermore, there was a tendency for the DEHP concentration profile to follow the clay content profile probably due to the binding of DEHP to clay particles or organic substances in the clay. DEHP seems to be eluted from sand.

Table 4.1. The concentration of phthalates in soil.

Region	Soil	Comments	µg/kg dry weight (dw)			
			DBP	BBP	DEHP	DINP
Roskilde - average(avg.) of 4 replicates (replic,) in 5 depths	natural[1]	preserved, not cultured for 100 years	2-8	0.3	4-27	3-17
	agri[1]	ecological cultivation	1.6-2.7	0.5	14-32	4-34
	agri[1]	manure	1.3	0.1-1	1-18	3-13
	agri[1]	fertiliser	1.1-2.1	0.1	9-20	4-35
	agri[1]	sludge amended, medium (0.9 t/ha/y)	<1.5	0.3	6-18	1-16
	agri[1]	sludge amended, low (0.7 t/ha/y)	1.6-2.3	<0.2	17-23	3-9
	agri[1]	sludge amended, heavily (17 t/ha/y)[5]	280-760	25-41	590-1,700	93-220
	agri[1]	sludge amended, heavily (17 t/ha/y)[6]	230-830	7-51	550-3,400	63-910
	agri[1]	run-off zone from sludge storage	1-39	0.3-29	5-670	1-110
Århus -1 sample of 25 sub-samples	natural [2]	sandy soil	<10	<5	<5	na
	natural [2]	clay soil	<10	<5	<5	na
	agri [2] (n=3)	sandy soil	<10	<5	13 (1)	na
	agri [2] (n=3)	sandy soil, sludge(10-20 t/ha)	<10	<5	14-27	na
	agri [2] (n=2)	clay soil	21 (1)	<5	16 (1)	na
	agri [2] (n=2)	clay soil, sludge (14-45 t/ha)	13 (1)	<5	34-120	na
Askov -avg. of 4 replic.	agri [3]	clay soil	13	<5	54	na
	agri [3]	sandy soil	10	<5	17	na
Jyndevad -avg. of 2 replic.	agri[4]	Sandy soil, artificial fertilisers	45-140	na	83-270	na

[1] Vikelsøe et al. (1999) - detection limits: DBP = 1.5 µg/kg dw; DEHP = 1 µg/kg dw; BBP = 0.2 µg/kg dw; DINP = 2 µg/kg dw.
[2] Boutrup et al. (1998) - detection limit: DBP = 10 µg/kg dw; DEHP = 5µg/kg dw; BBP = 5 µg/kg dw.
[3] Krogh et al. (1996) - detection limit: DBP =10 µg/kg dw; DEHP = 5 µg/kg dw; BBP = 5 µg/kg dw.
[4] Grøn et al. (1999) - detection limit: DBP = 20-40 µg/kg dw; DEHP = 100 µg/kg dw (the soil is used in the investigation of uptake by plants).
[5] Heavily amended with sludge in 25 years, but changed to fertilisers 6 years before sampling.
[6] As [5] but sampled 2 years later.
<value: Below detection limit.
na: Not analysed.
n: Number of locations.
( ): Number of positive findings (only if it differ from the number of locations(n)).

In another investigation (Boutrup et al., 1998), the concentrations of DBP, BBP and DEHP were measured in 5 agricultural soils with different clay/sand content (3 sandy and 2 clay soils) with and without sludge amendment. Two, not cultivated soils (natural soils) with similar soil properties were also included. The soils were located in the Århus region in Jutland. The values given in Table 4.1 are results from analysis of one pooled sample consisting of 25 sub-samples from each soil. DBP and DEHP were only found in some of the locations and therefore, the number of positive findings is given in brackets.

Table 4.1 also presents data from two agricultural soils located near Askov (Krogh et al. (1996) where the results are averages of four replicate samples and from a sandy soil from southern Jutland (Grøn et al., 1999) used in an investigation on the uptake of DEHP by plants. The latter results are from two replicate measurements.

The concentrations of DBP, BBP, DEHP and DINP are in the same order of magnitude for all soils from Roskilde, except from the heavily sludge amended soil where the concentration are 10 to 100 times higher. The results from the other investigations are not as extensive, but the concentration levels are comparable. The last investigation, however, show concentrations as high as the heavily sludge amended soils from Roskilde. The observation by Vikelsøe et al. (1999) that DEHP to a certain extent is more strongly bound to clay than to sand is supported by the other two investigations where higher concentrations of DEHP are found in clay soil.

4.2.2 Sediment

Several studies of the concentrations of DBP, BBP and DEHP in both marine and freshwater sediment have been found. In the following text, they are reported separately.

Marine sediment

One investigation of the concentrations of phthalates in marine sediment in Denmark was carried out in the Little Belt Region (Lillebæltsamarbejdet, 1998). In the investigation, two main categories of sediments were included. One category was coastal water sediment sampled close to outflow from point sources, mouth of streams in bays and in small coves. The other category included sediments from the open sea and from open bays. The upper 5 cm of the sediment was sampled and five sub-samples were taken at each location and mixed prior to the analysis. The concentrations are given in Table 4.2, both as an average of all the samples from each category of sediments and as the minimum and maximum concentrations. DBP and DEHP were found in all samples, whereas BBP was only detected in 4 and 8 samples from open sea and coastal water, respectively. The concentrations of BBP in the other samples were below the detection limit (10 µg/kg dry weight).

Phthalates in the marine sediment around Århus have also been investigated (Boutrup et al., 1998; Århus Amt, 2003). In the first investigation, sediment from 6 different sites were analysed and in the second investigation, the concentrations of DEHP, DBP and BBP were measured in sediment from 10 sites. Some of the sample sites were included in both investigations and they are classified as coastal sediment samples since they were sampled in bays. The extent of exposures of the investigated sample sites ranged from not exposed and diffusely exposed to sites receiving discharge water. In both investigations, the samples were taken in the upper 2 cm

of the sediment. Multiple sub-samples (not the same number for all locations) were pooled prior to analysis.
The concentration of phthalates was also analysed in sediment from Århus harbour (Boutrup et al., 1998), where relatively high concentrations of DBP, BBP and DEHP were found.

Also sediment sampled in Roskilde Fjord and Isefjord (Vikelsøe et al., 2001) has been categorised as coastal water. The sediment samples were taken from the upper 2-5 cm of the sediment. From most locations, replicate samples were measured, but only the average from each location is given in the publication. The minimum and maximum values in Table 4.2 are therefore minimum and maximum averages from each location. The real maximum value actually measured at one specific sampling time was higher and the minimum value lower, but these data were not reported. The highest concentration of all three substances was found at the location closest to the outlet from a wastewater treatment plant. The concentration decreased as the distance to the outlet increased.

Especially in the coastal water sediments there is a broad range of measured concentrations and the concentrations in coastal water sediment from Lillebælt reach higher concentrations than the rest of the locations. The other results are in the same order of magnitude, but the concentration in Roskilde Fjord sediment is more similar to the concentration in open sea sediments than to the other coastal water sediments.

Table 4.2. The concentration of phthalates in marine sediment.

Region	Category of sediment	Value	µg/kg dry weight (dw)		
			DBP	DEHP	BBP
LittleBelt[1]	Open sea and bays (n=22)	avg	123 (22)	117 (22)	18 (4)
		range	42-320	49-450	13-21
	Coastal water (n=44)	avg	229 (44)	1,201 (44)	33 (8)
		range	35-2,400	31-16,000	10-120
Århus[2]	Harbour (n=1)		190	5,400	1,300
Århus[2]	Coastal water (n=6)	avg	na	287 (5)	na
		range		82-490	
Århus[3]	Coastal water (n=10)	avg	na	237 (3)	5 (2)
		range		130-320	5
Roskilde, Isefjord[4]	Coastal water (n=6)	avg	118 (6)	548 (6)	6 (6)
		range	43-143	21-724	3-7

[1] Lillebæltsamarbejdet (1998) - detection limit: DBP = 10 µg/kg dw; DEHP = 10 µg/kg dw; BBP = 10 µg/kg dw.
[2] Boutrup et al. (1998) - detection limit: DBP = 100 µg/kg dw; DEHP = 50 µg/kg dw; BBP = 20 µg/kg dw.
[3] Århus Amt (2003) - detection limit: DBP = 50 µg/kg dw; DEHP = 50 µg/kg dw; BBP = 5 µg/kg dw.
[4] Vikelsøe et al. (2001) - detection limit: DBP = 48 µg/kg dw; DEHP = 43 µg/kg dw; BBP = 10 µg/kg dw.
n: Number of locations.
( ): Number of positive findings.
na: Not analysed.

Fresh water sediment

Three of the studies measuring the concentration of phthalates in the marine sediment also included the concentration of phthalates in fresh water sediments (Boutrup et al., 1998; Århus Amt, 2003; Vikelsøe et al., 2001). The results are summarised in Table 4.3.

In the study by Boutrup et al. (1998), sediments from lakes and streams were investigated. A lake with no direct source of exposure and several lakes receiving wastewater were included. In this study, the data from different locations were pooled. The samples were taken the same way as the marine sediment samples previously described.
As with the marine sediment, the results from Roskilde Fjord (Vikelsøe et al., 2001) are averages of multiple samplings on each location and are therefore, minimum and maximum averages from the locations. Some of the reported minimum averages from Roskilde are below the limit of detection. This could be due to the fact that also the data below detection limit has been included in the averages.

The concentration of the three measured phthalates is lower in the lakes and streams connected to Roskilde Fjord compared with the lakes and streams in Århus Amt. There are no comments on the extent of exposure of the streams and lakes in Roskilde, but a lower extent of exposure could be the explanation of the differences in the concentrations of phthalates. The concentrations of DEHP in streams and lakes in Roskilde are more comparable to those in the lake in Århus Amt with no direct sources.

Table 4.3. The concentration of phthalates in fresh water sediment.

Region	Sediment from	Comments	Value	µg/kg dry weight (dw)		
				DBP	DEHP	BBP
Århus[1]	Lake (n=5)	no direct source	n=1	<100	310	<20
		wastewater outlet	avg.	288 (4)	2,335 (4)	36(2)
			range	130-630	640-2,500	27-44
	Stream (n=5)	wastewater outlet	avg.	150 (2)	2,928 (4)	83 (2)
			range	140-160	75-6,900	35-130
Århus[2]	Lake (n=17)		avg.	118 (4)	5,923 (13)	26 (5)
			range	100-130	100-68,000	14-54
Roskilde[3]	Lake (n=3)		avg.	36 (3)	96 (3)	3 (3)
			range	3-67	14-124	1.4-5.5
	Stream (n=2)		range	3-17 (2)	75-180 (2)	1.7-3.8 (2)

[1] Boutrup et al. (1998) - detection limit: DBP = 100 µg/kg dw; DEHP = 50 µg/kg dw; BBP = 20 µg/kg dw.
[2] Århus Amt (2003) - detection limit: DBP = 50-100 µg/kg dw; DEHP = 50-100 µg/kg dw; BBP = 5-20 µg/kg dw.
[3] Vikelsøe et al. (2001) - detection limit: DBP = 48 µg/kg dw; DEHP = 43 µg/kg dw; BBP = 10 µg/kg dw.
n: Number of locations.
( ): Number of positive findings.
<value: Below detection limit.

4.2.3 Water

One study with measurements of the concentration of phthalates in marine water will be presented together with the results from measurements in freshwater (Table 4.4).

In the investigation of phthalates in sea, stream and lake water from Roskilde Fjord (Vikelsøe et al., 2001), the concentrations during the season were measured. From each location, multiple samples (not the same number for all locations) were taken during one year, but only the averages of all samples are given. Thus, as with the sediment samples from the same investigation, the presented minimum and maximum values in Table 4.4 are the minimum and maximum average of the multiple samples taken on each location. Since only averages of all seasonal samples are available, the seasonal variation is not visible.

Table 4.4. The concentration of phthalates in water.

Region	Location	Value	µg/m^3		
			DBP	DEHP	BBP
Århus[1]	River (n=2) normal flow		<500	<200	<200
	flood	avg	<500	710 (2)	<200
Roskilde[2]	Stream (n=4)	avg	100 (1)	300 (4)	100 (4)
		range		120-730	30-160
	Tap water (n=1)		160	11,000	460
Denmark[3]	Stream (n=30)	median	na	670 (5)	na
		max.		1,500	
Roskilde[4]	Stream (n=4)	avg	nd	250 (4)	3 (4)
		range		110-410	2-5
	Lake (n=1)		11	410	13
	Bay (n=5)	avg	6 (4)	90 (5)	3 (5)
		range	2-31	70-190	2-7
Denmark[5]	Groundwater, upper (n=18)	median	380 (6)	na	na
		max.	810		
Denmark[6]	Groundwater (n=547)	median	1,200 (44)	na	na
		max.	8,100		

[1] Boutrup et al. (1998) - detection limit: DBP = 500 µg/m^3; DEHP and BBP = 200 µg/ m^3.
[2] Vikelsøe et al. (1998) - detection limit: DBP = 20 µg/ m^3; DEHP = 90 µg/ m^3; BBP = 0.3 µg/ m^3.
[3] Bøgestrand (2001) - detection limit: DEHP = 500 µg/ m^3.
[4] Vikelsøe et al. (2001) - detection limit: DBP = 30 µg/ m^3; DEHP = 25 µg/ m^3; BBP = 3 µg/ m^{3}l.
[5] Granth et al. (2001) - detection limit: unknown.
[6] GEUS (2001) - detection limit: unknown.
n: Number of locations.
( ): Number of positive findings.
<value: Below detection limit.
na: Not analysed.
nd:not detected

In the other investigation from Roskilde (Vikelsøe et al., 1998), the same streams were investigated, but not with seasonal measurements. Furthermore, tap water was included in order to find sources of contamination during the analysis. The values have relevance for the evaluation of human exposure through the drinking water.
There is no specific information on the depth of sampling in any of the studies. This could be of importance since xenobiotics are not distributed evenly through the water column. Investigations indicate that phthalates are concentrated in the surface water (Thomsen and Carlsen, 1998).

The two streams in Århus Amt were sampled as point samples from each stream at two different run-off situations: normal and flood. The detection limits were high (e.g., 200 µg DEHP/m^3) compared with some of the other investigations and only at flood, it was possible to detect DEHP.

In the national monitoring program of the aquatic environment (NOVA), the concentration of DEHP has been measured in 30 streams throughout Denmark (Bøgestrand, 2001). Only at five locations was DEHP detected maybe because the detection limit was high (500 µg/m^3) compared to some of the other investigations (e.g. 25 or 90 µg/m^3).
The concentration of DBP in the groundwater has also been measured during NOVA 2003 from both the groundwater monitoring areas (GEUS, 2001) and the agricultural watershed catchment areas (Granth et al., 2001). DBP was detected in 6 out of 18 investigated monitoring stations from 5 agricultural watershed catchment areas (Granth et al., 2001), and in 44 out of the 547 groundwater monitoring stations (GEUS, 2001).
The concentrations of phthalates - especially DEHP - in the tap water and DBP in the groundwater are very high compared to the concentrations in the other water samples. The concentrations of BBP in the streams located close to Roskilde are higher when measured in Vikelsøe et al. (1998) compared with the measurements in Vikelsøe et al. (2001); this is, however, not the case for DEHP where the concentrations are similar.

4.2.4 Air

To our knowledge, there are no measurements on the concentrations of phthalates in ambient air in Denmark. Based on measurements of the deposition of phthalates (see next section), Løkke and Rasmussen (1983) estimated an air concentration of DEHP and DBP in Denmark. The estimated values were 22 ng/m^3 and 9 ng/m^3, respectively.

Table 4.5. The concentration of phthalates in indoor air.

Location	Value	ng/m^3	
		DBP	DEHP
Classroom (n=1)	winter/spring	1,200/810	110/280
Day care centre (n=1)	winter/spring	1,350/1,010	1,050/160
Offices (n=4)	avg	820	300
	range	570-1,080	180-540

From Clausen et al. (1999)
n: Number of locations

Indoor air concentrations of phthalates have been measured in 4 offices, one school classroom, and a day care centre in Denmark (Clausen et al., 1999). All locations were considered as non-industrial environments.
The air was sampled during 4-8 hours on a single day in the winter and again in the spring. For the school and day care centre, both values are presented. For the 4 offices, the average and the minimum and maximum values for all measurements are presented (Table 4.5).

4.2.5 Deposition

In several studies, the deposition rate has been measured (Table 4.6). In the study by Vikelsøe et al. (1998), the measurements were made during a winter season (from fall to spring). Therefore, the range of values contains the seasonal variation, where the highest deposition rates are measured in the fall and the lowest in the early spring. The authors stated that the used method underestimate the dry deposition and during winter, also the wet (snow) deposition.

Table 4.6. The deposition rate of phthalates.

Locality	Comment	Value	µg/m^2/day			
			DBP	DEHP	BBP	DINP
Lille Valby	during 1 year[1a]	avg. (n=18)	0.58	0.63	0.05	0.05
	only winter season[1b]	range (n=8)	0.12-0.85	0.33-1.48	0.03-0.11	na
Ulfborg[2] (n=1)	regional		1.55	1.10	0.03	na
Århus[2] (n=1)	regional, town		1.82	0.43	0.13	na
Lyngby[3] (n=1)	non-industrial		0.8	1.90	na	na
København[3] (n=1)	150 m from plasticiser plant		2.2	4.70	na	na

[1a] Vikelsøe et al. (1999)
[1b] Vikelsøe et al. (1998)
[2] Boutrup et al. (1998)
[3] Løkke and Rasmussen (1983)
na: Not analysed
n: Number of locations

In Boutrup et al. (1998), the deposition rates are measured twice at two different locations. The location Ulfborg was considered to represent the diffuse deposition of phthalates with no direct point source and the measurements from Århus to represent the more direct deposition from the city (Boutrup et al., 1998). In this investigation, the measurements are expected to represent both the wet and dry deposition. The deposition of phthalates close to a point source (a plasticiser plant) was measured by Løkke and Rasmussen (1983) by analysis of snow samples.
Except for the investigation close to the point source, where deposition rates are high (Løkke and Rasmussen, 1983), the depositions are quite similar in the other investigations.

4.2.6 Sludge

There are several studies, where the concentration of phthalates in sludge from Danish sewage treatment plants (STPs) has been analysed. From these studies, it seems that the concentration of phthalates in sludge does not necessarily depend on the size of sewage treatment plant, or on the fraction of wastewater from industry or private households. Contribution of phthalates to wastewater from many sources (diffuse) rather than from certain point sources seems to be the determining factor since there are no clear correlations between the concentration of phthalates in the sludge and the size, type or wastewater source of the STP.

Table 4.7. The concentration of phthalates in sludge.

Region	Value	mg/kg dry weight (dw)			
		DBP	DEHP	BBP	DINP
Denmark [1] (n=20)	avg	3.88 (7)	37.9 (20)	0.18 (13)	na
	range	<0.02-26	39-170	<0.02-0.74	
Århus Amt[2] (n=6)	avg	0.55 (6)	25.2 (6)	0.17 (5)	na
	range	0.26-1.30	9-49	<0.05-0.41	
Denmark[3] (n=18-33)	avg	0.2 (10)	21.2 (33)	0.27 (8)	8.1 (16)

[1] Kristensen et al. (1996) - detection limit: DBP = 20 µg/kg dw; DEHP = 20 µg/kg dw; BBP = 20 µg/kg dw.
[2] Boutrup et al. (1998) - detection limit: DBP = 100 µg/kg dw; DEHP = 100 µg/kg dw; BBP = 50 µg/kg dw.
[3] MST (2001) - detection limit: DBP = 20-500 µg/kg dw; DEHP = 20-500 µg/kg dw; BBP = 20-500 µg/kg dw.
n: Number of locations.
( ): Number of positive findings.
-: Below detection limit.

Data from three investigations are reported (Table 4.7).
In the first study, the 20 investigated STPs represent typical Danish STPs as regards the type and size. The samples were taken as 7-10 sub samples. The amount of sludge corresponded to 6 hours of sludge production. The sub samples were mixed before analysis (Kristensen et al., 1996).
In the second investigation, the measurements were made on sludge samples from six STPs in one to three replicates (Boutrup et al., 1998).
In the national monitoring program of the aquatic environment (NOVA 2003), data were collected from a total of 35 STPs that in total received around 45% of the sewage in Denmark. The sludge (Table 4.7) and the inlet and outlet (Table 4.8) from municipal STPs were investigated (MST, 2001). Four replicate week samplings of the inlet and outlet in each STP were made. Furthermore, 7-11 purely industrial wastewater outlets were also analysed for the concentrations of phthalates in the wastewater. The reported values are those where more than 50% of the results were above detection limit.

The concentrations in the sludge found in all investigations are within the same range. DEHP is the only phthalate that was found in the outlet of the municipal STPs. The concentrations of the

other phthalates were below the detection limit. In the industrial outlets, the concentration of both DBP and DEHP was above the detection limit.

Table 4.8. The concentration of phthalates in inlet and outlet of STPs.

Source of wastewater		Value	µg/l			
			DBP	DEHP	BBP	DINP
Municipal[1] (n=16)	inlet	avg	1.19	17.7	0.99	0.22
	outlet	avg	-	1.88	-	-
Industrial[1] (n=7-11)	outlet	avg	0.6	1.3	-	-

[1] MST (2001) - detection limit: DBP = 0.1-0.5 µg/l; DEHP = 0.1-0.7 µg/l; BBP = 0.1-0.5 µg/l; DINP = 0.1-0.2 µg/l.

The fraction of degraded phthalates in the STPs has been estimated in two separate investigations (Table 4.9). The degradation of the substances was estimated as the difference between the amount let into the plant and the amount directed to the wastewater and sludge. Therefore, the fraction of degraded substance also includes any release to the atmosphere.

Table 4.9. The fraction of phthalates directed to water, sludge or degraded in the STPs.

Destination	Fraction of inlet			
	DBP	DEHP	BBP	DINP
directed to sludge[1] (n=6)	0.03-0.076	0.30-0.34	0.053-0.09	na
directed to water[1] (n=6)	0.004-0.42	0.03-0.07	0-0.42	na
degraded in the STP[1] (n=6)	0.54-0.92	0.63-0.66	0.54-0.95	na
degraded in the STP[2] (n=4)	0.6-0.98	0.2-0.7	0.8-0.99	0.8-0.9
directed to sludge[3] (n=3)	0.08	0.30	0.05	na
directed to water[3] (n=3)	(0.15)	0.07	(0.37)	na
degraded in the STP[3] (n=3)	0.77-0.92	0.63	0.58-0.95	na

[1] Kristensen et al. (1996)
[2] Grütner et al. (1996)
[3] Boutrup et al (1998)
(value): The value is estimated on basis of measurements below the detection limit.
na: Not analysed.

4.3 The food

The types of food of interest for this study are basic foodstuffs and include dairy products, meat, fish and crops produced in Denmark. Unfortunately, not many data on the concentrations in

Danish foodstuffs are available; one survey on the uptake of DEHP in barley and carrots (Grøn et al., 1999) and one investigation of the concentrations in milk (Petersen, 1999). Petersen (1999) estimates also the total daily intake of phthalates from food.

4.3.1 Crops

The survey on the uptake of selected xenobiotics by plants presents data on the concentrations of DEHP in barley plants and carrots. The study focuses on the uptake when sludge is used as a fertiliser and therefore, 7 different levels of sludge amendment (0-90 tons dry weight/hectare) were investigated. There was no significant effect of sludge added to the soil on the concentration of DEHP in the plants. Thus, in Table 4.10, the results from two replicate analyses from all sludge treatment are pooled.
The plants were grown in a sandy soil from Southern Jutland. The bioavailability of xenobiotics is assumed to be higher in a sandy soil compared with a soil with higher content of clay or organic matter. The concentrations of DEHP (and DBP) in the soil, where the carrots and barley were grown, were at the same levels as those in heavily sludge-amended soils analysed in other investigations (Table 2.1), even though no sludge had been applied to the soil prior to the analysis. When increasing amounts of sludge were applied, the concentration of DEHP also increased.
The authors commented that the high concentration of DEHP in the rind of the carrot could be explained by the fact that even after cleaning, soil adheres to the rind and therefore, DEHP in the soil is included in the analysis.

Table 4.10. The concentration of DEHP in crops and vegetables.

Plant	Part of plant	mg/kg dry weight (dw)
		DEHP
barley	roots	0.3-1.3
	leaves	<0.3-0.6
carrots	rind	0.8-6.3
	heart	0.3-1.6
	leaves	0.2-2.3

From Grøn et al. (1999) - detection limit: DEHP = 0.3 mg/kg dw.

4.3.2 Dairy products

The concentration of DEHP in Danish milk measured by Petersen (1999) is presented in Table 4.11 together with data on the concentrations of DEHP in two other Danish dairy products analysed by other researchers (Sharman et al. (1989) as reported in Petersen (1999)). Furthermore, the estimated total daily intake from food for humans in Denmark is given. The estimate is based upon analysis of phthalates in 29 different meals collected by test persons during 24 hours and normalised to a daily diet of 10 mega Joule energy.

The concentration of DEHP in the milk was mainly below the detection limit (0.05-0.2 mg/l), except from a few samples where it ranged from 0.05 to 1.4 mg/l. The concentration of DEHP in the products does not only represent what is excreted in the milk, but also the amount of DEHP that has migrated from any plasticiser in contact with the milk, cheese or butter during processing, packing etc.

There exist one Norwegian study (Sharman et al., 1994 – quoted from Petersen, 1999) on the concentration of DEHP in milk and cream that can be used as a supplement to the Danish data. The concentration found in that study was 0.05 mg DEHP/kg milk (1% fat) and is similar to what was found by Petersen (1999).

Table 4.11. The concentration of phthalates in dairy products.

Product	mg/kg wet weight		
	DBP	DEHP	BBP
milk, 3,6% fat (n=15)	na	<0.05-0.14	na
cheese (n=1)	na	3.1	na
butter (n=1)	na	3.5	na
	mg/person/day		
Daily intake, averages[1] (n=29)	0.13-0.29	0.19-0.3	0.02-0.03
Daily intake, max. value	0.72	1.1	0.32

[1] Minimun and maximum averages are the result of two different ways of using the values below the detection limit and below determination limit (see Petersen (1999)). n: Number of products.
na: Not analysed.

5 Comparisons of measured and estimated concentrations

In this section, the predicted environmental concentrations (PECs) and the concentrations in food estimated by EUSES (presented in section 3) will be compared with the measured concentrations in the environment and food (presented in section 4).

5.1 The environment

5.1.1 Soil

The local PECs for DEHP, DBP and DINP in agricultural soils and the concentrations measured in heavily sludge amended soils are in the same range, with maximum local PECs being a bit higher than maximum measured concentrations in heavily sludge amended soils. The amount of sludge applied to the local agricultural soil in the EUSES calculations is set to 3,000 kg/ha/year. This figure is somewhat lower than the amount applied to the investigated soils; there are no

further information on the extent of exposure of the investigated soils or the sludge applied to the soils.

The estimated regional concentrations are compared with the data for soil receiving no sludge. For DEHP and DBP, the regional PECs (8.0 and 4.4 µg/kg dry weight (dw), respectively) are within the broad range of measured concentrations (1-54 and 1-21 µg/kg dw, respectively), but at the lower end of the range. Also for DINP, the regional PEC (1.1 µg/kg dw) seems to be a slight underestimation of the actual background level of DINP in Danish soils since it is similar to the lowest measured value (1 µg/kg dw).

There are no measured data of DIDP.

5.1.2 Sediment

For DINP and DIDP, no PECs have been estimated for the marine compartment (water and sediment) and there are no measured data for fresh water and fresh water sediment.

For DEHP, the regional PEC in sediment (3,480 µg/kg dw) is above the measured concentration in sediment from a lake with no direct sources (310 µg/kg dw) and also above the concentration found in sediments from lakes and streams in the Roskilde region (maximum: 124 µg/kg dw); however, no information about the extent of exposure has been given. The regional PEC in sediment is at the same level as the average concentration measured in sediment from lakes and streams in Århus Amt receiving wastewater outlet (2,335, 2,928 and 5,923 µg/kg dw), but the range of these concentrations are very broad and relatively few of the concentrations are that high.
It is more or less the same picture for DBP, where the regional PEC for DBP (634 µg/kg dw) in sediment is at the same level as measured values from Århus Amt and higher than in the lake with no direct sources (<100 µg/kg dw).

For both DEHP and DBP, the maximum local PEC (77,800 and 9,270 µg/kg dw, respectively) in sediment is around 10 times higher than the maximum measured value (6,900 and 630, respectively). However, there is no information whether any DEHP/DBP consuming industry is situated close to any of the sample sites and thus, the sample sites where the maximum concentration has been measured are not necessarily representative for the local exposure levels of the substances.

5.1.3 Surface and groundwater

The regional PEC for DEHP in surface water (296 µg/m^3 (total)) seems to be a reasonable estimate when compared with the average measured concentrations from the different locations; the estimated value is in the middle of the range of average measured concentrations (90-710 µg/m^3).The regional PEC for DBP (1,160 µg/m^3 (total)) is much higher than the few available measured concentrations of DBP in fresh water (10-100 µg/m^3). In the investigation where the highest value was found, the concentration of DBP in the three other investigated streams was below the detection limit (20 µg/m^3).
The local PECs in surface water exceed the measured concentrations for the specific substances, but the sample sites are not referred to as representative for local exposure levels.

There are no measured concentrations of DINP and DIDP in surface water.

The estimated regional concentration of DBP in groundwater (34.5 µg/m^3) is 10 times lower than the measured median concentration (380 µg/m^3) in the upper groundwater from the monitoring stations in the agricultural catchment areas and it is 35 times lower than the concentration found in the groundwater monitoring areas (median: 1,200 µg/m^3). Even though the concentration of DBP was below the detection limit in a lot of the samples from monitoring stations, the estimated regional concentrations is still an underestimation of a realistic worst-case groundwater concentration.
In EUSES, the concentration in the groundwater is actually the concentration in the pore-water of the agricultural soil. This is supposed to be a worst-case assumption since transformation and dilution in deeper soil layers is neglected. At least in the case of DBP, it is not a worst-case estimation.
The estimated maximum concentration in groundwater for the local scenario of the formulation of adhesives etc. is higher than the measured maximum concentration, but there is no information on the extent of exposure of the groundwater monitoring stations and it is therefore uncertain whether the measured concentrations are representative for a regional or a local exposure level.
There are no measured concentrations of DEHP, DINP and DIDP in groundwater.

5.1.4 Air

There are no measurements of the ambient air concentration of phthalates in Denmark, but Løkke and Rasmussen (1983) made estimations of the ambient air concentration based on measured deposition of DEHP and DBP. These estimations for DEHP (22 ng/m^3) and DBP (9 ng/m^3) are around 4 and 6 times higher than the EUSES estimated regional air concentrations (5.3 ng/m^3 and 1.5 ng/m^3, respectively).

The measured indoor concentrations are not comparable with the estimated concentrations in ambient air. But since the estimated concentration in ambient air is used to estimate the daily uptake of the phthalates via inhalation and because many hours are spent indoor, it is interesting to look at the differences between the estimated concentration in ambient air and the measured indoor concentrations.
The measured indoor concentrations of DEHP (e.g., 300 ng/m^3) and DBP (e.g., 830 ng/m^3) are one and two orders of magnitudes higher, respectively, than the estimated concentrations in ambient air. Therefore, it is likely that the actual uptake of DEHP and DBP via inhalation is higher than the estimated regional uptake. For some of the local scenarios for both DEHP and DBP, estimated ambient air concentrations are at the same levels as the measured indoor concentrations. The daily uptake via inhalation in these local scenarios is much higher than the estimated regional uptake (almost 100 times for DEHP and 1000 times for DBP).

By assuming that the average person during a day spend more hours indoor than outdoor, it is possible to estimate the daily uptake of DEHP via inhalation of air using an average air concentration of 0.3 ng/l (from indoor measurements) instead of the estimated concentration in ambient air of 0.0053 ng/l.
The daily uptake via inhalation of air would then be (0.3 µg/m^3*20 m^3/day / 70 kg)*0.75/0.5 = 0.13 µg/kg/day instead of the value 0.0023 µg/kg/day estimated by EUSES. This is a 50-fold

increase and it would further result in an increase of the total daily intake by around two times. A separate consumer exposure scenario for exposure via indoor air is performed in section 10 and therefore, no further details will be included at this point.

5.1.5 Deposition

EUSES predicts only the local deposition of phthalates close to point sources (1000 m). Therefore, only the concentrations of DEHP and DBP measured in samples taken 150 m from a plasticiser plant are comparable with the estimated concentrations. The highest estimated local deposition rate of DBP (4.4 µg/m^2/day) is at the same level as the measured value (2.2 µg/m^2/day), whereas the highest estimated local deposition of DEHP (98.9 µg/m^2/day) is around 20 times higher than the measured value (4.7 µg/m^2/day).
The other measurements of the deposition of DEHP and DBP and the measurements of DINP have been performed from locations more comparable to the regional scale; these are within the very wide range of estimated local deposition rates.

5.1.6 Sludge and wastewater

As for deposition, EUSES only estimates the concentration of substances in sludge and wastewater for the local scenarios and does not estimate a regional value.
Again, as for deposition, the ranges of estimated local concentrations in sludge are so wide that it is no surprise that the measured values are within the range of predicted local concentration. Furthermore, there is no prediction of the concentration in purely industrial sludge and there are no predictions from a local municipal wastewater scenario to compare with.

The measured concentrations of DBP and DEHP in outlets from 7-11 industries (0.6 and 1.3 µg/kg dw, respectively) are much lower than many of the estimated local concentrations in effluent wastewater (maximum estimated concentrations are 250 and 100 µg/kg dw, respectively). In the investigation of the concentrations of DBP and DEHP in industrial outlets, no information is provided on whether these phthalates are used by the industries.

The estimated fate of the substances in the regional STP can be compared with the measured fractions of substance that are directed to air and to water, or are degraded. There seems to be at least some accordance between the measured and the estimated fate of the substances in the STP even though there are differences. The measured data show that DEHP to a higher extent is directed to the sludge compared with DBP, whereas the fraction of DBP that is degraded in the STP reaches higher levels than the fraction of degraded DEHP. The single investigation of DINP shows a degradation rate as high as for DBP (up to 90%) whereas the estimated degraded fraction of DINP is much lower (10%), instead DINP is mainly directed to the sludge (84%).

5.2 Food

5.2.1 Dairy products

In the few milk samples where DEHP was detected, the concentration was much higher than what has been estimated as the regional concentration in cow's milk by EUSES (0.05-1.4 mg/kg wet weight (ww) compared to $7.2*10^{-6}$ mg/kg ww). Even in the local scenario, where the highest concentration of DEHP was predicted in the milk, the estimated concentration is only 0.0047 mg/kg ww. As already mentioned, the measured concentration could be high because of contamination with phthalates during processing in the dairy and that is not included in EUSES predictions. The estimated concentration could be low because of an underestimation of the amount excreted in the milk.

5.2.2 Vegetables

The measured concentration of DEHP in both carrots and barley exceeds the regional estimated concentrations (0.2-6.3 mg/kg dw compared to 0.002 (leaves) / 0.007 (roots) mg/kg ww - conversion factor from dry weight (dw) to wet weight (ww) = 0.25). The estimated maximum concentration at local exposure levels (1.2 (leaves) / 3.8 (roots) mg/kg ww) is at the same level as the measured values.
Some of the crops were grown in heavily sludge amended soil with resulting increasing concentration of DEHP in the soil, but this did not influence the level of DEHP in the plant as it was more or less the same as in plants grown in soil receiving no sludge. However, the concentration of DEHP in this particular soil was higher (83-270 µg/kg dw) even before sludge amendment than the average in other soils not receiving sludge (see Table 4.1). Even though no correlation between DEHP concentration in the soil and the concentration of DEHP in the plant at relatively high soil concentrations was found, it is possible that a relationship at lower soil concentrations exist. Thereby, crops grown in soil containing less phthalates than in the study by Grøn et al. (1999) are likely also to contain less DEHP than the measured concentrations.
The origin of the DEHP measured in the leaves of the plants is probably mainly by atmospheric deposition and independent of the concentration in the soil. The atmospheric concentration of DEHP in the study was not mentioned.

5.2.3 Fish

No Danish measurements on the concentration of phthalates in fish are available.
According to the EU-RAR of DEHP (2001), measured levels of DEHP in fish vary between a few µg/kg and 19,000 µg/kg wet weight (ww). This interval is based on different investigations as e.g., a recent study from Austria (Pfannhauser et al., 1997 cited in DEHP, 2001) where 180 fishes (trout, brown trout, char, carp and eel) were collected from 58 different locations. DEHP was found in 71 of the samples. The highest measured concentration was 2,600 µg/kg ww and the 90^{th}-percentile was approximately 500 µg/kg ww. In another study where phthalates were measured in 25 fishes in The Netherlands, the concentration of DEHP ranged from <1 µg/kg ww to approximately 300 µg/kg ww (RIC, 2000 – cited in DEHP, 2001). In the same investigation, the concentrations of DINP and DIDP in the fishes were below the detection limit (10 µg/kg ww) (RIC, 2000 - cited in DEHP, 2001).

The estimated regional concentration of DEHP in fish (191 µg/kg ww) seems to be reasonable, but not worst case, compared to these investigations. The estimated concentration of DINP and DIDP (10 µg/kg ww and 3 µg/kg ww, respectively) is at or below the detection limit in the investigation.

5.2.4 Meat

No Danish measurements on the concentration of phthalates in meat are available. To our knowledge, no investigations from other northern European countries are available, that could be used instead for the comparison with estimated concentrations in meat.

5.2.5 Tap water

The measured concentration of DBP in Danish tap water (160 µg/m^3) is lower than the estimated regional concentration in drinking water (574 µg/m^3), whereas for DEHP, the measured concentration value is much higher than the estimated regional concentration (11,000 µg/m^3 compared with 56.9 µg/m^3). Whether the investigated water samples are representative for the drinking water in Denmark is uncertain, since only one single measurement has been made.

5.3 The total daily intake

The regional estimated total daily intakes of DEHP (0.19 µg/kg bw/day) and DBP (0.1 µg/kg bw/day) are approximately 20 and 40 times lower, respectively, than the mean daily intake calculated based on the measured concentration in meals (4 µg/kg bw/day for both DEHP and DBP) (Petersen, 1999). The highest estimated local daily intake of DEHP (20 µg/kg bw/day) and DBP (60 µg/kg bw/day) are at the same level as the maximum measured daily intake (16 µg/kg bw/day and 10 µg/kg bw/day, respectively).
As already mentioned in the section 4.3.2 on the concentration of DEHP in cow's milk, food can be contaminated with phthalates through contact with phthalate containing materials. The investigated meals that are the basis for the calculations of daily intake (Petersen, 1999) may also contain phthalates that have migrated to the food during processing, packaging etc. This source of phthalates in food is not included in the EUSES estimations, which may therefore give a lower estimate of the total daily intake of man by exposure via the environment. However, this scenario is included in the consumer exposure assessment and thus, in the evaluation of the combined exposure, see Part 3.

The estimated fractions of the daily intake from the different sources to the total daily intake are summarised in Table 5.1.
For all four substances, the fraction of daily intake via leaves and roots vegetables is high. For DEHP, the intake from fish is also a major source of exposure whereas for DBP, water contributes significantly. For DINP and DIDP, almost all the sources contribute significantly to the total daily intake.
According to the EUSES estimates in the EU-RAR of DINP, fish and root crops are the dominating exposure pathway. In the present assessment, these sources are also important but together with several other sources as well (Table 5.1).

Table 5.1. The fraction of the total daily intake from different sources. The fractions are from the present EUSES estimations with the Danish regional profile (EUSES) and also from two refined estimations (estim. 1 and 2; see next section, 5.3.1).

Sources	DBP		DEHP			DINP	DIDP	BBP
	EUSES	estim. 1	EUSES	estim. 1	estim. 2	EUSES	EUSES	estim. 1
Water	**0.251**	0.03	0.0134	0.012	**0.11**	0.0088	0.0024	0.02
Fish	0.015	0.00008	**0.75**	**0.66**	**0.10**	**0.494**	**0.26**	0.05
Leaves	**0.34**	**0.13**	0.0991	**0.12**	**0.53**	0.0567	0.064	**0.6**
Roots	**0.38**	**0.83**	**0.125**	**0.20**	**0.13**	**0.206**	**0.35**	**0.3**
Meat	0.00785	0.003	0.00045	0.0004	0.00007	**0.11**	**0.13**	0.02
Milk	0.00754	0.003	0.00041	0.0005	**0. 12**	**0.10**	**0.12**	0.02
Air	0.00315	0.001	0.0119	0.014	0.002	0.0215	0.079	0.003

bold: The main sources of exposure (the fractions beyond 0.1)

5.3.1 Refined estimations of the total daily intake

The refined estimations of the regional total daily intake will be based on the available measurements from Denmark. Since there are very few measurements of the concentration of DINP and DIDP, the estimations will only be performed for DEHP and DBP. BBP has not beenincluded in the previous EUSES predictions, but data on BBP in the Danish environment are available and therefore, a total daily intake of BBP will be estimated on the basis of these measured environmental data. The physico-chemical properties for BBP appears in Table 2.5. Furthermore, the bioconcentration factor (BCF) for fish will be set to 449 in accordance with the EU-RAR of BBP (Draft report 2002).

The refined estimations will be made in two different ways, estimation 1 and 2. In estimation 1, EUSES estimates the regional total daily intake based on measured environmental concentrations and thereby, on estimated concentrations in the various foodstuffs. In estimation 2, EUSES estimates the regional total daily intake based on the measured concentrations in the various foodstuffs. The details are described below.

In estimation 1, the measured concentrations of DBP and DEHP in the environmental compartments that best represent the model compartments will be used. Based on the previously mentioned bioaccumulation and bioconcentration factors, partition coefficients etc., EUSES estimates the concentrations in the different foodstuffs and then the total daily intake. As a worst-case assumption, the highest measured concentration from each environmental compartment will predominantly be used (see the values in Table 5.2).

With respect to soil, “agricultural soil not heavily amended with sludge” represents agricultural soil, “non agricultural soil” represents natural soil, and the input value for the concentration in industrial soil is taken from “heavily sludge amended soil” (see Table 4.1).

The concentration in air estimated by Løkke and Rasmussen (1983) will be used as the input value for the concentrations in ambient air. There are no data on the concentration of BBP in ambient air, but in indoor air, the concentration of BBP is similar to the concentration of DBP. Therefore, it will be assumed that the concentration of BBP in ambient air also is similar to the DBP concentration.
The measured concentrations of phthalates in sediment differ a lot in the two reported investigations (see Table 4.3) and it is therefore difficult to select the concentration as input value in the refined estimations. Since the PECs of both DBP and DEHP in sediment is at the higher end of the measured range (see Table 3.1), the PECs will be used in the refined estimations. For BBP where no PEC exists, the average concentration in the sediment in streams measured by Boutrup et al. (1998) will be used as input value.
The input value for the groundwater concentration of DBP will be the highest average measured value. For BBP and DEHP, EUSES estimates the groundwater concentration. The surface water concentration of DEHP used in estimation 1 will be the maximum measured concentration. For DBP, the only measurement above the detection limit will be used as input value even though it is lower than the concentration estimated in the original EUSES calculation.

Table 5.2. Concentrations in the environmental compartments and in food used in the refined estimations of the daily intake by EUSES.

Compartment	Estimation	DBP	DEHP	BBP
Agri. soil (µg/kg)	1	21	54	35
Natural soil (µg/kg)	1	8	27	0.3
Industrial soil (µg/kg)	1	830	3,400	910
Surface water (µg/l)	1	0.1	0.73	0.16
Sediment (µg/kg)	1	PEC (634)	PEC (3,480)	83
Air (µg/m^3)	1,2	0.009	0.022	0.009[1]
Ground water (µg/l)	1	1.2	-	-
Drinking water (µg/l)	2	-	11	0.46
Leaves (mg/kg ww)[2]	2	-	0.2	-
Root (mg/kg ww)[2]	2	-	0.2	-
Milk (mg/kg ww)	2	-	0.05	-
Fish (mg/kg ww)[3]	2	-	Est. 1 (0.613)	-
Meat (mg/kg ww)[3]	2	-	Est.1 ($1.02*10^{-4}$)	-

[1] Data from DBP, see text.
[2] A conversion factor of 0.25 has been used to extrapolate from dry weight (dw) to wet weight (ww) (0.8 mg/kg dw = 0.2 mg/kg ww).
[3] The concentration in fish and meat used in the second estimation will be those estimated in the first estimation based on measured concentrations in the environmental compartments.

Secondly, in estimation 2, the few available measured concentrations of the DEHP in food and water will be used. Since no Danish measurements of the concentration in meat and fish are

available, the concentration in meat estimated by EUSES on the basis of the concentrations in the Danish environment (estimation 1) will be used.
The input value for the measured concentration of DEHP in milk will be 0.05 mg/kg; it should be kept in mind that the DEHP concentration in many of the milk samples was below this value. On the other hand, other dairy products with a higher content of fat contain more DEHP. The mean concentration of DEHP in the heart of the carrots will be used as the input value for the measured concentration in roots, and the mean concentration of DEHP in the barley and carrot leaves as the input value for the measured concentration in leaves (see Table 5.2). These values are expressed as concentrations per kg dry weight. When used as input values in EUSES, they have to be expressed as concentrations per kg wet weight and a conversion factor of 0.25 will be used to convert dry weight to wet weight (assuming 75% water content) (EC, 1996). The single measurement of the concentration of DEHP in tap water and the highest measured concentration of DEHP in fish will be used as the input value for the measured concentration in drinking water and fish, respectively.

The total daily intake of DEHP by adults and children increased around three times and the daily intake of DBP increased 16 times when the measured environmental concentrations were used as input data in EUSES to estimate the concentration in food (estimation 1) instead of the predicted regional environmental concentrations originally estimated by EUSES (see Table 5.3). The total daily intake of BBP based on the measured concentrations of BBP in the environment is very similar to the daily intake of DBP.
The total daily intake of DEHP increased around 30 times when measured data on the concentration of DEHP in fish, milk, crops and drinking water were used as input data to estimate the daily intake (estimation 2) compared with the original regional EUSES estimation. This refined estimated total daily intake of DEHP by adults is similar to the measured mean daily intake (Petersen, 1999) (Table 5.3), where the intake via inhalation and drinking water was not included. But the estimated daily intake via inhalation is, when an air concentration of 0.022 µg DEHP /m^3 is used, not a major route of exposure (Table 5.1).

The importance of the different sources is similar whether the total daily intake is estimated solely by EUSES, or environmental concentrations are added to the model (see Table 5.1). In the estimated total daily intake based on measured concentrations of DEHP in food (estimation 2), almost all sources contribute significantly to the daily intake.
As mentioned previously, the original EUSES predictions yield a lower value for the total daily intake than the measured intake (Petersen, 1999). When the estimation is based on measured environmental concentrations (estimation 1), the estimated value for the total daily intake is increased. For DEHP, the total daily intake estimated by EUSES based on measured environmental concentrations (estimation 1) is still somewhat below the measured mean value of the daily intake for adults (Petersen, 1999), but for DBP, it is at the same level. Also, the estimated total daily intake of BBP based on the measurements of BBP in the environment is at the same level as the measured total daily intake. In the estimation based on measured concentrations of DEHP in several foodstuffs (estimation 2), the daily intake of DEHP is similar to the measured mean total daily intake (Petersen, 1999).

Table 5.3. The total daily intake from the different estimations, measurements, and from the EU-RARs.

Total daily intake	mg/kg bw/day				
	DEHP	DBP	BBP	DINP	DIDP
EUSES, reg., adult	$1.9*10^{-4}$	$1.0*10^{-4}$	-	$1.6*10^{-5}$	$9.2*10^{-6}$
EUSES, reg., child, 1-6 y	$8.7*10^{-4}$	$4.9*10^{-4}$	-	$8.0*10^{-5}$	$4.6*10^{-5}$
EUSES, reg., child, 7-14 y	$3.8*10^{-4}$	$2.1*10^{-4}$	-	$3.5*10^{-5}$	$2.0*10^{-5}$
EUSES, max. local, adult	0.02	0.06	-	$5.1*10^{-3}$	$2.9*10^{-3}$
EUSES, max. local, child, 1-6 y	0.10	0.40	-	0.03	0.02
EUSES, max. local, child, 7-14 y	0.04	0.16	-	0.01	$6.8*10^{-3}$
Estim. 1, reg., adult	$7.1*10^{-4}$	$1.6*10^{-3}$	$9.7*10^{-4}$	-	-
Estim. 1, reg., child, 1-6 y	$3.4*10^{-3}$	$8.0*10^{-3}$	$5.9*10^{-3}$	-	-
Estim. 1, reg., child, 7-14 y	$1.4*10^{-3}$	$3.5*10^{-3}$	$2.4*10^{-3}$	-	-
Estim. 2, reg., adult	$4.5*10^{-3}$	-	-	-	-
Estim. 2, reg., child, 1-6 y	0.026	-	-	-	-
Estim. 2, reg., child, 7-14 y	0.011	-	-	-	-
Measured[1], adult, means	$2.7–4.3*10^{-3}$	$1.8–4.1*10^{-3}$	$2.8–4.3*10^{-4}$	na	na
Measured[1], adult, max.	0.016	0.01	0.0046	na	na
EU-RAR[2], reg., adult	$1.73*10^{-3}$	$3.59*10^{-4}$	-	$1.0*10^{-3}$	$2.0*10^{-3}$
EU-RAR[2], reg., child	0.018	-	-	$6.5*10^{-3}$	0.013
EU-RAR[2], max. local, adult	0.085	0.0925	-	0.028	0.027
EU-RAR[2], max. local, child	0.312	-	-	0.156	0.166

[1] Petersen (1999) – see also Table 4.11.
[2] EU-RAR of DBP (2001); EU-RAR of DEHP (2001); EU-RAR of DINP (2001); EU-RAR of DIDP (2001).
-: Not estimated.
na: Not analysed.

5.3.2 Total daily intake from the EU-RARs of DEHP, DBP, DINP and DIDP, and from other investigations

In Table 5.3, the regional total daily intake estimated by EUSES in the EU-RARs is presented (DBP, 2001; DEHP, 2001; DINP, 2001; DIDP, 2001).

Concerning DEHP, the regional total daily intake in the EU-RAR is between what has been estimated on the basis of the measured concentrations in the environment (estimation 1) and the estimation based on measured concentrations in foodstuff (estimation 2). The maximum daily intake from the local scenarios is very similar in the EU-RAR of DEHP (2001) and in the present assessment. As in the present assessment (see Table 5.1), the predominant contribution to the total daily intake of DEHP for adults as estimated in the EU-RAR of DEHP (2001) is from fish (52%) and root crops (18%), but also from dairy products (24%).

For DBP, the estimated regional total daily intake in the present assessment is at the same level as the regional total daily intake estimated in the EU-RAR of DBP (2001), whereas the only refined estimation of the regional total daily intake of DBP (estimation 1) is somewhat higher. According to the EUSES estimations made in the EU-RAR of DINP (2001) fish and root crops are the dominating exposure pathways for DINP. In the present study they are also important pathways, but together with several other pathways (table 5.1).

For DINP and DIDP, the maximum local total daily intake estimated in the EU-RARs DINP, 2001; DIDP, 2001) is around ten times higher than the maximum local total daily intake estimated for Denmark in the present assessment. The regional daily intake of DINP and DIDP is approximately 100 times higher than the regional daily intake estimated for Denmark.
As mentioned before, the used amount of DINP and DIDP is expected to have increased over the last years. Therefore, the regional tonnage used in the present emission assessment may be too low compared with the currently used tonnage of these two phthalates, which consequently may result in a underestimation of the total daily intake. Furthermore, it is very likely that the values estimated for the total daily intake would increase if the estimates could be based on measured concentrations of DINP and DIDP in the environment as it has been seen for DEHP and DBP.

Beside the EU-RARs (Table 5.3), several other studies have estimated the daily intake via the environment (food, water, air) of especially DEHP, DBP and BBP.

A total daily dietary intake of DEHP of 3-30 μg/kg bw/day has been assessed for an adult of the general population in the United States (Doull et al., 1999). The range of the estimated adult exposure to DEHP via the environment in the present assessment depending on input data in EUSES is similar (0.2-20 μg/kg bw/day; the highest value is the maximum local daily intake). The EUSES estimations also include exposure via ambient air and drinking water, but according to the EUSES estimations, both ambient air and water (except for the estimation based on measured concentrations in food) contribute relatively little to the total exposure (Table 5.1).

MAFF estimated the exposure to phthalates through dietary intake based on a survey of the concentration of phthalates in fatty food (MAFF, 1996). According to MAFF (1996), the contribution from other food groups was expected to be negligible. This is not in accordance with the present EUSES estimations where other food groups contribute as well (Table 5.1).
The 97.5 percentile of estimated daily dietary intake was 20 μg BBP, 300 μg DEHP, and 31 μg DBP per person/day (0.3, 4.2, and 0.4 μg/kg bw/day, respectively, if the body weight is assumed to be 70 kg) (MAFF, 1996).
For DBP, the MAFF estimate is in the low end of the range of the present EUSES estimations (0.1-60 μg DBP/kg bw/day, where the highest value is the maximum local daily exposure to DBP or DEHP). However, the Danish tonnage data for DBP in the present assessment were scarce and the use of DBP is probably descending, which may have resulted in an overestimation of the exposure to DBP.
For BBP, the MAFF estimate is a bit lower than that in the present EUSES estimation (0.97 μg BBP/kg bw/day based on measured environmental concentrations).
For DEHP, it is very similar to the exposure estimated on basis of measured concentrations in food and water (estimation 2: 4.5μg/kg bw/day).

The international Program on Chemical Safety (IPCS) has also estimated the total daily dietary intake of DBP and BBP. The estimates are based on a Canadian market-basket survey of 98

different food types. For adults, the intake of DBP was 7 μg/kg bw/day (IPCS, 1999). The intake of BBP for adults was 2 μg/kg bw/day and it could be up to three-fold higher for infants and children (IPCS, 1999). In the present assessment, in general the exposure to children (1-6 years) via food etc. is approximately 6 times higher than for adults. Both estimates (IPCS, 1999) are higher than the present estimates based on measured environmental concentrations, but for DBP it is lower than the maximum estimated exposure.

Exposure via food, water, and air to both DEHP and DBP by different age groups in the population of Canada has also been estimated (Chan and Meek, 1994; Meek and Chan, 1994). As in the present study, the maximum exposure was estimated for young children (0.5-4 years old) to 19 μg DEHP/kg bw/day and 5 μg DBP/kg bw/day. For both substances, food was the major source of exposure, followed by indoor air and drinking water and the lowest exposure was via ambient air and soil. Exposure via indoor air is not included in the exposure estimates by EUSES. Therefore, the values from the Canadian survey are not totally comparable with the present EUSES estimations. Anyhow, the Canadian exposure to DEHP is similar to the exposure estimated in the present assessment based on measured concentrations in food and water (26 μg DEHP/kg bw/day (1-6 years)). The Canadian exposure to DBP is similar to the exposure estimated in the present assessment based on measured concentrations in the environment (8 μg DBP/kg bw/day (1-6 years)).

The present estimated regional total daily intake of BBP based on measured concentrations of BBP in the environment is at the same level as the estimated total daily intake of BBP by Effting and van Veen (1998) based on assumed concentrations in Dutch food. As in the present assessment of DEHP and DBP, the authors found that EUSES underestimates the concentration of BBP in food and thereby, the total daily intake of BBP. Instead they predicted the total daily intake of BBP based on assumptions regarding the concentration of BBP in Dutch food (the assumptions is based on measurements in food particular from the United Kingdom). Effting and van Veen (1998) also found, in concordance with the present assessment, that for BBP, the intake of crops is a major source of the total daily intake of phthalates.

In general, the present estimations of exposure to DEHP, DBP and BBP are at the same level as the exposure estimated in other investigations although the range of both the estimated exposures in the present assessment as well as in other investigations covers around one order of magnitude. It is mainly the exposure estimated based on measured concentration in the environment (and foodstuff for DEHP) that are comparable to the other investigations, whereas the maximum local daily intake based on the original EUSES estimations in general are higher. To our knowledge, the exposure via the environment to DINP and DIDP is only evaluated in the EURARs of DINP (2001) and of DIDP (2001) and as already mentioned, both the regional and the maximum local total daily intake estimated in the present assessment is between 10 to 100 times lower than in the EURARs.

6 Discussion of exposure via the environment

6.1 The Danish country profile

The use of a specific Danish profile only changed the exposure assessment of DEHP slightly (see section 3.3.2). The conclusion that the use of specific regional parameters in EUSES instead of the EUSES defaults values only lead to small improvements of the predicted environmental concentrations (PECs) is in concordance with the results obtained in other studies.
Schwartz et al. (2000) assessed the aquatic fate of polycyclic musk fragrance (HHCB) by using both the EUSES standard scenario and different site-specific scenarios. Based on a comparison of estimated values with measured data, they concluded that there was only a slight improvement in the exposure estimations by replacing the defaults values in the EUSES standard region with a specific regional profile. Considerable improvement was obtained by using more specific consumption quantities instead of the standard 10% of the European tonnage as the regional tonnage, and measured partition coefficients and bioconcentration factors. Similar results were found in a more comprehensive study with several substances and including all environmental compartments (Berding et al., 2000).
The relatively small improvement obtained by the use of specific regional profiles instead of the default values in these studies could be explained by either a low degree of influence of the regional parameters on the exposure assessment or if the specific region (in both studies the specific region was the North Rhine Westphalia) is very similar to the standard region in EUSES.
As already mentioned, some of the regional parameters in the present exposure assessment showed a high impact on the exposure estimations (see section 3.3). Furthermore, a recent study where the regional parameters were changed from the standard region to a region representing the Northern or Southern Europe, the PECs changed a lot more than when changing the parameters to the North Rhine-Westphalia region (Berding et al., 2002). The range between the “North” and “South” estimations of the concentration in any compartment was up to 2 orders of magnitude.

6.2 Predicted environmental concentrations

In general, the regional PECs of DEHP and DBP in many of the environmental compartments were within the range of available measured concentrations. However, the PECs were not always worst-case concentrations, e.g., in soil, the estimated concentration was at the lower end of the range of measured values. Only for DBP in surface water did the estimated concentration exceed all measured concentrations. Comparisons of estimated and measured concentrations of DIDP and DINP were only possible for the soil compartment since for all other compartments, no measured data are available.

The reasons why the intended conservative EUSES does not predict worst-case concentrations for the phthalates in the present assessment as expected could be poor data input into the model, unrepresentative measurements, or the models in EUSES could be inappropriate to predict concentrations of phthalates.
The uncertainty of the values of the physico-chemical properties and also the degradation parameters of the phthalates is well known. Also, data on the emission are old and limited in

number and therefore, several assumptions have been made concerning the regional tonnage and the emissions. Furthermore, due to lack of data, the emission via waste has not been included, thereby underestimating the true emission.
It is generally expected that the EUSES model will overestimate the exposure. Berding et al. (2000) included DEHP in a study where estimated and measured concentrations were compared. In general, the estimated concentrations, especially if they were estimated by the standard EUSES, were overestimated. Also, by the use of more precise parameters (including regional specific parameters, measured degradation parameters etc.), the predicted concentrations, in general, were somewhat higher than measured concentrations. Only for the concentration in soil, an underestimation was observed.
Regarding the reliability of the measurements there is an agreement between the results obtained in different investigations for some compartments (e.g. soil), whereas for other compartments (e.g. the sediment), there are big differences between the results from the various investigations. But, as already mentioned, the extent of the investigations differs a lot and it is difficult to judge, even for the compartments where several investigations are available, whether they cover the spatial and temporal variations.

6.3 Concentrations in foodstuff and the total daily intake

The estimated concentrations of DEHP in foodstuffs, entirely made by EUSES, were much lower than the few available measured concentrations of DEHP in Danish dairy products and crops. Also, the total daily intake of DEHP and DBP appears to be underestimated compared to the measured values, whereas the estimated concentration of DBP in drinking water was in accordance with the single available measured concentration. Comparisons of estimated and measured concentrations of DINP and DIDP were not possible since no measured data are available.

The general conclusion in the present assessment, that EUSES apparently underestimates the concentration in food and the total daily intake of the phthalates, is in agreement with Effting and van Veen (1998) who found that EUSES underestimated the concentration of BBP in food and the total daily intake of BBP in the Netherlands.

Again, as with the PECs, several reasons for the discrepancies can be found. The starting point of the calculations of the concentration in the food is PECs that may already be underestimates (see previous section). Also, the parameters used to estimate the uptake of substances in crops, the bioconcentration factor in fish, and the bioaccumulation in meat and milk can be incorrect and thereby EUSES predicts too low concentrations in food. Furthermore, the measured concentrations, especially in roots, leaves and water, are very limited in number and, as mentioned before, the measured concentration in milk and the total daily intake also include phthalates entering the food during processing, via contact to packaging materials etc. This is not included in the EUSES estimations of the concentrations in food and the total daily intake.

The estimated total daily intake increased when PECs were replaced by measured environmental concentrations. The replacement of estimated concentrations in foodstuffs with the few available measured concentrations resulted in a further increase in the estimated total daily intake. In order to improve the exposure assessment of humans via the Danish environment, there is a need for

further data regarding measured concentrations, especially of phthalate concentrations in various foodstuffs, but also of phthalate concentrations in the various environmental compartments.

In the present assessment, several exposure levels via the environment have been estimated depending on the input data used for the estimations. In the attempt to estimate the combined exposure via environment and via consumer products (Part 3), there is a need to choose relevant exposure levels. The maximum total daily intake for the local scale based solely on EUSES estimations is the highest of all the estimated values. Therefore this has to be considered in the evaluation of the combined exposure.
When the total daily intake is estimated on the basis of measured concentrations in the environment and in the food, a more reliable estimate will be obtained in theory. However, it is uncertain whether the measured concentrations also represent a “high exposure local environment” and for many of the environmental media, the measured data are very scarce. In spite of these limitations, measured concentrations are considered to be more realistic values than estimated values and therefore, the total daily intake estimated on basis of measured concentrations in the environment and in the foodstuffs also has to be taken into account in the combined exposure assessment (Part 3).

PART 2: Exposure via consumer products

7 Introduction

The phthalates are present in a variety of consumer products on the Danish market (see Appendix 7 for a detailed list).

Since the phthalates are not chemically bound in the materials, they may be released from the product during their lifetime. The products are continuously emitting phthalates even though the intensity of emission is not expected to be linear over the product lifetime. New products are expected to release higher amounts of phthalates compared with older products. In the present assessment, no differences in release during ageing of the product will be considered.

Consumer exposure can occur via several pathways, by different routes, and in different situations. The complexity of the consumer exposure to phthalates makes the assessment of the exposure difficult and it is impossible to include all possible pathways and situations. In the present assessment, 6 different scenarios, that are reasonable to occur, will be considered and the exposure from each scenario will be estimated. The 6 scenarios have been chosen on basis of the detailed list of phthalate containing products on the Danish market (Appendix 7) and the consumer exposure scenarios included in the relevant EU-RARs DEHP, 2001; DINP, 2001; DIDP, 2001; DBP, 2001). Not every possible situation will be included and other scenarios could likely also be of interest. Some of them are mentioned in section 14.

The exposure from the following 6 scenarios will be estimated (see also Appendix 8).

- Toys – oral, dermal (DEHP, DINP, DIDP)
- Infant formulae and baby food – oral (All)
- Indoor air – inhalation (All)
- Clothing (gloves, footwear etc.) – dermal (DEHP, DINP, DIDP)
- Paints, adhesives etc. – inhalation, dermal (All)
- Nail polishing (DBP)

Data are not available for all exposure pathways and for those, where data are available, they are in general very limited. A lot of assumptions and generalisations are therefore necessary to take into account in order to quantify the exposure via the given pathways; the exposure estimates should therefore be interpreted with reservations.

The bioavailability is not included in the exposure estimations and therefore, the estimates can only be considered as the external exposure.

8 Exposure via toys

Phthalates have been used as major components in PVC toys and baby equipment as e.g., teethers.
In Denmark, the content of phthalates in toys and other childcare articles in the age group of 0-3 years is regulated by two Statutory Orders of the Danish Ministry of Environment (Miljøministeriets bekendtgørelse nr. 151 af 15/03/1999; Miljøministeriets bekendtgørelse nr. 975 af 27/11/2002). According to the regulation, toys are not allowed to contain more than 0.05% (w/w) phthalates (diesters of *o*-phthalic acid). From investigations of toys on the Danish market (Rastogi and Worsøe, 2001), it is evident that concentrations higher than 0.05% occur (see next section). Furthermore, there is no regulation of the content of phthalates in toys meant for children older than 3 years.
Both the dermal exposure during the child's handling of the toys and especially for the young child, the oral exposure during chewing and biting in the toy are of concern. The physical chewing and the presence of saliva result in the extraction of the phthalates as well as do sweat following dermal contact.
The oral exposure via toys will be estimated based on measured migration rates of DINP (Könemann, 1998) and mouthing times (Groot et al., 1998). A measured dermal absorption rate of DEHP (Deisinger et al., 1998) will be used to estimate the dermal exposure via toys.

8.1 Measured concentrations of phthalates in toys

Rastogi and Worsøe (2001) have measured the concentration of phthalates in toys on the Danish market. In 2001, 20 products of toys meant for children in the age 0-3 years were collected from the Danish retail outlets. The different toys were all assumed to contain phthalates and the collection included dolls, teethers, rattles, animals, books etc. The plastic parts of the products were analysed in duplicates for their content of different phthalates. The analytical method could not separate DINP and DIDP and the content of DIDP was therefore reported as DINP when both substances were present in a product. One or more phthalates were found in 9 out of the 20 products. In 8 of the investigated products, the content was higher than the allowed concentration (0.05%). The measured concentrations are listed in Table 8.1. Different parts of each product were analysed; therefore, the number of samples with DEHP (n=10) exceeds the number of products where phthalates were found.

Table 8.1. The amount of DINP, DIDP, DEHP and DBP in 20 products of toys for children in the age 0-3 years.

	Content (mg/g)			
	DINP/DIDP	DINP	DEHP	DBP
No. of positive samples	7	9	10	1
Mean	430	200	130	1.5
Range	353 – 550	4.1– 361	0.6 – 302	-

Rastogi and Worsøe, (2001)

The results are in agreement with a Nordic investigation of 18 products of toys on the Nordic market (Throne-Holst, 2001). DINP was found in 15 of the 18 products and in 14 products, the concentration was above the allowed concentration in Denmark (0.5 mg/g). The concentrations ranged between 0.4 and 610 mg/g (0.04-61%). DEHP was detected in 12 products at concentrations between 0.03 and 390 mg/g (0.003 - 39 %); in 9 products, it was above the allowed concentration in Denmark (0.5 mg/g).

8.2 Measured migration of phthalates from toys

Several investigations of the migration of phthalates from toys have been performed (Könemann,1998; Rastogi et al. 1997; Steiner et al., 1998; Chen, 1998). The migration studies have been carried out by static or dynamic *in vitro* extraction methods. Also *in vivo* tests have been performed. Human volunteers chewed or sucked on soft PVC samples and the phthalate concentration in collected saliva was determined (Könemann, 1998; Steiner et al., 1998). In general, the recovery from the static *in vitro* methods is lower than from the dynamic *in vitro* methods and the recovery from the dynamic *in vitro* methods is lower than from the more realistic *in vivo* methods of chewing (CSTEE, 1998).
The use of different methods makes it difficult to compare results and also to decide which of the results that best represent the realistic exposure of children. The development and validation of a standard method is very important and there are ongoing discussions of an *in vitro* method based on dynamic extraction in a simulant (JRC, 2001) as a standard method, but a standard method is still not finally adopted. However, when considering exposure of children, it seems reasonable to assume that studies performed in human saliva better represent real-life situations than the *in vitro* tests. Therefore, only the migration of phthalates obtained from *in vivo* studies will be considered in the present exposure assessment.

8.2.1 Measured migrations of phthalates from *in vivo* studies

In the following text, the unit of the measured migration rates will generally be the one used in the original investigation. In Table 8.2 however, the measured migration rates from the different investigations are presented with the same unit (μg/cm^2/h) in order to facilitate a comparison of the results from the various investigations.

In the investigation by Könemann (1998), the release of DINP from PVC samples was measured. Three different specimens (2 toys and one standard PVC disk) were tested in ten different test persons as a minimum. The test persons were both sucking and biting on the specimens. All three specimen had surfaces of 10 cm^2 and they contained 38-43% DINP. The release ranged from 0.3-8.9 μg DINP/minute with an average release for each of the three specimens of 1.38 μg/minute, 2.44 μg/minute, and 1.63 μg/minute, respectively.
Steiner et al. (1998) measured the release of DINP and DEHP from PVC sheets containing approximately 32% DEHP or PVC teethers containing approximately 36% DINP. The average release of DEHP was 793 μg/dm^2 for 3 hours sucking. The average release of DINP after only 1 hour of sucking was 830 μg/dm^2.
Chen (1998) measured the migration of DINP from five different toys. A disk from each toy (10.3 cm^2) was tested in two test persons. Each test person chewed and sucked 4 times 15 minutes on the disk. The maximum migration of DINP into the saliva for any one individual and

time period was 826.48 µg/hour. The average migration rate for all 4 time periods for individuals ranged from 63.22 to 596.64 µg/hour.
It should be mentioned that released phthalates from the toy might be hydrolysed by saliva forming monoesters of released phthalates. These monoesters are not included since only the the parent compounds (diesters) in the saliva have been measured.

Table 8.2. Measured migration of DEHP and DINP from toys.

Study by	Value	Comment	Migration (µg/cm^2/hour)	
			DINP	DEHP
Könemann (1998)	Max.	sucking, chewing	53.4	-
Steiner et al. (1998)	Avg.	only sucking	8.3	2.64
Chen (1998)	Max. avg.	sucking, chewing	59.6	-

8.2.2 Relevance of the migration measurements for toys on the Danish market

The content of DINP in the products investigated by Könemann (1998) and Steiner et al. (1998) and the content of DEHP in the products investigated by Steiner et al. (1998) is at the same level as that found in toys on the Danish market (Rastogi and Worsøe, 2001).
DIDP was also found in the toys on the Danish market (Rastogi and Worsøe, 2001). Even though the exact content of DIDP in the products could not be measured due to the overlapping chromatograms of DINP and DIDP, DIDP seems to be present in toys on the Danish market. It is therefore reasonable to estimate the exposure to DIDP via toys as well. A release rate of DIDP has not been measured and therefore, it will be assumed that because of the physico-chemical similarities between DINP and DIDP, the release of DIDP from toys is similar to the release of DINP from toys.
In general, DBP is not added intentionally to toys and it was only found in a low concentration (0.15%) in one of the investigated toys on the Danish marked. Therefore, in the present exposure assessment, the exposure to DBP via toys will be considered as negligible.

8.3 Measured mouthing times

Groot et al. (1998) investigated the mouthing time of children. The individual differences in mouthing times were large, also within age groups. The maximum mouthing time among the children was approximately 3 hours/day, which was observed for one child in the age group from 6-12 month. This age group also showed the maximum average mouthing time (44 minutes/day) and they spent most of the total time mouthing on toys compared to the younger children (3-6 month) that mainly mouthed their fingers.
In the older age groups, the maximum mouthing time decreased to 53 and 31 minutes/day for the 12-18 month old and 18-36 month old children, respectively.

8.4 Estimated exposure

8.4.1 Oral Exposure

The oral exposure will be estimated as follow:

$$E_{oral} = \frac{m * A_{surface} * t}{bw}$$

E_{oral}: the oral exposure (µg/kg bw/day)
m: the migration rate of phthalate from toys (µg/cm^2/hour)
$A_{surface}$: the surface area of product (cm^2)
t: the duration of exposure (hours/day)
bw: the body weight (kg)

It is assumed that for the youngest children (6-12 month), the duration of exposure is 3 hours/day and for the older children (12-36 month), the duration is 30 minutes/day (based on Groot et al., 1998). According to CSTEE (1998), the body weight of the 6-12 month old children is 8 kg. In the present assessment of the exposure via the environment, the body weight of the 1-7 years old children has also been set to 8 kg in order to included even the smallest children in the assessment. Therefore, in this part of the exposure assessment, 8 kg will be used as the body weight of both the 6-12 month old and the 12-36 month old children. Furthermore, it is assumed that the children are mouthing 10 cm^2 of the toys (as suggested by CSTEE (1998)).

The estimated exposure is presented in Table 8.3. With increasing age of the child, the body weight increases and the mouthing time decreases. Therefore, the oral exposure is highest for the young children (6-12 month) and lower for older children. Since no measured migration rate of DIDP is available, it will be assumed that it is similar to the migration rate of DINP and therefore, the exposure to DIDP equals that of DINP.

Table 8.3. The estimated daily exposure to DEHP and DINP via mouthing on phthalate containing toys based on results from human *in vivo* studies.

Study by	Exposure (µg/kg bw/day)			
	6-12 month		12-36 month	
	DINP	DEHP	DINP	DEHP
Könemann (1998)	200.25	-	33.4	-
Steiner et al. (1998)	31.13	9.91	5.19	1.65
Chen (1998)	223.5	-	36.2	-

[1] Based on the release rate of DINP.
See Table 8.2 for the migration rates used in the estimations.

8.4.2 Dermal exposure

The phthalates can be absorbed through the skin on the hands during handling of the toy, while during mouthing the toy, they can be absorbed through the lips and the skin around the mouth.

The dermal exposure will be estimated as follows:

$$E_{dermal} = \frac{a_{dermal} * A_{contact} * t}{bw}$$

E_{dermal}: the dermal exposure (µg/kg bw/day)
a_{dermal}: the dermal absorption rate (µg/cm^2/hour)
$A_{contact}$: the surface area of contact (cm^2)
t: the duration of exposure (hours/day)
bw: the body weight (kg)

Deisinger et al. (1998) measured a dermal absorption rate of DEHP of 0.24 µg/cm^2/hour (see section 11 about gloves, clothes etc.). According to the EU-RARs of DIDP (2001) and of DINP (2001), a factor of ten is applied to extrapolate from the absorption rate of DEHP to the absorption rate of DIDP and DINP (see section 11). The surface area of contact is assumed to be 100 cm^2 (according to CSTEE (1998)). The duration of dermal exposure is assumed to be the same as the mouthing time (see above) since during mouthing, the children are in dermal contact with the toy. For children from 6-12 month old, the duration of dermal exposure is therefore 3 hours/day and for children from 12-36 month old, it is 0.5 hours/day (Groot et al., 1998). The body weight of both groups of children is set to be 8 kg (see previous section).

The dermal exposure to DEHP for children from 6-12 month old is thus estimated to 9 µg/kg bw/day and for DINP and DIDP, it is 1 µg/kg bw/d. For the older group of children (12-36 month), the estimated dermal exposure to DEHP is 1.5 µg/kg bw/day and for DINP and DIDP, it is 0.15 µg/kg bw/day.

Dermal exposure via handling of toys can also occur for children older than 3 years as e.g., in contact with inflatable toys, modelling clays etc. No duration of contact is known for older age groups and in the present assessment, the exposure of 1-3 years old children will be assumed to represent the exposure to children from 1-7 years old as well.
For the 7-14 years old children, dermal exposure via clothes etc. is estimated in section 11. It will be assumed that this exposure scenario also covers other dermal exposure pathways than clothes as e.g., modelling clay etc. because it is a general scenario where the duration of exposure is set to 2 hours per day and the surface area of contact is the area of the hands, and because no specific parameters are available for a dermal toy exposure scenario for children of this age.

Dermal exposure could also occur from the use of inflatable products like swim rings, beach balls or swimming pools. In Denmark, it is only allowed to use phthalates in swim rings, wings and vests until the 1st of January 2004 (Miljøministeriets bekendtgørelse nr. 975 af 27/11/2002). However, the use of phthalates in other inflatable products like beach balls, inflatable swimming pools etc. made for children older than 3 years is not restricted.

The Danish Environmental Protection Agency has made an investigation of the emission of phthalates to water from different toys, including inflatable products, on the Danish market used by children (MST 2003, unpublished data). The investigated toys contained between 24-31% of total phthalates (w/w), mainly DINP or DEHP. The test duration was 24 hours and the migration to water was tested in duplicates. The emission of phthalates to water at room temperature was below the detection limit for all the investigated phthalates (1 μg/cm^2/24 hours). However, when the samples were incubated at 30°C with UV light corresponding to the amount of UV light on a summer day and the water was rotated, DEHP or DINP could be detected in the water. The maximum measured migration of DEHP was 69.6 μg/cm^2/24 hours from an inflatable swimming pool; the migration of DINP was 55.2 μg/cm^2/24 hours. The maximum measured migration rate of DINP was 124.8 μg/cm^2/24 hours from a dinosaur, which corresponds to a migration of 0.2% of the phthalate in the product (MST, unpublished data). It is likely that the migration of the phthalates occurs mainly in the beginning of the incubation, but whether the migration mainly occurs within 1, 10 or 24 hours is uncertain.
Assuming a swimming pool containing 10 cm of water for 24 hours (or as many hours as it takes to emit the amount emitted during the 24 hours test), the concentration of DEHP in the water will be 0.55 μg/l based on the maximum migration rate from the swimming pool. Since it is a swimming pool, the whole body of the child will be exposed. The dermal absorption rate (0.24 μg/cm^2/hour) of DEHP from PVC film (containing 40.4% DEHP (w/w) (Deisinger et al., 1998), which is used to estimate the uptake via dermal contact to toys in the present assessment, is not expected also to represent the dermal absorption of DEHP via diffusion from water. Therefore, the exposure to phthalates migrated to water will not be further quantified.

8.5 Comparisons with other investigations

8.5.1 Oral exposure

The EU-RAR of DINP (2001) concludes that since migration data from Könemann (1998) are consistent with those of Chen (1998) and that these may be more representative for the European market of toys, the maximum migration rate from Könemann (1998) is the most appropriate one to use to estimate the oral exposure to DINP (200 μg/kg bw/day, children 0-3 years).
Even though *in vivo* measurements of the migration of DEHP are available, the maximum migration rate of DINP from Könemann (1998) is used in the EU-RAR of DEHP (2001). This is because the migration rates of DEHP (Steiner et al. 1998) are obtained only after sucking and not also by chewing. When comparing migration rates of DINP after sucking with those after sucking and chewing, it is obvious that chewing increases the migration of DINP and that could also be very likely for the migration of DEHP.
The maximum migration rate of DINP from Könemann (1998) is also used in the EU-RAR of DIDP (2001). The migration rate of DIDP has been measured *in vitro*, but as the amount of phthalates migrating from PVC measured by static *in vitro* methods is lower than the amount measured by dynamic *in vitro* methods, which again is lower than the amount measured by *in vivo* methods, the migration rate of DINP is used instead.
The estimated exposures to DEHP and DIDP are therefore the same as that estimated for DINP: 200 μg/kg bw/day (DEHP, 2001; DIDP, 2001).
In the present assessment, the same approach has been taken as in the EU-RARs. When the exposure from different scenarios is combined (see Part 3), the estimated exposure of DINP via

toys based on data from Könemann (1998) (see Table 8.3) will be used as the oral exposure estimate for both DEHP, DINP and DIDP.

A fact sheet for the estimation of the exposure via toys has been made (Bremmer and van Veen, 2002). In the fact sheet, basic information has been collected in order to be able to assess the exposure of children to substances in toys. Fact sheets are made to act as a source of data for the users of the program CONSEXPO (CONSumer EXPOsure).
Bremmer and van Veen (2002) has suggested to use a lower migration rate and a shorter mouthing time than in the present assessment for children's exposure via mouthing toys. They suggested to use the highest average migration rate (2.44 μg/10 cm^2/minute) from the study by Könemann (1998) whereas in the present assessment, the maximum measured migration rate (8.9 μg/10 cm^2/minute) from the same study was used in order to estimate worst case exposure. Bremmer and van Veen (2002) suggested a mouthing time based on the same study as in the present assessment (Groot et al., 1998), but instead of the maximum measured total mouthing time (3 hours, including the time spent mouthing non-toys, but not dummies), they calculated a 75 percentile and included only the time spent mouthing toys (in total 74 minutes). Furthermore, they also suggested to use the mouthing time measured for the 1-3 month old children that in general spend less time mouthing than the 6-12 month old children. No matter what category of mouthing objects (toys meant for mouthing, other toys, or non-toys), these could potentially contain phthalates and therefore, in the present assessment, it has been decided to use the total mouthing time for the group of children that spend most time on mouthing.
If exposure via toys is estimated based on the data from Bremmer and van Veen (2002), the exposure to DINP would be 22 μg/kg bw/day; this is almost 10 times lower than the estimate obtained in the present assessment.

8.5.2 Dermal exposure

In the present assessment, the same approach has been taken as in the EU-RARs for the dermal exposure estimates.

In Bremmer and van Veen (2000), exposure parameters from several different dermal exposure scenarios have been suggested. One of the scenarios described the dermal exposure to DINP via contact with the ground sheet of a tent. Bremmer and van Veen (2000) have suggested a contact duration of 3 hours, a frequency of 48 times per year, and an average surface area of contact of 1457 cm^2 (estimated as a combination of the surface area of contact when children are crawling and lying on the ground sheet). An upper limit of leaching of DINP has been suggested based on the measured leaching rate of DINP during mouthing on toy (0.244 μg/cm^2/minute) made by Könemann (1998). The authors considered this value as being not very reliable and pointed out the necessity of determining the rate empirically. If instead, the dermal absorption rate measured by Deisinger et al. (1998) was applied and the estimated average yearly dermal exposure to a child (1-7 years) via contact with the ground sheet of a tent was 1.7 μg/kg bw/day (0.024 μg/cm^2/hour * 1457 cm^2 * 3 hours * (48/365) / 8 kg).
This value is only a bit higher than the estimations made in section 8.4.2 where the information of contact is from CSTEE (1998) and Groot et al. (1998).

9 Exposure via infant formulae and ready-to-use baby food

Phthalates occur as contaminants in infant formulae and ready-to-use baby food. The infant formulae and baby food can be contaminated via the environment or via processing and packaging.
All the phthalates are present in the environment (see Part 1 "Indirect exposure via environment") and therefore, potentially they can also occur in the infant formulae and ready-to-use baby food.

The Danish legislation on food contact materials (Fødevareministeriet, 2002) is essentially an implementation of the EU legislation in this field (EEC, 1989). BBP and DBP appear on the list of authorised substances allowed for lacquered films of regenerated cellulose film, but only in limited amounts (2 and 3 mg per dm^2 of surface area, respectively). The phthalates mentioned in this report do not appear on other lists of authorised or non-authorised substances in the directives. However, the Scientific Committee for Food (SCF) has evaluated the phthalates and the tolerable/temporary tolerable daily intakes per kg body weight for these substances are listed in the so-called "Synoptic document" (EU-Commission, 2003). In practice, these values can be used in the evaluation of phthalate migration from all types of food contact materials taking in consideration the following clause in the legislation: "Materials and articles must be manufactured in compliance with good manufacturing practice so that, under their normal or foreseeable conditions of use, they do not transfer their constituents in quantities, which could endanger human health, etc.".

According to the Danish Plastic Industry (1996) and MST (1999), DEHP is present in food industry hoses (mainly imported) and, according to the EU-RARs (DINP, 2001; DBP, 2001; BBP, Draft report 2002), particularly DEHP, DBP and BBP can be used in food packaging materials.
The exposure via infant formulae and ready-to-use baby food will be estimated based on measurements of the concentrations of DEHP, DBP and BBP in different brands on the Danish market (Petersen and Breindahl, 2000) as well as English investigations of DINP and DIDP (MAFF, 1998).

9.1 The concentration of phthalates in infant formulae and baby food

The concentration of DEHP, DBP, and BBP in different brands of infant formulae and ready-to-use baby food containing fruit, vegetables, cereals and meat has been measured (Petersen and Breindahl, 2000). All the brands were on the Danish market at the time of the investigation. Six out of the 11 brands of baby food and 8 out of the 11 brands of infant formulae contained one or more of the phthalates. In Table 9.1, the highest detected concentration of DEHP and BBP in baby food and infant formulae is given. DBP was only detected in baby food. In infant formulae, the concentration of DBP in all brands was below the detection limit (0.01-0.1 mg DBP/kg wet weight).
In the present assessment of the exposure via infant formulae and baby food, the maximum detected concentration of DEHP, BBP and DBP in baby food and of DEHP and BBP in infant formulae will be used as reasonable worst cases. For DBP in infant formulae, the upper detection limit (0.1 mg DBP/kg wet weight) will be considered as a worst case.

Table 9.1. The maximum measured concentration of DEHP, DBP and BBP in infant formulae and ready-to-use baby food.

Product	No. of products	Max. measured concentration (mg/kg wet weight)		
		DEHP	DBP	BBP
Infant formulae	11	0.06 (5)	-	0.01(2)
Baby food	11	0.63 (3)	0.04 (4)	0.005 (4)

From Petersen and Breindahl (2000).
( ): Number of positive findings.
-: Below the detection limit (upper detection limit: 0.1 mg/kg).

In a MAFF survey, the concentrations of phthalates were measured in a total of 39 individual samples of 14 different brands (powder and ready-to-use) (MAFF, 1998). DEHP was the most abundant phthalate and was found in 23 of the 39 products in concentrations ranging from 0.05 to 0.44 mg/kg in dry powder and of 0.015 mg/kg in ready-to-use formulae. DBP and BBP were detected in 8 of the 39 samples and the concentrations ranged from <0.05 to 0.09 and from <0.003 to 0.015 mg/kg in dry powder, respectively (MAFF, 1998).

When preparing infant formulae, 1 g powder is dissolved in approximately 6.5 ml water (according to the EU-RAR of DINP, 2001). Thus, the estimated maximum concentration of e.g., DEHP in the formula (wet weight (ww)) based on data from the MAFF survey would be (0.44 mg/kg dry powder * 1 kg dry powder / 6.5 kg water = 0.07 mg/kg formula (ww)). This is very close to the maximum measured concentration in the Danish survey (see Table 9.1).

DINP and DIDP were not detected in any of the samples in the MAFF survey (MAFF, 1998). The detection limit for both substances was 0.1 mg/kg dry powder. Due to lack of measured concentrations of DINP and DIDP in infant formulae in Denmark and since the content of DEHP in infant formulae on the English market seems to be consistent with the content of DEHP in infant formulae on the Danish market, the detection limit (0.1 mg/kg dry powder) of DINP and DIDP in the MAFF survey will be used in the present exposure assessment.

9.2 Estimated exposure from infant formulae, baby food and diet

In Denmark, it is recommended by the National Health Service that infants from 0 – 6 month old exclusively drink breast milk, if possible. If breast-feeding is not possible, infant formulae can be used instead (Sundhedstyrelsen, 2002). After the first half year of life, the diet becomes more diverse. In addition to infant formulae or breast milk, they also eat e.g., ready-to-use baby food and the same type of food as adults, but not in the same quantities. Therefore, in the present assessment, the exposure to phthalates via infant formulae and baby food will be estimated for two age groups. One group includes the infants, from 0-6 month old. Since no Danish measurements of the phthalate concentration in breast milk are available, only the exposure via infant formulae will be considered for this age group. The other group includes the infants older

than 6 months. For this age group, the exposure via ready-to-use baby food and via other food (vegetables, meat, cereals etc.) will be considered in addition to the exposure via infant formulae.

9.2.1 Exposure via infant formulae and baby food

The exposure via infant formulae or ready-to-use baby food (Table 9.2) will be estimated as follows:

$$E_{oral} = \frac{C * i}{bw}$$

E_{oral}: the oral exposure (µg/kg bw/day)
C: the concentration of phthalate (µg/kg) in infant formulae or baby food
i: the daily intake of infant formulae or baby food (kg/day)
bw: the body weight (kg)

According to Petersen and Breindahl (2000), the recommended total daily intake of the investigated infant formulae is 600-750 ml. According to the EU-RAR of DINP (2001), the total daily intake of infant formulae is somewhat higher. The intake is depending on the age of infants, but as an average, the daily intake is stated to be 894 ml/day for children 0-6 month old and 900 ml/day for children older than 6 months (DINP, 2001).
In a German survey of the consumption of commercial infant food products, the intake of formulae by 118 infants at the age of 3 months was 47% of the total food intake (805±144 g/day). The intake of breast milk was also 47%. In total, the intake of milk products was 757±135 g/day (Kersting et al., 1998). Assuming a child that is not breast fed and therefore exclusively eats infant formulae, the intake of approximately 900 ml/day for children younger than 6 month (according to the EU-RAR of DINP, 2001) seems to be a realistic worst case.
In the Copenhagen Cohort study, 2-month old not breast fed infants (n=12) consumed 810 g/day of infant formulae (mean value). At the age of 4 month, the average consumption of infant formulae by not breast fed children (n=24) was 783 g/day (Michaelsen, 1997). These data are in agreement with the German study.

According to the German survey, the total intake of milk products (infant formulae and breast milk) by 6 month old infants was 525±93 g/day. For 9 month old children, it was 248±50 g/day (Kersting et al., 1998). Michaelsen (1997) found that not breast fed children at the age of 9 months (n=49) consumed 123±21g/day of infant formulae. Taking the results of these studies into account, a total intake of 900 ml infant formulae per day as suggested in the EU-RAR of DINP (2001) for children older than 6 months seems to be too high. In the present assessment, an intake of 525 g/day from Kersting et al. (1998) will be used as a more realistic worst case.

For DINP and DIDP where the concentrations were measured per kg dry weight (MAFF, 1998), a total daily intake of 140 g dry powder (corresponds to 900 ml (DINP, 2001)) and 80 g dry powder (corresponds to 525 ml) will be used for the children younger than 6 months and the children older than 6 months, respectively.
According to the German survey, the intake of commercial baby food was 181±30 g/day and 207±41 g/day for infants at the age of 6 and 9 months, respectively. The daily intake of baby

food is, according to Petersen and Breindahl (2000), assumed to be one jar weighing from 135-250 g. In the present assessment, a daily intake of 250 g/day will be used as a realistic worst case.

The daily exposure of infants from 0-6 months and infants older than 6 months (and until they stop to eat special infants and baby food) via infant formulae and baby food is presented in Table 9.2.

Table 9.2. Exposure of infants to phthalates via infant formulae and ready-to-use baby food.

	Age group	Body weight (kg)	Daily intake	Exposure (µg/kg bw/day)				
				DEHP	DBP	BBP	DINP	DIDP
Infant formulae	0-6 month	5.5	900 ml/140 g	9.8[1]	16.4[2]	1.6[1]	2.5[3]	2.5[3]
Infant formulae	>6 month	8	525 ml/80 g	3.9[1]	6.6[2]	0.7[1]	1.0[3]	1.0[3]
Baby food	>6 month	8	1 jar, 250 g	19.6[1]	1.25[1]	0.16[1]	-	-
Total	>6 month			23.5	7.85	0.86	1.0	1.0

[1] Based on measured concentrations from Petersen and Breindahl (2000).
[2] Based on the maximum detection limit from Petersen and Breindahl (2000).
[3] Based on the detection limit from MAFF (1998).

9.2.2 Exposure via other food than infant formulae and ready-to-use baby food

In addition to infant formulae, breast milk and ready-to-use baby food, infants older than 6 months also eat meat, vegetables etc. that is included in the assessment of indirect human exposure via the environment (see Part 1). Therefore, estimated exposures via the environment also have to be included when considering infant exposure via food. In the German survey, the children at the age of 9 and 12 months consume 43% and 62% (w/w), respectively, of other food than commercial infant food and breast milk (family (table) food, home prepared infant food etc.) (Kersting et al., 1998).

In the present assessment, it will be assumed that the infants older than 6 month, in addition to the infant formulae and the baby food, also eat half the amount of food as children from 1-6 years (age group used in the assessment of the exposure via the environment). The estimated maximum total daily intake for the local scale is expected to be a worst case estimate and for DEHP, it is at the same level as the maximum measured total daily intake (see section 5.3) (Petersen and Breindal, 2000). Therefore, 50% of the maximum local total daily intake of DBP, BBP, DEHP, DINP and DIDP by children (1-6 years) will be used in order to include the contribution from food other than infant formulae and ready-to-use baby food in the exposure of infants (see combined exposure Part 3 Tables 16.1 – 16.5).

9.3 Comparisons with other investigations

In the EU-RAR of DEHP (2001) and in MAFF (1998), the estimated exposure to DEHP via infant formulae is 13 and 8 µg/kg bw/day for infants 0-3 months old and 6 months old, respectively.
Since the MAFF detection limits of DINP and DIDP are used as concentrations in infant formulae in the present assessment as well as in the EU-RARs of DINP (2001) and of DIDP (2001), the estimated daily exposure to the two substances are the same in both the present assessment and in the EU-RARs (2.5 µg/kg bw/day and 1.8 µg/kg bw/day for infants 0-3 months old and older than 6 months, respectively).
In the EU-RAR of BBP (Draft report 2002), the estimated daily intake of BBP via infant formulae is between 0.1 and 8.7 µg/kg bw/day depending on the age of the infants and the concentration of BBP in infant formulae used to estimate the intake. In the risk characterisation of BBP, 0.15 µg/kg bw/day is used, which is around 10 times lower than in the present assessment. In MAFF (1998), the exposure of new-borns to BBP is estimated to be 0.2 µg BBP/kg bw/day.
Exposure via infant formulae is not included in the EU-RAR of DBP (2001), but the intake via breast milk is estimated to be maximum 6 µg/kg bw/day for infants from 0-3 months old. The estimate of the exposure to DBP via infant formulae for infants 0-6 months old in the present assessment is almost three times higher, but it is likely to be an overestimation since it is based on the highest detection limit of DBP (0.1 mg/kg wet weight). In MAFF (1998), the estimated exposure of new-borns to DBP from infant formulae is 2.4 µg/kg bw/day.

10 Exposure via indoor air and dust

All phthalate containing products contribute to the total phthalate concentration in the air – indoor as well as outdoor. The direct human exposure from products that mainly are used outdoor (e.g., roofing, tarpaulins etc.) is expected to be low and is not considered further in the present assessment. Phthalate containing products (flooring, wall covering, cables, shower curtains, furniture etc.) used indoor will release phthalates to the indoor air. The relatively limited volume of the indoor air compartment can result in a relatively higher exposure to humans than outdoor.

10.1 The concentration of phthalates in consumer products

An investigation on the concentration of DEHP, DBP, BBP, and the sum of DINP and DIDP in PVC products sold on the Danish market has been carried out (Pors and Fuhlendorff, 2001). The chemical analyses were made in duplicates on different types of bags, gloves, vinyl floors, shower curtains, and vinyl tapestry. Mainly DEHP was found in the products, but also DIDP and DINP were found in many products and with contents up to 59% (w/w). In Table 10.1, the measured concentrations for each product group are presented.

Table 10.1. The concentration of phthalates in different PVC products on the Danish market.

Product group	No. of products	Concentration of (g/kg)			
		DEHP	DINP/DIDP	DBP	BBP
Gloves	4	230-420 (3)	585 (1)	-	20-33 (2)
Bags	3	120-210 (3)	110 (1)	-	1.5 (1)
Shower curtains	3	62-230 (3)	85.5 (1)	-	-
Vinyl floors	5	47-160 (3)	6.4-310 (3)	15.5 (1)	0.9-20 (3)
Vinyl tapestry	4	67-100 (2)	230-260 (2)	-	-
Vinyl tiles	2	89.5 (1)	270 (1)	-	-

(): Number of positive samples.
- : Below the detection limit: 0.1 mg/kg for all the phthalates.

10.2 Measured concentrations of phthalates in air

Phthalates are emitted to the indoor air from the above-mentioned products as well as from many others plasticiser containing building- and interior materials. Since the phthalates are substances with a low vapour pressure and a high adsorption coefficient, they are readily adsorbed to particles. Therefore, the estimation of the exposure via indoor air will be based on both measured gas phase concentrations of phthalates emitted from interior materials (Afshari et al., in preparation) and measured concentrations of phthalates in dust from Danish homes and schools (Clausen et al., 2003; Clausen et al., in preparation). To separate the estimations of exposure into the concentration of phthalates in the vapour phase and the amount adsorbed to particles is not correct. The concentration in each phase is dependent on the concentration in the other phase since the phthalates adjust in equilibrium between the phases. However, due to the available data, the concentration of phthalates in the indoor air will in the present assessment be estimated as the phthalates in the vapour phase combined with the phthalates adsorbed to particles.

Several fractions of particulate matter exist. The particulate matter can be sedimented on floors and other horizontal surfaces and will in the following be referred to as “sedimented dust”. The particulate matter can also be suspended in the air (“airborne dust”), but only a fraction of the airborne dust will have the potential to be inhaled. In general, the potential for a particle to be inhaled will be determined by the size of the particle, but it also depends on a lot of other factors such as e.g., electrostatic properties, shape of the particle etc. As a rough guide, particles with aerodynamic diameters below 100 µm have the potential to be inhaled (TGD 2003). This fraction will in the following be referred to as the “inhalable fraction”. Particles with aerodynamic diameters below 5 µm are most likely to settle in the tracheobronchial and pulmonary regions (TGD 2003) and this fraction is referred to as the “respirable fraction”.

10.2.1 The concentration of phthalates in dust

Table 10.2 shows the concentration of phthalates adsorbed to sedimented dust in Danish homes and schools measured by Clausen et al. (2003; in preparation). DEHP is the predominant phthalate adsorbed to dust in the homes. The average amount of DEHP adsorbed to dust in

schools is almost four times higher than in private homes. In the investigation, there was no correlation between the concentration of phthalates adsorbed to dust and the presence or absence of phthalate containing PVC floors.

Other (not Danish) investigations of the amount of phthalates in dust from homes are also presented in Table 10.2. The concentrations of DEHP and BBP adsorbed to dust in the Danish homes are comparable to what has been found in most of the other investigations (Øie et al., 1997; Pöhner et al., 1997; Butte et al., 2001), whereas the amount of DBP is 2-4 times higher in the Danish homes compared with the other investigations (Øie et al., 1997; Butte et al., 2001). In the investigation of house dust in UK, the concentrations of phthalates are lower than in the other investigations. In this investigation, the concentration of DIDP has also been measured (Santillo et al. 2003).

Table 10.2. The concentration of phthalates in sedimented dust (µg/g dust) from Danish schools and homes compared with other investigations.

Country	Comments	No. of samples	DEHP		DBP		BBP		DINP	DIDP
			mean	95th percentile	mean	95th percentile	mean	95th percentile	mean	mean
DK[1]	schools	15	3214	7063	-	-	-	-	-	-
DK[2]	homes	25	858	2595	382	887	121	307	-	-
Norway[3]	homes	38	640		100		110		100	-
Germany[4]	homes	286		2600		370		270	-	-
Germany[5]	homes	272		2000		-		320	-	-
UK[6]	homes	29	192		-		57		49	21

[1] Clausen et al. (2003)
[2] Clausen et al. (in preparation)
[3] Øie et al. (1997)
[4] Butte et al. (2001)
[5] Pöhner et al. (1997)
[6] Santillo et al. (2003)
-: Not analysed
blank: Value not stated in the reference

The concentration of phthalates in sedimented versus inhalable dust

The measurements of the concentration of phthalates in dust in Danish homes and schools have been made on sedimented dust. Sedimented dust could contain particles from phthalate containing material. Thereby, the concentration of phthalates in sedimented dust could be expected to be higher than in airborne and inhalable dust.
Øie et al. (1997) measured both the concentration in sedimented dust and in suspended particulate (PM_{10}) dust. The PM_{10} is 10 µm size particles and therefore, they have the potential to be inhaled. In 6 of the 38 investigated homes (Table 10.2), in addition to sedimented dust, also

PM_{10} was collected and the concentration of phthalates was measured. The average amount of phthalates adsorbed to the PM_{10} was 600 µg DEHP/g dust, 370 µg DBP/g dust, 140 µg BBP/g dust, and 0 µg DINP/g dust. For DEHP and BBP, the concentrations measured in this inhalable dust fraction are very similar to the concentrations measured in sedimented dust (Table 10.2) whereas for DBP, it is almost 4 times higher; DINP was only detected in the sedimented dust. Furthermore, the amount of DEHP and BBP adsorbed to the sedimented dust samples was correlated to the amount adsorbed to PM_{10} whereas for DBP, there was no correlation. The authors suggested that sedimented dust samples are good surrogates for PM_{10} with respect to BBP and DEHP, but not for DBP. Therefore, in the present study, the 95th percentile for the concentration of DEHP and BBP in the sedimented dust measured by Clausen et al. (2003; in preparation) will be used as the concentration in inhalable dust as a realistic worst-case scenario. Regarding DBP, the actual concentration of DBP adsorbed to inhalable dust may be different. Danish data on the concentration of DBP in air (ng/m^3), including DBP adsorbed to airborne dust (Clausen et al., 1999, see section 10.4) are available; however, regarding the concentration of DBP in dust, only data on sedimented dust are available from Denmark.
Therefore, as for DEHP and BBP, the 95th percentile for the concentration of DBP in sedimented dust measured by Clausen et al. (in preparation) will be used. No data are available from Denmark on the concentration of DINP and DIDP in dust. In the present assessment, the concentration of DINP in dust from Øie et al. (1997) will be used, whereas for DIDP, the measured concentration from UK will be used (Santillo et al., 2003).

10.2.2 The amount of dust

Kildesø et al. (1998) measured the amount of dust in a Danish school, in a kindergarten and in an administration building. The measurements were made in two different rooms at each location, they were repeated once a week during 28 weeks, and the sampling time lasted from the morning until the afternoon. According to the authors, the sampling was performed by two different methods in order to measure both the amount of total airborne dust and the respirable fraction. Whether the authors used the same definition of respirable dust as in the present assessment (particle size < 5µm) is uncertain since the aerodynamic diameter of the sampled particles is not mentioned.
The results from the study are presented in Table 10.3.

Table 10.3. The amount of dust in Danish schools.

	Amount of airborne dust[1] (µg/m^3)					
	Schools		Offices		Kindergarten	
	total	respirable	total	respirable	total	respirable
Mean	174	102	63.5	54.4	267	133
Max. value in 95% confidence interval	203	120	75.8	64.6	294	148

[1] Kildesø et al. (1998)

The values should, according to the authors, be realistic estimates of the mean concentration of airborne dust during a school day (Kildesøe et al., 1998), but they also mention that humans may be exposed to higher concentrations than the measured concentration of airborne dust. Due to personal movements, sedimented dust will be resuspended and humans will be exposed to it. Since the sampling of airborne dust was by stationary monitoring and not by personal monitoring, the real exposure to dust may be higher than the measured concentrations of airborne dust (Kildesøe et al., 1998). Clayton et al. (1993) found that the personal exposure (measured with a personal exposure monitor) exceeded the measured airborne level (measured with a stationary indoor monitor) by approximately 50%. Therefore, in the following calculations, the concentration of respirable dust will be set at 50% higher than the measurement concentrations from Kildesøe et al. (1998) in order to include the personal resuspension of sedimented dust.

As already mentioned, the study made by Kildesøe et al. (1998) did not include measurements of airborne dust from private homes, but data exist on the amount of sedimented dust in private homes in Denmark (Gravesen et al., 2002). The amount of sedimented dust in private homes (0.4 g/m^2) was around 2 times higher compared to the amount of sedimented dust in schools (0.16 g/m^2) (Gravesen et al., 2002). Whether this also is the case for the airborne dust or the respirable fraction is unknown. According to Kildesø et al. (1998), there is not necessarily a positive correlation between the amount of dust on surfaces and the amount of airborne dust. In the present assessment, the amount of respirable dust in schools (Kildesøe et al., 1998) will also be used as the amount of respirable dust in private homes.

10.2.3 The vapour-phase concentration of phthalates

The vapour phase concentration of DEHP and DBP, emitted from 9 commonly used products, has been measured (Afshari et al., in preparation). The products included PVC flooring and skirting, electric cables, and floor wax etc. and the emission from the products was measured in test chambers. It was not possible to estimate emission rates of DEHP and DBP from the different products partly due to the low vapour pressure of the phthalates and the high degree of sorption of both DEHP and DBP to the interior surfaces of the chambers.
During 150 days, the vapour phase concentration of DEHP, emitted from the various products, varied between 0.2 and 1.2 $\mu g/m^3$. The lowest concentration was measured in the chamber containing polyolefine flooring with or without wax as well as wallpaper, whereas the highest emission was observed from an el-wire and PVC skirting.
The highest concentration of DBP (20 $\mu g/m^3$) was measured in the test chamber containing polyolefine flooring covered with wax. It reached maximum around day 10 before it declined (Afshari et al., in preparation). Except from this very high emission from polyolefine flooring covered with wax the concentration of DBP in the chambers containing the other products, was lower than for DEHP. This indicates that DBP was a contaminant in the product and not added as plasticiser since an air concentration higher than that of DEHP would be expected due the higher vapour pressure of DBP. The maximum concentration measured was 0.2 μg DBP/m^3 emitted from PVC skirting and polyolefine flooring without wax during the first 60 days of the experiment.

10.3 Estimated exposures of phthalates in air

10.3.1 Exposure via indoor air

In the present study the daily human inhalatory exposure to phthalates in indoor air will be estimated as follow:

$$E_{inh} = \frac{C_{air} * V_{inh} * t}{bw}$$

E_{inh}: the inhalation exposure (µg/kg bw/day)
C_{air}: the concentration of phthalate (µg/m^3) in air
V_{inh}: the inhalation rate (m^3/day)
t: the exposure duration (hours/day)
bw: the body weight (kg).

The assumptions made for each of the above-mentioned parameters are as follow:

bw and V_{inh}:
Three age groups will be considered: Adults, children (1-6 years) and children (7-14 years). The body weights and inhalation rates presented in section 2.1.4 will be used in the estimations.

t:
According to ECETOC (2001), humans spend 22 hours/day indoor in the United Kingdom; it will be assumed this value also applies to both Danish adults and children.

As it is possible, on the basis of the measurements made by Clausen et al. (2003), also to predict the air concentration of DEHP in schools, there is a need to know the daily school hours for children. According to ECETOC (2001), children spend 8 hours/day at school. For Danish schoolchildren, this value is too high as they on average spend 6 hours/day in school. This value will be used in the exposure assessment of DEHP to children (7-14 years old). Consequently, a value of 16 hours/day (22 hours/day - 6 hours/day) will be used as the time children spend indoor in other buildings than school (incl. private residences). The younger children (1-6 years) will not be regarded as school children.

C_{air}:
The concentration of the individual phthalate in air (C_{air}) includes both the vapour-phase concentration and the concentration adsorbed to respirable dust. Therefore, C_{air} will be estimated as follow:

$$C_{air} = C_{vapour} + (C_{dust} * R_{dust})$$

C_{vapour}: the concentration of the phthalate in the vapour phase (µg/m^3)
C_{dust}: the concentration of the phthalate adsorbed to the dust (µg/g)
R_{dust}: the concentration of respirable dust in air (g/m^3).

As mentioned previously, the 95th percentile of the concentration of DEHP, DBP and BBP in the dust (Clausen et al., 2003; Clausen et al., in preparation) will be used as the concentration adsorbed to dust (C_{dust}). For DINP, the estimation of exposure via the inhalation of dust will be made on the basis of the mean concentration of DINP in dust in the Norwegian study (Øie et al., 1997). For DIDP, the mean dust concentration of DIDP measured in UK will be used (Santillo et al., 2003). This last investigation in general showed lower concentrations of phthalates in dust compared to the other investigations.

The maximum value of the 95% confidence interval of respirable dust (Kildesø et al., 1998) multiplied by a factor of 1.5, because personal resuspension of sedimented dust has to be taken into account (Clayton et al., 1993), will be used as the amount of respirable dust (R_{dust}).

The maximum measured value in Afshari et al. (in preparation) will be used as the vapour-phase concentration of DEHP (C_{vapour}); this is probably an overestimation since DEHP will adsorb to surfaces and dust in the indoor environment.
For DBP, two different concentrations will be used in order to estimate both a general exposure via indoor air and a more specific and only occasional occurring exposure via indoor air during and after waxing the floor. For the general exposure scenario, the concentration used is the maximum measured value (0.2 μg/m^3) apart from the very high concentration measured in the chamber containing polyolefine flooring covered with wax. For the specific exposure scenario, the concentrations used is the higher concentration of DBP emitted from polyolefine flooring covered with wax (20 μg/m^3). In PROBAS, DBP is registered in polishing wax etc. (Flyvholm, 2001).

Table 10.4. Estimated indoor air concentrations, C_{air}.

Fraction	Concentration (μg/m^3)						
	DEHP		DBP		BBP	DINP	DIDP
	school	homes	homes	Poly. + wax	homes	homes	homes
Adsorbed to dust, (C_{dust}+R_{dust})	1.27[1]	0.47[2]	0.16[2]	0.16[2]	0.05[2]	0.02[3]	0.004[4]
Vapour-phase, (C_{vapour})	1.2[5]	1.2[5]	0.2[5]	20[6]	(0.05)[7]	(0.02)[7]	(0.004)[7]
Total, C_{air}	2.47	1.67	0.36	20.16	0.1	0.04	0.008

[1] Based on data from Clausen et al. (2003) and Kildesø et al. (1998).
[2] Based on data from Clausen et al. (in preparation) and Kildesø et al. (1998).
[3] Estimated on the basis of a mean concentration of DINP in dust in Øie et al. (1997).
[4] Estimated on the basis of a mean concentration of DIDP in dust in Santillo et al. (2003).
[5] Afshari et al. (in preparation).
[6] The maximum vapour phase concentration obtained by emission from polyolefine flooring covered with wax (Afshari et al., in preparation).
[7] Based on the assumption that the vapour phase concentration is similar to the particulate concentration in air – as seen for DEHP (schools) and for DBP (homes).

For BBP, DINP and DIDP where no measured vapour-phase concentrations are available, it will be assumed that the vapour-phase concentration of these phthalates is similar to the concentration adsorbed to respirable dust in air. This assumption is based on the results reflected in Table 10.4 for DEHP (school) and for DBP (home) where the concentration adsorbed to dust (C_{dust}+R_{dust}) is approximately the same as the vapour-phase concentration (C_{vapour}); this is also in accordance with Øie et al. (1997) (see next section 10.4).

It will be assumed for both adults and children that the concentration of the phthalates in the private homes is more or less similar to the concentration found in many other buildings where they spend time during a day (non-industrial environments apart from the school) and the exposure can therefore be estimated on basis of the measured concentrations in dust at homes.

10.3.2 Exposure via car interior

Phthalates can be present in the air in cars due to emission from phthalate containing car interior. The inhalation exposure via car interior is included in some of the EU-RARs (DEHP, 2001; DINP, 2001; DIDP, 2001).
According to the EU-RAR of DEHP (2001), the air concentration of DEHP in six different cars has been measured at room temperature. In two of these cars, DEHP was detectable. In a new car, the DEHP concentration was 9.6 µg/m^3; at day 20 and at day 40, it was 5.2 and 1.8 µg/m^3, respectively. Because of the scarce data on the concentrations in cars, the car interior exposure estimations in the EU-RAR of DEHP (2001) (and in the EU-RAR of DINP (2001) and the EU-RAR of DIDP (2001)) are based on the same air concentrations of the phthalate as in the exposure estimations via indoor air. The duration of exposure (the time spent in cars) is assumed to be 24 hours minus the hours spend indoor, e.g., 4 hours for adults and 2 hours for children.
In the present assessment, a separate exposure scenario via car interior will not be made. Instead the time spent indoor for both children and adults will be set to 24 hours (for children, 6 of these hours are spent in school) and thereby, the time spent in cars is included. It is a conservative scenario as regards the duration of exposure, since it is assumed that humans are either indoor or in cars, never outdoor.

10.3.3 Estimated exposures

On basis of the above-mentioned assumptions, the estimated human exposures to phthalates in indoor air are presented in Table 10.5.

As mentioned, the estimated exposure based on the emission of DBP from polyolefine flooring covered with wax is an occasionally occurring scenario rather than a general one. It will mainly be relevant for shorter periods of time during application and approximately up to two weeks after application as the concentration of DBP declined 10 days after the application of the wax to the flooring according to Afshari et al. (in preparation). The estimated exposure via polyolefine flooring covered with wax should therefore be interpreted as µg/kg bw/day per application rather than µg/kg bw/day. If an yearly average exposure should be estimated, the frequency of use has to be defined.

Table 10.5. The exposure via inhalation of phthalates in air.

Age group	Exposure estimate(µg/kg bw/day)							
	DEHP			DBP		BBP	DINP	DIDP
	school	home	total[1]	home	poly. + wax	home	home	home
adult	-	0.48	0.48	0.10	5.76	0.03	0.01	0.002
children (1-6 years)	-	1.94	1.94	0.42	23.4	0.12	0.05	0.09
children (7-14 years)	0.31	0.63	0.94	0.18	10.1	0.05	0.02	0.004

[1] The sum of the exposure at school and at home.

10.4 Comparisons with other investigations

According to Clausen et al. (1999), the indoor air concentration of DEHP and DBP (including both vapour phase concentration and the amount adsorbed to particles) in a school, in a kindergarten, and in 4 offices was between 0.11-1.05 µg DEHP/m^3 and between 0.57-1.35 µg DBP/m^3 (see Table 4.5). These measured total concentrations of phthalates in indoor air are at the same level as those estimated in the present assessment (Table 10.4) although the measured values of DEHP are a bit lower than the estimated values and the measured values of DBP are a bit higher.

Øie et al. (1997) found that the exposure to DEHP adsorbed to suspended particulate matter (PM_{10}) was one to three times higher than the vapour phase exposure. In the present assessment, the exposure to DEHP and DBP adsorbed to particles is 0.4 to 1.1 times higher than the vapour phase exposure (except the specific polyolefine flooring covered with wax scenario) (Table 10.4).

The total exposure to airborne DEHP in the present assessment (Table 10.5) is around 10 times less than the estimation in the EU-RAR of DEHP (2001) (4.4 µg/kg bw/day for adults and 22.4 µg/kg bw/day for children). This is mainly due to the worst-case approach in the EU-RAR, where the saturated vapour pressure of DEHP is used to predict the vapour phase concentration (5.3 µg/m^3 at 20ºC) and the amount adsorbed to airborne particles is set to three times that of the vapour phase concentration, in agreement with Øie et al. (1997) (total air concentration 21 µg/m^3).
This approach is also used in the exposure estimations via indoor air to DINP and DIDP (DINP, 2001; DIDP, 2001). The saturated vapour pressure of DINP is 10 µg/m^3, the total concentration in air is therefore 40 µg/m^3, and the estimated exposure for adults is therefore 8.3 µg/kg bw/day. The saturated vapour pressure of DIDP is 5 µg/m^3, the total concentration in air is therefore 20 µg/m^3, and the estimated exposure for adults is 4.2 µg/kg bw/day.

In a combined exposure assessment of DEHP and DBP for the Canadian population, the maximum exposure via indoor air was estimated for children (5-11 years old) for DEHP to 1.2 µg/kg bw/day and for DBP to 1.1 µg/kg bw/day (Chan and Meek, 1994; Meek and Chan, 1994).

These values are quite similar to the exposure via indoor air estimated in the present assessment (1.9µg DEHP/kg bw/day and 0.4 µg DBP/kg bw/day (1-6 years)).

10.5 Other routes of exposure

Exposure to the phthalates following dermal contact with e.g., flooring could be an additional route of exposure, especially for children. Furthermore, the children crawl and play on the floor and thereby they may be more exposed to the phthalates in the sedimented dust than adults. Dermal exposure could also occur during polishing or waxing of the floor or cleaning of interior materials containing phthalates, especially if the cleaning is carried out with hot water and bare hands. Dermal exposure of both adults and children via contact with phthalate containing products will be estimated in section 11.

11 Exposure via gloves, clothes, footwear etc.

As mentioned previously, the phthalates are used in a variety of consumer products that are in contact with the human skin including plastic gloves, water proof clothes, artificial leather in footwear, clothes, seats, and furniture etc. (see Table 10.1 and Appendix 7). DEHP, DINP and DIDP are those phthalates that are registered in some of the above-mentioned products (see Appendix 7) and they are also found in high content in gloves on the Danish market (Table 10.1). It is more uncertain whether DBP and BBP are present in clothes, artificial leather etc. on the Danish market (see Appendix 7). DBP was not found in gloves on the Danish market (Table 10.1) (Pors and Fuhlendorf, 2001). In the same investigation, BBP was detected in 2 of the 5 gloves, but in concentrations much lower than for DEHP and for DINP/DIDP (maximum 3.3% (w/w)). Therefore, only dermal exposure to DEHP, DINP and DIDP, but not to DBP and BBP, via gloves, clothes, footwear etc. will be considered in the present assessment. Dermal exposure to BBP and DBP via gloves is not included in the EU-RAR of DBP (2001) or in the EU-RAR of BBP (Draft report 2002).

The human exposure to the phthalates is a function of several parameters as e.g., the duration of contact, the area of the skin in contact with the plasticiser-containing product, the amount of phthalate at the surface of the product, and the penetration of the phthalate through the skin. In the present exposure assessment, the dermal exposure to DEHP, DINP and DIDP via the mentioned consumer products will be estimated according to the EU-RARs (DEHP, 2001; DINP, 2001; DIDP, 2001) based on a study performed by Deisinger et al. (1998).

11.1 Dermal absorption rate

Deisinger et al. (1998) investigated the absorption of DEHP contained as a plasticiser in PVC-plastic film through rat skin *in vivo*. Sheets of the PVC-film were applied on the shaved back of 8 rats in two separate experiments and the mean dermal absorption rate derived from the study was 0.24 µg DEHP/cm^2/hour. The content of DEHP in the PVC film was 40.4% (w/w).

In the investigation of the content of phthalates in gloves on the Danish market (Pors and Fuhlendorf, 2001), 3 out of 4 gloves contained phthalates in contents between 23-42% (w/w) (Table 10.1). Deisinger et al. (1998) stated that the PVC film was 'heavily plasticised'; nevertheless, the content of DEHP in the PVC films seems to be realistic also for products on the Danish market.

It has been shown that DIDP are ten times less absorbed through the skin than DEHP (Elsisi et al., 1989). Assuming that, because of the physico-chemical similarities between DINP and DIDP, it is also the case for DINP, the dermal absorption rate of DINP and DIDP will be assumed to be 0.024 µg/cm^2/hour.

11.2 Estimated exposure

The dermal exposure will be estimated as follow:

$$E_{dermal} = \frac{a_{dermal} * A_{contact} * t}{bw}$$

E_{dermal}: the dermal exposure (µg/kg bw/day)
a_{dermal}: the dermal absorption rate (µg/cm^2/hour)
$A_{contact}$: the contact surface area (cm^2)
t: the duration of exposure (hours/day)
bw: the body weight (kg)

Assuming, as in the EU-RARs, a situation, where a person (adult) is wearing phthalate-containing gloves (the surface area of hands is 840 cm^2 (TGD 2003)) for 2 hours a day, the dermal exposure to DEHP for an adult would be 5.8 µg/kg bw/day. Similarly, the dermal exposure to DINP and DIDP would be 0.58 µg/kg bw/day.

The dermal exposure to phthalates is also relevant for children even though they normally do not wear PVC gloves. They can be exposed via other products as e.g., waterproof clothes, textile prints etc. and also via contact with other products than clothes (PVC floors, artificial leather on furniture etc.). According to an investigation made by the Danish Environmental Protection Agency, a set of waterproof clothes for children on the Danish marked contained 24% (w/w) of DEHP (MST 2003, unpublished data).
It is difficult to make one specific dermal contact scenario for children because the group of products that children potentially could be in dermal contact with is diverse and none of the products appear to be the one for which contact is more likely than the others. Furthermore, data describing children's contact to specific products are lacking (duration, frequency, and contact surface area). Therefore, the assumptions used to estimate the dermal exposure of adults will also be used to estimate the dermal exposure of children. Thus, the assessment will not cover one specific dermal exposure scenario of children, but instead an average scenario covering several different pathways to dermal exposure of children.

As for adults, it will be assumed that the exposed body surface area of children corresponds to the area of the hands. According to USEPA, but modified by Bremmer and van Veen, 2002, the

total body surface area of a 1.5-year old child is 0.480 m^2 and approximately 5 % of the total body surface of a child (240 cm^2) corresponds to the surface area of the hands of children. For a 6.5-year old child, the total body surface is 0.841 m^2 (USEPA – modified by Bremmer and van Veen, 2002) and 5% of the total body surface is 420 cm^2.
Furthermore, both the duration of the exposure and the dermal absorption rate used to estimate the children exposure will be as for adults (2 hours/day and 0.24 µg/cm^2/hour, respectively). The dermal exposure of children (1-6 years) will therefore be 14.4 µg/kg bw/day for DEHP and 1.4 µg/kg bw/day for DINP and DIDP. The dermal exposure of the older group of children (7-14 years) will be 7.8 µg/kg bw/day for DEHP and 0.78 µg/kg bw/day for DINP and DIDP.

11. 3 Comparisons with other investigations

The present estimates of the dermal exposure to DEHP, DIDP and DINP via gloves etc. of adults are almost identical to the exposure estimated in the EU-RARs of the substances (DEHP, 2001; DINP, 2001; DIDP, 2001) except for the use of a different body weight of an adult. In the EU-RARs, the body weight of an adult is 60 kg, whereas in the present assessment, the average body weight of adults is set at 70 kg and therefore, the dermal exposure to DEHP in the EU-RAR is estimated to be 6.7 µg/kg bw/day (DEHP, 2001) and the dermal exposure to DINP and DIDP is estimated to be 0.67 µg/kg bw/day (DINP, 2001; DIDP, 2001).

The only dermal exposure scenario for children included in the EU-RARs is via toys (DEHP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002), see sections 8.4.2 and 8.5.2.

12 Exposure via use of paints, lacquers, adhesives, sealants etc.

In the PROBAS (Flyvholm, 2001), the five phthalates are recorded in paints, adhesives, fillers etc. and exposure to phthalates during use of these products can therefore be expected. Chemical products have to be labelled if they contain more than 0.5% of substances classified as CMR-substances (carcinogenic, mutagenic and/or toxic to reproduction). DBP and DEHP are classified as toxic to reproduction. It is therefore expected that the use of DEHP and DBP in chemical products will decrease and that other phthalates not classified as CMR-substances (e.g. DINP or DIDP) will be used instead (MST, personal communication, 2003). In glues used in private homes, the phthalates have almost entirely been substituted with other substances (MST, 2000), but according to MST (personal communication, 2003), DBP is still used in glues.

When phthalate-containing paints, fillers etc. are used, the exposure route to phthalates is by inhalation and by dermal absorption due to spilling on fingers etc.

Since no data are available on the emission of phthalates from paints or adhesives, the consumer exposure via paints, adhesives etc. in the present assessment will be estimated by the use of CONSEXPO 3.0 (van Veen, 2001). CONSEXPO 3.0 (CONSumer EXPOsure) is a computer-modelling tool that contains the components contact, exposure and uptake. The contact component specifies the frequency of use, the duration of actual use, and the duration of contact

with the product. The exposure component contains various models to estimate the concentration of the compound in the medium that directly contacts the human body and these estimation models include inhalatory, dermal and oral routes of exposure. The uptake component estimates the amount taken up through the skin, the lungs or the gastrointestinal wall.

In the present assessment, only the exposure from one general scenario representing the exposure via both paints, adhesives, fillers etc. will be estimated. The products differ slightly in the content of phthalates but since they are used in a similar manner and since no specific exposure data exist for any of the products, only one general exposure scenario will be made.

A general exposure fact sheet on paints has been made (Bremmer and van Veen, 2000) where basic information has been collected in order to be able to assess the consumer exposure to substances in paints. Fact sheets are made to act as a source of data for the users of CONSEXPO. In the present assessment, the data from the fact sheet for paints (Bremmer and van Veen, 2000) will be used to assess the inhalatory exposure to phthalates in paints. As mentioned, it will be assumed that an exposure scenario based on data from the fact sheet for paints also is relevant for the exposure during use of lacquers, adhesives, sealants etc.

12.1 Estimated exposure

12.1.1 Inhalatory exposure

CONSEXPO is divided into three parts: the contact, the exposure, and the uptake. In the following text, some of the parameters defining the contact, the exposure, and the uptake are presented. Further details on parameter settings can be found in Appendix 9.

According to Bremmer and van Veen (2000), the parameters defining the contact scenario for paints (solvent rich, brushing) are a use-frequency of 6 times/year, a total contact duration of 3 hours and 18 minutes, and an actual use duration of 3 hours. The exposure is estimated with the "evaporation due to painting" scenario. It is based on a release area (painted area) of 2.5 m^2 and a room volume of 20 m^3.
The uptake by inhalation is estimated with the fraction model assuming 100% absorption and an inhalation rate (as in the other exposure scenarios in the present assessment) of 20 m^3/day for an adult.
According to the EU-RARs (DEHP, 2001; DBP, 2001; DINP, 2001; DIDP, 2001), the average content of phthalates in paints is 5% and in adhesives etc., it is 10%. Therefore, in order to be certain that the exposure via paints, adhesives etc. is not underestimated, an average content of 10% will be assumed in the present assessment.

The outcome of the assessment of the inhalation exposure is given in Table 12.1. The uptake of DBP is more than 100 times higher than the uptake of the other phthalates. This higher exposure is probably due to the higher vapour pressure of DBP than of the other phthalates.
The exposure via use of paints etc. is estimated exclusively for adults since children are not expected to use paint or other of the mentioned products.

Table 12.1. The estimated inhalatory and dermal exposure via use of paint etc. for adults.

Route of exposure		DBP	DEHP	DINP	DIDP	BBP
Inhalation[1]	Mean event concentration, air (μg/m^3)	102	0.36	0.63	0.54	0.42
	Average uptake per event (μg/kg bw/day)	4.0	0.014	0.025	0.021	0.017
	Yearly averaged uptake (μg/kg bw/day)	0.066	$2.3*10^{-4}$	$4.1*10^{-4}$	$3.5*10^{-4}$	$2.7*10^{-4}$
Dermal	Yearly averaged uptake (μg/kg bw/day)	0.034[2]	$3.4*10^{-3;\,2}$	$3.4*10^{-4;\,2}$	$3.4*10^{-4;\,2}$	0.032[1]

[1] Estimated in CONSEXPO
[2] Estimated based on measured dermal absorption rates

12.1.2 Dermal exposure

The dermal exposure via spilling of paints, lacquers, adhesives etc. on fingers will be estimated by two different approaches due to differences in available data on each substance. For DEHP, DINP, DIDP and DBP, the dermal exposure is estimated based on measured dermal absorption rates whereas for BBP, the dermal exposure is estimated by using CONSEXPO.
For DEHP, the dermal absorption rate of 0.24 μg/cm^2/hour measured by Deisinger et al. (1998) will be used. They measured the possible transfer of DEHP to rat skin from PVC plastic film and not paints etc. (section 8.4.2 and 11.1), but it will be assumed that the transfer of DEHP from paints will be similar.
For DINP and DIDP, a factor of ten will be applied to the dermal absorption rate for DEHP measured by Deisinger et al. (1998) resulting in a absorption rate of 0.024 μg/cm^2/hour, as also done in the other dermal exposure estimations (section 8.4.2 and 11.1).
Dermal absorption of DBP, has also been investigated. After dermal exposure to rats, absorption occurred with ca. 60% of the dose being excreted in urine within 7 days. An *in vitro* study with undiluted DBP indicated slower absorption by human skin (2.40 μg/cm^2/hour) than by rat skin (93.35 μg/cm^2/hour) (Scott et al., 1987). In the present assessment, the dermal absorption rate by human skin (2.40 μg/cm^2/hour) will be used.
For BBP, a dermal absorption rate is not reported in the literature, but in an exposure assessment of BBP made by Effting and van Veen (1998), a fitted skin permeability of $3.2*10^{-7}$ cm/minute has been estimated by CONSEXPO. This skin permeability can be used in CONSEXPO to estimate the dermal exposure to BBP (see Appendix 9 for details on parameter setting).

As for the inhalation exposure and according to the fact sheet for paints (Bremmer and van Veen, 2000), an use-frequency of 6 times/year, a total contact duration of 3 hours and 18 minutes, and an actual use duration of 3 hourswill be used in the present assessment for all phthalates. The surface area of contact will be assumed to be 20 cm^2 (Effting and van Veen, 1998) and as for the

inhalation exposure, an average content of 10% of phthalates will be assumed. The estimated dermal exposure via paints, adhesives etc. (yearly averaged) is presented in Table 12.1.

12.2 Comparisons with other investigations

The exposure via adhesives is included in the EU-RAR of DBP (2001). The inhalatory exposure was estimated to be 0.34 µg/kg bw/day by the use of CONSEXPO. The dermal exposure via adhesives is also estimated in the EU-RAR of DBP (2001), but not via direct contact with the adhesives, only via contact with air containing phthalates. In the EU-RAR of DBP (2001), it is concluded that the dermal exposure is low and therefore not taken into account. None of the other EU-RARs (DEHP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002) have included exposure via paints, adhesives or other similar scenarios.

In the human exposure assessment of BBP made by Effting and van Veen (1998), the exposure to BBP via paints and adhesives is assessed by the use of CONSEXPO as in the present assessment. They estimated a yearly averaged inhalatory uptake of BBP of $4.8*10^{-4}$ µg/kg bw/day via use of adhesives and $6.6*10^{-5}$ µg/kg bw/day via use of floor finish. The exposure is at the same level as that estimated in the present assessment ($2.7*10^{-4}$ µg/kg bw/day). Effting and van Veen (1998) used more or less the same parameters defining the scenario as in the present assessment, but they used different physico-chemical properties, especially the vapour pressure of BBP was different.
The estimated averaged dermal uptake of BBP was $4.6*10^{-3}$ µg/kg bw/day via use of adhesives and $3.8*10^{-4}$ µg/kg bw/day via use of floor finish (Effting and van Veen, 1998). This is around ten and 100 times lower than that estimated in the present assessment (0.32 µg/kg bw/day).

13 Exposure via nail polish

American NGOs (Haulihan et al., 2002) investigated the content of phthalates (DEP, DMP, DBP, BBP and DEHP) in 72 different cosmetic products (lotions, fragrance, hair spray etc.) by analysis. Phthalates were found in 52 products ranging from trace amounts up to nearly 3 % of the product formulation. As regards the phthalates in the present assessment, DBP, DEHP and BBP were only found in a few products with relatively low average concentrations (see Table 13.1).

In the American investigation, DBP was mentioned as an ingredient in 16 out of 24 nail polish products (from information on ingredients labels and in patent records) and the stated average concentration was 5% (Haulihan et al., 2002). As the only phthalate of those assessed in the present report, DBP is registered in cosmetic products in PROBAS (Flyvholm, 2001) and according to the EU-RAR of DBP (2001), DBP is mainly present in nail polish.

Recently, it has been decided in EU to restrict the use of CMR-substances (carcinogenic, mutagenic and/or toxic to reproduction) in cosmetics. DBP and DEHP are classified as toxic to reproduction. BBP is not yet classified, but in the EU-RAR of BBP (Draft report 2002), BBP is assessed to have reprotoxic effects.

DINP and DIDP were not included in the American investigation (Haulihan et al., 2002). According to the EU-RAR of DINP (2001), DINP is not assumed to be present in cosmetics.

Table 13.1. The concentration measured in 72 cosmetic products.

	DEHP	DBP	BBP
No of positive findings	3	6	4
Type of products	fragrance	fragrance, hair spray, deodorant	fragrance, hair spray
Average (%)	0.001	0.03	0.0014
Maximum (%)	0.0025	0.08	0.0046

From Haulihan et al. (2002).

The only exposure scenario in relation to the use of phthalates in cosmetic products in the present assessment will be a nail polish exposure scenario for DBP. The exposure route via nail polish is by inhalation as well as by dermal contact with the product and via air (DBP, 2001). The average concentration of DBP in the nail polish is reported to be 5% (Houlihan et al. 2002). Since no measured data are available, CONSEXPO 3.0 will be used to estimate the exposure.

13.1 Estimated exposure

The parameters defining the contact scenario are, according to van Veen (1996) (cited in the EU-RAR of DBP), a use-frequency of twice a week (TGD 1996), the total contact duration is 10 minutes, and the actual use duration is 5 minutes. The exposure is estimated with the "evaporation from mixture" scenario. The release area (nails) is 20 cm^2 and the room volume is 20 m^3.
The uptake by inhalation is estimated with the fraction model assuming 100% absorption and an inhalation rate (as in the other scenarios in the present assessment) of 20 m^3/day for an adult. Further details on parameter settings can be found in Appendix 10.
The outcome of the assessment of the inhalation uptake is an average inhalation exposure per event of 0.006 µg/m^3. The average uptake per event is 1.2 * 10^{-5} µg/kg bw/day of application and a yearly average uptake of 3.4*10^{-6} µg/kg bw/day.

The dermal exposure is expected to be even less as it is mainly dermal exposure via air and the dermal absorption rate is low.

13.2 Comparisons with other investigations

The exposure to DBP via the use of nail polish is also evaluated by the use of CONSEXPO in the EU-RAR of DBP (2001). The average inhalatory exposure per event was estimated to be 0.004 µg/m^3 and the yearly average uptake was estimated to be 2 * 10^{-6} µg/kg bw/day. The estimated exposure in the EU-RAR is a little bit lower than in the present assessment. All the parameters

defining the exposure scenario appear to be similar in the present assessment and in EU-RAR of DBP (2001). Probably the older version of CONSEXPO used in the EU-RAR of DBP (2001) can explain the small difference.

14 Other exposure pathways

Exposure to phthalates via other pathways than the above-mentioned can also occur, but for various reasons the exposure via these pathways will not be quantified in the present report. In the following, some of those pathways will be mentioned.

14.1 Medical devices

According to the Danish Plastic Industry (1996), DEHP and DINP are also used in medical devices mainly in the manufacturing of flexible tubes, containers etc. Not only medical devices but also dental soft-lining materials can contain high amounts of phthalates. The exposure to phthalates via medical devices will not be considered in the present report since it is only a relevant pathway for a sub-population an thus, not a general consumer exposure scenario. Furthermore, due to the widespread use of these articles in variable medical treatments, it is difficult to make a general exposure assessment.

The assessment of exposure via medical devices is included in the EU-RAR of DEHP (2001). Four different situations with exposure from medical products were assessed: long-term haemodialysis, long-term blood transfusion in adults and in children, and neonatal transfusions. The average daily exposure used in the risk characterisation was 3.1, 0.03, 0.075 and 1.7 mg/kg bw/day for the four situations, respectively, and the conclusion drawn was that there is concern for all end-points and a need for limiting the risks (DEHP, 2001).

The American Food and Drug Administration, Centre for Devices and Radiological Health, has made a comprehensive assessment of DEHP released from PVC medical devices (FDA, 2001). Of all the various medical procedures assessed by FDA, the maximum dose of DEHP received by adults (trauma or surgical patients) was 8.5 mg/kg bw/day via blood transfusion. The maximum dose received by neonates was 22.6 mg/kg bw/day via exchange transfusion (FDA, 2001).
If a person is exposed to phthalates via medical devices, this pathway seems to contribute more to the total exposure than any other pathway of exposure.

14.2 Food contact materials

The contamination of food with phthalates can occur during processing, handling, transportation and packaging (including “secondary” storage in food storage articles), see section 9 for further details.

The phthalates monitored in Danish food by Petersen (1999) could be environmental contaminants in the food or they could be contaminants from processing and packaging.

Therefore it is difficult to make an exposure assessment of food contact materials alone. When using measured data of processed and packed food for the exposure assessment, the phthalates migrating from food contact materials to the food is likely to be included. Therefore, in the present assessment, there will be no attempt to make a separate food contact material exposure scenario.

The exposure via food is estimated in Part 1 of the report "indirect exposure via the environment", section 3. The exposure via food estimated exclusively by EUSES is without any contribution from food contact materials. The further exposure estimation in EUSES based on the few measured data of DEHP in food from Denmark includes the exposure via food that could have been in contact with DEHP containing food contact materials. For the other phthalates (DBP, DINP, DIDP and BBP), no exposure estimation is made in EUSES on basis of measured data in foodstuff.

14.3 Modelling clay

The Danish Environmental Protection Agency has made an investigation of the content of phthalates in modelling clay on the Danish market and the emission of phthalates during heating of modelling clay (MST, 2002).
Two traditional modelling clays were investigated for the content of phthalates. One of them contained a negligible amount of an unidentified phthalate whereas the other did not contain phthalates. The total phthalate content in 4 modelling clays for oven hardening ranged from 16-24%. Of the phthalates assessed in the present report, DINP occurred in one modelling clay (9.7% w/w). BBP was also detected in one modelling clay (3.7% w/w), whereas two products contained DBP and DEHP, but in lower concentrations (maximum 0.02% and 0.6% w/w, respectively). DIDP was not present in any of the investigated modelling clays(MST, 2002).
BBP and DINP were the only phthalates that were emitted from clays at 130°C, which is the recommended oven temperature for hardening of the modelling clay. The emission was 3.8 and 0.4-0.5 mg/kg modelling clay, respectively. At 200°C, the emission of BBP and DINP was much higher than at 130°C (1000 and 600 mg/kg modelling clay, respectively) and small amounts of DBP and DEHP were also emitted from some of the products(MST, 2002). So, children using modelling clay can be exposed to phthalates, especially BBP and DINP according to the investigation by the Danish EPA (MST, 2002), during dermal contact with modelling clays and by inhalation during hardening of the modelling clay.

Because no specific data on the phthalate exposure via modelling clays are available, the dermal exposure to phthalates via modelling clays can only be estimated by the use of the dermal absorption rate of 0.24 µg/cm^2/hour measured by Deisinger et al. (1998) and used in the present assessment for the dermal contact to various products (section 11.2) and toys (section 8.4.2). This estimation will, however, not be made in the present report. Instead, it is assumed that the dermal exposure via playing with modelling clays is included in the two other dermal contact scenarios, see sections 8.4.2 and 11.2.

The inhalatory exposure during hardening of the modelling clay in the oven can be estimated as follows.
Assuming that 0.5 kg of modelling clay has to harden in the oven at 130°C, the recommended temperature, 1.9 mg BBP will be released. If this amount distributes evenly in a room (20 m^3),

the concentration will be 0.095 mg/m^3 and the exposure of a child (7-14 years) will be 4.75 μg/kg bw/day (for 24 hours of exposure following hardening). However, the emitted BBP will not distribute evenly in the air since, as mentioned previously, the emitted BBP will primarily adsorb to particles rather than be present in the vapour phase. Furthermore, the duration of the emission will only last approximately half an hour whereas the duration of exposure will be longer since the emitted BBP does not disappear just because the oven is turned off. The exact duration is not known. Bremmer and van Veen (2002) suggested that the frequency of use of modelling clay (traditional) is once a week. If it is assumed that it can be the same use frequency of modelling clay meant for hardening in the oven, the yearly inhalatory exposure can be estimated to 0.7 μg/kg bw/day.

PART 3: Combined exposure, via the environment and consumer products

15 Introduction

The phthalates are used in a wide range of products and because of the diffuse emissions from all the products, humans are likely to be exposed via many different pathways and both oral, inhalatory and dermal exposure occur. Ideally, the combined exposure to a particular phthalate is the sum of all these individual exposure pathways and exposure routes. However, it is uncertain whether all exposure pathways have been identified and not all of the identified pathways can be quantified properly. Furthermore, exposure via the working environment is not included in this report. Therefore, the combined exposure estimated in the present assessment only represents an indication of the exposure level via the environment and via consumer products and thus only represents the total exposure of a human to a particular phthalate partly.

Besides the combined exposure to one particular phthalate as e.g., DEHP, it is also important to consider the combined exposure to all phthalates. Exposure to several phthalates in combination occurs because various products contain more than one phthalate and because humans use and are surrounded by many products concomitantly. The effect of the combined exposure is still uncertain, but the possibility of additive or maybe synergistic effects is present.
In addition to the exposure to those phthalates assessed in the present report, humans are also exposed to other phthalates as e.g. DEP.

16 Estimated combined exposure

The combined exposure to DEHP, DBP, DINP, DIDP and BBP estimated in the present assessment are presented in Tables 16.1 – 16.5, respectively. The combined exposure estimates are obtained from the exposure estimates in the different consumer exposure scenarios combined with the estimated exposures via the environment. It should be kept in mind that the estimated exposure via the environment includes both oral and inhalatory exposure, but the inhalatory exposure only constitutes a minor part of the total daily intake (Table 5.1). Therefore, in Tables 16.1 – 16.5, the exposure via the environment is considered as being oral exposure.
More than one estimate on the exposure via the environment are available for some of the substances as the exposure has been estimated on basis of measured concentrations in the environment (DEHP, DBP, BBP) and food (DEHP) as well as on the regional and local exposure estimations based solely on the EUSES calculations (DEHP, DBP, DINP and DIDP). In Tables 16.1 – 16.5, the estimated maximum local daily intake based on the EUSES calculations is presented together with the exposure estimated on basis of measurements. When estimating the combined total exposure, the maximum local daily intake is used, except for BBP, where the exposure estimations have only been made on basis of measured concentrations of BBP in the environment. The exposure estimates based on these measured concentrations in the environment of DEHP and DBP are 30-40 times lower than the estimated maximum local

exposure of DEHP and DBP. Therefore, the estimated total exposure to BBP is lower than the exposure to any of the other substances.

In Tables 16.1 – 16.5, the combined exposure according to the EU-RARs of each substance has also been included.

The bioavailability of the substances has not been taken into account in the exposure estimates except for the dermal exposure scenarios (toys and clothes etc.) where a measured dermal absorption rate has been used to estimate the exposure. Therefore, the estimated exposure can only be considered as the external exposure for the other routes of exposure.

The present assessment shows that food is the dominant pathway of exposure to the five selected phthalates, no matter how the EUSES estimations were made, i.e., based on PECs, measured environmental data, or measured concentrations in food. For the young children, oral exposure through the mouthing on toys containing DEHP, DINP, and DIDP also plays a significant role. However, due to the present regulation of use of phthalates in toys to young children (up to 3 years old), there seems to be a discontinuously use of phthalates in toys that are supposed to be used by the youngest children and therefore, the mouthing on such toys (rattles, teethers etc.) may not be a significant route of exposure for this age group. Yet, phthalates are not regulated in toys for older children and therefore, the oral exposure through mouthing on toys can still be significant.
Both dermal and inhalation exposure is lower than the oral exposure. In general, the dermal exposure has been estimated to be higher than the inhalation exposure via indoor air except for DBP.

The combined exposure to all the five phthalates assessed in the present report is presented in Table 16.6. In the combined exposure to all phthalates, the maximum local total daily intake has been carried forward from Tables 16.1 – 16.5, except for BBP where no local estimations have been made (Table 16.5). Therefore, the total daily intake estimated for BBP on basis of measured environmental concentrations, has been carried forward instead. For the other substances, the other EUSES estimations of the exposure via the environment (based on measured values) are not carried forward since they are lower than the maximum local total daily intake.
The combined exposure to the five phthalates has been estimated to be between 0.1 mg/kg bw/day and 1 mg/kg bw/day for adults and 6-12-month old children, respectively. For adults, the main exposure is to DEHP and DBP, whereas for the young children, also DINP and DIDP contribute significantly (via toys). In general, the exposure to DBP exceeds the exposure to DEHP due to the higher indirect exposure via the environment to DBP than to DEHP (maximum local total daily intake). As mentioned before, it should be noted that the quantitative data on the tonnage of substances used in Denmark may not represent the situation at present. Especially quantitative data on the use pattern of DBP were scarce and several assumptions had to be made to generate the quantitative data. Also the quantitative data on the use pattern on DINP and DIDP may be encumbered with uncertainty since they are not very recent and the amount used of DINP and DIDP is expected to have increased over the last years. Therefore, the regional tonnage used in the present emission assessment may be too low compared with the currently used tonnage of these two phthalates, which consequently may result in an underestimation of the total daily intake.

Table 16.1. The combined oral, inhalatory and dermal exposure to DEHP via all evaluated pathways for adults and children.

DEHP	Exposure (µg/kg bw/day)											
	Adults			Children (6-12 month)			Children (1-6 years)			Children (7-14 years)		
	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal
Toys	-	-	-	200.3	-	9.0	33.4[8]	-	1.5	-	-	-[9]
Infant form., baby food	-	-	-	23.5	-	-	-	-	-	-	-	-
Indoor air	-	0.5	-	-	(1.9)[6]	-	-	1.9	-	-	0.9	-
Clothes, gloves, etc.[1]	-	-	5.8	-	-	-	-	-	14.4	-	-	7.8
Paints etc.	-	$2.3*10^{-4}$	$3.4*10^{-3}$	-	-	-	-	-	-	-	-	-
Via environment – max local [2,3]	20	-	-	(50)[7]	-	-	100	-	-	40	-	-
Via environment - measured environ. conc.[3]	0.7	-	-	(1.7)[7]	-	-	3.4	-	-	1.4	-	-
Via environment – measured conc. in food[3]	4.5	-	-	(13)[7]	-	-	26	-	-	11	-	-
Measured, max [4]	16	-	-	-	-	-	-	-	-	-	-	-
Total	20[5]	0.5	5.8	273.8	1.9	9.0	133.4	1.9	15.9	40	0.9	7.8
Total EU-RAR	16[5]	5.3	6.7	200	24.4	9.0	85[10]	-	-	-	-	-

[1] A general scenario that potentially could include dermal exposure to a lot of different phthalate containing products.
[2] The highest estimated value of exposure via the environment.
[3] This estimated exposure via the environment also include inhalatory exposure, but it constitutes only a minor part of the total daily intake and the intake is therefore almost only oral.
[4] From Petersen (2000).
[5] The maximum estimated local daily intake is used to estimate the total exposure.
[6] From children 1-6 years (bw = 8 kg; V_{inh} = 9.3 m^3/day).
[7] 50% of the exposure of children (1-6 years) (see section 9.0).
[8] The oral exposure via toys is estimated for 1-3 years old children.
[9] Dermal exposure to toys can also be relevant for 7-14 years old children (e.g., inflatable products, modelling clay etc.), but it is assumed that this is included in the dermal exposure through clothes etc.
[10] Only exposure via environment.

Table 16.2. The combined oral, inhalatory and dermal exposure to DBP via all evaluated pathways for adults and children.

DBP	Exposure (µg/kg bw/d)											
	Adults			Children (6-12 month)			Children (1-6 years)			Children (7-14 years)		
	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal
Infant formulae, baby food	-	-	-	7.9	-	-	-	-	-	-	-	-
Indoor air	-	0.1	-	-	(0.4)[7]	-	-	0.4	-	-	0.18	-
Indoor air, floor wax	-	5.8	-	-	-	-	-	-	-	-	10.1	-
Paints etc.	-	0.07	0.03	-	-	-	-	-	-	-	-	-
Nail polish	-	$3.4*10^{-6}$	-	-	-	-	-	-	-	-	-	-
Via environment -max local[2,3]	60	-	-	(200)[8]	-	-	400	-	-	200	-	-
Via environment – measured environ. conc.[3]	1.6	-	-	(4.0)[8]	-	-	8.0	-	-	3.5	-	-
Measured, max[4]	10	-	-	-	-	-	-	-	-	-	-	-
Total	60[5]	0.2 (6.0)[6]	0.03	208	0.4	-	400	0.4	-	200	0.18 (10.3)[6]	-
Total EU-RAR	92.5[5]	-	-	-	-	-	-	-	-	-	-	-

[1] A general scenario that potentially could include dermal exposure to a lot of different phthalate containing products.
[2] The highest estimated value of exposure via the environment.
[3] This estimated exposure via the environment also include inhalatory exposure, but it constitutes only a minor part of the total daily intake and the intake is therefore almost only oral.
[4] From Petersen (2000).
[5] The maximum estimated local daily intake is used to estimated the total exposure.
[6] The exposure without the floor wax scenario (the exposure inclusive the floor wax scenario).
[7] From children 1-6 years (bw = 8kg; V_{inh} = 9.3 m^2/day).
[8] 50% of the exposure of children (1-6 years) (see section 9.0)

Table 16.3. The combined oral, inhalatory and dermal exposure to DINP via all evaluated pathways for adults and children.

DINP	Exposure (µg/kg bw/d)											
	Adults			Children (6-12 month)			Children (1-6 years)			Children (7-14 years)		
	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal
Toys	-	-	-	200.3	-	1	33.4[8]	-	0.2	-	-	-[9]
Infant formulae, baby food	-	-	-	1.0	-	-	-	-	-	-	-	-
Indoor air	-	0.01	-	-	(0.05)[6]	-	-	0.05	-	-	0.02	-
Clothes, gloves, etc.[1]	-	-	0.6	-	-	-	-	-	1.4	-	-	0.8
Paints etc.	-	$4.1*10^{-4}$	$3.4*10^{-4}$	-	-	-	-	-	-	-	-	-
Via environment – max local[2,3]	5.1	-	-	(15)[7]	-	-	30	-	-	10	-	-
Total	5.1[4]	0.01	0.6	216	0.05	1	63.4	0.05	1.6	10	0.02	0.8
Total EU-RAR[4,5]	20			410 (6 month-3 years)			20 (3-6 years)			20		

[1] A general scenario that potentially could include dermal exposure to a lot of different phthalate containing products.
[2] The highest estimated value of exposure via the environment .
[3] This estimated exposure via the environment also include inhalatory exposure, but it constitutes only a minor part of the total daily intake and the intake is therefore almost only oral.
[4] The maximum estimated local daily intake is used to estimated the total exposure.
[5] The total exposure is the sum of all exposure pathways by all exposure routes and the bioavailability of DINP has been taken into account.
[6] From children 1-6 years (bw = 8kg; V_{inh} =9.3 m^2/day).
[7] 50% of the exposure of children (1-6 years) (see section 9.0).
[8] The oral exposure via toys is estimated for 1-3 years old children.
[9] Dermal exposure to toys can also be relevant for 7-14 years old children (e.g., inflatable products, modelling clay etc.) but it is assumed that this is included in the dermal exposure through clothes etc.

Table 16.4. The combined oral, inhalatory and dermal exposure to DIDP via all evaluated pathways for adults and children.

DIDP	Exposure (µg/kg bw/d)											
	Adults			Children (6-12 month)			Children (1-6 years)			Children (7-14 years)		
	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal
Toys	-	-	-	200.3	-	1	33.4	-	0.2	-	-	-
Infant formulae, baby food	-	-	-	1.0	-	-	-	-	-	-	-	-
Indoor air	-	0.002	-	-	(0.009)[6]	-	-	0.009	-	-	0.004	-
Clothes, gloves, etc.[1]	-	-	0.6	-	-	-	-	-	1.4	-	-	0.8
Paints etc. (during use)	-	$3.5*10^{-4}$	$3.4*10^{-4}$	-	-	-	-	-	-	-	-	-
Via environment – max local[2,3]	2.9	-	-	(10)[3]	-	-	20	-	-	6.8	-	-
Total[4]	2.9	0.002	0.6	210	0.009	1	53.4	0.009	1.6	6.8	0.004	0.8
Total EU-RAR[5]	20			400 (6 month-3 years)			20 (3-6 years)			20		

[1] A general scenario that potentially could include dermal exposure to a lot of different phthalate containing products.
[2] The highest estimated value of exposure via the environment.
[3] This estimated exposure via environment also include inhalatory exposure, but it constitutes only a minor part of the total daily intake and the intake is therefore almost only oral.
[4] The maximum estimated local daily intake is used to estimated the total exposure.
[5] The total exposure is the sum of all exposure pathways by all exposure routes, and the bioavailability of DIDP has been taken into account.
[6] From children 1-6 years (bw = 8kg; V_{inh} = 9.3 m^2/day).
[7] 50% of the exposure of children (1-6 years) (see section 9.0).
[8] The oral exposure via toys is estimated for 1-3 years old children.
[9] Dermal exposure to toys can also be relevant for 7-14 years old children (e.g., inflatable products, modelling clay etc.) but it is assumed that this is included in the dermal exposure through clothes etc.

Table 16.5. The combined oral, inhalatory and dermal exposure to BBP via all evaluated pathways for adults and children.

BBP	Exposure (µg/kg bw/d)											
	Adults			Children (6-12 month)			Children (1-6 years)			Children (7-14 years)		
	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal	oral	inhalation	dermal
Infant formulae, baby food	-	-	-	1.2	-	-	-	-	-	-	-	-
Indoor air	-	0.03	-	-	(0.12)[1]	-	-	0.12	-	-	0.05	-
Paints etc. (during use)	-	$2.7*10^{-4}$	0.03	-	-	-	-	-	-	-	-	-
Via environment - measured environ. conc.	0.97	-	-	(2.9)[2]	-	-	5.9	-	-	2.4	-	-
Total	0.97	0.03	0.03	4.1	0.12	-	5.9	0.12	-	2.4	0.05	-

[1] From children 1-6 years (bw = 8kg; V_{inh} = 9.3 m^2/day).
[2] 50% of the exposure of children (1-6 years) (see section 9.0).

Table 16.6. The total exposure to the five phthalates from all the assessed pathways.

	Exposure (µg/kg bw/day)															
	Adults				Children (6-12 month)				Children (1-6 years)				Children (7-14 years)			
	oral	inhal.	dermal	Total	oral	inhal.	dermal	total	oral	inhal.	dermal	total	oral	inhal.	dermal	total
DEHP	20	0.5	5.8	26.3	274	1.9	9.0	285	133.4	1.9	15.9	151	40	0.9	7.8	48.7
DBP	60	0.2	0.03	60.2	208	0.4	-	208	400	0.4	-	400	200	0.2	-	200
DINP	5.1	0.01	0.6	5.7	217	0.05	1	218	63.4	0.05	1.6	65.1	10	0.02	0.8	10.8
DIDP	2.9	0.002	0.6	3.5	210	0.009	1	211	53.4	0.01	1.6	55.0	6.8	0.004	0.8	7.6
BBP	0.97	0.03	0.03	1.0	4.1	0.12	-	4.2	5.9	0.12	-	6.0	2.4	0.05	-	2.5
All	89.0	0.75	7.1	**96.8**	913	2.5	11	**927**	656	2.5	19.1	**678**	259	1.2	9.4	**280**

17 Comparison of the estimated combined exposure with other investigations

The present assessment shows that food is the dominant pathway of exposure to the five selected phthalates. This finding is in accordance with many other assessments in which food is reported to be the major source of the general population exposure to many of the phthalates (CERHR, 2000 a-e; Chan and Meek, 1994; Meek and Chan, 1994; ATSDR, 2001; ATSDR, 2002). However, most of these references do not quantify the exposure. It is also in accordance with the EURARs of DINP, DIDP and DEHP where the exposure to adults via the environment (almost entirely via food) contributes most to the total exposure (DINP, 2001; DIDP, 2001; DEHP, 2001).

According to ATSDR (2001), some dairy products, e.g., fish and seafoods, can be the major source of exposure to DBP if these foods comprise a large part of the diet. If they are not important sources in the diet, ambient air is likely to be the most important source of exposure to DBP. In the present assessment, no separate estimations has been made for the exposure via ambient air as it is included in the total exposure via the environment, but as already mentioned previously, ambient air does not seems to play any significant role in the total exposure in the present EUSES estimations of the exposure via the environment (see Table 5.1).

It has been reported that the second most important source of exposure to phthalates is indoor air (CERHR, 2000 a-e; Chan and Meek, 1994; Meek and Chan, 1994; ATSDR, 2001). Also according to the EURARs of DINP and DIDP, the exposure via indoor air is the second most dominating pathway (DINP, 2001; DIDP, 2001). In the present assessment, the exposure via dermal contact with phthalate containing products is in general estimated to be higher than the inhalatory exposure via indoor air. This finding is in accordance with the EURAR of DEHP (2001) where the dermal exposure via contact to gloves etc., in contrast to the EURAR of DINP and DIDP, is higher than the exposure via inhalation of indoor air.

For young children, the present assessment has shown that the estimated oral exposure through the mouthing on toys containing DEHP, DINP, and DIDP also seems to contribute significantly to the estimated total exposure; even more than the exposure via the environment and indoor air. In the EURARs of DEHP, DINP and DIDP, in which the exposure of children via toys has been estimated, the oral exposure via toys constitutes quantitatively the most important pathway (DEHP, 2001; DINP, 2001; DIDP, 2001).

Effting and van Veen (1998) estimated a combined environmental and residential exposure to BBP by the use of EUSES and CONSEXPO. The average total daily uptake for adults was 1.1 μg/kg bw/day (worst case: 7.5 μg/kg bw/day). This is very close to the estimated combined exposure to BBP in the present assessment (1.0 μg/kg bw/day). In the combined exposure, Effting and van Veen (1998) assessed the environmental exposure including ambient air as well as the residential exposure from vinyl products (indoor air) and different consumer products (adhesives, paints and hair spray). The authors concluded that the estimated exposure from the various sources as well as the various routes of exposure was at the same order of magnitude and therefore, the exposure to BBP does not seem to be dominated by one specific source or single route of exposure. In the present assessment, the exposure via the environment contributes

significantly to the total combined exposure compared to the exposure via indoor air and via the use of paints, lacquers etc.

Recently another approach to investigate the exposure to phthalates than estimating the exposure via measured concentration in several media has been applied (Blount et al. 2000). Phthalate monoester metabolites were quantified in urine sampled during 1988-1994 from 289 adult humans. The monoesters with the highest urinary level were monoethyl phthalate, monobutyl phthalate, and monobenzyl phthalate reflecting exposure to diethyl phthalate, dibutyl phthalate, and benzyl butyl phthalate, respectively. The concentrations of mono 2-ethylhexyl phthalate (MEHP) and mono isononyl phthalate (MINP) were lower. This could indicate low exposure to these substances but it could also be explained by differences in storage, metabolisms or excretion of these two substances compared to e.g., DBP and BBP. Furthermore, since DINP is a mixture of several isomers, also several urinary metabolite isomers are expected to be excreted. Only one DINP monoester metabolite isomer was included in the measurements and therefore, the internal dose of DINP may be underestimated.

The urinary levels of phthalate monoester metabolites measured by Blount et al. (2000) have been used to estimate the exposure to the corresponding diester phthalates (David, 2000; Kohn et al., 2000). The 95 percentile of the estimated total daily intake of DEHP was 3.1 μg/kg bw/day, of DBP 6.9 μg/kg bw/day, of BBP 3.3 μg/kg bw/day, and of DINP 1.1 μg/kg bw/day. These estimated intakes include all possible pathways and routes of exposure.
In the present assessment, the estimated total exposure to DEHP and DBP, including all pathways and routes of exposure, is a lot higher (26.3 and 60.2 μg/kg bw/day, respectively). As already mentioned, these high figures are mainly due to the fact that in the estimation of the total exposure, the maximum local daily intake estimated by EUSES was used as the exposure via the environment. There is no information in Blount et al. (2000) on the exposure level of the test-persons, so it is uncertain whether they resemble adults exposed at a local level in the EUSES (adults living close to point sources) or rather adults exposed at a regional level. If the exposure via the environment used in the combined exposure is based on the measured environmental concentrations instead of the maximum local daily intake, the total exposure to DEHP and DBP is 7 and 1.8 μg/kg bw/day, respectively. This is much closer to the estimated exposure made by David (2000) and Kohn et al. (2000) based on the urinary monoester metabolites. For both DINP and BBP, the exposure estimated by David (2000) and Kohn et al. (2000) is at the same level as in the present assessment (5.7 and 1.0 μg/kg bw/day, respectively).

CPC (2003) has also measured the urine levels of phthalate monoester metabolites in persons aged 6 years and older. In general, the levels of excreted monoester metabolites (of DEHP, DBP, BBP, DINP) were found to be higher in 6-11 years old children compared to that in the older age groups (12-19 years old; >19 years old). The urinary monoester metabolites are only a fraction of the totally excreted urinary phthalate metabolites, which again are only a fraction of the internal dose as excretion also takes place via faeces. Therefore, it is unknown whether the differences in the urine level of monoester metabolites between age groups reflect differences in exposure between age groups or rather reflect differences in metabolism between age groups.
The present assessment, where the estimated exposure to DEHP, DBP BBP and DINP was higher for children than for adults, supports the likeliness of children being more exposed to phthalates than adults are.

PART 4: Discussion and conclusions

18 Indirect exposure via the environment

The indirect exposure via the environment has been assessed by the use of EUSES. Instead of the standard default input parameters representing a standard region in EU and used in the EU-RARs, a set of input parameters specific for Denmark has been used. Based on the specific Danish profile, the Danish tonnages of the substances and specific usages, and substance specific information as given in the EU-RARs of each phthalate, predicted environmental concentrations (PECs) and exposures have been estimated. Measured concentrations in the Danish environment and in Danish food obtained by monitoring studies have been compared with the estimated concentrations and exposures. For a refinement of the EUSES exposure estimations, monitoring data from Denmark have also been used as input data in EUSES. Furthermore, it has been evaluated whether the exposure assessments in the EU-RARs of the phthalates apply for Denmark as well.

Unfortunately, there are only few measured data on the concentrations of DINP in the Danish environment and to our knowledge, no data on the concentrations of DIDP. Therefore, the comparison between measured and estimated concentrations has mainly been possible for DEHP and DBP. Monitoring data on BBP are also available for some compartments, but since the exposure to BBP via the environment was not assessed in the EUSES, no comparison has been made between measured and estimated concentrations.

18.1 The environment

18.1.1 Soil

The regional PECs of DEHP and DBP (8.0 and 4.4 µg/kg dry weight (dw), respectively) are within the broad range of measured concentrations (1-54 and 1-21 µg/kg dw, respectively) in soil receiving no sludge, but at the lower end of the range. Also for DINP, the regional PEC (1.1 µg/kg dw) seems to be a slight underestimation of the actual background level of DINP in Danish soils since it is similar to the lowest measured value (1-35 µg/kg dw).

The local PECs of DEHP, DBP and DINP in agricultural soils and the concentrations measured in heavily sludge amended soils are in the same range, with maximum local PECs being a bit higher than maximum measured concentrations in heavily sludge amended soils.

There are no measured data of DIDP in soil, whereas measurements of BBP in soil are available.

18.1.2 Sediment

For DEHP, the regional PEC in fresh water sediment (3,480 µg/kg dw) is above the measured concentration in sediment from a lake with no direct sources (310 µg/kg dw) and also above the concentrations found in sediment from lakes and streams in the Roskilde region (maximum: 124

µg/kg dw); however, no information about the extent of exposure has been given. The regional PEC in sediment is at the same level as the average concentration measured in sediment from lakes and streams in Århus Amt receiving wastewater outlet (2,335, 2,928 and 5,923 µg/kg dw), but the range of these concentrations are very broad and relatively few of the concentrations are that high.
It is more or less the same picture for DBP, where the regional PEC for DBP (634 µg/kg dw) in sediment is at the same level as measurements from Århus Amt and higher than in the lake with no direct sources (<100 µg/kg dw).

For both DEHP and DBP, the maximum local PEC (77,800 and 9,270 µg/kg dw, respectively) in sediment is around 10 times higher than the maximum measured value (6,900 and 630, respectively). However, there is no information whether any DEHP/DBP consuming industry is situated close to any of the sample sites and thus, the sample sites where the maximum concentration has been measured are not necessarily representative for the local exposure levels of the substances.

There are no measured data of DINP and DIDP in fresh water and fresh water sediment; only for marine water and sediment, but the marine compartment is not included in the EUSES models. There are measurements of BBP in fresh water and fresh water sediment.

18.1.3 Surface and groundwater

The regional PEC for DEHP in surface water (296 µg/m^3 (total)) is in the middle of the range of average measured concentrations (90-710 µg/m^3), and therefore seems to be a reasonable estimate.
The regional PEC for DBP (1,160 µg/m^3 (total)) is much higher than the few available measured concentrations of DBP in fresh water (10-100 µg/m^3). In the investigation where the highest value was found, the concentration of DBP in the three other investigated streams was below the detection limit (20 µg/m^3).

The local PECs in surface water exceed the measured concentrations for the specific substances, but the sample sites are not referred to as representative for local exposure levels.

There are no measured concentrations of DINP and DIDP in surface water. Measurements of BBP in surface water are available.

The estimated regional concentration of DBP in groundwater (34.5 µg/m^3) is 10 times lower than the measured median concentration (380 µg/m^3) in the upper groundwater from the monitoring stations in the agricultural catchment areas and it is 35 times lower than the concentration found in the groundwater monitoring areas (median: 1,200 µg/m^3). Even though the concentration of DBP was below the detection limit in a lot of the samples from monitoring stations, the estimated regional concentrations is still an underestimation of a realistic worst-case groundwater concentration.
In EUSES, the concentration in the groundwater is actually the concentration in the pore-water of the agricultural soil. This is supposed to be a worst-case assumption since transformation and dilution in deeper soil layers is neglected. At least in the case of DBP, it is not a worst-case estimation.

The estimated maximum concentration in groundwater for the local scenario of the formulation of adhesives etc. is higher than the measured maximum concentration, but there is no information on the extent of exposure of the groundwater monitoring stations and it is therefore uncertain whether the measured concentrations are representative for a regional or a local exposure level.

There are no measured concentrations of DEHP, BBP, DINP and DIDP in groundwater.

18.1.4 Air

There are no measurements of the ambient air concentration of phthalates in Denmark, but Løkke and Rasmussen (1983) made estimations of the ambient air concentration based on measured deposition of DEHP and DBP. These estimations for DEHP (22 ng/m^3) and DBP (9 ng/m^3) are around 4 and 6 times higher than the EUSES estimated regional air concentrations (5.3 ng/m^3 and 1.5 ng/m^3, respectively).

Measured indoor air concentrations are available, but they are not comparable with the estimated concentrations in ambient air.

18.1.5 Deposition

EUSES predicts only the local deposition of phthalates close to point sources (1000 m). Therefore, only the concentrations of DEHP and DBP measured in samples taken 150 m from a plasticiser plant are comparable with the estimated concentrations. The highest estimated local deposition rate of DBP (4.4 µg/m^2/day) is at the same level as the measured value (2.2 µg/m^2/day), whereas the highest estimated local deposition of DEHP (98.9 µg/m^2/day) is around 20 times higher than the measured value (4.7 µg/m^2/day).
The other measurements of the deposition of DEHP and DBP and the measurements of DINP have been performed from locations more comparable to the regional scale; these are within the very wide range of estimated local deposition rates.

There are also measurements of the deposition of BBP, but no measurements of the deposition of DIDP.

18.1.6 Sludge and wastewater

As for deposition, EUSES only estimates the concentration of substances in sludge and wastewater for the local scenarios and does not estimate a regional value.
Again, as for deposition, the ranges of estimated local concentrations in sludge are so wide that it is no surprise that the measured values are within the range of predicted local concentration. Furthermore, there is no prediction of the concentration in purely industrial sludge and there are no predictions from a local municipal wastewater scenario to compare with.

The estimated fate of the substances in the regional STP can be compared with the measured fractions of substance that are directed to air and to water, or are degraded. There seems to be at least some accordance between the measured and the estimated fate of the substances in the STP

even though there are differences. The measured data show that DEHP to a higher extent is directed to the sludge compared with DBP, whereas the fraction of DBP that is degraded in the STP reaches higher levels than the fraction of degraded DEHP. The single investigation of DINP shows a degradation rate as high as for DBP (up to 90%) whereas the estimated degraded fraction of DINP is much lower (10%), instead DINP is mainly directed to the sludge (84%).

18.1.7 The Danish country profile

The use of a specific Danish profile only changed the exposure assessment of DEHP slightly (see section 3.3.2). The conclusion that the use of specific regional parameters in EUSES instead of the EUSES defaults values only lead to small improvements of the predicted environmental concentrations (PECs) is in concordance with the results obtained in other studies (Schwartz et al., 2000; Berding et al., 2000), but the degree of improvement of the predicted environmental concentrations (PECs) also depends on how similar the specific region and the standard region are (Berding et al., 2002).

18.1.8 Environmental concentrations - conclusions

In general, the regional PECs of DEHP and DBP in many of the environmental compartments were within the range of available measured concentrations. However, the PECs were not always worst-case concentrations, e.g., in soil, the estimated concentration was at the lower end of the range of measured values. Only for DBP in surface water did the estimated concentration exceed all measured concentrations. Comparisons of estimated and measured concentrations of DINP were only possible for the soil compartment since for all other compartments, no measured data were available.
No measured concentrations of DIDP in the environment were available. Measured concentrations of BBP were available, but no EUSES estimates were made.

The reasons why the intended conservative EUSES does not predict worst-case concentrations for the phthalates in the present assessment as expected could be poor data input into the model, unrepresentative measurements, or the models in EUSES could be inappropriate to predict concentrations of phthalates.
The uncertainty of prediction of the physico-chemical properties and also of the degradation parameters of the phthalates is well known. Also, data on the emission are old and limited in number and therefore, several assumptions have been made concerning the regional tonnage and the emissions. Furthermore, due to lack of data, the emission via waste has not been included, thereby underestimating the true emission.
It is generally expected that the EUSES model will overestimate the exposure. Berding et al. (2000) included DEHP in a study where estimated and measured concentrations were compared. In general, the estimated concentrations, especially if they were estimated by the standard EUSES, were overestimated. Also, by the use of more precise parameters (including regional specific parameters, measured degradation parameters etc.), the predicted concentrations, in general, were somewhat higher than measured concentrations. Only for the concentration in soil, an underestimation was observed.
Regarding the reliability of the measurements there is an agreement between the results obtained in different investigations for some compartments (e.g. soil), whereas for other compartments (e.g. the sediment), there are big differences between the results from the various investigations.

But, as already mentioned, the extent of the investigations differs a lot and it is difficult to judge, even for the compartments where several investigations are available, whether they cover the spatial and temporal variations.

18.2 Foodstuffs and drinking water

18.2.1 Dairy products

In the few milk samples where DEHP has been detected, the concentration was much higher than what was estimated as the regional concentration in cow's milk by EUSES (0.05-1.4 mg/kg wet weight (ww) compared to $7.2*10^{-6}$ mg/kg ww). Even in the local scenario, where the highest concentration of DEHP was predicted in the milk, the estimated concentration was only 0.0047 mg/kg ww. The measured concentration could be high because of contamination with phthalates during processing in the dairy and that is not included in EUSES predictions. The estimated concentration could be low because of an underestimation of the amount excreted in the milk.

There are no measured concentrations of DBP, DINP, DIDP and BBP in dairy products in Denmark.

18.2.2 Vegetables

Measured concentrations of DEHP in crops are only available from one investigation where barley and carrots were grown in heavily sludge amended soil. The measured concentration in both carrots and barley exceeded the regional estimated concentrations (0.2-6.3 mg/kg dw compared to 0.002 (leaves) / 0.007 (roots) mg/kg ww). The estimated maximum concentration at local exposure levels (1.2 (leaves) / 3.8 (roots) mg/kg ww) is at the same level as the measured values.

There are no measured concentrations of DBP, DINP, DIDP and BBP in crops in Denmark .

18.2.3 Fish

No Danish measurements on the concentration of phthalates in fish are available.

The estimated regional concentration of DEHP in fish (191 µg/kg ww) seems to be reasonable, but not worst case, compared to measured concentrations of DEHP is fish (between a few µg/kg and 19,000 µg/kg ww) from other European countries.
The estimated concentration of DINP and DIDP (10 µg/kg ww and 3 µg/kg ww, respectively) is at or below the detection limit (10 µg/kg ww) for the measured concentrations.

18.2.4 Meat

No Danish measurements on the concentration of phthalates in meat are available. To our knowledge, no investigations from other northern European countries are available, that could be used instead for the comparison with estimated concentrations in meat.

18.2.5 Tap water

The measured concentration of DBP in Danish tap water (160 µg/m^3) is lower than the estimated regional concentration in drinking water (574 µg/m^3), whereas for DEHP, the measured concentration is much higher than the estimated regional concentration (11,000 µg/m^3 compared with 56.9 µg/m^3). Only one single measurement has been made and it is doubtful whether one investigated water sample represents the drinking water in Denmark.

There are no measured concentrations of DINP and DIDP in Danish tap water, whereas for BBP one measurement is available.

18.2.6 Concentrations in foodstuffs and drinking water - conclusions

Very few measurements have been made on the concentration of phthalates in food and drinking water in Denmark. The estimated concentrations of DEHP in foodstuffs, entirely made by EUSES, were much lower than the few available measured concentrations of DEHP in Danish dairy products, and crops.
Also, the only available measured concentration of DBP in drinking water was lower than the estimated concentration whereas for DEHP, the measured concentration was much higher than the estimated one.
No measured data are available on the concentrations of BBP, DINP and DIDP except for one measured concentration of BBP in drinking water.

The general conclusion in the present assessment, that EUSES apparently underestimates the concentration in food of the phthalates, is in agreement with Effting and van Veen (1998) who found that EUSES underestimated the concentration of BBP in food in the Netherlands.

Again, as with the PECs, several reasons for the discrepancies can be found. The starting point of the calculations of the concentration in the food is PECs that may already be underestimates. Also, the parameters used to estimate the uptake of substances in crops, the bioconcentration factor in fish, and the bioaccumulation in meat and milk can be incorrect and thereby, EUSES predicts too low concentrations in food. Furthermore, the measured concentrations, especially in roots, leaves and water, are very limited in number and, as mentioned before, the measured concentration in milk and the total daily intake also include phthalates entering the food during processing, via contact to packaging materials etc. This is not included in the EUSES estimations of the concentrations in food.

18.3 The total daily intake

The regional estimated total daily intakes by adults of DEHP (0.19 µg/kg bw/day) and DBP (0.1 µg/kg bw/day) are approximately 20 and 40 times lower, respectively, than the mean daily intake calculated based on measured concentration in meals (4 µg/kg bw/day for both DEHP and DBP) (Petersen, 1999). The highest estimated local daily intake of DEHP (20 µg/kg bw/day) and DBP (60 µg/kg bw/day) are at the same level as the maximum daily intake calculated based on measured concentration in meals (16 µg/kg bw/day and 10 µg/kg bw/day, respectively).
The investigated meals that are the basis for the calculations of daily intakes (Petersen, 1999) may also contain phthalates that have migrated into the food during processing, packaging etc. These sources of phthalates in food are not included in the EUSES estimations, which may therefore give a lower estimate of the total daily intake by man from exposure via the environment.

For all four substances, the fraction of daily intake via leave - and root vegetables is high. For DEHP, the intake from fish is also a major source of exposure whereas for DBP, water contributes significantly. For DINP and DIDP, almost all the sources contribute significantly to the total daily intake.

18.3.1 Refined estimations of the total daily intake

Refined estimations of the regional total daily intake have been performed in EUSES based on the available measured concentrations of phthalates in the Danish environment. Since there are very few measurements for DINP and DIDP, the refined estimations were only performed for DEHP, DBP and BBP. BBP was not included in the previous EUSES predictions, but data on BBP in the Danish environment are available and therefore, a total daily intake of BBP was estimated on the basis of these measured environmental data.
For DEHP, a second refined estimation of the regional total daily intake has also been performed in EUSES based on measured concentrations in the various foodstuffs.
Table 5.3 summarises the results of the refined estimations of the total daily intake.

The total daily intake of DEHP by adults and children increased around three times (from 0.19 to 0.71 µg/kg bw/day for adults; from 0.87 to 3.4 µg/kg bw/day for children 1-6 years old; and from 0.38 to 1.4 µg/kg bw/day for children 7-14 years old) and the total daily intake of DBP increased 16 times (from 0.1 to 1.6 µg/kg bw/day for adults; from 0.49 to 8.0 µg/kg bw/day for children 1-6 years old; and from 0.21 to 3.5 µg/kg bw/day for children 7-14 years old) when the measured environmental concentrations were used as input data in EUSES to estimate the concentration in food instead of the predicted regional environmental concentrations (PECs) originally estimated by EUSES.
The total daily intake of BBP based on the measured concentrations of BBP in the environment was very similar to the estimated total daily intake of DBP.

The total daily intake of DEHP increased around 30 times (from 0.19 to 4.5 µg/kg bw/day for adults; from 0.87 to 26 µg/kg bw/day for children 1-6 years old; and from 0.38 to 11 µg/kg bw/day for children 7-14 years old) when measured data on the concentration of DEHP in fish, milk, crops and drinking water were used as input data to estimate the daily intake compared with the original regional EUSES estimation.

This refined estimated total daily intake of DEHP by adults (4.5 µg/kg bw/day) is similar to the measured mean daily intake (2.7-4.3 µg/kg bw/day) (Petersen, 1999) (Table 5.3), where the intake via inhalation and drinking water was not included; the estimated daily intake via inhalation is not a major route of exposure in the present EUSES estimations.

18.3.2 Total daily intake from the EU-RARs of DEHP, DBP, DINP and DIDP, and from other investigations

Concerning DEHP, the regional total daily intake in the EU-RAR is between what has been estimated on the basis of the measured concentrations in the environment and the estimation based on measured concentrations in foodstuff. The maximum daily intake from the local scenarios is very similar in the EU-RAR of DEHP (2001) and in the present assessment. For exact values, see Table 5.3.
As in the present assessment, the predominant contribution to the total daily intake of DEHP for adults as estimated in the EU-RAR of DEHP (2001) is from fish (52%) and root crops (18%), but also from dairy products (24%).

For DBP, the estimated regional total daily intake in the present assessment is at the same level as the regional total daily intake estimated in the EU-RAR of DBP (2001), whereas the only refined estimation of the regional total daily intake of DBP based on the measured concentrations in the environment is somewhat higher; for exact values, see Table 5.3.

According to the EUSES estimations made in the EURAR of DINP (2001), fish and root crops are the dominating exposure pathways for DINP. In the present study, they are also important pathways, but together with several other pathways.
For DINP and DIDP, the maximum local total daily intake estimated in the EU-RARs (DINP, 2001; DIDP, 2001) is around ten times higher than the maximum local total daily intake estimated for Denmark in the present assessment. The regional daily intake of DINP and DIDP is approximately 100 times higher than the regional daily intake estimated for Denmark. For exact values, see Table 5.3.
The amount of DINP and DIDP is expected to have increased over the last years. Therefore, the regional tonnage used in the present emission assessments may be too low compared with the currently used tonnage of these two phthalates, which consequently may result in a underestimation of the total daily intake. Furthermore, it is very likely that the values estimated for the total daily intake would increase if the estimates could be based on measured concentrations of DINP and DIDP in the environment as it has been seen for DEHP and DBP.

The present estimated regional total daily intake of BBP based on measured concentrations of BBP in the environment (see Table 5.3) is at the same level as an estimated total daily intake of BBP by Effting and van Veen (1998) based on assumed concentrations in Dutch food. They found, in concordance with the present assessment, that for BBP, the intake of crops is a major source of the total daily intake of phthalates.

In general, the estimations of the total daily intake of DEHP, DBP and BBP in the present assessment are at the same level as the intakes estimated in other investigations (Doull et al., 1999; MAFF, 1996; IPCS, 1999; Chan and Meek, 1994; Meek and Chan, 1994; Effting and van Veen, 1998) although both the range of intakes estimated in the present assessment and in other

investigations is around one order of magnitude. It is mainly the total daily intakes estimated based on measured concentration in the environment (and foodstuff for DEHP) that are comparable to the other investigations, whereas the regional total daily intake estimated by EUSES is lower and the maximum local total daily intake in general is higher.

18.3.3 Total daily intake - conclusions

As for the concentrations in the environment (PECs), also the total daily intake of DEHP and DBP appears to be underestimated by the use of EUSES compared with the measured total daily intake (Petersen, 1999). This is in agreement with Effting and van Veen (1998) who found that EUSES underestimated the concentration of BBP in food in the Netherlands, and thereby also the total daily intake of BBP. As for the concentrations in food, several reasons for the discrepancies can be found, see section 18.2.6.

The estimated total daily intake increased when PECs were replaced by measured environmental concentrations. The replacement of estimated concentrations of DEHP in foodstuffs with the few available measured concentrations resulted in a further increase in the estimated total daily intake of DEHP. In order to improve the exposure assessment of humans via the Danish environment, there is a need for further data regarding measured concentrations especially of phthalate concentrations in various foodstuffs including crops that, according to the EUSES estimations, contribute significantly to the daily intake via the environment, but also of phthalate concentrations in the various environmental compartments.

In the present assessment, several exposure levels via the environment have been estimated depending on the input data used for the estimations. The maximum total daily intake for the local scale based solely on EUSES estimations is the highest of all the estimated values. Therefore the local exposure has to be considered in the evaluation of the combined exposure. When the total daily intake is estimated on the basis of measured concentrations in the environment and in the food, in theory a more reliable estimate is obtained. However, it is uncertain whether the measured concentrations also represent a “high exposure local environment” and for many of the environmental media, the measured data are very scarce. In spite of these limitations, measured concentrations are considered to be more realistic values than estimated values and therefore, the total daily intake estimated on basis of measured concentrations in the environment and in the foodstuffs also has to be taken into account in a combined exposure assessment.

19 Exposure via consumer products

Consumer exposures have been estimated for several different exposure scenarios: toys, building materials etc., infant formulae and baby food, artificial leather and gloves, paints etc., and nail polish. The estimation of the exposures are different in the various scenarios depending on the available data, but exposures have generally been estimated based on e.g., total amounts in the consumer products, emission data from products (e.g., toys, building material) as well as measured concentrations (e.g., infant formulae and baby food, house dust). Furthermore, the

computer exposure modelling program CONSEXPO has been used for estimations of dermal exposures to nail polish and paints.

19.1 Exposure via toys

Phthalates have been used as major components in PVC toys and baby equipment as e.g., teethers.
From investigations of toys on the Danish market (Rastogi and Worsøe, 2001), it is evident that concentrations higher than the maximum concentration allowed in toys and other childcare articles in the age group of 0-3 years (0.05% w/w) occur; there is no regulation of the content of phthalates in toys meant for children older than 3 years. Both the dermal exposure during the child's handling of the toys and especially for the young child, the oral exposure during chewing and biting in the toy are of concern.

The migration of phthalates from toys into saliva has been investigated in different *in vivo* studies. In accordance with the EU-RAR of DINP (2001), the migration data from Könemann (1998) (which are consistent with those of Chen 1998) are considered as being most representative for the European market of toys and therefore, the maximum migration rate (8.9 µg/10 cm^2/minute) from Könemann (1998) has been used to estimate the oral exposure to DINP in the present assessment. Due to lack of measured migration rates for DEHP and DIDP, the migration rate of DINP has also been used to estimate the oral exposure via toys to these two phthalates. The maximum measured total mouthing time (3 hour) measured by Groot et al. (1998) for 1-3 month old children has been used as the duration of mouthing. The estimated oral exposure to DINP, DEHP and DIDP via mouthing on toys is therefore similar (200 µg/kg bw/day).
This is likely to be a worst-case scenario and less conservative values for both the migration rate (2.44 µg/10 cm^2/minute) and mouthing time (74 minutes), but based on the same investigations as in the present assessment, have been suggested (Bremmer and van Veen, 2002). These suggested values led to almost 10 times lower estimated oral exposure (22 µg/kg bw/day) than the present assessment as well as in the EU-RARs of DINP, DIDP and DEHP.

In the present assessment, the same approach has been taken as in the EU-RARs for the dermal exposure estimates where a measured dermal absorption rate of DEHP (0.24 µg/cm^2/hour) has been used (Deisinger et al., 1998). The dermal exposure to DEHP for children from 6-12 month old is thus estimated to be 9 µg/kg bw/day and for DINP and DIDP, it is 1 µg/kg bw/d. For the older group of children (12-36 month), the estimated dermal exposure to DEHP is 1.5 µg/kg bw/day and for DINP and DIDP, it is 0.15 µg/kg bw/day.

As for the oral exposure via toys, exposure parameters from several different dermal exposure scenarios have been suggested by Bremmer and van Veen (2000). One of the scenarios described the dermal exposure to DINP via contact with the ground sheet of a tent. Using the dermal absorption rate measured by Deisinger et al. (1998), the estimated average yearly dermal exposure to DINP for a child (1-7 years old) via contact with the ground sheet of a tent was 1.7 µg/kg bw/day, which is around 10 times higher than the estimated dermal exposure in the present assessment, but only a bit higher than the estimations made in section 8.4.2 where the information of contact is from CSTEE (1998) and Groot et al. (1998).

19.2 Exposure via infant formulae and ready-to-use baby food

Phthalates occur as contaminants in infant formulae and ready-to-use baby food. The infant formulae and baby food can be contaminated via the environment and/or via processing and packaging.

Based on measured concentrations of DEHP in infant formulae and baby food on the Danish market, the estimated daily intake of DEHP via infant formulae is 9.8 and 3.9 µg/kg bw/day for infants 0-6 months old and older than 6 months, respectively, and via baby food 19.6 µg/kg bw/day for children older than 6 months. The estimated daily intake of DEHP via infant formulae is in accordance with those estimated by EU-RAR of DEHP (2001) and MAFF (1998) (13 and 8 µg/kg bw/day for infants 0-3 months old and 6 months old, respectively).

The estimated daily intake of BBP via infant formulae, based on measured concentrations in products on the Danish market, is 1.6 and 0.7 µg/kg bw/day for infants 0-6 months old and 6 months old, respectively. In the EU-RAR of BBP (Draft report 2002), a daily intake of 0.15 µg/kg bw/day is suggested to be used in the risk characterisation, which is around 10 times lower than that in the present assessment. In MAFF (1998), the daily intake of BBP in new-borns has been estimated to be 0.2 µg BBP/kg bw/day.

The estimated daily intake of DBP via infant formulae, based on measured concentrations in products on the Danish market, is 16.4 and 6.6 µg/kg bw/day for infants 0-6 months old and 6 months old, respectively. These estimations are almost three times higher than the daily intake via breast milk (6 µg/kg bw/day) estimated in the EU-RAR of DBP (2001). The estimation in the present assessment is likely to be an overestimation since it is based on the highest detection limit of DBP (0.1 mg/kg wet weight). In MAFF (1998), the estimated daily intake of DBP in new-borns from infant formulae was 2.4 µg/kg bw/day.

The estimated daily intakes of DINP and DIDP (2.5 µg/kg bw/day and 1.8 µg/kg bw/day for infants 0-3 months old and older than 6 months, respectively) in the present assessment as well as in the EU-RARs (DINP 2001; DIDP 2001) are based on the MAFF detection limit of DINP and DIDP in infant formulae.

19.3 Exposure via indoor air and dust

Phthalate containing products (flooring, wall covering, cables, shower curtains, furniture etc.) used indoor will release phthalates to the indoor air. The exposure to the phthalates via indoor air has been estimated based on measurements of the concentration of phthalates in dust as well as on vapour phase concentrations.
In the present assessment, the total exposure to airborne DEHP has been estimated to be 0.48 µg/kg bw/day for adults and 1.94 µg/kg bw/day for children. This is around 10 times lower than the estimated values in the EU-RAR of DEHP (2001) (4.4 µg/kg bw/day for adults and 22.4 µg/kg bw/day for children). This is mainly due to the worst-case approach in the EU-RAR of DEHP (2001).

A similar worst-case approach is also used in the exposure estimations via indoor air to DINP and DIDP (8.3 and 4.2 µg/kg bw/day, respectively) (DINP, 2001; DIDP, 2001) and therefore, the

estimated exposure via indoor air in the present assessment (0.01 and 0.002 µg/kg bw/day, respectively) are much lower than those in the EURARs.

According to Clausen et al. (1999), the indoor air concentration of DEHP and DBP (including both vapour phase concentration and the amount adsorbed to particles) in a school, in a kindergarten, and in 4 offices was between 0.11-1.05 µg DEHP/m^3 and between 0.57-1.35 µg DBP/m^3. These measured total concentrations of phthalates in indoor air are at the same level as those estimated in the present assessment (1.67 µg DEHP/m^3 and 0.36 µg DBP/m^3).

Øie et al. (1997) found that the exposure to DEHP adsorbed to suspended particulate matter (PM_{10}) was one to three times higher than the vapour phase exposure. In the present assessment, the exposure to DEHP and DBP adsorbed to particles was in general 0.4 to 1.1 times higher than the vapour phase exposure.

19.4 Exposure via gloves, clothes, footwear etc.

The phthalates are used in a variety of consumer products that are in contact with the human skin including plastic gloves, water proof clothes, artificial leather in footwear, clothes, seats, and furniture etc. The exposure via dermal contact to these products has been estimated based on a measured dermal absorption rate of DEHP (0.24 µg/cm^2/hour) (Deisinger et al., 1998).

The total dermal exposure to DEHP has been estimated to be 5.8 µg/kg bw/day by assuming that an adult is wearing phthalate-containing gloves 2 hours a day.
Similarly, the dermal exposure to DINP and DIDP has been estimated to be 0.58 µg/kgbw/day by assuming that the dermal absorption rate of DINP and DIDP is 10 times less than that of DEHP.

The dermal exposure to phthalates is also relevant for children as they can be exposed via products such as e.g., waterproof clothes, textile prints etc. and also via contact with other products than clothes (PVC floors, artificial leather on furniture etc.). It is impossible to make one specific dermal contact scenario for children and therefore, it was assumed that the exposed body surface area of children corresponds to the area of the hands. Furthermore, both the duration of the exposure and the dermal absorption rate used to estimate the exposure to adults was also used to estimate the children's exposure. The dermal exposure of children (1-6 years) was estimated to 14.4 µg/kg bw/day for DEHP and 1.4 µg/kg bw/day for DINP and DIDP. The dermal exposure of the older group of children (7-14 years) was estimated to 7.8 µg/kg bw/day for DEHP and 0.78 µg/kg bw/day for DINP and DIDP.

The present estimates of the dermal exposure of adults to DEHP, DIDP and DINP via gloves etc. are almost identical to the exposures estimated in the EU-RARs of the substances (DEHP, 2001; DINP, 2001; DIDP, 2001) as the same approach has been used except for the use of a different body weight of an adult.
For children, no dermal scenarios, except from the dermal exposure via toys, have been included in the EU-RARs.

19.5 Exposure via use of paints, lacquers, adhesives, sealants etc.

All five phthalates are recorded in paints, adhesives, fillers etc. (Flyvholm, 2001) and exposure to phthalates during use of these products can therefore be expected. Since no data are available on the emission of phthalates from paints or adhesives, the exposure via paints, adhesives etc. has been estimated by the use of CONSEXPO 3.0 (van Veen, 2001). Both dermal and inhalatory exposure has been assumed to occur. In general, both the estimated dermal and inhalatory exposures are relatively small (ranging from 0.066 to $2.3*10^{-4}$ µg/kg bw/day) where the highest estimated exposures were obtained for DBP.

Except from the EU-RAR of DBP (2001), none of the other EU-RARs (DEHP, 2001; DINP, 2001; DIDP, 2001; BBP, Draft report 2002) have included exposure via paints, adhesives or other similar scenarios. In the EU-RAR of DBP (2001), the inhalatory exposure was estimated to be 0.34 µg/kg bw/day by the use of CONSEXPO, whereas it was concluded that the dermal exposure is low and therefore not taken into account.

In the human exposure assessment of BBP made by Effting and van Veen (1998), the exposure to BBP via paints and adhesives was also assessed by the use of CONSEXPO as in the present assessment. They estimated a yearly averaged inhalatory uptake of BBP of $4.8*10^{-4}$ µg/kg bw/day via use of adhesives and $6.6*10^{-5}$ µg/kg bw/day via use of floor finish. The exposure is at the same level as that estimated in the present assessment ($2.7*10^{-4}$ µg/kg bw/day).
The estimated averaged dermal uptake of BBP was $4.6*10^{-3}$ µg/kg bw/day via use of adhesives and $3.8*10^{-4}$ µg/kg bw/day via use of floor finish (Effting and van Veen, 1998). This is around ten and 100 times lower than that estimated in the present assessment (0.32 µg/kg bw/day).

19.6 Exposure via nail polish

As the only phthalate of those assessed in the present report, DBP is registered in cosmetic products in PROBAS (Flyvholm, 2001) and, according to the EU-RAR of DBP (2001) and Haulihan et al., (2002), DBP is mainly present in nail polish.

Since no data are available on the emission of phthalates from nail polish the exposure has been estimated by the use of CONSEXPO 3.0 (van Veen, 2001).

The yearly average uptake from inhalation was estimated to $3.4*10^{-6}$ µg/kg bw/day; the dermal exposure is expected to be even less.

The estimated exposure ($2*10^{-6}$ µg/kg bw/day)in the EU-RAR of DBP (2001) is similar to that in the present assessment.

19.7 Other exposure pathways

Exposure to phthalates via other pathways than the above-mentioned can also occur, e.g., via medical devices, food contact material, modelling clay; however, exposure via these pathways has not been quantified in the present report.

20 Combined exposure

The exposures from all the pathways and all the routes estimated for indirect exposure via the environment as well as for consumer exposure have been combined in order to quantify the combined exposure via the environment and via consumer products.

The present assessment shows that food is the dominant pathway of exposure to the five selected phthalates, no matter how the EUSES estimations were made, i.e., based on PECs, measured environmental data, or measured concentrations in food. This finding is in accordance with many other assessments in which the food is reported to be the major source of the general population exposure to many of the phthalates (DINP, 2001; DIDP, 2001; DEHP, 2001; DBP, 2001; CERHR, 2000 a-e; Chan and Meek, 1994; Meek and Chan, 1994; ATSDR, 2001; ATSDR, 2002).

For the young children, oral exposure through the mouthing on toys containing DEHP, DINP, and DIDP also plays a significant role. However, due to the present regulation of use of phthalates in toys to young children (up to 3 years old), there seems to be a discontinuously use of phthalates in toys that are supposed to be used by the youngest children and therefore, the mouthing on such toys (rattles, teethers etc.) may not be a significant route of exposure for this age group in the future. However, phthalates are not yet regulated in toys for older children and therefore, the oral exposure through mouthing on toys can still be significant.

Both dermal and inhalation exposures are lower than the oral exposure. Other studies have reported that the second most important source of exposure to phthalates is via indoor air (DINP, 2001; DIDP, 2001; CERHR, 2000 a-e; Chan and Meek, 1994; Meek and Chan, 1994; ATSDR, 2001); however, in the present assessment the dermal exposure via contact with phthalate containing products (with gloves as an example) has been estimated to be higher than the inhalation exposure via indoor air, except for DBP. This is in accordance with the EURAR of DEHP (2001)

Estimates of exposure to DEHP, DBP and BBP based on measurements of urinary metabolites of the parent compounds (Blount et al., 2000; David, 2000; Kohn et al., 2000), and therefore including all possible pathways and routes of exposure, are at the same level as the estimated combined exposures to DEHP, DBP and BBP in the present assessment if the exposure via the environment is based on measured concentrations in the environment.

Besides the combined exposure to one particular phthalate as e.g., DEHP, also the combined exposure to all phthalates has been considered. For adults, the main exposure is to DEHP and DBP, whereas for the young children, also DINP and DIDP contribute significantly (via toys). In general, the exposure to DBP exceeds the exposure to DEHP due to the higher indirect exposure via the environment to DBP than to DEHP (maximum local total daily intake). As mentioned before, it should be noted that the quantitative data on the tonnage of substances used in Denmark might not represent the situation at present. Especially quantitative data on the use pattern of DBP were scarce and several assumptions had to be made to generate the quantitative data. Also the quantitative data on the use pattern on DINP and DIDP may be encumbered with some uncertainty since they are not very recent and the amount used of DINP and DIDP is expected to have increased over the last years. Therefore, the regional tonnage used in the

present emission assessment may be too low compared with the currently used tonnage of these two phthalates, which consequently may result in an underestimation of the total daily intake.

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Abbreviations

BAF	- bioaccumulation factor
BBP	- benzylbutyl phthalate
BCF	- bioconcentration factor
bw	- body weight
DBP	- di-butyl phthalate
DEHP	- di-(2-ethylhexyl) phthalate
DINP	- di-isononyl phthalate
DIDP	- di-isodecyl phthalate
dw	- dry weight
EU-RAR	- European Union Risk Assessment Report
EUSES	- The European Uniform System for Evaluation of Substances
$K_{air\text{-}water}$	- air-water partition coefficient
K_{oc}	- organic carbon / water partition coefficient
K_{ow}	- octanol/water partition coefficient
MAFF	- Ministry of Agriculture, Fisheries and Food
MST	- Danish Environmental Protection Agency (MiljøSTyrelsen)
NOVA	- National monitoring program of the aquatic environment
PE	- person equivalents
PEC	- predicted environmental concentration
PM_{10}	- particulate matter <10μm
PROBAS	- The Danish Product Register
PVC	- polyvinylchloride
SCF	- surface correction factor
TGD	- Technical Guidance Document
STP	- sewage treatment plant
ww	- wet weight
w/w	- weight to weight

APPENDICES

APPENDIX 1 - Average food consumption rates (kg/d) from Denmark

APPENDIX 2 - Import and export of pure phthalates from Statistic Denmark

APPENDIX 3 - Emission factors for the different use categories – from the A and B tables in EC (1996)

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APPENDIX 6 - Local PECs, concentrations in food groups and total daily intake

APPENDIX 7 - Overview of phthalate containing products, some of which are available to consumers

APPENDIX 8 - Overview of the consumer exposure assessment scenarios

APPENDIX 9 - Parameters used in CONSEXPO defining the exposure via paints etc.

APPENDIX 10 - Parameters used in CONSEXPO defining the exposure via nail polish

APPENDIX 1

Average food consumption rates (kg/day) from Denmark (Andersen et al. (1995))

Main category	Food intake (kg/day)		
	Adult	Child (7-14 y)	Child (1-6 y)
	average	average	average
Meat	0.114	0.098	0.050
Poultry	0.018	0.018	0.012
Fish	0.024	0.015	0.013
Milk etc.	0.350	0.552	0.510
Cheese	0.032	0.024	0.014
Fruit	0.166	0.189	0.175
Vegetables	0.229	0.200	0.122
Cereals	0.211	0.214	0.166
Water etc.	2.022	0.787	0.514

From Andersen et al. (1995)

APPENDIX 2

Import and export of pure phthalates from Statistic Denmark (Statistic Denmark, 1992 - cited by Hoffmann; Statistic Denmark, 2002).

Group		Tons	
		1992	2001
Dibutyl *ortho*-phthalates 2917.31.00	Import	454	82.1
	Export	4	0.2
	Total	450	81.9
Dioctyl *ortho*-phthalates 2917.32.00	Import	1,482	1,053.4
	Export	0	0.5
	Total	1,482	1,052.9
Dinonyl- or didecyl *ortho*-phthalates 2917.33.00	Import	175	1,076.1
	Export	0	2,692.3
	Total	175	-1,616.2
Di-isononyl- and di-isodecyl *ortho*-phthalates 2917.34.10	Import	3,227	
	Export	0	Not existing
	Total	3,227	
Esters of phthalic acids except those mentioned above 2917.34.90	Import	166	
	Export	15	Not existing
	Total	151	
Esters of *ortho*-phthalic acids, except dibutyl-, dioctyl-, dinonyl- and didecyl *ortho*-phthalates 2917.34.00	Import		913.3
	Export	Not existing	101.0
	Total		812.3
Phthalic anhydride 2917.35.00	Import	1,670	1,199.8
	Export	9	0
	Total	1,672	1,199.8
Plasticisers, composed, for rubber or plastic 3812.20.00 (I 2002; sum of 3812.20.10 and 3812.20.90)	Import	1,019	576.3
	Export	0	5.2
	Total	1,019	571.1
Total, plasticisers, pure substances		6,504	3,718 (without the data on dinonyl- and didecyl *ortho*-phthalater

Dibutyl *ortho*-phthalates = DBP
Dioctyl *ortho*-phthalates = DEHP
Dinonyl- or didecyl *ortho*-phthalates (DINP, CAS nr. 28553-12-0 also synonym with dinonyl phthalate; didecyl phthalate is not included in IUCLID)
Di-isononyl- and di-isodecyl *ortho*-phthalates = DINP and DIDP
Esters of phthalic acids except those mentioned above – (incl. BBP)
Esters of *ortho*-phthalic acids, except dibutyl-, dioctyl-, dinonyl- and didecyl *ortho*-phthalates – (incl. BBP, DINP and DIDP)

Import of PVC, semi-manufactured (with (compound) and without (resin) plasticiser)

Group		Tons	
		1992	2001
PVC, un-manufactured, softened 3904.22.00	Import	-	15,636[2]
	Export	-	719.9[2]
	Total		14,916.1[2]
PVC compound	Total	10,000[1]	
Estimated amount of phthalates imported as un-manufactured, softened, PVC (PVC compound)		3-5,000 t [1]	Ca. 4,500 t (30% phthalate)
PVC, un-manufactured, not mixed with other compounds 3904.10.00	Import		35,942.3[2]
	Export		281.1[2]
	Total		35,661.2[2]
PVC Resin, for production of PVC compound	Total	14,000-18,000[1]	

[1] Hofmann (1996)
[2] Statistic Denmark (2002)

APPENDIX 3

Emission factors (A-tables) and the fraction of the main local source and days of emission (B-table) for the different phthalates and use categories – the A- and B-tables are from EC (1996),

DEHP:

The emission factors (A-tables) and the fraction of the main local source and days of emission (B-table) for the each use pattern

Use pattern	Industry Category	Use category	Life cycle	Mc.	Air	Wastewater	Soil	Fraction of the main local source	Days of emission
Polymer plasticiser	Polymers industry (11)	Softener (47)	Prod.	III	0.00001	0.02	0.0001	1	300
			Proc.	II	0.01	0.001	0.0005	0.05	300
Paints, lacquers etc.	Paints, lacquers and varnishes industry (14)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0	0.001	0.005	0.15	300
			Priv. use		0	0.001	0.005	$8*10^{-7}$	300
Adhesives (+ sealants)	Engineering Industry (16)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	II	0.0001	0.01	0.005	0.6	138
Printing Inks	Pulp, paper and board industry (12)	Solvent (48)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.05	0.0005	0.0015	0.33	300
Ceramics	Others (15)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.001	0.1	0.01	1	1

DBP:

The emission factors (A-tables) and the fraction of the main local source and days of emission (B-table) for the each use pattern

Use pattern	Industry Category	Use category	Life cycle	Mc.	Air	Wastewater	Soil	Fraction of the main local source	Days of emission
Polymer plasticiser	Polymers industry (11)	Softener (47)	Prod.	III	0.00001	0.02	0.0001	1	300
			Proc.	II	0.01	0.001	0.0005	0.1	300
Paints, lacquers etc.	Paints, lacquers and varnishes industry (14)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0	0.001	0.005	0.15	300
			Priv. use		0	0.005	0.005	$4*10^{-6}$	300
Adhesives (+ sealants)	Engineering Industry (16)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	II	0.0001	0.01	0.005	0.6	240
Printing Inks	Pulp, paper and board industry (12)	Solvent (48)	Form.	III	0.0025	0.02	0.0001	0.8	300
			Proc.	III	0.05	0.0005	0.0015	0.33	300
Grouting Agents	Others (15)	Others (55)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.001	0.1	0.01	1	1

DINP:

The emission factors (A-tables) and the fraction of the main local source and days of emission (B-table) for the each use pattern

Use pattern	Industry Category	Use category	Life cycle	Mc.	Air	Wastewater	Soil	Fraction of the main local source	Days of emission
Polymer plasticiser	Polymers industry (11)	Softener (47)	Prod.	III	0.00001	0.02	0.0001	1	300
			Proc.	II	0.005	0.001	0.0005	0.15	270
Paints, lacquers etc.	Paints, lacquers and varnishes industry (14)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0	0.001	0.005	0.3	200
			Priv. use		0	0.001	0.005	$4*10^{-8}$	200
Adhesives (+ sealants)	Engineering Industry (16)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	II	0.0001	0.01	0.005	0.6	138
Printing Inks	Pulp, paper and board industry (12)	Solvent (48)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.05	0.0005	0.0015	0.33	80
Ceramics	Others (15)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.001	0.1	0.01	1	1

DIDP:

The emission factors (A-tables) and the fraction of the main local source and days of emission (B-table) for the each use pattern

Use pattern	Industry Category	Use category	Life cycle	Mc.	Air	Wastewater	Soil	Fraction of the main local source	Days of emission
Polymer plasticiser	Polymers industry (11)	Softener (47)	Prod.	III	0.00001	0.02	0.0001	1	300
			Proc.	II	0.005	0.001	0.0005	0.15	80
Paints, lacquers etc.	Paints, lacquers and varnishes industry (14)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0	0.001	0.005	0.6	160
			Priv. use		0	0.001	0.005	0	1
Adhesives (+ sealants)	Engineering Industry (16)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	II	0.0001	0.01	0.005	0.8	115
Printing Inks	Pulp, paper and board industry (12)	Solvent (48)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.05	0.0005	0.0015	0.33	300
Printing inks, textile	Textile processing industry (13)	Softener (47)	form.	III	0.0025	0.02	0.0001	1	300
			proc.	III	0.05	0.85	0.005	0.75	1
Ceramics	Others (15)	Softener (47)	Form.	III	0.0025	0.02	0.0001	1	300
			Proc.	III	0.001	0.1	0.01	1	1

APPENDIX 4

Emission during end-use of phthalate containing products

4.A The surface correction factor (SCF) and technical lifetime of different use patterns. The values are used to estimate the total emitting surfaces of each use pattern.

	Average thickness (mm)	Emission sides	SCF	Technical lifetime
Roofing material[1]	1.5	1	1	20
Paints, lacquers etc. [2]	0.04	1	38	7
Adhesives, sealants etc.[3]	1.5	1	1	20
Printing inks[4]	0.0015	1	1000	1

[1] Available study.
[2] DEHP (2001). A SCF of 10 are used in EU-RAR of DINP and DIDP for paints, lacquers etc.
[3] DEHP (2001), DINP (2001) and DIDP (2001).
[4] DEHP (2001). No emission to air from printing ink in the other EU-RARs.

4.B Estimated emission during end-use of each product group to air, wastewater, surface water and soil.

DEHP in	Tons/year	Surface area (m^2)	Wastewater	surface water	Soil	Air
PVC[1]	9,540	-	23.9	15.3	15.3	3.8
Paints, lacquers etc.	30	4,245,360[2]	4.2[4]	2.1[6]	2.1[6]	0.04[7]
Adhesives, sealings etc.	230	2,447,200[2]	2.4[4]	1.2[6]	1.2[6]	0.02[7]
Printing inks	34	18,088,000[2,3]	0.2[5]	-	-	0.34[7]
Total			30.7	18.6	18.6	4.2

[1] See appendix 4C
[2] (volume of DEHP)*(the area of 1 ton roofing material = 532 m^2)*SCF*technical lifetime
[3] Only used to estimate the emission to air. The emission to water is estimated on the basis of tonnage
[4] 1.98 g/m^2/year*surface area*fraction used indoor (0.5)
[5] Volume of DEHP*emission factor (0.06)*fraction remained after primary treatment (0.1)*recycling rate (0.5)*retention of phthalate in paper (2)
[6] 1.98 g/m^2/year*surface area* fraction used outdoor (0.5)*fraction directed to surface water/soil (0,5)
[7] 9.5 mg/m^2/year*surface area

DINP in	Tons/year	Surface area (m^2)	Wastewater	Surface water	Soil	Air
PVC[1]	1,350	-	3.4	2.2	2.2	0.5
Paints, lacquers etc.	12	1,698,144[2]	0.9[4]	0.4[6]	0.4[6]	0.02[7]
Adhesives, sealings etc.	12	127,680[2]	0.07[4]	0.03[6]	0.03[6]	0.001[7]
Printing inks	12	6,384,000[2,3]	0.07[5]	-	-	0.1[7]
Total			4.4	2.6	2.6	0.6

[1] See appendix 4C
[2] (volume of DINP)*(the area of 1 ton roofing material = 532 m^2)*SCF*technical lifetime
[3] Only used to estimate the emission to air. The emission to water is estimated on the basis of tonnage
[4] 1.05 g/m^2/year*surface area*fraction used indoor (0.5)
[5] volume of DINP*emission factor (0.06)*fraction remained after primary treatment (0.1)*recycling rate (0.5)*retention of phthalate in paper (2)
[6] 1.05 g/m^2/year*surface area* fraction used outdoor (0.5)*fraction directed to surface water/soil (0,5)
[7] 9.5 mg/m^2/year*surface area

DIDP in	Tons/year	Surface area (m^2)	Wastewater	Surface water	Soil	Air
PVC[1]	400	-	1.0	0.6	0.6	0.2
Paints, lacquers etc.	2	283,024[2]	0.2[3]	0.07[4]	0.07[4]	0.003[5]
Adhesives, sealings etc.	36	383,040[2]	0.2[3]	0.1[4]	0.1[4]	0.004[5]
Printing inks	-	-	-	-	-	-
Total			1.4	0.8	0.8	0.2

[1] See appendix 4C
[2] (volume of DIDP)*(the area of 1 ton roofing material = 532 m^2)*SCF*technical lifetime
[3] 1.05 g/m^2/year*surface area*fraction used indoor (0.5)
[4] 1.05 g/m^2/year*surface area* fraction used outdoor (0.5)*fraction directed to surface water/soil (0,5)
[5] 9.5 mg/m^2/year*surface area

DBP in	Tons/year	Surface area (m^2)	Wastewater	Surface water	Soil	Air
PVC[1]	2,171	-	6.0	3.5	3.5	0.9
Paints, lacquers etc.	447	63,255,864[2]	62.6[4]	31.3[6]	31.3[6]	0.2[7]
Adhesives, sealings etc.	400	4,256,000[2]	4.2[4]	2.1[6]	2.1[6]	0.04[7]
Printing inks	200	106,400,000[2,3]	1.2[5]	-	-	2.2[7]
Total			74.0	36.9	36.9	3.3

[1] See appendix 4C
[2] (volume of DBP)*(the area of 1 ton roofing material = 532 m^2)*SCF*technical lifetime
[3] Only used to estimate the emission to air. The emission to water is estimated on the basis of tonnage
[4] 1.98 g/m^2/year*surface area*fraction used indoor (0.5)
[5] Volume of DBP*emission factor (0.06)*fraction remained after primary treatment (0.1)*recycling rate (0.5)*retention of phthalate in paper (2)
[6] 1.98 g/m^2/year*surface area* fraction used outdoor (0.5)*fraction directed to surface water/soil (0,5)
[7] 9.5 mg/m^2/year*surface area

4.C Emission factors of phthalates during end-use of PVC products.

From detailed estimations of the emission from several different PVC product groups made in the EU-RARs of of DEHP (2001), EU-RAR of DINP (2001) and EU-RAR of DIDP (2001) the total emission from all PVC product groups has been used in the present study to estimate a general emission factor for PVC products. The estimated emitted amount used in the present study is also presented.

Emission fractions (%)				
	Wastewater	Surface water	Soil	Air
DEHP	0.28	0.14	0.14	0.04
DINP	0.23	0.19	0.19	0.04
DIDP	0.23	0.15	0.15	0.04
Average	0.25	0.16	0.16	0.04

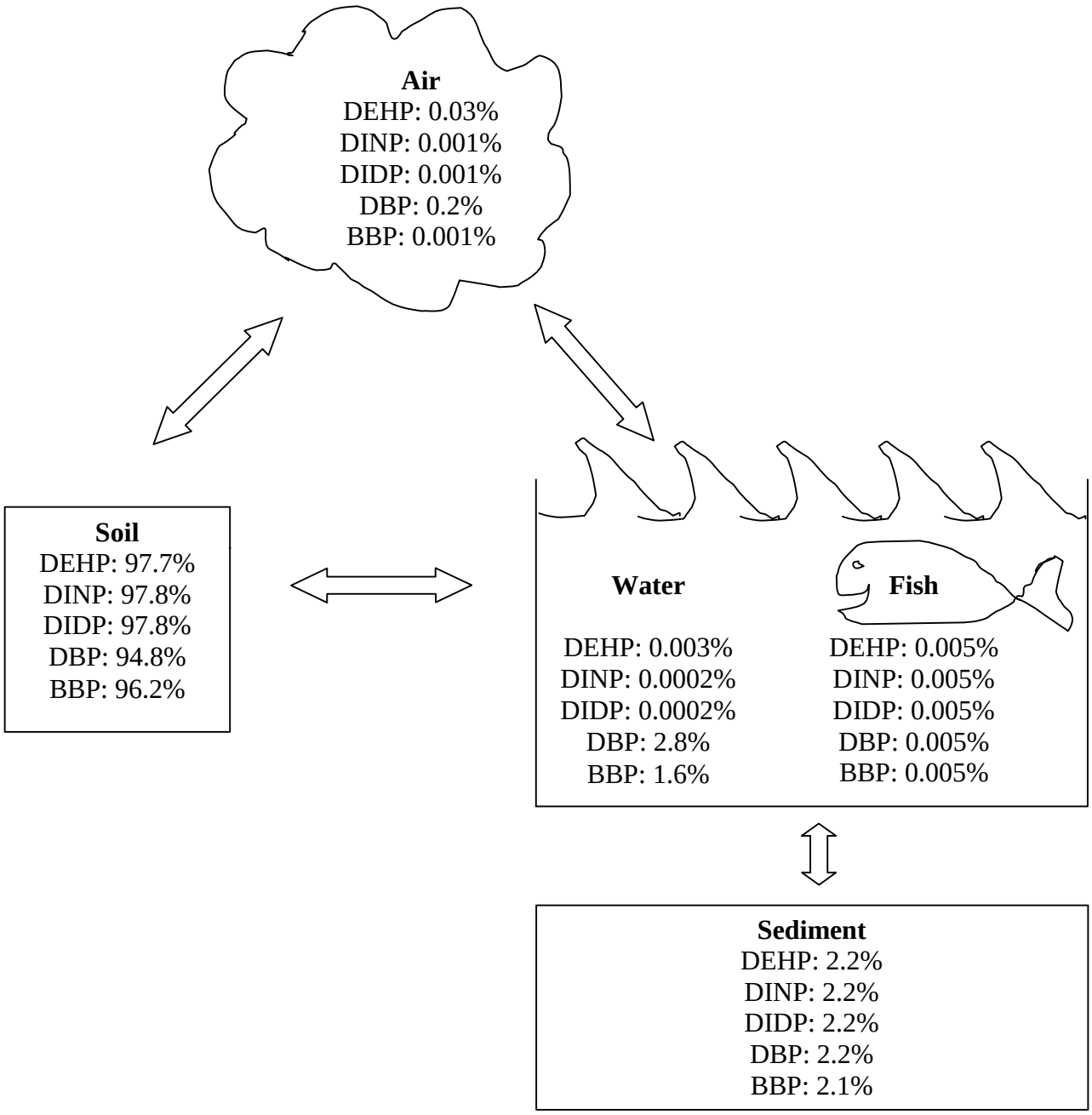
Total amount emitted (tons/year)				
	Wastewater	Surface water	Soil	Air
DEHP	23.9	15.3	15.3	3.8
DINP	3.4	2.2	2.2	0.5
DIDP	1.0	0.6	0.6	0.2
DBP	6.0	3.5	3.5	0.9
Total	34.3	21.9	21.9	5.5

4.D Emission factors used by Hoffmann (1996) to estimate the emission during use of products.

Type of product		Fraction emitted to (assumed life time)	
		Air	Water
PVC	Floor and wall covering	0.01 % (10 years)	0.01-0.1%(10 years)
	cables	0.01 – 0.1 % (10 years)	?
	Tubes etc.	0.01-0.1 % (5 years)	0.01-0.1 % (5 years)
	Other products	0.01-0.1 % (0-5 years)	0.01-0.1 % (0-5 years)
Lacquers, paint etc.		0.01 (10 years)	0.01-0.1%(10 years)
Adhesives		-	5-10% (private) 1-5% (industrial)
Fillers		?	?

APPENDIX 5

The distribution of DEHP, DINP DIDP, DBP and BBP estimated in the fugacity-based environmental equilibrium partitioning model, EQC, version 1.01, Level 1.



APPENDIX 6

The estimated local concentrations of DEHP in the different environmental compartments, in the wastewater, in the different food groups and the local total daily intake of DEHP.

DEHP	PVC		Paints, lacquers etc.		Printing inks		Adhesives etc.	
	PROC	FORM	PROC	PRIV USE	FORM	PROC	FORM	PROC
Annual average local PEC in air (total) (µg/m^{-3})	3.64	0.0624	0.00526	0.00526	0.07	0.427	0.443	0.0155
Annual average total deposition flux (mg/m^{-2}/d)	0.0989	0.00158	0.000000186	9.94E-13	0.00179	0.0115	0.0121	0.000337
Annual average local PEC in surface water (µg/m^{-3})	919	1100	234	228	1210	236	6900	2180
Local PEC in sediment during emission episode (µg/kg dw)	9970	12000	2200	2120	13300	2210	77800	50400
Local PEC in agric. soil (total) averaged over 180 days (µg/kgdw)	618	597	15	10.8	675	32.7	4510	2870
Local PEC in grassland (total) averaged over 180 days (µg/kg dw)	605	404	13.6	10.8	457	47.4	3030	1910
Local PEC in groundwater under agricultural soil (µg/m^{-3})	187	181	4.56	3.26	205	9.9	1,370	868
Concentration in untreated wastewater (mg/l)	0.135	0.17	0.00125	6.65E-09	0.193	0.00157	1.3	0.832
Concentration of chemical (total) in the STP-effluent (mg/l)	0.0109	0.0137	0.000101	5.37E-10	0.0156	0.000127	0.105	0.0671
Concentration in dry sewage sludge (mg/kg)	324	407	2.98	0.0000159	461	3.75	3120	1990
Concentration in effluent exceeds solubility	Yes	Yes	No	No	Yes	No	Yes	Yes
Local concentration in wet fish (mg/kg)	0.772	0.922	0.197	0.191	1.02	0.198	5.79	1.83
Local concentration in root tissue of plant (mg/kg)	0.519	0.501	0.0126	0.00903	0.567	0.0274	3.78	2.41
Local concentration in leaves of plant (mg/kg)	1.16	0.0199	0.00167	0.00167	0.0223	0.136	0.141	0.00503
Local concentration in drinking water (µg/m^{-3})	0.00023	0.000274	0.0000585	0.0000569	0.000303	0.0000589	0.00172	0.000868
Local concentration in meat (wet weight) (mg/kg)	0.0158	0.000306	0.0000245	0.0000242	0.000344	0.00185	0.00219	0.000234
Local concentration in milk (wet weight) (mg/kg)	0.00474	0.0000918	0.00000736	0.00000727	0.000103	0.000555	0.000656	0.0000703
Local total daily intake for humans, adults (mg/kg bw/d)	0.0171	0.00272	0.000214	0.000198	0.00305	0.00197	0.0195	0.00996
Local total daily intake for humans, 1-6 years (mg/kg bw/d)	0.102	0.0125	0.000986	0.000911	0.014	0.0118	0.0898	0.0444
Local total daily intake for humans, 7-14 years (mg/kg bw/d)	0.041	0.00553	0.000429	0.000396	0.00621	0.00467	0.0397	0.020

The estimated local concentrations of DBP in the different environmental compartments, in the wastewater, in the different food groups and the local total daily intake of DBP.

DBP	PVC		Paints, lacquers etc.		Printing inks		Adhesives, sealants etc.	
	Proc	Form.	Proc	Priv. Use	Form	Proc	Form	Proc
Annual average local PEC in air (total) (µg/m^{-3})	1.66	0.682	0.00164	0.0015	0.306	2.48	0.763	0.0194
Annual average total deposition flux (mg/m^{-2}/d)	0.00296	0.00123	0.000000249	9.97E-12	0.000549	0.00444	0.00137	0.0000347
Annual average local PEC in surface water (µg/m^{-3})	1700	19200	1700	1150	9230	1230	21,400	7070
Local PEC in sediment during emission episode (µg/kg dw)	654	8330	656	414	3960	450	9,270	3660
Local PEC in agric. soil (total) averaged over 180 days (µg/kg dw)	79.2	2460	75.4	0.188	1100	17.7	2,760	1010
Local PEC in grassland (total) averaged over 180 days (µg/kg dw)	61.6	1750	53.5	0.188	783	20.7	1,960	716
Local PEC in groundwater under agricultural soil (µg/m^{-3})	624	19400	594	1.48	8690	140	21,700	7950
Concentration in untreated wastewater (mg/l)	0.0616	2.03	0.062	0.00000248	0.908	0.00923	2.27	0.832
Concentration of chemical (total) in the STP-effluent (mg/l)	0.00675	0.222	0.00679	0.000000272	0.0995	0.00101	0.249	0.0911
Concentration in dry sewage sludge (mg/kg)	56.2	1850	56.5	0.00226	828	8.42	2070	759
Concentration in effluent exceeds solubility	No	No	No	No	No	No	No	No
Local concentration in wet fish (mg/kg)	0.00305	0.0346	0.00306	0.00207	0.0166	0.00221	0.0384	0.0127
Local concentration in root tissue of plant (mg/kg)	0.196	6.11	0.187	0.000467	2.73	0.044	6.83	2.5
Local concentration in leaves of plant (mg/kg)	3.4	1.41	0.00348	0.00308	0.631	5.1	1.57	0.0413
Local concentration in drinking water (µg/m^{-3})	0.000848	0.0194	0.00085	0.000574	0.00869	0.000615	0.0217	0.00795
Local concentration in meat (wet weight) (mg/kg)	0.215	0.0905	0.000282	0.000224	0.0406	0.322	0.101	0.00326
Local concentration in milk (wet weight) (mg/kg)	0.068	0.0286	0.000089	0.0000709	0.0128	0.102	0.032	0.00103
Local total daily intake for humans, adults (mg/kg bw/d)	0.0408	0.039	0.000741	0.0000653	0.0175	0.0602	0.0436	0.00968
Local total daily intake for humans, 1-6 years (mg/kg bw/d)	0.269	0.215	0.0035	0.000311	0.0963	0.399	0.24	0.0463
Local total daily intake for humans, 7-14 years (mg/kg bw/d)	0.105	0.0905	0.00158	0.00013	0.0406	0.155	0.101	0.0208

The estimated local concentrations of DINP in the different environmental compartments, in the wastewater, in the different food groups and the local total daily intake of DINP.

DINP	Adhesives, sealants etc.		Printing inks		Paints, lacquers etc.			PVC
	Form	Proc	Form	Proc	Form	Proc	Priv. Use	Proc
Annual average local PEC in air (total) (µg/m^{-3})	0.0237	0.00153	0.0237	0.15	0.0237	0.000839	0.000819	0.772
Annual average total deposition flux (mg/m^{-2}/d)	0.000557	0.0000286	0.000557	0.00342	0.000557	0.000000454	6.05E-14	0.0178
Annual average local PEC in surface water (µg/m^{-3})	317	132	317	15.4	317	17.4	12.9	269
Local PEC in sediment during emission episode (µg/kg dw)	6,190	18,700	6,190	391	6,190	318	209	5,820
Local PEC in agric. soil (total) averaged over 180 days (µg/kg dw)	232	709	232	13.3	232	5.6	1.39	242
Local PEC in grassland (total) averaged over 180 days (µg/kg dw)	167	506	167	16.2	167	4.39	1.39	206
Local PEC in groundwater under agricultural soil (µg/m^{-3})	40.5	124	40.5	2.33	40.5	0.979	0.244	42.3
Concentration in untreated wastewater (mg/l)	0.0681	0.21	0.0681	0.00208	0.0681	0.00125	2E-10	0.0638
Concentration of chemical (total) in the STP-effluent (mg/l)	0.0056	0.0173	0.0056	0.000171	0.0056	0.000103	1.64E-11	0.00525
Concentration in dry sewage sludge (mg/kg)	171	529	171	5.23	171	3.14	0.000000503	161
Concentration in effluent exceeds solubility	Yes	Yes	Yes	No	Yes	No	No	Yes
Local concentration in wet fish (mg/kg)	0.266	0.111	0.266	0.0129	0.266	0.0146	0.0108	0.226
Local concentration in root tissue of plant (mg/kg)	0.198	0.608	0.198	0.0114	0.198	0.0048	0.00119	0.207
Local concentration in leaves of plant (mg/kg)	0.00238	0.000157	0.00238	0.015	0.00238	0.0000843	0.0000823	0.0776
Local concentration in drinking water (µg/m^{-3})	0.0000792	0.000124	0.0000792	0.00000384	0.0000792	0.00000434	0.00000323	0.0000674
Local concentration in meat (wet weight) (mg/kg)	0.0188	0.0179	0.0188	0.0828	0.0188	0.000623	0.00051	0.431
Local concentration in milk (wet weight) (mg/kg)	0.00594	0.00565	0.00594	0.0262	0.00594	0.000197	0.000161	0.136
Local total daily intake for humans, adults (mg/kg bw/d)	0.00107	0.00236	0.00107	0.000858	0.00107	0.0000336	0.0000172	0.0051
Local total daily intake for humans, 1-6 years (mg/kg bw/d)	0.00526	0.0114	0.00526	0.00522	0.00526	0.000164	0.0000855	0.0302
Local total daily intake for humans, 7-14 years (mg/kg bw/d)	0.00234	0.00516	0.00234	0.00219	0.00234	0.0000724	0.0000372	0.0127

The estimated local concentrations of DIDP in the different environmental compartments, in the wastewater, in the different food groups and the local total daily intake of DIDP.

DIDP	PVC	Paints, lacquers etc.		Adhesives, sealants etc.	
	Proc	Form	Proc	Form	Proc
Annual average local PEC in air (total) (µg/m^{-3})	0.23	0.00537	0.00158	0.0701	0.00579
Annual average total deposition flux (mg/m^{-2}/d)	0.00555	0.000107	0.000000425	0.00192	0.000154
Annual average local PEC in surface water (µg/m^{-3})	95.9	65.2	5.66	1110	436
Local PEC in sediment during emission episode (µg/kg dw)	6850	1270	129	21800	22200
Local PEC in agric. soil (total) averaged over 180 days (µg/kg dw)	250	45.5	5.12	771	784
Local PEC in grassland (total) averaged over 180 days (µg/kg dw)	177	31.3	4.33	516	521
Local PEC in groundwater under agricultural soil (µg/m^{-3})	43.7	7.95	0.895	135	137
Concentration in untreated wastewater (mg/l)	0.0638	0.0113	0.000624	0.204	0.208
Concentration of chemical (total) in the STP-effluent (mg/l)	0.00636	0.00113	0.0000622	0.0204	0.0208
Concentration in dry sewage sludge (mg/kg)	167	29.8	1.64	536	546
Concentration in effluent exceeds solubility	Yes	Yes	No	Yes	Yes
Local concentration in wet fish (mg/kg)	0.0806	0.0548	0.00475	0.931	0.366
Local concentration in root tissue of plant (mg/kg)	0.214	0.039	0.00439	0.661	0.672
Local concentration in leaves of plant (mg/kg)	0.00772	0.00018	0.0000531	0.00235	0.000195
Local concentration in drinking water (µg/m^{-3})	43.7	8.15	0.895	139	137
Local concentration in meat (wet weight) (mg/kg)	0.0496	0.00207	0.000445	0.0307	0.0187
Local concentration in milk (wet weight) (mg/kg)	0.0157	0.000656	0.000141	0.00971	0.0059
Local total daily intake for humans, adults (mg/kg bw/d)	0.0013	0.000189	0.0000229	0.00314	0.0026
Local total daily intake for humans, 1-6 years (mg/kg bw/d)	0.0067	0.00907	0.000113	0.015	0.0126
Local total daily intake for humans, 7-14 years (mg/kg bw/d)	0.00293	0.000405	0.0000501	0.0067	0.00569

APPENDIX 7

Overview of phthalate containing products, some of which are available to consumers.

Product type	DEHP	DBP	BBP	DINP	DIDP	Scenario
Waterproof clothes[1]	x			x	x	Clothing
Gloves, industry[1]	?	?	?	?	?	Clothing
Shoes, Boots, waders etc.[1]	x	?	?	x	?	Clothing
Textile print[3]	?	?	?	?	?	Clothing
Waterbed[1]	?			x		Clothing
Furniture (artificial leather)[1,2]	?			x		Clothing
Cosmetics[3]		x				Cosmetic
Sealants[3]	x	x	x	x	x	Paints, adhes. etc. (during use)
Paints and lacquers[3]	x	x	x		x	Paints, adhes. etc. (during use)
Adhesives[3]	x	x	x	x	x	Paints, adhes. etc. (during use)
Glues[3]						Paints, adhes. etc. (during use)
Grouting agents[3]	x	x	x			Paints, adhes. etc. (during use)
Hoses, (food) industry[1]	x					Food
Support in tops, lids etc.[1]	?	?	?	x	x	Food
Flooring, wall covering etc.[1,3]	x		x	x		Indoor air
Bags, suitcases etc[1]	x			x		Indoor air
Office items…[1]	x			x		Indoor air
Cables, electrical components etc.[1,3]	x	x		x	x	Indoor air
Profiles[1]	x			x		Indoor air
Table cloth, shower curtain etc.[1]	x	?	?	?	?	Indoor air
Carpet tiles, mats etc.[1,3]	x	x		x		Indoor air
Adhesive tape, foils etc.[1]	x	?	?	?	?	Indoor air
Printing inks, dyestuff [3]	x	x	x	x	x	Indoor air
Rubber[4]	?	?	?	?	?	Indoor air
Silicone[4]	?	?	?	?	?	Indoor air
Polisher, wax etc.[3]		x				Indoor air, waxing
Cars, interior[1]	x				x	Included in indoor air
Cars, undercoating etc.[1]	x				x	Outdoor (no scenario)
Tarpaulin[1]	x			x		Outdoor (no scenario)
Fenders, maritime products[1]	?	?	?	x	?	Outdoor (no scenario)
Roofing etc.[1,3]	x	x	x	x		Outdoor (no scenario)
Medical device[1]	x			x		Medical (no scenario)
Toys[1]	x			x	x	Toys
Covering, swimming pools[2]	x	x				Water; dermal, oral
Garden hose[1]	x					Water; dermal, oral

[1] Danish Plastic Industry (1996)
[2] MST (1999)
[3] Flyvholm (2001)
[4] MST (2000)

APPENDIX 8

Overview of the exposure assessment scenarios

Toys

Route:
- oral
- dermal

Data:
- measurements on the concentration of phthalates in toys on the Danish marked
- measurements on the leaching rate during mouthing of the toys
- mouthing time
- dermal absorption rates.

Infant formulae and baby food

Route:
- oral

Data:
- measurements on the concentration of phthalates in infant formulae and baby food
- the daily intake of the infant formulae and baby food
- estimations of the exposure via the environment (EUSES estimations)

Indoor air and dust – inhalation

Route:
- inhalation

Data:
- measurements of the concentration of phthalates in dust
- measurements on the air concentration of phthalates emitted from PVC products
- the amount of dust indoor

Clothing (gloves, footwear etc.)

Route:
- dermal

Data:
- measurements on the concentration of phthalates in clothing e.g. gloves
- dermal absorption rates

Paints, adhesives etc.

Route:
- inhalation
-dermal

Data:
-CONSEXPO, fact-sheet paint
-dermal absorption rates

Cosmetics (nail polish, DBP)

Route:
- inhalation

Data:
- CONSEXPO

APPENDIX 9

Parameters used in CONSEXPO defining the inhalation exposure via paints etc.

Scenario	Parameter	Inhalation	Dermal
Contact	Frequency	6 times/year [1]	6 times/year [1]
	Total duration	198 min [1]	198 min [1]
	Duration of use	180 min [1]	180 min [1]
Exposure	Name of model	*evaporation due to painting*	*fixed volume of product*
	Release area	2.5 m^{2} [;1]	-
	Room volume	20 m^{3} [;1]	-
	Ventilation rate	12 m^{3}/hr [1]	-
	Product amount	250 g [1]	-
	Product density	1.18 g/cm^{3} [;1]	1.18 g/cm^{3} [;1]
	Weight fraction	0.1 [2]	0.1 [2]
	Fraction to upper layer	0.1 [1]	-
	Layer exchange rate	0.4 l/min [1]	-
	temperature	20ºC [1]	-
	molecular weight matrix	450 g/mol [1]	-
	applied product volume	-	0.24 cm^{3} [;3]
	dilution before use	-	100%
Uptake	Name of model	*fraction model*	*Diffusion model*
	Absorbed fraction	1.0 [1]	-
	inhalation rate	20 m^{2}/day [2]	-
	Contact area	-	20 cm^{2} [;3]
	Blood flow at contact area	-	2 cm^{3} [;3]

[1] Bremmer and van Veen (2000), Factsheet for paints
[2] values used in the exposure assessment via the environment (different from the values from the factsheet)
[3] Effting and van Veen (1998)

APPENDIX 10

Parameters used in CONSEXPO defining the inhalation exposure to BBP in nail polish

Scenario	Parameter	Inhalation
Contact	Frequency	104 times/year [1]
	Total duration	10 min [1]
	Duration of use	5 min [1]
Exposure	Name	*evaporation from mixture*
	Release area	20 cm^{2} [;1]
	Room volume	20 m^{3} [;2]
	Ventilation rate	12 m^{3}/hr [2]
	Weight fraction	0.05 [3]
	temperature	20ºC [2]
	molecular weight matrix	100 g/mol [1]
Uptake	Absorbed fraction	1.0 [1]
-fraction model	inhalation rate	20 m^{2}/day [3]
Mean event concentration, air (mg/m^{3})		$5.99*10^{-6}$
Average uptake per event (µg/kg bw/ day of application)		$1.19*10^{-5}$
Average uptake per year (µg/kg bw/day)		$3.38*10^{-6}$

[1] EU-RAR of DBP (2001)
[2] Bremmer and van Veen (2000), Factsheet for paints
[3] Values used in the exposure assessment via the environment (different from the values from the factsheet)