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Phthalates in preschool dust

the relation between phthalates and parameters in the preschool environment

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1. Summary

Children are constantly exposed to many chemicals via the products they come in contact with in their everyday life. One chemical group is phthalates, the most commonly used plasticisers in the world. Phthalates are used mainly in PVC plastic products like floors, toys, food containers and wallpaper but they are also used in rubber, glue, paint, cables etc. Since these chemicals are weakly chemically bound to the PVC they can leak and migrate to the air, food, water and skin. Children are exposed to phthalates mainly through food, but because of the hand to mouth behaviour they are also exposed via dust inhalation and dust ingestion. About ten years ago regulations of the most toxic phthalates in toys and child care products were implemented in the EU and from February 2015 it is a general prohibition for the use of DEHP (diethylhexyl phthalate), DnBP (di-n-butyl phthalate), DiBP (diisobutyl phthalate) and BBzP (butyl benzyl phthalate). DiNP (diisononyl phthalate) is a relatively new phthalate and it has replaced DEHP to some extent in floors and toys. In phthalate free products DINCH (1,2-cyclohexane dicarboxylic acid diisononyl ester) is one of the plasticizer replacing phthalates. The banned phthalates have been shown to cause adverse effects on reproduction and recent research also investigates links between phthalate exposure and asthma and allergy symptoms in children. More experimental animal studies are needed to further investigate the asthma and allergy correlation.

In the present project dust samples were collected from 30 preschool indoor environments in the Stockholm city area to evaluate the levels of the six substances mentioned and the phthalate DEP (diethyl phthalate). The main aim of this thesis project was to search for relations and links between the phthalate concentrations in dust and parameters from the indoor environment in preschools. Studied parameters are *e.g.* construction year, floor type, cleaning routines and quantity of toys and furniture made of plastic or foam. The project was commissioned by the Swedish EPA and performed at the Institute of Environmental Medicine (IMM) at Karolinska Institutet.

Preschools were recruited and dust was sampled from areas above floor level in a play room on a filter attached to a vacuum cleaner. Areas close to plastic materials were avoided as much as possible but sometimes the amount of dust in the play room was not enough without sampling close to plastics. A questionnaire with questions about the indoor environment was answered by the preschool director or a preschool teacher. The dust was analysed by the Swedish Environmental Research Institute and the phthalate concentrations in the dust were similar to those found in previous studies. Preschool indoor parameters from the questionnaire were translated into a numerous code and transferred to a database for statistical analyse. Statistical analyses were made using Mann-Whitney U-tests and Spearman's rank correlation tests. A negative relation was found between dust phthalate (DEHP and DnBP) concentration and construction year. Also rooms with old PVC floors had higher concentrations of DEHP and DnBP in dust than rooms with new PVC floors. There was also a trend that dust from preschools with PVC floors had higher DiNP concentration than dust from preschools with other floor types. The preschools that used foam mattresses for resting had higher DiNP concentrations than those with no foam mattresses. Most preschools had new foam mattresses, which could indicate a more common use of DiNP in new mattresses or mattress

covers compared to old mattresses that contains more DEHP. The four Waldorf preschools that participated had lower DiNP dust concentrations than the other preschools, which was expected since Waldorf orientation includes using as little plastic material as possible. No relation was found between the phthalate dust concentrations and the quantity of toys made of soft plastic in the sampled area.

Many preschools had made a plastic inventory where they removed old and soft plastic toys and material. Also many preschools recently replaced old foam mattresses used for resting. This could be due to the big chemical focus in the media and authorities and the brochures that have been sent out the last couple of years about what preschool can do to decrease the chemical exposure of children. This interest and awareness seen in the preschools was positive and hopefully the trend spreads to more preschools. Since children spend a big part of their time at preschools it is an important mission for society and the government to decrease the exposure to hazardous chemicals there. Hopefully what has been done so far is just the beginning.

2. Introduction

2.1 Background

The focus and awareness of health effects of chemicals has greatly increased during the last decades. A focal point has been to evaluate and decrease potentially toxic chemicals that are used everywhere around us in clothes, toys, food, electrical devices and furniture among other things. During fetal life, infancy, childhood and adolescence, the development of organ systems undergo critical time points when chemical exposures can cause more severe effects than in adulthood. For example, young children can be more sensitive than adults to chemicals with toxic effects on the nervous system, hormone system and immune system because of the on-going development of these organ systems (Chance, 2001). In addition, children are more exposed to chemicals than adults because they eat, drink and breathe more in relation to their body weight. Also, children are more exposed to dust than adults because of their behavioural patterns, such as more pronounced hand to mouth behaviour and more time spent playing on the floor (Pohl *et al.* 2005). In their everyday life, children are exposed to many chemicals via the products they come in contact with. Adverse effects may emerge and the health outcome depends on the chemical dose and potency and if the exposure happens during critical developmental periods (Selevan *et al.* 2000). Chemical exposure from children's close environments is a current issue that has been in focus over the past decade in Sweden and in the EU. In fact, over the past decade the EU-regulations that affect chemicals has become stricter.

The use of chemicals is connected to lifestyle factors and to the high consumption of products. For example, in the year 2005 the toy turnover in Sweden was 40 000 tonnes or 30 kg per child and year (SCB, 2005). Phthalates is a group of synthesised chemicals used in many products in children's close environments, mainly in polyvinyl chloride (PVC) plastics in toys, flooring, food containers and textiles (KemI, 2014a). Phthalates have been shown to be endocrine disrupting chemicals that can cause adverse effects on the reproductive system. They are also suspected to play a role in the development of other diseases, such as asthma and allergy. Even though a number of phthalates recently have been subjected to restrictions within the EU, there are many phthalates still in use. In children's close environments, *e.g.* in preschools, old toys and furniture that might not meet the new standards may still be used. The hypothesis is that given the high number of products in the preschool environment in general, and of old products in particular, the chemical exposure of children in these environments might be of concern.

In this pilot project dust samples were collected from 30 preschool indoor environments in the Stockholm city area in the spring 2015 to analyse and evaluate the levels of phthalates in the dust samples. The six studied phthalates were diethylhexyl phthalate (DEHP), diethyl phthalate (DEP), di-n-butyl phthalate (DnBP), diisononyl phthalate (DiNP), diisobutyl phthalate (DiBP) and butyl benzyl phthalate (BBzP). In addition, a phthalate-substitute was studied;1,2-cyclohexane dicarboxylic acid diisononyl ester (DINCH). The Stockholm city Environment Department will continue the dust sampling from an additional 70 preschools during the autumn of 2015. In addition to the evaluation of phthalates in preschool environments, this thesis project is part of a larger project, commissioned by the Swedish

Environmental Protection Agency (Swedish EPA) and performed at the Institute of Environmental Medicine, Karolinska Institutet, that will result in a time trend evaluation of the chemical exposure of Swedish children. In this larger project the dust sampling will be complemented with urine sampling and hand wipe sampling from 100 children between 3.5 and 4.5 years of age attending the studied preschools. In the larger project the samples will be analysed for more chemicals than phthalates.

The main aims of this thesis project are to:

- Evaluate a test method for dust sampling in preschools and develop a preschool questionnaire used for dust sampling

- Evaluate the phthalate concentrations in dust from 30 preschools

- Identify and evaluate factors that influence phthalate levels in dust in the preschool environment

2.2 Phthalates

There are 50-100 plasticisers in commercial use in the EU today and phthalates are the most common ones. One million tonnes of phthalates are produced every year in the EU, out of which 90 % are used in PVC products (ECPI, 2015). Phthalates are derived from phthalic acid and they are used mainly in PVC plastic products like floors, toys, food containers and wall papers. Other products containing phthalates are binding agents, rubber, glue, paint, cables, solvents etc. (Heudorf *et al.* 2007). Since the phthalates are only very weakly chemically bound to the PVC they can leak and migrate to the air, food, water, skin etc. Children are exposed to phthalates mainly through food, but because of the hand to mouth behaviour they are also exposed via dust inhalation and dust ingestion (Heudorf *et al.* 2007). The chemical structures of the six phthalates that are analysed in this study are shown in Figure 1.

DEHP was the most commonly used phthalate a few decades ago and still is in some parts of the world. However, it has been an important goal worldwide to decrease the use of DEHP because of the toxic effects. The phthalate DiNP has been a suitable alternative to DEHP, for example in floors, because both phthalates are persistent (ECHA, 2010). Around 2010, DiNP was used to a higher extent than DEHP in Europe. The use of DEHP in Western Europe decreased from 595 000 tonnes/year in 1997 to about 210 000 tonnes/year in 2009. The total use and production of plasticisers including phthalates has decreased in Europe, but since many industries that produce and use PVC have moved to countries outside of Europe the global use and production increase (ECHA, 2010). Today, when consumers want phthalate-free products, phthalates like DiNP can be replaced by other esters with similar properties, such as DINCH (The Danish EPA, 2010). Surveys from several European countries show that DINCH is one of the most common phthalate alternatives used in toys and child care articles, *e.g.* it has been found in inflatable toys and changing pads (The Danish EPA, 2010).

It is difficult to find information about what products specific phthalates are used in. The most common phthalates in PVC worldwide are DEHP, DiNP and diisodecyl phthalate (DiDP) (The Danish EPA, 2010). PVC is most widely used in the building industry, in everything from facades, cables, floors and walls (KemI, 2014b). In the beginning of this century, Swedish producers of cables and cords started to replace the phthalates banned in 2015 with other phthalates. PVC floors have a smooth surface and are easy to clean and therefore they are commonly used in hospitals, schools, preschools etc. New PVC floors used in Sweden often contain DiNP and DiDP and in phthalate free floors DINCH is a common plasticizer. Old PVC floors commonly contain DEHP. Furniture with leatherette, plastic furniture, the underside of carpets, oilcloths etc usually contain phthalates. In Sweden and Denmark DiBP has been found in furniture and both DEHP and DiNP have been found in bath toys. In toys, phthalate alternatives are found in higher levels than real phthalates. In PVC floors (no production year mentioned) DiNP and DEHP have been found (KemI, 2014a). DEP is mainly found in personal care products and cosmetics (Koniecki *et al.* 2011).

In July 2011 the EU toy safety directive (2009/48/EC) was implemented in all the member countries (EUR-Lex, 2009). Among other things, this directive includes a requirement for CE-marking, which means that the products should meet the safety regulations of the EU. The chemical regulations in the directive started to apply in July 2013. There are no specific regulations about phthalates in the directive but it says that the chemical content of toys is not allowed to cause any health risks for people with a focus on children (EUR-Lex, 2009). From February 21st 2015 it is a general prohibition for the use of DEHP, DnBP, BBzP and DiBP in the EU if not a special permission is given (EU-commission, 2014). The mentioned phthalates are listed on EU's Authorisation list and the prohibition applies for all products in the EU market (ECHA, 2015a).

The chemical legislation in the EU is called REACH (Registration, Evaluation, Authorisation and restriction of Chemicals) and came into force in 2007. Since then DEHP, DnBP and BBzP are forbidden in toys and childcare products above 0.1 % of the weight mass. Childcare products include products with the purpose to help children sleep, relax, feed, chew or to help with their hygiene (ECHA, 2013a; Reach, Annex XVII, point 51). They are also forbidden in electrical products for consumers (ECHA, 2013a; Reach, Annex XVII, point 30). The phthalate DiNP is forbidden above 0.1 % of weight mass in child products that children can put in the mouth (ECHA, 2013a; Reach, Annex XVII, point 52). DiBP is forbidden in electrical products and is listed on ECHAs' Candidate list of Substances of Very High Concern (SVHC), which means that companies within the EU that produce, import or sell products that contains >0.1% of the substance are obligated to inform the consumers about the content (ECHA, 2015a). It should be easier to implement a chemical restriction in REACH than it is today. The implementation costs between 5-10 million Swedish crowns or about 0.5-1 million Euro (KemI, 2014a). Fewer resources are required to restrict chemicals in the EU toy safety directive (2009/48/EU) and the RoHS-directive (2011/65/EU) than in REACH. The companies that produce or sell products have the responsibility for the safety of the product and for the correct chemical marking. One example is CE-marking, which means that the article meets the safety requirements of the EU (KemI, 2014a).

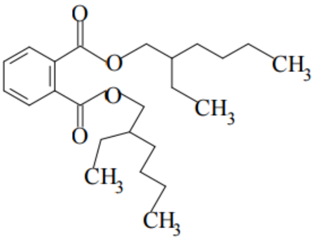
DEHP

IUPAC name: Bis(2-ethylhexyl)phthalate

Cas number: 117-81-7

Molecular formula: $C_{24}H_{38}O_4$

Structural formula:



ECHA, 2008c

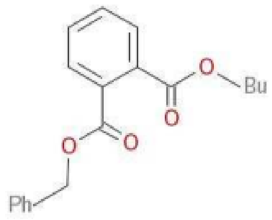
BBzP

IUPAC name: Benzyl butyl phthalate

Cas number: 85-68-7

Molecular formula: $C_{19}H_{20}O_4$

Structural formula:



ECHA, 2008b

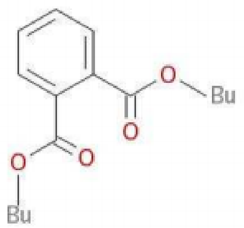
DnBP

IUPAC name: Dibutyl phthalate

Cas number: 84-74-2

Molecular formula: $C_{16}H_{22}O_4$

Structural formula:



ECHA, 2008d

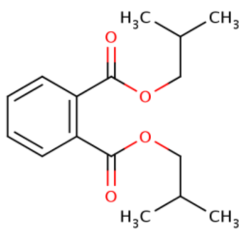
DiBP

IUPAC name: Diisobutyl phthalate

Cas number: 84-69-5

Molecular formula: $C_{16}H_{22}O_4$

Structural formula:



ECHA, 2009

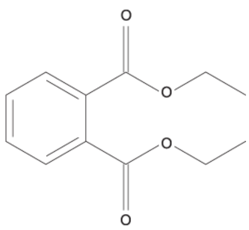
DEP

IUPAC name: Diethyl Phthalate

Cas number: 84-66-2

Molecular formula: $C_{12}H_{14}O_4$

Structural formula:



ECHA, 2015b

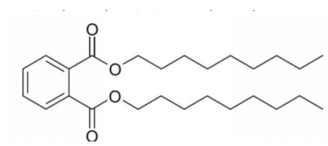
DiNP

IUPAC name: Diisononyl phthalate

Cas number: 68515-48-0, 28553-12-0

Molecular formula: $C_{26}H_{42}O_4$

Structural formula:



ECHA, 2013b

Figure 1. Chemical structures of the six phthalates in the study.

2.3 Toxic effects of phthalates

The most studied and recognised toxic effect of phthalates is that they can impair reproduction and fertility. The phthalates DEHP, BBzP, DnBP and DiBP are categorised by ECHA as reproductive toxicants that after long term exposure may cause harm to the unborn child and possibly impair fertility (EU-commission, 2014). Phthalates are endocrine disrupting chemicals with anti-androgenic properties. It has been shown that DEHP is reprotoxic and can affect the testicular function and the weight of male reproductive organs (ECHA, 2008). BBzP has been shown to cause developmental effects like decreased testis weight and decreased fertility after *in utero* exposure (ECHA, 2008b; Nagao *et al.* 2000). The phthalate DnBP has been shown to cause an abnormal development of the Sertoli cells and may cause testicular adverse effects (Fischer *et al.* 2003). DiBP may cause adverse effects on male reproduction and developmental toxicity (ECHA, 2009) and DiNP has been shown to cause adverse testicle effects in animals (Clewell *et al.* 2011; Boberg *et al.* 2011). DINCH has been shown not to be carcinogenic, reprotoxic or developmentally toxic and to have a low acute toxicity (The Danish EPA, 2010).

Another effect that has been investigated recently is the link between phthalate exposure via indoor air and dust and asthma and other allergic symptoms (Jaakkola & Knight, 2008). It was concluded to be a low risk of adverse effects for humans in a non-occupational situation after phthalate inhalation (Larsen et al. 2004). DEHP and the phthalate metabolite MEHP (Mono-2-ethylhexyl phthalate) inhalation studies have shown respiratory effects in mice and rats at exposure levels high above human indoor levels (Larsen *et al.* 2004; Klimisch *et al.* 1992). Repeated exposure to high concentrations of DEHP via inhalation has been shown to elevate the levels of inflammatory cells, lymphocytes and neutrophils in the lungs in mice (Larsen *et al.* 2007) but the risk of adverse pulmonary effects after inhalation of air and dust have been shown to be small (Hansen *et al.* 2007). Monophthalates, such as MEHP, can cause immunosuppression *in vitro* in incubated human epithelial cells (Jepsen *et al.* 2004) and *in vivo* in mice injected subcutaneously (Larsen *et al.* 2001). In summary, these experimental studies point towards a low risk of adverse respiratory effects from phthalate dust exposure.

Epidemiological studies, in general, show positive associations between phthalate indoor exposure and asthma and allergic symptoms in children. Several case reports have shown that exposure to PVC fumes and DEHP fumes in the working environment are very likely to cause airway irritation, asthma and other respiratory adverse effects (Brunetti & Moscato 1984; Butler *et al.* 1981; Pauli *et al.* 1980). Other epidemiological studies have found positive associations between phthalates in house dust and allergic symptoms or asthma (Hsu *et al.* 2012; Kolarik *et al.* 2008; Bornehag *et al.* 2004). In the study by Bornehag *et al.* (2004) BBzP in dust from children’s rooms were associated with rhinitis and eczema and DEHP in dust was associated with asthma. Associations between the presence of PVC floors and asthma and allergic symptoms have been shown in children from Sweden (Larsson *et al.* 2010; Shu *et al.* 2014), Norway (Jaakkola *et al.* 1999), Finland (Jaakkola *et al.* 2000) and Russia (Jaakkola, 2004). The incidence of symptoms was higher in homes where PVC floors were used in the child’s room. These epidemiological studies indicate that phthalates and PVC floors might be linked to respiratory adverse effects.

The exact mechanism behind these respiratory effects is not fully established. However, one *in vitro* study with human bronchial epithelial cell lines suggests that a mechanism behind the respiratory effects can be that phthalates induce inflammatory factors in the bronchial smooth muscle cells and increase the proliferation and migration of the cells, which leads to structural changes of the airways (Kuo *et al.* 2011).

2.4 A non-toxic environment

One environmental goal in Sweden is to achieve a "non-toxic environment" and a big part of that objective includes decreasing the toxic exposure of children. Among other things, the Swedish Chemical Agency (SCA) develops criteria for non-toxic and sustainable procurement for schools and preschools which will be a focus area 2015-2020 (KemI, 2014c). Children are exposed to phthalates, brominated flame retardants, perfluorinated compounds, organophosphate compounds etc. in their daily preschool environment (KemI, 2013a). They are exposed *e.g.* through food, toys, furniture, floors, carpets, food containers, textiles, electronic devices and mattresses.

On commission from the Swedish government, the SCA has developed an action plan on how to reach a "non-toxic environment", especially with a focus on children and youths (KemI, 2014c). The SCA has published several reports about chemical exposure of children and one is about the chemical exposure in the preschool environment (KemI, 2013a). In this report, dust samples were taken from areas above the floor in a new preschool built in 2012, an old preschool from 1987 and an apartment preschool from 1986 (KemI, 2013a). Two dust samples per preschool were analysed for DnBP, DiBP, BBzP and DEHP. In the new preschool, DEHP was the only phthalate found in levels above the detection limit, and the concentration was about 50 µg/g dust. In the old preschool, all phthalates except BBzP were above detection limit and in the apartment preschool all phthalates were above detection limit. The DEHP concentrations were higher in the old preschool than in the apartment preschool (KemI, 2013a). The SCA also analysed Swedish toys in 2012-2013 from 44 companies that produce, import or sell toys (KemI, 2013b). In this study, 23 out of 211 toys (11%) had too high concentrations of the restricted phthalates. Too high concentrations were found in *e.g.* dolls, pens, arrows, costumes and dress-up clothes and inflatable toys. Overall the knowledge about the chemical legislation for toys was low and many companies had limited awareness about the detailed rules for toys in REACH, the EU toy safety directive (2009/48/EU) and the RoHS-directive (2011/65/EU) that includes electrical toys (KemI, 2013b).

The SCA states that it is crucial to highlight the issue of chemical exposure to children in the UN and the EU and to focus on education to increase the knowledge of sustainable use of chemicals. After a web based questionnaire in Sweden, it was concluded that especially parents to small children wanted to receive more comprehensible and accessible information about chemicals in purchased products and the related health risks (KemI, 2014c). During the last few years several brochures addressed to parents, schools, preschools and municipalities have been made. One example is an easy-to-read brochure made by Stockholm city Environment Department containing ten suggestions towards a "non-toxic" preschool (Stockholms stad, 2014). The tips include getting rid of or replacing soft plastics, electronics and foam products, having suitable cleaning routines, choosing ecological food, avoiding canned food and not heating food in plastic containers (Stockholms stad, 2014). In Sweden both Stockholm and Göteborg municipalities have developed chemical plans with guidelines on how to decrease the chemical exposure. In the chemical plan of Stockholm it is written that Stockholm city should cooperate with municipalities, scientists and organisations to develop methods to perform and monitor measures to decrease the exposure of children (Stockholms stad, 2013). Among other things, there are plans to educate the directors and teachers in preschools, to work with procurement and monitor the chemical exposure of children and to make a pilot study with "chemically smart families".

Children are exposed to the same product chemicals as adults plus those present in toys and childcare products. Many hazardous chemicals can be found in air and dust because they leak from products, furnishing and construction (The SSNC, 2013). Restrictions for phthalate concentrations in products are mainly applied for toys, electrical and chemical products but children can also be exposed from products that are not restricted, such as floors, shower curtains, imitation leather (The SSNC, 2013). In 2013, the Swedish Society for Nature Conservation (The SSNC) started a project called "operation non-toxic preschool" where they made an inventory at 129 preschools in 41 municipalities in Sweden (The SSNC, 2014). The project showed that children in Swedish preschools are exposed to many products that contain hazardous chemicals. Most preschools, about 75 %, had more plastic toys than wooden toys, half the preschools had toys that were not produced to be toys, a third used electronic devices as toys and 75 % had foam mattresses with plastic covers that may contain phthalates (The SSNC, 2013). The report by SSNC (2014) gives preschools advice on how to decrease children's exposure to hazardous chemicals, similar to those in the brochure by the Stockholm city (2014). Among the general advice are:

* Choose products produced within the EU

* Remove soft plastic toys

* Don't accept toys from parents

* Don't let children play with things that are not made to be toys

* Look for environmental labelling before purchase

* Choose more ecological food

* Don't use wet wipes and vinyl gloves when changing napkins

In addition, there are more detailed tips about specific product groups. When it comes to plastics and phthalates is it advised to avoid soft plastic, especially old soft plastic that smells, not to use plastic in combination with heated food or drink, avoid recycled plastic and plastic produced outside the EU (The SSNC, 2014).

2.5 Phthalate concentrations in dust

There are a number of studies performed that examine phthalate concentrations in indoor dust and several of them in a preschool environment. The three methods often used for collection of dust are to vacuum areas above the floor, vacuum the floor and collect dust from vacuum cleaner bags. In general, concentrations of phthalates in dust are higher in preschool indoor environments than in bedrooms (Langer *et al.* 2010; Bergh *et al.* 2011). Bekö *et al.* (2013) estimated the mean ingestion of dust from a Danish study performed by Langer *et al.* (2010) with 500 children 3-6 years old. The intake of DEHP present in dust was estimated to 1.04 µg/kg bw/day. The corresponding intakes of DEP, DnBP, DiBP and BBzP were 0.074, 0.067, 0.092 and 0.041 µg/kg bw/day, respectively (Bekö *et al.* 2013). In the study by Langer *et al.* (2010) urine samples were also collected from the children. The concentration of phthalate metabolites in the urine were positively correlated to the phthalate concentrations of indoor dust, except for DEHP (Langer *et al.* 2014).

3. Methods

3.1 Recruitment of preschools

Directors of preschools were contacted by phone and invited to participate in the study. Participation included both dust sampling and recruitment of children for urine and hand wipe sampling. When a preschool accepted the invitation to participate in the study a time for dust sampling was booked. Of the contacted preschools 34 of 53 or 64 % accepted the invitation. Dust samples were collected from 30 preschools between February and March 2015. The districts in Stockholm visited were Bromma (n=12), Kista-Husby-Akalla (n=8), Kungsholmen (n=9), Södermalm (n=1) and Älvsjö (n=1). Four preschools had a pedagogical approach called Waldorf, which includes an indoor and outdoor environment with very little plastic and foam materials and instead a focus on natural materials and fabrics. Two preschools had an outdoor pedagogical approach called "ur och skur", which means that the children spend more time outdoor than in regular preschools.

3.2 Preparing materials and filters

Before each preschool visit a sampling bag was prepared that contained the filters, contact information and a map to the preschool, the inspection questionnaire, aluminium foil, plastic bags for the dust samples, nitrile gloves and a rule to measure rooms and mats. The weight of the cellulose filters used was 0.5 g (± 0.02 g) each but they were preweight to get the precise weight. Nitrile gloves and sterile tweezers were used when weighing the filters and the filters were wrapped in aluminium foil after the weighing to avoid contamination. The filters used for the bisphenols and phthalates analyses were purified for 10 minutes in 10 mL 99.9 % methanol (Chromasolv, Sigma-Aldrich) to remove possible contamination. After purification they were weighed again. Four blank filters and ten field blank filters were also weighed and wrapped in aluminium foil. The blank filters were used during chemical analysis to correct for contamination from the production of the filters and the handling of the filters from the weighing and receive the correct phthalate dust concentrations. The field blank filters were used to correct for contamination, if any, from the handling of the filters at the dust sampling to receive the correct phthalate dust concentrations.

3.3 Sampling

As described in the study by Björklund *et al.* (2009) dust samples were collected by an industrial strength vacuum cleaner (Alto AERO 840). For every preschool one dust sample was collected for the analyses of phthalates and bisphenols. A cellulose filter was attached to a styrene-acrylonitrile holder and inserted in a nozzle made of polypropylene attached to the intake nozzle of the vacuum cleaner. The lid of the filter holder was removed before sampling and afterwards the lid was put back and the holder was wrapped in aluminium foil and sealed in a plastic bag. Dust was collected until the filter was completely covered. When the field blank filters were sampled the vacuum cleaner was immediately switched off and the filter removed. The samples were stored in a -20 °C freezer at Karolinska Institutet. To eliminate large particles from the sample a strainer was placed on the filter. Dust was collected from playrooms in departments for 3-4 year old children. Dust was mainly collected from window frames, door frames, shelves and on top of cupboards. Sampling close to plastic materials was avoided as much as possible. When the dust quantity was not enough additional dust was vacuumed from other places with as little plastic materials as possible, like mouldings and metal ventilation pipes. In a few preschools, dust from two small adjacent rooms was combined. Field blank samples were taken at every third preschool.

3.4 Questionnaire

After the dust was collected in a preschool the director of the preschool or preschool teachers answered questions from the questionnaire orally. The questions covered *e.g.* the construction year of the building, cleaning routines, quantity of plastic or foam toys and furniture, type of mattresses and pillows used for rest, size and type of the room, type of floor and number of electrical devices in the room. The visit took about 1-2 hours in total. The questionnaire was evaluated after the first preschool visits and questions were changed or removed and answering alternatives were added.

3.5 Chemical analysis

The samples were analysed at the Swedish Environmental Research Institute (IVL) using a valid method described in the study performed by Bergh *et al.* (2012). The only difference was the dust extraction method. In the analysis of the present study the dust samples were extracted from the filters using microwave-assisted extraction and in the study by Bergh *et al.* (2012) an ultra sound bath was used. In the microwave-assisted extraction dust was placed inside pre-cleaned high quality Teflon vessels together with the extraction solvent (10ml acetone: n-hexane, 1:1 v/v) and heated to a controlled temperature with microwave power. After the high productivity microwave-assisted extraction, plasticizers were exported from the matrix. The dust weights in the present study ranged between 2.5 and 206 mg and the limit of detection of the analysed phthalates were 0.001-0.25 µg/g dust.

3.6 Database development and statistics

The IBM SPSS Statistics (version 22) was used for the statistical analyses. A database was created consisting of the preschool inspection questionnaires, the dust sampling forms and the results from the chemical analyses. The questionnaires and sampling forms were encoded and manually added to the database.

The data was not normally distributed according to the D'Agostino-Pearson omnibus test, therefore the nonparametric tests Mann-Whitney U-test and Spearman's rank correlation test were used. Box plots were made for the most significant and interesting relations. Of all the parameters in the questionnaire, 16 were analysed in binary groups to compare the concentrations of six phthalates and the alternative DINCH. For the "scale variables" (*e.g.* construction year, m^2 per child and year of the floor) the Spearman's rank correlation test was used to search for relations with the concentrations of the seven chemicals. Significant relations were further evaluated by Scatter plots.

3.7 Preschool characteristics

The preschools were of different areas, construction years and number of children. Of the 30 preschools four were of Waldorf orientation and a total of nine were private. Eight preschools were a part of an apartment building. The number of children in the preschools ranged from 17 to 115, with a mean of 59 children. In the departments where the dust samples were taken the number of children ranged from 10 to 35 with a mean of 19 children. Preschool total indoor area ranged from 90 to 1380 m^2 and the size of the room where the dust sample were taken ranged from 10 to 63 m^2. The number of square meter per child ranged from 5.3 to 16.4 with a mean of 10.3 m^2/child. Construction year ranged from 1890 to 2013. To include renovation in the analysis, the construction year was replaced with the year of renovation if a large renovation had been made on floors, roofs, walls etc after the year 2000. If no renovation had been performed after 2000, the construction year was not replaced. Using this definition, the "construction or renovation year" ranged from 1890 to 2014. A selection of descriptive statistics is shown in table 1.

The cleaning routines varied between the preschools. Half the preschools had a hired cleaner and half had a cleaning company. A majority of the preschools were cleaned five days a week and at night time. The last major house cleaning had been performed 3-50 months before the dust sampling except for the newest preschools where no major house cleaning had been done yet. Floor cleaning had been performed in 21 preschools using polish (n=10) or wax (n=3) and in eight preschools it was unclear whether polish or wax had been used. In 11 preschools toys from parents were accepted but there was a general trend to stop accepting toys from parents and get rid of these toys if they had been accepted in the past.

The indoor furniture and toys were made of a variety of materials and several preschools had started to get rid of products that might contain chemicals of concern. Half the preschools used mattresses at rest, five used sheepskins and ten used another material or did not use any mattresses at all. As few as five preschools had foam mattresses in the sampling room, on the floor or in a wardrobe, and 19 had foam mattresses somewhere in the room or elsewhere in the department where the samples were taken. Interestingly, many preschools had replaced the old mattresses with new ones during the last year, which seemed to be an intervention that had been prioritised in a few districts. Only four out of 19 preschools had mattresses that were older than eight years. The floor types in the sample rooms were PVC (n=13), linoleum (n=14) or wood (n=3). Out of the 13 PVC floors eight were from later than 2007, which means that the majority were new.

Plastic furniture were present in 13 of the sampled rooms and five of them were from before 2007. A majority of the plastic furniture was stools and chairs. In 13 of the sampled rooms, foam furniture were present and seven rooms had a foam sofa. The amount of plastic toys in the room, lego and pearls excluded, ranged from 0-8.5 crates (dimension: 20x30x25 cm) with a mean number of 2.9 crates. The sampled rooms in the Waldorf preschools and one with outdoor pedagogical approach contained no plastic toys. When including toys in the whole department, a range from 0-14 crates of plastic toys was found. Foam toys in the room ranged from 0-4 crates (mean 1.5 crates). The whole department had a range of 0-14.5 crates of foam toys. Out of the 30 preschools 17 had no foam toys. Electrical devices are common in preschool areas and 17 preschools had two or more devices in the sampling room, seven preschools had one device and six preschools had no devices.

One preschool participated in the project "operation non-toxic preschool" performed by the SSNC and 9 preschools have done their own chemical inventories where *e.g.* soft plastic toys were removed etc. In 21 rooms the sampling was made close to plastics, mostly containers or consumable material, because it was impossible to find enough dust other ways.

Table 1. The number of preschools (n) in each category and the minimum, maximum and mean result of the variables.

Variabel	n*	Min.	Max.	Mean
No. of children in preschool	30	17	115	59
No. of children in department	30	10	35	19.1
Preschool area (m^2)	29	90	1380	633.9
Area (m^2) per child	29	5.3	16.4	10.3
Room area (m^2)	30	9.9	63	36
Construction or renovation year	30	1890	2014	1982
Year the floor was inserted	24	1974	2014	1999
Year the PVC floor was inserted	13	1975	2013	2002
No. of plastic furniture	13	1	9	2.9
No. of crates with plastic toys in room	30	0	8.5	2.9
No. of crates with plastic toys in the entire department	30	0	14	5
No. of crates with foam toys in room	30	0	4	1.5
No. of crates with foam toys in the whole department	30	0	14.5	2.1
Age (year) or youngest child	30	1.5	4	3.1
Age (year) or oldest child	30	4	7	5.5

* Number of preschools that answered the specific question.

4. Results

4.1 Concentrations of phthalates in dust

The analysed chemical concentrations in dust showed a wide range for all compounds except for DEP (table 2). A sample with a concentration of 4603 µg DEHP/g dust was considered to be an outlier and was removed from the analyses of DEHP. This extremely high DEHP concentration was obtained when the sample was taken from a cable cord, which is presumed to have contaminated the sample. The field blanks did not show any contamination that affected the results.

Table 2. Descriptive statistics of the minimum (min), maximum (max), mean and standard deviation (SD) of the concentrations (µg/g dust) in dust from the 30 preschools.

Chemical	n*	Min.	Max.	Mean	SD
DEHP	29	52.4	945.0	425.1	268.1
DEP	30	0.30	1.6	0.7	0.25
DnBP	30	1.3	92.7	26.8	23.2
DiNP	30	57.8	5605.2	1045.3	1362.0
BBzP	30	1.0	220.3	38.2	52.0
DiBP	30	1.0	71.7	11.1	15.1
DINCH	30	9.6	3681.6	244.9	699.3

* Number of preschools

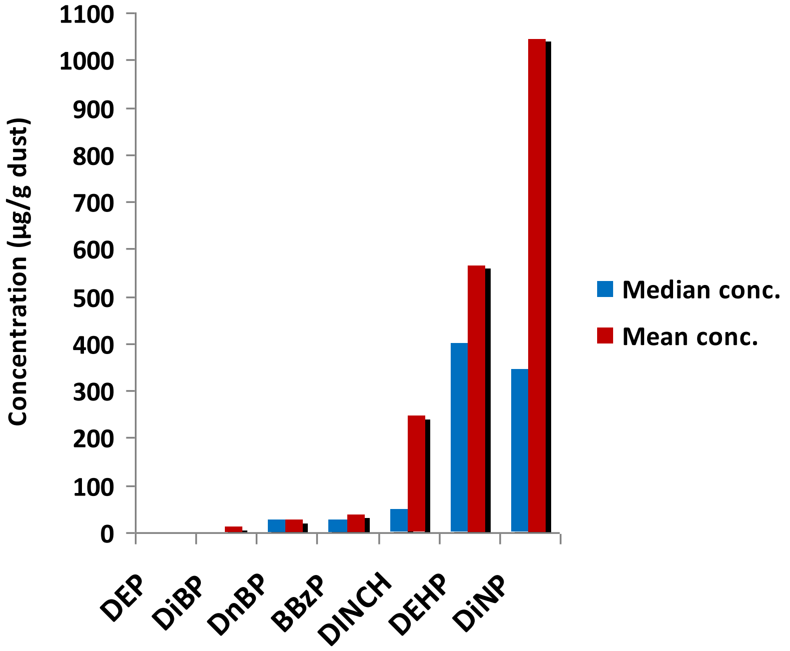


Figure 2. The median and mean phthalate concentrations (µg/g) of DEHP (diethylhexyl phthalate), DEP (diethyl phthalate), DnBP (di-n-butyl phthalate), DiNP (diisononyl phthalate), DiBP (diisobutyl phthalate), BBzP (butyl benzyl phthalate) and DINCH (1,2-cyclohexane dicarboxylic acid diisononyl ester) in dust from 30 preschools. The DEP mean and median concentrations (1-2 µg/g) were to low to be shown in the figure.

4.2 Relations and correlations

From the questionnaire 16 parameters were analysed for relations with the concentrations of the 7 chemicals DEHP, DEP, DnBP, DiNP, BBzP, DiBP, and DINCH. The parameters were if the preschool was a Waldorf preschool, had an area of <10m2 per child, had a construction or renovation year <1985, cleaning >4 times per week, had a major cleaning during the last year, had PVC floor in the sampled room, accepted toys from parents, had foam mattresses for resting, had a room size <35 m^2, had plastic or foam furniture, had >1 crate of plastic toys, had >1 crate of foam toys, had >19 children in the department, had done a plastic inventory and if the sampling was made close to plastic. The parameters and the mean and median concentrations of the two binary groups of each parameter are shown in table 3. Chosen results are described and shown together with scatterplots and boxplots in figure 3-7.

An area of less than 10 m^2 per child was related with high DnBP concentrations and major house cleaning during the last year was related with high DEP concentrations. Preschools that accept toys from parents showed a relation with high concentrations of DnBP. There was no link between high phthalate concentrations in dust in the preschools where the dust sampling was performed close to plastic materials. Preschools that had made a plastic inventory had lower concentrations of DiNP than other preschools.

No relation was found between phthalates in dust and the number of crates with plastic or foam toys in the sampling room, nor with plastic furniture or foam furniture in the sampling room. There were no significant relation between the preschools that clean every day compared to those that clean more seldom or between any of the 16 preschool parameters and the concentrations in dust of BBzP, DiBP or DINCH.

Table 3. The mean and median phthalate concentrations (µg/g) in dust from different preschools. The Mann Whitney test was used to see if there was a significant difference in phthalate concentrations between the groups "yes" and "no".

Parameter		n#	DEHP	DEP	DnBP	DiNP	BBzP	DiBP	DINCH
Waldorf preschool?	Yes	4	261.5/150.6	0.93/0.78	45.5/47.6	**115.3/115.5***	20.1/8.1	5.3/3.3	94.8/77.4
	No	26	444.0/398.7	0.70/0.66	23.9/20.6	**1188.4/459.8***	41.0/26.1	12.0/7.2	268.0/43.3
Area <10m² per child?	Yes	12	483.4/458.7	0.81/0.68	**38.5/28.7***	729.9/256.7	51.6/22.8	9.5/7.8	52.3/47.1
	No	17	403.9/397.5	0.68/0.63	**19.8/15.1***	1324.0/630.0	30.5/26.2	12.7/5.5	382.6/42.5
Construction or renovation year <1985?	Yes	16	**535.4/591.4***	0.74/0.68	**36.9/29.6****	681.7/330.9	43.5/26.5	14.7/7.8	62.1/46.2
	No	14	**306.4/270.4***	0.72/0.63	**15.2/9.5****	1461.0/524.2	32.2/16.2	7.0/5.5	453.8/54.5
Cleaning >4 times per week?	Yes	19	433.9/323.1	0.69/0.63	19.0/15.1	1076.1/333.4	49.8/29.4	10.7/5.5	340.9/38.1
	No	10	408.5/457.5	0.80/0.68	40.2/27.4	992.1/563.7	18.2/11.2	11.8/6.8	79.1/55.7
Major house cleaning during the last year?	Yes	16	492.3/456.4	**0.81/0.78****	30.7/26.5	823.4/330.9	52.4/29.0	12.3/8.5	68.9/45.4
	No	12	372.9/265.9	**0.62/0.57****	25.8/22.3	1372.9/459.8	25.3/20.4	11.0/5.5	216.2/47.1
Is it PVC floor in the room?	Yes	13	330.4/298.4	0.64/0.58	22.8/12.3	1793.2/1551.7	20.2/16.2	8.9/3.1	479.6/48.3
	No	17	502.2/473.6	0.79/0.67	29.8/26.1	473.5/277.1	44.3/26.2	12.8/8.1	65.4/44.1
Accept toys from parents?	Yes	11	485.5/458.7	0.69/0.64	**35.9/34.6***	906.8/333.4	46.5/19.7	17.8/8.0	125.7/44.1
	No	19	388.3/311.1	0.75/0.69	**21.5/14.3***	1125.5/356.0	33.4/26.2	7.2/5.5	313.9/48.3
Foam mattresses in the department?	Yes	19	443.0/456.4	**0.64/0.63***	24.8/25.6	**1480.3/692.3****	42.1/26.0	9.3/6.8	351.8/48.3
	No	11	391.2/306.3	**0.87/0.87***	30.3/22.2	**294.0/170.7****	31.6/18.8	14.2/5.4	60.1/31.3
Room size <35 m²?	Yes	14	395.8/298.4	0.80/0.69	33.7/28.3	1275.5/302.7	49.8/22.8	15.8/8.4	54.4/38.5
	No	16	449.0/429.3	0.67/0.64	20.8/14.7	844.0/355.4	28.1/23.6	7.0/5.5	411.5/61.1
Plastic furniture in the room?	Yes	13	456.9/428.1	0.67/0.63	28.0/19.1	1103.1/355.0	28.7/16.2	14.1/6.7	395.0/50.0
	No	17	402.8/323.1	0.77/0.67	25.9/25.6	1001.2/333.4	45.5/26.2	8.8/6.8	130.1/38.9

* = p-value <0.05 ** = p-value <0.01

= one outlier was removed from DEHP analysis which means that the total number of DEHP analysis was 29.

Table 3 continues.

Parameter		n	DEHP	DEP	DnBP	DiNP	BBzP	DiBP	DINCH
Does the room have foam furniture?	Yes	14	418.4/320.0	0.78/0.63	26.9/23.9	1279.7/344.2	33.6/23.6	6.3/5.5	132.3/44.4
	No	16	431.5/399.8	0.68/0.68	26.7/22.3	840.3/342.2	42.3/23.3	15.2/7.4	343.4/46.2
Does the room have >1 crate of plastic toys?	Yes	19	413.0/397.5	0.69/0.67	20.9/15.1	1262.4/356.0	44.0/26.0	9.8/5.5	341.6/42.5
	No	11	448.2/405.8	0.78/0.66	37.1/27.4	670.5/185.1	28.3/21.2	13.4/8.9	77.8/55.7
Does the room have >1 crate of foam toys?	Yes	10	441.6/389.7	0.67/0.65	28.3/30.1	1580.4/344.7	43.8/26.5	11.3/6.7	522.9/36.0
	No	20	416.5/397.5	0.76/0.68	26.1/20.6	777.8/316.0	35.4/20.4	11.0/6.8	105.9/49.2
Do the department have >19 children?	Yes	14	404.9/348.4	0.65/0.63	24.5/14.7	1103.2/524.2	35.3/17.5	9.1/7.2	377.6/46.3
	No	16	444.0/400.0	0.80/0.68	28.8/25.9	994.7/291.0	40.8/28.2	12.8/6.1	128.7/46.2
Has a plastic inventory been made?	Yes	11	368.5/311.2	0.81/0.66	28.3/18.8	819.2/185.1	34.6/26.0	6.2/5.4	149.6/83.8
	No	19	455.0/399.8	0.68/0.67	25.9/25.6	1176.2/563.8	40.3/21.2	13.9/7.6	300.1/42.5
Was dust sampling made close to plastic?	Yes	21	446.2/426.9	0.70/0.67	26.6/25.6	1240.0/563.7	44.6/21.2	12.3/7.6	262.3/38.9
	No	9	378.4/242.4	0.78/0.66	27.3/22.2	591.2/175.2	23.3/26.0	8.3/4.3	204.3/66.5

* = p-value <0.05   ** = p-value <0.01

4.3 Construction or renovation year

In the analysis, the construction year was replaced by the renovation year if renovation had been performed after the year 2000, otherwise the construction year was used. As seen in the scatterplots in figure 3 and the boxplots in figure 4, concentrations of DEHP and DnBP in the dust were negatively related with construction/renovation year (p<0.01, Spearman´s test). Figure 4 shows that the preschools constructed or renovated before 1985 had higher phthalate concentrations in the dust compared with the preschools constructed or renovated after 1999 (no preschools were built between 1985 and 1999).

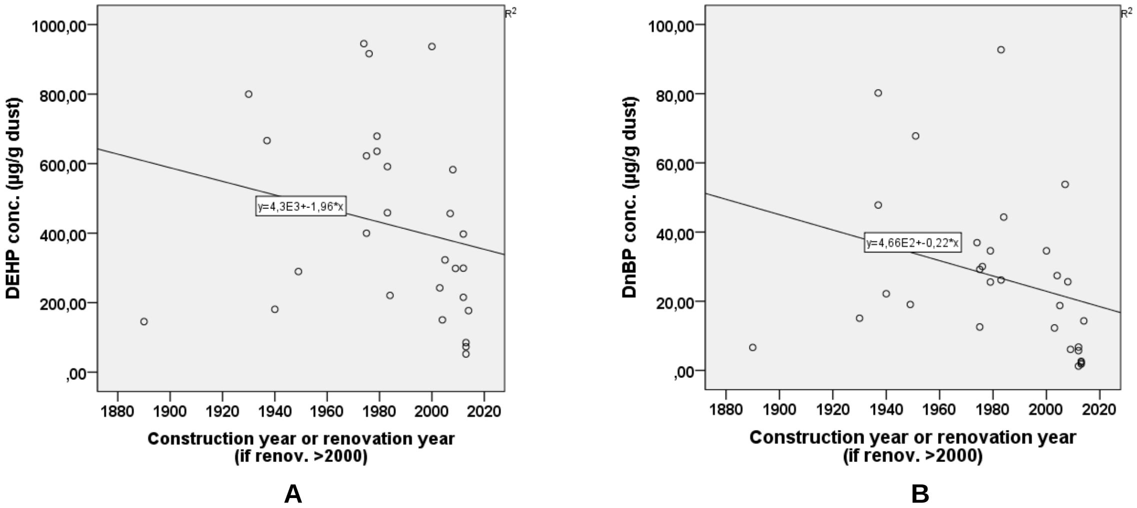


Figure 3. A) DEHP (diethylhexyl phthalate) and B) DnBP (di-n-butyl phthalate) concentrations in dust in relation to construction or renovation year. The p-values 0.006 (A) and 0.003 (B) were obtained using Spearman's rank correlation test.

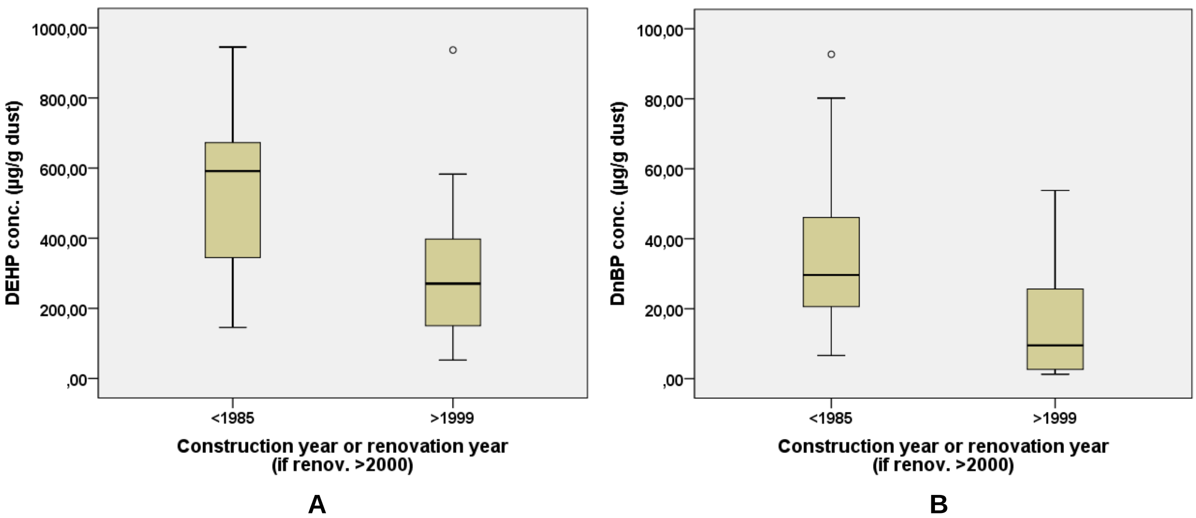
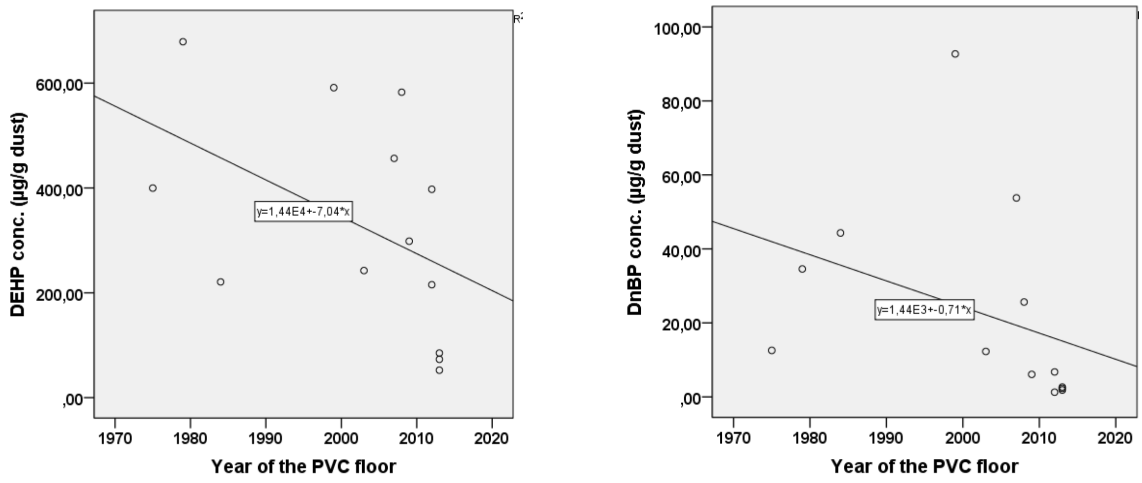


Figure 4. A) DEHP (diethylhexyl phthalate) and B) DnBP (di-n-butyl phthalate) concentrations in relation to a construction or renovation year 1890-1988 and 2000-2014. The preschools with phthalate concentrations far from the rest of the group are symbolised with circles.

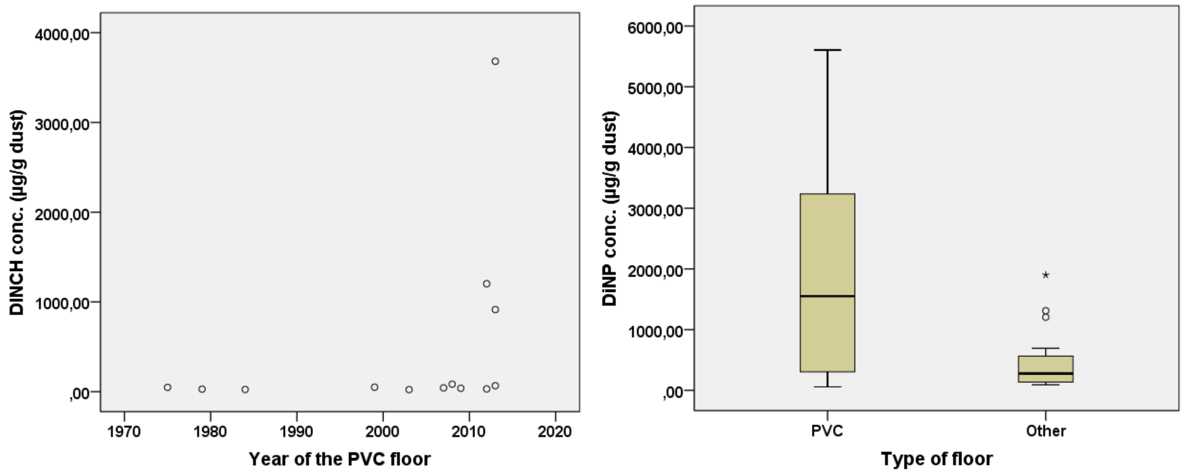
4.4 Floor type and year the PVC floor was inserted

In the 13 preschools with PVC floor, the floor was inserted between 1975 and 2013. As seen in figure 5 it was a negative relation between year of the PVC floor and DEHP (A) and DnBP (B) concentrations in dust with p-values of <0.01. Figure 6A shows the dust concentrations of the phthalate alternative DINCH in preschools with PVC floors. Three of the preschools with floors from later than 2011 showed high concentrations of DINCH in dust, whereas in the preschools with PVC floors from before 2011 the DINCH concentrations in dust were negligible. The thre preschools with PVC floor and high DINCH concentrations (914-3682 µg/g dust, figure 6A) were the only preschools with higher DEHP concentrations than 250 µg/g dust. As seen in figure 6B the DiNP concentrations in dust from preschools with PVC floor were significantly higher than those in dust from other preschools (p-value <0.05).



A B

Figure 5. A) DEHP (diethylhexyl phthalate) and B) DnBP (di-n-butyl phthalate) concentrations in relation to year of the PVC floor. The The p-values 0.006 (A) and 0.002 (B) were obtained using Spearman's rank correlation test.



A B

Figure 6. DINCH (1,2-cyclohexane dicarboxylic acid diisononyl ester) concentrations in relations to year of the PVC floor is shown in A and DiNP (diisononyl phthalate) concentrations in dust from preschools with and without PVC floor is shown in B. The preschools with phthalate concentrations far from the rest of the group in B are symbolised with circles and asterisks.

4.5 Other results

The preschools with foam mattresses present in the department where the dust samples were taken had significanlty higher ($p<0.01$) DiNP concentrations in dust than those without mattresses (figure 7A). In figure B it is shown that the preschools that had made a plastic inventory had lower DiNP concentrations than the preschools that had not made an inventory. Waldorf preschools had significantly lower ($p<0.05$) DiNP concentrations in dust than other preschools (figure 7C).

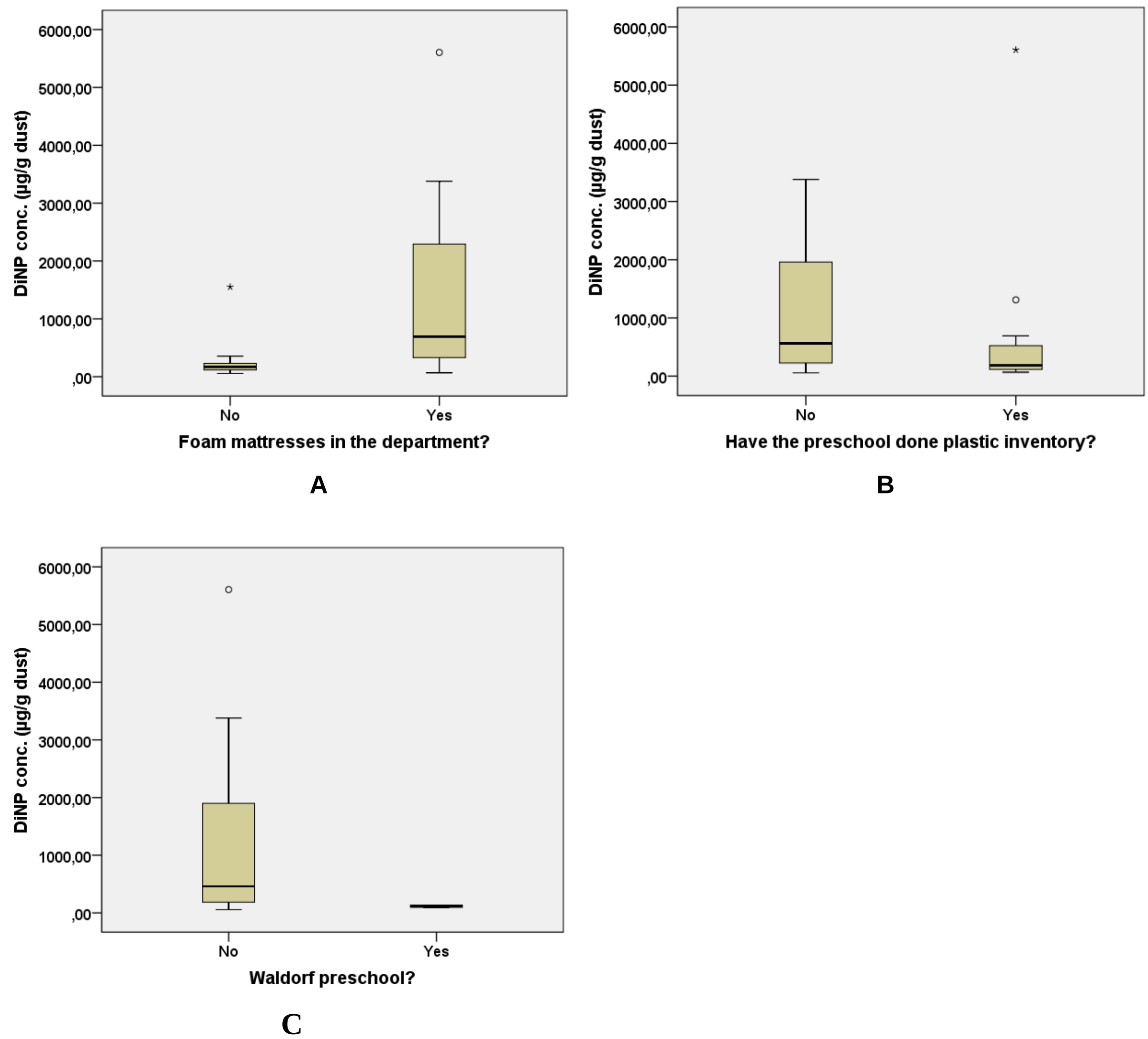


Figure 7. A) DiNP (diisononyl phthalate) concentrations in dust from preschools with or without foam mattresses, B) DiNP concentrations in dust from preschools that had performed or not performed a plastic inventory and C) DiNP concentrations in dust from preschools with and without a Waldorf pedagogical approach. The preschools with phthalate concentrations far from the rest of the group are symbolised with circles and asterisks.

4.6 The questionnaire

The preschool inspection questionnaire generally worked well for the preschool visits. However some questions did not result in enough variation in the answers, *e.g.* no preschool had plastic wallpapers, plastic carpets or used cernit clay. The preschool questionnaire was gradually optimised and changed throughout the inspections. After the 30 preschool visits and dust samplings were finished the answers were evaluated and a suggestion was made of which questions to prioritise when the sampling continues in the autumn of 2015. These suggestion included removing certain questions about cleaning routines, pillows, blankets and decreasing the number of answer alternatives of *e.g.* the material of mattresses and pillows.

5. Discussion

5.1 Construction and renovation year

Preschools built before 1985 had higher DEHP and DnBP dust concentrations than preschools constructed or renovated after 1999. There were also negative correlations between DEHP/DnBP concentrations in dust and construction/renovation year. This was expected because the levels of phthalates in building materials have most likely decreased over time. Another explanation to this negative relation could be a lower phthalate concentration in *e.g.* toys and furniture in new than in old preschools. A similar link was found in a study performed by Bornehag *et al.* (2005), with higher DEHP concentrations in dust from building with the construction year <1960 compared to buildings with the construction year >1960.

5.2 PVC floor

Furthermore, it was expected that preschools with PVC floors should be associated with higher concentrations of phthalate in the dust than preschools with other floor materials (linoleum or wood). Spearman´s test showed that among preschools with PVC floors the highest concentrations of DEHP and DnBP in dust were found in those with oldest PVC floors, probably due to replacement by other chemicals in new PVC floors. No preschool with a PVC floor from before 2011 contained DINCH in the dust but three preschools with PVC floors from after 2011 contained the phthalate alternative. This might indicate that alternatives like DINCH are more common in newly produced PVC floors since it can be a marketing advantage to be able to label the floor phthalate free. Since DINCH was the only phthalate alternative analysed, other alternatives might have been used as well in new PVC floors. In conclusion DINCH is a new phthalate alternative only found in dust from new preschools with new PVC floors.

5.3 Mattresses, inventory and Waldorf preschools

PVC floors often contain phthalates and these chemicals are also found in foam mattress covers at preschools (KemI, 2013a). The preschools with foam mattresses in the department where the dust samples were taken had higher DiNP concentrations than the preschools with no foam mattresses, which was expected since most foam mattresses were new and should not contribute to the dust concentrations of the banned phthalates (DEHP, DiBP, DnBP and BBzP). The relation between DiNP concentrations in dust and foam mattresses might be an indication of a more frequent use of DiNP in mattress covers in recent years.

The concentrations of DiNP were lower in the preschools that had performed a chemical inventory, often with a focus on soft plastic toys, clothes, bags, paints etc, compared to the preschools that had not done an inventory. The fact that many preschools had made a plastic inventory and replaced the old mattresses could be due to the extensive chemical focus in the media, authorities and the brochures that have been sent out in the last couple of years about what preschools can do to decrease the chemical exposure of children.

Waldorf preschools were associated with low DiNP concentrations in dust. This was expected because a part of the Waldorf pedagogics is to use non-plastic materials in the preschool.

5.4 Plastic and foam products

It was expected to find a positive relation between phthalate concentrations in dust and amount of plastic products (toys and furniture), but this was not the case. This can be because the plastic furniture mostly was stools and other small furniture and they might not have affected the concentrations much since the amount of plastic was small. The hard plastic toys generally contain much less plasticizers than the soft toys and the counting of plastic toys was therefore supposed to only include soft plastics. However, it was difficult to differentiate between soft and hard plastics and in many cases both soft and hard plastic toys were included in the counting. Also, the age of the toys matter as there are more hazardous phthalates in old soft plastic toys. Potentially, a relation between soft plastic toys and phthalate levels in dust can have been obscured by the inclusion of hard plastic toys. The fact that it was a relation between preschools that performed a plastic inventory and high DiNP concentrations in dust was interesting since a big focus on an inventory was to remove plastic toys. When the parameter "has a plastic inventory been made" showed such a relation, the same was also expected for the parameter "does the room have >1 crate of plastic toys".

The fact that no relation was seen between phthalate concentrations in dust and foam furniture or toys might indicate that there are not much phthalates in foam products. Plastic covers of foam products may contain phthalates but not many toys or furniture had plastic covers. In addition, 17 preschools had no foam toys in the sampling room and in the preschools with foam toys present in the room, there was only a small variation in the amount of the foam toys.

5.5 Other results

There was a negative relation between floor area per child and concentration of DnBP in the dust. This link was not expected but the wear of the floor might be increased if the number of children per m^2 is increased, and might contribute to higher DnBP levels. High DnBP concentrations were significantly related with the preschools that accept toys from parents, which was expected and indicates that these toys may contain soft plastic from before the ban of the four phthalates. Several authorities and the SSNC recommend preschools to stop accepting toys from parents, as written in previously mentioned brochures, and most preschools visited had done so or planned to do so. In a few preschools that no longer accepted toys from parents, these products were still found in the department and needed to be cleared out.

A negative relation between cleaning frequency and phthalate concentration in the dust was expected but not found. The cleaning routines varied and it was difficult for several preschools to know the exact routines. The everyday cleaning routines usually includes vacuuming and mopping of the floors but no cleaning of areas above floor level where the dust samples were taken. It was observed that the preschools with their own hired cleaner were more satisfied with the cleaning than the preschools that hired cleaning companies. The preschools that had performed major house cleaning during the last year before the dust sampling had higher DEP concentrations in the dust than preschools without such a major house cleaning. This was not expected since the major house cleaning does include areas above floor level as opposed to the everyday cleaning. An explanation to the narrow range of the DEP concentration in dust and the very low concentrations could be that DEP mainly is found in personal care products and cosmetics and those products are not very common in preschools (Koniecki *et al.* 2011).

It is unknown to which extent phthalate alternatives are used in different products but it is obvius that children are exposed to them especially via plastic products. Even if it is better to use them than phthalates banned in 2015, the allowed phthalates and alternatives might also cause adverse effects after long time of exposure. It is difficult to measure effects of combined exposures and far from all chemicals have been tested enough. Chemicals are often not on the label of products and it should be mandatory to label products with all added substances, even those present in low concentrations. The consumers have the right to know and to make decisions based on correct and reliable information from the producers.

5.6 Previous studies

Four studies, within the same field as the present one have been performed by vacuuming areas above floor level. A Danish study analysed dust from 151 preschools and from 500 children's bedrooms for five phthalates; DEHP, DEP, DnBP, DiBP and BBzP (Langer *et al.* 2010). Dust was collected in 2008 from non-floor surfaces with filters mounted to a vacuum cleaner. DEHP was found in all samples in higher concentrations than the other phthalates. The four other phthalates were detected in more than 90 % of the preschool samples. Except for DEP the dust concentrations of all phthalates was higher in the preschools than in the bedrooms. In a Swedish study, the levels of the same six phthalates as in the present pilot study were analysed in dust collected from 346 children's bedrooms (Bornehag *et al.* 2004). Dust was collected in 2001-2002 from mouldings and shelves using cellulose filters mounted to a vacuum cleaner. The levels of the five phthalates present in the more recent Danish study by Langer *et al.* (2010) were lower in both preschools and bedrooms than those in bedrooms in the study presented by Bornehag *et al.* (2004). Two more studies have been performed by vacuuming areas above floor level. A Swedish study measured phthalates in dust and air 2008 from ten preschools, ten homes and ten offices (Bergh *et al.* 2011). In the preschools, dust was sampled in playrooms and all samples were taken from shelves, windows and doors. In 2004-2005 a Bulgarian study was performed measuring phthalate concentrations in dust from the bedrooms of 184 children (Kolarik *et al.* 2008). This method, to vacuum areas above floor level, was the most common one of the studies presented in table 5.

Other sampling methods were used in three studies, two German and one South Korean. In the first German study dust was collected in 2011-2012 from 63 preschool floors and the median DEHP concentration was 888 µg/g dust (Fromme *et al.* 2013). The same research group performed a study in 2000-2001 where ten phthalates were measured in dust from vacuum cleaner bags from 59 apartments (Fromme *et al.* 2004). In the South Korean study the concentrations of phthalates in dust were measured in 2007-2008 from floors and furniture in 84 preschools (Kim *et al.* 2011). The concentrations of phthalates in dust found in the present pilot study are similar to those presented in other studies (table 5).

Table 5. Concentrations (mean/median, µg/g dust) of phthalates in studies sorted by year of performance and number of preschools in the study (n). *=mean **=geometric mean.

Reference	Country	Origin of dust and year of dust sampling	n	DEHP	DEP	DnBP	DiNP	BBzP	DiBP
This pilot study	Sweden	Preschools, 2015	30	564/399	0.73/0.67	27/24	1045/344	38/24	11/6.8
Fromme *et al.* 2013	Germany	Preschools, 2011-2012	63	1973/888	3.4/1.4	30/21	745/302	21/6.0	39/20
Bergh *et al.* 2011	Sweden	Preschools, 2008	10	2000/1600	6.9/4.2	190/150	-	47/31	9.1/3
Bergh *et al.* 2011	Sweden	Homes, 2008	10	980/680	11/3.7	130/130	-	31/17	6.0/4
Langer *et al.* 2010	Denmark	Childrens' bedrooms, 2008	500	220/210	3.1/1.7	8.1/15	-	4.2/3.7	16.6/27
Langer *et al.* 2010	Denmark	Preschools, 2008	151	540/500	1.9/2.2	30/38	-	16.4/17	18.2/23
Kim *et al.* 2011b	South Korea	Preschools, 2007-2008	84	358.2*	13.4*	107.8*	-	336.3*	-
Kolarik *et al.* 2008	Bulgaria	Childrens' bedrooms, 2004-2005	177	960**	350**	7860**	-	320**	-
Fromme *et al.* 2004	Denmark	Homes, 2001-2002	59	775.5/703.4	44.6/6.1	56.6/47	-	86.1/29.7	-
Bornehag *et al.* 2004	Sweden	Childrens' bedrooms, 2001-2002	346	1310/770	3.1/0.0	226/150	639/41	319/135	97/45

5.7 Comparisons with previous studies

The most relevant previous studies for comparison to the present pilot study are the ones that measured phthalate concentrations in preschool dust collected from areas above the floor, i.e. the studies performed by Langer *et al.* (2010) and Bergh *et al.* (2011). As shown in table 5 the Langer study from 2010 collected dust from 151 preschools and because of the large amount of preschools included, that study is the most relevant one for comparisons. The mean and median phthalate concentrations in dust in the present study were similar to the concentrations presented by Langer *et al.* (2010) for DEHP, DnBP and DiBP, higher for BBzP and lower for DEP. This could indicate that the usage of phthalates in products is similar today as in 2008 even though a stricter EU regulation and legislation have been implemented. It takes time for phthalate products to be phased out from the market. Compared to the study made by Bergh *et al.* (2011) the present pilot study showed lower concentrations of DEHP, DEP and DnBP and the same magnitude of BBzP and DiBP. However, the comparisons between studies should be made with caution mainly because of methodological differences.

The study performed by Fromme *et al.* (2013) analysed dust from vacuum cleaning the floors playrooms in preschools. The study showed higher concentrations of DEHP than this present study and about the same levels of the rest of the chemicals. In the South Korean study (Kim *et al.* 2011b) the dust was sampled from a mix of floor and furniture and the concentrations of phthalates in dust in the present pilot study were at the same magnitude for DEHP but lower for DEP, DnBP and BBzP than in the study by Kim *et al.* (2011b).

The Bulgarian study by Kolarik *et al.* (2008) showed higher levels of all four phthalates tested in dust from children's bedrooms than in the present study, a 100-fold higher concentrations of DEP and DnBP and 2-10 times higher levels of DEHP and BBzP. Why the levels presented in this study are much higher than in the rest of the studies remain unclear. The remaining four studies with dust collected from homes or children's bedrooms show that the present pilot study generally has lower or similar concentrations of the phthalates. However, the concentrations of BBzP in dust reported by Langer *et al.* (2010) where lower than those of the present study.

The present study showed higher DiNP concentrations than the two other studies that measured this phthalate; Bornehag *et al.* (2004) and Fromme *et al.* (2013). The number of comparable studies where DiNP has been analysed are limited but a comparison between the previous studies and the present indicate that the DiNP concentrations in dust are higher today than before. It could be because the preschools generally have more products with DiNP now than before. Many of the other phthalates measured are banned today and it is therefore expected that the DiNP concentrations in dust are slightly higher today than in previous studies.

5.8 Recommendations for future studies

If the study was made again it might be possible for the preschools to be recruited in a more effective way. It is also a good idea to find out how much dust that is required for the chemical analyses before the preschool visits. Some chemicals might exist in very low concentrations, and therefore a larger dust quantity is required.

Children are exposed to all dust including the dust on cords and plastics and one dust sample had a high DEHP concentration because of sampling on a cord. This observation shows the importance of collecting dust from areas with as little plastic products as possible and it is something to really focus on this autumn when this project continues in an additional 70 preschools. Maybe a future study could include sampling from plastic materials to investigate relations with the indoor environment parameters.

Regarding the ethics it was important to have a permission to take dust samples at the preschools. The exact levels and the comparisons between specific concentrations found in given preschools must not be made public. It could be difficult for the preschools to handle the phthalate concentrations in dust, especially for one with higher levels than the others. The fact that a preschool have the highest levels does not necessarily mean that there is a health risk. It is important to have a plan before the project starts on how to handle the communication of results.

6. Conclusions

The method used in this pilot project was found to be suitable and the phthalate concentrations were comparable to those presented in previous studies. Several factors that influence phthalate concentrations in dust were identified and based on them the following conclusions can be made:

- Preschools in old buildings show a tendency of higher phthalate concentrations in dust than in preschools in new buildings.
- PVC floors, especially old ones, most likely contribute to phthalates in dust.
- Foam mattresses covers might increase the levels of phthalates in dust.
- The Waldorf preschools in the study generally had lower phthalate concentrations in dust than other preschools.

A recommendation is to do a plastic inventory and remove especially old soft plastics. Both preschools and parents can get inspiration from the SSNC (2014) report. Note that the report is about more chemical groups than just phthalates. Another recommendation is, if possible, to choose another floor type than PVC. Several of the new preschools had higher concentrations of the phthalate alternative DINCH than old preschools, which can indicate that the PVC producers make more phthalate free products today than before.

Given the amount of products in the preschool environment in general, the chemical exposure of children in these environments might be of concern. However, a number of preschools had removed old toys and foam mattresses, which shows an ongoing process and increasing awareness of chemical exposures of children in the preschool environment.

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