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EFFECT OF EFFLUENTS FROM TEXTILE INDUSTRIES ON HUMANS AND ENVIRONMENT

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ABSTRACT

Production in textile industries consists of multistep chain process which involves production of a large number of heterogeneous compounds. It is also one of the greatest consumers of water in production of per kg of the material. It significantly contributes to environmental pollution by discharging wastewater rich in hazardous chemicals, such as azo-dyes, flame retardants, formaldehyde, dioxins, biocides and heavy metals, which in different ways pose threats to public health. Chemicals, harmless or dangerous, can accidentally or purposely enter and leave the textile mill during different steps of the manufacturing process. They can end up in goods placed on the market, either intentionally to give specific characteristics to the article (color, softness, flame and crease resistance, or water repellent properties), or unintentionally as residual materials from the production (traces of toxic and carcinogenic compounds can often be found in commercial dyes). Several textile operations lack quality-control systems for contaminants present in the raw fibre, left in the finished product, handled during the manufacturing processes or formed due to the use of high temperature, alkaline conditions, powerful oxidizing agents, etc. Significant amounts of hazardous compounds have been found in wastewater effluents from textile production Moreover, clothes are worn in close contact to the skin and, if chemicals are present in the garments, wearing them is a possible route for human exposure. Increasing environmental awareness has recently pushed an enhancement of international standards and regulations concerning quality, safety and sustainability of the textile industry.

KEYWORDS: Azo-dyes, Flame retardants, Formaldehyde, Dioxins, Biocides, Sustainability, Hazardous.

INTRODUCTION

Several textile operations lack quality-control systems for contaminants present in the raw fibre, left in the finished product, handled during the manufacturing processes or formed due to the use of high temperature, alkaline conditions, powerful oxidizing agents, etc. Significant amounts of hazardous compounds have been found in wastewater effluents from textile production plants.[1-3] Moreover, clothes are worn in close contact to the skin and, if chemicals are present in the garments, wearing them is a possible route for human exposure. Increasing environmental awareness has recently pushed an enhancement of international standards and regulations concerning quality, safety and sustainability of the textile industry. Since 2006 the European Union (EU) has adopted the Registration, Evaluation, Authorization and Restriction of Chemicals (REACH) regulation to protect human health and the environment from risks posed by chemicals.[4] EU introduced restrictions, applied to any compound on its own, in a mixture or in an article, which limit or ban manufacture, placing on the market and/ or use of a substance.[4] Nevertheless, a comprehensive characterization of organic contaminants present in fabrics and the related environmental impact are hindered due to the large number of intermediaries involved in the production steps, and to the extensive and rapid change in the chemicals usage, caused by fashion trends. For many of the newly introduced substances, especially dyes, environmental and health impact data are scarce. Furthermore, many of the used compounds might not fall under the obligation for registration, since their consumption does not exceed one metric ton. However, if they have low chemical and biological degradation rates, they might accumulate in biological tissues and their concentrations magnified in organisms exposed to contaminated environments. This points out the strong need for developing analytical methods to effectively detect and control pollutants discarded into the environment as well as hazardous compounds, which consumers could be exposed to during the use.

The textile chain starts with the production of raw fibres, which can be classified into two general categories. The categories are Natural (cotton, wool, silk) and Artificial (petrochemical origin, regenerated cellulose). Pesticides and herbicides might be used during the cultivation of cotton, as well as biocides and fungicides during its transportation and storage, while parasiticides are commonly applied on sheep to control external parasites.[5,6] Even though their concentrations in the final products are too low to be detected, these contaminants can be present in the raw material and released from it during heating or scouring processes on the fibre; hence, moving these into the aquatic environment, potentially create pollution problems. Pentachlorophenol, an organochlorine pesticide, also used as fungicide during transportation, has been detected in textiles.[7] Synthetic fibres may also contain very broad categories of impurities; e.g. solvents (when raw material undergoes solvent-spinning processes to make the fibre), by-products of polymerization processes (monomers, oligomers, catalysts, and solvents), and additives (antistatics, lubricants).[8] Alcohols (C14-C22), esters of carboxylic acids (C14-C24), hydrocarbons (C14-C18), carboxylic acids (C16-C24), and phthalate esters have been detected in wastewaters from synthetic fibre dyeing and finishing operations, as well as in extracts from synthetic fibres such as polyester, acrylic and nylon-6.[8]

Pre-treatment processes (desizing, scouring, bleaching, and mercerizing) are performed on the fibre in order to remove undesired material and improve the affinity for dyeing, printing and finishing treatments.[9] Each production step, briefly described below, implies the usage and the discharge of vast array of chemicals.[10] Desizing is a process used to remove size material from textile.[11] It can represent an environmental problem when the compounds used, which might be found in effluents from the processing plants, are difficult to emulsify or hardly biodegradable, like silicon oils.[12]

Hydrophobic substances, natural or added, such as oils, waxes, gums, which do not absorb the dyes or which are dyed with different colour, result in considerably impaired dyeing. These compounds have to be removed before the dyeing, by a process of Scouring.[12] This process uses compounds such as anionic or non-ionic surfactants (e.g. alcohol ethoxylate, and alkyl phenol ethoxylate), dispersing and reducing agents or chelating agents for removing metal ions.[12] Bleaching is a chemical process to remove unwanted, extraneous substances in order to improve the whiteness of the textile.[13] The properties of bleaching agents depend on the type of fibre and often a combined bleaching treatment is needed. The most frequently used compounds are hypochlorite or chlorite and hydrogen peroxide together with many auxiliary compounds such as stabilizer, co-stabilizer, wetting agents, activators, and anti-corrosion agents.[14] Mercerizing is a chemical procedure that improves the dye ability and increases the strength, dimension stability and lustre. Several compounds like alcohol sulphates, anionic surfactants, and cyclohexanol can be used.[15]

Dyeing is the process to colour textile material. This operation can be done during various stages (on the fibre, yarn, fabric and product) alone or together with other treatments.[16] It involves preparation of the dye, dyeing, fixation, washing, and drying. Dyes contain chromophoric (usually double-bonds, aromatic and heteroaromatic rings, responsible for coloring properties) and auxochromic groups (groups forming salts, responsible for dyeing properties).[17] With regard to the structure and usage, dyes are classified into azo-dyes, anthraquinone dyes, acidic (for wool, nylon, silk) or basic, disperse dyes (used for polyester and other synthetics material), reactive and direct dyes (used for cotton and viscose), and metal complex dyes (generally chromium or cobalt complexes).[18] The issue of carcinogenicity related to azo-dyes has already been addressed.[19] Mainly used for yellow, orange, and red coloring, they represent the majority of the synthetic dyestuffs. The structure contains one or more azo-bonds, which can be cleaved, releasing potentially carcinogenic aromatic amines.[20,21] After dyeing treatment, a large amount of the non-fixed dye leaves the dyeing units together with other auxiliary substances added during the process, resulting in a contaminated effluent water stream, which should be treated to remove the chemicals before being released into the environment. Several of these compounds are non-biodegradable and are released non-transformed into the wastewater system.[22] Wastewater released after the dyeing process might contain, beside dyes, dyeing additives like ethoxylates, alkylphenol ethoxylates, retarders for cationic dyes, dispersing agents, ethylenediamine tetraacetate and many others.[23]

Printing is the process where pigments, which are generally insoluble, are applied on the textile to give specific patterns, e.g. text on the garment. Possible pollutants from printing are: dyes or pigments, and organic solvents. Finishing encloses all those treatments, mechanical and chemical, performed on fibre, yarn, or fabric, in order to improve look and texture.[24] Possible pollutants released due to finishing processes are: formaldehyde, resins, flame-retardants, antistatics, softenings, crosslinking agents and biocides.[25] Release of chemicals from textile usage Effluents of textile-dyeing plants are extremely difficult to treat due to their high content of pollutants such as polycyclic aromatic hydrocarbons, heavy metal ions, surfactants, dyes, solvents, detergents and recalcitrant compounds.[26-28] Dyes and other contaminants[29], octyl- and nonylphenol, their ethoxylates and carboxylates derivate have been detected in textile wastewater treatment plants, as well as in stream effluents and surface waters.[30-32] Bioassay toxicity tests made on the chemicals released to water from various textile plants[33], together with studies on the modifications of the habitat[34], revealed a need for implementation and enforcement of the strategy for monitoring and protecting the streams. Several studies on chemicals in textile have been made during the years. They include biocides[35], hexabromocyclododecane flame-retardants[36], organophosphorous pesticides[37], per- and polyfluoroalkyl substances[38-40], polycyclic aromatic hydrocarbons[41,42], formaldehyde[43], nonylphenol ethoxylate and the release through laundering azo-dyes and their reduction products, polychlorinated dibenzo-p-dioxins, dibenzofurans and octachlorodibenzofuran. Dioxins have been shown to be present in dyes and to magnify their concentration in textile after the dyeing processes; they were also formed during textile processing and incineration.[32]

Hazardous substances, which remain in the fabric, may, via diffusive transport, be transferred to the stratum corneum of the skin, leading to 8 dermal absorption and systemic exposure. Textile clothes, bed linen and towels are articles of every-day use, which come in close contact with the human skin. The skin is the largest human organ, which has a vast area exposed to the environment. Molecules with a weight less than 1000 Da and a log KOW between 0.7 and 5.9 have the potential to cross the epidermal barrier, with maximum absorption between 1 and 2.[44-45] Experiments to show transfer of organic compounds from contaminated textile to the outermost layers of the skin have been made. Klasmeier et al.[44] which demonstrated the transfer to skin from different cotton T-shirts containing high levels of hepta- and octachlorinated dibenzo-pdioxins and dibenzofurans. In a scientific study by Gallagher et al.[45], volunteers were made to wear shorts and T-shirts/tank tops during 5 minutes of mild exercise, which made them sweat; subsequent analysis of the skin revealed, for some of the tested persons, the presence of benzothiazole on the back (covered by textile) but not on the forearms (not covered by textile), indicating the release of chemicals from clothes. A study proving the transfer of polychlorinated dibenzo-p-dioxins from textile to the skin has also been made[44]; however, systemic exposure is only possible if the compounds are able to overcome the penetration barrier posed by the viable epidermis. An in vivo experiment performed by Blum et al. in 1978[46] demonstrated that a chemical present in clothes can enter the human body via dermal absorption. In this study pajamas containing 5% tri (2,3-dibromopropyl) phosphate (a flame retardant, Mw = 697.6 Da, Log KOW = 3.71), was worn by children during sleep (8 hours exposure period). The following morning a fifty-fold increase in the metabolite 2, 3-dibromopropanol was detected in the urine. After changing to a pajama free of the substance, the urine concentration of the metabolite slowly decreased, but, after five days, it was still twenty times higher compared to the initial concentration. So far, the risks shown to be related to textile exposure are mainly dermatitis caused by azo-dyes.[47,48]

However, a large number of chemicals found in textiles (perfluorinated compounds (PFCs), organophosphorous pesticides, polycyclic aromatic hydrocarbons (PAHs), dioxins, nonylphenol ethoxylate and phthalates) can pose more severe risks to human health like cancer, immunotoxicity, as well as reproduction and development hazards.[49-51] Even if exposure to chemicals from textiles is supposed to take place through skin contact, other up-take routes cannot be ignored. By evaporation, substances can migrate into the air, or due to wear, fibres can be released to the air giving additional routes of human exposure through inhalation or ingestion. Not only clothes, but several other textile articles are present in the indoor environment and can all summed up to entail an exposure source of potentially high importance. Compounds in the fabric can be washed-out during laundry and, via wastewater and sewage plants, reach the aquatic environment.

Quinolines

Quinoline is an organic base belonging to the group of azaarenes, heterocyclic aromatic compounds with one or more nitrogen atoms placed in the aromatic ring, The principal sources of quinoline are coal tar and petroleum, and the main uses as a chemical intermediate, corrosion inhibitor and as a solvent for resins and terpenes may result in its release to the environment through various wastewater streams. Its industrial application related to textiles is the manufacture of dyes, such as 2- and 4-methyl derivatives, that are precursors to cyanine dyes.[52] Like pyridine derivatives and other azaarenes, quinoline is often reported as an environmental contaminant. Its presence has been associated with effluents from processing shale oil or coal facilities[53], and it has been found in urban air particulate matter and tobacco smoke.[54] Because of its relatively high solubility in water (quinoline is slightly soluble in cold water but dissolves easily in hot water), it has significant potential to cause water contamination .Its presence has been shown in ground water, lake and marine sediments ,and quite recently in dye processing plants effluents. Due to its log octanol/water partition coefficient (log KOW) of 2.03, quinoline has potential to be absorbed through skin. It has been shown to cause skin irritation and chromosome aberrations in rat liver.[55] Even though no carcinogenicity studies on humans are available, Quinoline structure shows that quinoline and some of its methylated isomers induce hepatocellular carcinosarcoma in mice, and EPA (Environmental Protection Agency) has classified it as a Group B2 chemical, a probable human carcinogen.

Benzothiazoles and Benzotriazoles

Benzothiazole (BT) and benzotriazole (BTri) are aromatic heterocyclic compounds where benzene is fused with a five member ring, which contains sulfur and/or nitrogen as heteroatoms, Benzothiazole, mostly as derivative with the 2-position of the thiazole ring being substituted, finds its application in rubber production as vulcanization accelerators, and in other industry applications as biocides. These compounds are also commonly used as corrosion inhibitors, while 2-hydroxyphenyl derivatives of benzotriazoles are used in textiles as Benzotriazole UV stabilizers (BUVS).[56, 57] BTs and BTris are rather water-soluble, but not readily degradable, thus, they are not completely broken down in wastewater treatment plants and a significant part reaches surface water.[58, 59] BT occurrence is widespread; it has been found in municipal and household wastewater[59,60], whilst BUVSs have been found in fish[59], clams, oysters, and gastropods.[60] BTri are released from automobile tires as well, and 2-Mercaptobenzothiazole (2-MBT) and BT are present in urban air, most likely due to tire tread abrasion.[94] Their acute toxicity in various test systems has been shown and also allergenic and irritating properties.[61] Benzotriazole was found to be mutagenic and estrogenic in aquatic organisms[62] and BUVSs can bioaccumulate in birds, fishes and invertebrates.[60] Due to the octanol-water partition coefficient, they can be absorbed by the skin. Transfer to skin experiment showed that benzothiazoles are released from textile clothes[56], moreover both classes of compounds have been detected in urine samples collected from general populations.[63]

Aromatic Amines

Aromatic amines have one or more aromatic rings connected to one or more amino groups; the simplest is represented by aniline, Depending on the number of aromatic rings or/and the substituents bonded to the aromatic ring, they have different environmental and health related impacts. Sources can be oil refining processes, diesel engine exhaust, synthetic polymers, textile and hair dyes, rubber, pesticides, and pharmaceuticals. Their use related to textile industry is chiefly in the production of azodyes, whose chemical structure is more complex than that of the parent amines]. However, when the dye is absorbed and metabolized in the body, the cleavage of the vulnerable azo-bond releases the amine, causing high risks to human health.[20] Certain dyes have a low degree of percutaneous absorption, nevertheless, bacteria of the human skin have been demonstrated to metabolize azo-dyes in to aromatic amines which are known to easily penetrate the skin. Studies on workers exposed to 2-naphthylamine, benzidine, and 4-aminobiphenyl showed association between human exposure to aromatic amines with an increased risk of urinary bladder cancer.[64,65] O-toluidine has also been suggested to be a bladder carcinogen. Workers exposed jointly to o-toluidine and 4,4’-methylene bis(2- methylaniline) showed 62-fold increase in bladder cancer risk.

Washing out of chemicals

Quinoline, benzothiazole, benzotriazole and derivative compounds are sparingly soluble in water but more easily soluble in hot water. Laundry can, thus, be a route of emission into the environment of contaminants present in textile. Analyses on samples before and after five and ten times washing were directed to quantify that emissions. Results showed an average loss of more than 50% for benzothiazole, whilst quinoline revealed a lower washout effect, probably due to the different usage, thus a diverse interaction with textile fibers. To estimate the emission into household wastewater for 5 kg of clothes, the average concentration of quinoline and benzothiazole and their respective average loss were used. In this way the amount of released benzothiazole was calculated to 0.5 g and the amount of quinoline 0.24 g. This suggests that laundry is a source of emission of these compounds into household wastewater. The loss of some compounds, e.g. quinolines, was slow (20% after ten washings), demonstrating that significant amounts of the chemicals remain in the clothes for a long time and thus have the potential of a chronic impact on human health.

Conclusions and future perspectives

Focus has been put on the release of hazardous chemicals into the environment and there is today a well-known environmental problem caused by the textile industry. However, substances remaining in finished textile products, in daily close contact with the body, have been neglected as a potential risk to human health. In this work a method for explorative screening of textile materials, with special emphasis on clothes, have been developed. Over hundred compounds were tentatively identified and more than forty were confirmed to be present in a set of sixty garments available on the global market. The hazards posed by the identified substances were primarily skin sensitization and irritation, but also reproduction toxicity, and proved or suspected carcinogenicity. Targeted analytical methods were developed for four groups of compounds: quinolines, benzothiazoles, benzotriazoles and aromatic amines, and were applied on the textile materials. Concentration differences for some of the target analytes was found to be related to the fiber type used in the textile. In particular, higher amounts of quinolines and benzothiazole were detected in textile manufactured mainly from polyester. Moreover, it is indicated that organic cotton and eco labelling are no guarantee that textiles are free from harmful chemicals. The release of these two groups of compounds during washing of the clothing textiles has been demonstrated. A discharge into household wastewater and further on to the aquatic environment is a most likely consequence. For compounds with a slow release during washing, a significant amount remain in the clothes have the possibility for a long term human exposure. Chemical residues from textiles have the potential to migrate from clothes to the human skin and be absorbed according to their size and octanol/water partition coefficient, and may thus cause local and/ or systemic effects. Harmless compounds or compounds with minor health effect could be metabolized by bacteria present on the skin, or if absorbed, be converted to harmful substances by hepatic enzyme systems. A combination of different toxic compounds could also enhance (or reduce) the health risk of the single substances

In order to get a better picture of the total exposure, not only clothes, but several other textile articles present in the indoor environment should be analyzed. Screening and targeting methods using LC/MS/MS with high resolution, using positive and negative electrospray ionization, as well as APCI, should be developed to include more hydrophilic compounds, which are more likely to be washed out and end up not in the sludge but rather go right into the aquatic system. Compounds for further research has to be selected by combining explorative chemical analysis with data on skin penetrating properties and toxicological effects, to put a special focus on compounds with a potential for human exposure. Transfer of the selected compounds from textile to the skin as well as their absorption needs to be investigated. Extraction could be made using simulated sweat and bioassay toxicity test made on the chemicals released from the fabric. Further research should also be directed towards analytical methods for determination of identified textile chemicals and/or their metabolites in human blood and/or urine. Methods for treating the textiles (e.g. carbon dioxide cleaners) to avoid having these compounds spreading into the environment should also be investigated.

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