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Published in:
Waste Management

Link to article, DOI:
[10.1016/j.wasman.2015.02.028](https://doi.org/10.1016/j.wasman.2015.02.028)

Publication date:
2015

Document Version
Peer reviewed version

[Link back to DTU Orbit](#)

Citation (APA):
Pivnenko, K., Eriksson, E., & Astrup, T. F. (2015). Waste paper for recycling: Overview and identification of potentially critical substances. *Waste Management*, 45, 134-142. <https://doi.org/10.1016/j.wasman.2015.02.028>

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Waste Paper for Recycling: Overview and Identification of Potentially Critical Substances

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19 **Abstract**

20 Paper product manufacturing involves a variety of chemicals used either directly in paper and pulp
21 production or in the conversion processes (i.e. printing, gluing) that follow. Due to economic and
22 environmental initiatives, paper recycling rates continue to rise. In Europe, recycling has increased
23 by nearly 20% within the last decade or so, reaching a level of almost 72% in 2012. With increasing
24 recycling rates, lower quality paper fractions may be included. This may potentially lead to
25 accumulation or un-intended spreading of chemical substances contained in paper, e.g. by
26 introducing chemicals contained in waste paper into the recycling loop. This study provides an
27 overview of chemicals potentially present in paper and applies a sequential hazard screening
28 procedure based on the intrinsic hazard, physical-chemical and biodegradability characteristics of
29 the substances. Based on the results, 51 substances were identified as potentially critical (selected
30 mineral oils, phthalates, phenols, parabens, as well as other groups of chemicals) in relation to paper
31 recycling. It is recommended that these substances receive more attention in waste paper.

32 Keywords: Hazardous substances; Paper; Priority pollutants; Recycling; Waste management

33

34

35 **Abbreviations**

36 BBP: Benzyl butyl phthalate

37 BPA: Bisphenol A

38 CAS: Chemical Abstracts Service

39 CEPI: Confederation of European Paper Industries

40 DBP: Dibutyl phthalate

41 DEHP: Diethylhexyl phthalate

42 DIBP: Diisobutyl phthalate

43 DIPN: Diisopropyl naphthalene

44 EDCs: Endocrine Disrupting Chemicals

45 EFSA: European Food Safety Authority

46 EuPIA: European Printing Ink Association

47 FDHA: Swiss Federal Department of Home Affairs

48 NIAS: Non-Intentionally Added Substances

49 PCBs: Polychlorinated biphenyls

50 PBT: Persistent, Bioaccumulative and Toxic

51 vPvB: very Persistent and very Bioaccumulative

52 ZELLCHEMING: Vereins der Zellstoff- und Papier-Chemiker und –ingenieure (German for:

53 Association of Chemical Pulp and Paper Chemists and Engineers).

54

55 **1. Introduction**

56 Paper recycling is one of the most well-established recycling schemes applied to waste materials
57 today. Recycled paper is an integral part of paper and pulp production, with estimated utilisation for
58 recycling in Europe of about 72% in 2012 (an increase of 20% from 2000) (CEPI, 2013a). In
59 addition to recycled paper being an important raw material for the paper industry (CEPI, 2013b), it
60 has also been demonstrated in several studies that paper recycling may offer significant
61 environmental benefits in a lifecycle perspective (Laurijssen et al., 2010; Villanueva and Wenzel,
62 2007). Thus, paper recycling may be regarded as beneficial from both a resource and an
63 environmental perspective and should be promoted as much as possible. However, increasing
64 concerns related to the presence of potential harmful chemical substances in paper have been voiced
65 within recent years (e.g. Biedermann et al., 2011b; Liao and Kannan, 2011; Pivnenko et al., 2013),
66 for example in relation to the migration of chemicals from packaging materials into food (e.g.
67 Begley et al., 2008; Biedermann et al., 2013; Gärtner et al., 2009; Lorenzini et al., 2013). While
68 further increasing paper recycling rates can undoubtedly be achieved in Europe, the quality of the
69 waste paper may ultimately decrease as more and more "marginal" paper fractions are collected for
70 recycling and the contents of harmful substances in paper thereby increase. A systematic overview
71 of the chemical substances potentially present in waste paper for recycling is therefore needed to
72 provide a basis for further evaluation of the quality of waste paper as a resource, and ultimately also
73 to maintain consumer acceptance of recycled paper in general.

74 Paper production and manufacturing operations generally consist of the following two phases:
75 i) paper and pulp production by the paper industry (i.e. different quality grades of paper) and ii)
76 paper product manufacturing by separate industries (e.g. periodicals, packaging materials, books,
77 etc.). Chemicals in waste paper may originate from a wide range of sources, namely intentionally
78 added (i.e. additives, inks, pigments, glues, etc.), part of a reaction and/or biodegradation or added

79 during the use phase of the paper or during the waste management phase (e.g. cross-contamination
80 from other waste materials during collection). Chemicals are added in order to improve the
81 production process itself and the quality or functionality of the final product. Starting with paper
82 production, chemicals are introduced through the use of synthetic additives, which include retention
83 aids, sizing agents, coatings, biocides, synthetic binders, etc. Synthetic additives represent slightly
84 more than 1% v/v of raw materials used in paper production (ZELLCHEMING, 2008), the largest
85 share of which (90% v/v) are functional additives (Moench and Auhorn, 2002) intended to be
86 retained in the paper product. The next step, where the paper is converted into a final product, may
87 include printing, dyeing, addition of adhesives and labels, etc. During the processing, chemicals
88 may dissolve and be removed via wastewater, volatilize and be released to air or remain in the solid
89 matrix and thereby be present in newly manufactured paper products. When waste paper is added to
90 the process, this may potentially introduce new substances from the use and waste management
91 phase. Knowing which potential partitioning a given chemical (or group of chemicals) will follow is
92 vital for identifying potentially critical substances which may end up being concentrated in the
93 fibres and be reintroduced into consumer products.

94 Recent studies have demonstrated that paper and paper products may contain high numbers of
95 chemical substances (BMELV, 2012; Bradley et al., 2008), most of which can be associated with
96 the printing industry, where more than 7,000 chemicals may be used in food-packaging ink
97 production alone (EuPIA, 2012). Nevertheless, very little quantitative information is available
98 regarding the presence of specific substances in paper products or waste paper potentially sent to
99 recycling. Most existing studies target a specific group of chemicals or paper products (e.g. Becerra
100 and Odermatt, 2012; Geens et al., 2012; Song et al., 2000; Trier et al., 2011), and attempting to
101 identify every single chemical present in paper has proved to be challenging (BMELV, 2012).

102 Although specific regulations covering paper food packaging do not exist, European
103 legislation on items (i.e. plastics, metal, paper, etc.) brought into contact with food prevents the use
104 of chemicals that could migrate into foodstuffs and adversely affect human health, as well as the
105 quality and nature of food (EC, 2004). This legislation covers paper packaging produced from
106 virgin fibres, but when paper is recycled, the producers may not be aware of the presence of any
107 specific chemicals added throughout the lifecycle of the paper. Consequently, the paper industry,
108 and the final output paper quality, is affected by the presence of chemicals in the recycled paper,
109 e.g. chemicals introduced during the use phase or via paper products from other countries. In 2012,
110 more than 5 million tonnes of paper (approx.11% of recycled paper) was imported into Europe from
111 the USA, Russia, Brazil, Canada, etc. for paper product manufacturing (CEPI, 2013a).

112 Without a comprehensive overview of which chemical substances should be prioritised in
113 relation to paper, and which substances should ultimately be avoided, it may not be possible in the
114 future to ensure both high recycling rates and at the same time a high quality of the paper products
115 based on recycled fibres. As direct and substance-by-substance analysis is not practically feasible, a
116 systematic screening of un-problematic chemicals is needed, in order to identify those substances
117 which may be considered most problematic and critical for the future recycling of paper.

118 The overall goal of this study is to provide a basis for systematically addressing the
119 recyclability of waste paper with respect to the potential presence of hazardous substances. The
120 specific objectives are: i) based on existing literature, to compile a list of chemical substances
121 potentially applied in paper production and paper product manufacturing, as well as chemicals
122 identified directly in paper, ii) based on a sequential hazard screening procedure to identify the most
123 critical chemicals from this list based on their harmfulness, physical-chemical properties and
124 biodegradability and iii) to evaluate potential implications related to the management of paper waste
125 and paper recycling.

126 **2. Methodology**

127 **2.1 Data sources for chemicals in paper**

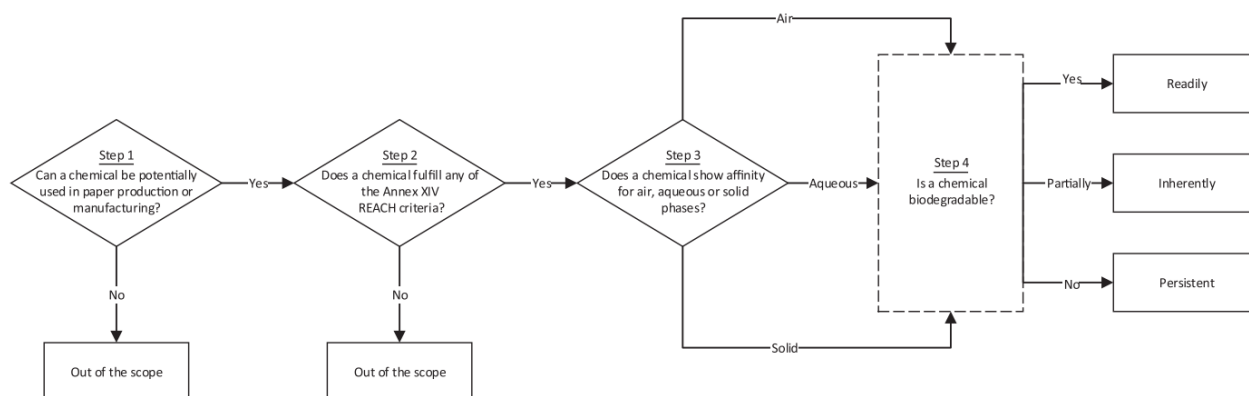
128 Information about chemical substances, used in either paper production or paper conversion, as well
129 as chemicals identified in actual paper product flows, was obtained from a range of data sources.
130 Chemicals used in pulp and paper production were obtained from national product registries
131 (KEMI, 2014; SPIN, 2013) and scientific assessments (Riskcycle, 2013; ZELLCHEMING, 2008),
132 as well as inventory data provided by the European Food Safety Authority (EFSA) (EFSA, 2012a).
133 Substances used by the printing industry were obtained from a recent Danish report (Miljøstyrelsen,
134 2011a), an inventory list of the European Printing Ink Association (EuPIA) (EuPIA, 2012) and
135 recent regulation issued by the Swiss Federal Department of Home Affairs (FDHA) (FDHA, 2005).
136 Although data obtained for paper printing could not be isolated from the printing of other materials,
137 the European printing industry belongs to a forest-based industrial sector, and the share of paper in
138 the printing industry is substantial. All of the abovementioned data sources predominantly reflected
139 European industry and research; this was not due to any selection of sources, but rather reflected
140 availability of state-of-the-art information and level of detail provided. No information could be
141 found related specifically to chemicals used in adhesives, so these were therefore only indirectly
142 included in the study as part of the analytical literature reviewed. Additionally, relevant scientific
143 literature addressing the composition of paper, paper products or waste paper was reviewed. While
144 the aim was not to provide an exhaustive review of all available literature, the focus was placed on
145 recent literature in order to relate any findings as best as possible to the current technological scope
146 of the paper industry. No geographical scope was applied to the selected studies, as paper is a
147 commodity traded on the global market with high volumes of paper, paper packaging and waste
148 paper being imported and exported on a yearly basis. In total, 25 scientific studies were reviewed.

149 Where available the concentrations of substances mentioned in the literature are also provided. See
150 Table 1 for a complete list of the data sources used in this paper.

151 Based on the abovementioned combination of information sources, a compilation of almost
152 10,000 chemical substances was obtained once duplicates were removed. To avoid ambiguity and
153 potential double-counting, only chemicals (or groups of chemicals) which could be assigned a valid
154 CAS (Chemicals Abstracts Service) registry number were included in the study.

155 **2.2 Criteria for identifying potential priority chemicals**

156 With the aim of identifying potentially critical chemicals that should be prioritised in relation to
157 paper recycling, a screening selection procedure was applied for those that may be considered most
158 harmful, most likely to be associated with paper fibres (and not volatilise or be released into the
159 water phase during re-pulping) and the most persistent in the environment. The procedure involved
160 the following four steps: 1) compiling an inventory of chemicals that may be used in the paper and
161 printing industries or which may have been identified in paper (corresponding to the list of about
162 10,000 substances mentioned above), 2) identifying potentially harmful chemicals, 3) identifying
163 chemicals primarily associated with solids (i.e. paper fibres) and 4) identifying chemicals
164 characterised as not readily biodegradable. Steps 1) through 4) were carried out consecutively,
165 thereby filtering out less problematic substances in relation to paper recycling. The remaining list of
166 chemicals therefore represented substances that should be prioritised in future scenarios
167 characterising paper and addressing paper recycling. See Figure 1 for an illustration of the
168 procedure.



169

170 **Figure 1.** Schematic representation of the methodology applied in selecting relevant chemicals of
171 interest.

172 In Step 2), chemicals were selected in accordance with Annex XIV of European REACH
173 Regulation (EC, 2006) and according to the following criteria: i) substances classified in hazard
174 class “carcinogenicity” (categories 1A and 1B, Carc. 1A or 1B) (EC, 2008), ii) substances classified
175 in hazard class “germ cell mutagenicity” (categories 1A and 1B, Muta. 1A or 1B) (EC, 2008), iii)
176 substances classified in hazard class “reproductive toxicity” (categories 1A and 1B, Repr. 1A or 1B)
177 (EC, 2008), iv) substances classified as “Persistent, Bioaccumulative and Toxic (PBT)” (according
178 to Annex III in (EC, 2006)), v) substances classified as “very Persistent and very Bioaccumulative
179 (vPvB)” (according to Annex III in (EC, 2006)) and vi) substances characterised as “Endocrine
180 Disrupting Chemicals (EDCs)” (WHO, 2002) for which scientific evidence of possible serious
181 effects on human health and/or the environment could be found. The chemicals selected based on
182 Step 2 included all substances fulfilling at least one of the abovementioned criteria. Only substances
183 with sufficient information available were selected in Step 2; in other words, those with non-
184 published or incomplete hazard assessments were not included. In practice this means that the
185 number of chemicals finally selected in this study might be underestimated, as future hazard

186 assessments of chemicals included in inventory list (Step 1) may reveal additional substances
187 fulfilling the Step 2 criteria.

188 Step 3) was based on the methodology described by Baun et al. (Baun et al., 2006), where
189 partitioning between phases is based on the potential of a given chemical to be adsorbed to solids, to
190 volatilise or to remain dissolved in the aqueous phase. Influence of particular paper production
191 processes (pulp, coating, drying, etc.) on phase distribution of substances is out of the scope of
192 the present work and was not considered.

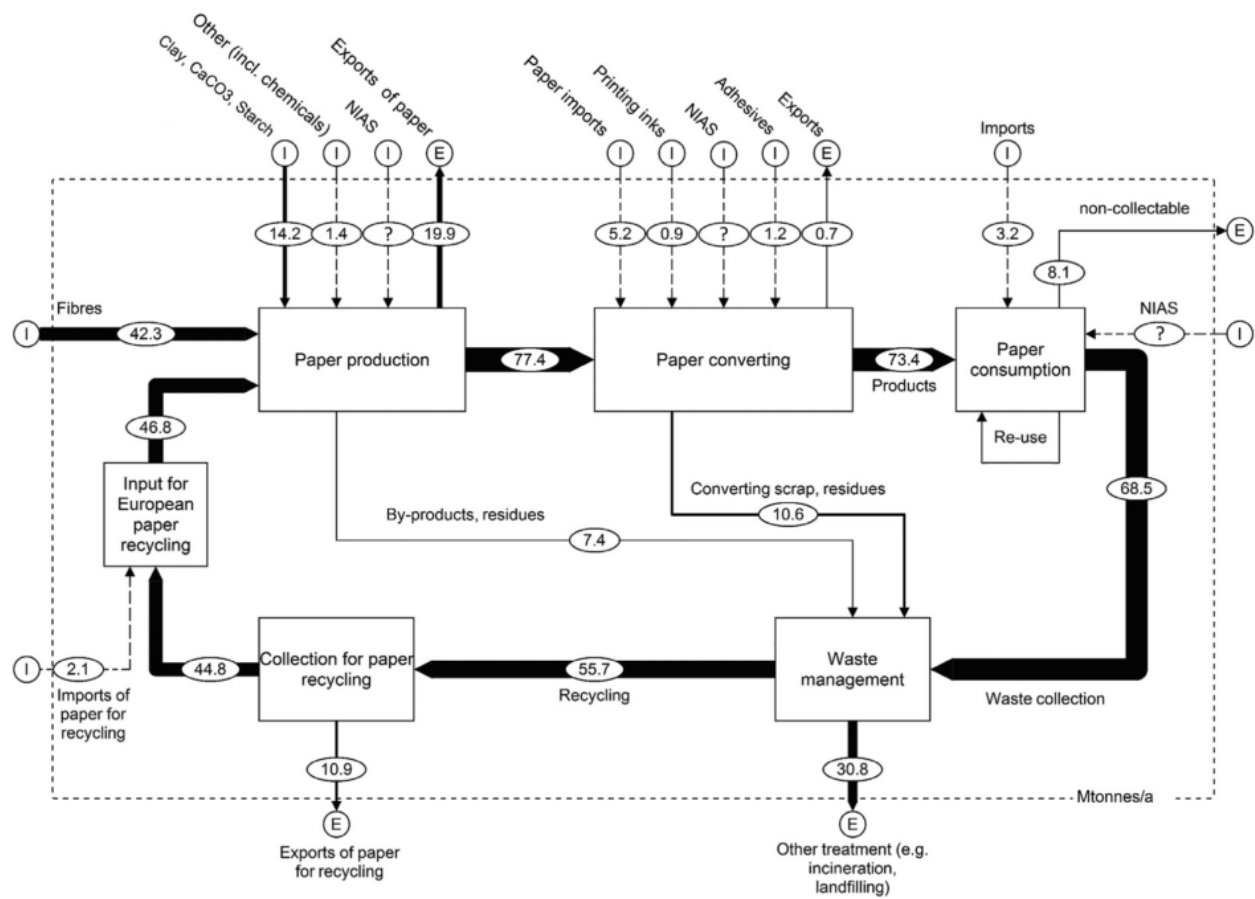
193 In Step 4) of the screening process, chemicals were assessed in accordance with their
194 biodegradability and then classified into persistent, inherently and readily biodegradable.
195 Classification was based either on the Biowin models 3 and 5, included in EpiSuite 4.1 (U.S. EPA,
196 2013) with cut-off values as presented by Baun et al. (Baun et al., 2006), or on scientific literature
197 providing experimental biodegradation results. Relevance of biodegradability of chemicals to
198 particular processes in paper recycling was not established, as the variations associated with
199 different steps of the paper lifecycle (i.e. paper production, manufacturing, use, waste paper
200 collection, re-processing, etc.) are potentially large.

201 **3. Results and Discussion**

202 **3.1 Overview of substances**

203 Figure 2 presents material flows of European paper recycling, indicating points where chemicals are
204 introduced into the loop. Most non-fibrous materials are introduced in the paper production step, but
205 they are almost entirely represented by non-hazardous naturally occurring substances such as clay,
206 CaCO_3 and starch. No quantitative data were available regarding chemicals added non-intentionally
207 into the loop (i.e. Non-Intentionally Added Substances, NIAS). An overview of each of the sources
208 contributing to the final list in Step 1 is presented in Table 1. Although the paper industry uses high

209 volumes of chemical substances (Figure 2), it is evident that a much higher variety of chemicals
 210 associated with paper products derives from printing (Table 1).



211
 212 **Figure 2.** Material flow of the European paper recycling loop. Dotted lines indicate points where
 213 chemicals are introduced (Based on (CEPI, 2013a, 2013b; EUPIA, 2013; FEICA, 2008.; ITC, 2014)
 214 and personal communication with the Confederation of European Paper Industries (CEPI)).

215 Due to the large number of chemicals identified in Step 1, attributing to each of them a
 216 potential use by industry is practically impossible. Nevertheless, most of the substances used in
 217 paper production can be attributed to fillers, binders, retention aids, wet/dry-strength agents,
 218 coaters, biocides, dispersers, etc. (ZELLCHEMING, 2008). In the printing industry the vast
 219 majority of chemicals are used as solvents, dyes, inks, pigments, binders, curing agents and photo-

220 initiators, plasticisers, surfactants, etc. (Miljøstyrelsen, 2011a) Only a small fraction (157) of the
 221 almost 10,000 substances could be identified in Step 2. Figure 3a presents the distribution of
 222 substances on Step 2 list in accordance with their use by industries throughout the lifecycle of
 223 paper. Only 10 of the chemicals are used exclusively in paper production (mainly biocides).
 224 Conversely, 133 chemicals were attributed to the printing industry, most of which are solvents and
 225 polymeric resins employed in inks, pigments and dyes. Chemicals which could not be attributed
 226 either to paper production or to the printing sector (14) could potentially be by-products or
 227 contaminants introduced into the production cycle through recycled paper.

228 **Table 1.** Data sources used in the study and their quantitative contribution to Step 1 list of
 229 chemicals.

Source	Number	Description	Industry	Reference
	of			
	chemicals			
Literature	348	Scientific literature providing analytical data on the identification or quantification of chemicals in paper and/or board	Paper and paper product manufacturing, and NIAS	(Biedermann et al., 2013, 2010; Binderup et al., 2002; BMELV, 2012; Bradley et al., 2008; Castle et al., 1997a, 1997b; Fierens et al., 2012; Gehring et al., 2004; LeBel et al., 1991; Liao and Kannan, 2011; Miljøstyrelsen, 2011b, 2003a, 2003b; Ozaki et al., 2004; Parry, 2001; Petersen et al., 2013; Poças et al., 2010; Riber et al., 2009; Rotter et al., 2004; Sipiläinen-Malm et al., 1997; Storr-Hansen and Rastogi, 1988; Sturaro et al., 2006; Vinggaard et al., 2000; Zheng et al., 2001)

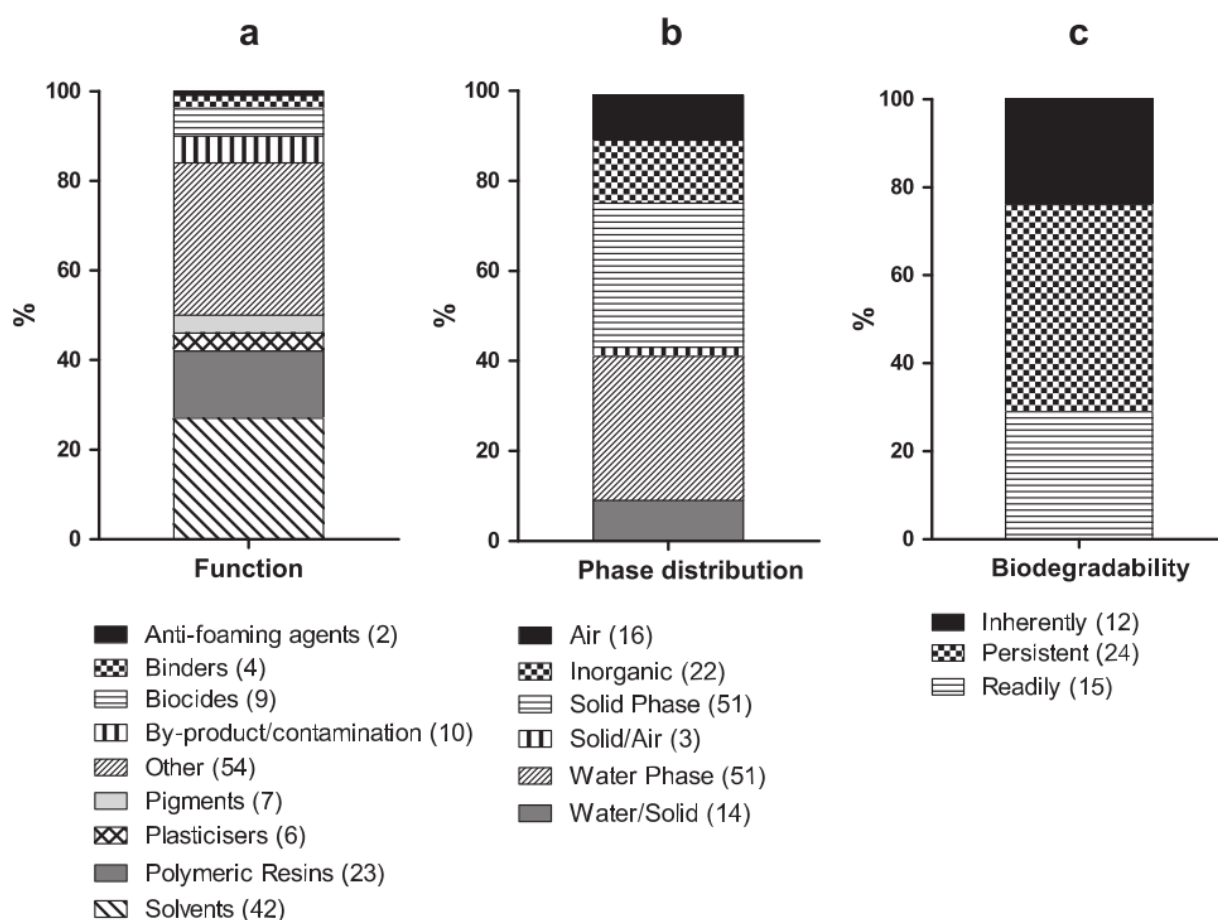
Danish register	product	75	Chemicals used in preparations of articles. Danish industry for pulp, paper and paper products	Paper and paper product manufacturing	(SPIN, 2013)
Swedish register	product	144	Chemicals used in preparations of articles. Swedish industry for pulp, paper and paper products	Paper and paper product manufacturing	(KEMI, 2014)
Danish Environmental Protection Agency		415	Inventory of chemicals used by the Danish printing industry	Paper product manufacturing*	(Miljøstyrelsen, 2011a)
RiskCycle		12	Database of chemical additives used in paper production	Paper manufacturing	(Riskcycle, 2013)
ZELLCHEMING		44	Chemical additives for the production of pulp and paper	Paper manufacturing	(ZELLCHEMING, 2008)
EFSA		223	Chemicals currently used in the manufacture of paper and board	Paper manufacturing	(EFSA, 2012a)
FDHA		4575	Chemicals permitted to be used in the manufacture of	Paper product manufacturing*	(FDHA, 2005)

			packaging inks	
EuPIA	3858	Inventory list of	Paper product	(EuPIA, 2012)
		chemicals used in the	manufacturing*	
		manufacture of food		
		packaging inks		
TOTAL:	9694	-	-	-

*not limited to paper matrix

Chemicals on the Step 2 list were assessed in relation to their potential partitioning between the air, aqueous and solid phases. It is evident from Figure 3b that most of the chemicals either have a higher affinity for a solid matrix (51) or remain dissolved in the water phase (51). Sixteen of the chemicals on the list are relatively volatile and could potentially escape through volatilisation in the production process, while an additional 22 substances on the list are inorganic, and although two of them (i.e. mercury and carbon disulphide) may partially volatilise, the distribution of the rest will depend very much on specific conditions in the paper processing stages (e.g. pH, redox conditions, presence of organic matter, etc.) and are difficult to predict. Chemicals remaining in the solid matrix are of particular interest in terms of paper recycling.

In the following step (Step 4) the biodegradability of the previously identified chemicals was assessed. As presented in Figure 3c, most of the substances that showed affinity for the solid phase were characterised as persistent (24), while the 27 remaining chemicals could be classified as inherently (12) and readily biodegradable (15).



244

245 **Figure 3.** Distribution of the use of chemicals from Step 2 list (3a); phase distribution of chemicals,
 246 i.e. Step 3 list (3b); biodegradability of chemicals associated with the solid phase, i.e. Step 4 list
 247 (3c).

248 While the list of potential priority chemical substances may not be exhaustive (as the
 249 screening can only be based on available information about substances), it nevertheless clearly
 250 indicates that concerns regarding their presence in paper and their potential accumulation in the
 251 paper lifecycle may be pertinent to a relatively small number. The list therefore forms a systematic
 252 basis for further research in relation to paper characteristics and paper recycling. The 157
 253 substances identified in Step 2 are grouped in the following according to their chemical structure

254 and then discussed in more detail. Individual tables listing each chemical substance according to
255 these groups can be found in the Supplementary Materials (Tables S1-S6).

256 **3.2 Mineral oils**

257 The commonly used term “mineral oils” refers to a mixture of components which originate from
258 crude oil refining processes. Mineral oils mainly contain straight and branched open-chain alkanes
259 (paraffins), alkylated cycloalkanes (naphthenes) and aromatic hydrocarbons (EFSA, 2012b), and
260 their final composition will depend largely on the initial composition of the crude oil, as well as the
261 refinery treatment (e.g. alkylation, hydro-treatment, cracking, extraction, etc.). Although the Joint
262 FAO/WHO Expert Committee on Food Additives (JECFA) recently withdrew previously
263 established acceptable daily intakes in relation to mineral oils (JECFA, 2012), the JECFA
264 assessment refers to highly-refined mineral oils free from aromatic hydrocarbons. On the other
265 hand, paper products were shown to contain technical-grade mineral oils which may include
266 aromatic hydrocarbons (Biedermann and Grob, 2010). Grob et al. (Droz and Grob, 1997) found
267 that, at least initially, printing inks (solvents in particular) used in paper products are the main
268 source of mineral oils in paper. Further studies have also posited that mineral oils may derive from
269 recycled waste paper (Biedermann and Grob, 2010; Biedermann et al., 2011b).

270 Out of the 157 chemicals included in the Step 2 list, 49 were classified as mineral oils (Table
271 S1 (Supplementary Material)) and characterised as carcinogens, while some are also mutagenic
272 substances. The chemicals presented herein are not single substances but rather mixtures of
273 substances containing various hydrocarbons. Being mixtures rather than single substances, mineral
274 oils can be used in a variety of applications in the industry, i.e. from solvents and as the basis for
275 polymeric resins through to lubricants and cleaning agents for machinery (EFSA, 2012b;
276 Miljøstyrelsen, 2011a). Most of the scientific studies currently available focus on mineral oil
277 content in paper used for food packaging (e.g. (Biedermann and Grob, 2010; Biedermann et al.,

278 2011a, 2011b; Droz and Grob, 1997)), as migration into foodstuffs remains one of the most
279 important sources of consumer exposure (EFSA, 2012b). As they are hydrophobic substances,
280 mineral oils may not be removed in water-based processes of paper recycling (i.e. pulping,
281 deinking, washing), remain in the solid matrix and have a high chance of persisting in the recycling
282 process and being reintroduced into newly manufactured products (BMELV, 2012). Such a scenario
283 is unlikely for some of the lighter mineral oils, which are expected to escape due to volatilisation in
284 e.g. paper drying step. A recent study (BMELV, 2012) showed that the deinking process reduces
285 insignificantly the concentration of mineral oils, while paper drying is the main process for their
286 removal (around 30% (Biedermann et al., 2011b)) – still resulting on average in 340 mg/kg (<C₂₄)
287 in unprinted food-packaging board produced (Biedermann and Grob, 2012). One study showed that
288 even the presence of a barrier (e.g. plastic foil) may not always prevent the migration of mineral oils
289 from packaging into a food product (Fiselier and Grob, 2012), and a biodegradation assessment has
290 shown that a significant number of mineral oils (15) can be classified as persistent, making
291 bioaccumulation relevant for some.

292 Due to the diversity of mineral oils, and the fact that they are mixtures, identifying and
293 quantifying single constituents (as the ones presented in Table S1 (Supplementary Material)) is
294 practically impossible. As a result, mineral oils are analysed instead as sum parameters (e.g. the
295 Hydrocarbon Oil Index), with fractioning based on the number of carbon atoms in the chemical
296 (Droz and Grob, 1997; Pivnenko et al., 2013) or fractioning between mineral oil saturated and
297 aromatic hydrocarbons (Biedermann and Grob, 2010; Biedermann et al., 2011b). The study
298 conducted by Pivnenko et al. (2013) showed the presence of mineral oils in all the analysed waste
299 paper fractions, with the highest concentrations (up to 1,800 mg/kg) identified in newspapers and
300 tissue paper. Similarly, among waste paper materials fed into the German recycling loop,
301 newspapers were identified as the main source of mineral oils (BMELV, 2012). Their presence in

302 newspapers can be attributed to solvents and processes used in cold off-set printing (Biedermann
303 and Grob, 2010), while mineral oils in tissue paper may indicate the introduction of chemicals
304 during the product's life span and waste management. Both studies mentioned above (BMELV,
305 2012; Pivnenko et al., 2013) present relatively stable concentrations of mineral oils in a variety of
306 board products, potentially indicating a contribution made by newspaper recycling.

307 **3.3 Phthalates**

308 Most phthalates are used as plasticisers in the preparation of printing inks, lacquers and dispersion
309 glues (BfR, 2007; CDC, 2009), though they can also be used as softeners in tissue paper
310 (Miljøstyrelsen, 2003a). From the Step 2 list, seven phthalates were identified (Table S2
311 (Supplementary Material)), the majority of which are classified as EDCs, while reproductive
312 toxicity is also attributed to some. Phase distribution assessment (Step 3) revealed that phthalates
313 may be retained in the paper and pulp solid matrices and could potentially follow the production
314 process until the final product. Benzyl butyl phthalate (BBP), Dibutyl phthalate (DBP) and
315 Diethylhexyl phthalate (DEHP) are classified as persistent, according to the criteria used. Table S2
316 (Supplementary Material) reveals the range of concentrations of phthalates quantified in paper, with
317 Diisobutyl phthalate (DIBP) reaching the highest concentrations (up to 120 mg/kg). A study
318 conducted by a German authority (BMELV, 2012) showed that phthalates were mainly present (up
319 to 35 mg/kg) in board, waste paper from offices, specialty paper and papers containing relatively
320 high amounts of glue. In contrast, newspapers, magazines and advertisements contained almost one
321 order of magnitude lower phthalate concentrations (BMELV, 2012). These results could potentially
322 indicate adhesives as the main source of phthalates in paper for recycling.

323 Experimental results involving four separate recycling facilities producing board for food
324 packaging indicated that in the recycling process, phthalates have a high affinity for paper fibres,
325 moving through the production line and then into final products (BMELV, 2012). Particularly, the

study showed that DIBP, DBP and DEHP have a tendency (on average) to accumulate in board produced from recycled paper. On the other hand, the same study showed that virgin fibre-based board contained phthalates in lower concentrations (<0.2 mg/kg) well below one order of magnitude.

3.4 Phenols

Among the 157 chemicals in Step 2, eight were identified as phenols (Table S3 (Supplementary Material)), all of which fulfilled the EDCs criteria. The use of phenols in the paper industry varies significantly; for example, Bisphenol A (BPA) is used as a developer in thermal paper and pentachlorophenol as a biocide in paper production (Mendum et al., 2011; ZELLCHEMING, 2008). Octylphenol, 4-nonylphenol and 4-tert-octylphenol are used in polymeric resins employed in ink preparation (EuPIA, 2012), while nonylphenol is part of some surfactants used in the printing (Miljøstyrelsen, 2011a). The majority of thermal paper is used in cash register receipts, which may contain up to 17,000 mg/kg of BPA (Miljøstyrelsen, 2011b). The remaining chemicals in Table S3 (Supplementary Material) show significantly lower concentration ranges (0.01-68.9 mg/kg) when compared to those of BPA (0.068-17,000 mg/kg).

Liao & Kannan (2011) detected BPA in the majority of 99 paper products analysed, which included magazines, paper towels, napkins, flyers, printing papers, etc., thus indicating potential spreading due to recycling. Similarly, another study (BMELV, 2012) found the highest concentrations of BPA in board packaging which was assumed to have the highest content of recycled paper. Structural BPA analogues (e.g. BPB, BPS, BPF, etc.) are available on the market, but the potential health effects of substitutes are still to be assessed in detail (Rosenmai et al., 2013). Phenols deserve special attention in terms of paper recycling, as nearly all of them demonstrate a high affinity to solids and are persistent, according to biodegradability criteria. The removal of BPA in the deinking process has been observed to be higher than 50%, but this still resulted in average

350 concentrations of BPA of 10 mg/kg in the board produced (BMELV, 2012). This was in contrast to
351 board based on virgin fibres, where no BPA was detected.

352 **3.5 Parabens**

353 Esters of *p*-hydroxybenzoic acid, or parabens, are commonly used as preservatives in a variety of
354 consumer products (Miljøstyrelsen, 2013). Butyl, ethyl, methyl and propyl parabens, identified in
355 Step 2 (Table S4 (Supplementary Material)) and which may be used as preservatives and biocides
356 by both the paper and the printing sectors (Miljøstyrelsen, 2011a; Vinggaard et al., 2000), are all
357 classified as EDCs and show a tendency to remain in aqueous solution. Hence, they can be expected
358 to be removed in the wet end of paper production. Only butyl and propyl parabens show a partial
359 affinity to solids, which may constitute an issue in paper recycling. Although no limit values for
360 chemicals in paper in Table S4 (Supplementary Material) were available, ‘no release of substances
361 in quantities which have an antimicrobial effect’ applies to food-contact paper, in accordance with
362 paper industry guidelines (CEPI, 2012). In a study investigating the oestrogenic potential of paper
363 for household use, parabens (methyl and propyl paraben) were identified only in samples of paper
364 made from virgin fibres (Vinggaard et al., 2000).

365 **3.6 Inorganics**

366 Out of the 157 chemicals, 22 substances were inorganic (Table S5 (Supplementary Material)).
367 Inorganic chemicals in general, and potentially toxic metals in particular, are used mostly in
368 pigment preparation and coatings (Miljøstyrelsen, 2011a). The presence of Hg could not be
369 attributed to any particular process, and it was therefore assumed to be the result of impurities
370 and/or contamination (Huber, 1997). Nevertheless, two studies addressing waste paper composition
371 found Hg in measurable concentrations (Riber et al., 2009; Rotter et al., 2004). Most of the
372 chemicals presented in Table S5 (Supplementary Material) have not been reported based on

analytical experiments but rather from inventory lists indicating their use by industry. Concentrations of Hg ranged from 0.01 to 0.386 mg/kg, Cd ranged from 0.02 to 0.3 mg/kg, while total Cr was found in the highest concentrations at between 1.1 and 92 mg/kg of paper. Since some pigments and dyes may contain Pb, one study showed that journals and magazines contained the highest concentrations (up to 400 mg/kg) of Pb in recyclable waste paper (BMELV, 2012). The same study also mentioned that the levels of Hg and Cd found were negligible. The limit values for Cd, Pb and Hg in paper and board intended for use in food packaging were set at 0.5, 3.0 and 0.3 mg/kg, respectively (CEPI, 2012).

Due to the nature of inorganic constituents in waste paper, their removal in the recycling process may vary. One relevant study (BMELV, 2012) indicated that newly produced paper products based on recycled paper may still contain considerable concentrations of Pb (up to 26 mg/kg), while concentrations of some metals (Sn, Sb) may even increase during paper recycling, potentially indicating release from machinery (BMELV, 2012). Nevertheless, the authors of the study indicated that the presence of potentially toxic metals in the concentrations measured should not pose health hazards, even if the paper is to be used for food packaging.

3.7 Other substances

The remaining substances not falling within the previous groups amounted to 67 out of the original 157 (Table S6 (Supplementary Material)). Although data on their identification in paper are scarce, several of the chemicals have been quantified in the scientific literature and reports (BMELV, 2012; Ozaki et al., 2004; Storr-Hansen and Rastogi, 1988; Zheng et al., 2001). Polychlorinated biphenyls (PCBs) are classified as “Persistent Organic Pollutants” and are no longer used in paper production (e.g. in the carbonless copy paper), as they were abolished in 1993 (Breivik et al., 2007). Nevertheless, PCBs may persist in the environment, for example accumulated in trees (Hermanson and Johnson, 2007) or other sources (e.g. books and archives), and they may therefore be

introduced into the paper production process. Diisopropyl naphthalene (DIPN) substitutes for PCBs in carbonless copy paper and may be used in other applications (Biedermann and Grob, 2012). It was shown that among the waste paper analysed, office paper contained the highest concentrations of DIPN (up to 1,400 mg/kg), indicating that specialty paper and the use of recycled paper are important sources thereof (BMELV, 2012). The study also showed that unconverted board made from recycled paper and intended for food packaging may contain DIPN ranging from 11 to 27 mg/kg.

Although, since the early 1990s, restrictions in developed countries on the use of elemental chlorine in the paper bleaching process have lowered the possibility of dioxin and furan formation (Ginebreda et al., 2012), these substances may still be detectable in paper products and other papermill outputs, albeit at very low levels (Latorre et al., 2005). The presence of dioxins and furans estimated in papermill effluent waters was in the range of approximately 1-10 ng/m³ (Latorre et al., 2005), while Munawar et al. identified both in lake sediments near pulp and papermill facilities (Munawar et al., 2000). Another study showed waste paper as the main source of dioxins and furans in a paper recycling facility (Santl et al., 1994). The issue is especially relevant for emerging economies, where potentially more lenient environmental legislations are applied and elemental chlorine may still be in use, thus resulting in detectable levels of dioxins and furans in pulp, paper and effluents (Thacker et al., 2007; Zheng et al., 2001).

The attention of the paper industry to some of the chemicals listed in Table S6 (Supplementary Material) has already been drawn, leading to setting limit concentration values for paper and board used in food packaging: DIPN is subject to tests only in products containing recycled paper, and these concentrations should be 'as low as technically possible' (CEPI, 2012). The same guidelines set the limit concentration of Mechler's ketone as low as 0.0016 mg/dm².

3.8 Implications for waste paper recycling and needs for future research

421 Although a relatively small number of substances were identified as critical (157 out of
422 approximately 10,000), there is a need for more information on their presence in waste paper
423 intended for recycling. Quantitative information on the presence of these substances could provide a
424 basis for establishing a priority list of chemicals to be monitored in waste paper prior to recycling as
425 well as in the final paper products. Although the paper industry has already placed focus on a range
426 of substances (e.g. BPA, BBP), the analytical methods needed to monitor others (e.g. substances
427 constituting mineral oils) are not readily available and represent a challenge for future research.
428 While the specific conditions of the paper recycling processes (i.e. temperature, pH, residence time,
429 etc.) may influence the distribution of chemical substances between the solid, air, and liquid phases,
430 more analyses are needed to fully document substance distributions.

431 A general lack of transparency related to the use of specific chemicals for example in the
432 printing industry contributes with uncertainty about the substance load associated with paper
433 products and thereby also with the subsequent quality of waste paper as a resource for recycling.
434 Many of the substances screened in this study could not exclusively be associated with paper
435 printing; however, the substances could not be excluded either based on available information.

436 While banning or gradual phasing out of critical substances in paper production may in the
437 future lead to less chemical substances in paper for recycling, increased source-segregation of
438 individual paper types may also be necessary to ensure a high quality of the paper actually collected
439 for recycling. The preliminary results also indicate the necessity of addressing material quality
440 when establishing target recycling rates. Too high levels of critical substances in waste paper may
441 ultimately mean that this paper should be routed to thermal treatment, thereby enabling the
442 destruction of persistent organic chemicals.

443 **4. Conclusions**

444 The literature review clearly demonstrated that paper and board products, as well as waste paper,
445 may potentially contain a large number of chemical substances, many of these associated with the
446 printing industry. From a total list of 10,000 identified chemicals potentially present in paper
447 products, 157 were classified as hazardous. Fifty-one of these substances were identified as critical
448 as they were likely to remain in the solid matrix during paper recycling and thereby end up in new
449 products based on recycled fibres. The analytical literature reviewed indicated presence of several
450 substances (e.g. phthalates, phenols) in higher concentrations in recycled paper when compared to
451 virgin-fibre based products. If such recycled paper products include food packaging, migration into
452 foodstuff is potentially possible. As almost half of these chemicals (24) are classified as persistent
453 and potentially bio-accumulating, this may pose a risk for consumers. Most of the 51 chemicals are
454 intentionally added during manufacturing, while some of the substances (5) could not be attributed
455 to any of the sectors within the paper industry. These substances may either be added unknowingly
456 by the industry, or originate from contamination of the paper during the use phase or during
457 collection and handling in the waste management phase. The study clearly demonstrates that there
458 is a need for more comprehensive quantitative data documenting the levels of potentially hazard
459 substances in paper sent to recycling as well as the final paper products. Based on the hazard
460 screening procedure, 51 substances have been identified as potentially critical. It is recommended
461 that analytical efforts are directed towards these substances.

462 **Acknowledgements**

463 The authors wish to thank the Confederation of European Paper Industries (CEPI) for facilitating
464 the data collection and providing constructive comments. Veronica Martinez Sanchez and Dr.
465 Alessio Boldrin (DTU Environment) are gratefully acknowledged for commenting on the
466 manuscript and engaging in constructive discussions. The Danish Research Council is acknowledge
467 for financial support through the 3R Research School and the IRMAR project.

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