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Dynamics of bisphenol A (BPA) and bisphenol S (BPS) in the European paper cycle: Need for concern?

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ABSTRACT

Bisphenol A (BPA) is an industrial chemical used as an additive in conventional point-of-sale thermal paper receipts. Due to BPA being an endocrine disruptor and a substance of very high concern, the European Union (EU) has proposed to ban its use in thermal paper from 2020. Potential similarities in toxicological profiles have raised concerns that the use of bisphenol S (BPS) as a substitute for BPA may result in yet another situation of a problematic chemical being distributed in consumer products. This study provides a comprehensive evaluation of the current knowledge of BPA and BPS use in thermal paper and, based on dynamic material and substance flow modeling, quantifies potential effects of the BPA ban on future BPA and BPS flows within the European paper cycle. Based on available data and the modeling of BPA and BPS flows, approximately 200 t of BPS are estimated to be present in the current European paper cycle. The modeling further demonstrated that by substituting 50% of BPA, BPS amounts in the European paper cycle would increase more than fivefold over a modeling period of 60 years. In the same time, more than 90 t of BPA would still be circulated in European paper products. BPA alternatives other than BPS should receive additional attention, as very limited quantitative data currently exist. The results of this study quantitatively demonstrate that chemical bans alone are not sufficient to ensure clean material cycles, and so the effective regulation of potential substitutes needs to be implemented in parallel.

1. Introduction

Bisphenol A (BPA) is an industrial chemical with global annual production surpassing 6,000,000 t in 2012 and anticipated growth in production of approximately 30% by 2020. BPA is used in a variety of applications, resulting in its use by a number of industries and its presence in material exchange between numerous sectors of economy (Jiang et al., 2017). BPA is primarily used as monomer in polymer production (i.e. polycarbonate or epoxy resins), used in manufacturing of plastic products (e.g. water bottles, food containers, polycarbonate "glass") or applied as coating (e.g. protective layer in tin cans). In addition, BPA is applied in form of an additive (developer) in thermal paper production, where it is used in its "free" (non-reacted) form. Developers in thermal paper are used in paper coating and react with heat sources by changing color and resulting in image development without the use of inks as in conventional printing (US EPA, 2014). Thermal paper is used primarily in conventional receipts in the retail sector (e.g. shop or supermarket receipts, ATM slips), potentially leading to human (consumer and professional) exposure to BPA through handling (Geens et al., 2011; Hehn, 2016; von Goetz et al., 2017). The majority of BPA present in paper material and products comes from its use in thermal paper, while BPA can potentially be used in ink and glue formulations (in form of e.g. bisphenol A diglycidyl ether (BADGE)) also applied on paper (Pivnenko et al., 2016a). Once BPA-containing paper is recycled, some BPA will be removed (primarily as paper sludge) during paper re-processing, while a fraction thereof may remain in the recycled materials and contaminate new paper products (Liao and Kannan, 2011a; Pivnenko et al., 2016a; Pivnenko et al., 2015b). Widespread use of BPA in conventional consumer products with high degree of potential exposure, and its endocrine-disrupting (hormonal) effects on living organisms, have led to gradual restrictions on BPA use and imports of selected BPA-containing products in parts of Europe, Asia, and the US (EU, 2011; NITE, 2003; State of Connecticut, 2013). Moreover, in June 2017, the Member State Committee of the European Chemicals Agency (ECHA) recognised BPA as an endocrine disruptor and a substance of very high concern (SVHC) (ECHA, 2017), leading to anticipated further restrictions on its use.

European paper production is a well-established example of an

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effective material recycling system in which about half of raw material needs originate from recycled fibers (CEPI, 2013). To ensure clean material flows and safe recycling, “problematic” chemical substances in the recycled materials should be limited (Pivnenko and Astrup, 2016). Recent literature has demonstrated that the selective recycling of “clean” material fractions, or the improved removal of substances during reprocessing, may not be sufficient to ensure BPA-free paper products without compromising quantitative recycling rates (Pivnenko et al., 2016a). Substance phase-out was therefore considered necessary.

In December 2016, the EU decided to set a maximum concentration limit of 200 µg of BPA per gram of thermal paper by 2020, effectively phasing out BPA use in thermal paper (EC, 2016a). While no substitution strategies have been proposed, existing and potential alternatives to BPA have been suggested (e.g. Pivnenko et al., 2015b; US EPA, 2014). There is a number of BPA alternatives that are already in use or can potentially substitute BPA in thermal paper, including phenol-based (e.g. bisphenol S (BPS), bisphenol E (BPE), bisphenol F (BPF)) and phenol-free (e.g. Pergafast® and urea urethane) chemical compounds. Several of the proposed BPA substitutes are chemical analogues to BPA with similar molecular structures (Usman and Ahmad, 2016). Recent research has reported that some BPA substitutes have equal or even greater toxicological effects, with endocrine disrupting mechanism of action similar to that of BPA (Chen et al., 2016; Eladak et al., 2015; Kinch et al., 2015; Usman and Ahmad, 2016). BPS is a prominent example of a BPA substitute (Pivnenko et al., 2015b; US EPA, 2014). It is a structural analog to BPA with similar *in vitro* metabolism, potencies, and mechanisms of action (Rochester and Bolden, 2015). In addition, it has reported adverse effects in humans and rodents (Eladak et al., 2015), aligned with a number of health hazards (Rochester and Bolden, 2015). Among other applications, BPS is registered in the EU for use in formulations for “industrial manufacture of paper” (EC, 2006), but current amounts in the European paper cycle are virtually unknown. Although concerns about the current and potential future substitution of BPA by BPS have been expressed (EC, 2016b), quantitative data on presence, partitioning and persistence of BPA and BPS in the paper material cycle is extremely limited (Pivnenko et al., 2016a; US EPA, 2014). Without clearly defined substitution strategy for BPA, increased use of potentially harmful alternatives can be expected. The question is, to what extent will the European ban ensure BPA-free paper flows, and how might levels of BPS develop over time? In view of limited data availability and restricted timeframe for BPA ban in the EU to enter into force, comprehensive literature overview and systematic modeling can offer useful insights for decision-making.

The aim of this study is to provide an evaluation of the available literature on presence of BPA and BPS in thermal paper and to investigate the effects of phasing-out BPA and phasing-in BPS, as its partial replacement, within the European paper cycle. The intention is to quantify and demonstrate systematically the consequences of recent BPA-related European legislation and the potentials for achieving a clean material cycle. The specific objectives are to: i) Provide a comprehensive overview and evaluation of available data on the presence of BPA and BPS in thermal paper; ii) Establish a dynamic model for BPA and BPS based on a state-of-the-art material flow model of the European paper cycle (Pivnenko et al., 2016a); and iii) Based on proposed European legislation, define scenarios for phasing-out BPA and determining potential consequences for BPA and BPS flows in the European paper cycle. The work illustrates the principles for estimating temporal changes associated with chemical substitution within material recycling loops—principles that could be applied to other cases where the presence of chemicals (e.g. conventional use, restrictions on use or introduction of new chemicals) in materials is of potential concern.

2. Materials and methods

2.1. Literature overview and evaluation

The existing literature concerning BPA and BPS in thermal paper receipts was reviewed from a global perspective. Scientific publications, public reports, patents, and ongoing projects were identified and evaluated. In total 29 experimental studies were identified, covering the period between 2009 and 2017. In case actual year of thermal paper sampling was not mentioned in a study, the publication year was used. The presence of either BPA or BPS in thermal paper may result from direct application of additives (e.g. developer in paper coating) or as contamination (e.g. through formulation impurities, paper recycling (up to 0.5 mg/g) or direct contact with bisphenol-rich thermal paper (up to 0.1 mg/g)) (Konkel, 2013; Liao and Kannan, 2011b; Pivnenko et al., 2015a, 2015b). However, an unbiased threshold for distinguishing between amounts of BPA or BPS added directly or via contamination has not been identified. Hence, all reported concentrations above the respective limits of detection (LODs) were accounted for in this work. Values below respective LODs were excluded from statistical analyses for simplicity and transparency, as common substitution methods may have limited applicability at high degrees of censoring (Antweiler, 2015, 2008).

2.2. Material and substance flow modeling

Flows of BPA and BPS in the European paper cycle were modeled, following the state-of-the-art model with a step-wise modeling approach (Pivnenko et al., 2016a). In brief, the stepwise model was based on a combination of static and dynamic material and substance flow analysis. Static paper flow analysis was carried out in a previous study (Pivnenko et al., 2016a) to quantify paper flows in Europe in 2012. Although there is some decrease in the European paper consumption between 2007 and 2009, consumption remained rather steady in the period 2012–2016 (CEPI, 2016). Hence, the earlier study concerning year 2012 can be considered representative of the current flows of paper in the European paper cycle. On the other hand, static substance flow analysis was expanded with new and detailed data concerning BPA and BPS amounts in end-of-life (EOL) paper fractions and thermal paper (Tables 1–4). In addition, the present work is aiming to reflect on the terms of recent European legislation proposing a ban of BPA use in thermal paper and focus explicitly on the modeling of BPA phase-out and BPS phase-in within the European paper cycle. The substance flow model is primarily based on chemical concentrations in the EOL paper and paper products as model input and output parameters, respectively. Figure A1 (Appendix A in Supporting Information) provides detailed overview of the inputs and outputs from each of the modeling steps. Definitions, key assumptions (further addressed in Section 2.5) and data sources for EOL paper fractions are outlined in Table 1 and final concentrations of BPA and BPS in discarded paper are presented in Table 2.

2.3. Scenario analysis

In paper products, BPA is primarily used as a developer in thermal paper production, but also as a plasticizer and plasticizer precursor for ink and glue formulations for paper conversion (Pivnenko et al., 2016a). A probable reduction in BPA use could lead to increased use of BPS and other alternatives, either in thermal paper only or in other paper products as well. Since BPS is potentially used as a partial substitute of BPA, the evaluation of BPS flows requires better understanding of BPA flows in the context of the newly proposed European legislation (i.e. scope, timeframe and maximum concentrations). In