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# Development of eco-efficient smart electronics for anti-counterfeiting and shock-detection based on printable inks

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**Key words:** life cycle assessment; electrochromism; piezoelectricity; anticounterfeiting; shock-detection; printed electronics; label; sensor;

## Abstract

Printed electronics are expected to meet an increasing demand for improved functionality and autonomy of products in the context of Internet-of-Things. With this trend the environmental performance of novel technologies is of growing importance. The current study presents Life Cycle Assessment of two novel devices: an anti-counterfeit label based on electrochromic display, and a shock-detection tag based on piezoelectric sensor, designed for the use in packaging of pharmaceuticals and luxury items to improve the safety and accountability in the supply chain. The devices are manufactured by means of energy-efficient printing techniques on low-cost flexible and recyclable paper substrate. Comprehensive cradle-to-grave analysis contributes with industrial-scale energy and material life cycle inventories and identifies main impact hotspots evaluated for a broad range of categories of ReCiPe midpoint (H) impact assessment method. Results show that major impact burdens are associated with NFC chip and RFID antenna while the impacts of solvents, process energy, electrochromic display/ piezoelectric sensor, Li-ion battery and substrate are comparatively small. In terms of their global warming potential, both the

anti-counterfeit label and shock-detection tag embody around 0.23 kg of CO<sub>2</sub>-eq. Several material-use reduction and material-substitution strategies are quantified and discussed for their potential to reduce high impacts of antenna.

## Introduction

Printed electronics aim to respond to increasing demands for connectivity, communication and information exchange in context of the Internet-of-Things (IoT), a network of “smart” everyday objects featuring enhanced functionality and device autonomy. Printed electronics (also referred as organic electronics) use traditional printing techniques and complex inks to fabricate electronic devices such as electronic displays, circuits, sensors, and electromagnetic tags <sup>1</sup>. Compared to manufacturing of traditional circuit boards, novel electronics entails fast and continuous manufacture on flexible substrates, featuring increased productivity, lower costs, and extended range of applications. These advantages are compounded by the continuous improvement of the printing techniques, the ink quality and the growing range of products requiring advanced functionalities <sup>2</sup>. With proliferation of these technologies there is a raising concern about their impact on the environment. Advanced organic and inorganic inks, although used in small amounts, embody high environmental impacts per unit of mass <sup>3</sup>, while printed electronics should be subjected to added scrutiny given their potential application for energy saving and renewable energy supply systems <sup>4,5</sup>.

Recent advancements in printed electronics have been focusing on development of two devices: an anti-counterfeit label (ACL), and a shock-detection tag (SDT). ACL can be attached to the products to enable authentication during their transportation and storage. This technology is highly relevant for many products prone to counterfeiting such as luxury items and pharmaceuticals <sup>6,7</sup>. This function is enabled by change of colour induced by a redox reaction within the electrochromic material embedded in electrochromic display (ECD). Under applied electrical potential the material switches from a neutral to a coloured state through either a reduction mechanism (cathodic coloration), an oxidation mechanism (anodic coloration), or a combination of two <sup>8</sup>, without emitting any source of light <sup>9-11</sup>. Traditionally, these materials were used to alter the transparency of glass to regulate penetration of light in self-dimming rear mirrors for cars, and smart windows for energy-efficient buildings <sup>5,12 13</sup>, and niche applications in dynamically tinting goggles <sup>14</sup>. Use of electrochromic materials to power low-energy displays are more recent and have find utility in various consumer electronics such as washing machines <sup>15</sup>, smart cards and labels, and medical appliances <sup>16</sup>.

SDT is designed to detect and record the damage of goods when exposed to shock, fall, or vibration during shipment. This function is enabled by piezoelectric materials embedded in piezoelectric sensor (PS) that generate electrical charge in response to an external mechanical stress. While piezoelectric materials have a widespread use in medical, aerospace, information technology and automotive industry

for sensor, actuators and oscillators for microprocessor-based systems <sup>17</sup>, their use in autonomous shock-detection devices is novel and prompted by emergence of polymeric piezoelectric materials. In comparison to traditional piezoelectrics based on inorganic ceramics or single crystals of lead zirconate titanate, barium titanate or lead titanate, the piezoelectric polymers are cheaper, processable on flexible substrate, and avoid the use of heavy metals.

The environmental performances of ACL and SDT are currently considered concomitant to their development and production (i.e., design optimization and upscaling) to realize opportunities for reducing environmental impacts of the new technologies at early stages of technological maturity <sup>18</sup>. Environmental impacts of new technologies are best studied using Life Cycle Assessment (LCA) <sup>19</sup>. This method enables to evaluate impacts of emissions to the environment throughout the life cycle of the product: production of raw materials, manufacture, product use, and end-of-life. In LCA, impacts of the emissions are evaluated for an extensive range of environmental impact categories to describe potential damages on human health, ecosystems, and resource use <sup>20</sup>. This is particularly relevant for avoiding burden shifting in assessment of more complex functional materials associated with diverse range of impacts often extending the scope of “popular” pollutants such as greenhouse gases.

Research into environmental impact of anticounterfeiting and shock-detection devices and integrating electrochromic and piezoelectric materials using LCA is conservative and it is hard to draw on any previous LCA studies that are directly related to the features or functionalities of ACL and SDT. Previous LCA studies on electrochromic materials focus on application in smart windows for energy saving which entails use and associated architectures of electrochromic devices quite different from that of ECD <sup>23,24</sup>. Several studies report development of “green” materials for anti-counterfeiting application, employing bio-based materials and solvents, and low-energy processes <sup>21,22</sup>, but evaluation of new materials is not supported by any formal environmental impact assessment. LCA studies of piezoelectric materials mostly involve research into benefits of lead-free piezoelectrics <sup>25</sup>, and synthesis and formulation of piezoelectric polymers <sup>26</sup>. Their use for shock-detection in sensory applications involve comparison between use of piezoelectric sensors with conventional strain gauges to indicate need for road maintenance <sup>27</sup>. These sensors have shown lower environmental impacts and overall better functional reliability.

With the attention to the existing gaps, the current study aims to assess environmental performance of ACL and SDT, contribute with comprehensive life cycle inventories for fabrication of two devices and identify opportunities for future impact mitigation. The notable feature of the new devices is an anticipated green design involving an avoidance of transparent conductive oxide such as indium tin oxide (ITO) and the choice of paper for a substrate as a bio-based and highly recyclable material <sup>28,29</sup>. ITO is generally used to build the transparent and conductive electrodes but employs energy-intensive deposition methods while indium is considered critical raw material <sup>30</sup>. Current assessment reveals if

some of these design features lead to anticipated improvements while exploring how the design can be further influenced to mitigate environmental impacts.

## **Methodology**

### *Goal and scope*

The study is carried out according to recommendation of ISO 14040 and 14044 standards for LCA studies, citing four stages of LCA: goal and scope, life cycle inventory analysis, life cycle impact assessment, and interpretation<sup>20,31</sup>. Referred standards and associated recommendations improve reliability, reproducibility and transparency of LCA studies.

### **Goal definition**

The goal of the current LCA study is to determine the environmental performance of two products based on printable electronics: anti-counterfeit label (ACL) and shock-detection tag (SDT). The study seeks to identify opportunities for impact mitigation and to investigate early product design optimizations while creating a reference point for future development of these technologies. The analysis was carried out for involved industry and research partners while it has a broader significance for the academic and industrial research in domain of printed electronics and IoT.

### **Product systems**

Although they serve entirely different functions, ACL and SDT are structurally very similar. They incorporate some identical components and are designed to communicate with smartphone that enables change of colour of the ECD and readings from the SDT. Schematic of different components in ACL and SDT are shown in Figure 1, and further breakdown of components of electrochromic display (ECD) and piezoelectric sensor (PS) are shown in Figure 2.

ACL integrates the ECD, substrate, radio-frequency identification (RFID) antenna, and near-field communication (NFC) chip. An ECD is built from successive layers namely: two electrodes conducting electricity, the electrochromic layer able to switch from one colour to another and the electrolyte as an ion conductor (Figure 2-a). All the layers are printed on a paper substrate with two encapsulation layers to protect the display from the environment and improve stability over time. Paper substrate acts as a low-cost and flexible substrate that facilitates easy printing of inks and attachment of other components. The choice of paper is also hoped to improve device recyclability.

The architecture of SDT features the PS, substrate, RFID antenna, NFC chip, and Li-ion battery. PS is made of two electrodes able to collect charges and a piezoelectric materials which accumulate electrical

charges under an applied stress (Figure 2-b). Similar to ECD, all the layers are printed on a paper substrate. The battery provides power input to the sensor, allows logging and time-tracking of data, and communicates with a smartphone upon activation of the tag. The battery and chip allow to track if a strain has been applied to the good during the whole transportation. The antenna and ECD/PS are printed using ink-based solutions via screen-printing technique while the chip and battery are bonded via pick-a-place equipment onto the substrate surface.

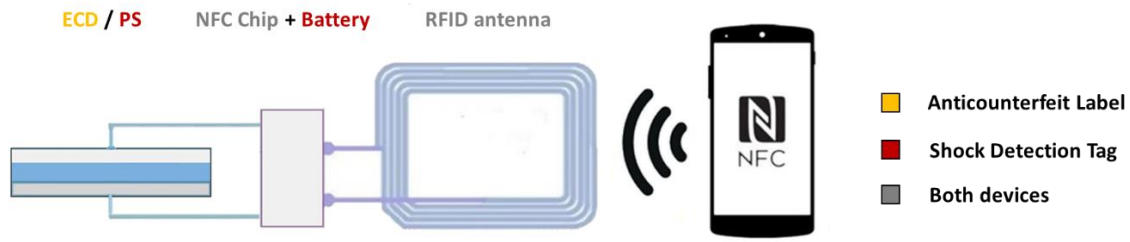


Figure 1. Schematic of anti-counterfeit label (ACL) and shock-detection tag (SDT)

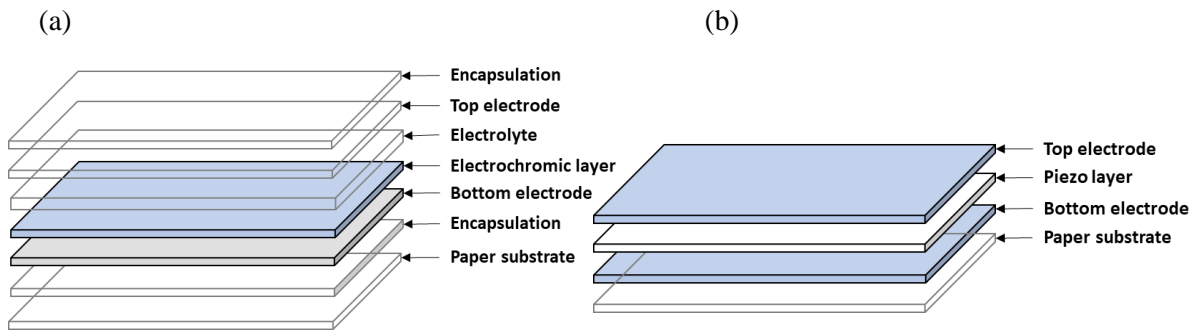


Figure 2. Schematic of (a) electrochromic display (ECD) and (b) piezoelectric sensor (PS)

Current technology corresponds to  $1\text{cm}^2$  of ECD that switches between a neutral to a coloured state in 2s under applied voltage of 3.8V giving an optical contrast ( $\Delta E^*$ ) higher than 15. PEDOT:PSS is used as electrochromic layer. This conjugated polymer shows a cathodic coloration from transparent/light blue at its oxidized state to dark blue in its reduced state<sup>32</sup>. The optical contrast is defined with the  $L^*$ ,  $a^*$ ,  $b^*$  chromaticity parameters which belong to the CIE colorimetric space and thanks to the equation (1).

$$\Delta E^* = \sqrt{(L_{\text{red}}^* - L_{\text{ox}}^*)^2 + (a_{\text{red}}^* - a_{\text{ox}}^*)^2 + (b_{\text{red}}^* - b_{\text{ox}}^*)^2} \quad (1)$$

With  $L_{\text{red}}^*$ ,  $a_{\text{red}}^*$ ,  $b_{\text{red}}^*$  represents the chromaticity parameters at the reduced state and  $L_{\text{ox}}^*$ ,  $a_{\text{ox}}^*$ ,  $b_{\text{ox}}^*$  represents the chromaticity parameters at the oxidized/neutral state.

## System boundaries

The analysis covers all stages in the life cycle of ACL and SDT: production of raw and auxiliary materials, manufacturing of the devices, their use, and end-of-life. Several processes of papermaking and coating are involved in producing the paper substrate, followed by processes of screen-printing of electronic components antenna, antenna tracks and ECD/PS, and finally a silicon chip bonding. In case of SDT, the printing of PS is also followed by poling of the piezoelectric materials and bonding of battery. End-of-life stage predicates two products are either recycled or landfilled. Recycling of ACL and SDT, as paper-based devices attached onto paper packaging, entail treatment in a paper recycling facility. Secondary paper pulp recovered through recycling is assumed to displace virgin production of pulp according to avoided burden approach in LCA <sup>33</sup>.

Cut-off to system boundaries applies to capital goods and foreground transportation emissions since those could not be envisioned at this stage of technological maturity and are anticipated to be very small given the weight of the devices. Impacts in the use phase are only associated with the energy required from smartphone to power ECD that is assumed to be negligible. Cut-off to system boundaries also applies to trace chemicals and impacts of some of the outputs of the recycling where the transport and fate of materials could not be adequately anticipated and modelled <sup>34</sup>. These assumptions are further detailed in the life cycle inventory section dedicated to the assumption and modelling of the end-of-life phase.

As a scenario analysis, we compare impacts of paper substrate with the more conventionally-used polyethylene terephthalate (PET) alternative. Generally, printed electronics typically use PET because of its high tensile strength, resistance to humidity, thermal stability and price compared to other plastics <sup>35</sup>. This comparison was expected to reveal importance of the choice of substrate in the life cycle of the device and potential advantages of paper substrate recyclability. The comparison is only carried out for ACL device with similar results expected for SDT.

Scenario analysis also explores strategies for mitigating impacts associated with the RFID antenna. Two strategies are compared with the default scenario (use of micro-silver ink for screen-printing of antenna, antenna tracks, and bottom electrode): 1) flexography printing using nano-particle silver ink, and 2) screen-printing using nano-particle copper ink. Flexography allows printing of antenna with thinner layer (3 – 3.5 $\mu$ m) to obtain similar conductivity of that from screen-printed antenna based on silver micro-particles (12 $\mu$ m layer thickness) resulting in the reduction of roughly three times in the use of ink. A scenario involving screen-printing with copper ink was borne out directly from this study as a printed copper antenna on a paper substrate has not been priorly succeeded in practice and published as an LCA inventory. Copper-based ink is deemed as a potentially good replacement for silver given lower costs and similar conductivity <sup>36</sup>, but given its drawbacks in terms of instability against oxidation and

high sintering temperature <sup>37</sup> it was not certain if this scenario was viable, unless proven. Several experiments were carried out to assert viability of this scenario and derive an inventory for this process (i.e., material uses and energy requirements for sintering necessary to model this scenario in LCA).

The environmental impact of demonstrators is determined for categories of ReCiPe Midpoint (H) impact assessment method <sup>38</sup>. Inventory of precursor materials was derived using the ecoinvent database version 3.6 for cut-off allocation approach <sup>39</sup>. This implies the use of attributional LCA approach (in distinction to a consequential approach in LCA) which was deemed the most suitable given the design-optimization focus in this study and the level of technological maturity of the devices. Classification and characterization of data are carried out using OpenLCA v1.10.2, an open-source LCA software (Green Delta, Berlin). Data for the foreground system were mostly collected empirically directly from the industry. In other cases, estimates were made to resemble prospective industrial-scale operations.

### **Function and functional unit**

Anti-counterfeiting function of the label could be stated as enabling product verification through change of colour over a period of product's transport and delivery to the final consumer. This functionality is conditioned by the number of times visible colour change can be induced, and the lifetime of the label. Therefore, we define functional unit as producing at least 20 times a visible chromacity change after receiving a 13.56 MHz signal (from the smartphone) over a period of 2 years. A single ACL device, deemed sufficient to enable a single product verification under described nominal conditions, was adopted as the reference flow.

The function of the SDT is to detect the shock or vibration during the transport that could cause the damage. Therefore, the functional unit is defined as detecting and recording any frequency above 13.56 MHz over the product's transportation, translating into a voltage signal readable by a smart phone. A single SDT is adopted as a reference flow.

### *Life Cycle Inventory*

Figure 3 shows process flowchart for the ACL and SDT. Inputs of resources and energy and outputs of waste arising in these processes (i.e., inventories) are used as a basis for environmental impact assessment model. Data is collected for four process stages described in this section with specific inventories disclosed in Supporting Information (SI) document:

- Production of substrate: Powercoat XD125 and PET (Table S-1)
- Printing of RFID antenna and ECD/PS (Table S-2 and Table S-3)
- NFC chip and Li-ion battery bonding (Table S-6 and S-7)



- End-of-life (Table S-9)

Geometries of components for each device are shown in Table 1, and further breakdown of components of ECD/PS provided in Table 2.

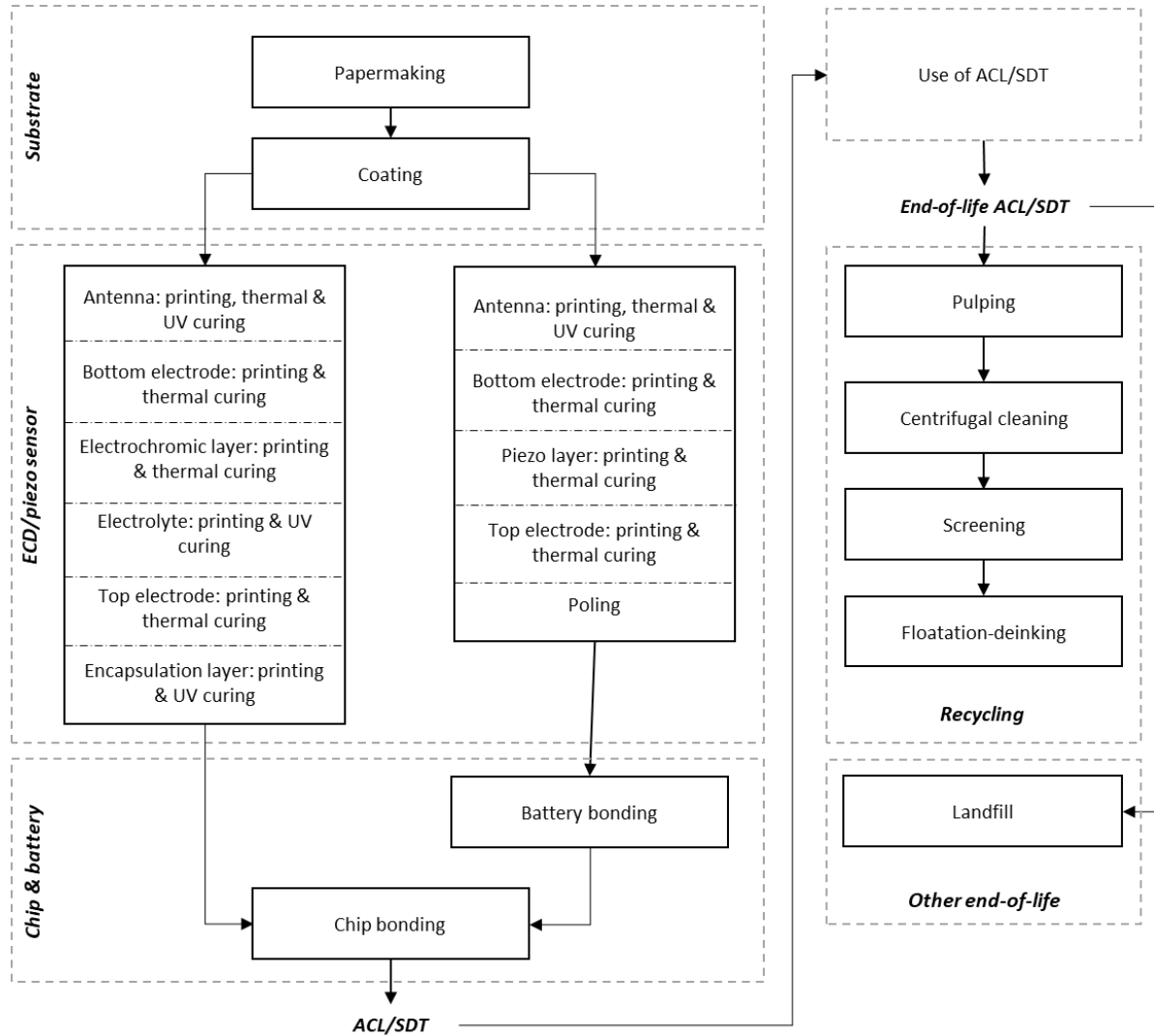


Figure 3. Processes in the life cycle of ACL and SDT.

Table 1. Dimensions and weight of ACL and SDT devices and components

		Device	Substrate	Chip	Battery	Printed inks
<b>Anti-counterfeit label</b>	Weight (g)	0.76	0.58	0.6	-	0.18
	Area (mm <sup>2</sup> )	4644	4644	1.66	-	-
	Volume (mm <sup>3</sup> )	-	-	5.54	-	-
<b>Shock-detection tag</b>	Weight (g)	1.62	0.68	0.6	0.86	0.08
	Area (mm <sup>2</sup> )	5400	5400	1.66	1157.31	-
	Volume (mm <sup>3</sup> )	-	-	5.54	486.07	-

Table 2. Dimensions of ECD and PS layers

Device component	Electrochromic display (ECD)			Piezo sensor (PS)		
	Printed surface (cm <sup>2</sup> )	Thickness (μm)	Volume (mm <sup>3</sup> )	Printed surface (cm <sup>2</sup> )	Thickness (μm)	Volume (mm <sup>3</sup> )
Antenna	11.6	13	15.08	12.1	13	15.73
Encapsulation	9.1	17	15.47	1	17	1.7
Antenna tracks	3.7	13	4.81	0.85	13	1.11
Insulation	7.1	20	14.2	-	-	-
Bottom electrode	3.7	10.5	3.89	7.8	1	0.78
Bottom electrode (carbon)	3.7	6	2.22	-	-	-
Electrochromic material	1.8	1	0.18	-	-	-
Piezo layer	-	-	-	9	5	4.5
Electrolyte	3.42	50	17.1	-	-	-
Electrolyte (photopolymer)	2.28	50	11.4	-	-	-
Top electrode	5.1	10.5	5.36	7.8	1	0.78
Encapsulation	7.1	20	14.2	-	-	-

### Production of substrate: Powercoat XD125 and PET

Raw material and emission inventory for Powercoat XD125 are constructed based on data provided by the manufacturer Arjowiggins France. Production of Powercoat XD125 undergoes stages of paper(base)-making and coating based on the production mill at Guarro Casas in Barcelona, for which inventory data was supplied as aggregated values. Two processes involve the use of additives (14.3 wt%) and calcium carbonate mineral filler (17.7 wt%), and two types of virgin eucalyptus-based paper pulps (68 wt%). Paper consumption is associated with productivity per SRA3 sheet (460\*320 mm<sup>2</sup>) which can supply substrate for 25 labels, or 10 tags, and Powercoat XD125 density of 125 g/m<sup>2</sup>.

Inventory for PET substrate considered in scenario analysis, was based on the datasets: “polyethylene terephthalate, granulate, amorphous”, and process for extrusion: “extrusion plastic sheets and thermoforming, inline”. The use of PET resin was based on projected thickness of 125 μm, manufacturing losses identical to Powercoat XD125 (16%), and losses due to extrusion (3.1%). For ACL, the weight of the PET substrate was approximated to be 2.377 g and PET resin use of 3.1 g.

### Printing of RFID antenna and ECD/PS

Screen-printing of antenna and ECD is carried out as a multistep process that involves: a) printing, drying and UV curing of the antenna, antenna tracks and bottom electrode b) printing and drying of electrochromic layer, c) printing and UV curing of electrolyte layer, d) printing and drying of the top electrode, and e) printing and UV curing of the encapsulation layer. Similar steps are carried out in

fabrication of PS. The main difference is the absence of electrolyte and electrochromic layer. Instead, there is a step of printing and thermally curing, and poling of the piezoelectric polymer.

Antenna, antenna tracks and bottom electrode (for ACL) are printed using either micro-particles-based silver ink (for default screen-printing) or nano-particles-based silver ink (for scenario involving flexography printing of antenna). Inventory for formulation of nano-particle-based silver and micro-particle-based silver particles constituting 70 wt% of ink is derived from secondary literature for production nano-particle of silver from metallic silver<sup>40</sup>. The application of nano-particle silver dataset to represent impacts of micro-particle silver was deemed acceptable given that production of micro-particle silver could be step in production of nano-particle silver<sup>41</sup>, and since impacts of silver ink is dominated (>95%) by production of silver metal itself which is expected to be used at high efficiency in formulation of either micro-particle or nano-particle-based silver inks. Electrochromic layer and electrodes for PS are printed using PEDOT:PSS ink. Dataset for PEDOT:PSS was built using secondary literature<sup>42</sup>. Thiophene precursor in the synthesis of PEDOT:PSS was modelled assuming continuous production from n-butane and sulphur at 560 °C<sup>43</sup>. Top and bottom electrodes of ECD are printed using carbon ink. The dataset for carbon-based ink was built by assuming 33 wt% ink solids based on graphite and 5 wt% binder. Insulation and encapsulation of components in the display, antenna and ECD was carried out using UV photopolymerization. Electrolyte ink was formulated combining LiTFSI:EmimTFSI and UV photopolymer (60:40 wt%). The impact of LiTFSI:EmimTFSI was represented by previously published inventory for LiTFSI<sup>44</sup>. Polymer polyvinylidene fluoride trifluoroethylene (PVDF-TrFe) ink was used as a piezo material for the sensor. The sensor is poled involving high voltage current in order to turn all the polymer chains in the same direction. Ethyl acetate and water are used as solvents for washing. Ethyl acetate is also considered in formulation of silver (20 wt%) and carbon ink (62 wt%). Waste outputs from printing involve wastewater from cleaning the printer mask and consumables supporting the printing process. Solid waste fraction is projected for incineration without energy recovery, and the wastewater is modelled as average municipal, following presumed neutralization and silver metal precipitation and recovery. We do not take credits for silver recovery as this has not yet proven and develop at the current pilot scale. The energy consumption of printing antenna and layers of ECD and PS are measured using wireless monitoring equipment E-cube-ZB35A0 and E-cube-ZB180A0 by Gulplug. Energy consumption is measured for different processes and printed components and disclosed in SI, Table S-4 and Table S-5.

The scenario for flexography printing of antenna takes into account only reduction of silver ink usage. The quantities of waste generated in the printing process, and energy uses during printing are taken analogous to the default screen-printing as the data of these flows were not available and since they have shown to have small influence on overall impacts of devices. However, in comparison to screen-printing, energy use in flexography is expected to be lower. Screen-printing is around 5 m/min and air

drying for 15min while flexography is carried out at the rate of 15 m/min with drying using near-infrared technique in less than 2 seconds.

The inventory for nano-copper ink used in scenario analysis relates to nano-copper ink formulation and sintering after screen-printing. Deriving of inventory constituted experimental work involving screen-printing of three types of nano-copper ink on paper, subsequent sintering and aging accompanied by measurement of resistance. Samples were sintered at temperatures for different ranges between 200-300 °C for a duration between 5 and 60s using a heat press, while aging tests were carried out in a climatic chamber for 475 h at 85% relative humidity emulating conditions of approximately 10 years in ambient conditions. Two inks are successfully printed but only one retains good conductivity (low resistance) after long aging. Thus, optimal operating conditions were determined for the 60s at 220°C combined with curing for 2 min at 80°C to achieve the resistance of 20.4  $\Omega/\square$  while maintaining the integrity of the paper substrate, which is similar to the resistance of the default silver antenna (22.7  $\Omega/\square$ ). Resistance of the copper antenna increases with aging only slightly more than silver (40% in comparison to 16 %). The composition of tested ink is reported to contain 85 wt% of solids (80 wt% nano-copper and 5 wt% acrylic resin binder) and the rest is assumed as solvent ethyl acetate (15 wt%). Chemical agents (<1%) assisting sintering are presumed negligible and not considered. Consumption of ink and energy for printing is taken identical to screen-printing of micro-particle silver ink due to the similar conductivity of printed antenna, the content of metals in the ink and antenna layer thickness. Formulation of nano-copper is modelled combining efficiencies as observed in Slotte et al. <sup>45</sup> for the synthesis of silver and copper through physical reduction following to UDE-1 arc-based scenario with reported LCA inventory for the formulation of silver nanoparticles <sup>40</sup>. Energy used for sintering and curing was approximated at 1.5 Wh.

### **NFC chip and Li-ion battery bonding**

Inventory for fabrication and bonding of the chip and battery are represented by ecoinvent datasets for silicon-based wafer NFC chip and Li-ion battery, and the use of adhesive and electricity for their bonding onto the substrate. Raw materials requirements for chip and battery are estimated according to their weights in the devices and use of adhesive to the areas they occupy on devices. The quantity of adhesive is approximated to cover entire area of the chip and 1/10 of the contact area with the battery. Energy for bonding is approximated based on power specifications of “pick-and-place” machine TAL15000. Bonding is carried out on per-item basis, so the energy for chip and battery bonding was assumed equal despite the differences in dimensions.

### **End-of-life**

We assume that 72 % of devices are recycled following conventional recycling process for the paper and remaining 28 % landfilled, taking into consideration rate of paper recycling in Europe <sup>28</sup>. These rates anticipate that devices would be characterized as a waste paper stream providing their attachment to paper packaging. For the landfill fraction we considered the background ecoinvent dataset for average municipal waste disposal.

The dataset for recycling was built combining data related to conventional paper recycling and potential removal rates of non-paper components of the devices tested in laboratory. The dataset for recycling of paper was built based on data reported for recycling company Greenfield in France. The paper recycling process involves several stages: pulping, centrifugal cleaning, screening, floatation and washing process. Inputs to the process include the use of water and surfactant, and energy in form of electricity and natural gas. Outputs of the process include solid waste sent to the landfill and wastewater disposed off to the wastewater treatment plant. The paper sludge from the floatation stage is utilized in agriculture (75 %) and brick manufacture (25 %). Impacts or benefits of use of paper sludge in brick manufacture was not accounted given limited information.

The impacts associated with non-paper components is estimated by solid waste fraction of non-paper components removed at screening stage and paper sludge removed at the floatation stage containing various printed components (see Table S-8, SI). In particular, we account for potential emissions to the soil from silver removed during floatation stage. Here, the impacts of silver to the soil reflect solely from the use of silver metal and do not consider the potential toxicity effects related to the physical properties of the compound (i.e., particle size). Impacts of PEDOT:PSS and electrolyte also removed at floatation are excluded so as part of the silver that re-enters paper production through recovery of secondary paper pulp. The flow of these fractions were difficult to plausibly anticipate and model given limitations pertaining to characterization of novel materials with current impact assessment methods in LCA.

## **Results and discussion**

### *Life cycle impact assessment*

The impact assessment results for two devices are shown in Table 3 and Table 4 for ACL and SDT, respectively. Impacts correspond to a life cycle of a single ACL and SDT device. Impact assessment values are also calculated separately for ECD and PS and included in SI, Table S-10 and Table S-11. The results are generated for 18 impact categories with intention to cover broad range of possible impacts.

Table 3. Impact assessment values of ACL

Impact category	Unit	Paper substrate	Chip fabrication & bonding	Printing antenna & ECD	End-of-life ACL	Total
Land use	m <sup>2</sup> a crop eq	5.00E-04	1.35E-03	5.35E-03	-1.79E-06	7.20E-03
Marine eutrophication	kg N eq	2.72E-07	1.18E-05	4.24E-06	3.47E-08	1.63E-05
Marine ecotoxicity	kg 1,4-DCB	7.79E-05	5.64E-03	1.99E-01	3.55E-04	2.05E-01
Freshwater eutrophication	kg P eq	4.27E-07	9.41E-05	2.00E-04	-4.51E-08	2.94E-04
Stratospheric ozone depletion	kg CFC11 eq	9.04E-10	9.54E-08	8.79E-08	8.75E-11	1.84E-07
Freshwater ecotoxicity	kg 1,4-DCB	5.99E-05	4.30E-03	1.25E-01	5.52E-04	1.30E-01
Human non-carcinogenic toxicity	kg 1,4-DCB	8.11E-04	7.52E-02	1.90E+00	1.94E+00	3.91E+00
Global warming	kg CO <sup>2</sup> eq	1.27E-03	1.58E-01	7.23E-02	1.33E-05	2.32E-01
Ionizing radiation	kBq Co-60 eq	2.80E-04	1.70E-02	2.32E-02	1.93E-04	4.07E-02
Fossil resource scarcity	kg oil eq	5.94E-04	4.02E-02	1.94E-02	1.54E-05	6.01E-02
Fine particulate matter formation	kg PM2.5 eq	3.66E-06	3.00E-04	2.15E-04	-2.05E-07	5.18E-04
Ozone formation, Human health	kg NO <sub>x</sub> eq	3.67E-06	3.20E-04	5.62E-04	-4.73E-06	8.81E-04
Mineral resource scarcity	kg Cu eq	5.27E-06	1.80E-04	1.86E-02	-1.44E-06	1.88E-02
Human carcinogenic toxicity	kg 1,4-DCB	5.49E-05	7.13E-03	1.44E-02	-2.92E-06	2.16E-02
Terrestrial acidification	kg SO <sub>2</sub> eq	5.39E-06	4.80E-04	4.75E-04	-3.31E-07	9.60E-04
Terrestrial ecotoxicity	kg 1,4-DCB	3.94E-03	1.39E-01	4.78E-01	-1.68E-04	6.21E-01
Ozone formation, Terrestrial ecosystems	kg NO <sub>x</sub> eq	3.75E-06	3.20E-04	5.70E-04	-7.53E-06	8.86E-04
Water consumption	m <sup>3</sup>	2.71E-05	1.86E-03	7.18E-04	2.19E-06	2.61E-03

Table 4. Impact assessment values of SDT

Impact category	Unit	Paper substrate	Chip fabrication & bonding	Printing antenna & PS	Battery fabrication & bonding	End-of-life SDT	Total
Land use	m <sup>2</sup> a crop eq	1.2E-03	1.4E-03	5.0E-03	1.4E-04	-2.5E-06	7.7E-03
Marine eutrophication	kg N eq	6.8E-07	1.2E-05	4.1E-06	3.4E-07	4.0E-08	1.7E-05
Marine ecotoxicity	kg 1,4-DCB	1.9E-04	5.6E-03	1.8E-01	1.4E-02	2.4E-04	2.0E-01
Freshwater eutrophication	kg P eq	1.1E-06	9.4E-05	1.9E-04	1.4E-05	-5.2E-08	2.9E-04
Stratospheric ozone depletion	kg CFC11 eq	2.3E-09	9.5E-08	8.1E-08	4.3E-09	1.0E-10	1.8E-07
Freshwater ecotoxicity	kg 1,4-DCB	1.5E-04	4.3E-03	1.1E-01	1.1E-02	3.8E-04	1.3E-01
Human non-carcinogenic toxicity	kg 1,4-DCB	2.0E-03	7.5E-02	1.7E+00	1.1E-01	1.3E+00	3.3E+00
Global warming	kg CO <sup>2</sup> eq	3.2E-03	1.6E-01	6.8E-02	5.2E-03	1.9E-05	2.3E-01
Ionizing radiation	kBq Co-60 eq	7.0E-04	1.7E-02	3.0E-02	9.1E-04	2.2E-04	4.9E-02
Fossil resource scarcity	kg oil eq	1.5E-03	4.0E-02	1.8E-02	1.5E-03	1.9E-05	6.1E-02
Fine particulate matter formation	kg PM2.5 eq	9.1E-06	3.0E-04	2.0E-04	3.5E-05	-2.3E-07	5.4E-04
Ozone formation, Human health	kg NO <sub>x</sub> eq	9.2E-06	3.2E-04	5.2E-04	2.2E-05	-5.5E-06	8.6E-04
Mineral resource scarcity	kg Cu eq	1.3E-05	1.8E-04	1.7E-02	3.2E-04	-1.7E-06	1.8E-02
Human carcinogenic toxicity	kg 1,4-DCB	1.4E-04	7.1E-03	1.9E-02	9.6E-04	-3.3E-06	2.7E-02
Terrestrial acidification	kg SO <sub>2</sub> eq	1.3E-05	4.8E-04	4.4E-04	9.5E-05	-3.6E-07	1.0E-03
Terrestrial ecotoxicity	kg 1,4-DCB	9.8E-03	1.4E-01	4.4E-01	5.7E-01	-1.9E-04	1.2E+00
Ozone formation, Terrestrial ecosystems	kg NO <sub>x</sub> eq	9.4E-06	3.2E-04	5.2E-04	2.3E-05	-8.7E-06	8.7E-04

<b>Water consumption</b>	m <sup>3</sup>	6.8E-05	1.9E-03	7.5E-04	7.6E-05	2.5E-06	2.8E-03
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It can be observed that the impacts are relatively high for the categories marine and terrestrial ecotoxicity, and human non-carcinogenic toxicity. Impact on these categories is mostly associated with the use of silver ink for the antenna, as well as disposal of silver (manifested as impacts in the end-of-life stage). In terms of their global warming potential, both devices embody around 0.23 kg CO<sub>2</sub>-equivalents (eq). These values for ECD are lower than those reported for LCD display of 0.886 kg CO<sub>2</sub>-eq, and 0.95 kg CO<sub>2</sub>-eq for LCD reported in the ecoinvent <sup>46</sup>. However, LCD displays can be used for functionally more demanding applications, such as televisions, computer monitors, instruments panels whereas the use of ECD would be preferred for ambient non-light-emitting displays thanks to its flexibility, energy-efficiency, easy integration and cost-effectiveness <sup>9</sup>. However, in applications where two types of displays could be used interchangeably, the ECD would represent a less carbon-intensive alternative.

### **Contribution analysis: ACL and ECD**

The relative impacts contribution of each component or process stage in the life cycle of ACL is shown in Figure 4 and contribution of each component from printing of antenna and ECD in Figure 5. The main impacts in the life cycle of ACL are created during the printing of antenna and ECD, and fabrication and bonding of chip. The use of paper substrate is comparatively very small and visibly manifests only for the category of land use. End-of-life impacts of ACL are negligible except on the category human non-carcinogenic toxicity that reflects on impacts of silver to the soil from the paper sludge disposal.

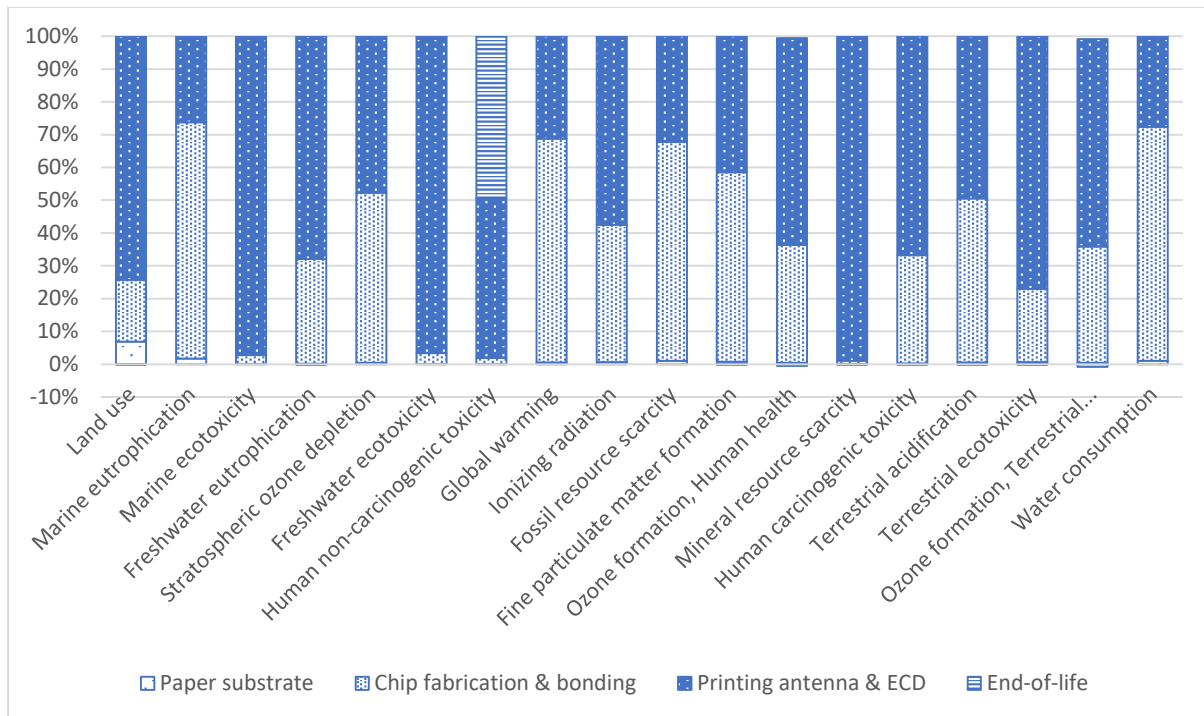


Figure 4. Contribution analysis: ACL

Further breakdown of impacts contribution of ECD (Figure 5) shows that the majority of the impacts are generated from the use of silver ink for printing of antenna, antenna tracks and ECD electrode. Impacts of silver ink are directly associated with the use of silver metal as a raw material in ink formulation (95-99 % of impacts from formulation of ink). Solid waste from printing occupies small share across most of the categories and has significant impact on the category of human carcinogenic toxicity. Impacts of direct process energy use are very low which is seen as one of the advantages of printed electronics in comparison to more conventional. Although, some processes are purposely avoided in the current design such as the use of sputtering that has high embodied energy and low material efficiency, and is commonly used deposition process in manufacturing of new flexible electronics<sup>47</sup>. For example, sputtering under vacuum is used for deposition of ITO in manufacturing of electrochromic windows and photovoltaics<sup>24</sup>. However, given large embodied impacts of silver as an alternative choice of electrode, it is not clear if trade-offs are justified from environmental perspective. Use of electricity for printing has only large impacts on the category of ionizing radiation due to electricity mix in France dominated by large share of nuclear energy<sup>48</sup>.



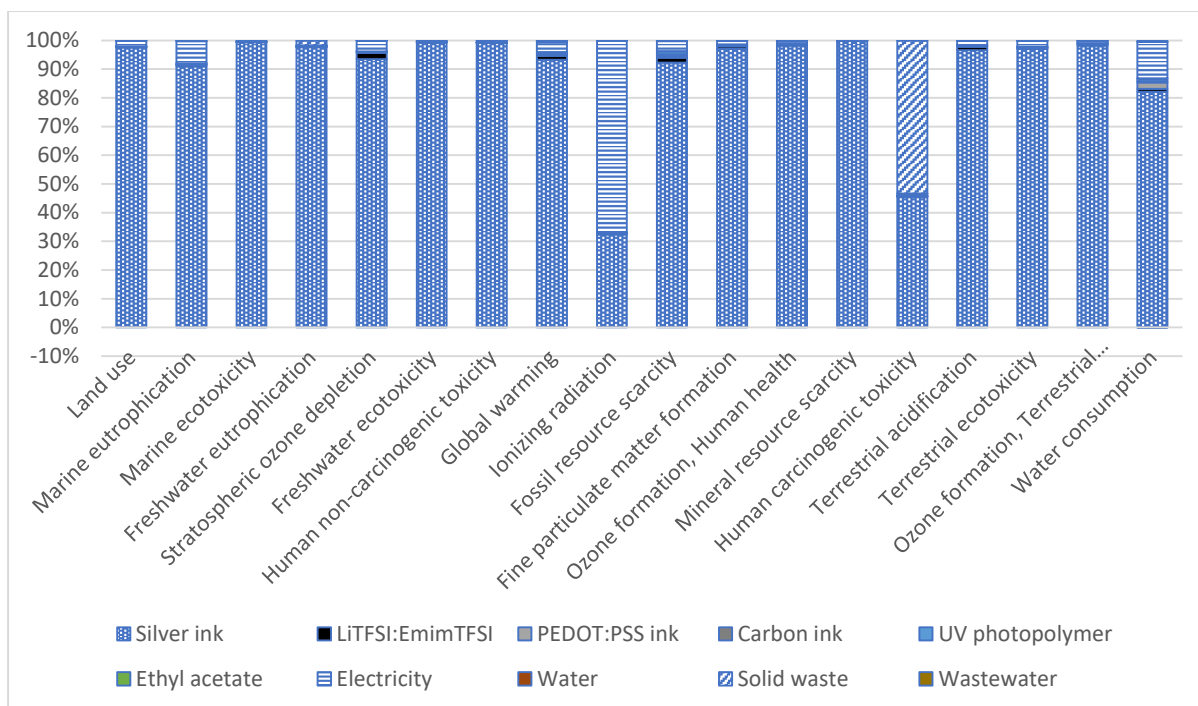


Figure 5. Contribution analysis: printing antenna and ECD

These findings are consistent with some observations noted in previous works on electrochromic and piezoelectric devices, although it is generally hard to draw these parallels given the vast range of possible architectures and applicable materials. Generally, it appears that the impacts of electrochromic materials are consistently low in comparison with other components in the architecture of electrochromic devices, while the impacts of substrate may vary. For example, in the study by Syrrakou et al. <sup>24</sup> and Papaefthimiou et al. <sup>49</sup>, the substrate and electrode based on fluorine-doped tin oxide constitute 56.5 % of total embodied energy while 24.5 % share of embodied energy was used in the manufacture of a propylene carbonate electrolyte <sup>24,49</sup>. Similarly, in Posset and Harsch <sup>23</sup>, a PET substrate contributes to 43 % of total energy consumption, followed by ITO electrode (33 %), the fabrication process (10 %) with electrolyte (4 %), and electrochromic materials (2 %) having minor contributions <sup>23</sup>. In our study, impacts of electrolyte are not significant (>1 % in all the categories), the impacts of the electrode (for ACL) are also prominent (around 20 % of impacts related to silver ink), while the impacts of a substrate in our study appear to be lower.

### Contribution analysis: SDT and PS

The relative impact contribution for SDT and integrated PS are represented in Figure 6 and Figure 7. Given structural similarities, the impact contribution of different components and process stages are similar to those observed for ACL. The most impactful stage is printing of the antenna and PS prompted by the use of silver for antenna and antenna tracks. Fabrication and bonding of the chip is the next most significant contributor. Impacts of battery fabrication and bonding are notable on the category of

terrestrial ecotoxicity. Analogous to ACL, the impacts of substrate and end-of-life stage are high on the categories of land use and human non-carcinogenic toxicity, respectively.

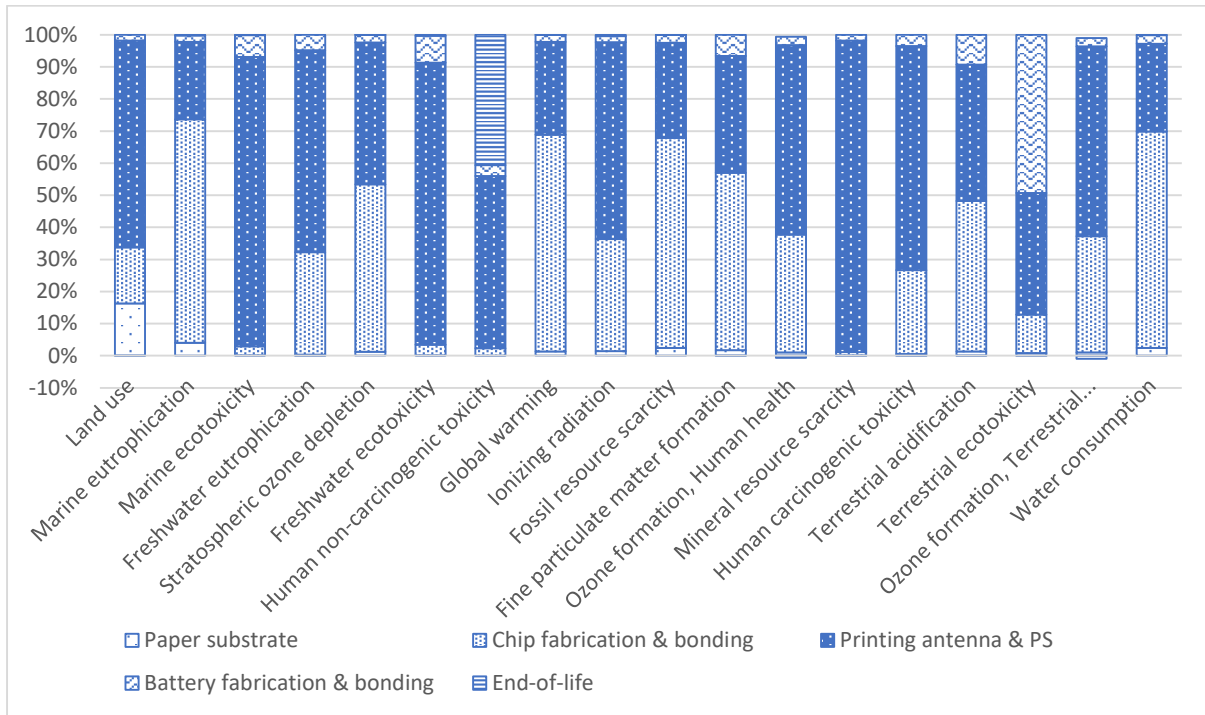


Figure 6. Contribution analysis: SDT

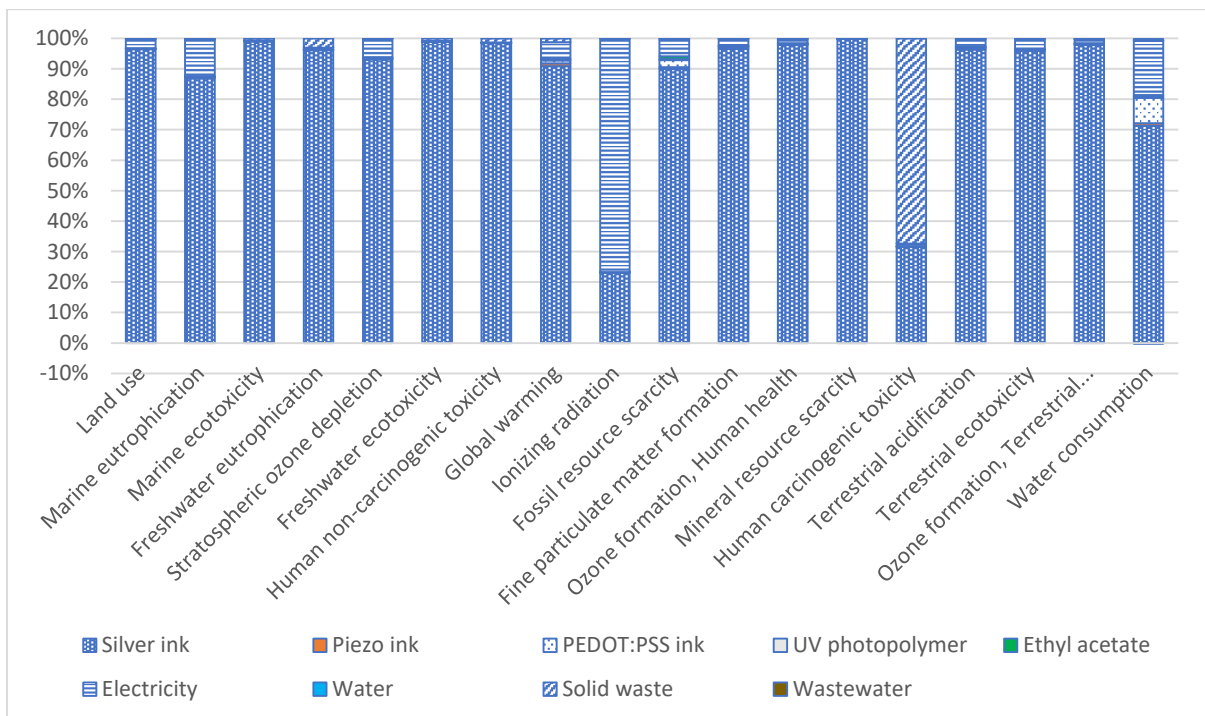


Figure 7. Contribution analysis: printing antenna and PS

## Interpretation

### Reducing the impacts of RFID antenna using flexography and nano-copper ink

Given the high impacts of silver ink used in the antenna, antenna tracks and bottom electrode, two strategies involving reduction and substitution of silver were compared with default ACL: flexography printing using nano-particle silver ink, and screen-printing involving nano-particle copper ink, Figure 8. The results are shown for ACL with similar results expected for SDT. Both scenarios result in a significant reduction of environmental impacts across all categories. Flexography printing of silver has a positive influence on environmental impacts by reducing requirements of silver ink, and due to reduction of silver impacts in the end-of-life stage. Hence, the most significant reductions are observed for toxicity-related categories and mineral resource scarcity. The substitution of micro-particle silver ink with nano-particle copper ink promotes even larger reductions in these categories. In the absolute terms, combined toxicity impacts involving ecotoxicity categories (terrestrial, marine, and freshwater) and human toxicity (carcinogenic and non-carcinogenic), of default scenario results in 4.89 kg of 1.4 DCB-eq (dichlorobenzene equivalents) that is reduced to 1.77 kg of 1.4 DCB-eq for flexography scenario (64 %) and to 0.77 kg of 1.4 DCB-eq for copper ink scenario (85 %). Both scenarios have a lesser influence on impacts of CO<sub>2</sub>-eq that induces small reduction (from 0.23 kg CO<sub>2</sub>-eq to 0.19 and 0.17 kg CO<sub>2</sub>-eq, respectively).

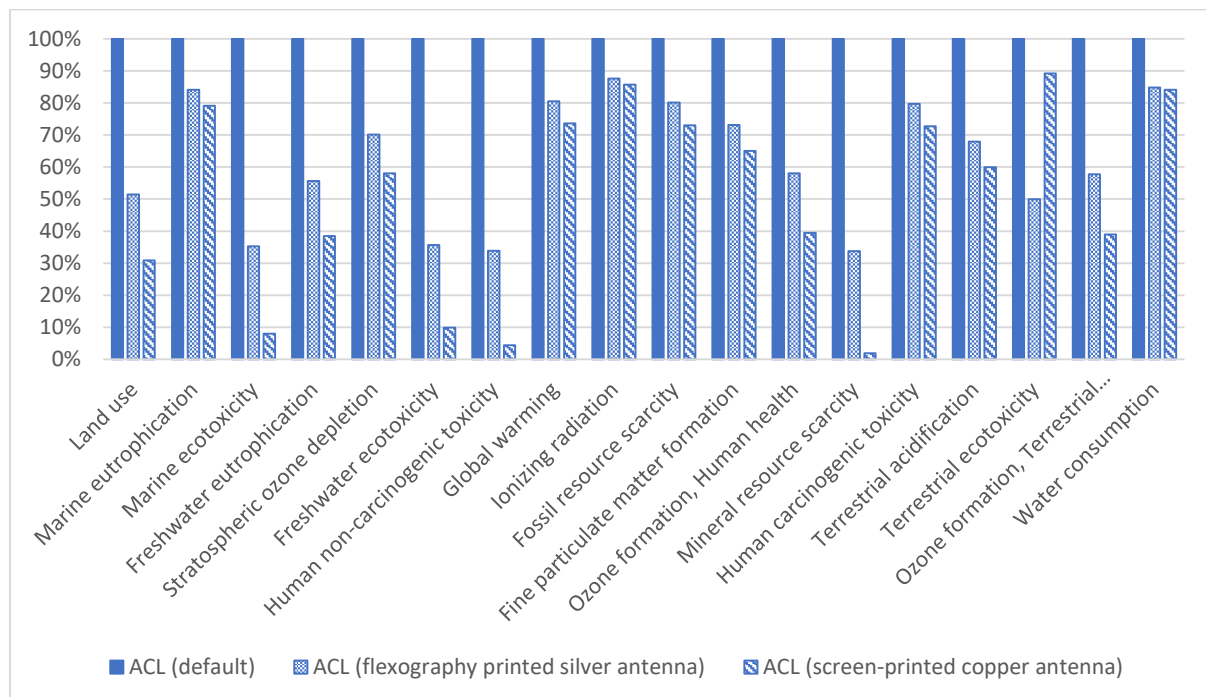


Figure 8. Comparison of screen-printed ACL with ACL printed combining flexography printing of antenna and screen-printing of ECD

**Influence of substrate on life cycle impacts**

Use of paper as a substrate is seen as advantageous from the perspective of circular economy and potentially more environmentally viable option to more commonly used plastic substrates. The paper is more readily recycled than plastics<sup>28,29</sup>, and the use of plastics in single-use products is seen as undesirable and faces uncertain future in Europe. Comparison is carried out between default ACL device and ACL based on PET substrate fully landfilled at the end-of-life. A comparison is also carried out between substrates only while excluding other device components (i.e., printed inks, chip, and battery), Figure 10.

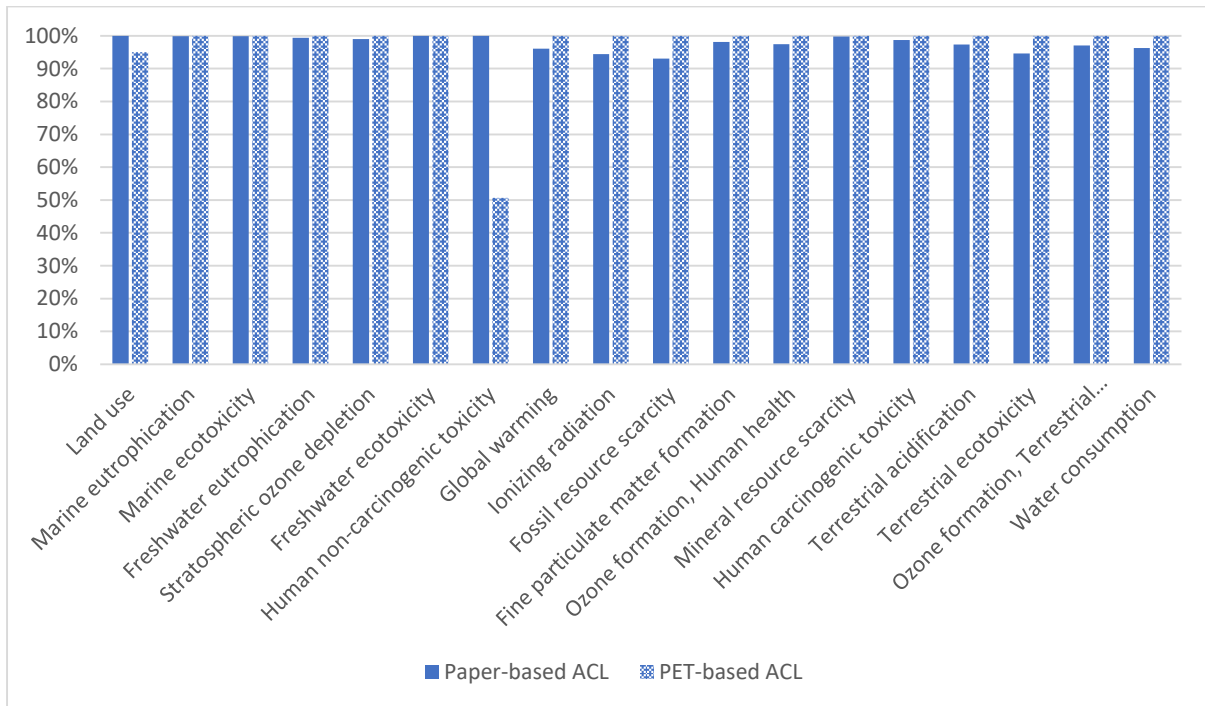


Figure 9. Comparison between ACL based on paper substrate and ACL based on PET substrate

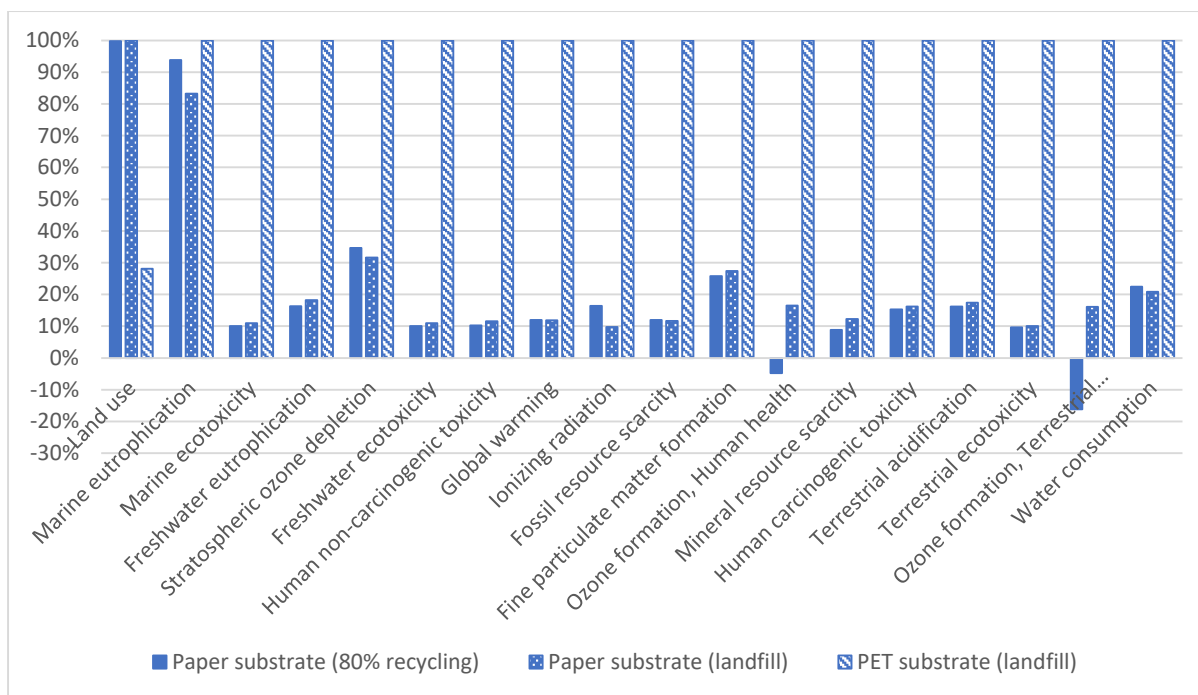


Figure 10. Comparison between cradle-to-grave scenarios for paper (Powercoat XD125) and PET substrate

The results show that paper has 80-90 % lower impacts in comparison to PET, for most categories while recycling has shown to further reduces the impacts of the paper. However, impact of substrate on overall burdens in the life cycle of ACL remain low for both materials (below 5%) with exception of the category human non-carcinogenic toxicity where landfilling scenario for PET-based device is preferred as it prevents emission of silver at the end-of-life.

Furthermore, while the current results clearly favour the paper substrate, any definite conclusions should bear in mind differences between the substrates in weight, investigated scope of end-of-life scenarios, and influence of their physical characteristics on durability and use of other materials in fabrication of the devices. The present comparison assumes a higher material requirements for the PET substrate (3.01 g in comparison with 0.736 g for paper), consistent with assumed substrate thickness of 125  $\mu\text{m}$ . Providing for the higher mass and calorific value, PET substrate would result in higher energy recovery if devices are incinerated, and have advantages over durability<sup>50</sup>. On the other hand, the PET substrate may require specific chemical or physical pre-treatment to achieve a good ink wettability prior to printing.

### Opportunities for eco-efficiency improvements and evaluation

The study has shown that both flexography printing and the use of copper ink constitute viable strategies for significant impact reduction and incentive to re-design and optimize the process in that direction, while the substitution of silver seems an altogether best strategy. Similarly, impact mitigation solutions

and opportunities could also focus on substitution of the NFC chip, as the second largest contributor in the life cycle of the devices. For example, the impacts of NFC chip for ACL could be mitigated by considering rectifiers based on zinc-tin-oxide instead of the silicon chip. The use of the battery and chip is necessary for SDT in order to keep track of a strain during the whole transportation, while the use of a chip for ACL has a simpler purpose that can be achieved using the rectifier. A printed rectifier would also enable a fully printed ACL without any bonding of components. Further impact mitigation could also be achieved by optimizing the design of the devices with their prospective end-of-life processes to minimize the impacts on the environment and maximize the recovery of components. For example, ongoing experiments on the recycling of devices in the paper recycling facility have shown that the choice of silver ink product could influence the silver uptake in the screening stage. Such evidence should be used to drive the selection of printing inks and other components.

To further broaden the understanding of the environmental performance of ACL and SDT, analysis can be extended in view of limitations of the current model, and by extending the scope to see how two devices fare in comparison with current technologies and given practical context of their use. Devices could be compared with conventional printed circuit boards, and micro and printable electronics<sup>51-53</sup>. However, any comparative assertions need to ensure an appropriate functional equivalency is achieved, which is not so straightforward for these relatively novel and unique technologies. For example, anti-counterfeiting functionality provided by ACL is more sophisticated in comparison to the printed bar codes, graphic labels, or Radio Frequency Identifier<sup>54,55</sup>. In comparison, printed ECD offers high-level security thanks to overt features such as the specific visual outlook, cost and complexity to reproduce<sup>56,57</sup>, consistent with the recommendation of European project BRIDGE<sup>7</sup>.

Furthermore, the environmental performance of these devices can be seen through indirect consequences of their role in preventing damages, product mishandling and counterfeiting while improving and regulating supply chains. For example, the anti-counterfeiting technology could reduce the usage of sub-standard, ineffective and harmful counterfeit medicines with potentially adverse effects on human health<sup>6</sup>, while better detection of damage can help optimize transportation routes and manners of handling resulting in less redundant and lost products. Therefore, the use of these technologies and their role on the products and the supply chains would result in indirect impacts and/or benefits that could be further analysed<sup>58</sup>.

The main limitations of the current study relate to the present end-of-life model that involves several uncertainties and simplifications. Uncertainties are associated with current end-of-life scenarios involving assumptions on landfilling and recycling rates. While we assume high recycling rates of paper in Europe, the estimated disposal rates may differ in practice depending on the location of product use. The simplifications correspond to the genericecoinvent waste inventories presently used to represent impacts of non-paper fractions such as the battery, chip, printed polymers and metals in the landfill.

The impact of these components could be better simulated by generating custom inventories using specialized modelling tools <sup>59</sup>. More precise modelling of the end-of-life stage should also explore potential toxicity impacts associated with materials particle size. Currently, impacts of copper and silver via paper sludge disposal are modelled based on their general reactivity and toxicity in nature, irrespective of particle size. However, a more thorough understanding of these aspects needs to overcome the limitations of current assessment methods to model new materials <sup>60</sup>.

## **Conclusion**

This study represents a first insight into the environmental impacts of two new printed electronic devices based on electrochromic and piezoelectric materials used for anti-counterfeiting and shock detection. The development of elaborate life cycle inventories and subsequent impact analysis allowed to observe that impacts of both devices are dominated by the production and end-of-life of RFID antenna associated with the use of silver, and production of NFC silicon chip, while the impacts of other components and process flows (i.e., waste disposal, Li-ion battery, sensor/display, and direct energy use) are comparatively small on most categories. Observations to these initial findings led to the development of several scenarios for reduction of impacts of RFID antenna involving flexography printing technique and substituting the silver ink with a copper-based nano-particles ink. Development and investigation of these scenarios involved dedicated experimental work to reach a proof-of-concept and establish preliminary data necessary for modelling. Both scenarios result in a significant reduction of impacts particularly on toxicity-related categories and could play a significant role in the reduction of environmental impacts of new devices. We have also shown that the choice of the substrate has a small influence on the overall impacts of devices and that the paper substrate appears preferable in comparison with PET. Insights into the environmental performance of new devices can be extended with consideration to the current modeling limitations, and in reference to comparative technologies for anti-counterfeiting and shock-detection given that functional equivalency is appropriately established. New inventories and observations generated throughout this study are hoped to serve as a reference point for the future development of piezo sensors and electrochromic displays.

## **Supporting information**

Tables detailing life cycle inventories of components and complementary impact assessment results.

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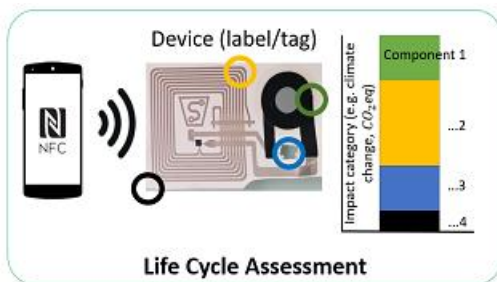
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Life cycle assessment reveals environmental impacts and most impactful stages in the life cycle of printed electronics for anti-counterfeiting and shock-detection.