



Plastic (PET) vs bioplastic (PLA) or refillable aluminium bottles – What is the most sustainable choice for drinking water? A life-cycle (LCA) analysis

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ABSTRACT

Nowadays, the most important tool to evaluate the environmental impact of both petro-plastics and bioplastics is the life cycle analysis (LCA). LCA determines the overall impact on the environment by defining, calculation and analyzing all the input and output directly related to production, utilization, and disposal of a product or a process. In this work, a LCA (cradle to grave) of bottles for drinking water was developed on three scenarios: polyethylene terephthalate (PET) bottles, as conventional packaging material for outdoor drinking water, polylactic acid (PLA) bottles, as alternative and innovative biodegradable packaging and aluminum bottle, as reusable and almost infinitely refilling packaging. As a result of LCA, ten impacts categories have been accounted for, among which the global warming potential (GWP, measured as kgCO₂ eq), the eutrophication potential (EP, measured as kgPO₄ eq.), human and eco-toxicity (HTP and ETP, measured as kg 1,4-DB eq.). The average drinking water consumption in Italy has been estimated in 1.5 L per day, corresponding to three 500 ml-plastic bottles and 1 refillable aluminum bottle. LCA has been firstly applied to a single bottle production and use, then to the daily and annual bottles consumption. PET bottles production and use assure the lower environmental impacts compared to PLA bottles, burdened by agricultural phase for corn cultivation, and to aluminum bottles, when the every-day washing with hot water or water and soap is comprehended. Moreover, including the end-of-life options into the analysis, PET recycling permits to reduce up to about 30% the GWP, whereas PLA composting does not lead to any GWP savings. In this study, aluminum bottle has been considered reusable for 2.5 years. The microbiological quality of water in one-way PET and PLA bottles has been compared with the refillable bottle rinsing with hot water and soap and only hot water, highlighting that the level of contamination is alarmingly increased in the latter case.

1. Introduction

To date, the weight of plastic items of any shape and size both in the daily life of people and at industrial level, is enormous (Rigamonti et al., 2014). Playing a key role in almost all sectors such as, for example, food storage, fashion and clothing industry, construction industry, pharmaceuticals and personal care products and many other applications, plastic has been rapidly become the third widespread material worldwide after cement and steel (Geyer et al., 2017). Recent data show a trend towards an increase in consumption and, therefore, wasting of plastics, i.e. in 2019 global plastic production reached 338 million tons, showing an increase of about 640% compared to 1975 (Matthews et al., 2021).

Lightness, malleability and flexibility are only few properties that

make plastic indispensable, along with the resistance against microbial attack or any other type of natural degradation (Katiyar et al., 2014). As a consequence of the linear economy concepts of “single-use” or “disposable” progressively diffused worldwide since the ‘80s, the biggest side-effect of its large use has been the huge amount of plastic waste released in the environment, exacerbated by globalization of supply chains, as well as by the growing importance of worldwide-level retailers (Schneiderman and Hillmyer, 2017). This has rapidly conducted the planet towards a condition of overall unsustainability, thought the degradation of ecosystems and danger for the survival of many animal and plant species, because of the persistence in environment of plastic materials (Comăniță et al., 2016; Wesch et al., 2016).

A promising alternative to petro-plastics are bioplastic, even called bio-based plastics or biopolymers, obtained from biomass such as corn

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and sugar cane, as part of the biorefinery concept (Shogren et al., 2019). In few years, they have been gaining a growing interest, because under suitable conditions they can be totally biodegradable to CO₂ in a matter of months and can contribute to carbon capture and storage (Lambert and Wagner, 2017). Bioplastics have characteristics similar to petro-plastics, but being biodegradable, they must not be disposed of or recycled but composted, closing the natural carbon cycle (Lamberti et al., 2020). These materials, such as polylactic acid (PLA) or bio-polyethylene, can be obtained directly from natural material, i.e. cellulose, starch or sucrose by fermentation and chemical synthesis starting from renewable biological monomers (Dedenaro et al., 2016), or directly produced by bacterial cultures, as polyhydroxyalkanoates and polyhydroxybutyrate (PHA and PHB) (Ciriminna and Pagliaro, 2020). However, global bioplastic production was still estimated at about less than 1% of the total plastic production, principally because the cost of research and development still makes up for a share of investment and has a great impact on material and product prices (Spierling et al., 2018; Zhao et al., 2020). In addition, it cannot be neglected that most bioplastics are currently produced from agricultural crop-based feedstocks (carbohydrates and plant materials), being not yet ideally aligned with the UN's sustainable development goals (SDGs), due to their competition for arable land, fresh water, and food production (Karan et al., 2019).

The ever-increasing problem of waste accumulation, along with the continuous exploitation of virgin resources, has recently promoted the transition to the circular economy, designed in such a way that all the flows of materials and waste are reintegrated into a cycle in order to be exploited again at the end of their life, mitigating the damages caused to natural capital depletion and improving its capacity to regenerate ecosystem services (Bucknall, 2020). The circular economy paradigm is based on the concept of life cycle thinking (LCT), as an approach where the whole life cycle of a product or a process is assessed from the moment of extraction of raw materials to the end of its life, to determine and quantify its environmental, costs, social and cultural impacts (Lazarevic et al., 2012). Among the tools of LCT, the life cycle assessment (LCA) is the most used quantitative methodology that supports the evaluations of the environmental impacts of a product, a process or a service, considering all stages of its life cycle, from cradle-to-grave (Petti et al., 2018). From a general point of view, the LCA allows to evaluate the interactions that a product or service has with the environment, considering its whole life cycle that includes the preproduction points (extraction and production of raw materials), production, distribution, use (including re-use and maintenance), recycling, and final disposal (Wolfson et al., 2019). LCA has emerged as a powerful method that can take into account the products' energy and resources consumed, as well as the generation of emissions and wastes (Tufvesson et al., 2013), and by now has been applied in a wide range of scientific research and industrial areas (Jacquemin et al., 2012; Curran, 2013; Tamburini et al., 2015; Lévassieur et al., 2016). LCA has been also highlighted by the Directive 94/62/EC as a tool able "to justify a clear hierarchy between reusable, recyclable and recoverable packaging and to legitimate environmental systems analysis for waste policy and strategy". Since the introduction of LCT into European policies, specifically in relation to the waste hierarchy, several LCA studies have been published on waste management systems (Bernstad & La Cour Jansen, 2012; Laurent et al., 2014a, 2014b; Khandelwal et al., 2019), including post-consumer plastic waste management (Toniole et al., 2013; Lazarevic et al., 2010; Hahladakis and Iacovidou, 2019), together with a wide literature on LCA of petro-plastics (Sanani and van der Meer, 2020; Dassisi et al., 2016) and bio-plastics (Kakadellis and Harris, 2020; Morão and de Bie, 2019) production. The majority of these studies concluded that recycling is usually the environmentally favorite treatment option for a single-polymer petroplastic material compared to incineration or landfilling, because in part it reduces the need of virgin materials, even though only 20% of used plastic has been now adequately recycled at EU level and 9% worldwide (d'Ambrières, 2019).

In the hierarchy of the circular economy, a priority of product reuse with respect to material recycling is recognized, as more value is retained (Milios, 2018). Reusing for as long as possible, from one side reduces the need for primary resources, from the other side diminishes the environmental impact of materials use, if the cycles are closed in sustainable ways. Refillable/reusable packaging systems have been successfully utilized for several decades, namely the glass bottles, due to the high turnover rates and the relative short transporting distances (Coelho et al., 2020).

One of sector where these new trends of packaging has influenced the consumers' preference is the bottled drinking water, especially in developed countries. Packaged drinking water has been taken as safe means of drinking water provision (Hawkins, 2017). Global packaged drinking water industry is estimated to be the most dynamic sector of all the food and beverage industry, and bottled water consumption is estimated to have reached nearly 100 billion gallons in 2017 (Tilahun and Beshaw, 2020). Bottled water is drinking water packaged generally in plastic bottles and regulated by national and local agencies (Pacheco-Vega, 2019). Among the common plastics used, polyethylene terephthalate (PET) is the most favorable packaging material for drinking water due to its chemical and physical stability (Gomes et al., 2019). It is forecasted that in 2021, more than 580 billion of PET bottles will be produced and consumed worldwide, representing more than 80% of total water packaging (Gu et al., 2020). Bioplastic bottles, usually made of PLA, still hold a tiny fraction of the total plastic market, including drinking water bottles, sharing less than 3%, principally due to their higher costs, but estimate growing in the coming years due to the desire and need to find non-petroleum-based polymers (Nampoothiri et al., 2010). In alternative, the choice of a more environmentally friendly package for drinking water has recently promoted the wide adoption of practices such as recycling and reuse. The return to reusable bottles, made of glass at home and of stainless steel or aluminum for the every-day outdoor activities, has been considered beneficial to the environment, and it has been becoming a new especially for young people, and the main argument is that these packages are refillable, allowing reuse by dozens of times (Piccirillo-Stosser, 2018).

The aim of this paper is to calculate and compare the environmental sustainability of production of one PET bottle vs. PLA bottle or aluminum refillable bottle for drinking water, and of use in a hypothetical one-year of time-frame. Based on the LCA approach, the environmental impacts of production from raw materials and the annual consumption of bottles or refilling in the case of using PET, PLA or aluminum bottles have been estimated, also expanding the boundaries to the end-of-life options for plastic bottles (open and closed loop recycling for PET and composting for PLA, in comparison to incineration and landfill) and to the impact of every-day washing with tap water and soap for aluminum bottles. The microbiological quality of water has been also evaluated for different scenarios of use.

2. Materials and methods

2.1. The case study

The average drinking water consumption in Italy has been estimated in about 1.5 L per day per adult (Nissensohn et al., 2017), with an average of 6 occasions of drinking spread throughout the day (Barraj et al., 2009). This can correspond to the use of three plastic bottle of 500 ml of capacity per day or two subsequent refilling of an aluminium bottle of 750 ml of capacity. Taking into account that the major part of PET bottles consumed are for one-way use and under the hypothesis of the same behaviour every day for a period of time of one year, we calculated the use of 1095 PET bottles or 1095 PLA bottles, wasted after use, and only one aluminium bottle, refilled twice a day. For the aluminium bottle, two every-day washing scenarios has been accounted for, with about 3 L of tap water and soap. In a circular economy perspective, there are several end-of-life options for PET bottles: thrown as unsorted waste

100% destined to incineration or, as worst case, to landfill, open-loop recycling obtaining a non-bottle grade quality PET that must be destined to other uses (i.e., fibres) and closed-loop recycling where PET is reprocessed at high temperature in order to increase its intrinsic viscosity to levels compatible with bottles forming process and to remove any possible residual organic contamination deriving from their previous use (Nessi et al., 2012). For the Italian law, the possibility to use up to 50% recycled PET for the manufacturing of mineral water bottles has been recently allowed (Intini and Kühtz, 2011).

PLA bottles could be incinerated as unsorted waste or, as biodegradable material, composted together with organic waste and end its life as organic fertilizer, the so-called compost, in soil (De Andrade et al., 2016). Aluminium can be 100% recycled, simply re-melting the metal and manufacturing it back into aluminium items (Stotz et al., 2017).

The LCA methodology was applied to this case study, according to the principles and the requirements provided, respectively, by the ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b) standards. According to them, LCA is composed of four major stages: goal and scope definition, inventory analysis, impact assessment, and interpretation, which are summarized in following sections.

2.2. Goal and scope definition

The aim of this work is to examine the environmental impact of production, use and end-of-life of PET and PLA bottles in comparison to aluminium bottle, in a time-frame of 1 year. According to the Joint Research Center of the European Commission (Nessi et al., 2018), we defined as functional unit (FU) the quantified performance of a product system that describes the function of the product, and as reference flow the amount of product needed in order to fulfil the defined FU. In this study, the FU that fully covers the function of containing beverage for consumption is “one year of use” corresponding to a reference flow of 1095 bottles of PET or PLA, and 0.4 refillable aluminum bottle, assuming an average lifetime of aluminum bottle of 2.5 years (930 days). The study is a “cradle-to-grave” LCA, i.e., it mainly covers all relevant process steps from raw material production to the final waste treatment or recycling. The general simplified processes flow diagrams

for the production of the three bottles are given in Fig. 1, including an indication of the system boundaries for the systems studied. As it can be seen from the flow charts, the system boundaries include:

- For PET, the polymer production starting from crude oil extraction;
- For PLA, the crop farming
- For aluminum, the metal extraction
- For PET and PLA, the production of polymer resins and the blow moulding for plastic bottles forming, as well as the punch pressing and finishing for aluminum bottle;
- For all three, the transportation of raw materials to production plants, by taking into account the tons carried per kilometer ($t \cdot km$);

Excluded from the system boundaries are retail of the bottles, production and transport of secondary and tertiary packaging, production and disposal of the infrastructure (machines, transport media, roads, etc.) and their maintenance. For aluminum, any end-of-life scenario has been accounted for, because of its lifespan of several decades, excluding breaking events (Thiel et al., 2013).

2.3. Life cycle inventory analysis (LCIA)

Data for the LCA analysis carried out in the present study are collected from scientific literature and industrial reports, mediated to be as much representative as possible of the processes described, or directly taken from the Ecoinvent® data base process (Ecoinvent Database,).

The production process of PET starts with the extraction of natural gas and oil, in order to obtain the monomers, i.e., monoethylene glycol (MEG) and purified terephthalic acid (PTA). The monomer synthesis stage required an oxidation reaction of paraxylene with acetic acid in the presence of catalysts and chemical additives to obtain PTA as a product (Chen et al., 2016; Gomes et al., 2019). In the case under study, the specific monomers synthesis reaction is also included in the Ecoinvent® database and was therefore inserted in the process according to the production of 1 PET or PLA bottle and 1 refillable aluminium bottle.

The synthesis of MEG starts from the production of the precursor ethylene oxide, following a hydrolysis reaction. All the data used were

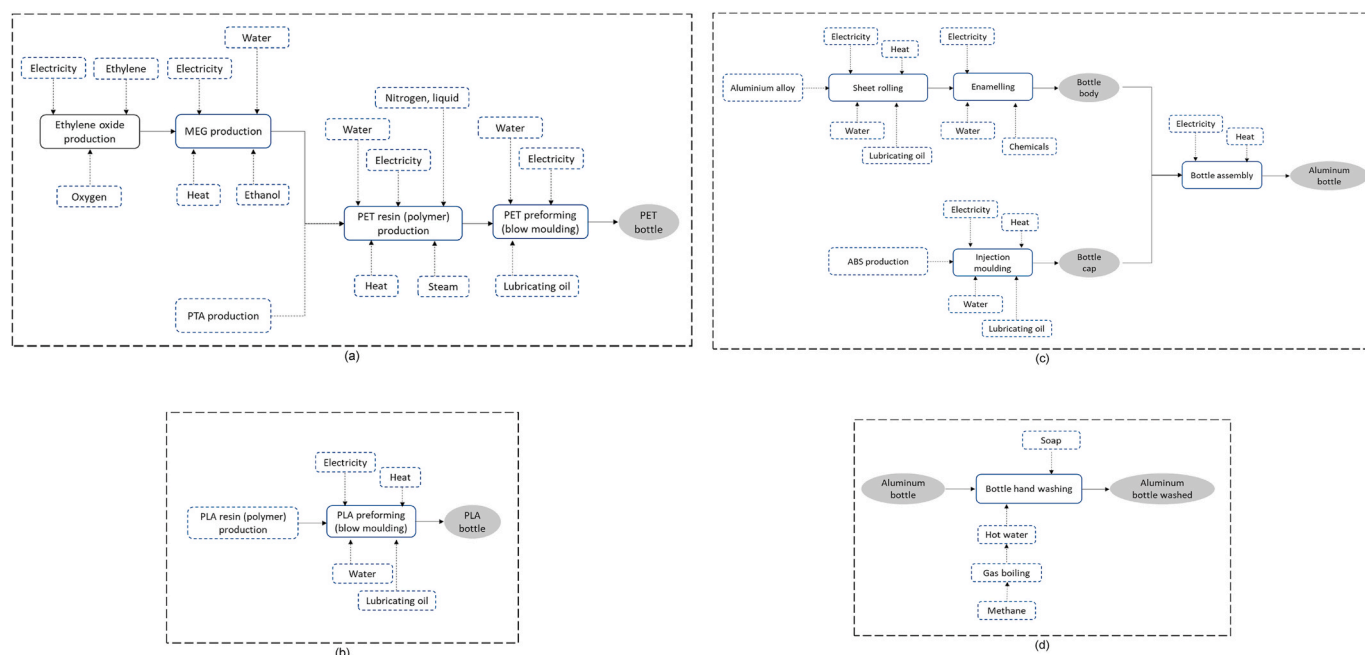


Fig. 1. System boundaries used in the life cycle assessment (LCA) of the production of PET bottles (a), PLA bottles (b), aluminum bottles (c) and washing (d) The boxes represent processes (solid box = foreground process; dashed box = background process) and grey circles represent products. (MEG = monoethylene glycol; PTA = purified terephthalic acid; PET = polyethylene terephthalate; ABS = acrylonitrile butadiene styrene).

averaged and referred to a 500 ml bottle with a weight of 19.10 g (Papong et al., 2014; Baidowska-Witos et al., 2020). Copolymerization was carried out to obtain PET granules, then subjected to the stretch blow moulding process (Benavides et al., 2018). The stretch blow moulding is a possible variant of the blow moulding process used for the tanks and containers production, where the polymer in granules is heated to the glass transition temperature, inflated and subsequently stretched in the upper part to generate the neck of the bottle (Giridharreddy and Tech, 2013). The stretch blow moulding data were obtained from the Ecoinvent® database and adapted to the FU. Material losses in an order of magnitude of about 10% between monomer production and polymerization process, as in the case of PTA, has been considered as negligible. All input materials have been reported in Table 1. Impacts have been calculated for 1 bottle and for 3 bottled, according to the daily average consumption of 1.5 L of drinking water in Italy. In addition, in order to consider the yearly use of PET bottles, 1 FU (1095 bottles) have been considered for the final impact calculations.

Regarding PLA production, a series of chemical products such as herbicides, pesticides and fertilizers listed in the Ecoinvent® database have been used for the cultivation of corn process. As foreground process, it incorporates the agricultural phase from seedling to harvesting, including the use of fertilizers, water and pesticides, corn drying, including the loss of water was taken into account to calculate the amount of dry cereal necessary for the production of a single bottle, glucose production, and fermentation to lactic acid, that is, the true precursor of PLA polymer. The PLA granules were subjected to the stretch blow moulding process (Papong et al., 2014). Considering 1 bottle, a quantity of PLA in granules of 11.28 g was necessary, that is the exact weight of 1 PLA bottle with a volume of 500 ml. Material losses in polymerization process was considered as negligible. All input materials necessary for the production of 11.28 g of PLA resin have been reported in Table 2. Impacts have been calculated for 1 bottle and for 3 bottled, according to the daily average consumption of 1.5 L of drinking water in Italy. In addition, in order to consider the yearly use of PLA bottles, 1 FU (1095 bottles) have been considered for the final impact calculations.

Data related to the aluminium extraction processes were included into the extraction process, already present in the Ecoinvent® database. The bottle production starts with the lamination of an aluminium disc to form a thin layer of metal that will form the body of the bottle that is then pressed and the neck formed. After a first washing, the preformed

Table 1

Life cycle inventory (LCI) of data for the production of 1 PET bottle of 500 ml of capacity, including primary and secondary data.

Input	Quantity	Unit	Unit process @ OpenLCA
SYNTHESIS OF ETHYLENE OXIDE			
Electricity	0.00175	kWh	Electricity, medium voltage APOS, RER
Ethylene	0.01580	kg	Ethylene, average APOS, RER
Oxygen	0.00879	kg	Oxygen, liquid APOS, RER
SYNTHESIS OF MEG*			
Ethylene oxide	0.01375	kg	
Electricity	0.00745	kWh	Electricity, medium voltage APOS, RER
Tap water	0.11804	kg	Tap water production, conventional with biological treatment APOS, RER
SYNTHESIS OF PTA**			
Purified terephthalic acid	0.01910	kg	Polyethylene terephthalate, granulate, amorphous APOS, RER
PET RESIN PRODUCTION			
MEG	0.00668	kg	–
PTA	0.01662	kg	–
Electricity	0.01185	kWh	Electricity, medium voltage APOS, RER
Heat	0.23894	MJ	Heat, district or industrial, natural gas APOS, RER
PET BOTTLE PRODUCTION			
PET RESIN	0.01910	kg	–
Stretch blow moulding	0.01910	kg	Stretch blow moulding APOS, RER

* MEG = monoethylene glycol; ** PTA = purified terephthalic acid.

Table 2

Life cycle inventory (LCI) of data for the production of 1 PLA bottle of 500 ml of capacity.

Input	Quantity	Unit	Unit process @ OpenLCA
SYNTHESIS OF PLA RESIN			
Poly lactide	0.01128	kg	Poly lactide, granulate, at plant APOS, RER
PLA BOTTLE PRODUCTION			
PLA RESIN	0.01128	kg	
Stretch blow moulding	0.01128	kg	Stretch blow moulding APOS, RER

bottle is internally coated with a thin layer of polyamide and then dried. Finally, a glazing step for colouring and differentiating the external design is carried out (Ciacci et al., 2014; Farjana et al., 2019). For the HDPE cap production, the injection moulding process already present in the Ecoinvent® database was used. The blowing process takes place intermittently and thus allows the formation of the characteristic screw around the cap, after heating and inflating (Giridharreddy and Tech, 2013). To calculate the electricity and heat consumed in the production process, data reported in an LCA study relating to the production of aluminium cans were used, and appropriately modified and converted according to the average weight of 1 bottle (Niero and Olsen, 2016). All input materials for bottle production have been reported in Table 3. Impacts have been calculated for 1 FU. In order to taking into account the daily and annual time-frame, the system boundaries have been extended to the bottle every-day washing (Table 4). The LCIA includes the amount of tap water used, the gas necessary for heating and the soap for 365 washing operation. As a reference, a gas methane boiler for domestic use of 24 kW of heating power with 85% yield, averagely consuming 0.2–1.4 m³/h of methane.

2.4. Impact assessment and interpretation of results

For the implementation of the impact assessment and the overall LCA modelling, the ReCiPe Midpoint (H) (PRé Consultants, Amersfoort, the Netherlands) method and the open-source package OpenLCA™ v.1.8

Table 3

Life cycle inventory (LCI) of data for the production of 1 aluminium bottle of 750 ml of capacity, including primary and secondary data.

Input	Quantity	Unit	Unit process @ OpenLCA
BOTTLE PRODUCTION			
BOTTLE BODY PRODUCTION			
Aluminium, alloy	0.00123	kg	Aluminium, wrought alloy APOS, GLO
Sheet rolling, aluminium	0.10590	kg	Sheet rolling, aluminium APOS, GLO
Coating powder	0.00605	kg	Coating powder APOS, RER
Enamelling	10.05943	m ²	Enamelling APOS, RER
Heat	850.5094	MJ	Heat, from steam, in chemical industry APOS, RER
Tap water	1.0000	kg	Tap water production, conventional with biological treatment APOS, RER
CAP PRODUCTION			
ABS	0.01300	kg	Acrylonitrile-butadiene-styrene copolymer APOS, RER
Electricity	0.22500	kWh	Electricity, medium voltage APOS, RER
Heat	6.66000	MJ	Heat, from steam, in chemical industry APOS, RER
Injection moulding	0.01300	kg	Injection moulding APOS, RER
BOTTLE PRODUCTION			
Electricity	2.05555	Kwh	Electricity, medium voltage APOS, RER
Heat	31.7038	MJ	Heat, from steam, in chemical industry APOS, RER
Bottle body	1	item	–
Cap	1	Item	–

Table 4

Life cycle inventory (LCI) of data for 1-day washing of 1 aluminium bottle of 750 ml of capacity, including primary and secondary data.

Input	Quantity	Unit	Unit process @ OpenLCA
BOTTLE WASHING			
Bottle	1	Item	–
Gas (methane) consumption	0.01350	m ³ /min	Heat production, natural gas, at boiler atm. low-NOx condensing non-modulating <100 kW central or small-scale APOS, RER
Soap, at plant	0.0283	kg	Soap production soap APOS, RER
Tap water, at user	3.0500	kg	Tap water, at user RER

(GreenDelta, Berlin, Germany) were used. An overview of the relevant impact categories reported in this study and what they describe are reported in Table 5.

2.5. Microbiological quality of water in PET bottle and PLA bottle vs aluminium bottle

A sample of water contained in the PET bottle and PLA bottle was withdrawal immediately after the cap opening and analysed as Pre-T₀. Then, at 40 min' interval for 6 times, about 80 ml of water was drunk directly from the bottle, in order to inoculate mouth bacterial charge into the water and simulate the usual outdoor drinking behaviour. Samples of water have been collected after the first drink (T₀), after 2 h (T₂) and after 4 h (T₄). 100 µl samples were collected, in order to avoid volume variations during the experiments. All assays have been carried out in triplicate.

In the case of aluminium bottle, the bottle was filled in the morning of the first day with tap water, filled again after 4 h, and then washed with hot water only and with hot water + soap, until the same double refilling the day after. again. At 40 min' interval for 6 times per day, about 125 ml of water was drunk directly from the bottle. 100 µl samples were collected as Pre-T₀, T₀, T₂ and T₄, as previously mentioned. Samples have been seeded at different dilutions on Petri dishes filled with Plate Count Agar medium and incubated for 48 h at 30 °C and then the colonies visually counted and data reported as colony forming units (CFU)/ml vs. time of sampling. All assays have been carried out in

Table 5

The impact categories with the corresponding abbreviations calculated in this study and their general description.

Impact category	Description
Global warming potential (GWP)	Indicator of potential global warming due to emissions of greenhouse gases to air
Ozone depletion potential (ODP)	Indicator of emissions to air that cause the destruction of the stratospheric ozone layer
Acidification of soil and water potential (AP)	Indicator of the potential acidification of soils and water due to the release of gases such as nitrogen oxides and sulphur oxides
Eutrophication potential (EP)	Indicator of the enrichment of the aquatic ecosystem with nutritional elements, due to the emission of nitrogen or phosphorous containing compounds
Photochemical ozone formation potential (POFP)	Indicator of emissions of gases that affect the creation of photochemical ozone in the lower atmosphere (smog) catalysed by sunlight.
Fossil fuels depletion potential (FDP)	Indicator of the depletion of natural fossil fuel resources
Human toxicity potential (HTP)	Impact on humans of toxic substances emitted to the environment
Eco-toxicity potential (ETP)	Impact on freshwater organisms of toxic substances emitted to the environment
Water depletion potential (WD)	Indicator of the amount of water required to dilute toxic elements emitted into water or soil
Land occupation potential (LOP)	Indicator of the land extension that is potentially subtracted to impact food production and thereby human health.

triplicate.

3. Results and discussion

3.1. LCA of production and of daily use of PET, PLA and aluminum bottles

The results of LCA related to bottles production, expressed per 1 bottle of different materials are summarized in Table 6. In addition, considering the different capacity of PET and PLA bottles in comparison with aluminum bottle, 3 PE T/PLA one-way bottles and 1 aluminum bottle refilled twice have been accounted for in impacts calculation, in order to cover the reference daily water consumption of 1.5 L. The impact categories units are related to the substance taken as a reference for each of them and in respect of which all other substances concurring to that impact category are converted to.

As regards the production process of the aluminum bottle, Table 5 shows the cumulative impacts for each impact category; all data are referred to the production of a single bottle. As it is well-known, aluminium bottle lifetime is accounted in several years, but it can be easily damaged by dents, so we hypothesize for a bottle an average lifetime of 2.5 years (930 days). In the idea that only one container per person is repeatedly used to meet the average daily amount of water drunk equal to 1.5 L, the impact due to daily use of aluminum bottle could be correctly divided by 930. It's worthwhile noting that the environmental impact for producing an aluminum bottle in all categories is at least one order of magnitude more than both plastic bottles for one item, on the contrary for daily consumes the impacts of aluminum bottle is consistently reduced.

It's worthwhile noting that the evaluation of the effective bottle reusability is very difficult, because, if in theory it can be used almost infinite times, in practice there is a factual probability of breaking or damaging event. A scenario of a lifetime of 25 years has been considered realistic for this study.

Concerning GWP, our results is comparable with values reported in literature, ranged from 0.091 to 0.156 kgCO₂eq for the production of 1 PET bottle (Papong et al., 2014; Perugini et al., 2005), whereas in several studies on corn-based PLA bottle production show lower GWP values, as 0.212 kgCO₂eq/bottle proposed by Gironi and Piemonte (2011) and 0.414 kgCO₂eq/bottle found in Nikolic et al. (2015), or 0.122 kgCO₂eq/bottle starting from cassava cultivation (Papong et al., 2014). This is probably due to the fact that in some studies CO₂ uptake during crop cultivation has been accounted for and subtracted to the final GWP values. In addition, it is very difficult to compare different crop-based PLA bottle production, because cultivation inputs (i.e., pesticides, fertilizers, field operations) are usually different with different effects on GWP. At the best Authors' knowledge, any study concerning LCA of reusable aluminum bottle has been published yet.

Specifically, for PET bottle production, the greatest contribution in almost all impact categories has been attributable to PET resin production (blue bars) followed by the stretch blow moulding for bottle forming (yellow bars). Within the PET resin production, about 50% of all impacts are caused by PTA production (Fig. 2). The main anthropic contribution to the greenhouse effect GWP) is due to the combustion of fossil fuels and it is expressed as kg of CO₂ equivalent. During the synthesis of PTA, the paraxylene production covers more than 13% of the overall GWP because of the use of many chemical additives.

On the other hand, water depletion potential is significant in the stretch blow moulding operation, reaching almost 45% the total potential water use (WDP). This is due to the high temperature of materials during the blow moulding that needs great amount of cooling water, which in turn influences also the other impact categories related to wastewater, as EP and ETP.

In the case of both HTP and ETP, a distinction must be made between acute toxicity, which can lead to the death of the target when the concentration of toxic in the environment is very high, and chronic with

Table 6

LCA of the production of 1 bottle of PET, PLA and aluminum bottles. The daily scenarios have also been calculated, considering the average drinking water consumption of 3 one-way PET and PLA bottles. In the case of aluminum bottle, hypothesizing an average durability of 2.5 years (930 days), the daily use corresponds to the 930th part of the overall impact due to its production.

Impact category	1 PET bottle	3 PET bottles (daily cons.)	1 PLA bottle	3 PLA bottles (daily cons.)	1 aluminum bottle	1 aluminum bottle (daily use)	Unit
Climate change (GWP)	0.134	0.403	0.616	1.847	11.921	0.01307	kgCO ₂ eq.
Eutrophication (EP)	2.28·10 ⁻⁵	6.85·10 ⁻⁵	5.90·10 ⁻⁴	17.70·10 ⁻⁴	0.011	1.12·10 ⁻⁵	kgPO ₄ eq.
Photochemical oxidant formation (POFP)	3.10·10 ⁻⁴	9.30·10 ⁻⁴	25.20·10 ⁻⁴	75.60·10 ⁻⁴	0.0279	3.06·10 ⁻⁵	kgNMVOC ^a
Ozone layer depletion (OLDP)	9.82·10 ⁻⁶	29.40·10 ⁻⁶	9.17·10 ⁻⁸	27.5·10 ⁻⁸	5.19·10 ⁻⁷	–	kgCFC-11 ^b eq.
Acidification (AP)	5.20·10 ⁻⁴	15.60·10 ⁻⁴	27.5·10 ⁻⁴	82.5·10 ⁻⁴	0.054	5.92·10 ⁻⁵	kgSO ₂ eq.
Fossil depletion (FD)	0.055	0.166	0.247	0.742	3.229	0.00354	kg oil eq.
Water depletion (WD)	6.99	20.97	8.92	26.76	477.0	0.52303	liters
Human toxicity (HTP)	0.050	0.151	0.218	0.653	5.486	0.00602	kg 1,4-DB ^c eq.
Eco toxicity (ETP)	2.15·10 ⁻³	6.45·10 ⁻³	9.86·10 ⁻³	29.58·10 ⁻³	0.266	2.92·10 ⁻⁴	kg 1,4-DB eq.
Land occupation (LOP)	2.01·10 ⁻³	6.03·10 ⁻³	32.28·10 ⁻³	96.84·10 ⁻³	0.160	1.75·10 ⁻⁴	m ²
Particulate matter formation (PMFP)	1.70·10 ⁻⁴	5.10·10 ⁻⁴	13.4·10 ⁻⁴	40.20·10 ⁻⁴	178.90·10 ⁻⁴	1.96·10 ⁻⁵	kg PM10 eq.

^a NMVOC = non methane volatile organic carbon.

^b CFC-11 = Trichlorofluoromethane.

^c 1,4-DB = 1,4 dichlorobenzene.

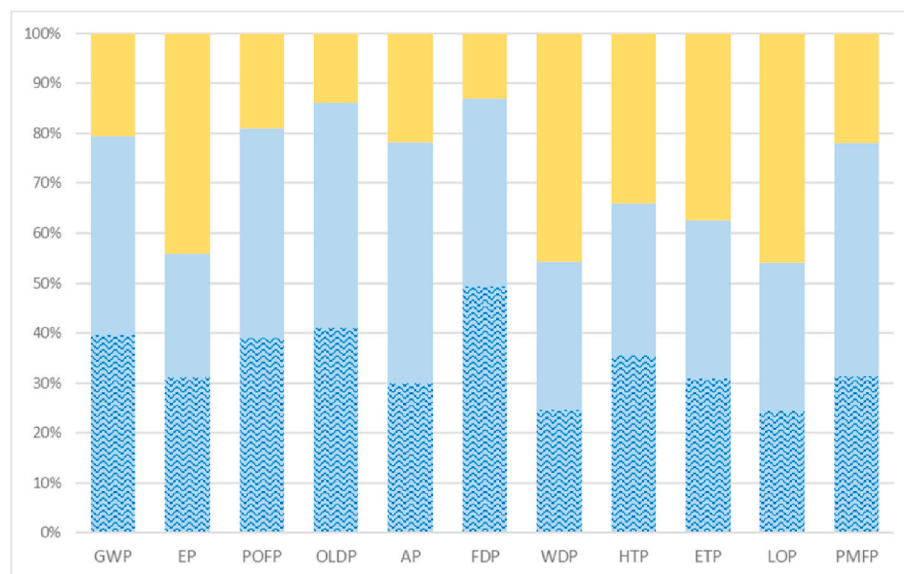


Fig. 2. Results of LCA method (ReCiPe® Midpoint (H)), expressed in terms of contribution trees for the impact categories considered in this study. The overall process is 100%, the yellow bars correspond to the contribution of the stretch blow moulding process for bottle forming; the light blue bars correspond to the PET resin production process, where almost 50% of impacts in all the categories is due to the PTA production (wavy blue bars). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

long-term effects caused by non-biodegradable substances and persistent which can be also bioaccumulated. These impacts can occur both on a global and regional scale depending on the diffusion factor, which makes very difficult to interpret the impact data in terms of effective action on human health. Emissions in atmosphere, generated by combustion in the factory, transport and due to processing machinery are the main contributor of ETP. For each toxic substance, the HTP and ETP is expressed taking as a reference the 1,4-dichlorobenzene equivalents.

As expected (Baldowska-Witos et al., 2020), PLA production is mainly burdened by PLA resin production, that contributes for more than 90% in all impact categories (Fig. 3), except for WDP, due to the stretch blow moulding operation, for the reasons mentioned above. Specifically, 91.08% of the overall GHG emissions occur during the corn production phase and are caused by the large amount of fertilizers and herbicides that are generally used in corn cultivation. The corn drying stage also generates GHG emissions, but in weight less than 1% of the overall impacts. The agricultural phase has to be retained the principal contributor to the environmental damages and impacts in all the categories. The use of fertilizers generates very high releases of nitrates and

phosphates into the environment, provoking very marked eutrophication phenomena (Hong et al., 2016). In the case of aquatic systems, this leads to a disproportionate algal proliferation that triggers a break in the ecosystem balance and can cause the death of the fish fauna by asphyxiation. The effects of this impact are usually regional, localized closed to areas of substances release (Reddy et al., 2018). In the production of biodegradable plastic, more than 95% of eutrophication is usually attributable to agricultural processing (Dietrich et al., 2017). Another relevant effect that raises concern for the human health is the thinning of the ozone layer. Ozone performs a protective function as it absorbs most of the harmful ultraviolet radiations with a high energy content, capable of interacting with biological molecules, such as DNA, and thus causing the onset of tumors and mutations both for aquatic ecosystems and terrestrial (Gaur et al., 2018).

The substances which this damage with a global effect has been attributed to, are the chlorofluorocarbons (CFCs). CFCs induce a chain mechanism that leads to the formation of chlorine and fluorine radicals, acting as catalysts in ozone decomposition (Burkholder et al., 2015). For both PET and PLA, the main contribution to the thinning of the ozone

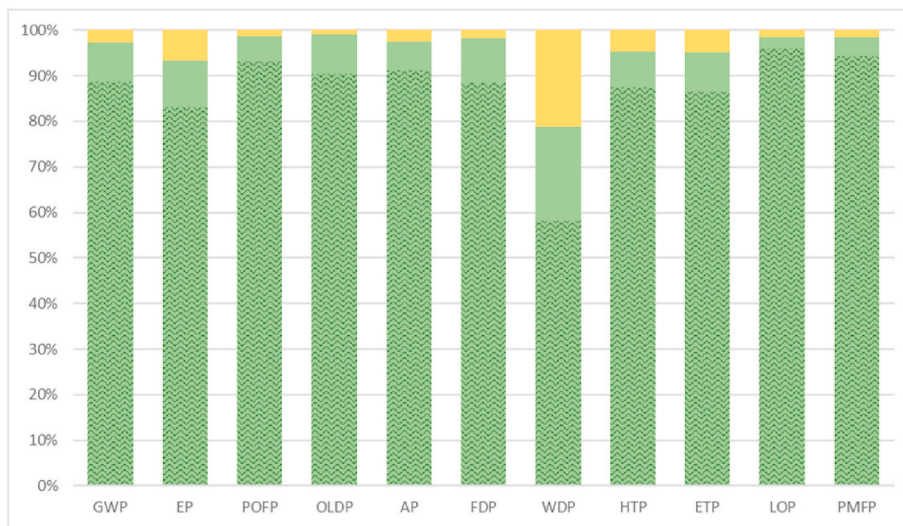


Fig. 3. Results of LCA method (ReCiPe® Midpoint (H)), expressed in terms of contribution trees for the impact categories considered in this study. The overall process is 100%, the yellow bars correspond to the contribution of the stretch blow moulding process for bottle forming; the green bars correspond to the PLA resin production process, where more than 90% of impacts in all the categories is due to the agricultural phase (wavy green bars). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

layer comes from the polymer synthesis step.

It is worthwhile noting that PLA bottle production has general overall higher impacts than PET bottle production, globally or regionally depending on the categories. The stretch blow moulding process presents low differences mainly attributable to the different quantity of resin required per unit of bottle produced. In fact, the average weight of a 500 ml PET bottle is 19.10 g while the average weight of a PLA bottle of the same volume is 11.28 g. This difference is due to the different density of the two materials and affects the environmental impacts of stretch blow moulding related to the demand of electricity for the extrusion and blow moulding process. This result is in agreement with the study carried out by Baldowska-Witos et al. and Nikolić et al. (2015) (Baldowska-Witos et al., 2020; Nikolić et al., 2015).

Aluminum bottle production consists in two main steps that is bottle body production and cap production (Fig. 4). It is self-explanatory that the greatest contribution is due to the manufacture of the bottle body, also including the aluminum extraction process. The body of the bottle participates to GWP for 78.94%, equal to the emission of 7.88 kg of CO2 equivalent, of which 66.10% is due to the energy needs of the processing plant and the process itself, while the extraction process contributes 7.60% due to the electrolysis processes and the heat required. It's

worthwhile noting the considerable impact of the electricity consumption for the bottle production (Fig. 4).

3.2. LCA of annual use scenarios

Taking into account the time-frame of one year, as mentioned before, the total amount of one-way PET and PLA bottles correspond to a total of 1095 pieces each, vs. 0.4 aluminum bottle (assuming 2.5 years of lifetime). The bottle refilling does not provide any additive impact, because of the use of the same bottle. In the case of aluminum bottle, the everyday washing with hot water + soap have been added (Fig. 5). Data on aluminum bottle washing have been obtained as an average of three hand washing with soap and tap water, assuming that at the end of day for the bottle washing the sink is not filled with water but the bottle is simply placed under the hot water flow. An average of 3 L of hot water for 1.2 min of bottle washing has been derived from experimental trials, where the amount of water used for washing has been recovered and measured several times. Our result was slightly less than the value reported, for example, by the US Geological Sciences (USGS, 2018), that is, as lowest value, 1.5/minute, corresponding to about 5 L/min. We excluded the scenario of the use of dishwasher, limiting our analysis to

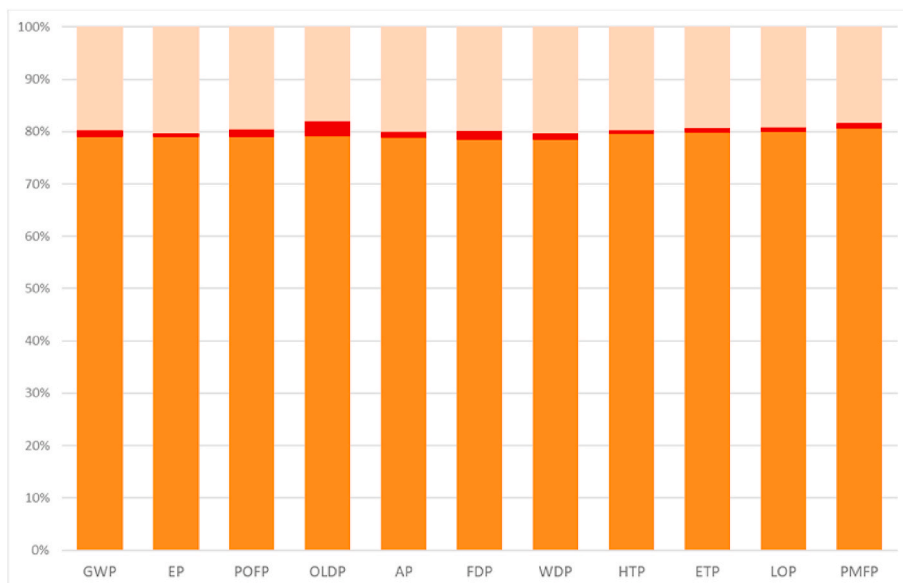


Fig. 4. Results of LCA method (ReCiPe® Midpoint (H)), expressed in terms of contribution trees for the impact categories considered in this study. The overall process is 100%, the pink bars correspond to the contribution of the electricity consumption during bottle forming process; the orange bars correspond to the bottle body and the red bars to the HDPE cap production process, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

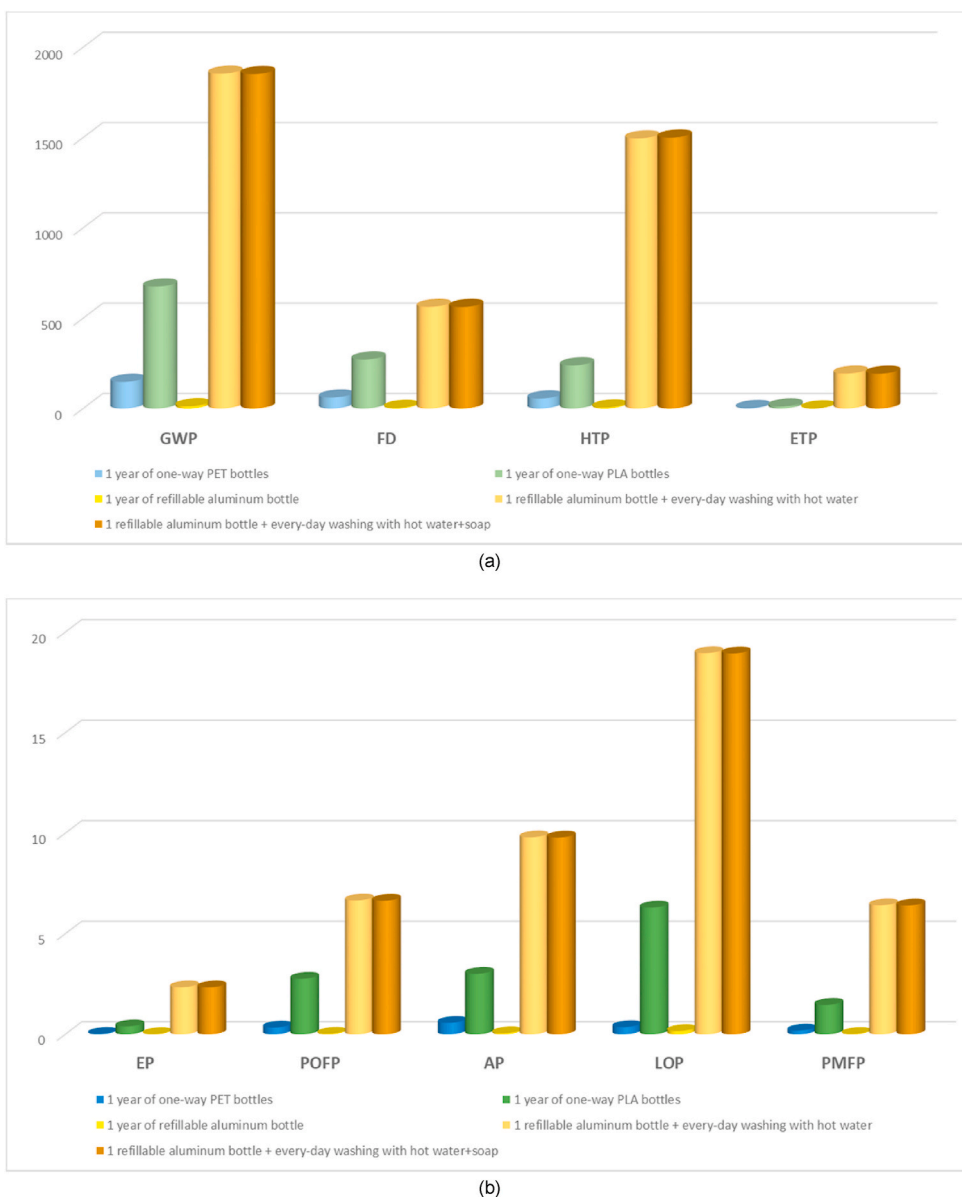


Fig. 5. – Impact categories related to 1 FU of PET or PLA bottles, and of aluminum refillable bottle, including daily washing with hot water, with or without soap.

the hand wash because of aluminium items cannot be dishwashed, differently from other materials for bottles, as steel or glass. Otherwise, it's worthwhile noting that currently automatic dishwashing is more superior as compared with manual dishwashing in terms of performance and resource consumption under the tested conditions. Furthermore, it points out that washing up dishes under running tap water is the most water-consuming manual dishwashing method of all investigated ones (Berkholz et al., 2013).

Observing the results for each impact categories, the high impact of gas consumption for water heating appears evident. The impact related to the use of soap, less than 0.1% and mainly due to the production process, is therefore negligible. The annual use of PET bottles has lower environmental impact than using PLA bottles, but, as expected, the use of 1095 one-way bottles (both of PET or PLA) for 1 year has a greater environmental impact than the use of a refillable aluminum bottle, in terms of GWP, as well as of all the other impact potentials that can have effects on the ecosystems and human health. Otherwise, when the refillable bottle every-day washing is included in the analysis, results are completely upset, resulting in an increase of at least two order of magnitude in each category.

In a circular economy perspective and taking into account the waste management hierarchy proposed by the EU, the waste reduction is surely the main priority, which favors the use of a refillable bottle instead of any disposable materials, but even considered the use of only one bottle per year, the annual impact of washing is severe. In addition, the end-of-life scenarios suggested by the EU provide the application of the principles of recycling or recovery (in this order of priority, after reduction and reuse) for packaging materials, as always preferable options compared to landfilling.

In this study, three end-of-life scenarios for PET and PLA bottles have been considered:

- open-loop and closed-loop recycling, compared to 100% unsorted waste destined to 100% incineration or 100% landfill (as worst scenarios), for PET
- 100% composting and 100% incineration (as worst case of unsorted waste), for PLA.

Regarding the aluminum bottle, no final provisions were hypothesized because in the time frame considered for the impact study, one

year, it was assumed that it does not reach the end of its life but is continuously reused.

For the evaluation of the environmental impact deriving from PET incineration and landfill, the processes present in the Ecoinvent® database, referred to 1 kg of PET and GWP was chosen as the impact category. The analysis revealed that landfill disposal causes the input of 0.0801 kgCO₂ eq/kg PET, whereas incineration 2.0326 kgCO₂ eq/kg PET, corresponding to 1.53·10⁻³ and 38.8·10⁻³ kgCO₂ eq per bottle, and 1.7 and 42,5 kgCO₂ eq. per year, respectively. The greater impact of incineration is due to the incineration plant itself and to the production of energy for combustion, which does not occur in landfills, where waste is simply collected and accumulated. PET bottle recycling allows for the production of secondary PET granules or flakes of non-bottle grade quality destined to other uses, as for fibers (open-loop recycling) (Shen et al., 2010). After undergoing a solid state poly-condensation (SSP) process, where the granules are heated at temperatures below the melting point in order to increase their intrinsic viscosity to levels compatible with the injection moulding process and to remove any possible residual organic contamination deriving from their previous use, PET can be reused for bottle production (closed-loop recycling) (Chilton et al., 2010).

Using the *avoided burdens method* (Finnveden et al., 2009), Nessi S. et al. (Nessi et al., 2018) evidenced that the open-loop PET recycling to fibers could allow an environmental benefit of about -24% of the overall GHG emissions, due to the lower use of virgin PET, while the closed-loop an environmental benefit of about -5% of the overall GHG emissions, due to the impact of the processes that re-convert PET to the bottle-grade. This method is based on the assumption that the amount of secondary material or product obtained from recycling processes allows for the avoidance of the primary production of the same or of a lower amount of that material or product. The avoided burdens of such primary production processes are hence credited to the system itself. Applied to our study, it means that the scenario of PET recycling permits to save an amount of CO₂ eq. ranging from 35.3 (open-loop option) to 7.3 (closed-loop option) kg on 1-year basis.

Analyzing the end-of-life scenarios of the PLA bottles, two options have been evaluated, 100% incineration in case of management as unsorted waste, or 100% composting in the case of differentiated waste collection as municipal organic fraction. Incineration does not lead to any benefit to the environment in terms of avoided impacts, because the material is totally burned, releasing indeed about 20% of extra GHG emissions (Razza and Innocenti, 2012). The composting route, often indicated as the best option and green alternative that encourages the

production of bioplastics, according to the study by Nessi S. et al., does not bring any environmental benefit as the bottle is totally degraded and therefore no fraction is recycled and reused for the synthesis of secondary bottles. This means that in the initial phase of the production chain of bioplastic bottles there is no reduction both in the quantity of virgin material and in the agricultural phase, the most impacting production stages. Otherwise, it is worthwhile noting that the carbon released during PLA composting is the biogenic carbon taken up by atmosphere during maize growth stage, so the net emissions of composting process is zero. Moreover, the production of compost can be considered advantageous because of its use as organic fertilizers in place of chemical fertilizers nitrogen and phosphorous-based (Larney et al., 2006). Assuming a yield in compost of about 30% from PLA (i.e., 0.33 kg of compost/1 kg of processed PLA), the use of compost as fertilizer would return about 0.3 kg CO₂ eq/kg compost (De Andrade et al., 2016). Based on these data, we can see a small emissions credits for composting alternative: in 1 year from 12.3 kg di PLA (1095 bottles), about 4 kg of compost is produced, and, even hypothesizing its use in maize organic fertilization, it returns only about 0.5 kg of CO₂ eq.

3.3. Microbiological quality of water

Fig. 6 shows the result obtained from the microbiological analysis of water sampled on the PET and PLA bottles, respectively. As expected, in both cases Pre-T0 samples are microbiologically pure, as reported on the official analysis on the bottle labels. A low microbial contamination has occurred in 4 h, probably due to the inoculum provided by saliva during the contact between the mouth and the bottle neck. After 4 h, empty bottles were thrown away and new bottles have opened, starting each time with microbiologically pure water. Final contamination has not exceeded the log₂ order or magnitude of microbial charge, for both PET and PLA bottles. For plastic bottles, microbiological quality of water has been monitored for 6 h after opening, under the hypothesis that each closed bottle opened has the same characteristics, so the use of 3 bottles per day means for three times the repetition of the same trend of microbial charge.

For aluminium bottle, results are reported in Fig. 7 and have evidenced some interesting difference compared with the previous cases. In this case, we have hypothesized a scenario where the bottle is filled in the morning of Day1, drunk with mouth in 6 h, then washed at the end of Day 1 and stored upside-down over the wash-basin until the morning of Day 2 when it is refilled and re-drunk in 6 h.

Here, Pre-T0 sample showed a slight contamination, probably due to

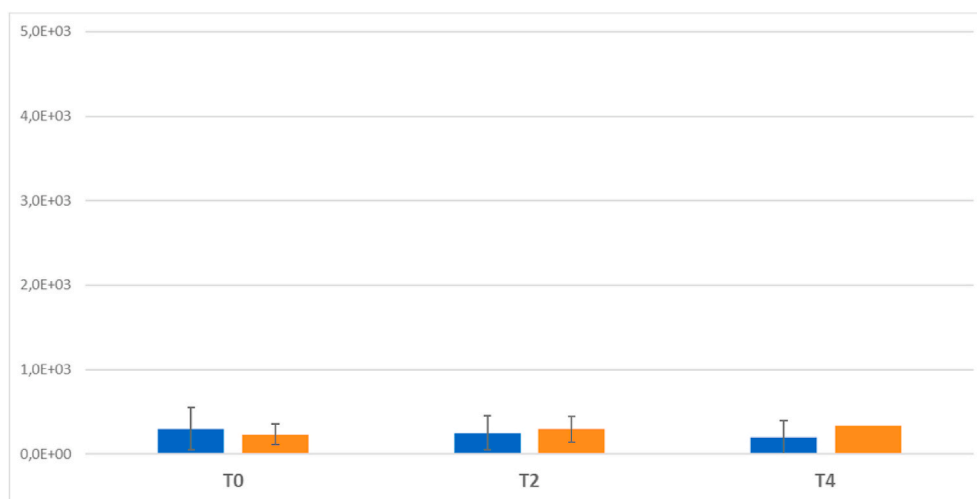


Fig. 6. Microbial contamination (expressed in CFU/ml) within 0, 2 and 6 h of opening, in a scenario of drinking every 30 min, for one PET bottle (blue bars) and PLA bottle (orange bars). PreT0 sampling resulted in 0 contamination (microbiologically pure) in both bottles immediately before the opening and are not reported in the graph. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

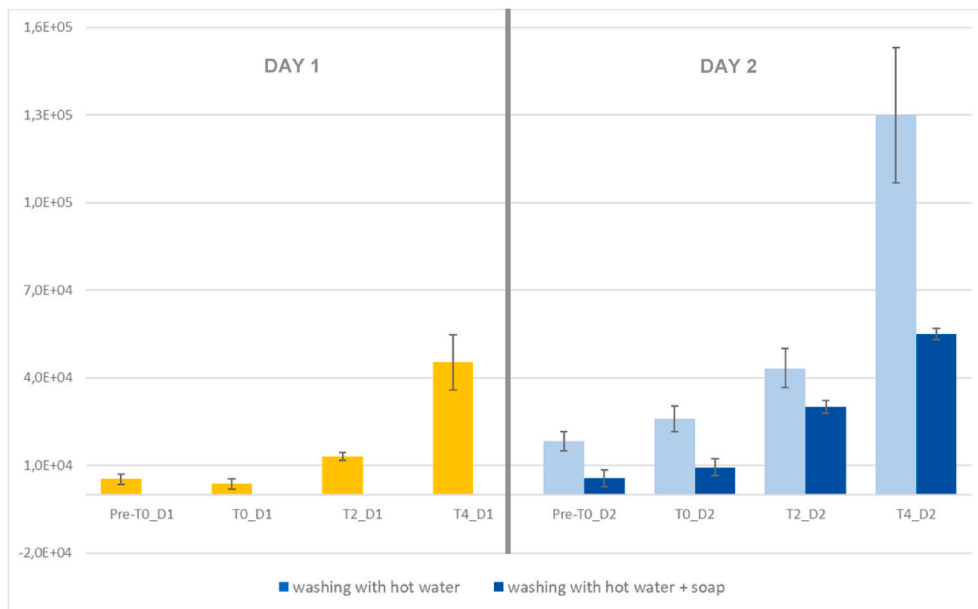


Fig. 7. Microbial contamination (expressed in CFU/ml) immediately after the tap water filling (Pre-T0) and within 0, 2 and 6 h of opening, in a scenario of drinking every 40 min, for aluminium refillable bottle (yellow bars) in a Day 1 use. The light blue bars represent a hypothetical Day 2 scenario of tap water bottle re-filling after washing with hot water only; the dark blue bars represent a Day 2 scenario of tap water bottle re-filling with tap water after washing with hot water and soap. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the fact that the bottle is not sterilized and could be exposed to ambient microbial charge. Then, in 4 h it has reached a contamination of $4.5 \cdot 10^4$ CFU/ml. In the first scenario, at the end of the Day 1, the bottle is rinsed with hot water and put away until the Day 2 in the morning, when it is refilled. Before starting to drink, the microbial contamination is increased of about one order of magnitude, from $5.0 \cdot 10^3$ CFU/ml to $1.8 \cdot 10^4$ CFU/ml, reasonably because the internal surface of the bottle has acted as inoculum surface for microorganisms. During the Day 2, the final concentration of microorganisms in the water has reached the value of $1.5 \cdot 10^5$ CFU/ml and it could be expected that in Day 3 and subsequent, the effects of the self-inoculum due to the microorganisms remaining in the bottle would be destined to increase. In the scenario of bottle washing with hot water and soap, it appears evident the positive effect of soap on the overall microbial charge. At the end of Day 2 the contamination is anyway higher than in the case of PET or PLA bottles, but lower than in the previous case.

Various previous studies (Oliphant et al., 2002; da Silva et al., 2008; Sun et al., 2017; Mills et al., 2018) have shown that refillable drinking water was less safe or could be contaminated with bacteria that could harm human health. Reusable drinking water bottles are consistently humid and are easily contaminated via the user's hands and mouth, which are not devoid of microorganisms, especially the normal microbial flora of the skin and mouth. This provides a perfect medium for bacteria survival and multiplication, especially as these drinking water bottles are held at room temperature (21–25 °C) for several hours (Lawlor et al., 2009). Improperly cleaned water bottles may present a potential contamination risk and thus be considered a risk for foodborne illness (Shruti et al., 2020).

4. Conclusions

In conclusion, the analysis of environmental impact of bottles for drinking water have suggested that the production of a single bottle alone could conduct to a misleading conclusions, because in this case a wider framework is strictly necessary to understand the environmental consequences potentially deriving from the use of plastic, bioplastic or aluminum. In particular, whereas the analysis of a single bottle production suggested major impacts to be paid by the aluminum bottle, in the scenario of daily use, as well as expanding to a number of bottles for a 1 year-use, the use of aluminum bottle becomes advantageous because of its reusability, in comparison with the need of 1095 PET or PLA

bottles. Otherwise, taking into account the every-day washing with soap of the aluminum bottle, the environmental impacts in all categories could be revised in favor of one-way plastic bottles, because the water heating has burdens significantly on the environmental performance of the aluminum bottle, i.e. increasing the GWP of more than two order of magnitude. Moreover, PET bottles if properly recycled can assure an environmental benefit due to virgin material savings that permit lower burdening on natural resources depletion, in a circular economy perspective. PLA bottles sustainability pays for the great impact of agricultural phase, even if part of the impacts could be mitigating by the closed cycle of compost as organic fertilizer. Considering the microbiological quality of water, the use of one-way plastic/bioplastic bottles seems to assure a higher quality, especially compared with the refillable bottle rinsing only with water. From one side, reusable bottles can be more environmentally and economically friendly, because consumers can repeatedly refill them. But, the availability of nutrients released by the contact with mouth and saliva seems to be the principal determinants of microbial growth in drinking water. Currently, there are no guidelines recommended for personal water bottle cleaning frequency and sanitation. The results of this study, albeit need of further investigations, can be used to create awareness and educate the public on the importance of maintaining proper hygiene practices around these reusable water bottles. Moreover, a proper information about the circular economy best practices that permits a proper recycling of traditional PET bottles can help citizens and consumers to consciously approach that fundamental issue for the future of the planet.

Credit author statement

Stefania Costa, Methodology and Data curation. Letizia Battistella, LCA analysis and data collection. Daniela Summa, Microbiological analysis and Data collection. Giuseppe Castaldelli, Original draft reviewing. Elisa Anna Fano, Editing and reviewing. Elena Tamburini, Conceptualization and Supervision

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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