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Material selection and prediction of solar irradiance in plastic devices for application of solar water disinfection (SODIS) to inactivate viruses, bacteria and protozoa

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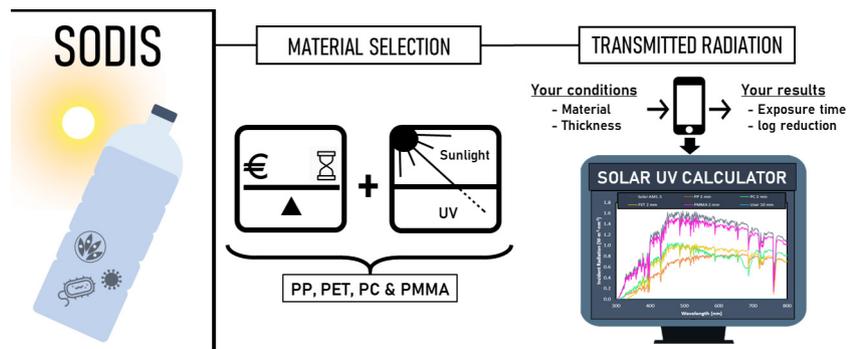
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HIGHLIGHTS

- Revision of mechanical and optical properties, weathering and costs of plastics
- Selection of optimal materials for SODIS container manufacturing
- Development of a tool for estimating UV radiation available inside containers
- Predictive evaluation of bacteria, viruses and protozoa solar inactivation
- Freely available *Solar UV Calculator* for custom evaluation of solar irradiance

GRAPHICAL ABSTRACT



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ABSTRACT

Solar water disinfection (SODIS) is a simple, inexpensive and sustainable Household Water Treatment (HWT) that is appropriate for low-income countries or emergency situations. Usually, SODIS involves solar exposure of water contained in transparent polyethylene terephthalate (PET) bottles for a minimum of 6 h. Sunlight, especially UVB radiation, has been demonstrated to photoinactivate bacteria, viruses and protozoa. In this work, an in-depth study of the optical and mechanical properties, weathering and production prices of polymeric materials has been carried out to identify potential candidate materials for manufacturing SODIS devices. Three materials were ruled out (polystyrene (PS), polyvinyl chloride (PVC) and polyethylene (PE)) and four materials were initially selected for study: polymethylmethacrylate (PMMA), polypropylene (PP), polycarbonate (PC) and polyethylene terephthalate (PET). These plastics transmit sufficient solar radiation to kill waterborne pathogens with production costs compensated by their durability under solar exposure.

A predictive model has been developed to quantitatively estimate the radiation available for SODIS inside the device as a function of the material and thickness. This tool has two applications: to evaluate design parameters such as thickness, and to estimate experimental requirements such as solar exposure time. In this work, this model evaluated scenarios involving different plastic materials, device thicknesses, and pathogens (*Escherichia coli* bacterium, MS2 virus and *Cryptosporidium parvum* protozoon). The developed *Solar UV Calculator* model is freely available and can be also applied to other customized materials and conditions.

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1. Introduction

In 2015, the General Assembly of United Nations announced the Sustainable Development Goals (SDGs) to “end poverty in all its forms” (United Nations (UN), 2015). That same year, there were almost 900 million people without access to safe drinking water (United Nations (UN), 2018) which is one of the priority themes of the SDGs. SDG 6 “Clean water and sanitation” aims to “ensure availability and sustainable management of water for all”. Provision of drinking water is the first challenge that must be approached and is considered the first target of the goal titled “Achieve access to safe and affordable drinking water”. Most people without access to safe drinking water live in low-income countries with few financial resources, thus, they require low-cost, sustainable and simple systems for Household Water Treatment (HWT). Moreover, these countries are located in regions where solar irradiance levels are very high throughout the year. Therefore, a suitable HWT alternative for them is the use of solar water disinfection (SODIS), commonly carried out in 1–2 L polyethylene terephthalate (PET) bottles exposed to solar radiation for a minimum of 6 h (McGuigan et al., 2012). This process is based on the harmful effect of UV radiation and its synergy with temperature on pathogens (Joyce et al., 1996; McGuigan et al., 1998; Romero et al., 2011; Gómez-Couso et al., 2012; Castro-Alfárez et al., 2017).

SODIS photoinactivation processes can follow direct and indirect mechanisms and are strongly affected by the different UV spectral ranges and specific microbiological features of the pathogens (bacteria, viruses or protozoa).

- i) Direct photoinactivation is an endogenous process that takes place when a constituent of the microorganism itself (e.g. nucleic acids, proteins or other macromolecules) absorbs photons, inducing changes to their chemical structure. For example, nucleic acids absorb radiation of wavelengths below 320 nm. As a result, many viruses can be photoinactivated with the UVB component of the solar spectrum (Lytle and Sagripanti, 2005; Mattle et al., 2015; Nelson et al., 2018). Similarly, *Cryptosporidium parvum* protozoon has been also proved to be inactivated under solar light, predominantly by the direct genome damage produced by the UVB radiation (Liu et al., 2015; Nelson et al., 2018). Bacterial pathogens are also sensitive to direct photoinactivation by solar UVB radiation (Jagger, 1985).
- ii) Indirect photoinactivation occurs when a photo-produced reactive intermediate (PPRI) (e.g. hydrogen peroxide, hydroxyl radical, singlet oxygen, carbonate radical and superoxide) damages microorganism components. Endogenous indirect inactivation takes place when PPRI are generated from internal sensitizers. For viruses, insufficient generation of PPRI produced by a few internal sensitizers makes endogenous indirect damage negligible and slower than direct damage (Love et al., 2010). When only UVA and visible radiation is available, indirect damage produced by internal sensitizers can slowly damage protozoon components (Liu et al., 2015; Nelson et al., 2018). Several studies confirm the importance of indirect endogenous mechanisms for bacteria when internal sensitizers (catalase, alkyl hydroperoxide reductase (Ahp) and peroxidase enzymes) are illuminated with wavelengths across the solar UV (UVA and UVB radiation) and visible ranges, promoting the formation of PPRI such as reactive oxygen species (ROS) (Nelson et al., 2018). For example, in *Escherichia coli* endogenous damage decreases as wavelength increases, with negligible effects for wavelengths above 400 nm. In contrast, *Enterococcus faecalis* extends its susceptibility up to 500 nm (Silverman and Nelson, 2016). In such cases, direct and indirect endogenous damage can interact and, for this reason, both are often studied simultaneously. Furthermore, if the sensitizer is external, then the indirect photoinactivation is exogenous. This mechanism is only possible if the water contains these sensitizers. For example, as external PPRI are not present, indirect

exogenous photoinactivation does not happen in pure water (Nelson et al., 2018). In other water matrices, the photoinactivation of viruses and bacteria can be enhanced under UVA and visible radiation (Kohn and Nelson, 2007; Romero et al., 2011; Mattle et al., 2015). However, it is a complex mechanism that depends on many factors (pathogen species, physiological state, radiation wavelength, type of sensitizers and its relationship with the pathogen) (Maraccini et al., 2016). For protozoa, exogenous indirect damage is generally negligible due to the thick resistant oocyst-wall (Liu et al., 2015; Nelson et al., 2018).

Summarizing, protozoa and viruses are mainly photoinactivated by direct endogenous mechanisms caused through the action of UVB radiation while bacteria are damaged by direct and indirect endogenous processes through the action of UVA and UVB radiation. In addition, if the water is not pure, exogenous indirect damage can enhance solar disinfection for bacteria and viruses but not for protozoa.

Consequently, the efficiency of SODIS processes for the different types of pathogens is strongly affected by the transmission spectra of the materials used for the devices. Usually, SODIS treatment is carried out using PET bottles due to their low-cost and accessibility. Concerns about chemical contaminants from plastic migration have been addressed by previous studies (Wegelin et al., 2001; Ubomba-Jaswa et al., 2010a). Alternative materials have been successfully evaluated, such as polypropylene copolymer (PPCO), polycarbonate (PC) and polystyrene (PS) 1 L bottles (Fisher et al., 2012), polyethylene (PE) bags (Lawrie et al., 2015), polymethylmethacrylate (PMMA) (Ubomba-Jaswa et al., 2010b; Reyneke et al., 2020) and glass reactors (Kalt et al., 2014; Mac Mahon and Gill, 2018; García-Gil et al., 2018) fitted with compound parabolic collectors (CPC). The design of SODIS systems requires the selection of optimal materials for the manufacturing of the devices considering the UV transmission spectra but also the mechanical properties, resistance to weathering and production cost. Moreover, the predictive estimation of the SODIS efficacy requires an accurate calculation of the radiation intensity and spectrum inside the devices. For these reasons, the goals of this work are the selection of the optimal candidate materials for SODIS device manufacturing and the estimation of solar available radiation and its spectral distribution as a function of the material and wall thickness. For the latter, a tool called the *Solar UV Calculator* has been developed and evaluated to predict SODIS under different scenarios of plastic materials, device thicknesses and target pathogens (*E. coli* bacterium, MS2 virus and *C. parvum* protozoon). This *Solar UV Calculator*, was developed as part of the WATERSPOUTT H2020 project which aims to develop HWT technologies based on the SODIS process for African settlements in rural areas of Malawi, Ethiopia, Uganda and South Africa. The *Solar UV Calculator* is freely available to any potential user interested in the evaluation of SODIS devices with customized materials and conditions or even for other solar applications based on spectral transmission calculations.

2. Materials and methods

2.1. Mechanical properties

Selection of candidate plastics for the manufacturing of SODIS devices has been carried out through an exhaustive review of literature data. Only transparent plastics and without any kind of UV-stabilizer were considered in the study. Three sets of plastics were studied depending on their photostability:

- Poorly photostable plastics (Polystyrene (PS), polyvinyl chloride (PVC), polypropylene (PP) and polyethylene (PE)).
- Moderately photostable plastics (Polyethylene Terephthalate (PET) and polycarbonate (PC)).
- Highly photostable plastics (polymethylmethacrylate (PMMA)).

Mechanical properties such as tensile strength, elasticity, impact resistance and weathering resistance (humidity, temperature and solar radiation) as well as degradation mechanisms of photooxidation, were analysed. Finally, manufacturing costs were also considered using Ecoinvent and Plastic Europe Databases (Frischknecht et al., 2005; PlasticEurope, 2012).

2.2. Optical properties

The optical properties of candidate plastic materials were experimentally evaluated by recording their transmission spectra with a UV-Vis-NIR spectrophotometer (Varian Cary 500, Palo Alto, California, USA). Three independent and replicated measurements for different thicknesses of the plastic samples were considered.

2.3. Solar UV calculator tool

A predictive tool called the *Solar UV Calculator* has been developed to predict the total radiation available and its spectral distribution inside the plastic device as a function of the thickness and type of plastic. For this purpose, maximum solar radiation inside the device has been calculated by applying Eq. (1) in the solar UV-Visible range from 290 nm to 800 nm:

$$I_{inside}(\lambda) = I_{sun}(\lambda) \cdot T(\lambda) \quad (1)$$

where $I_{inside}(\lambda)$ is the monochromatic radiation intensity ($W \cdot m^{-2} \cdot nm^{-1}$) inside the device, $I_{sun}(\lambda)$ the monochromatic solar radiation intensity ($W \cdot m^{-2} \cdot nm^{-1}$) and $T(\lambda)$ is the transmittance of the plastic material at each wavelength (dimensionless). The transmittance (T) is directly related to the absorbance (A) of the material (dimensionless) by the well-known Eq. (2):

$$A(\lambda) = -\log(T(\lambda)) \quad (2)$$

whereas the absorbance is related to optical path length (L) in (m) by the Beer-Lambert law (Eq. (3)):

$$A(\lambda) = \beta(\lambda) \cdot L \quad (3)$$

where $\beta(\lambda)$ is the monochromatic extinction coefficient (m^{-1}) for wavelength λ , which depends on the material. For each type of plastic, the absorption spectrum was measured for three different thicknesses and the monochromatic extinction coefficients were obtained from the slope of the linear regression of absorbance vs thickness plot (for each wavelength).

Based on Eqs. (1), (2) and (3), the radiation intensity inside the device can be expressed as:

$$I_{inside}(\lambda) = I_{sun}(\lambda) \cdot 10^{(-Th \cdot \beta(\lambda))} \quad (4)$$

where the optical path length for the light transmission is the wall thickness of the device (Th) in (m).

2.4. Application of the Solar UV Calculator tool to different SODIS scenarios

The *Solar UV Calculator* tool was tested for the design of SODIS devices. For that, the influence of the thickness on the available radiation and its spectral distribution inside device was assessed. Evaluations were performed with selected plastic devices with a typical thickness of 2 mm for high-capacity devices (20 L) and 0.5 mm for low-capacity bottles (2 L). The ASTM G-173 reference AM 1.5 solar spectrum was considered for the incident solar radiation calculations. For the estimation of the UVB, UVA and Visible radiation, the integrals in the wavelength ranges of 290–320 nm, 320–400 nm and 400–800 nm were calculated, respectively.

The *Solar UV Calculator* tool was also tested for estimating the experimental requirements to ensure a specific pathogen inactivation. For that, a pseudo first-order kinetic model that relates the inactivation with the radiation intensity was used (Eq. (5)):

$$\frac{dC}{dt} = -k \cdot I \cdot C \quad (5)$$

By integrating Eq. (5), the expression for the logarithmic inactivation of the pathogen with time is obtained:

$$\ln\left(\frac{C}{C_0}\right) = -k \cdot I \cdot t \quad (6)$$

where C is the final concentration of the pathogen ($microorganisms \cdot mL^{-1}$), C_0 is the initial concentration of the pathogen ($microorganisms \cdot mL^{-1}$), k is the kinetic constant ($m^2 \cdot W^{-1} \cdot h^{-1}$), I is the incident radiation and t is the reaction time (h). For decimal logarithmic reductions, the napierian reduction has to be divided by 2.303.

To calculate the kinetic constant, the lethal doses (D) for a specific inactivation logarithmic reduction of three model pathogens (*E. coli* bacterium, MS2 virus and *C. parvum* protozoon) were found in the literature (Fisher et al., 2012; Lawrie et al., 2015; Abeledo-Lameiro et al., 2017). The three disinfection experiments were carried out under natural sunlight, were performed at low water temperature and in clear water to avoid inactivation speeding up due to temperature or exogenous damage and ensure that the damage is only product of solar radiation. Moreover, no attenuation coefficient is required in clear water as many authors describe a diffuse attenuation coefficient related to the water composition (Kirk, 1994; Craggs et al., 2003).

Finally, with Eq. (7) (combination of Eqs. (4) and (6)) the minimum exposure time for a specific thickness or the maximum thickness for a specific time reaction can be calculated for a set logarithmic reduction.

$$\ln\left(\frac{C}{C_0}\right) = -k \cdot I_{sun}(\lambda) \cdot 10^{(-Th \cdot \beta(\lambda))} \cdot t \quad (7)$$

Using Eq. (7), the minimum exposure time was calculated for a specific thickness of 0.5 mm and the minimum thickness was obtained when the exposure time is 6 h, the reaction time recommended in the SODIS manual with cloudless sky (Luzi et al., 2016).

3. Results and discussion

3.1. Selection of candidate plastic materials based on mechanical properties

The determination of the tensile stiffness of a plastic prior to breaking or permanently deforming and the energy required to fracture a plastic (toughness), ascertained by the measurement of the tensile modulus of elasticity and impact resistance, respectively, are critical mechanical properties for any plastics to be used as devices material. However, in addition to them, resistance to weathering is also critical. The harmful effects of weather exposure on plastics have been attributed to photo-degradation or photo-oxidation processes by UV light and the action of oxygen (Gillen and Celina, 2017). In general, polymer degradation is usually controlled using UV-stabilizers that block UV-transmission through plastic or avoiding the exposure to high temperatures, water or oxygen. However, in the case of SODIS processes, blocking UV is not desirable, while oxygen is dissolved in the water and present in the surrounding air. Consequently, the plastic materials candidate for SODIS devices should be pure plastics, without any kind of UV stabilizer. In this sense, poorly photostable plastics are generally dismissed and moderately photostable polymers will require regular replacement (Feldman, 2002).

Polystyrene (PS), polyvinyl chloride (PVC) and polyolefins such as polypropylene (PP) and polyethylene (PE) are poorly photostable polymers. General Purpose Polystyrene (GPPS), the only transparent PS,

exhibits high stiffness, but brittle behaviour (Nexant's ChemSystems Solutions, 2006). PS becomes brittle when chain scissions are produced by radiation action and O₂ addition to chain radicals (Wypych, 2015a). PVC can be either flexible and malleable or rigid and brittle (Titov, 1984). When PVC is exposed to natural weathering, mechanical properties such as tensile strength, elasticity, and impact resistance decline, and the material becomes increasingly discoloured, indicating lower transparency (Feldman and Barbalata, 1996). In the case of polyolefins, degradation by photooxidation proceeds by a free radical chain mechanism in four steps: initiation, propagation, chain branching, and termination. This is initiated by the presence of hydroperoxides and their decomposition leads to the formation of other radicals, chain scissions and crosslinking. Although the degradation mechanism of PE and PP by photooxidation is similar, the initiation, termination, and chain-branching reaction rates are different. Thus, PP chains contain tertiary hydrogens that make the formation of intermediate hydroperoxides much easier and faster. In addition, the reaction rates of the termination step for PE are 100–1000 times higher than for PP (Wypych, 2015b). Overall, these polymers have a lifetime less than one year when exposed to weathering without any stabilizer. Non-stabilized PE becomes exceedingly brittle within a few weeks (Rånby, 1993; Feldman, 2002). PP and HDPE are used in the production of many products (estimated at 10 and 6.3 million tonnes in 2017 by Plastics Europe Market Research Group, respectively) due to their low production prices valued at 8.25€/kg and 7.77 €/kg (data from Database Plastic Europe (PlasticEurope, 2012)).

Polyethylene Terephthalate (PET) and polycarbonate (PC) are considered to be moderately photostable polymers. These plastics have considerable lifetimes in the outdoors without stabilizers. PC is a tough and transparent plastic material with excellent strength, stiffness, impact resistance, and resistant to high temperature, that suffers a slow attenuation of these properties under photodegradation by UV radiation. Humidity does not affect PC ageing, however, it produces a synergistic effect when UV radiation initiates photooxidation. At lower humidity, the reaction happens at a slower rate but UV penetrates deeper into the polymer, resulting in a thick layer of degraded material. At higher humidity, a thin photooxidized layer is quickly formed which shields the core of the polymer from further damage by UV radiation (McKee, 2019). Turton and White (2014) confirmed experimentally that photodegradation decreases with the depth of unstabilized PC (Turton and White, 2014). PET is a strong, stiff engineering plastic with excellent machining characteristics, and chemical resistance. PET is often used for food applications where low moisture absorption, low thermal expansion, resistance to staining, or resistance to cleaning chemicals is required. Most PET photodegradation occurs at the surface. Photooxidation produces hydroperoxides that lead to chain scission causing the ageing of the material (Wypych, 2015b). PET can be transparent in the amorphous state with physical and chemical resistance and good hardness although it degrades upon weathering exposure (McKee, 2019). Photodegradation of PET and PC leads to more brittle materials with eroded surfaces and lower transmittance. Wall thickness is obviously an important factor for both polymers. Some authors have studied the suitable lifetime of polymer ageing taking into account the distribution of the reaction products across the width of the material and the problems related with the use of Fick's law for estimating this goal (Audouin et al., 1994). Wall thickness has to be sufficiently high to ensure that photodegradation reactions do not affect the core of the plastic piece but as thin as possible to allow maximum UV transmission to the interior. The production prices for PET and PC are estimated at 13.9 €/kg and 19.6 €/kg respectively (data from Database 2.0 Ecoinvent (Frischknecht et al., 2005)).

Finally, polymethylmethacrylate (PMMA) is a highly photostable polymer typically used without any stabilizer for extended (many years) outdoor applications (McKee, 2019). PMMA has a number of advantages over other candidate materials including outstanding weathering, resistance to UV radiation, transparency, hardness, rigidity

Table 1

Summary of mechanical properties and production costs for the selected plastic as candidate material for manufacturing SODIS devices.

	Resistance	Photostability	Durability	Production cost
PS	Low	Low	Very low	–
PVC	High	Low	Very low	–
PE	High	Low	Very low	Very low
PP	High	Low	Low	Very low
PC	High	Moderate	Reasonable	Moderate
PET	High	Moderate	Reasonable	Moderate
PMMA	High	High	High	High

and dimensionally stable. In addition, PMMA weakly absorbs UV radiation producing some chain scission and the formation of methyl methacrylate monomer. However, these reaction products hardly impair the stability of PMMA due to the fact that the monomers can combine again (Rånby, 1993). PMMA also has a good impact strength. Its main disadvantage is its high price: 28.9€/kg (data from Database 2.0 Ecoinvent (Frischknecht et al., 2005)). A summary of the mechanical properties for PS, PVC, PE, PP, PC, PET and PMMA are provided in Table 1.

Summarizing, PS, PVC, PE and PP are poorly photostable and non-durable polymers that makes photostabilization necessary for outdoors. However, PP with 1% of UV stabilizers remains stable for long periods, consequently maintains transparency and can also be easily and cheaply replaced. For this reason, PP has been considered as a possible plastic material for SODIS devices while PS, PVC and PE were excluded as candidate materials for the manufacturing. The production prices for PET and PC are higher than that of the PP, however it is compensated by durability. Finally, considering weathering resistance and UV radiation transmission PMMA could be the optimal material for manufacturing SODIS devices. Furthermore, although its production price exceeds that of the rests of polymers described, again, this is compensated for by durability. Therefore, on the basis of mechanical properties, PP, PET, PC and PMMA were the four candidate materials considered for manufacturing SODIS devices.

3.2. Optical analysis of candidate plastic materials

SODIS relies on the damage caused by solar UV radiation to pathogens. However, pathogen susceptibility varies with the wavelength and microbiological features. Thus, the wavelengths transmitted inside the SODIS device is an important factor for disinfection performance. Consequently, the transmission spectra of the four materials selected on the basis of their mechanical properties were experimentally analysed and are shown in Fig. 1. PP transmits radiation from 235 nm,

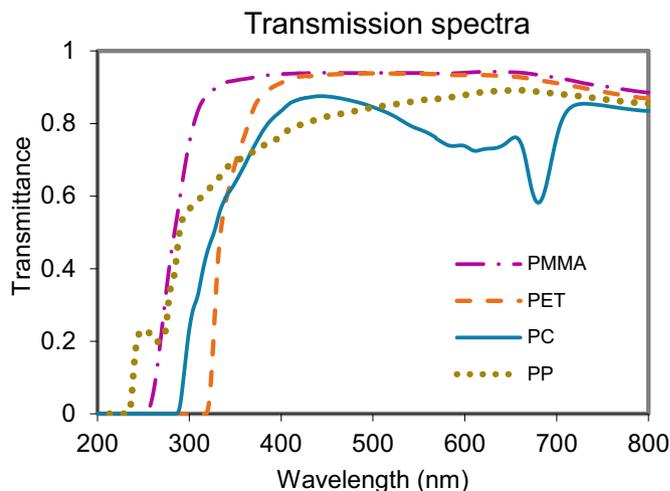


Fig. 1. Transmission spectra of PMMA, PP, PC and PET.

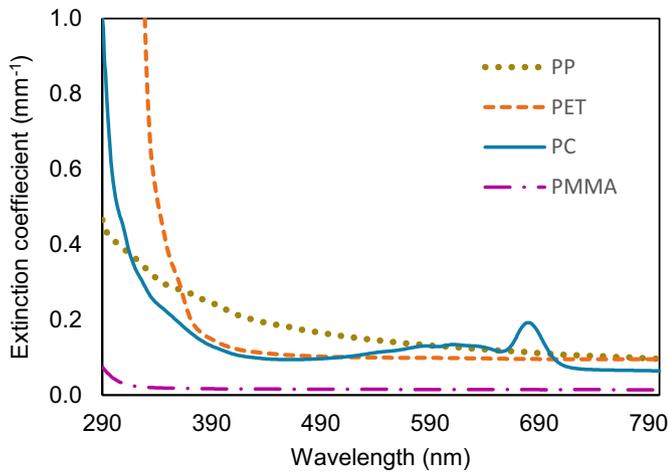


Fig. 2. Spectral distribution of the experimental extinction coefficients for the studied polymers.

PMMA from 250 nm and PC from 290 nm. At first sight, PP is more transparent than PMMA and both of them are more transparent than PC. However, the solar radiation reaching the Earth's surface does not include wavelengths below 290 nm, and therefore this wavelength range can be overlooked in the analysis. For wavelengths of solar spectra, transmittance is higher in the case of PMMA and therefore better for manufacturing SODIS devices than PP, followed by PC. Nevertheless, all three polymers were found to be suitable plastics for SODIS process considering optical properties, since they transmit UVB and consequently, cause the inactivation of protozoa, viruses and bacteria. PET blocks UVB radiation transmission and, as a result, viruses and protozoa inactivation is difficult (Busse et al., 2019).

3.3. Solar UV Calculator tool

Both mechanical and optical properties, critical for manufacturing SODIS devices, are directly related to the wall thickness: the higher thickness, the higher resistance to deformation or breakage and the lower transmission. Consequently, it is necessary to achieve a compromise between both of them to estimate the optimal thickness for the walls of the device. The developed *Solar UV Calculator* allows the assessment of the impact of thickness on the transmission of radiation. The tool estimates the available radiation inside device using Eq. (4),

where Th is the thickness of the device and $\beta(\lambda)$ is the extinction coefficients as a function of wavelengths for every specific plastic. The extinction coefficients ($\beta(\lambda)$) for the candidate plastics were obtained using Eq. (3) and are shown in Fig. 2.

PET only allows the transmission of visible and UVA radiation due to the high values of extinction coefficient for wavelengths between 290 and 320 nm (UVB radiation). PC, PP and PMMA all allow transmission of UVB, UVA and visible radiation. For wavelengths lower than 290 nm, the dramatic increase of the extinction coefficients indicates that this radiation is completely absorbed. However, this fact is hardly significant since the solar spectrum reaching the Earth surface does not include wavelengths below 290 nm. In contrast, for wavelengths higher than 290 nm, the values of PP and PC extinction coefficients are higher than that of PMMA. Therefore, the PMMA will transmit more solar radiation than PP and PC plastics for equal thickness.

The *Solar UV Calculator* has been evaluated and tested for two different purposes: i) to evaluate design parameters such as the thickness; and, ii) to estimate experimental requirements of the SODIS process, such as the solar exposure time.

3.3.1. Evaluation of design parameters

The thickness of the plastic material is determined primarily by mechanical resistance considerations but also by the device size/volume. The thickness of the plastic material should provide enough resistance to carry the water inside. However, the thickness of the plastic material has a critical effect on the optical properties of the material and therefore on the solar radiation available inside the device for SODIS purposes. When thickness increased, the transmittance decreased for all polymers. However, due to the differences in the extinction coefficients, the influence of the thickness on the available radiation inside device is very dependent on the material. In order to show the importance of estimating the transmitted radiation before manufacturing, predictions were performed with devices with a typical thickness of 2 mm for high-capacity devices (20 L) and 0.5 mm for low-capacity bottles (2 L). For a thickness of 0.5 mm the incident radiation (transmitted radiation) seems to be very similar for each polymer (Fig. 3). However, from the numerical data shown in Table 2 it follows that PET does not transmit UVB radiation and this is a critical factor for protozoa and viruses inactivation. PMMA transmits a high percentage of the solar radiation UVB (97%) and UVA (97.9%), while the behavior of PP and PC is very similar transmitting around 65% of UVB radiation and 70–80% of UVA solar radiation. However, when the thickness is increased up to 2 mm, transmitted UVB radiation decreases strongly to 17% in the case of PP and PC. Even though only 0.26–0.29 W/m² of the UVB is available

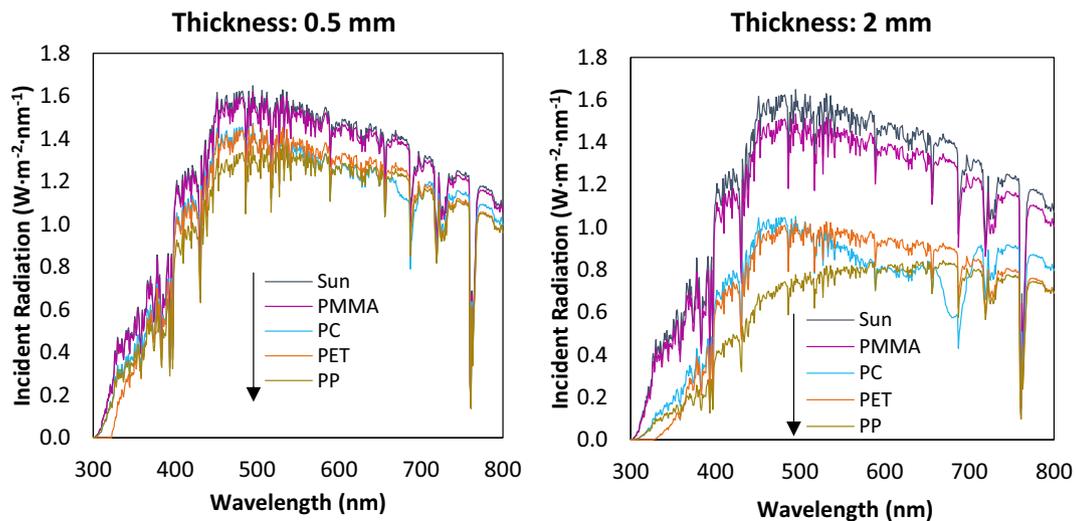


Fig. 3. Spectral incident/transmitted radiation for a thickness of 0.5 mm (left) and 2 mm (right). The arrow indicates the order of the lines from the top to the bottom.

Table 2
UVB, UVA, Visible and total incident/transmitted radiation for thickness values (Th) of 0.5 mm and 2 mm.

	Solar	PP				PC			
	(W·m ⁻²)	Th = 0.5 mm		Th = 2 mm		Th = 0.5 mm		Th = 2 mm	
		(W·m ⁻²)	%						
UVB (280–320 nm)	1.63	1.06	64.9	0.26	17.7	1.05	64.5	0.29	17.5
UVA (320–400 nm)	45.6	33.2	72.8	12.9	28.3	37.05	81.0	20.1	44.0
Visible (400–800 nm)	543	462	85.2	288	53.0	480	88.3	333	61.3
UV-Visible (280–800 nm)	590	497	84.1	301	51.0	518	87.7	353	59.8
	Solar	PMMA				PET			
	(W·m ⁻²)	Th = 0.5 mm		Th = 2 mm		Th = 0.5 mm		Th = 2 mm	
		(W·m ⁻²)	%						
UVB (280–320 nm)	1.63	1.59	97.0	1.45	88.5	0.00	0	0.00	0
UVA (320–400 nm)	45.6	44.7	97.9	42.0	92.0	31.9	70.0	14.5	31.8
Visible (400–800 nm)	543	534	98.3	506	93.3	484	89.1	342	62.9
UV-Visible (280–800 nm)	590	580	98.2	550	93.2	516	87.3	356	60.3

inside the device, it is still a useful result due to its potential to inactivate protozoa and viruses. The transmitted UVA radiation also decreases, reaching values similar to the transmitted UVA radiation by PET with 2 mm of thickness. In contrast, PMMA behavior hardly changes due to its low extinction coefficient spectra maintaining high levels of UVB and UVA incident radiation (88.5% and 92.0% of the solar irradiance).

To sum up, PMMA, PP and PC transmit UVB radiation and a higher percent of UVA; however, the thickness must be taken into account due to the fact that a small increase in the thickness can cause a disproportionately large decrease of the radiation available for SODIS inside the device.

3.3.2. Estimation of experimental requirements for SODIS applications

A lethal UV dose must be attained inside the device during SODIS in order to achieve complete disinfection. Moreover, the susceptibility of pathogens to be photoinactivated is wavelength-dependent; therefore, the lethal UV dose required is different for each pathogen (Mattle et al., 2015; Silverman and Nelson, 2016; Busse et al., 2019).

3.3.2.1. MS2 virus. As it was previously explained, in general, viruses only can be photo-damaged by UVB radiation. Fisher et al. (2012) found that MS2 needed a lethal UVB lethal dose of 12.83 kJ/m² (0.31 W/m² during 11.5 h) to achieve three-log inactivation under sunlight conditions using polypropylene copolymer (PCCO) bags transparent to the complete UV region. Taking into account Eq. 6, kinetic constant of the virus inactivation was estimated to be 5.38 · 10⁻³ cm²/mj.

Using the results of the Solar UV Calculator, the kinetic model was employed to estimate the required solar exposure time to ensure 3-log inactivation of the virus for a minimum thickness of 0.5 mm. In the case of PP and PC, the exposure time was very similar (3.36 and 3.40 h, respectively) due to its similar transmission in the UVB region. For PMMA, disinfection is achieved in only 2.24 h as a result of its low extinction coefficient and its high transmission in the UVB range. Finally, since PET does not transmit UVB radiation, the viruses cannot be inactivated by solely photoactivated processes in PET devices (Table 3). The maximum thickness of the devices to guarantee virus inactivation for an exposure time of 6 h was also calculated. As expected, in the case of PP and PC devices the thickness was almost the same (1.17 mm and 1.16 mm). For PMMA devices, the thickness reached a value of 16.8 mm, a value extremely high that assures resistance and

durability. Again, the opacity of PET to UVB radiation range makes us conclude that PET cannot be considered a good candidate for device manufacture if only photoinactivation is considered (Table 4). The available radiation inside each device for this scenario is shown in Fig. 4.

3.3.2.2. Cryptosporidium parvum protozoan. When UVB is present in the solar spectrum, protozoan photoinactivation is mainly mediated by these wavelengths. Abeledo-Lameiro et al. (2017) found that under sunlight conditions, *C. parvum* viability decreases to 35% of its original value with a UVB dose of 32.29 kJ/m² (5 h at an average UVB radiation of 0.78 W/m²). Fitting to a first order kinetic model (Eq. (6)), the kinetic constant was 3.25 · 10⁻⁴ cm²/mj. The thickness and the exposure time effect were also evaluated for *C. parvum* photoinactivation. PET was not evaluated due to the opacity to the UVB radiation. Although UVA radiation can also inactivate the protozoa slowly, a synergistic temperature contribution (above 37 °C) is then required (Gómez-Couso et al., 2010), since the temperature of the experiments of Abeledo-Lameiro et al. (2017) was always below 35 °C. For a reduction in *C. parvum* oocyst global viability of up to 50% with a thickness of 0.5 mm, the exposure time was determined, resulting in 5.58, 5.64 and 3.72 h for the PP, PC and PMMA, respectively (Table 3). The required solar exposure time of PP and PC devices is very close to the “standard” SODIS time of 6 h. The thickness required to achieve a 50% the oocyst viability in 6 h was determined. The maximum thicknesses of devices were 0.58 mm for PP, 0.57 mm for PC and 8.24 mm for PMMA (Table 4). Again, as a result of the high UVB transmission of PMMA, *C. parvum* is inactivated quicker when the water is exposed in PMMA than in PP and PC devices, which have similar transmittances and, therefore, similar thickness to achieve the same oocyst viabilities. Available radiation inside each device for this scenario is shown in Fig. 5.

Table 3

Required solar exposure time (h) with a thickness of 0.5 mm to reach three-logarithmic units inactivation of MS2 virus and *E. coli* bacterium and 50% viability of *C. parvum* oocysts.

	PP (h)	PC (h)	PMMA (h)	PET (h)
MS2	3.36	3.40	2.24	–
<i>C. parvum</i>	5.58	5.64	3.72	–
<i>E. coli</i>	0.97	0.90	0.69	1.52

Table 4

Maximum thickness (mm) to reach three-logarithmic units inactivation of MS2 virus and *E. coli* bacterium and 50% viability of *C. parvum* oocysts in 6 h.

	PP (mm)	PC (mm)	PMMA (mm)	PET (mm)
MS2	1.17	1.16	16.8	–
<i>C. parvum</i>	0.58	0.57	8.24	–
<i>E. coli</i>	3.10	4.22	47.2	2.47

3.3.2.3. Escherichia coli bacterium. Bacteria can be inactivated with UVB and UVA solar radiation so both radiation ranges must be considered for determining the SODIS inactivation time Lawrie et al. (2015) found diverse UV lethal doses for the inactivation of *E. coli*, *Enterococcus* spp. and *Clostridium perfringens* in plastic bags (PE and PET) with different transmission spectra. For this reason, it is necessary to know the sensitivity coefficient of the pathogen for each wavelength $P(\lambda)$ for determining its wavelength contribution ($m^2 \cdot W^{-1} \cdot h^{-1}$), Silverman and Nelson (2016) developed a biological weighting function (BWF) to estimate the sunlight inactivation rates of *E. coli*. Therefore, Eq. (6) is modified as follows:

$$\ln\left(\frac{C}{C_0}\right) = -P(\lambda) \cdot I(\lambda) \cdot t \tag{8}$$

Considering the sensitivity coefficients from Silverman and Nelson (2016) and the developed tool, Eq. (9) (derived from Eqs. (6) and (8)) was applied to the two scenarios to estimate the effect of the thickness, plastic material and exposure time in the *E. coli* inactivation by SODIS technology.

$$\ln\left(\frac{C}{C_0}\right) = -P(\lambda) \cdot I_{sun}(\lambda) \cdot 10^{(-Th\beta(\lambda))} \cdot t \tag{9}$$

The solar exposure time required to ensure a 3-log inactivation for a thickness of 0.5 mm was very similar for PP and PC (0.97 and 0.90 h, respectively) as a result of their similar transmission across the whole UV region. Inactivation using PMMA was achieved in only 0.69 h due to its low extinction coefficient and its high transmission in the UV range. Finally, PET cannot transmit UVB radiation, and UVA slowly inactivates bacteria. In this case, 1.52 h were required to achieve a three-log inactivation (Table 3). The maximum thickness of the devices has also been

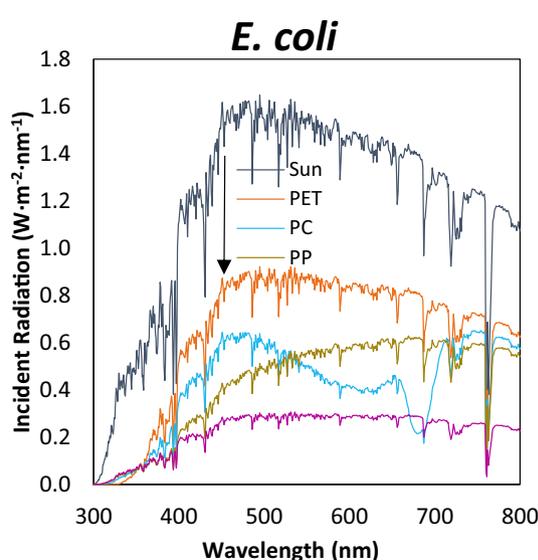
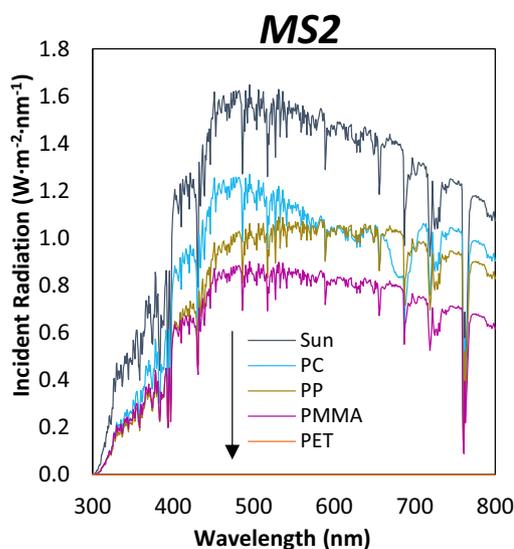


Fig. 4. Spectral incident radiation required to reach three-logarithmic units inactivation of MS2 virus and *E. coli* bacterium in 6 h for the maximum thickness values. The arrow indicates the order of the lines from the top to the bottom.

C. parvum

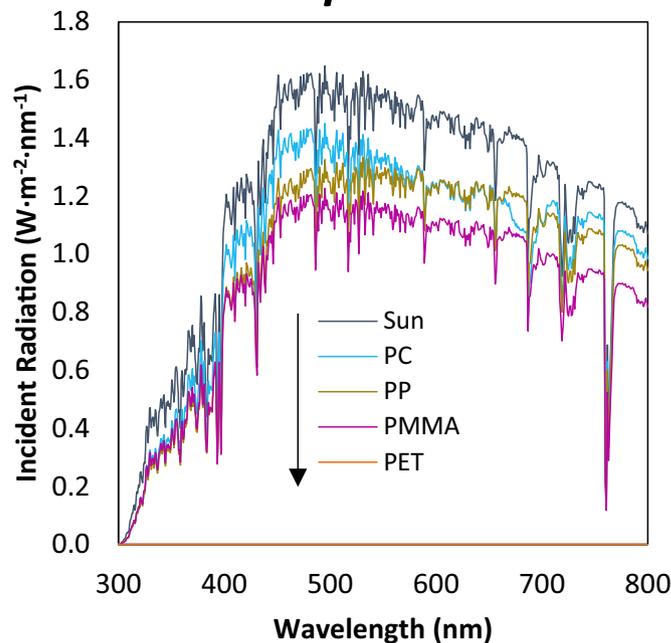


Fig. 5. Spectral incident radiation required to reach 50% viability of *C. parvum* oocysts in 6 h for the maximum thickness values. The arrow indicates the order of the lines from the top to the bottom.

calculated for a three log-inactivation in an exposure time of 6 h. Obviously, the widest thickness allowed is for PMMA material (47.20 mm) due to its high UV transmission in the UVA and UVB range, followed by thickness of PP and PC (3.10 mm and 4.22 mm) because of the combination of moderated UVA and UVB transmission, and finally maximum thickness for PET was 2.47 mm as a result of only UVA transmission (Table 4). Available radiation inside each device for this scenario is shown in Fig. 4.

To sum up, considering the solar exposure time and plastic thickness, PMMA provides the best results to be used as device for SODIS process. However, PC and PP also meet the requirements for solar water

disinfection due to the fact that they are able to transmit UVB radiation and therefore, bacteria, viruses and protozoa can be inactivated. Finally, if considering solely endogenous and non-thermal processes, then PET devices are suitable only for bacterial photoinactivation.

4. Conclusions

PP, PC, PMMA and PET have been shown to be suitable candidate materials for solar water disinfection devices: PP has low durability but can be economically replaced, PET and PC have moderate durability and production costs, and PMMA is an expensive plastic but relatively unaffected by weathering. From the optical point of view, with the exception of PET in the UV range, they all achieve the required UV transmission.

The *Solar UV Calculator* presented in this work has been proved to be a useful tool to evaluate design parameters and estimate experimental requirements for solar water disinfection in suitable plastic devices. Taking into account the thickness and the spectral transmission of the materials, the tool allows the estimation of the radiation available for SODIS inside the device and especially its spectral distribution, since the sensitivity of pathogens are strongly dependent on the wavelength range. The *Solar UV Calculator* was able to evaluate design parameters such as thickness, type of plastic, and radiation spectra. For example, for a thickness of 0.5 mm, the transmitted solar UV-A radiation was 97.9%, 70%, 72.8% and 81.0% for PMMA, PET, PP and PC, respectively, whereas the transmitted solar UV-B radiation was 97%, 0%, 64.9% and 64.5% for the same plastics. Furthermore, PMMA was found to be less sensitive to changes in thickness due to its low extinction coefficients, thus, the radiation transmitted remains high. The *Solar UV Calculator* was an essential tool for the estimation of the experimental requirements such as solar exposure time or inactivation level for three model pathogens (*E. coli* bacterium, MS2 virus and *C. parvum* protozoon). As an example, for a thickness of 0.5 mm and a 3-log inactivation of bacteria, the required solar exposure time was determined at 0.69 h for PMMA, 1.52 h for PET, 0.97 h for PP and 0.90 h for PC.

Finally, although the *Solar UV Calculator* has been validated for the specific materials and pathogen scenarios used in this study, it is freely available as Supplemental Data to any potential user interested in the evaluation of SODIS processes with other materials and conditions, or even to the evaluation of other solar processes subjected to a strong spectral dependence on materials transmission.

CRedit authorship contribution statement

Ángela García-Gil: Investigation, Writing - original draft. **Cristina Pablos:** Investigation, Methodology, Writing - review & editing. **Rafael A. García-Muñoz:** Methodology, Validation, Writing - review & editing. **Kevin G. McGuigan:** Funding acquisition, Validation, Writing - review & editing. **Javier Marugán:** Conceptualization, Funding acquisition, Validation, Writing - review & editing.

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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Appendix A. Supplementary data

The developed Solar UV Calculator is made freely available for anybody interested in performing their calculations with user-defined materials defined by its transmission spectra.

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