Time of protection equivalence principle to allow design barrier layers for promoting the use of recycled materials for food contact

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Abstract

European and national environmental protection policies have programmed the forthcoming end of single-use plastics, including food packaging. Because plastic materials account for 50% of plastic waste, closed-loop recycling seems imperative. Still, plastics other than PET (Polyethylene terephthalate) are not widely recycled for food contact due to safety concerns. Among them, post-consumer polyolefins are heavily formulated, degraded, and contaminated by the previously contacting product. Using recycled materials behind a functional barrier (FB) could resolve the issue under specific provisions, but neither the European regulation nor the US FDA offer guidance concerning functional barriers evaluation and use. Mathematical modeling is the only viable method recognized by European and American agencies to evaluate recycled material under usage conditions. This study numerically explores the possibility of combining two effects: decontamination of the recycled material and a functional barrier to allow safe use of the recycled material.

1-Introduction

The circularity of food packaging is part of the European Green Deal pact and Directive (EU) 2019/904. The end of single-use plastic packaging is planned in Europe. In France, the new anti-waste law N°220-105 imposes the return to food contact of all plastic packaging from January 1st 2025. Recycled materials intended to be in contact with food must be as inert as virgin food-contact materials, as stated in the EU Regulation (EC) 1935/2004 and must comply with a strict evaluation of recycling processes by the European Food Safety Authority (regulation (EC) 282/2008). According to the European Commission, the current rules are not favorable to the development of the circular economy (Ellen MacArthur Foundation 2020). Nowadays, only PET is authorized globally within the European Union. The polymer is relatively low formulated and glassy, all grades are initially suitable for food contact and contamination levels in post-consumer PETs remain low (3 mg/kg value accepted as a maximum (Franz and Welle 2020)). Nevertheless, recycled PET alone cannot replace all packaging applications. It is appropriate for packaging beverages in bottles (water, sodas, acid juices) but not adapted to applications requiring hot filling, pasteurization, sterilization, and vacuum cooking. The reason is that the glass transition temperature of PET is between 76°C and 81°C, which means that it cannot be used above 72°C. Other polymers, particularly polyolefins, isotactic polypropylene, and high-density polyethylene, should be used to cover at least 2/3 of the current plastic packaging applications. They have extremely varied origins, they are highly formulated, and only additives belonging to the positive list of the EU regulation (EC) 10/2011 and not causing excessive migration are authorized for food contact. Post-consumer polyolefins are more contaminated by chemicals and higher molecular weight compounds, with a decreasing efficiency of decontamination (vacuum evaporation) as the molecular weight increases (Palkopoulou *et al*. 2016). Therefore, they have far less favorable opinions from the European Food Safety Authority (EFSA) and the US FDA (Geueke *et al.* 2018). Reconciling environmental and health safety objectives requires exploring alternative uses for recycled material, particularly behind a functional barrier. The concept of a functional barrier is initially based on the idea of a diffusion barrier that delays migration without changing the final fate (Feigenbaum. 2005). This barrier is not absolute but only temporary. It must be appropriately chosen and scaled to ensure food safety. Modeling tools are already widely used in Europe, USA, and China. They are recognized in European (Hoekstra *et al.* 2015) and American (Schwope *et al.* 1990) practical guides and their state of the art has been taken up in the reference book Zhu *et al.* (2019). The use of a functional barrier is already accepted in the European plastic regulation (regulation (EC) 10/2011). The difficulty for the plastic material is related to contaminants whose nature, toxicity, and quantities are not known a priori. The analysis must be generic and robust.

The working hypothesis is based on a dimensionless formulation that allows comparing a solution with or without a barrier for various levels of decontamination. This approach potentially covers all thermoplastics, substances, geometries, and uses. The novelty of the described concept lies in the fact that a lower decontamination rate is envisaged in the presence of a functional barrier when using non-authorized materials for food contact. The text is organized as follows: the next part presents the mathematical model and the approximate solutions and the description of a case-study. The results are discussed in the third section. In particular, the aim is to show solutions where the barrier does not add waste and discuss the barrier's performance. The conclusions are finally summarized.

2-Materials & Methods

Acceptable thresholds

EFSA set rules to evaluate food contact materials regarding the risk of exposure to non-evaluated substances. Post-consumer recycled materials are a significant source of unknown chemicals potentially harmful. An acceptable level of exposure can be established according to analytical, chemical structure analogy (e.g., Cramer class) or a regulation threshold (e.g., the US TOR defined under 21 CFR 170.39), as summarized in Table 1.The most conservative thresholds have been jointly defined by EFSA and WHO and are coined thresholds of toxicological concern (TTC). For genotoxic substances, the TTC value corresponds to a maximum chronic daily exposure of 0.0025 $\mu g^{-1}.kg^{-1}_{BW}.day^{-1}$. Its conversion into maximum acceptable concentration in food reads:

$$C_{thresh}^{TTC,genotoxic} = 0.0025\ \mu g.kg_{body\ weight}^{-1}.day^{-1} * \frac{body\ weight}{daily\ intake} \tag{1}$$

A bodyweight of 60 and 5 kg is taken for adults and infants, respectively, with 1 and 0.75 kg daily intake respectively (Barthélémy et al. 2014). As discussed by Zhu *et al.* (2019), the $C_{thresh}^{TTC,genotoxic}$ value of 0.15 mg·kg^{-1} for adults is far below the detection limit of the most commonly used analytical techniques, around 10 mg·kg^{-1}. Only modeling and numerical simulation can demonstrate that this threshold is not exceeded for low molecular weight surrogates. Current scenarios that EFSA considers use concentrations in recycled streams of 3 and 10^3 mg·kg^{-1} as conservative values for PET and polyolefins, respectively.

Table 1: Applicable TTC's for food contact according to the type of consumer.

TTC	TTC adult (µg/kg of food)	TTC infant (µg/kg of food)
With genotoxicity	0.15	0.017
TOR	1	
Detection Limit	10	
Organophosphates and carbamates	18	2
Cramer class III	90	10
Cramer class II	540	60
Cramer class I	1800	200

Mass transfer model

Without a loss of generality, the studied cases described mass transfer between recycled material (layer indexed $j = 2$) and food (layer indexed $j = 0$) separated by a functional barrier with an index $j = 1$. Mass transfers are described in one dimension, as shown in Figure 1. The thicknesses are denoted $\{l_j\}_{j=0,1,2}$ with an equivalent food thickness $l_0 = \frac{V_F}{A}$, where V_F is the food volume and A is the surface area in contact.

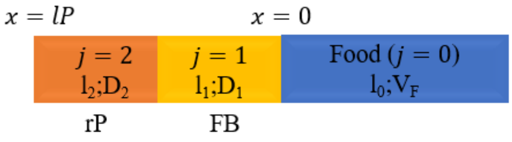


Figure 1: Layer indexing for mass transfer modeling.

The transport model of contaminants is like the one used in (Nguyen et al., 2013) and (Zhu *et al*. 2019). It fulfils the assumptions recommended to evaluate the compliance of plastic materials intended to be in contact with food according to the European Regulation (EC) 10/2011. It assumes a diffusion-solubilization model in each layer and the food with a Henry-like isotherm with a constant Henry coefficient k_i and a diffusion coefficient D_i. By denoting the cumulated thicknesses: $l_0^\Sigma = 0$, $l_1^\Sigma = l_1$, $l_2^\Sigma = l_1 + l_2$, and the concentration profile $c_{x,t}$, the transport assumptions read in the polymer:

$$\frac{\partial c_{x,t}}{\partial t} = D_i \frac{\partial^2 c_{x,t}}{\partial x^2} \; for \; l_{i-1}^\Sigma < x < l_i^\Sigma \; and \; i = 1,2$$

$$D_1 \left.\frac{\partial c_{x,t}}{\partial x}\right|_{x=l_1^\Sigma-\text{Ò}} = D_2 \left.\frac{\partial c_{x,t}}{\partial x}\right|_{x=l_1^\Sigma+\text{Ò}} \; with \; \text{Ò} \; \rightarrow 0 \; and \; t > 0 \tag{2}$$

$$\frac{c_{x=l_1^\Sigma-\text{Ò},t}}{c_{x=l_1^\Sigma+\text{Ò},t}} = \frac{k_2}{k_1} \; with \; \text{Ò} \; \rightarrow 0 \; and \; t > 0 \; and \; \left.\frac{\partial c_{x,t}}{\partial x}\right|_{x=l_2^\Sigma} = 0 \; when \; t > 0$$

An implicit model is used for the food layer ($j = 0$) and implemented as boundary layer of the third kind, also known as Robin boundary condition at the food-packaging interface ($x = 0$):

$$j = -D_1 \left.\frac{\partial c_{x,t}}{\partial x}\right|_{x=-\epsilon} = h_m\left(c_{x=\epsilon,t} - C_F^t\right) = h_m\left(\frac{k_1}{k_0} c_{x=-\epsilon,t} - C_F^t\right) \text{ with } \epsilon \rightarrow 0, \;\; C_F^t = C_F^{t=0} + \frac{1}{l_0}\int_0^t j \cdot dt \tag{3}$$

This condition enables to account for the accumulation in the food and the partition coefficient between the food in contact and layer 1 $\left(\frac{k_1}{k_0}\right)$. Far from the interface, the concentration in the food, C_F^t, is assumed to be uniform. Close to the interface, h_m is the mass transfer coefficient, with units in $m^2 \cdot s^{-1}$ and representing the mass transfer resistance on the food side. The amount within the boundary layer is assumed to be negligible compared to C_F. The initial concentration in the recycled layer at $t = 0$ is given by Equation (4) where $0 \leq \Delta < 1$ is the decontamination rate and C_P^{max} the maximum concentration in the source of recycled materials.

$$c_{x,t=0} = (1 - \Delta) C_P^{max} \; for \; l_1^\Sigma < x < l_2^\Sigma \tag{4}$$

Dimensionless mass transfer model

Migration from layer 2 to layer 1 starts during processing, as we showed (Dole et al., 2006), but coupling heat and mass transfer complicates the extrapolation of results from specific cases to more general ones. The difficulty can be partly circumvented by introducing a dimensionless formulation and cumulative time effects, as shown in (Zhu et al., 2019). The dimensionless time can accommodate the variation of D_2 with time due to temperature:

$$Fo = \int_0^t \frac{D_2(t)dt}{l_2^2} \tag{5}$$

By introducing the dimensionless position $x^* = \frac{x}{l_2}$, concentration $c^* = \frac{c_{x,t}}{C_P^{max}}$ and diffusivities $D_i^* = \frac{D_i}{D_2^{t\to\infty}}$, the two first system equations become:

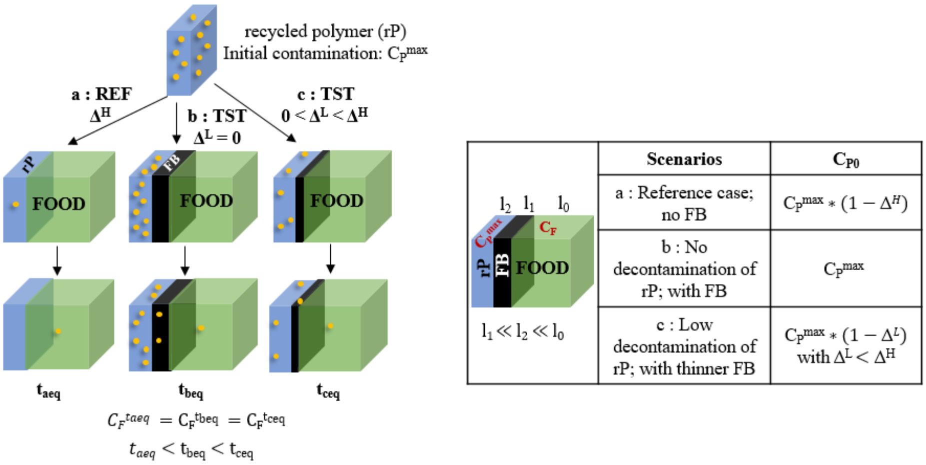
$$\frac{\partial c^*}{\partial Fo} = D_i^* \frac{\partial^2 c^*}{\partial x^{*2}} \; for \; \frac{l_{i-1}^{\Sigma}}{l_2} < x^* \leq \frac{l_i^{\Sigma}}{l_2} \; and \; i = 1,2 \tag{6}$$

$$D_1^* \frac{\partial c^*}{\partial x^*}\bigg|_{x^*=\frac{l_1^{\Sigma}}{l_2}-\grave{o}} = D_2^* \frac{\partial c^*}{\partial x^*}\bigg|_{x^*=\frac{l_1^{\Sigma}}{l_2}+\grave{o}} \; with \; \grave{o} \to 0 \; and \; Fo > 0$$

By introducing the dimensionless mass Biot number $Bi = \frac{h_m l_2}{D_2^{t\to\infty}}$ and by considering $C_F^{t=0} = 0$, the food boundary condition becomes:

$$j^* = -D_1^* \frac{\partial c^*}{\partial x^*}\bigg|_{x^*=-\grave{o}} = B_i\left(\frac{k_1}{k_0} c_{x=\grave{o}}^* - C_F^*\right) \; with \; \grave{o} \to 0 \; and \; C_F^* = \frac{C_F^t}{C_{Pmax}} = \frac{l_2}{l_0}\int_0^{Fo} j^*.dFo \tag{7}$$

Food contamination is maximized when $\frac{k_1}{k_0} = \frac{k_2}{k_0} \to \infty$. This condition corresponds to a functional barrier acting only as a diffusion barrier (no solubility barrier) and a much higher chemical affinity for the food.



Scenarios	C_{P0}
a : Reference case; no FB	$C_P^{max} * (1 - \Delta^H)$
b : No decontamination of rP; with FB	C_P^{max}
c : Low decontamination of rP; with thinner FB	$C_P^{max} * (1 - \Delta^L)$ with $\Delta^L < \Delta^H$

Figure 2: Studied scenarios with decontamination only REF; with decontamination and FB TST. ($C_{P_{max}}$: Initial concentration in the packaging; C_F: Concentration in food; $\Delta_{\{H,L\}}$: Decontamination level with H: High and L: Low; t_{eq}: Time at which concentration in food is the same in each system a,b,c).

For a given recycled stream (the same C_P^{max}) and plastic design (same l_2 and D_2 values), different values of C_F^* are therefore expected for different decontamination rates Δ and functional barrier thicknesses l_1. Two situations (see Figure) are particularly relevant, denoted REF (reference) and TST (alternative):

- REF: No barrier ($l_1 = 0$) but a decontamination level, Δ^H.
- TST: Barrier ($l_1 > 0$) but a lower decontamination level, $\Delta^L < \Delta^H$.

The reference situation $C_F^*(\Delta, l_1 = 0)$ accepts a trivial approximate solution from Eq. 4.20 in Crank's book (Crank, 1975):

$$C_{F,REF}^* \approx min\left(2\sqrt{\frac{Fo}{\pi}}, 1\right)\frac{1-\Delta^H}{l_0^*} \tag{8}$$

The situation $\boxed{\text{TST}}$ verifies $\boldsymbol{C_{F,TST}^*} < \boldsymbol{C_{F,REF}^*}$ when $Fo \ll Fo_{lag}$ with Fo_{lag} given by: $Fo_{lag} = \frac{l_1^*}{6D_1^*}$

When $Fo > Fo_{lag}$ and $l_1 \ll l_2$, a rough solution of $\boxed{\text{TST}}$ is given by:

$$C_{F,TST}^* \approx min\left(\frac{D_1^*}{l_1^*}\left(Fo - Fo_{lag}\right), 1\right)\frac{1-\Delta^L}{l_0^*} \tag{9}$$

From Eqs. (8) and (9), the condition $\boldsymbol{C_{F,REF}^*} = \boldsymbol{C_{F,TST}^*}$ corresponds to:

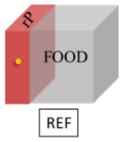
$$Fo \approx \frac{\left(l_1^* + \sqrt{{l_1^*}^2 + Fo_{lag}\pi\left(D_1^*\frac{1-\Delta^L}{1-\Delta^H}\right)^2}\right)^2}{\pi\left(D_1^*\frac{1-\Delta^L}{1-\Delta^H}\right)^2} \xrightarrow[Fo_{lag}\to 0]{} \frac{4{l_1^*}^2}{\pi\left(D_1^*\frac{1-\Delta^L}{1-\Delta^H}\right)^2} \xrightarrow[\Delta^L\to\Delta^H]{} \frac{4}{\pi}\frac{{l_1^*}^2}{{D_1^*}^2}\ such\ that\ C_{F,REF}^* = C_{F,TST}^* \tag{10}$$

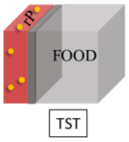
As a rule of thumb, ${l_1^*}^2/D_1^*$ should be chosen greater than $\left(\frac{1-\Delta^L}{1-\Delta^H}\right)^2$. All effects are non-linear and should be explored rigorously before proposing a solution for a given polymer or application. The system is also strongly constrained. Since it is preferable to minimize the thickness of the barrier ($l_1^* < 1$) to minimize wastes, D_1^* should be also lower than 1 ($D_1^* \ll l_1^*$). That is to say that the functional barrier should be chosen among the best polymer or coating barriers and not interfere with the subsequent recycling of the material.

Studied case and simulation methodology

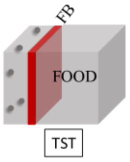
The studied case corresponds to 100µm thick PP trays covered internally with a 10µm PET layer. The simulation details are shown in Table 2. The considered contaminants of recycled PP are homologous aromatic surrogates: toluene, biphenyl and p-terphenyl, with a 1000 mg.kg^{-1} initial concentration and decontamination levels (Δ^H, Δ^L) range from 0% up to 99.9% (vacuum evaporation). Toluene is used as the reference evaluation substance with an acceptable threshold set to $C_{thresh}^{TTC,genotoxic}$.

Table 2: Parameters considered for simulations (TOL: toluene, BIP: biphenyl, TERP: terphenyl)





Layer 2: recycled polypropylene, 40°C				Layer 1 (FB): dry PET (A), 40°C				Layer 1 (FB): plasticized PET (B), 40°C			
α(T, Tg)	6 (b)			α(T, Tg)	7 (c)			α(T, Tg)	7 (c)		
l(µm)	100 (e)			l(µm)	10 (e)			l(µm)	10 (e)		
D(m².s^{-1})	TOL	BIP	TERP	D(m².s^{-1})	TOL	BIP	TERP	D(m².s^{-1})	TOL	BIP	TERP
	10^{-13} (a)	2.7e^{-15} (b)	1.6e^{-16} (b)		3.5e^{-18} (c)	1.6e^{-19} (b)	1.4e^{-20} (b)		5e^{-16} (c)	2.2e^{-17} (b)	2e^{-18} (b)
C_0(µg.kg^{-1})	10^6 (e)			C_0(µg.kg^{-1})	0			C_0(µg.kg^{-1})	0		
$\frac{k_2}{k_0}$	1 (d)			$\frac{k_1}{k_0}$	1 (d)			$\frac{k_1}{k_0}$	1 (d)		
L	10^{-2}			L	$10^{-2}*(l_1+l_2)/l_2$						
Bi	10^4 (d)										



(a) from Eun et al. 2014; (b) from Fang et al. 2013; (c) from Zhu et al. Soft Matter, 2019.
(d) conservative scenario according to Vitrac, O.,Hayert, M. (2005).
(e) Typical value for trays and bottles.

As detailed by Zhu et al. (2019), all simulations were carried out with the open-source FMECAengine (Vitrac and Nguyen 2014-2019). (Fang et al., 2013) et al. (2013) methodology was used to scale diffusion coefficients of linearly repeated aromatic jumping units according to Equation (11), M_0 refers to the molecular weight of reference solute, toluene in this case.

$$\frac{D(M,T)}{D(M_0,T)} = \left(\frac{M}{M_0}\right)^{-\alpha_{(T-Tg)}} \tag{11}$$

3-Results & Discussions

Protection time equivalence of functional barrier with direct food contact

Simulated contamination kinetics for REF and TST conditions with toluene are plotted in Figure 3a. As predicted by Eq. (10), a minimum thickness of the functional barrier can offer the same protection for the consumer as a decontamination level Δ_H (equivalent times are marked by intersections). This protection is useful and robust only if it fulfils three conditions: (i) it is lasting longer than lag time (pure diffusion barrier), (ii) it does not double the amount of wastes ($l_1^* < 1$) and (iii) $C_F < C_{thresh}^{TTC,genotoxic}$. Food shelf-life considerations are not introduced here, but it is more and more accepted to adjust the effective shelf life with the acceptable chemical risk for the consumers. For polyolefins and low molecular weight compounds (with high diffusivities), adding a functional barrier is not enough and a decontamination step is essential to get a protection equivalent to $\Delta^H > 0.99$ or more. The depicted REF condition (dark red curve) describes a thermodynamic equilibrium achieved in about 2.3 days. That is to say that the concentration in food is entirely determined by $C_P^{max}(1-\Delta^H)/l_0^*$ after 3 days. It corresponds to a value 6.7 times larger than $C_{thresh}^{TTC,genotoxic}$ for REF at the decontamination level of $\Delta^H = 0.9999$ which is the maximum value feasible realistically by the industry (for 0.999, the threshold is exceeded 67 times). The missing protection can be brought by combining a high Δ^L and a functional barrier delaying the equilibrium. As a rule of thumb and to avoid the production of extra-wastes, the virgin functional barrier (layer 1) should be more barrier than the recycled polymer (layer 2). In the depicted TST condition, lag-time is comparable to equilibration time for toluene (1.5 days) and almost negligible. The protection is brought in the permeation regime when the concentration level is almost linear with time. In this regime, the linearity highlights those ten times higher protection (10 times longer shelf-life or service-life for the material) and requires either to multiply by ten the thickness or to divide by ten the factor 1-Δ^L.

Solute size effects

For bigger solutes, simulated conditions are plotted in Figure 3 b and c. As expected, a shift appears toward higher protection time equivalences. Table 3 show a report of those equivalent times for toluene and biphenyl, according to decontamination levels ΔL for TST and ΔH for REF situation. During a certain amount of time, the TST alternative solution is even safer than the REF condition. The risk assessment of a functional barrier relies on two conditions: (i) it has to be at least as safe as a highly decontaminated monolayer (today no recycling process is authorized and able to do such a thing, this is why the use of functional barriers has a big interest) and (ii) a compromise has to be found between the condition (i) and the fact that for large contaminants there is no need to target higher protection times than small ones since they migrate faster, allowing in some applications to lower the initial decontamination of large contaminants. The existence of a substance-dependant scaling law for equivalent times aims to target the decontamination needs according to the profile and extract good dimensioning practices to avoid considering large contaminants. The thickness of the functional barrier needs to be sufficient to accommodate shelf life and the storage of the material before use.

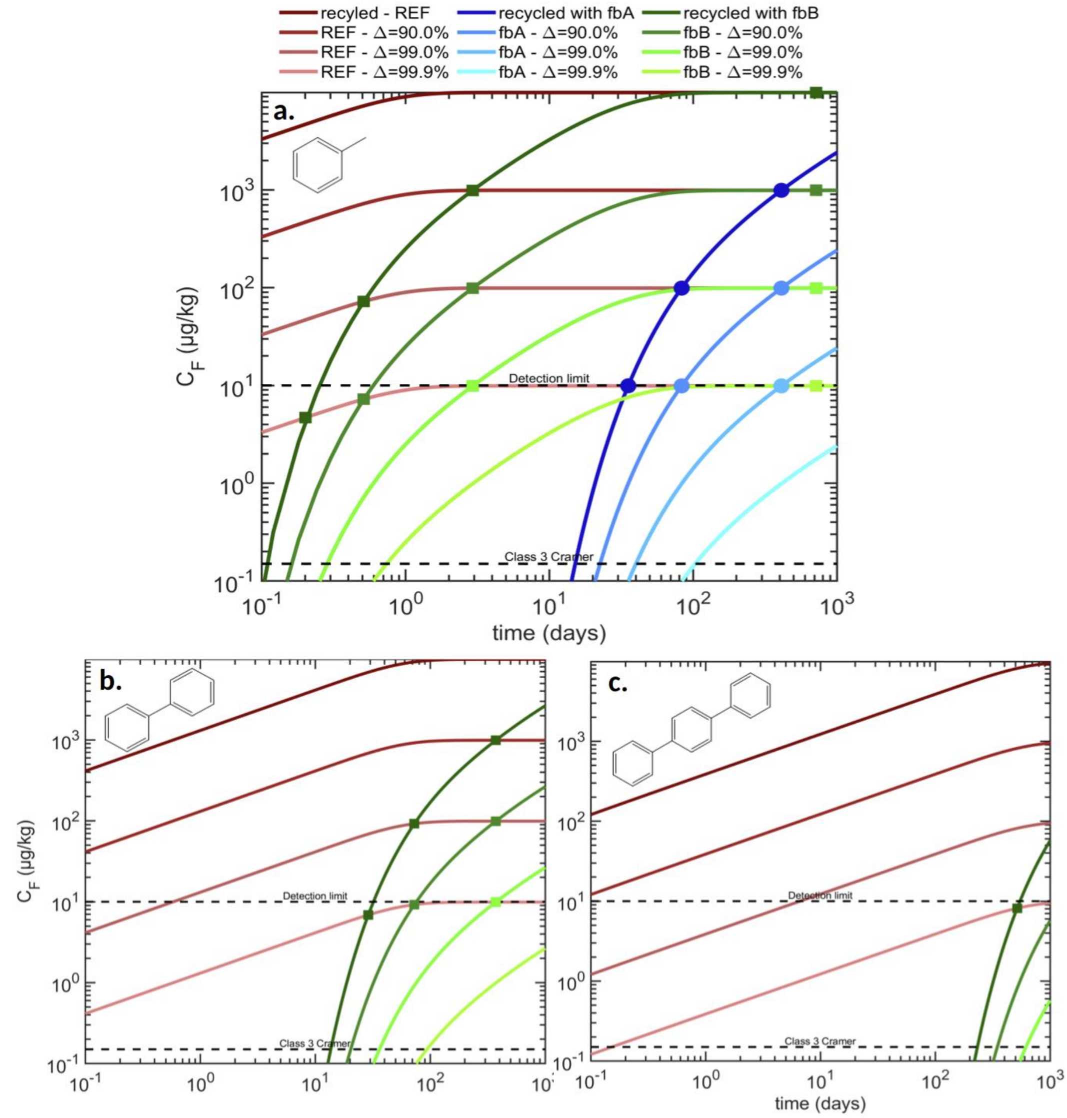
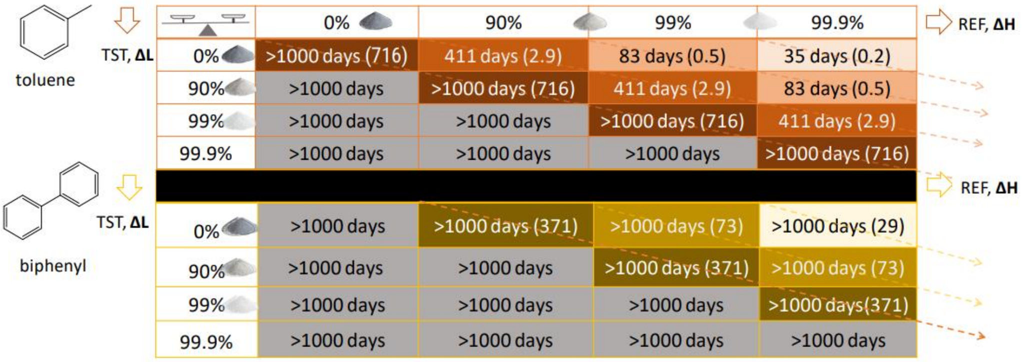


Figure 3: a-b-c: Simulated contamination kinetics of toluene, biphenyl, terphenyl, respectively for $\boxed{\text{REF}}$ and $\boxed{\text{TST}}$ conditions (blue: FB = dry PET; green: FB = plasticized PET).

Table 3: Equivalent safety times reported for simulations with toluene and biphenyl (Values in parenthesis are for plasticized PET)



	TST, ΔL	0%	90%	99%	99.9%	REF, ΔH
toluene	0%	>1000 days (716)	411 days (2.9)	83 days (0.5)	35 days (0.2)	
	90%	>1000 days	>1000 days (716)	411 days (2.9)	83 days (0.5)	
	99%	>1000 days	>1000 days	>1000 days (716)	411 days (2.9)	
	99.9%	>1000 days	>1000 days	>1000 days	>1000 days (716)	
biphenyl	0%	>1000 days	>1000 days (371)	>1000 days (73)	>1000 days (29)	
	90%	>1000 days	>1000 days	>1000 days (371)	>1000 days (73)	
	99%	>1000 days	>1000 days	>1000 days	>1000 days(371)	
	99.9%	>1000 days	>1000 days	>1000 days	>1000 days	

4-Conclusions

Conservative rules have been identified for homologous aromatic surrogates to assess the risk of using non-authorized materials for food contact behind functional barriers. The development of this concept is driven by environmental imperatives and must be supported by additional safeguards other than relative decontamination level. It addresses for the first time the idea of equivalent protection combining the decontamination of small and volatile contaminants by mechanical recycling (washing and evaporation under vacuum) and protection of food from larger contaminants by a barrier layer. The functional barrier applies to all solutes but all don't need to be decontaminated at the same rate. Multicriteria optimization and rigorous validation are in process and would be a way to cover the practical impossibility of measuring contamination at the acceptable toxicological thresholds.

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