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The impacts of plastic products on air pollution - A simulation study for advanced life cycle inventories of plastics covering secondary microplastic production

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ABSTRACT

A framework, SeqPlast, to include secondary microplastic (MP) formation due to photooxidation into current Life Cycle Assessments (LCA) from cradle-to-grave of products. The framework details how to incorporate secondary MP formation into the plastic's life cycle as related to the plastic's use phase, and location specific waste treatment and how to connect the impacts with current life cycle impact assessment methodologies (LCIA). Plastics, now ubiquitous in the environment are a potential source of emerging pollution and have been shown to have negative effects of various species. Thus, the framework consist of 1) a degradation module, which accounts for micro- and nano- plastic formation with dynamic degradation of microparticles, 2) an emissions module accounts for the potential of the plastic particles to be emitted to air, and 3) an impacts module which connects the various emissions to existing LCIA methods. The framework allows for quantification of secondary microplastic in an LCA context and for further characterization of the impacts at endpoints in terms of human health, and allows for a high level of regionalization, both in terms of input data and characterization of impact damages. SeqPlast was tested on a case study of mulch film which showed that the per kg contribution to particulate matter (PM) and other impacts is low. The impacts vary largely depending on the degradation rate and ranged from 9.24×10^{-6} to 0.00043 kg PM equivalents per kg of mulch film, depending on either a slow or fast degradation rate, respectively. The impact came mostly from the littered fraction, which was estimated to be 10% of the products weight after the use phase, in Europe. The degradation rates due to UV degradation were low, 0.034 $\mu\text{g}/\text{year}$ for a slow degradation rate, compared to values derived from the literature, 0.345 $\mu\text{g}/\text{year}$ for a fast degradation rate during littering, that included other sources of degradation, such as abrasion. However, when these impacts are scaled up to the European consumption of plastics and monetized, it is evident that even small increases of PM are costly for society and could potentially amount to millions of dollars per year in human health damages. Further research efforts should focus on filling data gaps, such as microplastic losses during production, recycling and potentially incineration, as well as degradation kinetics including other degradation factors such as freeze/thawing, wet/drying in combination with UV degradation.

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Symbol	Definition
D_{total}	The effective damage dose to a polymer material
E_o	The incident UV radiation dose on a polymer surface
ϕ	The number of instances it takes before a photon is absorbed on the polymer
t	The time of exposure
V	Volume
r	Radius
ρ	Density
i	Timestep of exposure
V_i	Volume change of a particle
V_{i-1}	Volume of particle in the previous timestep
k	Degradation rate
m	Mass
d	Diameter
j	A particle
$M_{i,j}$	Mass of particles in the given timestep
$M_{tot, PMx}$	Total mass of particles in a relevant particle size definition, PMx
$M_{tot, cum}$	Cumulative mass of particles in the particle size definition over the whole time horizon $i=n$
MP_{lift}	Lifting potential of a microplastic particle MP
V_0	Initial velocity of the microplastic
g	The gravitational constant
K	Air resistance of a particle
α	Angle of oblique roll
f	Fluid viscosity of air

1. Introduction

Plastic pollution has in the past decade come under the microscope of the public eye and the scientific community. Scientific studies examining various facets of plastic pollution have flooded the literature (Li et al., 2020). Unlike other types of pollution, the problems caused by plastics are visible, which has perhaps motivated various countries to set targets for the elimination or reduction of some plastic products, such as plastic bags and disposable items (Sonnemann and Valdivia, 2017). In May 2018, a new proposal was passed by the European Commission, banning several one-time use plastic items, and imposing stricter regulations on several other plastic products (European Commission, 2018a). According to Giacovelli, (2018) there are now 60 countries that have either banned or imposed taxes on single use plastics. This puts into evidence how important the topic has become, both in terms of public perception and political willingness to act. Yet, if plastics must be regulated due to increased evidence of negative impact (Plastic Soup Foundation, 2019), then their impacts must also be quantified. Plastics, which have been designed to optimize different material properties, such as flexibility, hydrophobicity, elasticity, and durability, among others, are well understood under laboratory conditions. However, when plastics are littered in the environment and exposed to the elements i.e. the combination of intermittent exposure to solar radiation, cooling, heating, drying, and rain, they begin to degrade, fragment and create so called microplastics. These are referred to as secondary microplastic, as opposed to primary microplastics, which are purposely produced in the micron size for use in different products e.g. microbeads in beauty products. In turn, more and more studies of micro- and nano-plastic present evidence for the negative effects that these particles can cause on marine fauna (Auta et al., 2017; Crawford and Quinn, 2017; Lambert et al., 2014). By recent estimates, the amount of plastic entering the marine environment could be around 4.8 to 12.7 million MT in 2010 and is projected to increase by an order of magnitude by 2025 in a business as usual case (Jambeck et al., 2015). Furthermore, an estimate for global mismanaged plastic predicts ca. 100 million MT per year in 2020 on average and reaching 220 million MT per year in 2060 if no preventative actions are taken to minimize the spill of plastic (Lebreton and Andrady, 2019).

Many approaches exist to measure the impacts of plastic products. Among these, impacts from the resource intensity, and waste treatment parts of the life cycle of plastic are in development, especially to address the impacts of plastic products in a circular economy (Sassanelli et al., 2019; Vinante et al., 2021). When it comes to plastic, due to their persistent nature and relevance in society, it becomes necessary to evaluate them taking into account their contribution as a source of emerging pollutants (Avio et al., 2017). In this context, the less visible facet of plastic pollution, i.e. the issues caused by microplastics (MPs) and nanoplastics (NPs), is an expanding area of study. MPs and NPs, which may be present as particles or synthetic fibers starting in the nanometer range up to 5 mm, have been described as ubiquitous in the environment, by several authors (Allen et al., 2019; Brahney et al., 2020; Lambert et al., 2014). These particles have been found everywhere from the poles to remote corners of the Pacific Ocean (Eriksen et al., 2018, 2014; Shahul Hamid et al., 2018). Their presence has been observed in a large variety of places e.g. on sea salt, honey, seafood, beer, breast milk and placenta (Liebezeit and Liebezeit, 2014, 2013; Wright and Kelly, 2017). Evidence of MPs' impact on human and ecosystem health is just beginning to emerge (Browne et al., 2008; Carbery et al., 2018; Galloway and Lewis, 2016; Rillig et al., 2017; Wright and Kelly, 2017). As more information comes to light, accounting for these impacts by means of life cycle assessment (LCA), a standardized methodology (Finkbeiner et al., 2006) designed to quantify the final damage of various substances, can be a step towards devising effective measures to regulate plastics. LCA is currently being used globally as a tool to support decision making in a policy context (Sonnemann et al., 2018). As it has moved from the academic realm, it has been adopted by for example, companies and governmental institutions, such as the European Commission's Joint Research Center, motivating various project like the LCA Initiative (Töpfer, 2002). More recently, developments that incorporate life cycle thinking in the policy sector include the UN's Sustainable Development Goals, including goal 12 for sustainable consumption and production patterns (UNEP et al., 2012). Furthermore, life cycle assessment is being used to evaluate the performance of the circular economy from a material flow analysis perspective, end of life treatment and eco-efficient design of products (Sassanelli et al., 2019). Life cycle thinking is being used to support green procurement and provide validity to greenhouse gas (GHG) claims, as it is the case for the Renewable Energy Directive recast (REDII)(European Commission, 2018b), and the Product Environmental Footprint (PEF) certification scheme. Thus, it seems a natural progression to employ LCA for quantification of potential problems due to plastics. Though LCA has come a long way, a need for further standardization is important and has been highlighted by various authors (Finkbeiner, 2013; Klöpffer, 2012). This is even more relevant when trying to quantify the multi-layered, interdisciplinary problems into which plastic develops when losses occur in nature. LCA provides a method to evaluate the effects of various planned and already in effect policies, such as the European ban on single-use plastics (European Commission, 2018a), South Korea's Plastic Waste Control Plan (), Peru's restriction of single use plastics at heritage sites, Canada's future ban on single use plastic and San Diego's ban of styrofoam containers in restaurants (Howard et al., 2019). All of these actions have impacts due to the reduction of plastic waste or the introduction of alternatives to plastic products, which can be assessed holistically with LCA, though it requires extra guidance to include the effects of emerging pollution from plastics.

To date, a few attempts at devising numerical models to track losses of plastic in the environment and provide a plastic footprint have been published (Kawecki and Nowack, 2019; Peano et al., 2020; Ryberg et al., 2019), and several methodologies are in devel-

opment for providing an inventory of plastic losses (Boucher et al., 2019). These are useful methods to derive total global quantities of plastic lost as macro-plastic and micro-plastic. However, to the best of our knowledge, most of these methodologies fail to make a connection between plastic loss and final impact, most of them do not consider secondary MP formation and none of them take into consideration the decomposition changes (size changes and gas products) that mismanaged plastic will experience throughout its lifetime and end of life (EoL). Tying secondary MP formation to global loss rates is indeed useful and needed to derive final concentrations of plastics in various environmental compartments. This in turn, allows for deriving damage indicators, such as species loss due to MP ingestion. Among the approaches, Ryberg et al., (2019) calculate global plastic losses starting from the amounts produced of each polymer type, consumption rates and waste treatments across the globe. However, secondary MP are only included for selected types of plastic i.e. for tire abrasion, road markings, city dust, weathering of marine coatings, plastic production and fibers from textile washing. Kawecki and Nowack (2019), apply a probabilistic material flow analysis approach to plastics produced worldwide and estimate microplastic based on the few reports of particles in the literature, which among other originate from construction sites, production, and bottled water. Both approaches include somewhat arbitrary estimates of the fate of secondary MP and neither of them link the particles to final impact or damage indicators. Furthermore, these studies apply a top down approach i.e. from production rates to loss (Ryberg et al., 2019) or product type to loss (Kawecki and Nowack, 2019), rather than estimating degradation kinetics for various types of polymers, excluding overwhelming evidence that MPs are formed in any polymer surface exposed to weathering (Chamas et al., 2020; Lambert et al., 2014; Vasile, 2000). Decoupling from global plastic losses and instead tying MP formation and fate to natural phenomena such as, rainfall, temperature, wind speed and sunlight hours, as well as exposure time and conditions associated with waste treatment at relevant geographical locations gives the following benefits. It allows for a broad application of the framework, since it reduces the data needs to meteorological data that is readily available worldwide and degradation characteristics of the polymer type, which can be developed for all polymers. Thus, it facilitates product specific inventory development since the MP formation potential is dependent on polymer type. And, the inventory can readily be linked to existing Life Cycle Impact Assessment (LCIA) methods, though only in the case of particulate matter formation and photochemical ozone formation (see section 2.2.3 for more details). To date, no LCAs link small micro and nano particles with particulate matter formation impacts on human or ecosystem health. Attempts to standardize inventory collection which, can be linked to a specific region of the world is also one of the missing components of most work found in the plastic loss inventories. Thus, many questions remain unanswered, such as how various levels of waste treatment infrastructure and practice relate to plastic losses, or how various topographies influence redistribution of plastic in the environment, final fate and impact.

Presently, there are no guidelines on how to collect country specific inventory and include the impacts from MPs and NPs in LCAs of products. Moreover, the initial stages of plastic degradation leading to MP formation in the natural environment are poorly understood, difficult to measure and rarely quantified in LCAs. The issues related to permanence and thereby evolving impact potential of fragmented plastics i.e. size changes of micro particles over time, is also lacking in the literature. Therefore, the aim of this work is to, create a framework, *SecuPlast*, applicable at polymer type level for taking inventory of secondary MPs and NPs that expresses: regional differences in waste treatment, includes the time dependency of MP formation and emission, as well as the

subsequent size changes of MPs in the environment (a dynamic degradation approach), and quantifies the impacts to human health with best available knowledge. The framework is demonstrated and tested on a case study of low-density polyethylene (LDPE) mulch film, in two different countries.

2. Method

2.1. Research Method

The research methodology followed for this work is a mixture of quantitative and qualitative methods that were used to achieve the work's purpose. The goal of this study was to understand the mechanisms governing micro and nanoplastic formation and emission into the atmosphere, and to integrate these emissions into the LCA methodology. One of the questions that this work seeks to answer is, are there physical phenomena occurring during the life cycle of plastic products which give an indication of how much MP and NP will be formed in a given time horizon? And, based on this, which components must be included in the inventory of plastic products to give a complete picture of the impacts? Thereby, an extension and refinement of the already standardized quantitative LCA methodology for gathering inventory of plastic products was carried out. The process followed was to define the variables and domain, as well as interrelations and relationships between the variables identified. The research is thereby, interpretivist in nature. The following research steps were planned: a literature review to identify 1) key variables governing degradation kinetics of conventional and bioplastics, 2) key processes dominating atmospheric emissions MP and NP, 3) key characteristics of MPs and NPs in relation to human health impacts. Based on the literature review a framework for inventory development was conceptualized, referred to as *SecuPlast*, and the pathway to deriving impacts from the new MP and NP emissions was delineated. The resulting empirical model made by following the conceptual framework was tested on a case study and the results were compared to the available literature, in order to answer the initial research questions. The conceptualized framework is described in the following section and details the resulting methodology to follow so that air pollution impacts from plastic products can be added into future LCAs. The results sections details the results obtained from applying the framework to the case study.

2.2. *SecuPlast* – the Conceptual Framework

Fig. 1 shows a visual representation of the conceptual framework developed to account for plastic impacts. The model consist of a degradation module, an emission module, and an impact module.

The degradation module includes UV damage to polymer surfaces by polymer type and plastic product usage. The model then accounts for further degradation of the polymer surface, as a function of exposure time, as well as consecutive degradation of the particles generated in the first instance i.e. month 1, year 1 or other time step. In this module, it is possible to link the degradation to a specific country, city or region by the use of site-specific irradiation data. Likewise, the emission module takes into account site-specific wind conditions and land cover, in order to predict a share of particles emitted to various compartments. Waste treatment capacity in the given location, is an important component of the inventory. Average waste treatment data is used in the model, which calculates a different potential for MP and NP formation depending on waste treatment methods, as well as influencing the possible share of plastic littered. Finally, the impact module takes the substances inventoried in the previous modules and produces impacts for the new emissions using conventional product system

Fig. 1. Conceptual framework for consistent inventory building for plastic loss and impacts in the environment. Dashed rectangle is the system boundary of the plastic's life cycle. The uppermost area shows various degradation pathways for polymers, of which photo-degradation and oxidation have been included in this study. Degradation mechanisms included in the model are progressive damage of the polymer surface, particle formation and degradation and particle accumulation through time. The purple arrows labeled ICEX (inter-compartment exchange) visualize the MPs potential for redistribution and emissions to air, the dashed arrows show sinks.

modelling software and the Life Cycle Impact Assessment methodology for assigning impacts to elemental flows.

3. Degradation module

3.1. Particle formation and size distribution in UV conditions

Photo-degradation of different polymer matrices is extensively covered in the literature (Allen, 2010; Carlsson and Wiles, 1969; Geuskens et al., 1977; Rånby and Rabek, 1975). Modelling studies of for example, epoxy coatings have been used in the past to predict the lifetime of coatings when exposed to weathering, as well as particle distributions (Hinderliter and Croll, 2005; Kiil, 2012; Lu et al., 2018). The approach taken by Lu et al (2018) is chosen here to approximate the initial degradation for polymers due to UV light. As defined in said work, the damage function to a polymer material is given by:

$$D_{total}(t) = E_0 * \phi * t \quad (1)$$

Where D_{total} ($J m^{-2}$) is the effective damage dose to the polymer material with respect to E_0 , the incident UV radiation dose ($W m^{-2}$), t , the time of exposure (s), and Φ , the quantum yield (dimensionless), which is the number of times an event occurs per photon absorbed, or in this case it can be interpreted as the ratio of bonds broken per photon at a given wavelength (Kiil, 2012). The quantum yield may be adjusted for each polymer by using the wavelength of excitement for the photosensitive groups known to

each polymer (Lu et al., 2018; Martin et al., 2002). For example, phenoxy groups in aromatic epoxy absorb the wavelength interval of 300–340 nm, which corresponds to a quantum efficiency of 10^{-4} (Kiil, 2012). Whereas polystyrene's photo-oxidation is initiated by absorption by phenyl chromophore at 260 nm (Yousif and Haddad, 2013) and polypropylene (PP) absorbs at 370 nm (Singh and Sharma, 2008). Absorption by the material is changed by the polymer's state, for example, partially oxidized polystyrene (PS) was shown to absorb at a wavelength of 450 nm (blue visible light), which can penetrate deeper in the ocean and, thus, has implications for the lifetime of PS in the marine environment (Ward et al., 2019). A study of polyethylene (PE) films, showed PE absorbs UV at 216 nm, which suggests the possibility of chromophore sites as C=C, C=O and C-O bonds (Kuria Kamweru et al., 2014). Otherwise, internal and external impurities in PE formulations, such as residues and catalyst containing Al, Ti and Cl, are known to absorb at wavelengths of 290 nm and above, which would correspond to 10^{-4} quantum efficiency (Gijssman et al., 1999; Gijssman and Doezman, 1996). In general quantum efficiencies for various polymers range from 10^{-2} to 10^{-4} (Guillet, 1985).

Equation 1 can be used to derive damage functions for the polymer material which may show a linear, power law or exponential response (more details available in Martin et al., (2002) and Lu et al., (2018)). Finally, equation 1 can be rewritten to derive a weight loss or thickness loss rate (k) for the material in question. The linear response is used in this work (see Lu et al., 2018). Furthermore, the material loss occurs at the very surface of the poly-

mer species, with ablation zones reportedly present from 2–100 μm in thickness, such that polymer delamination happens gradually from the very surface of the polymer towards the inside (Hinderliter and Croll, 2005).

The model generates randomized particle diameter distributions equating the total mass loss (thickness) for the polymer material due to the UV dose received. Assuming particles are spherical, a number of particles can be calculated using the density, ρ , of the polymer material and the particle's volume. Where, the volume of a sphere is given by:

$$V = \frac{4}{3}\pi r^3 \quad (2)$$

and V/ρ , gives the mass (μg) of each particle in the randomized particle size distribution. For simplification purposes, particles are assumed to be spherical, but it is clear that microplastic particles found in the environment come in different shapes, with some shapes possibly being more damaging than others (Nel et al., 2006).

A first particle set is generated by the model at timestep i , where i in this work is years, but is dependent on the waste treatment pathway of the product. For litter pieces and particles, i may be 100 years or 500 years, depending on the time horizon of the LCA and the degradation rate k . On the other hand, for sanitary landfill, an average UV exposure rate of 208 days per year was calculated based on management conditions for European landfills i.e. on average uncovered 4 days per week (European Commission, 2018). These conditions, for landfill, represent the worst case scenario of exposure for plastics that are disposed of by sanitary landfilling. With each subsequent year after $i = 1$, the polymer in question will proceed with polymer delamination i.e. a new set of particles will be generated each year for the given time horizon, until $i = n$. For all particles in particle set $i = 1$, and all particle sets formed after, +1 to $i = n$, the volume change of each particle V_i , due to subsequent UV exposure is given by Eq 3:

$$V_i = V_{i-1} - V_{i-1} * k \quad i = 1, \dots, n \quad (3)$$

Where the volume of a particle in a specific year V_{i-1} , at the timestep i is decreased each year by k , with the thickness loss rate as per the term $V_{i-1} * k$ subtracted from V_{i-1} . Once all volume changes have been calculated for each particle, the volumes are recalculated into particle masses, m , with the relationship $m = V/\rho$.

Thereafter, the sum of all particles that have reached sensitive sizes at the completion of the timestep are compiled into relevant damage size categories based on their diameter, d , i.e. $d < 2.51 \mu\text{m}$, $d > 2.5$ and $d < 10 \mu\text{m}$. So that the mass of each of the relevant damage size categories is given by the sum of the mass of each particle j , over timestep i , where $M_{i,j}$ fulfills the diameter condition in Eq 4.

$$M_{\text{tot}, \text{PMx}} = \sum_{i,j} M_{i,j} \text{ for } M_{i,j} \text{ with } d \leq 2.5 \mu\text{m} \text{ and } 2.51 > d \leq 10 \mu\text{m} \quad (4)$$

Then the cumulative mass of particles, $M_{\text{tot}, \text{cum}}$, with sensitive diameter sizes derived from each new particle set generated for $i=1, \dots, n$, is given by:

$$M_{\text{tot}, \text{cum}} = \sum_i \sum_j M_{i,j} \quad (5)$$

Particle diameter distributions for the starting particle set are adjusted to experimentally observed values whenever possible, since it has also been shown to be polymer specific, e.g. polystyrene (PS) has been proven to produce nano-sized particles after just 14 days of 24h light exposure (Lambert and Wagner, 2016). However, particle distributions reported in the literature are highly dependent

on, among other, detection method and collection equipment. This makes it difficult to determine a universal size distribution for particle formation (Eo et al., 2018), or a steady state as is often used in LCA (Rosenbaum et al., 2017). A summary of particle distributions found in the literature, derived from table S1 in the Supplementary Information (SI), in different settings i.e. marine environment, soil, sediment etc., is used in the degradation module for the randomly generated particles. In Table 1, 3 possible distribution sets which were used to test the model are presented with set 1) summary of particle size distributions derived from real life measurements in the literature, set 2) a higher proportion of smaller particles in the set, and set 3) a set with a lower proportion of smaller particles.

3.2. Degradation into gaseous compounds

Gaseous products of decomposition such as CO_2 , H_2O and CH_4 , are part of the degradation module and should always be considered in relation to plastic degradation. It is well known that the final products of decomposition of various plastics are CO_2 , H_2O and CH_4 , but that there can also be intermediate products during degradation (Bastioli, 2014). Also, depending on degradation conditions being aerobic or anaerobic, the species of decomposition gases will differ (Royer et al., 2018; Singh and Sharma, 2008). Furthermore, morphology of the polymer i.e. whether it is a film, powder or fiber will have influence on the rate of degradation (Royer et al., 2018). Also, any biomass produced in the process due to microbial activity will also mineralize during the assessment's time horizon. For this work, gas production rates presented in Royer et al. (2018) are applied to the case study of mulch film, since this work presents gas production rates under various conditions, representing various possible fates of the particles or litter pieces, such as burial in soil, decomposition in marine water, or air environment under light or dark conditions. Average gas production rates under various conditions used for the case study are presented in Table 2.

It is important to note that the production rates for gases used for the case study (presented in Table 2) are not the only possible compounds emitted during the degradation of plastic materials and that even new plastic products have been shown to emit volatile organic compounds (VOCs) (Even et al., 2019; Lattuati-Derieux et al., 2013; Lomonaco et al., 2020; Shen et al., 2020; Webb et al., 2012). A recent study, looked at the VOC profiles of common plastic consumer goods and found that out of a restricted list of VOCs, softer plastics (PE, PS, and PVC) emitted larger amounts of, among other, cyclohexanone, 2-Phenyl-2-propanol and toluene, in comparison to harder plastics (Even et al., 2019). Furthermore, the authors calculated piece specific emission rates with various exposure scenarios and obtained quantities of VOCs far below the European Lowest Concentration of Interest values (EU-LCI) for indoor air quality. However, they pointed out that their results are based on a scenario where 1 toy emits VOCs in a room of 30 m^3 , which does not allow for exceeding the EU-LCI guidelines. However, when the breathing area is reduced to 1 m^3 , representing the area around a child playing with the toy, and the sensitivity of the child is taken into account, then guidelines may be exceeded (Even et al., 2019). Yet another study, compared the VOC profiles of virgin material to that of marine plastic recovered from a beach (Lomonaco et al., 2020). It found that virgin PE produced 100 ng/g acrolein, 200 ng/g propanol, and 2500 ng/g methyl vinyl ketone, while marine PE emitted 20, 50, and 2500 ng/g of the respective VOCs. The same study showed virgin PS was a trace source of benzene, toluene, ethylbenzene, and benzaldehyde at levels from 10–30 $\mu\text{g/g}$ and PP is a source of methyl vinyl ketone and methyl propenyl ketone (4 and 10 $\mu\text{g/g}$, respectively), while marine PP emitted 600 ng/g methyl propenyl ketone. Importantly, some of these VOCs are known harmful compounds and

Table 1

Summary of particle distribution sets used in the case study. Set 1 corresponds to particle distributions summarized from real life measurements found in the literature (see table S1, in SI). Set 2 and 3 correspond to a situation with higher proportion or lower proportion of small particles, respectively.

Set 1		Set 2		Set 3	
Size class median (μm)	% by size class	Size class median (μm)	% by size class	Size class median (μm)	% by size class
148	66%	125	75%	125	45%
488	25%	375	15%	500	21%
2000	9%	1500	10%	2375	34%

Table 2

Average gas production rates for aged LDPE estimated from Royer et al 2018.

	Unit	Air		Water	
		Dark	Light	Dark	Light
Methane (CH ₄)	C (nmol/g/d)	0.37	0.27	0.00	5.40
Ethylene (C ₂ H ₄)	C (nmol/g/d)	0.14	48.16	0.00	5.10
Ethane (C ₂ H ₆)	C (nmol/g/d)	0.21	0.16	0.00	1.40
Propene (C ₃ H ₆)	C (nmol/g/d)	0.06	0.24	0.00	1.10

thereby the previously mentioned study concluded that plastics are a potential unrecognized source of toxic and harmful trace gases (Lomonaco et al., 2020). Furthermore, all compounds detected by the methods used in these studies could only identify and quantify a fraction of the peaks detected by Gas Chromatography-Mass Spectroscopy (GC-MS) methods used i.e. from 100–400 peaks representing distinct VOCs a total of 12 to ca. 140 peaks were identified (Even et al., 2019; Lomonaco et al., 2020).

3.3. Emissions Module

Consider a MP source situated on the ground. The most important parameters, responsible for emission of the MPs from the surface into air are (1) the speed of the wind that the MPs is exposed to and (2) the size and shape of the MPs.

In this work the wind speed for one point in Denmark and one point in Italy over 11 years (time resolution of 1 hour) was obtained for 10 meter wind data, i.e. data for wind speeds at an altitude of 10 meter, from ERA4 from European Centre for Middle range Weather Forecasts (ECMWF). These data were transformed into two meter wind data, i.e. data for wind speeds at an altitude of 2 meter, for 7 different categories of land use for Denmark and Italy applying the wind shear formula (Holton, 2004). The land use categories considered were bare soil, built area, cropland, forest, sand, shrub land, and water. For the second parameter (the shape) it was assumed that all the MPs were spherical.

The spatial resolution applied in our approach (i.e. national average) for the wind speed is in good alignment with the inventory data typically applied in LCA (e.g. Ecoinvent (Ecoinvent 2021)), where the highest possible spatial resolution equals national resolution. Application of average data including time weighted averages are also considered common practice in LCA, which most often seeks to assess a typical/normal/average situation (see e.g. Tian (2018)).

Based on these assumptions a 2-dimensional micro-physical model was developed (Hansen et al., 2021). The model is based on the theory of the oblique roll of the spherical object under the influence of air resistance. As input to the model, data from the MPs size distributions generated by the degradation module for 1 kg of plastic mulch film was used. The angle for the oblique roll in the model was 40°. This angle was chosen based on a study by Rice et al., (1995).

The 2-dimensional micro-physical model determines whether the MPs will lift into the air or stay on the ground. The share emit-

ted to air (in the range 0–10 μm) is considered a potential exposure to human health via inhalation. The final emission equation, MP_{lift} , in meters (Hansen et al., 2021) is given by:

$$MP_{lift} = \frac{-m}{K} * \ln \left(\frac{mg}{K} * \frac{1}{V_0 \sin(\alpha) * \frac{-mg}{K}} \right) \quad (6)$$

Where m is the mass of the MP particle (μg), V_0 is the particle's initial velocity (m/s), α is the angle of oblique roll (40°), g is the gravitational constant (9.8 m/s²), and K is the resistance through air of a particle with a mass, m , and initial velocity, V_0 , varied to represent average wind conditions of the location and associated average land cover, with a weighted average of equation 6. Air resistance, K , of the particle is further given by the equation, $K = 6\pi f r$, where, f is the fluid viscosity of air (kg/m*s), and r is the radius of the given MP particle (μm). The remainder of the particle distribution is likely captured in soil or emitted to water ecosystems. There is a growing body of literature on the possible final fate of macro plastics and MPs in the environment (Allen et al., 2019; Barrows et al., 2018; Eriksen et al., 2014; Kawecki and Nowack, 2019). Due to the uncertainty related in determining the final fate of the MPs in the upper end of the size distribution in the environment, in this work, the model only considers emissions to air. Particles that are not emitted to air and are not in the 0–10 μm range are shown as inventory only and no further estimates are done to determine their fate or characterization. For more detailed information on the impacts the larger size fraction of MP might have on ecosystems, the reader is referred to Saling et al., (2020), and Lavoie et al., (2021) where effect factors have been derived for a mass of MPs or NPs entering the aquatic compartment (Saling et al., 2020). Considering the long residence time of plastic in the environment and the light density of plastic materials, which increases the likelihood of their transport between compartments, it was deemed less likely that plastic sinks are permanent. Final fate for MPs and macro-plastics have been given various estimated values from only 10% reaching water bodies for tire abrasion particles (Ryberg et al., 2019), to between 10–40% of mismanaged plastic reaching the ocean (Jambeck et al., 2015). More sophisticated modelling methods have been applied to predict sedimentation and buoyancy of plastic particles (Besseling et al., 2017) and more granular estimates of plastic losses to watersheds (Lebreton and Andrady, 2019). It is clear from the wide range of values for plastic ending up in a particular compartment that this is a highly speculative area, but also that MPs have a high propensity for long distance travel as has been evidence by the work of Allen et al., (2019) and Brahney et al., (2020).

3.4. Impacts Module

PM is a known air pollutant causing health effects in populations worldwide. PM consisting of “coarse particles” with aerodynamic diameters <10 μm, fine particles <2.5 μm and ultrafine particles of aerodynamic diameter <0.1 μm cause among other, respiratory damage due to, but not limited, to their small size, since their chemical composition and shape is also relevant. Fine particles and ultrafine particles are the most damaging since their small

size allows them to penetrate deep in the lung. Ultrafine particles can penetrate the alveolar capillary membrane and reach any other organ in the body through the bloodstream. Thus, particles, though their point of entry is the respiratory track can lead to systemic effects causing damage in the rest of the body (Schraufnagel et al., 2019). Furthermore, the final damage of PM is not only size related, but also depends on their composition and structure. PM serves as a vector for heavy metals or other chemicals, depending on the source and may interact with allergens to induce asthmatic attacks (Baldacci et al., 2015). In the context of MPs, additives such as bisphenol A and phthalates are just a few of the substances that may gain entry via inhalation. Furthermore, the biopersistence of MPs is another cause for concern, since they have been shown to accumulate in lung tissue, in for example lung biopsies of textile workers (Gasperi et al., 2018). Within the LCIA area, mature characterization models are available that arrive at a final damage caused per unit of PM emitted. Thus, no further steps were taken to characterize the impact from MPs on human health, since we rely on the existing characterization models for this purpose and on the fact that the most important factor determining the damage of the particles generated by the degradation model is their size (Schraufnagel et al., 2019). Though it is acknowledged that the combination of size/composition/shape of MPs are all important parameters determining final impact on human health, these parameters are not thus far well characterized, and have not been included in this work. The emission module describes the share of particles of the whole particle distribution that is emitted to air or stays on the ground. The mass of the share emitted to air in the particle range of 0–10 µm is then used as input for the characterization models for further quantification of its induced impacts. The characterization factors used for the various size fractions resulting from the degradation module i.e. <2.5µm, and <10µm are documented in the SI, table S2. Due to the degradation of the particles, some of the initial mass of particles disappears, either because it has become a gaseous compound and/or a very small nanoparticle which contributes very little in mass to the overall burden of PM expressed as mass in the given time horizon. The impacts expressed in the results consider the remaining mass of the particles as being capable of causing an PM impact. On the other hand, few studies have characterized the damage that losses of plastic might have on ecosystems (Everaert et al., 2018; Lavoie et al., 2021; Lechner and Ramler, 2015; Redondo-Hasselerharm et al., 2018; Saling et al., 2020; Woods et al., 2019). For the most part, effect studies have been done in the laboratory with often higher concentrations than what is found in nature (de Sá et al., 2018). Toxicological effects of PE and PS particles have been observed in the laboratory, but are highly dependent on concentration. On the other hand, various studies have found negative effects on *Daphnia magna* communities and benthic organism at environmentally relevant concentrations (Anbumani and Kakkar, 2018; Bour et al., 2018; Redondo-Hasselerharm et al., 2018). Effects to marine organisms reported in the literature are many and include effects at the 1) molecular level, such as impacts to cell metabolism, gene expression, and cell signaling, 2) systems level such as, digestive, neurological or immune functions, and 3) effects at the organism level such as, growth, reproduction and/or feeding success (for more information please see, Franzellitti et al., 2019). In vitro toxicity of plastic consumer products was analyzed in order to benchmark toxicity from conventional polymers and bio-polymers (Zimmermann et al., 2020, 2019). The authors found the endpoints affected (cytotoxicity, estrogenicity, antiandrogenicity, and oxidative stress) by the products made from the various commodity polymers varied within polymer type, but was generally lower for products made from HDPE and PET (Zimmermann et al., 2019). Baseline toxicity was the highest for PVC, PUR-based products (also affecting most endpoints) and PLA, whereas baseline toxicity for

Table 3

Average regional waste treatment for Denmark and Italy. “-Slow” scenarios include MP formation via UV degradation while the “-base” scenarios do not include impacts from MPs.

	Recycling	Incineration	Landfill	Litter
DK-base	37%	60%	3%	0%
DK-slow	23%	65%	2%	10%
IT-base	31%	33%	36%	0%
IT-slow	28%	29%	32%	10%

PE, PS and LDPE based products varied too much to make generalizations about their toxicity. This may be due to differences in the formulation of the various products of the same polymer type including intermediates, solvents, process regulators and additives that may differ in the formulations (Zimmermann et al., 2019). Notably, bio-polymers exhibited comparable levels of baseline toxicity as their petroleum counterparts e.g. PE vs Bio-PE. Though it was shown that there was a significant difference with regard to inducing estrogenicity, with bio-polymers having a less pronounced effect than petro-polymers for this endpoint (Zimmermann et al., 2020). In this work, it has not been possible to add the toxicity effects described in the above mentioned to the impacts module and impacts on species are not included. For the latest developments on characterization of impacts of microplastics on ecosystems, the reader is referred to Lavoie et al., (2021) and Saling et al., (2020).

3.5. The case study

Mulch film, which is typically used in agricultural applications for raising the temperature of soil and weed control was chosen as the product to test the SeqPlast model. As the model quantifies degradation due to UV damage, it was important to choose a product that is outside most of the year, or at times when not disposed of properly because it might be ploughed into fields or left on the field, all of the year (Steinmetz et al., 2016). For this purpose, an LCA was carried out that includes all life cycles of this product, that is to say the production of LDPE, use phase i.e. extending the film on agricultural land for 1 year, and end of life (EoL) with the following options: recycling, incineration, landfilling in sanitary landfill under European conditions, and litter. The baseline did not include litter as a possible end of life and did not include MPs formation during landfilling or during the use phase. Transport was excluded from all scenarios.

The LCA was performed from cradle-to-grave, with the system boundary extending from LDPE manufacturing gate to EoL gate. Co-products from EoL, that is to say, new LDPE granules from recycling and energy production from incineration, were not included in the study since this can be better categorized as an accounting LCA or situation A micro-level decision support i.e. it answers questions related to specific products, which are not expected to change production capacities (European Commission - Joint Research Centre, 2010). The goal of the study is to explore the difference in impacts when including or not including impacts from MPs and littered macro-plastic, which also becomes MPs, concerning eco-design of products.

Two European regions were assessed, Denmark and Italy. The differences in waste treatment of these regions can be seen in Table 3. The recycling rates include a reject rate of 13% at sorting facilities and 17% of the incoming plastic waste at recycling facilities (European Commission, 2019), which was estimated as the average European reject rate.

A littering rate of 10% was applied to the non-baseline scenarios. It is difficult to estimate with accuracy how much of the plastic consumed in one region is littered or mismanaged. A littering rate of 2% was used by (Jambeck et al., 2015) for all European countries,

while the share of mismanaged plastic varied from 15–40% of the waste produced by coastal populations. However, a recent study of mismanaged plastic waste with a spatial resolution of 1 km for the world, quantifies Europe's mismanaged plastic at ca. 11% of annual plastic waste generation or 3.3 million tons yearly (lower and upper range of 1.3–9.1 Mt) (Lebreton and Andrady, 2019). Thus, 10% was deemed a more appropriate value for this work, though this value was varied to test the sensitivity of the littering rate for overall results. No losses of MPs were modelled for the recycling, and incineration portion of the waste management. This is due to lack of data, but should be included in the future as more information becomes available and is an essential component of any plastic loss inventory for LCA. Using local conditions for waste management is key to obtain realistic plastic losses. This is demonstrated by the various publication that use data on waste management to derive plastic losses for various regions (Jambeck et al., 2015; Kawecki and Nowack, 2019; Ryberg et al., 2019).

The sensitivity of the model was also tested in regard to degradation rate. Values for degradation in the work of Chamas et al., (2020), which include degradation rates for several polymers under various degradation conditions, were used as a point of comparison for the UV degradation rates calculated in the degradation module.

UV irradiance data was obtained from Solar Radiation Data website (SoDA) (Global Modeling and Assimilation Office (GMAO), 2015) for 3 points in each country of interest representing a wide coverage of latitudes in each country. UV data was averaged on a monthly basis for the three locations and used as input in the degradation module. Furthermore, the dosage of UV is adjusted to each specific EoL option, depending on the product of interest by equation 1. Land use cover data, which is used in the emissions module to adjust wind conditions to the surface roughness of the area, was obtained from Eurostat (Eurostat, 2020).

The calculations were done using the data analysis tool R (R Core Team, 2013) where several scripts were developed for the assessment. Data generated from all three modules served as input to the LCA software OpenLCA (GreenDelta, 2019) which was supplied with the Ecoinvent v3.6 database (Wernet et al., 2016) and was used to construct the LCA model. Two characterization methods were used for the life cycle impact assessment: ReCiPe Hierarchist midpoint and endpoints (Huijbregts et al., 2017) and ILCD 2011 Midpoint+ (European Commission - Joint Research Centre - Institute for Environment and Sustainability, 2012). ILCD midpoints were then characterized to damage as endpoints with regionalized values for disability adjusted life years (DALY) (Verones et al., 2016). The damage at endpoints was then monetized using budget constrained ability to pay, which is the least uncertain way to assign a value to various damages (Weidema, 2009). Based on this method, a value of 110 000 USD₂₀₀₃ per DALY was used (Dong et al., 2019; Weidema, 2009).

4. Results

4.1. Model Results

The model provides various insights into the resulting degradation kinetics, fate, and damage of MP particles formed through the degradation of plastic items. The case study focused on examining the fate and damage of particles arising from 1 kg of LDPE typically used as mulch film in agriculture to raise the temperature of the soil, protect against weeds, etc. (Steinmetz et al., 2016). It successfully links the waste treatment infrastructure and land cover of various countries to the final emission of MPs to the different compartments, making it possible to assess MP impacts in a regionalized manner.

4.2. Degradation rates and wind analysis

Summary statistics for wind data used in the emissions model per land cover is presented in Table 4. For simplification purposes, the 1st quantile was used as the minimum (Min) wind speed vector for the model and the maximum values were used as the high (Max). This is done to get as wide a representation as possible for wind speeds for each of the locations assessed, which are then implemented in the emissions module as minimum and maximum values for wind speed.

Results from the degradation module predict a total yearly damage factor of 0.49 and 0.65 (dimensionless) for DK and IT respectively and a yearly damage factor of 0.28 and 0.37 (dimensionless) under landfill conditions, due to UV degradation. These degradation rates are used in the rest of the work as suggested by Lu et al. (2018) and explained in 3.1.1 to derive a yearly mass loss as particle size distributions. The values agree with a higher overall irradiance falling on the Italian territory than in Denmark.

4.3. Particle spreading into various compartments

Surface roughness in combination with wind velocity determines the particle's ability to lift off the ground and potentially become a source of particle exposure for human health. Fig. 2 shows how the various types of surfaces affect whether particles are emitted to air or stay on the ground, as a function of wind speed.

It is clear from Fig. 2 and Fig. 3 that the smallest particles, those that are responsible for respiratory damage to human health (<10µm), are able to take off the ground under all wind conditions and land covers tested, as shown by the red distribution in conditions of low wind in Fig. 2. By contrast, Fig. 3 shows the diameter size of particles that are not able to lift off the ground. This includes the largest particles, mostly above 2500µm in diameter, for conditions of high wind (Max), with the exception of built up areas. Note, smaller particle ranges (purple) are also able to stay on the ground for low wind speed conditions (Min), though, particles of diameters lower than 10 µm are predicted to be emitted to air by the model.

Important to note is the fact that forest cover acts as a sink for MP and NP particles, since none of the particles are able to leave the ground (data not shown). This has recently been described in measured atmospheric deposition rates of MPs, as the “canopy comb out” effect produced from forests, i.e. particles collect on plant leaves as a function of leaf area index and subsequently wash out after precipitation events (Klein and Fischer, 2019). This has important repercussion for geographical areas dense in forest cover.

The model groups particle by diameter sizes into different compartments, i.e. ground and air. The particles ending in the air compartment are further grouped into three groups according to size: PM₁₀, PM_{2.5}, and large particles in the air fraction (>10.1 µm in diameter).

The variation of the mass of particles ending in each of the compartments and standard deviation can be seen in Table 5.

In general, higher variation is observed under high Danish wind conditions (Max). The percentage of particles ending up in the air and ground compartments under Max conditions was 77% and 23%, while for Min conditions it was 6% and 94%, respectively. Furthermore, the percentage ending up in the PM₁₀ fraction was higher for Min conditions than Max, since the mass fraction of the particle distribution ending up in the air compartment for low wind conditions is lower, thus PM₁₀ is a larger share of the air compartment, 1.7×10^{-3} vs 1.4×10^{-4} . The standard deviation for all PM shares is modest, thereby, the model is very stable and predicts a more or less uniform share ending up as PM for all life cycle stages

Table 4

Summary statistics of hourly wind speed in meters per second. Summary statistics are adjusted by the wind shear formula to 2 m wind speed, depending on land use type.

Denmark						
Land use type	Built Area	Cropland	Shrub land	Sand	Bare soil	Water
Min	0.00	0.00	0.00	0.00	0.00	0.00
1st Qu.	2.40	1.37	2.23	3.20	2.90	3.22
Median	3.44	1.96	3.18	4.57	4.15	4.61
Mean	3.58	2.04	3.32	4.76	4.32	4.80
3rd Qu.	4.64	2.65	4.30	6.17	5.60	6.21
Max	11.63	6.63	10.76	15.46	14.02	15.56

Italy						
Land use type	Built Area	Cropland	Shrub land	Sand	Bare soil	Water
Min.	0.01	0.01	0.01	0.02	0.02	0.02
1st Qu.	0.75	1.31	1.21	1.74	1.58	1.75
Median	1.09	1.91	1.77	2.54	2.30	2.55
Mean	1.23	2.16	1.99	2.87	2.60	2.89
3rd Qu.	1.58	2.77	2.57	3.69	3.34	3.71
Max.	5.71	10.01	9.26	13.31	12.07	13.40

Fig. 2. Violin plot of particle distribution count generated during sanitary landfilling of LDPE. Blue particles, with diameters higher than $\sim 30 \mu\text{m}$ stay on the ground to various degrees depending on surface roughness. Red particles in the lower part of the particle distribution ca. $< 30 \mu\text{m}$ are lifted into the atmosphere even under low wind speed conditions (Min) while orange particles show the diameter sizes lifted into the air under high winds (Max) and include few particles of sizes $> 500 \mu\text{m}$ in diameter.

assessed (Landfill, Litter, and the Use phase). About $5.03 \times 10^{-6} \%$ of the mass is in the range of $\text{PM}_{2.5}$ with Min conditions and $4.14 \times 10^{-7} \%$ for max conditions. The mass share of PM_{10} and $\text{PM}_{2.5}$ is very low, which is quickly understood when taking in consideration the mass of a single particle i.e. 4.83×10^{-13} and 7.55×10^{-15} kg, for a particle of 10 and $2.5 \mu\text{m}$ in diameter, respectively, and a density equivalent to PE. Thus, $\sim 99\%$ of the particle mass emitted to the air compartment belongs to particles of diameters $> 11 \mu\text{m}$, which are heavier.

4.4. Particle evolution through the 100 year horizon

It is not enough to consider the effects of plastic particles for one year of degradation, since the formed particles in year 1 will continue to degrade over the following years, and in the case of littering, the release of a 1st set of particles in year 1 allows deeper layers of the polymer to be exposed to further degradation. Thus, in the subsequent years after the 1st set of particles is formed and is removed from the polymer surface, additional sets of particles

Fig. 3. Violin plot of particle distribution count generated during littering of LDPE. Showing only particles that stay on the ground depending on high wind speed (max) and low wind speed (min).

Table 5

Share of mass of particle distribution emitted to air or remaining on the ground depending on wind intensity. Percentage share refers to the average mass in each compartment out of the total mass of the particle distribution in each life cycle (use phase, landfill and litter) and standard deviation in parenthesis.

Compartment	Wind Intensity	
	Min	Max
Ground	94% ($\pm 1.43 \times 10^{-03}$)	23% ($\pm 2.37 \times 10^{-02}$)
Air	6% ($\pm 1.43 \times 10^{-03}$)	77% ($\pm 2.37 \times 10^{-02}$)

are formed and continue to degrade the polymer. Additionally, all subsequent sets of particles continue to decrease in size during the 100 year horizon.

At a constant degradation rate, the 1st set of particle continues to degrade through 100 years, shown in Fig. 4. Starting with an average diameter of ca. 130 μm at year 1, the 1st particle set reaches an average diameter of ca. 40 μm in year 100. The particle distribution after 100 years has a considerably higher amount of particles in the damaging diameter range 0–10 μm , than after 1 year of degradation.

To test the model, in order to see when the particle distribution would reach ca. 10 μm on average, the time horizon with the baseline degradation rate for Denmark was extended. Additionally, the degradation rate was tested with the values presented by Chamas et al., (2020), since they use a very similar approach, but include additional types of degradation, other than UV, and accelerating factors of degradation in their model. Fig. 5A shows the influence of a higher degradation rate on the size distribution of littered particles, hereafter referred to as the Fast degradation rate. The Fast degradation rate is higher by a factor from 6–20 depend-

ing on the life cycle phase, but is on average 10 times higher in magnitude than the slow degradation rate for Denmark i.e. on average 0.027 $\mu\text{g}/\text{year}$ for a slow degradation and 0.279 $\mu\text{g}/\text{year}$ for a fast degradation rate. After about 18–20 years, the particle distribution with the faster degradation rate reaches an average size of 10 μm . This is in stark contrast to Fig. 5B, where the particle distribution reaches ca. 10 μm on average after 200 years. This observation provides evidence for the importance of knowing how fast material degradation will occur under natural environment conditions.

The evolution of the total mass littered in comparison to the mass of particles in the PM 0–10 μm range can be seen in Fig. 6. Fig. 6A and Fig. 6B show the logarithmic mass degradation. During this time step the mass of both PM ranges increases during the 100 year period and in Fig. 6A it is a slow linear increase, while in Fig. 6B the increase is sharp at the beginning and begins to level off toward the end of the time period. The slow increase in Fig. 6A is due to bigger particles in the particle distribution degrading and becoming part of the PM range. The sharper increase in Fig. 6B considers also the additional mass of additional particles sets being generated in the polymer and adding to the share of PM₁₀ and PM_{2.5}. The same pattern is observed again in Fig. 7, which shows the littered piece and PM particle mass evolution with the fast degradation rate described above. In the latter, it is possible to see the completion of the fate of the particle distribution. That is to say, a sharp increase in the PM levels followed by a drop as the total polymer mass continues to degrade and approaches zero. This pattern also explains the reason why a large part of the mass never becomes PM, since as PM increases there is also a share of the mass that essentially disappears from the particle distribution i.e. with constant degradation there is also a constant elimination of mass from the particle distribution or the mass of some of the particles becomes so small that it is practi-

Fig. 4. Density plot of the 1st set of particles formed due to UV degradation through a 100 year horizon.

Fig. 5. Density plots of particle distributions degrading through time, for A) a faster degradation rate as per Chamas et al. (2020) and B) the baseline degradation rate for Denmark with an extended time horizon of 200 years. The tails of the particle distributions have been cut at 300 μm and 500 μm for A and B, respectively.

cally undetectable. This is similar to experimental results observed by Song et al. (Song et al., 2017) and colleagues who could only account for around 24% of the initial mass of polymer after accelerated weathering experiments, and inferred that the unaccounted mass was in the submicron range and thereby undetectable.

After the 100 year period, and even when all particle sets are taken into account, PM_{10} makes up 0.024% and 1.24% of the initial littered mass for slow and fast degradation, respectively, while $\text{PM}_{2.5}$ makes up $8.66 \times 10^{-07}\%$ and 0.012% of the initial mass for slow and fast degradation, respectively. That said, it is worth noting that the increase of PM_{10} and $\text{PM}_{2.5}$ within one year is of 592% and 376% respectively, for the Italian degradation rate during lit-

tering, and 280% and 194% for PM_{10} and $\text{PM}_{2.5}$, respectively, with the Danish degradation rate. However, the increase in PM_{10} and $\text{PM}_{2.5}$ mass is small, due to the very small masses of these particles and even when subsequent shedding of the polymer with a new particle set each year and the continued degradation of the sets thereof is accounted for, the mass of PM_{10} and $\text{PM}_{2.5}$ account for 2.2×10^{-5} kg and 8.07×10^{-8} kg, respectively, after 100 years of exposure. In comparison, the mass of PM_{10} is 5.6×10^{-7} kg and the mass of $\text{PM}_{2.5}$ is 1.9×10^{-9} kg after 1 year of exposure of the 1st particle set. Thus, the increase of PM is large, but does not amount to large masses due to the minuteness of particles.

Fig. 6. Degradation of total littered mass and formation of particulate matter in the 0-10 μm range for A) the 1st particle set as LogMass and total logmass of polymer and B) the 1st particle set and subsequent particle sets formed each year for 100 years as logMass.

Fig. 7. Degradation of the 1st set of particles with the Fast degradation in log scale.

4.5. LCA Results

SeqPlast relies mostly on recently published and thus existing characterization factors (CFs) for the developed impact assessment, i.e. substances tallied during the inventory are multiplied by a value (CF) which gives them a relative impact to a specific area of environmental interest a.k.a. an impact category. Two different characterization methods were used to quantify the impacts induced by the elemental flows tallied here from MP formation. The two characterization methods included are ReCiPe 2016 and ILCD 2011, which are both standardized LCA methods, but differ in their classification of elemental flows (grouping) and CFs. For example, ReCiPe groups elemental emissions of ethane into two impact categories, Ozone Formation contributing to Human Health and Ozone Formation contributing to Terrestrial Ecosystems, as kg of NO_x-equivalents. While ILCD groups ethane into one Photochemical Ozone Formation impact category, as kg of non-methane volatile organic compounds (NMVOC)-equivalents. On the other hand, ReCiPe assigns a CF of 36 kg CO₂ equivalents to 1 kg of CH₄ emissions (the current consensus value according to the IPCC), while ILCD assigns it a value of 26 kg of CO₂ equivalents. Furthermore, ReCiPe lacks a CF for PM₁₀ emissions, which are assigned a 0 contribution to particulate matter formation, while ILCD characterizes PM₁₀ as 0.228 or 0.406 kg of PM_{2.5}-equivalents, depending on population typology (high or low population density). Thus, due to these known methodological differences it was deemed necessary to use both characterization methods, since ReCiPe is expected to underestimate impacts arising from particles, while ILCD will underestimate the global warming potential of the emissions of GHGs from plastic degradation, which are also included in SeqPlast.

The degradation of plastic in the environment leads to the creation of small particles, the release of NMVOCs such as ethane, ethylene, and propene and the release of the potent GHG, methane. These emissions were inventoried throughout the whole life cycle of 1 kg of LDPE used as mulch film and compared to a situation where the impacts from the degradation of plastic are ignored. The emissions contribute to three/four impact categories, listed first for ILCD and second for ReCiPe, i.e. particulate matter/fine particulate matter formation, Climate change/Global warming potential, and Photochemical Ozone Formation/Ozone formation for human or terrestrial ecosystems. The sensitivity of the model was tested for two variables, 1) an increase in the amount of littering, and 2) a fast or slow degradation rate.

4.6. Comparison to mulch film without MPs

Relative to a case where additional impacts of plastic are not taken into account, the addition of MP and NPs changes the overall impacts of the 1 kg of mulch film very little, as seen in Fig. 8. As explained previously, the ReCiPe method does not assign an impact to most of the particles formed due to degradation, since these are in the PM₁₀ range and it only accounts for PM_{2.5}. Photochemical ozone formation is also slightly underestimated. Thus, results from the ILCD method are given more focus from this point forward.

Looking at the contribution of each life cycle stage, here defined as the production of LDPE, the use of LDPE as mulch film and finally the disposal of mulch film via the waste treatment options of recycling, landfilling in sanitary landfill, controlled incineration, and finally the possibility of littering the mulch film. Fig. 9 shows the contribution of each of the above named life cycle stages to three impact categories for LDPE without additional impacts (None panel) and for LDPE with the impacts of MPs under Danish degradation conditions (Slow panel).

In comparison to production, recycling and incineration, the inventoried impacts coming from MPs and emission of NMVOCs and GHGs during the decomposition of LDPE in the environment, are

small. Littering is the most burdensome of the MPs impacts, but accounts roughly for 1% of the impact for particulate matter formation and photochemical ozone formation. However, when the fast degradation rate is tested for sensitivity with the values for degradation reported by Chamas et al. (2020), noteworthy changes can be seen in particulate matter formation. With a faster degradation rate of MPs, the particulate matter burden increases by 40% compared to the MPs-less case, and 38% compared to the Slow degradation case. Moreover, the impacts of littering are now the second most burdensome impact contributing to particulate matter, second only after the production phase (Fig. 10, Fast).

4.7. Increased littering

Increasing the degree of littering, from the baseline value of 10% to 50%, 75% and 100% of the polymer, has the counterintuitive effect of lowering the overall impacts of the three impact categories studied. This is because when a larger share of LDPE is littered, a smaller share of the plastic is treated via recycling, incineration and landfilling. And, as can be observed in Fig. 9, conventional EoL treatment options incur significant burdens. The LCA shows that results are rather impervious to a change in the littering rate, at least for the baseline “slow” degradation rate. A change of 40%, from 10% to 50% in the littering rate, incurs an increase of a mere 4% in the final impact of the three impact categories.

4.8. Regional differences

Particulate matter emissions from Italy are higher than for the Danish case, suggesting that the higher UV degradation rate in Italy is a more important factor than the forest cover of the locations assessed. Italy has a 33% forest cover compared to Denmark's 11.4% of total land cover. The overall impact of waste treatment in Italy is lower, due mostly to a lower incineration rate. However, a higher rate of landfilling in Italy is visible, as there are higher impacts from the landfill contribution for Italy than Denmark.

4.9. Final damage assessment of MP impact

The increase in Disability Adjusted Life (DALY) years in Denmark, in comparison to the case when no MP impacts are included is of 0.06% per kg of LDPE mulch film, while species loss increases by about 0.01% for all scenarios. By contrast, for Italy the increase in DALY is of 0.5% per kg of LDPE in comparison to the Italian baseline. For the fast degradation tested under Danish conditions, the increase is of ca. 2.4% in comparison to the Danish baseline.

When expressed in monetary terms (USD), the per kg contribution of monetized damage is low, only a few cents per kilo of LDPE, though more noticeable for IT and also for a faster degradation rate. Though these amounts may seem small when observed on a per kilo basis, it is informative to think about them in terms of, for example the EU's consumption of LDPE. Scaled up to European level, the additional impacts of plastic considered costs the EU between 3.5 million USD and 133 million USD per year in human health damages for a slow and fast degradation, respectively, with a 10% littering rate. If similar damages are considered for the whole of the EU's consumption of plastic (though this is better assessed on a per type of polymer basis), then the damage to human health could potentially cost from 20 million to 755 million USD in damage to species. These values, though highly uncertain, should be considered carefully and refined as new information from risk assessment studies becomes available to LCA.

5. Discussion

The SeqPlast model may be a useful first step in generating complete inventories of plastic pollution that do not over-

Fig. 8. Relative change in Photochemical ozone formation, Particulate matter and Climate change potential relative to when degradation of plastics as MPs in the environment is not accounted. Difference between two life cycle impact assessment methods, ILCD and ReCiPe.

Fig. 9. Contribution from each life cycle stage to the impacts of 1 kg of LDPE from cradle-to-grave for the plastic without additional impacts from the formation of MP, a.k.a. "None" for no additional degradation in the environment, and 1 kg of LDPE with MP impacts, here dubbed "Slow" according to the rather slow degradation for the baseline Danish case.

look the contribution of small particles. The conceptual framework presented can be applied to any plastic product that experiences photooxidation, granted further data is needed for other polymers especially on potential gaseous decomposition products when degradation occurs in the natural environment and when degradation includes biological and mechanical abrasion pathways. Furthermore, it was shown with the case study that it is fairly easy to perform the assessment in a regionalized manner, which is es-

pecially important for particulate matter contribution, since this is, for the most part, a localized phenomena (though it is also well known that PM can also travel far and wide (Lin et al., 2014)).

The case study showed that on a per kg basis, MPs formed during the use phase and landfilling are negligible in their potential for PM formation with the slow degradation rates predicted by the degradation module. Littering of the LDPE, however, has the potential to have a high contribution to particulate matter forma-

Fig. 10. Contribution from each life cycle stage to the impacts of 1 kg of LDPE from cradle-to-grave for the plastic with a fast degradation rate (left) and slow degradation rate (right) with Danish waste treatment characteristics. Sensitivity was tested for only particulate matter formation.

Fig. 11. Monetized damage to human health per kg of mulch film. Derived from DALY valued as 110,000 USD2003 per DALY. Scenarios with -base at the end do not include additional impacts from plastic litter and MPs. Slow refers to a slow degradation of plastic in the environment (only UV), and fast corresponds to a faster degradation rate, including various degradation factors.

tion with a fast degradation rate and is not insignificant even with a slow degradation rate. The continued degradation of the particle distribution results in a large increase of particles in the PM_{10} and $PM_{2.5}$ ranges, but this increase does not show through in the LCA model, suggesting the PM formation impact category for both the ReCipe and ILCD methodologies may be highly unsusceptible to changes. The latter is a concern, since the LCIA methods routinely used for LCA should accurately represent the potential problems arising from PM. A CF factor of 0 for PM_{10} is incomprehensible given the evidence in literature for the importance of particles with aerodynamic diameters of $<10 \mu m$. Health problems arising from PM range from cardiovascular, to cerebrovascular and respiratory diseases, where small increases in PM have been shown to cause large increases in for example, cardiopulmonary mortality and respiratory admission (Anderson et al., 2012). However, due to this characterization approach, the ReCipe methodology underestimates the damage caused by PM by an order of magnitude lower compared to the ILCD method.

In terms of NMVOCs, plastic litter has the potential to be an important source of NMVOCs, which are precursors to tropospheric ozone formation and contribute to air pollution. This is true for LDPE which has been shown to degrade into compounds such as ethane and ethylene when exposed in air or water. If we contemplate the large quantities of plastic floating in the ocean and the recent predictions of future mismanage plastic worldwide, which are thought to reach 210 million tons on average per year by 2060 (Lebreton and Andrady, 2019) it becomes clear that even small quantities on a per kg basis present a potentially large diffuse source of NMVOCs and the same can be said for PM formation. In turn, this increases the need for appropriate regulation that takes emerging pollutants into consideration and takes necessary actions to prevent adverse health effects with e.g. the precautionary principal. The model presented here does not include MP and NP occurring during production and recycling of plastic products. A potential way to alleviate this data gap would be to require producers to declare any MP in their value chains, which would in turn help scientist create complete mass flow analysis of plastic losses and increase our understanding of impacts on both humans and ecosystem health. An important aspect of the model is the inclusion of a littering fraction, since it is evident that plastic leak to the environment regularly. This aspect, may help highlight alternative products with no MP potential, such as some types of biodegradable plastics and show this added benefit.

Furthermore, only a handful of VOCs have been quantified for the inventory in this work. Whereas, studies have shown the presence of harmful VOCs in commodity polymers (Even et al., 2019; Zimmermann et al., 2019) and naturally aged plastic products (Lomonaco et al., 2020). Moreover, effects from migration of toxic substances during the use phase of plastic products are an important aspect to include in future models. Evidence of these toxic substances inducing non-specific toxicity, from food contact materials, as well as for non-food contact materials, exists (Zimmermann et al., 2019). Furthermore, evidence of the presence of many chemicals that may cause endocrine disruption, oxidative stress and antiandrogenicity in both petro- and bio-based polymers is also available (Zimmermann et al., 2020). In terms of exposure assessments, it has been pointed out that though a large variety of

VOCs might be emitted from plastic material, different VOCs may trigger the same toxicological end points and thus, the mixture of VOCs might be more toxic than any one VOC distinctively assessed (Even et al., 2019). As Zimmermann et al (2020) point out, it is imperative to know what the plastic product formulation contains in order to be able to “design out” the negative properties, and it is impossible to do this as long as plastic formulations are kept as proprietary information, as they are today.

Furthermore, this work shows that particles of diameters $<10\ \mu\text{m}$ will be emitted to air where they will be a source of exposure. In this regard, the valuation of externalities presented in this work, which was estimated using a budget constrained ability to pay, may be useful in drawing attention to the high potential mismanagement plastics may have in terms of monetary loss due to an increase in hospitalization and other costly health interventions. However, more work is still needed on the valuation of plastic products on ecosystems. A valuation that includes ecosystem services might be better suited to show potential economic loss due to domino effects in the animal kingdom.

Many questions remain unanswered in relation to plastic losses in the environment and their impacts. Enabling comparison of different plastic types and their uses in LCA, which include impacts from secondary MPs, is of paramount importance in relation to environmental performance assessments of e.g. circular economic models and biotechnologies. Development of complex inventory models enabling and qualifying assessments of such complex systems, including elementary flows of biologically active materials and compounds, is however a long process stretching over several decades as illustrated for other inventory models, such as PestLCI i.e. an inventory model designed to quantify pesticide emissions from arable land, which has now been in development for over 3 decades (see Birkved and Hauschild, (2006); Dijkman et al., (2012); Tang et al., (2020)). Hence, this first attempt to include elementary flows of MPs and materials capable of generating MPs in the environment is somewhat similar to the first attempt to initially quantify pesticide emissions from arable land for LCA based on model predictions (Birkved and Hauschild, 2006). It is evident that models like SecuPlast and PestLCI have to be (and are) updated as new data and knowledge is provided.

This work shows a viable way to include some of the impacts derived from the formation of secondary MPs from plastic products through their life cycles, which can be adjusted as variable such as the degradation rate are clarified by other research. It is worth noting that the model is highly flexible in terms of input data. A starting point for the calculation can be the degradation rate ($\mu\text{g}/\text{year}$) from which point the model can calculate the dynamic evolution of PM as detailed in section 3.1.3. The latter is an important aspect, as it was seen that UV degradation alone results in lower degradation values than those seen in other work. A particularly pressing research need will, thereby, be to determine with higher accuracy the degradation rate to which plastic products are subject when mismanaged, since this was seen to be a sensitive parameter of the contribution to PM. Furthermore, more studies are necessary to evaluate which gaseous compounds currently being emitted from other polymers types which undergo degradation in the natural environment and to what extent these contribute to different impacts e.g. global warming, NMVOCs, PM, etc. Lastly, this work analyzed the potential of particles to cause damage due to their small size, but the chemical composition of particles is an important, however, unassessed factor that may worsen the toxicological effects of PM. The addition of other chemicals and additives into plastic products which may then be inhaled should continue to be explored and integrated to LCA models, which may then be useful in improving product design or in providing guidance to decision makers.

6. Conclusion

A straightforward framework to account for secondary MP formation, emission and impacts throughout the life cycle of plastic products was presented. The framework provides guidance on which factors to include in order to perform regionalized accounting and characterization of secondary MP impacts from an air pollution perspective, and the link between the elemental flows to endpoint damages by using existing LCIA methodologies. The case study showed that the contribution of MP to PM is low with a slow degradation rate, but may be an important contributor with a faster degradation rate that includes accelerating factors for polymer degradation in the natural environment. Furthermore, it was shown that small particles ($<10\ \mu\text{m}$) are emitted to air under various land roughness and even at low wind speeds, and have the potential to become a source of exposure to human health. Decomposition of plastic in the environment may also be an important potential source of NMVOCs, which might contribute to increase air pollution. A valuation of damages at LCIA endpoints showed that when scaled up to European level, even small increases of PM are costly for society and could potentially amount to millions of dollars per year in human health damages. Though the model includes inhalation impacts to human health due to MP formation, much work is needed to make a complete assessment of plastic impacts. Future research should focus on determining all physical parameters that affect the degradation kinetics of plastic products in nature. Furthermore, obtaining missing data on losses of MP during production and recycling of plastic products as well as, quantifying impacts to ecosystem is an important data gap that requires further work. In this regard, it would be valuable to provide a robust valuation method that includes ecosystem services that are affected due to uncontrolled losses of plastic. The continued integration of these factors into LCA will continue to benefit policy makers that can rely on LCA results in order to make the correct choices for future generations, in relation to eco-design of environmentally friendly plastic products.

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.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:[10.1016/j.spc.2021.07.008](https://doi.org/10.1016/j.spc.2021.07.008).

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