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An urgent call to think globally and act locally on landfill disposable plastics under and after covid-19 pandemic: Pollution prevention and technological (Bio) remediation solutions

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ABSTRACT

Landfilling and illegal waste disposal have risen to deal with the COVID-19 potentially infectious waste, particularly in developing countries, which aggravates plastic pollution and inherent environmental threats to human and animal health. It is estimated that 3.5 million metric tonnes of masks (equivalent to 601 TIR containers) have been landfilled worldwide in the first year, with the potential to increase global plastic municipal solid waste by 3.5%, alter biogas composition, and release $2.3 \times 10^{21}$ microplastics to leachates or adjacent environments, in the coming years. This paper reviews the challenges raised in the pandemic scenario on landfills and discusses the potential environmental and health implications that might drive us apart from the 2030 U.N. sustainable goals. Also, it highlights some innovative technologies to improve waste management (from collection to disposal, waste reduction, sterilization) and mitigates plastic leakage (emission control approaches, application of biotechnological and monitoring/computational tools) that can pave the way to environmental recovery. COVID-19 will eventually subside, but if no action is taken in the short-term towards effective plastic policies, replacement of plastics for sustainable alternatives (e.g., biobased plastics), improvement of waste management streams (prioritising flexible and decentralized approaches), and a greater awareness and responsibility of the general public, stakeholders, industries; we will soon reach a tipping-point in natural environments worldwide.

1. Introduction

Since early 2020, worldwide public health and economy have been severely affected by the COVID-19 pandemic, an acute respiratory disease caused by a highly infectious novel coronavirus - the SARS-CoV-2 (also known as coronavirus 2) [1]. By July 2021, the COVID-19 disease affected over 187 million people and caused 4.0 million deaths worldwide [2]. During this health crisis, the protection of lives and livelihoods has become a priority for governmental decisions and actions. The World Health Organisation (WHO), Centres for Disease Control and Prevention, and local governments have announced several guidelines to reduce the spread and health risks associated with COVID-19, including frequent home, regional or state-wide quarantine, restricted travelling, handwashing, and social distancing [3,4]. Besides, the use of personal protective equipment (PPE) such as surgical and medical masks, non-medical face masks (including self-made or commercial masks of cloth, cotton, among others) and face shields, was highly recommended for ordinary citizens. In contrast, other PPEs (including gloves, goggles) became mandatory for frontline health workers [5].

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As COVID-19 intensified all over the world, the use and consumption of PPE and other single-use-plastics (SUP) increased drastically, resulting in a massive upstream PPE supply chain disruption, a drawback on prevailing SUP bans or restrictions in several countries (e.g., Canada, some states in the U.S.A), and downstream waste disposal challenges [5]. Natural environments (e.g., beaches, rivers, seas), which at the beginning of the pandemic benefited from reducing litter and improved water quality from decreased tourism [6], are now becoming tainted with COVID-related waste [7]. A widely known example is related to the dozens of disposable masks observed in a 100 m stretch in Soko’s islands beach, Hong Kong (compared with only one or two items observed per month) [8]. In Kenya, this litter type was present in beaches under the concentration of 0.1 item m⁻² and represented 0.43% of total items but reached 55.1% in urban beaches [9].

Alongside, a significant share of COVID-19 plastic waste (particularly PPE, gloves, and plastic materials discarded by ordinary citizens as mixed waste) is being landfilled [10,11], instead of being incinerated as recommended/prioritised by several international and national organisations (as reviewed by Parashar et al. [12]). Landfills are, thus, becoming overloaded with COVID-19 waste, which (in the long run) can result in space crash, illegal dump, and the release of toxic pollutants [13]. This is of particular concern in developing countries, such as Cambodia, the Philippines, India, Indonesia; where uncontrolled landfilling and indiscriminate dumping were prevailing before COVID-19 [11,14,15]. In this sense, this article aims to address the challenges raised in the pandemic and post-pandemic world on landfills, including potential environmental and health implications that might drive us apart from the 2030 U.N. sustainable goals. Also, it highlights some innovative mitigation technologies and improved management strategies that can pave the way to environmental recovery. Such integrative but focused discussion (on landfills) has been missing in recent publications e.g., [5,6,9,12,13,16,17].

2. Plastic waste generation during covid-19 pandemic and implications on landfills

Lebreton and Andrady [18] predicted a world production above 200 megatons of municipal plastic waste in 2020 under a business-as-usual-scenario for plastic consumption, of which 43% would remain mismanaged (i.e., ending up in landfills, open dumps or littered in natural environments). However, the COVID-19 pandemic induced a significant change in waste dynamics and composition, mostly due to a considerable worldwide increment in infectious waste [12,19].

In March 2020, the WHO estimated a global demand of 89 million facemasks per month [20]; but such estimation soon became surpassed. One month after COVID-19 being declared a pandemic (i.e., on April 2020), Germany alone was demanding 17 million FFP facemasks and 45 million surgical masks per month [21]; and after eight months (in November) Germany’s Federal Ministry of Health was preparing to distribute 290 million masks to healthcare facilities until 25th December to suppress the second wave [22]. In Italy, and according to the information released on 12th November 2020 by the Extraordinary Commissioner on the free distribution to Regions and Autonomous Provinces, 1,040 million face masks were distributed by health personnel, law enforcement agencies, public service providers, Public Administration, nursing homes, local public transport and police; of which 57 million were used in seven days [23]. In the UK, 7,800 million PPE items have been distributed from March to November 2020 to health and social care services, namely adult social care providers, wholesalers, community pharmacies, dentists, and local resilience forums [24-26]. Added to these numbers is the voluntary or mandatory use of PPE by the public outside healthcare services, which significantly impact municipal solid waste (MSW) composition. In Saudi Arabia, facemask consumption has been estimated to be 5,336–38,426 million per year [27]. In Asia, face mask use can reach 2,300 million per day, resulting in 16,659 tonnes of medical waste per day [16]. Another significant contribution to plastic waste generation during the pandemic is related to disposable plastics for COVID-19 diagnosing. For instance, worldwide, 15,000 tonnes of waste was generated from Polymerase Chain Reaction (PCR) tests up to August 2020, with 97% of PCR material referred for incineration [19].

Plastic waste generated in health/medical care facilities, laboratories and other contaminated health/social facilities should be treated and managed in accordance with the international/country/state law on hazardous waste (e.g., EU law on waste, especially Directive 2008/98/EC, articles 17, 23, 24, and 25 on hazardous waste), i.e., be incinerated/disinfected followed by safe disposal (e.g., sanitary landfilling of the ashes). Whereas developed countries (or countries with high income and well-implemented/distributed incineration/waste-to-energy facilities) can manage COVID-19 medical waste properly. Some successful examples include South Korea, India, and Spain. After the first outbreak, the Ministry of Environment of South Korea released “the extraordinary measures for safe waste management and disposal”, which included (among other guidelines) daily incineration of COVID-related waste (where before could be saved for 7 days) [28]. China deployed mobile waste treatment stations along with the plan to convert industrial waste disposal plants into bio-medical waste (BMW) treatment facilities due to no industrial activity during the lockdown period; whereas in Catalonia, Spain, the existing incinerators facilities have put on the job to a priority disposal of medical waste [28].

Conversely, other countries struggled to follow such proper procedures, disposing of such medical waste in landfills or open dumps [12]. Some Asian countries (e.g., Thailand, Philippines, India) are known to dump solid wastes in open landfills due to the scarcity of resources for waste management, with an increased public health risk from the spread of infectious waste from the pandemic, in addition to the recurrent environmental problem [16].

Household solid waste (or municipal solid waste – MSW), on the other hand, should be double-bagged to be further incinerated (preferably) or, as the last resource, landfilled [3,12], with a minor share being potentially recycled after disinfection. Before the COVID-19 pandemic, landfilling was already the common practice of waste management all over the world, with developing (or low income) countries presenting a higher landfill rate (Fig. 1) [29], as it remains an easy, low-tech, and low-cost method compared to incineration and recycling [30]. Thus, such a waste disposal route remains popular for MSW during the COVID-19 pandemic [31].

At the beginning of this health crisis, particularly during the lockdown, the MSW decreased in most cities, particularly those with a higher tourism rate. For example, Milan (Italy) decreased MSW generation by 27.5% [32], and Barcelona (Spain) decreased by 25.0% [33]. Nevertheless, the world plastic share in MSW and MSW generation seemed to increase as the economy started to re-establish still under the persistence of COVID-19. For example, in Romania, the MSW amount increased ten times from the 26th of February to 15th June 2020 [34]. This is not surprising as the worldwide demand for, use, and consumption of PPE remain high to prevent transmission.

Based on the current consumption patterns, one can estimate a worst-case scenario waste generation resulting from disposable face-masks wearing. Considering a global population of 7,800 million, the average use of disposable masks by 73.7% of the population (based on updated information from Covid.wordometer.info), and the estimated need of 1 mask hab⁻¹ day⁻¹, this would translate into the necessity of 5,746 million disposable face masks per month (corresponding to 2,097 billion per year (see Table 1). Considering that PPE is mostly disposed in mixed waste and applying a global landfilling rate of 42% [35,36], it is expected that 293 thousand tonnes per month, or 3,524 thousand tonnes per year of disposable masks (4 g) might be landfilled worldwide, with greater pressure upon developing countries (e.g., India, Bangladesh) (Table 1). The worldwide monthly estimation of mask-related waste generation for landfills is actually in line with the 4,312 tonnes of COVID-19 related wastes (mostly collected as mixed waste) in Romania
from 25th February to 15th June 2020 (~39 tonnes day$^{-1}$; 1,176 tonnes month$^{-1}$) – which translated to the world population, it would result in 478 thousand tonnes month$^{-1}$ with 206 thousand tonnes month$^{-1}$ being landfilled) [34]. To PPE consumption adds up the single-use plastics (SUP, particularly plastic packaging), which was projected to grow by 5.5% due to pandemic response [37] as a result of postponed or withdrawal of several national, state-wide and/or international plastic policies [5].

In 2017, about 2,010 million tonnes of MSW were globally generated [37]. Thus, assuming a global landfill rate of 42% (resulting in 840 million tonnes), the PPE contribution to MSW would only be approximately 0.4%. However, concerns arise when considering the plastic share of MSW. According to the World Bank [38], the global share of plastic waste in MSW is about 12%, resulting in a contribution of 3.5% of PPE in the plastic share of MSW being globally landfilled in 2020. Notwithstanding, such contribution is variable. Countries such as Sweden will likely have < 1% of PPE contribution on plastic share in their MSW (which most of them was being recycled – mechanically or chemically); whereas in Portugal and Canada, such pressure can be higher > 4% Table I). In addition, according to the World Bank [38], the global landfilling rate can be even higher and aggravated in the coming years if no mitigation action is taken.

The increased pressure on landfilling (either from reducing recycling activities, the increased use of plastic packaging, and the general use of PPE) may compromise sustainable development goals. For instance, the European Union targeted landfilling to a maximum of 10% of MSW and recycling to a minimum of 65% by 2030 [39]. However, only in 2020, illegal plastic waste disposal has risen by 280% worldwide, and the global recycling rate is estimated to decrease by 5.1% [40].

3. Public health implications of landfilling in the post-covid world

Countries that relied on landfills as major disposal routes (e.g., Brazil, China, USA, India, Fig. 1) are, therefore, receiving intense loads of MSW daily (with a substantial contribution from PPE and SUP). Thus, such intense loads can exhaust landfills capacity, which likely results in space crush, plastic leakage, and leaching of toxic chemicals [13]. In addition, instead of attempting to reduce the amount of residues landfilled following a circular economy, the current pandemic may increase the need for more landfills, which require increased land use with higher entrenchment in the natural world.

A brief overview of landfills’ environmental implications (which affect human and animal health) can be depicted in Fig. 2.

3.1. Plastic leakage, dust generation, propensity for landfill fire

While environmental agencies recommend a daily coverage of residues in landfills (e.g., the Portuguese Environmental Agency [41]), this may not be possible in all cases around the world. Intense accumulation of waste on open landfills is known to provide breeding sites, burrows and nutrient supply for opportunistic species (e.g., rats; [42]). For instance, white storks (Ciconia ciconia) have been reported to feed on landfill sites, including in the short moments between the arrival and coverage of waste, with municipal waste comprising 68.8% of the diet of these animals in Spain (Avila, Salamanca, Zamora) [43]. A considerable amount of landfill waste was also observed in overwintering gull species (Herring gulls Larus smithsonianus, great Black-backed gulls Larus marinus, and Icelandic gulls Larus glaucoides) [44]. Thus, organisms relying on waste for food supply can end up entangled or ingesting plastic waste affecting their survival, feeding, health status, or fitness. The frequent ingestion of landfilled waste by the overwintering seagulls (although most is regurgitated) has been associated with a significant decrease in their reproduction and significantly increased chemical body burdens [44]. This is not surprising as plastics can absorb and carry heavy metals, organic compounds [45], and pathogens [46]. Despite its activity under landfill conditions not being currently known, SARs-CoV-2 can persist face masks for up to 21 days at 20 °C [47]. Thus, landfill waste can transport many contaminants and pathogens, posing a severe risk to animal and human health, especially when considering the role of larger organisms that may feed on this waste and act as carriers/vectors of pathogens. For instance, common seagulls in Porto, Portugal, are known reservoirs for multidrug-resistant Escherichia coli [48].

Public health hazards caused by open landfills are not limited to pathogens and adverse effects on biota. Landfilling generated dust and fires, contributing to unfavourable odour and air pollution around these sites [49]. Particulate matter with an aerodynamic diameter of >30 μm (such as microfibres released from masks; [50] can be carried out by wind up to 100 m from the source; whereas particles with a diameter 30–10 μm and <10 μm can be deposited as far as 250–500 m and 1 km,
Table 1
Estimated contribution by disposable masks to waste generation in several countries worldwide, and its share for landfills when considering the 2017 landfill rate. Data obtained from worldometer.com (population). OCDE.stat (landfill rate). Eurostat (landfill rate). Statistica.com (mask usage). World Bank (MSW and % plastics/country).

| Country       | % of masks usage | Population (millions) 2020 | Nr masks/day (kt/day)1 | Waste generation (kt/year) | Landfill rate (%) | Landfilled waste (kt/year) | MSW generated (kt) in 2017 | % plastics in MSW | Contribution of PPE to plastics share in MSW (%) | Landfilled containers Day⁻¹ |
|---------------|------------------|---------------------------|------------------------|---------------------------|------------------|---------------------------|-----------------------------|-----------------|-----------------------------------------------|-----------------------------|
| Australia     | 58.00            | 8.90                      | 5.16                   | 0.02                      | 7.54             | 52.88                     | 3.99                        | 13,751.00       | 7.61                                         | 0.72                        |
| Brazil        | 76.00            | 209.50                    | 159.22                 | 0.64                      | 232.46           | 72.55                     | 168.66                      | 79,890.00       | 13.50                                         | 2.16                        |
| Canada        | 78.00            | 37.59                     | 29.32                  | 0.12                      | 42.81            | 69.29                     | 29.66                       | 25,100.00       | 3.00                                         | 5.69                        |
| France        | 86.50            | 66.99                     | 57.95                  | 0.23                      | 84.60            | 21.59                     | 18.27                       | 33,400.00       | 9.00                                         | 2.81                        |
| Japan         | 76.00            | 126.47                    | 96.12                  | 0.38                      | 140.33           | 0.99                      | 1.39                        | 43,980.00       | 11.00                                        | 2.90                        |
| Italy         | 93.90            | 60.36                     | 56.68                  | 0.23                      | 82.75            | 25.71                     | 21.28                       | 29,560.00       | 11.60                                        | 2.41                        |
| Mexico        | 85.00            | 126.20                    | 107.27                 | 0.43                      | 156.61           | 66.46                     | 104.08                      | 53,100.00       | 10.90                                        | 2.71                        |
| South Africa  | 90.00            | 57.78                     | 52.00                  | 0.21                      | 75.92            | 29.00                     | 22.02                       | 18,460.00       | 7.10                                         | 5.79                        |
| Spain         | 96.40            | 46.96                     | 45.27                  | 0.18                      | 66.09            | 53.59                     | 35.42                       | 22,020.00       | 9.00                                         | 3.34                        |
| United Kingdom| 97.00            | 66.65                     | 64.65                  | 0.26                      | 94.39            | 16.87                     | 15.92                       | 30,910.00       | 20.20                                        | 1.51                        |
| USA           | 78.00            | 328.20                    | 255.99                 | 1.02                      | 373.75           | 52.16                     | 194.96                      | 243,720.00      | 9.50                                         | 1.61                        |
| Portugal      | 90.00            | 20.28                     | 18.25                  | 0.07                      | 26.65            | 49.58                     | 13.21                       | 5,000.00        | 10.72                                        | 4.97                        |
| Denmark       | 63.50            | 5.80                      | 3.68                   | 0.02                      | 5.38             | 0.84                      | 0.05                        | 4,720.00        | 1.61                                         | 7.8                         |
| Finland       | 54.10            | 5.50                      | 2.98                   | 0.01                      | 4.34             | 0.92                      | 0.04                        | 2,810.00        | 1.45                                         | 10.66                        |
| Norway        | 34.00            | 5.30                      | 1.80                   | 0.01                      | 2.63             | 3.47                      | 0.09                        | 3,950.00        | 2.25                                         | 2.96                        |
| Germany       | 74.60            | 83.02                     | 61.93                  | 0.25                      | 90.42            | 0.25                      | 0.23                        | 51,050.00       | 13.00                                        | 1.26                        |
| Sweden        | 12.10            | 10.23                     | 1.24                   | 0.01                      | 1.81             | 0.44                      | 0.01                        | 4,600.00        | 6.85                                         | 0.57                        |
| China         | 83.00            | 1,439.32                  | 1,194.64               | 4.78                      | 1744.17          | 72.55                     | 1,265.45                    | 220,400.00      | 9.80                                         | 8.08                        |
| World         | 73.70            | 7,800.00                  | 5,746.43               | 22.99                     | 8,389.79         | 42.00                     | 3,523.71                    | 2,010,000.00    | 12.00                                        | 3.48                        |

1) Assumptions: consumption of 1 mask per day; mask weight of 4 g (e.g. Missism Disposable 3 Layer Breathable Mask. Elastic EarLoop and Metal Nose Wire Clip); a compressed volume of 163 cm³ for 20 disposable masks (lab tested); Average container 6.69 m³.
Kt: thousand tonnes.
respectively [51]. As part of these particles, microplastics from landfills could be resuspended and contaminate the nearby areas, an issue that requires more attention in the future [52]. Airborne microplastic contamination in large cities is already recognised, for instance, with outdoor air concentrations of 0.9–4.2 microplastic particles m$^{-3}$ in Shanghai mostly originating from textile and abrasion of plastics [53]. Long-term exposure to high concentrations of airborne microplastics, or exposure of susceptible individuals, may lead to airway or interstitial inflammatory responses in the lung, coursing with dyspnoea [54]. Besides a public health threat, these airborne microplastics can deposit and contaminate other matrices, such as soil and water. In Yantai, China, a yearly deposition of airborne microplastics of 23 trillion particles or 0.9–1.4 tonnes is expected in 100 km coastline [55]. The contribution of landfill resuspension to airborne microplastics, and its impacts on public health and environmental contamination, have not yet been addressed and require further attention in future studies.

In addition to resuspension and direct release, landfills fires can also contribute to air pollution. Landfill fires, caused by heat released from intense aerobic biologic activity, will likely increase soon due to global warming and the increasing loads of COVID-19 waste. The World Health Organization (WHO) estimates that air pollution exposure causes 7 million deaths annually [56]. Both dust emissions and landfill fires are already known to significantly harm the environment and human health due to emissions of heavy metals, dioxins, PCBs, and furans [30]. Therefore, further landfilling of wastes and consequent generation of related air pollution in the surroundings can further exacerbate these numbers. Higher effects will be felt by communities surrounding landfilling sites, which may translate into an increased risk of low birth weight, congenital disabilities, and certain types of cancer [57]. Communities living near landfill sites are usually those with lower incomes, which are already burdened by many stressors (e.g., poor nutrition, lower access to health care), exacerbating social injustices.

3.2. Biogas and landfill leachates generation

Another environmental concern related to intense landfilled waste is the formation of biogas and leachates. Biogas starts forming 2–3 years after waste landflling due to waste degradation and relies on waste composition, environmental conditions, and landfill age [30]. Such process emits a considerable amount of greenhouse gases (GHG; 1.9% of global GHG in 2016), although it can be reduced with an efficient energy recovery facility usually required (e.g., Directive 31/1999/CE). However, in most countries, particularly in developing countries, uncontrolled landfills are prevailing [11,14]. Thus, the environmental footprint of landfills will likely be aggravated in the post-COVID scenario.

Disposable masks are mostly made with electrospun nanofibers from a diverse polymeric material (such as PP) and start losing properties (e.g., static electricity that confers the original filtering performance) when exposed to, for instance, water or moisture, losing their integrity and releasing micro- and nano-fibres along with hazardous chemicals as observed by Saliu et al., and Sullivan et al., [58,59]. As smaller are the plastic particles (e.g., micro- nano-sized), the higher is their potential to be biodegraded by microorganisms, and such biodegradation processes releases gas (mainly CO$_2$, and depending on their biobased content, also CH$_4$, H$_2$), likely contributing to landfill biogas. Recent studies highlight this hypothesis, with plastics biodegradation under simulated landfill conditions affecting biogas composition [60]. Thus, considering that the COVID-19 pandemic altered MSW that is now counting with significant contribution of PPE, it is likely to affect both biogases and leachates. In addition to their production and transportation, PPE landfilling contributes to additional GHG release, which should be further addressed through a Life Cycle Assessment (LCA) to pursue more sustainable alternatives and practices.

Leachates start forming after the first waste disposal and intense rainy seasons (particularly in poorly covered landfills), as they are resultant not only during biodegradation processes but also through desorption/lixiviation from solid wastes (plastics, metals, among others) [60]. World landfills can release on average 5 m$^3$ ha$^{-1}$ d$^{-1}$ of severely contaminated leachates [30], and their composition often consists of nutrients (primary nitrogen), pharmaceuticals, other organic compounds, heavy metals [61] and microplastics [62]. With billions of disposable masks (mostly composed of plastics) ending up in landfills, microplastics release will increase in the future. Disposable masks under mechanical abrasion (although in aquatic medium) evidenced the release of thousands of microfibres along with leachable metals (i.e., lead up to 6.79 µg/L, cadmium up to 1.92 µg/L, antimony up to 393 µg/L, and copper up to 4.17 µg/L) [58]. In simulated landfill environments, plastic wastes composed of PP and a PE and PP composite (which is, in fact, the major component of disposable masks) attained a weight loss of up to 10% during approximately one year [63]. Thus, landfilled disposable masks (made of polypropylene, PP) will fragment into micro- and nano-plastics and degrade through fluctuating temperatures and pH, deep-seated fires, physical stress, and microbial activities [64] releasing, concomitantly, leachable hazardous chemicals. A PP piece that has been landfilled for 5 years revealed colonisation
signs with viable microorganisms, oxidation confirmed by carbonyl and hydroxyl indexes, increased crystallinity, delamination, surface cracks, and the formation of microplastics of diameters under 0.4 – 6.9 µm [65]. While conditions vary with landfills, waste mixture, and each plastic type, a worst-case scenario of facemasks’ landfilling can be estimated. The previously 3,524 thousand tonnes per year of disposable masks (made mostly made of PP) that might be landfill in the world (Table 1), considering a decomposition of 10% weight over a year [63], would generate an amount of 2.3x10^21 microplastic particles (here assuming just the formation of particles with 7 µm in size) after a year of landfilling (see Supplementary Data).

Aside from hazardous chemicals, as previously mentioned, landfill leachates can contain considerable concentrations of pathogens, as the avian influenza virus (H6N2) that can remain infective from 30 to > 600 days in landfill leachates [66]. Thus, being hydrophobic particles with a resistant carbon backbone, such small-sized microplastics can carry hazardous chemicals and pathogens [67], while supporting the growth of biofilms/microbiota with a high abundance of antibiotic resistance genes [68]. This fact will exacerbate the adverse effects of microplastics on biota (e.g., [69]), affecting ecosystem services and functioning, and human health, when released to the environment.

Therefore, landfill leachates should be carefully processed to avoid aerosol formation during aeration or flushing in the leachate treatment plant [30], and avoid the release of potentially contaminated microplastics. Several technologies are available to treat landfill leachates (as well as wastewaters) via advanced oxidative treatments (e.g., ozonation), photocatalytic treatments, biological processes, physical–chemical processes, among others [70]. Nevertheless, these can be ineffective with some small-sized microplastics that can have adsorbed contaminants/pathogens, which calls for more research and innovative technology [62], as explored in the next section. Without a proper mitigation treatment, such emissions of pollutants (either solid, gas, or liquids) produced in solid urban waste landfill sites can last approximately three decades or even centuries after the landfill site is closed [71,72], with continuous loads to the surrounding environments [30].

3.3. Geomorphological implications

If we consider the predicted 5.8 billion masks of disposable face-masks consumed and discarded per day, of which 2.4 billion is eventually ending up on landfills, this will result in approximately 601 TIR containers landfilled daily around the world (Table 1). In small but highly COVID-19 impacted countries such as Portugal, it would result in 2.23 TIR containers being landfill daily. Along with the technogenic disasters above mentioned (e.g., landfill fires, chemical substance leakage, among others), landfills overload and increased number of illegal dumps during COVID-19 (as it is happening in developing countries such as India; [11]) will likely result in concerning morphological changes and geohistorical impacts of local character.

Landfill sites are often large underground structures with a complex mixture of municipal waste. Yet, most of them (particularly in the tropical and subtropical areas) are often placed in former sand, gravel or peat pits, wetlands, or waterlogged areas, where former excavations and drainage system complicates the collection of the leachate generated by the infiltrating precipitation [73]. Such deposits often result in the formation of landfill leachate plumes that impose a risk to downgradient water bodies; and consequently a threat to animal and human health [73]. Furthermore, when these underground structures encompass a greater land-use than expected (due to COVID-19 pandemic), they might also imply significant long-term geomorphic changes in various geomorphic features, such as riverbed and shoreline migration meanders and old riverbeds, as depicted in several geomorphometric analysis [74]. Such transformations of landscapes will eventually affect the ecological integrity of the area (including biodiversity loss) and interfere with the local microclimate.

4. Strategies to reduce covid – 19 plastic waste being landfilled

Even though a vaccination programme against COVID-19 had been accelerated in several countries (see https://vaccine-schedule.ecdc.europa.eu), it remains a slow process towards global herd immunity. Thus, the use of PPE and disposable plastics for COVID-19 diagnosis and treatment will prevail, at least in the following semester. Based on our predictions, the amount of plastic waste generated and mismanaged during the COVID-19 pandemic is staggering, mostly due to a lack of efficient planning and policy intervention on plastic waste management. This will aggravate plastic pollution worldwide if no action is taken immediately. Thus, it is imperative to start developing/implementing robust policies and sustainable approaches/initiatives to improve plastic waste management to reduce their adverse environmental and human health effects. The scientific community has presented several recommendations to governments, policymakers, corporate sectors, and the general public to overhaul the existing plastic waste management paradigm and motivate appropriate actions [12,75,76]. Among such recommendations, it is highlighted the need to decrease plastic waste generation and increase recycling, which eventually decreases landfills and open dumps, allowing the implementation of proper mitigation/remediation strategies.

4.1. Sustainable production and use of PPE and SUP

Several strategies can be put in use to reduce PPE and SUP waste generation significantly. Implementing strategies of public health protection beyond the use of PPE and SUP contribute to the reduction of waste production. For instance, the WHO recommends minimising the need for PPE through social distancing practices [77]. In healthcare, this translates into the use of telemedicine, physical barriers (e.g., glass windows), and restricted areas. The same principles can be applied to the general public by restricting the need to access public places (e.g., by implementing remote working).

Along with PPE, the use of plastics in packaging increased during the pandemic. Both cases (i.e., PPE and general SUP) can benefit from improvements in design, such as reducing the amount of plastic used or substituting it for eco-friendlier alternatives whenever possible. In the case of PPE, reusable alternatives (e.g., cotton masks) or treatment of disposable PPE allows for reuse (e.g., N95 masks can be decontaminated by steaming, [78]) can reduce the amount of waste produced while still contributing to public health protection. Another alternative is the substitution of disposable plastics for bio-based solutions. For example, wheat gluten biopolymer (a by-product or co-product of cereal industries) can be electrosprun into nanofibre membranes and subsequently carbonised at over 700 °C to form a network structure, which can simultaneously act as the filter media and reinforcement for gluten-based masks [79]. Such gluten material can be reinforced with very low amounts of lanosol (a naturally-occurring substance for microbes resistance; <10 wt%) together with the carbonised mat and shaped by thermoforming to create the facemasks [79].

Several biobased solutions are also available for other SUP, such as packaging that increased substantially during the COVID-19 pandemic. For example, poly- hydroxyalkanoates (PHAs) and homopolymers such as polyhydroxybutyrates (PHBs) extracted from algae biomass can present similar physicochemical properties as petrochemical plastics applied in such applications (e.g., polypropylene, polyethylene, and poly-ethylene terephthalate), with increased potential for biodegradation when desired (as reviewed by Patricio Silva [80]).

Other strategies can be put into place to reduce plastic waste (even general waste) for landfilling. Governmental regulations may support the reduction of landfill streams, and diversion to other alternatives, by applying landfill taxes to municipalities based on waste being landfilled [75], providing recycling benefits to consumers (e.g., buy-back programs for bottles), or applying higher fees to mixed wastes than recyclables in a door-to-door collection or by using smart trash containers.
4.2. Improve PPE and SUP recycling/repurposing

Implementation of structured waste management procedures, especially for the separate collection of COVID-19 pandemic wastes, is deemed necessary. For example, the use of colour-coded bags by individual households for the disposal of PPEs. Further, colour-coded bins must be deployed at the community level to ensure proper collection and disposal of such used PPEs. In Montreal, Canada, and Guimaraes, Portugal, specific PPE-trash containers have been installed in several places around the city to motivate ordinary citizens to safely dispose of their masks and consider their potential decontamination for further recycling/repurposing [82,83].

Disinfection procedures of plastic wastes as PPE (e.g., U.V., ozone, heat, microwave, autoclave) and/or a quarantine period (>72 h) can allow safe recycling [27,76]. China, for example, applied on-site/mobile treatment facilities such as Sterilwave SW440 (applying microwave sterilisation at 110 °C, with a treatment capacity up to 80 kg/h as reviewed by [28]). After disinfection, biomedical plastic waste no longer threatened public health and could follow regular waste streams for proper end-of-life for these materials. Masks collected in specific bins can be thermo-recycled at 190–230 °C [84] or 300–400 °C (pyrolysis) [85,86], allowing the conversion of the polypropylene into liquid fuels that can be further used as a source of energy with similar to fossil fuels. Otherwise, it can be used for pellets manufacturing to make boxes, trays, etc. (e.g., UBQ Materials and TerraCycling enterprise [87], or can be used to make pavements [88].

Improved infectious waste treatment during pandemics, or other emergencies, can be promoted by creating guidelines based on the waste storage facilities (avoiding their use whenever possible) and increasing incineration capacity by installing more facilities, co-processing with other wastes, or by mobilising private facilities [89]. Germany and Sweden were able to couple with the intense loads of potentially infected waste from the COVID-19 pandemic due to their well-developed and distributed incineration (waste-to-energy) facilities, only relying on landfilling to bury the ashes (<1%).

4.3. Encourage plastic waste recycling (even during a pandemic)

Recycling companies worldwide were already facing an economic crisis due to the low cost of virgin plastics production compared with recycled plastics. However, this situation was severely aggravated during COVID-19 incited by the fear of virus transmission. The life-span of the virus varies for different surfaces, remaining active for more extended periods on smooth surfaces [90,91]. However, several disinfectants are used to eliminate disease vectors while handling the waste (see [28] for more details). The application of such disinfection approaches can then allow safe recycling, which should be encouraged. A successful example comes from Hong Kong; which government introduced two bonus schemes to encourage waste recycling: (i) One-off Rental Support Scheme that allowed recycling facilities to pay 50% of their rent (or up to HKD$25,000); (ii) One-off Recycling Industry Anti-Epidemic Scheme that supports the operational costs of recycling facilities at a rate of HKD$20 000 per month [92].

5. Landfilled plastics – technological approaches for mitigation purposes

The implementation of a biorefinery located on landfill sites (or near them) will help reducing plastic waste on-site and, indirectly, the costs of waste-to-energy plants (consequently lowering logistical and supply chain costs related to waste transportation and lowering operating and capital costs by using existing infrastructure) [95]. Plastics shredding followed by thermal processing [95], Fenton oxidation processing [96], or biological pre-treatment (e.g., Pseudomonas sp., Bacillus cereus, Bacillus pumilus, and Arthrobacter; [97] are also relevant to increase the life expectancy of the site. Thermal processing allows energy recovery, whereas Fenton processing and biological pre-treatment will facilitate plastic waste biodecomposition after landfilled. Another strategy to reduce plastics for landfills (here, only the waste volume) involves plastic compactors. Such technology melts plastic waste into a disk, reducing water and consequently the surface area available for biodegradation, adsorption of contaminants, and leaching of monomers and additives [98]. All the previously mentioned approaches require, however, the separation of plastic waste from mixed wastes. This process might also require prior decontamination (e.g., the Microwave technique as implemented in Sterilwave SW440 mobile facility used in China) to avoid the spread of infectious diseases (such as COVID-19).

5.1. Reduction and/or pre-treatment of plastic waste before landfilling

The implementation of a biorefinery located on landfill sites (or near them) will help reducing plastic waste on-site and, indirectly, the costs of waste-to-energy plants (consequently lowering logistical and supply chain costs related to waste transportation and lowering operating and capital costs by using existing infrastructure) [95]. Plastics shredding followed by thermal processing [95], Fenton oxidation processing [96], or biological pre-treatment (e.g., Pseudomonas sp., Bacillus cereus, Bacillus pumilus, and Arthrobacter; [97] are also relevant to increase the life expectancy of the site. Thermal processing allows energy recovery, whereas Fenton processing and biological pre-treatment will facilitate plastic waste biodecomposition after landfilled. Another strategy to reduce plastics for landfills (here, only the waste volume) involves plastic compactors. Such technology melts plastic waste into a disk, reducing water and consequently the surface area available for biodegradation, adsorption of contaminants, and leaching of monomers and additives [98]. All the previously mentioned approaches require, however, the separation of plastic waste from mixed wastes. This process might also require prior decontamination (e.g., the Microwave technique as implemented in Sterilwave SW440 mobile facility used in China) to avoid the spread of infectious diseases (such as COVID-19).

5.2. Acceleration of microbial degradation of landfilled plastics (including PPE)

Bioreactor technology is already a reality in several modern landfills, and it uses enhanced microbiological processes to transform and stabilise MSW constituents within 5–10 years, significantly increasing the organic waste decomposition, conversion rates, and process effectiveness than conventional landfills [99]. Bioreactors can operate under aerobic, aerobic-anaerobic or anaerobic conditions, where waste (including plastics) are converted to gas with energy recovery. Anaerobic landfill bioreactors allow a faster degradation, and biogas formed has a high methane concentration, but it also produces hydrogen sulphide and high ammonia levels compared to aerobic bioreactors [100]. Nevertheless, and independently of the bioreactor type, this technology extends the useful life of landfills by reducing, for instance, the need to site new facilities as biodegradation occurs. Such technology can be even improved for plastics degradation with the help of key microorganisms. Different actinomycetes, algae, bacteria, and fungi have proven to degrade persistent plastics by converting them into environmentally friendly carbon compounds, with key enzymes identified (see recent reviews such as [101–103]). So far, plastics degradation proved to be more efficient in the presence of a microbial consortium, such as Actinobacteria with Firmicutes (which are already present in anaerobic digesters, [64]) and Bacillus sp. and Pseudomonas sp.; and Brevibacillus agri (2 strains), Brevibacillus brevis, and Aneurinibacillus aneurinilyticus (with high potential for aerobic bioreactors) - where some bacteria use monomers and excrete byproducts that become substrates for others to grow [101–103]. Key enzymes involved in the biodegradation process includes laccase, manganese-dependent peroxidase and hydrolase (urease, protease, lipase). The degradation rate of plastics by naturally occurring microbes remains a relatively slow process, as it depends on several factors (e.g., polymer characteristics and environmental factors). A potential solution is modifying key enzymes through protein engineering to design microbial strains with better degradation efficiency. However, this approach requires in-depth knowledge on the biochemical and structural properties of such vital enzymes involved in plastics
biodegradation, which remains so far poorly covered. In addition, pre-
treatments and additives (e.g., nanoparticles) also seem to play a role
when improving microbes performance towards plastics degradation,
which also needs special attention [103].

5.3. Control and treatment of emissions (landfill gas and leachates)

With MSW receiving more plastic waste (PPE related), it is expected
that biogas formation will be affected. For instance, the presence of
HDPE, PP, and PS on food waste inhibited biogas production in anaer
obic digesters [104]. Yet, the structure of the plastic includes a carbon
backbone; thus, its biodegradation in landfills (which occurs at a lower
rate) will increase the share of CH$_4$, H$_2$, and ultimately CO$_2$ in a longer
run, along with potential other volatile compounds (e.g., added as ad-
ditives). Parallely, the presence of persistent plastics in landfill
conditions will fragment (before being microbiologically degraded),
originating microplastics (i.e., plastic debris < 5 mm in size), hazardous
chemicals from additives, and non-intentionally added substances that
will enter leachates constitution. These particles have already been re-
ported in landfill leachates from northern European countries and in
China [105,106], which raises concerns as these small particles are
known vectors of hazardous contaminants and pathogens [62].

By law, sanitary landfills should control and treat biogas and
leachates, but the technological approaches are mostly dependent on the
infrastructures’ financial support. The first approach to control leach-
ates formation and, to some extent, biogas release relies on selecting
landfill cover and multilayer liners. For example, novel technological
applications in the construction of multilayer liners involve the combi-
nations of waste (e.g., compacted plastics and fibre material [107]; and
geosynthetic materials (e.g., geosynthetic clay, granular bentonite,
additives). Parallelly, the presence of persistent plastics in landfill
applications in the construction of multilayer liners involve the combi-
nations of waste (e.g., compacted plastics and fibre material [107]; and
geosynthetic materials (e.g., geosynthetic clay, granular bentonite,

Table 2
Overview of the main (bio)technological approaches that are/can be implemented on landfills for biogas purification for further use, leachate microplastics removal, and ex-situ/in-situ plastic-waste bioremediation.

| Traditional approach |
|----------------------|
| Physical absorption (e.g., high pressured water scrubbing) |
| Chemical absorption (e.g., amine swing absorption) |
| Adsorption (organic: activated carbons, and inorganic: silica gels, aluminas, zeolites adsorbents) |
| Artificial soil filtration (~98%, size 50-500 µm)$^a$ |
| Sand bed filtration (70%, size 50-500 µm)$^b$ |
| Coagulation/floculation (47.82%, size < 1.2 µm)$^b$ |
| Coagulation/floculation + sedimentation + sand filtration + activated carbon filtration (81%)$^b$ |
| Coagulation/ floculation + flotation + sand filtration + activated carbon filtration (83%)$^b$ |
| Electrochemical oxidation$^b$ |
| Dissolved air flotation (up to 95%)$^b$ |
| Oxidation dich (up to 97%, size > 25 µm)$^b$ |

| Existing advanced approaches |
|-----------------------------|
| Membrane technology (only limited to small landfills) |
| Ozone (90%, particularly at temperatures > 35 °C)$^c$ |
| Conventional activated process (up to 95.6%, size 20-5000 µm)$^c$ |
| Sequencing Batch Biological Reactor (100%; size 50-500 µm)$^c$ |
| Membrane biological reactor (up to 99%, size > 20 µm)$^b$ |
| Reverse osmosis (90% removal)$^b$ |
| Advanced oxidation processes(e.g., photo-Fenton, O$_3$/UV, H$_2$O$_2$/UV, ultrasound (US), UV/US, and H$_2$O$_2$/US) |
| Ultrafiltration (42%)$^b$ |
| Dissolved air flotation (up to 95%)$^b$ |
| Microscreen filtration with disc filters (DFDS)$^b$ |
| Electrocoagulation (up to 99% at pH 7)$^b$ |

| Emerging approaches |
|--------------------|
| Cryogenic separation (-170°C, 80 bar)$^d$ |
| Biological upgrading methods$^d$ |
| Thermochemical technologies using supercritical water$^d$ |
| Dynamic membranes (99%)$^d$ |
| Biological degradation$^d$ |
| Photocatalysis (e.g., 98.40% for 400-nm OS)$^d$ |
| Constructions wetlands (88%)$^d$ |
| Granular activated carbon filtration, high doses of FeCl$_3$, H$_2$O/AlCl$_3$, H$_2$O, and ultrafiltration through polyvinylidene fluoride (PVDF) membranes (up to 40%)$^d$ |

$^a$ Tested in landfill leachates.
$^b$ Tested in wastewater treatment plants and water treatment plants but have the potential for landfill leachates.
$^c$ Untested.
$^d$ Tests in laboratory conditions.
Several (bio)technologies proved efficiency in collecting and processing/treating biogas and leachates, which has been scrutinised in recent critical reviews towards their efficiencies and drawbacks for cleaning and upgrading steps (e.g., [113]). For biogas, upgrading technologies for purification and concentration processes for its further use in numerous applications (e.g., electricity, liquid gas, fuel) include water scrubbing, cryogenic separation, physical absorption, chemical absorption, pressure swing adsorption, membrane technology, and biological upgrading methods (Table 2). Assuming that the presence of PPE (which composition is mainly PP, PE) will likely contribute to the increment of CO₂, CH₄ and heavy metals emissions [58,114], the most efficient technological approach highlighted in the literature to treat biogas enriched with such gases is chemical absorption scrubbing [113]. Such an approach achieves the highest purity for biomethane (CH₄ > 99%) with low losses (<0.1%) and high carbon dioxide (CO₂) elimination, all this without the need for pressurisation. Yet, it requires high investment, heat demand for regeneration, and it often undergoes corrosion and salt precipitation [113]. Cryogenic separation also allows a high purity for CH₄ and CO₂ is obtained as a byproduct [113]. However, it implies high costs for capital and operation, and it still under development to be implemented at a larger scale (such as landfills). For low-income countries, the cheapest (and easy to use) technology is adsorption (e.g., granular activated carbon, zeolites, metal-organic framework), which allow adsorbing relatively high quantities of CO₂ (dominant gas in aerobic landfill gas) and CH₄ to a greater extent under anaerobic conditions [100]. Nevertheless, the success of this application relies on low/absent moisture conditions.

Several treatments are also available for leachate treatments with the potential to remove microplastics, which includes photophysical and chemical processes, coagulation, reverse osmosis, dynamic membrane filtration, bioreactors/biological degradation, sequencing batch reactors, among others (Table 2). Among them, the sequencing batch reactor proved high efficiency (100%) for the removal of microplastics > 50 μm in size from landfill leachates [105]. Yet, such a technological approach has low efficiency in removing pathogens, requires skilled personnel and dependence on uninterrupted power supply (high maintenance). Other possibilities include microplastics photocatalytic degradation (e.g., with zinc oxide), which stands out as a viable and energy-efficient method (which also removes plastics at a nanoscale) [115]. However, some end products from photocatalytic degradation may impose a risk to both animal and human health. A solution may be among the countries with high income, its success in biomedical waste management, particularly during pandemic scenarios, to avoid overloading such facilities and illegal dumping. Although South Korea be among the countries with high income, its success in biomedical waste management relied on the implementation of extraordinary and tightened measures for safe waste disposal and management (previously applied against MERS) against COVID-19 even before being considered pandemic (i.e., January 28, 2020). Through a volume-based waste fee system (VBWFS) for MSW, South Koreans could purchase standard coloured bags for each type of waste (e.g., yellow for food waste; blue for general waste). During COVID-19, they had garbage bags labelled “waste for incineration” (here to include PPE) and “waste bag for landfill” still through the VBW system. This helped managing which waste was following to landfills while imposing correct public behaviour. Hong Kong, Korea, and Japan introduced bonus schemes to encourage waste recycling. In Wuhan, China, and Bangkok, Thailand, implemented specific bins “for facemasks only” (as implemented in Canada and Portugal) allow to collect masks for correct end-of-life, including repurposing safely. A similar strategy worldwide implemented, allied with a significant engagement of ordinary citizens, and basic infrastructure establishment and capacity improvement of the new proposed design of medical contaminated waste treatment would reduce the PPE amount going to landfills.

In addition, governmental actions should include the reinforcement of 3R’s (reduce, reuse, recycle) policies by implementing incentive/reward programs; engagement of the general public on recycling activities including PPE by providing specific bins for new recycling streams for such equipment; reorganization of municipal solid waste collection and handling strategies to promote recycling and make up for

5.4. Implementation of integrative monitoring programs

Along with the in-situ monitoring studies (mandatory in most countries; e.g., see [117] to assess quantity and composition of landfill biogas and leachates, it is also crucial to address their potential environmental risk. For this purpose, the implementation of frequent aerial, geomorphological/geodetic and/or geoelectrical surveys can provide essential insights on the impacts of landfills on their surrounding environments [118], along with the spatial displacement of landfill areas [119], and spread of contaminated plumes [120]. These studies can then be allied with integration software (e.g., RES2DINV ERT and Oasis Monta) modelling software, as serving as a proficient metric for delineating landfills’ impact on humans, ecosystems, and water-bearing structures, both at the ground surface and underground features [120]. Such risk assessment studies should be coupled to other metrics such as Plastic Waste Footprint (i.e., metrics that encompasses the impact of plastic on natural resources and contribution to greenhouse emission, plastic pollution, and climate change) and used as a tool for decision making/policy creation and public engagement, as it provides a numerical form of environmental burdens for use by non-specialists [121].

6. Final remarks

COVID-19 pandemic has led to significant disruption in plastic waste management, with severe environmental challenges. Landfills have been the most recurring disposal technology to deal with COVID-19 pandemic plastic waste that goes along MSW, particularly in developing countries. This is of particular concern when forecasting future pandemics scenarios, as they proved to be recurrent. It is time to rethink plastics (prioritising bioplastics) and current plastic waste management strategies while improving waste collection and treatment facilities and implementing strong and effective plastic policies towards a circular bioeconomy and environmental sustainability.

Landfills extinction is still a long road ahead, especially for developing countries as they have limited financial support to implement and prioritise recycling and waste-to-energy options. Thus, it is crucial for such countries to enforce and provide good policies and guidelines on MSW management, particularly during pandemic scenarios, to avoid overloading such facilities and illegal dumping. Although South Korea be among the countries with high income, its success in biomedical waste management relied on the implementation of extraordinary and tightened measures for safe waste disposal and management (previously applied against MERS) against COVID-19 even before being considered pandemic (i.e., January 28, 2020). Through a volume-based waste fee system (VBWFS) for MSW, South Koreans could purchase standard coloured bags for each type of waste (e.g., yellow for food waste; blue for general waste). During COVID-19, they had garbage bags labelled “waste for incineration” (here to include PPE) and “waste bag for landfill” still through the VBW system. This helped managing which waste was following to landfills while imposing correct public behaviour. Hong Kong, Korea, and Japan introduced bonus schemes to encourage waste recycling. In Wuhan, China, and Bangkok, Thailand, implemented specific bins “for facemasks only” (as implemented in Canada and Portugal) allow to collect masks for correct end-of-life, including repurposing safely. A similar strategy worldwide implemented, allied with a significant engagement of ordinary citizens, and basic infrastructure establishment and capacity improvement of the new proposed design of medical contaminated waste treatment would reduce the PPE amount going to landfills.

In addition, governmental actions should include the reinforcement of 3R’s (reduce, reuse, recycle) policies by implementing incentive/reward programs; engagement of the general public on recycling activities including PPE by providing specific bins for new recycling streams for such equipment; reorganization of municipal solid waste collection and handling strategies to promote recycling and make up for
the new PPE recycling streams; improvement of waste management facilities (priority should be given to flexible and decentralized approaches) through effective financial mechanisms; promotion of a sustainable assessment of technologies (SAT) for Best Available Technology (BAT) for waste treatment/management considering their technical, social, and economic aspects, along with the environmental performances. For instance, innovative and effective (bio)technologies and computational tools to improve landfills are already available and will continue to advance exponentially, but they must be prioritised in forthcoming financial programs. Alongside, monitoring and risk assessment of the impacts of landfills on-site on their surrounding environments is recommended, along with the implementation of frequent aerial and geomorphological surveys, to develop strict guidelines, limits and contingency plans.

Synergisms between academia-governments-stakeholders is also fundamental to develop sustainable alternatives and implement active mitigation and remediation measures. Equal importance is given to the general population’s involvement in education and science dissemination programs to support sustainable behaviour (e.g., preference for bio-based products, prioritizing recycling to close the loop), elucidate the environmental issues related to plastic pollution, and help phasing-out landfills by entailing a circular bioeconomy.

CRediT authorship contribution statement

Ana L. Patricio Silva: Visualization, Writing - original draft, Writing - review & editing. Joana C. Prata: Visualization, Writing - original draft, Writing - review & editing. Armand C. Duarte: Conceptualization, Supervision, Writing - review & editing. Damia Barcelo: Conceptualization, Supervision, Writing - review & editing. Teresa Rocha-Santos: Conceptualization, Supervision, Writing - review & editing.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

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