

Prospective Dynamic and Probabilistic Material Flow Analysis of Graphene-Based Materials in Europe from 2004 to 2030

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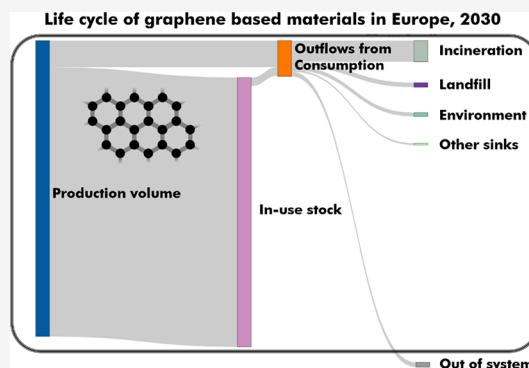
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ABSTRACT: As industrial demand for graphene-based materials (GBMs) grows, more attention falls on potential environmental risks. The present article describes a first assessment of the environmental releases of GBMs using dynamic probabilistic material flow analysis. The model considered all current or expected uses of GBMs from 2004 to 2030, during which time there have already been significant changes in how the graphene mass produced is distributed to different product categories. Although the volume of GBM production is expected to grow exponentially in the coming years, outflow from the consumption of products containing GBMs shows only a slightly positive trend due to their long lifetimes and the large in-use stock of some applications (e.g., GBM composites used in wind turbine blades). From consumption and end-of-life phase GBM mass flows in 2030, estimates suggest that more than 50% will be incinerated and oxidized in waste plants, 16% will be landfilled, 12% will be exported out of Europe, and 1.4% of the annual production will flow to the environment. Predicted release concentrations for 2030 are 1.4 ng/L in surface water and 20 $\mu\text{g}/\text{kg}$ in sludge-treated soil. This study's results could be used for prospective environmental risk assessments and as input for environmental fate models.

KEYWORDS: graphene, graphene-based material (GBM), material flow analysis, exposure modeling



INTRODUCTION

Since single graphene layers were first isolated, in 2004, they have been applied to numerous fields. Thanks to its high thermal conductivity¹ and electron mobility,² graphene can be used in composite materials, batteries, supercapacitors, and conductive inks. Due to their high chemical inertness, extreme mechanical strength, and high electrical conductivity, coatings made with graphene-based materials (GBMs) provide improved anti-erosion properties and resistance against lightning strikes to wind turbine blades.^{3–6} Although graphene oxide is the GBM most frequently investigated for its potential use in coatings,⁶ other forms (e.g., hydroxyl-functionalized graphene nanoplatelets) have also attracted interest for application in wind turbine coatings.^{4,5} The oil and drilling industries are also expected to benefit from GBMs.^{7,8} Adding GBMs can improve the rheology, fluid loss control, and lubricity of drilling fluids.⁹ Graphene's gas impermeability and adsorbability enable its use in filters.^{10,11}

Although the International Union of Pure and Applied Chemistry clearly defines graphene as a single layer of carbon atoms, the term “graphene” is often applied generically to a range of GBMs. To remedy this situation, the European Union's Graphene Flagship initiative has proposed a GBM classification framework based on the number of graphene

layers present, the average lateral dimension, and the atomic carbon/oxygen ratio.¹²

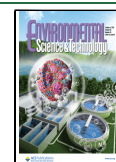
With its potential for use in a vast range of applications, the numbers of publications and patents concerning GBMs have increased exponentially.¹³ This is expected to continue in the coming years in tandem with the rapid growth in GBM research and development (R&D), increased industrial production capacity, and lower manufacturing costs.^{14,15} However, increased GBM use raises concerns about its potential release into the environment. When innovative technologies develop rapidly, progress toward suitable governance often lags behind.¹⁶ The safe and sustainable by design (SSbD) concept aims to involve safety considerations at an early stage of innovation processes to fill those gaps.^{16,17} Graphene technologies are still in the early stages of their development,¹⁸ and therefore, performing risk assessments based on SSbD approaches is suitable.

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Life cycle assessment (LCA) is one of the methods used to assess the environmental impact of products or materials from production and manufacturing through their use phase and on to end-of-life (EoL).¹⁹ Arvidsson and Molander reported that as the scale of graphene production increases—from the laboratory and pilot projects to the industrial scale—its overall environmental impacts are expected to decrease.²⁰ The majority of researchers and innovation advisors do not currently consider graphene a risk to the environment.²¹ However, based on comparisons with competing materials, such as graphite, molybdenum, and indium tin oxide, Beloin-Saint-Pierre and Hischer suggested that some of its uses were not the best environmental options.²² Although LCA is a useful tool for evaluating environmental impacts, the application of LCA to manufactured nanomaterials still shows limits due to the lack of life cycle inventory data and nano-specific characterization factors for toxicity-related impacts.²³

Environmental risk assessment is another tool for evaluating environmental impacts of materials, combining exposure and hazard assessment.²⁴ A number of ecotoxicity studies examining GBMs have shown their toxicity to various organisms, including bacteria, algae, invertebrates, and vertebrates.^{25–29} The GBMs that showed the greatest antibacterial activity were graphene oxide and reduced graphene oxide.³⁰ *Caenorhabditis elegans* also demonstrated different toxicity reactions to different forms (e.g., graphene oxide and reduced graphene oxide).³¹ On the other hand, Mottier and colleagues compared the effects of normalized size, mass concentration, and surface area on growth inhibition among amphibian larvae,³² concluding that the specific surface area of the carbon nanomaterials was the best descriptor of toxicity in that population.

As the second pillar of any environmental risk assessment,³³ an environmental exposure assessment should be performed to estimate predicted environmental concentrations. To the best of our knowledge, there were no previous predicted environmental concentrations for GBMs. The basis for quantifying environmental concentrations is information on the sources of the materials to the environment. Material flow analysis (MFA) is the established method of assessing the flows and stocks of materials throughout their life cycles³⁴ and quantifying environmental releases. It can also be used to estimate simplified predicted environmental concentrations that can be seen as the initial release concentration without consideration of transformation processes and final fate.³⁵ Gottschalk and colleagues were the first to fully incorporate probabilistic aspects into MFA, creating probabilistic material flow analysis (PMFA).³⁶ Dynamic probabilistic material flow analysis (DPMFA) was developed to consider the dynamics of the production volumes of materials and/or their transfer coefficients.^{37–42} DPMFA has been used to predict future flows of nanomaterials,³⁵ e.g., by considering different future use scenarios⁴³ or by modeling flows of novel materials, such as nanocellulose with high expected production growth rates.⁴⁴

Given the absence of data on environmental flows and exposure, the present study aimed to provide a basis for future environmental risk assessments of GBMs by estimating their release using a DPMFA and determining predicted release concentrations in different environmental compartments.

METHODS

Dynamic Probabilistic Material Flow Analysis. The present study used the DPMFA Python package originally developed by Bornhöft and colleagues,³⁸ which subsequent studies improved^{37,39} to account for temporal variations of production volumes and transfer coefficients. The present work performed an exposure assessment of graphene, multilayer graphene, graphene oxide, reduced graphene oxide, graphene nanosheets, graphene nanoribbons, and graphite nanoplatelets, sheets, and flakes, and we modeled flows of these materials across the European Union (EU-27), Switzerland, Norway, and the United Kingdom from 2004 to 2030. In this study, this spatial system boundary is referred as “Europe”. The complete life cycles of products containing GBMs were assessed from production and use to EoL. The model comprises seven technical compartments and five environmental compartments (Figure S1). It should be noted that “Elimination” refers to GBM transformation into carbon dioxide through oxidation. Two outflows from consumption account for the immediate outflow from consumption and the delayed release through the in-use stock. The codes, input files, and raw results are available for download at Zenodo.⁴⁵

Data Collection. Production Volume Estimation. Graphene production volume estimates used two sources. Future Markets’ Graphene Report 2020 provided low, medium, and high estimates for worldwide graphene production demand from 2018 to 2030.⁴⁶ Additionally, the Graphene Flagship initiative¹⁴ deliverable reported minimum, mean, and maximum estimates for global graphene production demand from 2010 to 2025. Other studies providing production estimates for graphene are available.⁴⁷ However, they did not specify the source of the data or the data collection method. Therefore, the current study considered only the Future Markets’ Graphene Report 2020 and the Graphene Flagship initiative deliverable.¹⁴ When both data sources were available for the same year, we calculated the inputs’ minimum, average, and maximum values to estimate the lower boundary, average, and upper boundary of annual worldwide graphene production volumes. For 2018–2030, both data sources were available. From 2004 to 2017, there were either no inputs or just a single input from the Graphene Flagship; therefore, we extrapolated graphene demand by assuming that the graphene demand curve’s slope reflected the slope of the yearly increase in the number of graphene patents.¹⁸ Worldwide graphene production demand in 2004—the year of graphene’s discovery⁴⁸—was set at zero. The lowest data variability (standard deviation divided by mean) was found in the years when only a single data input or no data input was available. European production volumes were scaled from worldwide estimates. The Graphene Flagship initiative reported Europe’s past and projected world market share in graphene, which was about 36% in 2015 and projected to decrease to 23% in 2025.¹⁴ To complete the time series, we assumed that this market share would remain at 23% until 2030. We also assumed that Europe’s world market share of graphene from 2004 to 2014 was the same as in 2015. Details of the European production volume estimates are summarized in the [Supporting Information](#).

Allocation to Product Categories. This study made changes to some product category allocations over time ([Supporting Information](#)). During graphene’s early developmental stage, most of its production volume was allocated to R&D ([Supporting Information](#)); however, the product

Table 1. Transfer Coefficients for Graphene-Based Materials (GBMs) during Their Use Phase, by Product Category^a

Product category	Reference, relevant material	Use release	Use release duration (years)	Use release schedule				Allocation after use release									
				X	Y1	Y2	Y3	Y4	...	Waste water	Air	Subsurface	Surface water	Roadsides	Elimination	Soil	
Sporting goods	Expert opinion	0															
Batteries	[40], CNT	0															
R&D	[54], CNT	0.05	1	1													
Electronics	[40], CNT	0															
Automotive	[42], CNT	0															
Paints and coatings	[55], CNT	0.01	7	0.9		0.1*(1/6)											0.25
Textiles	[40], CNT	0.03	3	0.5	0.3	0.2											
Drilling fluid	Expert opinion	0.1	1	1													
Medical applications	[56], nano-SiO ₂	0															
Tires	[41], tires	0.2112	4	0.25	0.25	0.25	0.25	0.25									
Construction	[42], nano-TiO ₂	0.01	80	0.9		1/79											
Filtration	Expert opinion	0.01	1	1													
Aerospace	[42], CNT	0															
Wind turbines	Expert opinion	0															

^aX values in the "Use release" column show the fraction of a product's GBM released during the use phase. The sum of "Use release" shown in this table and "EoL release" shown in Table 2 for each product category is 1. "Use release duration" is the number of years during which release occurs. "Use release schedule" values show the proportion of the total GBM released after a product enters the system in years 1 to 4. "Allocation after use release" describes where GBMs are released to during their use phase. Abbreviations: R&D: research and development, CNT: carbon nanotubes, TiO₂: titanium dioxide, SiO₂: silicon dioxide.

Table 2. Transfer Coefficients for Graphene-Based Materials (GBMs) during Their End-of-Life (EoL), for Various Product Categories^a

Product category	Reference, relevant material	EoL release		Lifetime distribution (normal) ($\sigma = \text{stdev}$)	Allocation to waste categories														
		1 - X	X		CDW	WindturbW	MMSW	WEEE	TextW	BattW	MedW	AviW	AutoW	TireW	DrillW				
Sporting goods	Expert opinion	1		Mean = 10; $3\sigma = 8$			1												
Batteries	[40], CNT*	1		Mean = 15; $3\sigma = 5$										1					
R&D	[54], CNT	0.95		$Y1 = 1.0$			1												
Electronics	[40], CNT	1		Mean = 8; $3\sigma = 8$					1										
Automotive	[42], CNT	1		Mean = 12; $3\sigma = 5$															1
Paints and coatings	[55], CNT**	0.99		Mean = 80; $3\sigma = 20$			0.34		0.05		0.26	0.06		0.1		0.18			
	[40], CNT	0.97		Mean = 3; $3\sigma = 2$								1							
Drilling fluid	Expert opinion	0.9		$Y1 = 1.0$															1
Medical applications	[56], SiO ₂	1		Mean = 8; $3\sigma = 5$										1					
Tires	[41], tires	0.7888		Mean = 4															1
Construction	[42], nano-TiO ₂	0.99		Mean = 80; $3\sigma = 20$			1												
Filtration	Expert opinion	0.99		$Y1 = 1.0$					1										
Aerospace	[42], CNT	1		Mean = 20; $3\sigma = 5$															1
Wind turbines	Expert opinion	1		Mean = 2 0; $3\sigma = 5$						1									

^a“1 - X” values in the “EoL release” column are the proportion of GBMs released during the product’s EoL. The sum of “Use release” shown in Table 1 and “EoL release” shown in this table, for each product category, is 1. Product lifetimes were assumed to be normally distributed. “Allocation to waste categories” describes how products are treated when they come to their EoL. “Allocation to waste categories” values indicate the proportion of material sent to each treatment. Abbreviations: R&D, research and development; CNT, carbon nanotubes; CDW, construction and demolition waste; WindturbW, wind turbine waste; MMSW, mixed municipal solid waste; WEEE, waste electrical and electronic equipment; TextW, textile waste; BattW, battery waste; MedW, medical waste; AviW, aviation waste; AutoW, automotive waste; TireW, tire waste; DrillW, drilling waste. *Modified; the original allocation covered batteries and sensors; therefore, in the current study, this is all allocated to battery waste. **Modified; the original allocation covered metals, which are not relevant for graphene, and therefore, the 10% that was allocated to the marine metals is allocated to aviation waste.

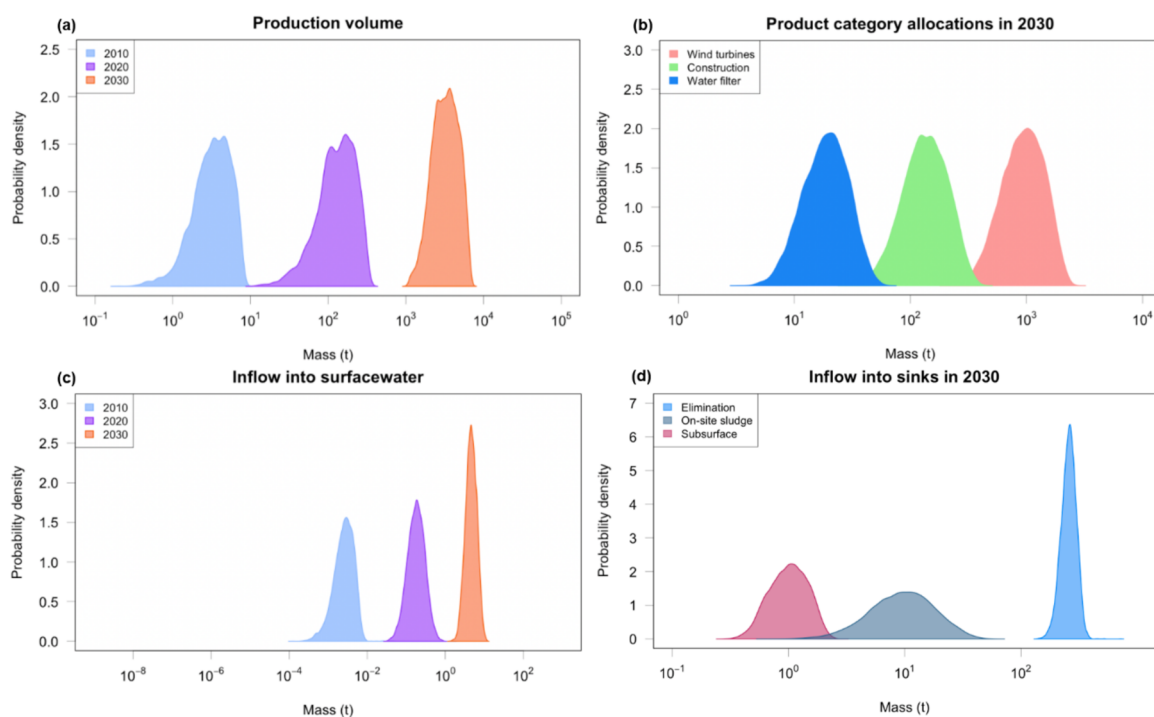


Figure 1. Example probability distributions of model inputs and results. (a) Probability distributions for GBM production volumes in 2010 (sky blue), 2020 (purple), and 2030 (orange). (b) Probability distributions of consumption of different product categories in 2030: water filters (blue), construction (green), and wind turbines (coral). (c) Probability distributions of inflows to surface waters in 2010 (sky blue), 2020 (purple), and 2030 (orange). (d) Probability distributions of inflow into each sink in 2030: the subsurface (pink), on-site sludge (grey), and elimination (blue).

categories identified by Future Markets' report did not specify the volume allocated to R&D.⁴⁶ We therefore estimated the share used in R&D based on Graphene Flagship's report,¹⁴ which described past and projected graphene market demand in different applications (original data from IDTechEx⁴⁹). Production volumes for the 13 remaining product categories were estimated based on the 24 product categories in Future Markets' report (Figure S5 and Table S2).⁴⁶ The product category for composites was subsequently divided into its two main applications of wind turbine blades and sporting goods. Graphene applications in the wind turbine blades market are expected to commence after 2022 (Table S3), and thus, no share is shown in Figure S5.

Future Markets only reported the shares of product categories from 2018. Table S3 summarizes the years when graphene made its market entry in the fields of electronics, batteries, wind turbine blades, sporting goods, water filters, paints and coatings, tires, and textiles, all identified from a literature research. We have assumed linear production volume growth for each product category from its year of market entry.

Transfer Coefficients. Transfer coefficients describing each product category's flows between compartments were identified from a literature search. Allocations for release after use were based on various references (Tables S4 and S5). The solid waste categories shown in Table 2 were updated based on the work of Rajkovic and colleagues.⁴⁰ The model required a variety of transfer coefficients, including release schedules and their post-use allocations and the proportions of those allocations. As most of the transfer coefficients were product-category-dependent, they were taken from previous DPMFA models.^{37–42} This is reasonable as most release processes are not specific to one nanomaterial but just depend on the release process and the matrix.⁵⁰ One unique

application of GBMs is as a drilling fluid additive. However, during their use phase in a borehole, these additives can be lost by diffusion through the rock, resulting in an undesirable direct release into the subsurface (Figure S6). This "circulation loss", including the additive loss, depends on various factors (e.g., soil permeability and borehole pressure). As Figure S7 shows, the present study's estimated rate of drilling fluid loss into the subsurface was based on a quantitative predictive model for circulation loss.⁵¹ Transfer coefficients related to tire applications and reprocessing were also updated based on Sieber et al.'s study (Tables 1 and 2).⁴¹

Predicted Release Concentration. The predicted release concentration constitutes a simplified predicted environmental concentration that takes into account the size of environmental compartments without considering environmental fate and transformation processes. In the current study, predicted release concentrations were estimated using MFA. Yearly predicted release concentrations into surface waters were derived from the mass of GBMs in surface water in each respective year. The surface water residence time for Europe (40 days) was used as a simplified method of estimating the cumulative quantities in those waters.⁵² Thus, the annual release into surface water was multiplied by a conversion factor of 40/365, and then this mass was divided by the volume of freshwater in Europe.⁵³ The masses of GBMs in natural and urban soils and sludge-treated soils were divided by the masses of these two soil categories in Europe to derive predicted environmental concentrations.⁴²

RESULTS

Probability Distributions. Due to its dynamic and probabilistic aspects, the model's flows, stocks, and sinks were calculated in the form of probability distributions.

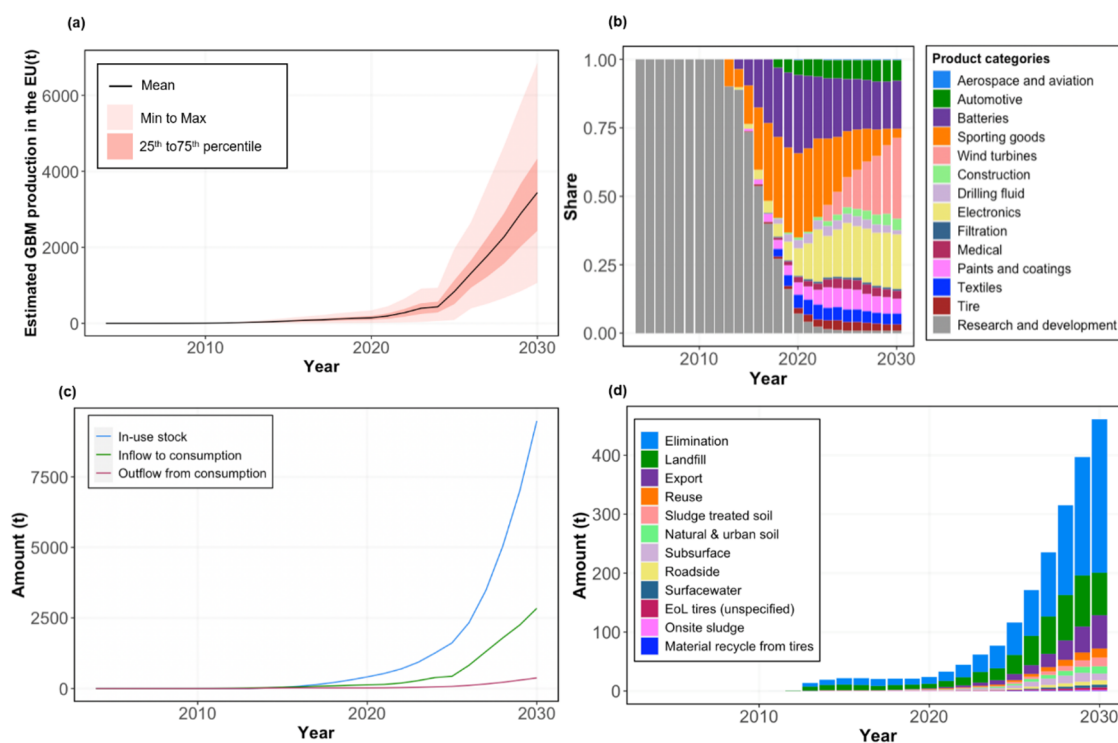


Figure 2. (a) Yearly GBM production. The red line shows the mean yearly GBM production in Europe. The lighter orange color represents the area between the minimum and maximum estimations for annual production, and the darker orange color indicates the range between the 25th and 75th percentiles. (b) Trend of allocations to product categories from 2004 to 2030. (c) Amounts of GBMs in in-use stocks, inflows into consumption, and outflows from consumption. (d) Dynamics of sinks and outflows leaving the system boundary (elimination and export) from 2004 to 2030.

Although the model enables the extraction of full probability curves based on 10,000 simulations, most of the figures and text refer to mean values for simplicity.

One of the model's inputs is the annual amount of GBM production, and this is given as a probability distribution (see Figure 1a for three specific years). Product category allocations are a type of transfer coefficient (another model input; see the Supporting Information) and also have a probabilistic nature. Figure 1b shows the 2030 probability distributions of the product allocations to three product categories as examples: water filters, wind turbine blades, and construction. The different product categories' probability distributions had similar standard deviations (curve widths), although the difference between the product categories' mean use volumes (biggest, wind turbines and smallest, aerospace) was rather large (86 times bigger). Water filters used very little of the total product allocation (1.9%). Figure 1c shows GBM inflows into surface waters in 2010, 2020, and 2030. The distribution curve for 2030 has the highest mean (an increase of inflows into surface waters over time) and a narrower shape (smaller standard deviation). Figure 1d shows the example of projected 2030 distributions of inflows into three sinks: elimination, on-site sludge, and the subsurface. Elimination, on the right side of the plot, is the sink with the greatest volume and smallest variance, whereas the on-site sludge sink shows a lower mean with a very big variance.

Flow Developments over Time. Figure 2a shows the development of the GBM production volume over time, illustrating the dynamic nature of the model that accounts for the dynamic inflows into the system boundary. Graphene production has increased continuously over this period, with a steep projected increase after 2024. For example, 2025's

projected production (828 t) is 14 times higher than 2015's (60 t), whereas the expected 2030 production (3461 t) is 24 times higher than 2020's (144 t). Product category allocations also show a dynamic perspective (Figure 2b). For the first nine years with records, GBMs were only used in R&D. One of the first GBM applications in commercial products was in sporting goods (e.g., tennis rackets and skis).^{57,58} Applications in batteries and electronics followed in subsequent years. Although the commercialization of GBM applications is still in its early stages, only 2.2% of GBMs were allocated to R&D in 2021, and this is expected to decrease to 0.76% in 2030.

Figure 2c shows the evolution of the amounts of in-use stock, inflows into consumption, and outflows from the use phase over time. During the GBM use phase (Figure 2c, in green), some product categories display delayed release; thus, only a small amount of a year's consumption will be released directly (outflow from consumption in red). The blue curve represents the accumulated amount of GBMs in the in-use stock.

Sinks are the final environmental compartments without outflows, indicating where GBMs exist at the end of the product life cycles. Figure 2d's stacked bar plot shows the evolution of the model's sinks over time. Elimination, landfill, and export represent the three largest amounts in the 10 sinks and 2 outflows leaving the system boundary. Materials recycled from tire applications represent the smallest sink across the period. Surface waters form the smallest of the environmental sinks.

Mass Flow Diagrams. Despite the current model's probabilistic nature, Figure 3 presents the GBM mass flows, stocks, and sink volumes for 2030 as mean values for simplicity. The vast majority (97%) of GBM consumption

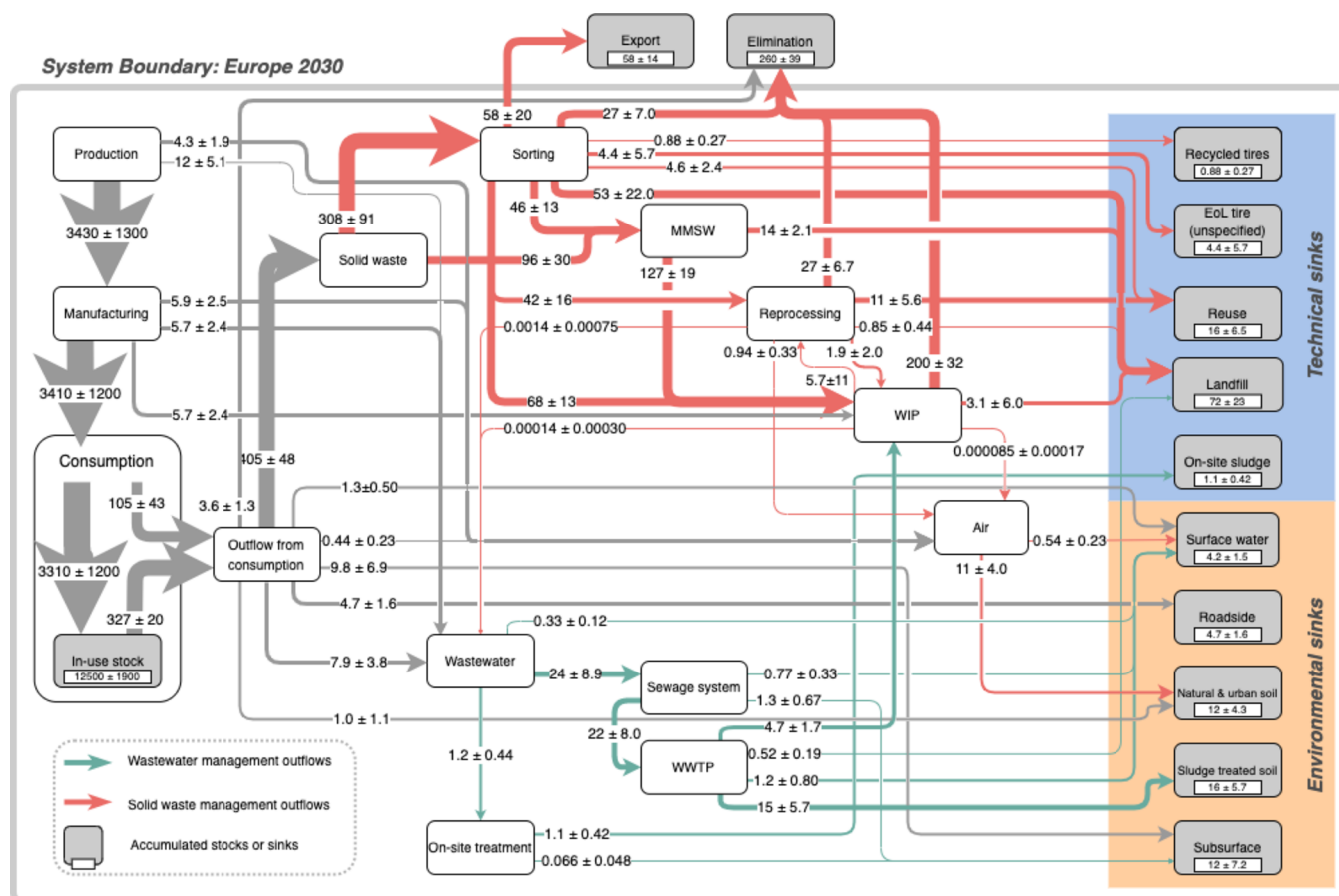


Figure 3. Projected European 2030 GBM flow diagram (in metric tonnes). The arrow thickness reflects the mean flow, and the mean and standard deviation are stated on each arrow. White boxes are compartments with outflows, whereas the gray boxes (with amounts) are sinks, accumulated stocks, or outflows leaving the system boundary (elimination and export). Gray box values show mean sink amount and standard deviation. Means bigger than 100 are rounded to two significant figures; means greater than or equal to 100 are rounded to three significant figures. Standard deviations are rounded to two significant figures. MMSW, mixed municipal solid waste; WIP, waste incineration plant; WWTP, wastewater treatment plant.

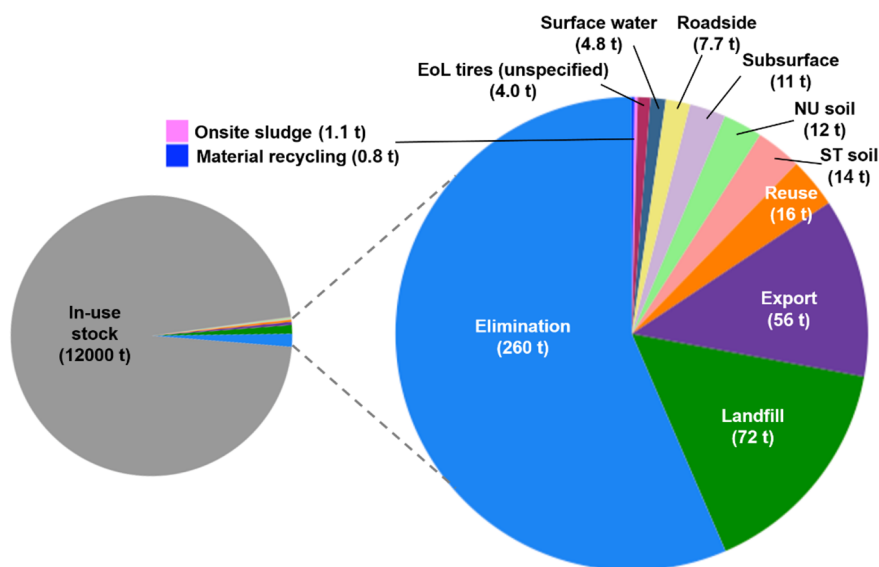


Figure 4. Overview of the final sinks, accumulated stocks, and outflows of GBMs in 2030. The left-hand pie chart shows accumulated stocks, sinks, and outflows leaving the system boundary. The right-hand pie chart compares the sizes of 10 sinks and the 2 outflows leaving the system boundary (i.e., elimination and export). Values are mean compartment sizes rounded to two significant figures. NU soil, natural and urban soil; ST soil, sludge-treated soil.

Table 3. Predicted GBM Release Concentrations 2010, 2021, and 2030 (Median, Mean, 5th, 25th, 75th, and 95th Percentiles)^a

	Units	P5	P25	Median	Mean	P75	P95
2010							
NU soil	pg/kg	4.0	8.0	12	13	18	25
ST soil	ng/kg	2.6	5.4	8.2	9.7	12	18
Surface water	pg/L	0.24	0.48	0.73	0.87	1.1	1.6
2021							
NU soil	ng/kg	0.29	0.54	0.79	0.86	1.1	1.6
ST soil	μg/kg	0.27	0.50	0.74	0.85	1.1	1.8
Surface water	ng/L	0.031	0.051	0.070	0.076	0.10	0.14
2030							
NU soil	ng/kg	8.2	12	16	16	20	27
ST soil	μg/kg	9.7	14	19	20	24	33
Surface water	ng/L	0.73	1.0	1.3	1.4	1.6	2.2

^aAll values rounded to two significant figures. NU soil: natural and urban soil; ST soil: sludge-treated soil.

remains in the in-use stock, mainly due to its application in wind turbine blades, batteries, and electronics. Most of the GBMs in wind turbine blades will flow into technical compartments during the EoL phase after their use (in an average of 20 years). Overall, 91% of consumption outflows goes to solid waste, and the rest is released, in descending amounts, to wastewater, the subsurface, roadside soils, elimination, surface water, natural urban soil, and the air. Thirty-four percent of solid waste goes to mixed municipal solid waste (MMSW), which mainly flows to waste incineration plants (WIP). Fifty-six percent (260 t) of all GBM going to sinks and flowing out of the system boundary is eliminated from the system, representing the incinerated (and thus oxidized) mass after waste incineration (Figure 4). Therefore, landfill, at 72 t, is the largest final sink in the system, with 74% of its inflow originating from sorting. Sludge-treated soil, at 16 t, is the environmental sink receiving the biggest amount of GBM due to transfers from wastewater treatment plant (WWTP) sludge. Surface waters form the environmental sink receiving the smallest amount (4.2 t).

In summary, the model's final sinks are landfill, natural and urban soil, sludge-treated soil, the subsurface, surface waters, roadsides, unspecified mass from EoL tires, on-site sludge, material recycled from tires, and mineral reuse. It is important to stress that the largest amount of GBM remains in the in-use stock, reflecting its delayed outflows from the consumption stage.

Predicted Release Concentrations. Predicted release concentrations were determined based on amounts released to surface waters, natural and urban soil, and sludge-treated soil (Table 3). The predicted 2030 release concentrations in Europe are 1.4 ng/L in surface waters, 16 ng/kg in natural and urban soil, and 20 μg/kg in sludge-treated soil. GBM concentrations in these sinks will thus have increased more than 1000-fold between 2010 and 2030.

DISCUSSION

Using a MFA model based on the complete life cycle of applications containing GBMs, we predicted those materials' complete flows through Europe's anthroposphere and into the environment. Production volume was one of the model's important inputs; however, only a limited number of sources have provided GBM production volumes in recent years. The information scarcity problem was an added challenge to estimating transfer coefficients because few empirical gra-

phene-specific data were available. Nonetheless, the present study integrated the transfer coefficients of other nanomaterials with similar physicochemical properties, like carbon nanotubes (CNTs). As GBMs and CNTs are both carbon-based nanomaterials, CNT-specific transfer coefficients were highly desirable data when no GBM-specific transfer coefficients were available. On the other hand, the low solubility of TiO₂ and SiO₂ also made their transfer coefficients reasonable candidates for the model. In tires, GBMs are used as an additive and remains in the rubber; thus, flows from the use and EoL of tires are characterized by application-specific transfer coefficients.⁴¹

The use of GBMs governs these flows during the complete life cycle of the products incorporating them. Identifying product categories was a crucial step toward representing GBM use, market shares, and distributions across various products. Although a couple of sources identified the product categories in which GBMs were used, their categorization methodologies were inconsistent. There was no way to integrate all the different data sources and their estimates of the proportions in each product category. Thus, the 13 product categories used in our model were identified based on the 24 product categories in Future Markets' report.⁴⁶ There were two main objectives in simplifying the product category. First, it was unnecessary to keep several product categories that go through similar use and EoL phases and releases. For example, the present study did not distinguish between supercapacitors and sensors but instead categorized both into the "electronics" product category. This was a reasonable approach considering the original product categories' use phase and EoL treatment. In identifying 13 product categories, this approach not only simplified the category but also fractionated them. Adhesives, for example, were reassigned to electronics, aerospace, automotive, and medical applications. Fractionating certain product categories enabled the model to consider the use and EoL phases more precisely for release estimations.

The present study considered "material recycled from tires" and "minerals reuse" as sinks. However, this could be refined with deeper research on the fate of GBMs during these recycling and re-use processes. Potential outflows from these sinks could then be identified, and possible environmental releases could be quantified and integrated into the model.

Although the in-use stock is not a sink, it was the biggest compartment for GBM every year. This is mainly due to wind turbine applications, which have a 20-year use phase. The EoL of wind turbine blades is a complex issue, and various potential

ways of recycling them have been discussed.^{59,60} The present study's model was built based on the fact that the most common current EoL process for disposing of wind turbine blades is currently the landfill.^{61–63} Note that many European countries forbid landfilling waste depending on its total organic carbon content,⁶⁴ and the graphene applications in wind turbine blade composites have the potential to contain very significant amounts.⁴⁶ When these blades are decommissioned after a 20 year life span, inflows of GBMs into recycling, waste incineration plants, or landfills will increase tremendously. However, given the huge efforts to develop recycling methods for wind turbine blades, it is likely that they will be able to enter recycling processes in a few years, significantly reducing mass flows into landfills during the EoL stage.⁶³ Several companies are currently developing methods to recycle wind turbine blades by mechanical, thermal, or chemical recycling technologies.⁶⁵ Although this form of recycling process is expected to bring bigger environmental benefit, the shredding during mechanical recycling may increase the release of materials from the product. Since the framework conditions for waste management will therefore change with regards to the circular economy, our model will require constant updating every time disposal practices change. Indeed, model updates will also be necessary as soon as new research provides more accurate GBM-specific transfer coefficients. For example, using a “best case approach,” we assumed that no GBMs would be abraded during the use of anti-erosion coatings because there is currently no data in the literature on average release rates (in t/year) from installed wind power stations. Release modeling should be updated as soon as more precise data based on field tests is publicly available.

The present study was the first to model and estimate the predicted release concentrations of GBMs into the environment. The analytical techniques used to quantify amounts of engineered nanomaterials and distinguish them from natural nanomaterials are limited.⁶⁶ Moreover, limited analytical methods for detecting and distinguishing GBMs from other carbonaceous materials in the environment form another challenge.⁶⁷ Thus, any validation of our mass flow and release model is currently impossible. An analytical method for measuring GBMs in the environment would need to reach a detection level in ng/L or ng/kg in soils, which is currently impossible.⁶⁷ Our model's results should therefore be treated with caution but could be used for prospective estimates of realistic concentrations in the near future.

After their release into the environment, GBMs are subject to a range of transformations. For example, GBMs have a strong adsorption capacity for solid particles,^{68,69} and in freshwater, they can react with other materials (e.g., inorganic ions, natural organic matter, and environmental pollutants).⁷⁰ Degradation, oxidation, reduction, and biotransformation are other possible chemical transformation pathways.^{71–74} However, the present study did not consider possible transformation and the fate of GBMs in the environment and only provided “release concentrations”, not predicted environmental concentrations. Therefore, the present study's results are representative of the area close to GBM emission sources and can be considered as input data for further fate modeling studies. Note that several fate models for engineered nanomaterials have been developed, such as SimpleBox4Nano⁷⁵ or NanoFate,⁷⁶ which need environmental releases as input values and could be adapted to GBMs in the near future.

Comparisons between GBM and CNT concentrations in the environment can be made. Wigger and Nowack estimated 2012's predicted environmental concentrations of multi-walled carbon nanotubes (MWCNTs) and single-wall carbon nanotubes (SWCNTs) in European freshwater to be 0.3 and 0.1 ng/L, respectively.⁷⁷ These values were reached with a total production of about 380 t/year of MWCNTs and 14 t/year of SWCNTs. The ratio of MWCNT production volume to concentrations in European freshwater (production volume (t)/concentration (ng/L) is in the same order of magnitude as GBMs in 2030 (GBM: 1714, MWCNT: 1266). Therefore, it could be concluded that the difference in European freshwater concentrations of MWCNTs and GBMs reflects the difference in those materials' annual production volumes. The ratio for SWCNTs was one order of magnitude smaller (140), reflecting more release from smaller production volumes than GBM. Both CNTs and GBMs have applications involving mostly limited releases during the use phase, with their major release flows occurring during EoL processes.

Release concentrations could provide crucial information for a first environmental risk assessment of GBMs and could be used as a replacement for predicted environmental concentrations in the absence of better data. Such a risk assessment would require a hazard assessment to be performed first to obtain a predicted no-effect concentration (PNEC) value for GBMs as no such values are currently available. A prospective assessment of the environmental risks of nanocellulose has been conducted using such an approach;⁴⁴ this is a material for which a huge production increase is expected in the coming years.

Nanomaterials like GBMs are produced and used in many different forms, and changes to those forms can occur during use and release. Many studies have shown that a nanomaterial's form can affect its environmental release, fate, behavior, and toxicity.^{78–81} To consider such processes, Adam and colleagues modeled the material flows of engineered nanomaterials in different pristine, matrix-embedded, transformed dissolved, and product embedded forms.⁸² Results from the specific MFA of crystal-form nanomaterials⁷⁷ and a size-specific DMFA, which considered the crystal and coating forms of TiO₂,⁸³ confirmed that the form of the nanomaterial affected the environmental exposure. Moreover, a form-specific and probabilistic environmental risk assessment of nano-Ag, nano-TiO₂, and nano-ZnO by Hong and colleagues showed that the form-specific approach could alter calculations of the environmental risks posed by engineered nanomaterials.⁵³ Several studies have reported different toxicological effects depending on the form of graphene,^{30–32} thus, a form-specific approach should be integrated into the next framework for the exposure and environmental risk assessment of GBMs. However, this approach will need information on the use of specific forms of GBMs (e.g., graphene, multilayer graphene, graphene oxide, reduced graphene oxide, graphene nanosheets, graphene nanoribbons, and graphite nanoplatelets, sheets, or flakes) in specific applications as the allocation of different forms to different product categories can only be done when different materials' functionalities can be assigned to different products.⁷⁷

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.2c04002>.

Input data for production volume estimations and details of European production volume estimations (XLSX)

Details about the modeling method: system boundary and material flow analysis diagram, production volume estimation, product categories, year of market entry, transfer coefficients, simplified release diagram of drilling fluid, transfer coefficient of vehicle tire applications, and transfer coefficient related to reprocessing (PDF)

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Notes

The authors declare no competing financial interest.

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