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Environmental attributes of fungal-like adhesive materials and future directions for bioinspired manufacturing

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ABSTRACT

This study evaluates the environmental impact of fungal-like adhesive materials (FLAMs), which are at the forefront of bioinspired manufacturing for sustainability and are thus well-poised to enable a circular economy because they can be sourced, manufactured, and degraded in almost any ecosystem, including urban environments. The main constituents of FLAMs (i.e., cellulose and chitin) are ubiquitous and available from diverse material sources, and the environmental impact of current production practices would vary depending on material sourcing. Here, cradle-to-gate life-cycle assessments were performed on four scenarios with different material sources to. provide insights into the environmental impact of the technology in its current state and identify critical areas to prioritize in future developments. Key contributors were material transportation and chemical use (i.e., sodium hydroxide), which significantly improved when FLAM production was based on regional suppliers and reduced the import distances of raw materials. Next, it was determined that cellulose selection must be based on regionally available pulping technology because using virgin or recycled cellulose fibers has a negligible impact on FLAM production. Finally, it was found that the use of locally produced chitosan from black soldier flies is a positive step toward regionalization and shorter distances. However, as a process performed at a lab scale, it requires the adoption of novel and more efficient biomass fractionation, such as those being implemented for other farmed arthropods (e.g., shrimp). Relative to similar materials, such as rigid polyurethane foam, lower impacts were observed in more than half the environmental indicators, positioning FLAMs in their current state as a more sustainable alternative, even without considering their unassisted compostability. The direction of future developments recommended here resonates with the research efforts on bioinspired manufacturing based on the convergence of ubiquitous resources, regional production, and optimized methods of extraction and fractionation of biomass resources, which we believe should be prioritized. The potential impact of the ongoing and proposed research efforts highlights the unique opportunity enabled by FLAMs and bioinspired manufacturing to minimize transportation and maximize the ecological integration of materials, which in turn present a unique prospect for the development of sustainable manufacturing.

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1. Introduction

Since the early nineteenth century, globalization has intertwined wealth, nations, and the state of our planet. The interdependence between the environment and prosperity entails a need for sustainable development in every country. Nations, which are united by the common fate of our planet and humanity, have established sustainable development goals to enable the continuation of our human story (Sachs et al., 2019). One such goal—responsible consumption and production—includes waste minimization and the efficient use of natural resources. This need for a change in production practices dovetails with trends toward circular economies (Geng et al., 2019), where doing more with the same initial resources leads to less waste and consequently demands fewer new resources. To this end, new materials and products are increasingly manufactured with renewable resources, process byproducts, and waste. However, without a paradigmatic shift from overdependence on synthetic resources that are not integrated within a natural system, the expression of an underlying

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human–nature material disconnection is merely suppressed (Ives et al., 2018). Given that no man-made material is capable of endless recycling without property degradation, waste will eventually be waste, albeit with prolonged utility. Hence, large-scale manufacturing with inherently biodegradable and renewable natural materials must be a priority alongside engineered synthetic materials to reduce the accumulation of waste and stress on thinning finite resources (Fernandez and Dritsas, 2020).

Manufacturing with unmodified molecules, such as cellulose, is prevalent throughout the world (e.g., paper). However, to improve the mechanical properties and ensure compatibility with large-scale manufacturing (e.g., injection molding), naturally occurring molecules must be transformed into materials that nature has difficulty decomposing (e.g., Polylactic Acid and Cornware). Nature's solution to improving mechanical properties is biocomposites, which have microscale hierarchical structures (e.g., wood and nacre) that are difficult to replicate in the laboratory, let alone on an industrial scale.

In the last decade, research on bioinspired manufacturing with natural materials has advanced and resulted in successful replication of some of these synergies of biological materials. Chitinous materials (which are composed of ubiquitous chitin molecules) have shown promise in the production of functional objects with common manufacturing methods (Fig. 1) (Fernandez and Ingber, 2014). To further augment their mechanical performance, secondary materials, such as insoluble proteins (e.g., silk fibroin) (Fernandez and Ingber, 2012) and cellulosic materials (e.g., cellulose fibers and wood flour) (Sanandiya et al., 2018), can be combined with chitinous materials to form bioinspired composites, which are promising material solutions for environmental sustainability and medical applications (Fernandez and Ingber, 2013). Specifically, fungal-like adhesive materials (FLAMs) replicate the material composition of fungal walls that contain chitin and cellulose. The resulting chitinous biocomposites have mechanical properties comparable to high-density polyurethane foam (PUR) and are practical in the contexts of traditional as well as additive manufacturing. Even without the use of organic solvents or modification of the original biomolecules, FLAMs are fully ecologically integrated, and they decompose without human intervention (e.g., recovery).

In addition to being inherently biodegradable, FLAM is also renewable and widely available. Chitin is the second most abundant polysaccharide on Earth and is nature's perennial solution for forming structural components across biological kingdoms (e.g., arthropods (Fabritius et al., 2010), fungi (Lenardon et al., 2010), and some vertebrates (Tang et al., 2015)). With natural reproduction, the steady state value for chitin is estimated at 10^{11} tons annually, which entails a high intrinsic recycling rate and low accumulation (Gooday, 1990). Commercially available chitin is processed from the exoskeletons of crustaceans, which are byproducts of the shellfish industry (Younes and Rinaudo, 2015). With up to 75% of crustacean mass becoming byproduct, an estimated 6–8 million tons of exoskeleton byproduct are generated globally per year (Hamed et al., 2016). These shells are post-processed to produce deacetylated chitin, or chitosan, for commercial use across industries, such as food packaging and biomedical industries (No and Meyers, 1995). Cities without established shellfish industries could produce chitin with simple organic waste management systems that valorize organic food waste with black soldier flies (BSFs) (Singh and Kumari, 2019). Besides converting waste into biomass, BSF reproduction produces larvae that are sold as feedstock and are an untapped, generalizable source of chitin.

Cellulose is the most abundant organic compound in the world. Found in cotton, wood, and plants, cellulose is typically used in the production of paper, textiles, and consumables. Cellulose fibers are produced from harvested softwood or hardwood and processed via different pulping methods. Global paper production alone is expected to grow to 482 million metric tons per year by 2030. Excluding consumable paper products, such as tissues or towels, used paper is generally collected and treated into recycled cellulose fiber for recombination with virgin pulp to produce new paper products. While the current recycling rate stands at 57%, incorporating more recycled fiber into cellulosic products is critical to minimizing the production footprint (Kinsella et al., 2018).

Hence, with ubiquitous raw materials that could be obtained from diverse waste or natural sources, FLAMs could potentially be produced and degraded locally in any urban environment to increase the use of waste without adversely impacting the environment (Sanandiya et al., 2020). While FLAMs are at the forefront of bioinspired manufacturing, and there are high expectations for their environmental benefits, the impacts of current production practices remain unverified. FLAMs are designed to be adapted to the surrounding ecosystem, but the versatility of the material sources for FLAM products could yield different environmental impacts; therefore, exploration of the main alternatives is warranted.

Unlike life-cycle assessment (LCA) studies of materials with similar composition, such as chitosan-based films and wood fiber reinforced or nano-cellulose reinforced composites, the material of study here is unique in its versatile application, its use of ubiquitous

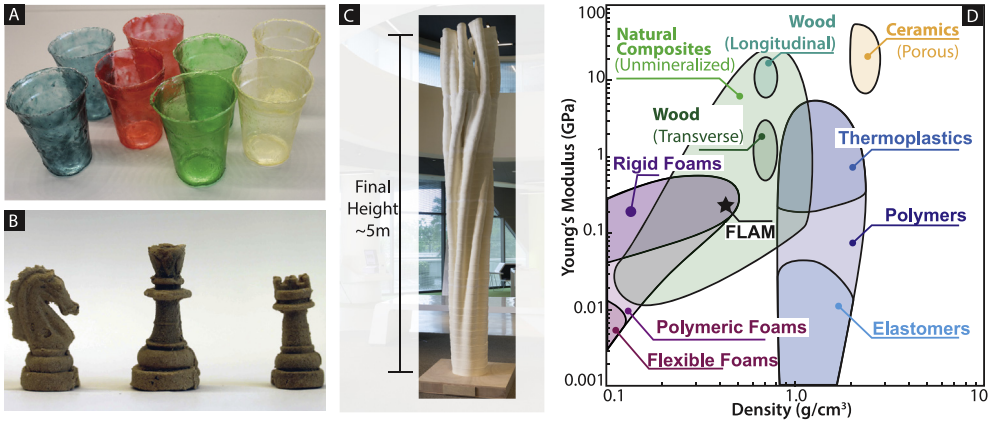


Fig. 1. Chitinous products and FLAMs. **(a)** Chitinous products are a potential alternative to petroleum-based plastics and can produce functional objects, such as cups. **(b)** Chitinous biocomposites with chitosan and wood flour are suitable for injection molding to produce artefacts of complex geometries. **(c)** Through additive manufacturing, FLAM could be used to produce organic forms, which can be assembled to form large structures. **(d)** Regarding its mechanical performance, FLAM is positioned between natural composites, such as wood and polymeric foams (e.g., rigid foams). In this study, rigid PUR foam was used as a comparison for considering similarities.

raw material within most local/regional circular systems, and its inherent biodegradability at the end of life. Hence, in this first LCA of the novel material and surrounding processes that are common to bioinspired chitinous manufacturing, we conducted cradle-to-gate LCAs to evaluate the environmental impacts associated with current and alternative FLAM production practices. Four different scenarios were considered: (A) the current practice of importing from international suppliers; (B) importing from regional suppliers; (C) using recycled cellulose fibers; and (D) using locally produced BSF-based chitosan.

The objectives of this study are the exploration of the current state of the production of FLAMs, its comparison with the production of a material of similar mechanical properties and uses (i.e., PUR foams), and the determination of those processes with the highest relative environmental impact in the production of FLAMs. Scenarios (A) and (D) evaluate the effects of unoptimized transportation and biomass conversion, respectively. Scenario (B) represents the most likely current scenario, whereas (C) considers the impact of using recovered biomass instead of primary sources. As a rather young and rapidly developing technology, the results of this study will help researchers identify which aspects of FLAM production should be prioritized in the technology's future development and during its regional implementation. The conclusion of this study provides specific examples of emerging technologies and processes that are either readily available or in development and can significantly improve the most environmentally impactful aspects of bioinspired manufacturing in general and FLAM production in particular.

2. Methodology

2.1. LCA methods

LCA is a standardized, systematic approach consisting of four phases: goal and scope definition, inventory analysis, impact assessment, and interpretation (ISO14040, 2006; ISO14044, 2006; ISO14045, 2012). With the established goals of assessing the environmental impact of FLAMs considering their different material sources and identifying significant contributors, this work's scope is limited to a partial life cycle from cradle to gate, as the anticipated application can be wide-ranging. For comparison against currently available similar materials, the functional unit was set as 1 kg of end material. In life cycle inventory modeling, the system boundary was based on an attributional model where the supply chain was analyzed. This decision was made considering that the purpose of LCA primarily accounts for existing production methods and supports decisions on the micro level, i.e., without large-scale consequences (European Commission - Joint Research Centre - Institute for Environment and Sustainability, March 2010). In addition, environmental credits or burdens from production processes were not allocated to the primary constituents, namely, waste or recyclable materials with low or no economic value. Hence, raw materials were considered "cut off" and available burden-free. The ReCiPe 2016 method, which is an improvement to predecessors, such as CML 2000 and Eco-indicator 99, was used for impact assessment to translate emissions from associated processes to midpoint or endpoint indicators using characterization factors (Huijbregts et al., 2017). Here, the calculation was approached with a hierarchist perspective and aggregated as midpoint indicators, such as climate change and acidification, to reduce the uncertainty associated with further aggregation in endpoint indicators. OpenLCA was used for matrix computation, such as formulation of process vectors, solving for scale, calculation of inventory vectors, and—ultimately— their multiplication with characterization vectors (Heijungs and Suh, 2002).

2.2. Case scenarios and system boundaries

To verify the environmental attributes of current production practices and simulate alternative scenarios of material sourcing, this study examines four case scenarios; two were based on the transportation of primary materials, and the two others were based on sourcing methods. The first scenario featured the current production practice of importing constituents from international suppliers; the second involved production with constituents from regional suppliers; the third was production with recycled cellulose fibers instead of virgin cellulose fibers; the last featured production with BSF-based chitosan. The required raw materials were the same across the four scenarios, and the varying factors were mainly material transport distance and the source of chitosan or cellulose fibers. The system boundaries are illustrated in Fig. 2.

2.3. Inventory data

2.3.1. FLAM production

This research used primary data on FLAM production in Singapore, which consist of two main imported constituents: chitosan from India and high-purity cellulose from Germany. The main process primarily involved dissolving as-received chitosan into 1% (v/v) acetic acid to prepare a chitosan solution (3% w/w), which was subsequently mixed with as-received cellulose fibers in an industrial dough mixer. While the chitosan-to-cellulose fiber ratio can differ, 1:8 was found to be the optimal ratio considering mechanical properties and compatibility with general manufacturing of the resulting composite. The amount of chitosan and cellulose fibers used per kilogram of FLAM was modeled with a triangular distribution of ±5% target amount to account for variability in production. The wet FLAM could then be dried under ambient conditions or at slightly elevated temperatures (50–60 °C) to accelerate the drying process and produce dried FLAM at approximately 27% its original weight. The transportation of chitosan and cellulose via air freight in the current production practice was assumed to originate from the airports nearest the suppliers, namely, Mumbai Airport (India) and Stuttgart Airport (Germany). With the destination point as Changi Airport (Singapore), Vincenty's formulae were used to approximate the distance between the two points.

Naturally, the primary and secondary data used in this study had an intrinsic variability and uncertainties arising from deviations in geographical, technological, and temporal differences as well as varying degrees of confidence in data completeness and source reliability (Ciroth et al., 2013). These uncertainties were described by qualitative indicators in a pedigree matrix and translated into empirical-based uncertainty factors (Weidema and Wesnæs, 1996). In addition to a basic uncertainty factor that accounts for variability even in a scenario of perfect data collection, a geometric standard deviation was aggregated, and the data were assumed to have lognormal distributions (Frischknecht et al., 2007).

Here, the transport distances were modeled to have a basic uncertainty factor of 2.0, as per Ecoinvent's recommendation (Frischknecht et al., 2007). For subsequent cases of FLAM production, the transport distances were assumed to be 3000 km for regional suppliers and 50 km for the specific case of local, laboratory chitosan production. In these cases, the distance represented the maximum distance between the production site and potential suppliers of raw material. Fig. 3 illustrates the current suppliers available within the assumed geographical radius.

The unit processes within Ecoinvent 3.5 database were used in modeling subprocesses, as the database is commonly recognized as the most consistent and compliant data source on the market. Primary data used in the inventory of the four different cases are summarized in Table 1.

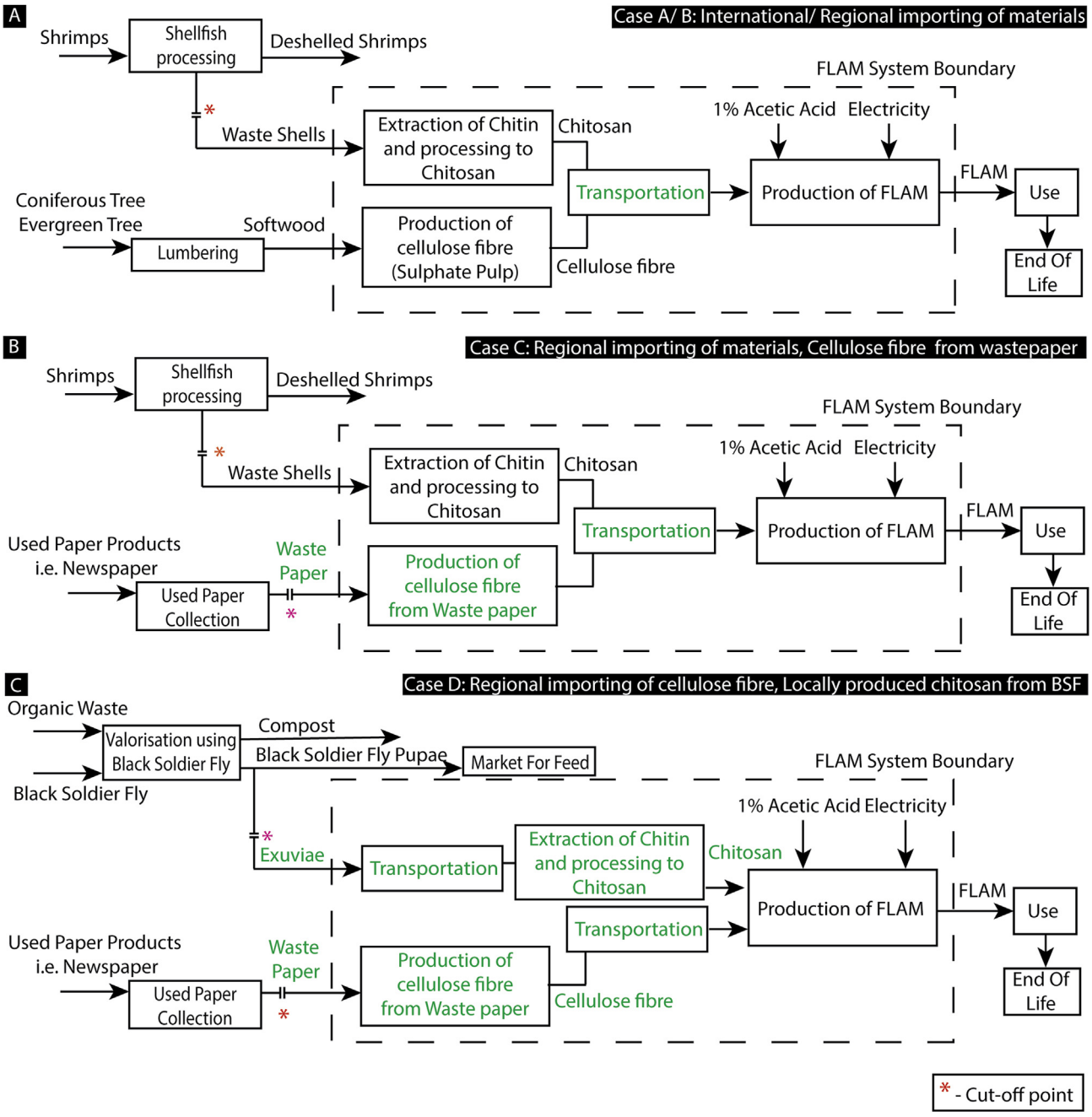


Fig. 2. System boundaries of four case scenarios. Green labels denote differentiating elements from Case A (current practice) **(a)** Case A represents current production practices, while Case B simulates the hypothetical scenario of regional suppliers. The systems have the same scope except the modification of material transport distance. **(b)** Case C is similar to Cases A and B except the simulated hypothetical use of recycled cellulose fiber. **(c)** Case D is similar to the previous cases except the hypothetical local laboratory production and use of chitosan. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

As shown in Table 1, the high-purity cellulose fiber production process in Germany was modeled by sulfate pulp (virgin cellulose fiber) production (Wernet et al., 2016), as both processes use harvested softwood and produce fibers that are low in contaminants (e.g., lignin). For the FLAM production in Case C, where recycled cellulose fibers produced from wastepaper were used, a modified unit process based on cellulose fibers used for blown-in insulation was utilized. The Ecoinvent unit process was modified to remove the use of boric acid and borate emission, given that recycled cellulose fibers would not require fire resistance (Peltola, 2004). Functionally, cellulose fibers recycled for insulation purposes can be used for FLAM production as well.

2.3.2. Chitosan and chitin production (India)

Secondary data from the life cycle inventory of Muñoz et al. (2018) were used to model chitosan and chitin production in India. However, only unit processes relevant to the supply chain of chitin and chitosan production were replicated, as the initial study was a consequential assessment, i.e., credited or discounted environmental impacts resulting from by-products. Subprocesses such as the diverted use of shrimp shells into fishmeal and avoided production of fertilizers due to protein sludge by-products were not considered in this study. The aforementioned work uses background data providers modeled with a consequential system from Ecoinvent database. Here that data was replicated using data

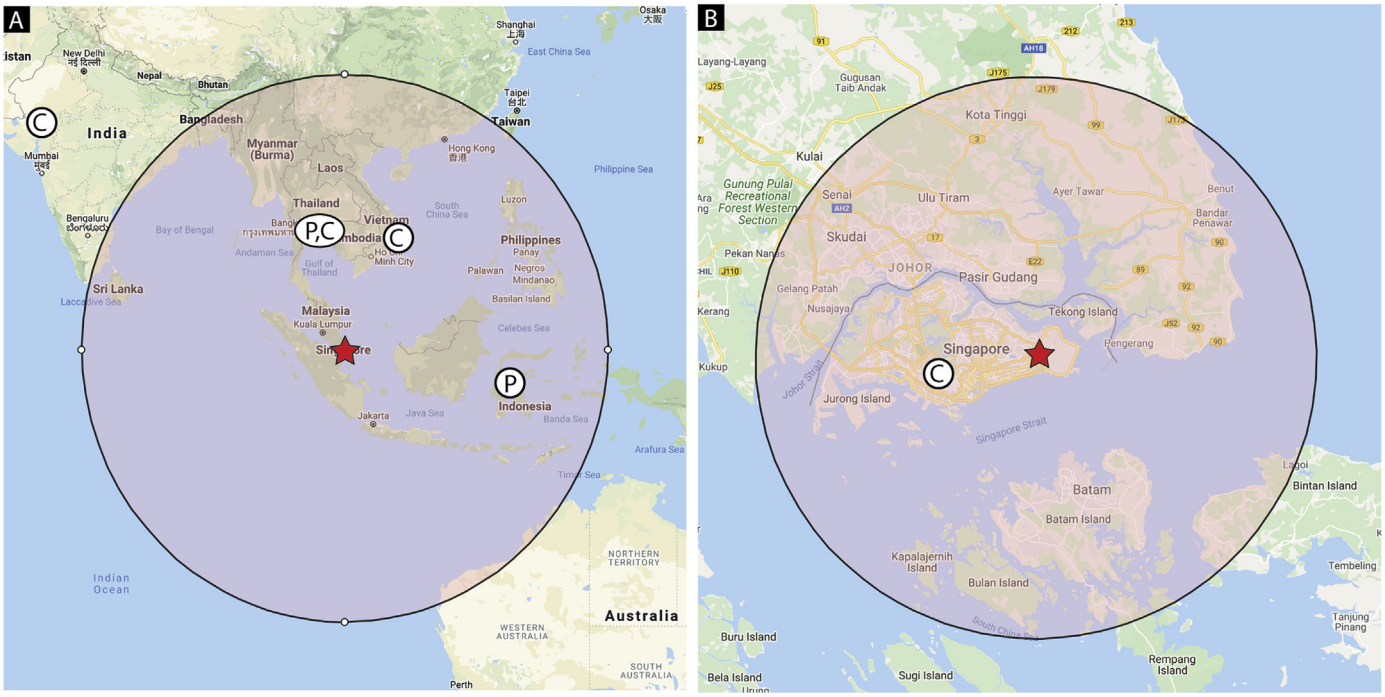


Fig. 3. Visualization of assumed geographical radius with respect to FLAM production site. **(a)** With a radius of 3000 km, chitosan producers ("C" markers) can be found in Vietnam and Thailand, while chemical pulps can be sourced from Indonesia and Thailand ("P" markers). **(b)** For the local production of chitosan, exuviae were taken from a black soldier fly (BSF) farm within a geographical radius of 50 km. Interestingly, the insect farm is in the heart of Singapore and demonstrates the viability of an urban (BSF) farm in developing or developed cities.

providers from the same database but modeled with a cut-off system, consistent with the goal of this attributional study.

Except the input process used in the market producing electricity, the replicated process was used to simulate India and regional suppliers. While the India suppliers in Case A used an electricity mix emulating the western India grid, the regional suppliers assumed an electricity grid that was similar to that of Vietnam. This was considering that there are existing chitosan suppliers in Vietnam and assuming that they performed similar chemical extraction processes of chitin/chitosan. A low score in data quality was used for regional suppliers because of the discrepancy that may be caused by data completeness and geographical correlations. The inventory data used in chitosan and chitin production are summarized in Tables 2 and 3, respectively.

2.3.3. Chitosan and chitin production (laboratory)

Primary data recorded from actual extraction processes were used to model laboratory-produced chitin and chitosan (Sanandiya et al., 2020). Low-voltage electricity from the Singapore power grid was assumed, given that equipment such as industrial dough mixers and magnetic stirrers are directly plugged into electricity outlets. Deionized water used in chitin production was differentiated as three entries due to its different uses, namely, dilution of sodium hydroxide, dilution of hydrochloric acid, and general washing. Material transport was assumed to be 50 km, which sufficiently covers the distance across Singapore.

3. Results and discussion

3.1. Case A: international supplier of constituents

While the production of FLAMs is anticipated to provide environmental benefits, unoptimized material sourcing generates harmful emissions that burden and negatively contribute to all midpoint indicators. As highlighted in Fig. 4, air transportation of materials to production sites over large distances nears or outweighs the contribution of raw material extraction across all indicators. Understandably, air travel across continents requires large quantities of aviation fuel, which is refined from crude oil and directly contributes to fossil depletion. The combustion of aviation fuels releases to the atmosphere gases and particulates, such as carbon dioxide, sulfur dioxide, nitrogen oxide, and volatile organic compounds that lead to climate change (CO_2 absorbs heat), terrestrial acidification (through wet or dry deposition), formation of photochemical oxidants, and other environmental indicators.

After air transportation, the most significant contributor is the production of sodium hydroxide, a chemical required during the deproteinization of shrimp waste during chitin production. As saturated brine is fed into different types of chlor-alkali processes (O'Brien et al., 2005), impurities such as vanadium, iron, magnesium, cobalt, and phosphate have to be removed to improve the efficiency of electrolysis or prevent severe process upsets (Hocking, 2005). When released into marine or freshwater environments, these metallic ions are toxic to aquatic biota and humans. In addition, excess phosphates can accelerate plant growth and lead to eutrophication. While cellulose fibers comprise a large portion in FLAMs, optimized production of sulfate pulp over more than 100 years has lowered the overall enviromental impact compared with that from the production of chitosan. This is evident, given that the environmental impact of sodium hydroxide does not increase the overall impacts of sulfate pulp production despite that chemical pulping utilizes sodium hydroxide as well. Hence, rather than the underlying production of chemicals, the quantity of chemicals used is more likely to require optimization. Given the environmental footprint from sodium hydroxide and hydrochloric acid production, the industrial process of producing commercial chitosan notably requires technological improvements if chitosan-based products are to be a key alternative to petroleum-based plastics.

Table 1
Summary of FLAM production process across all cases.

FLAM production						
Input						
Flow	Amount	Unit	Basic uncertainty	Data quality entry	Provider (common across all cases unless specified)	Remarks
Acetic acid, 98% solution state	0.97	g	1.05	(2; 4;1; 1;5)	Market for acetic acid, without water, in 98% solution state | Acetic acid, without water, in 98% solution state | Cutoff, S - GLO	
Chitosan	3	g	–	(2; 4;1; 1;5)	Cases A, B, C: Chitosan production (Mahtani Chitosan), Table 3 Case D: Chitosan production (laboratory), Table 4	Data assumed to have a triangular distribution with: min = 2.85000, mode = 3.00000, max = 3.15000
Electricity, low voltage	6.75E-06	MWh	1.05	(4; 4;1; 1;5)	Market for electricity, low voltage | Electricity, low voltage | Cutoff, S - SG	
Cellulose fibers	24	g	–	(2; 4;1; 1;5)	Cases A, B, D: Sulfate pulp production, from softwood, bleached | Sulfate pulp, bleached | Cutoff, S - RoW Case C: Cellulose fiber production, not inclusive of blowing in | Cellulose fiber, not inclusive of blowing in | Cutoff, S - Modified - RoW	Data assumed to have a triangular distribution with: min = 2.85000, mode = 3.00000, max = 3.15000 Case C: Inputs for borax, in-ground modified to 0 kg Output for all boron, borate, boric acid emission modified to 0
Material transport (chitosan, air freight)	3E-6*distance (Refer to remarks for assumed distance)	t*km	2	(4; 4;1; 1;5)	Cases A, B, C: Market for transport, freight, aircraft | Transport, freight, aircraft | Cutoff, S - GLO Case D: Market for transport, freight, lorry, unspecified | Transport, freight, lorry, unspecified | Cutoff, S - RoW	Case A: 3910 km Case B: 3000 km Case C: 3000 km Case D: 50 km
Material transport (cellulose, air freight)	2.4E-7*distance (Refer to remarks for assumed distance)	t*km	2	(4; 4;1; 1;5)	Market for transport, freight, aircraft | Transport, freight, aircraft | Cutoff, S - GLO	Case A: 10200 km Case B: 3000 km Case C: 3000 km Case D: 3000 km
Deionized water	96.03	g	1.05	(2; 4;1; 1;5)	Market for water, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW	
Output						
FLAMs	27	g	1.05	(2; 4;1; 1;5)		

Inefficiencies such as the total amount of water used can be improved, as the combined water consumption of tap water and deionized water is more than the total amount of water used in chemical pulping. Interestingly, inert material landfill from chitosan production has a net credit to natural land transformation. This is a cumulative result considering the transformation between non-natural and natural land use.

3.2. Case B: regional importation of constituents

In the second case study, the regional importation of chitosan and cellulose was assumed to be available within a geographical radius of 3000 km, and the production process of the producers was similar to that in Case A. The contributions from air freight transportation were reduced visibly and the environmental impacts were lower than those of rigid PUR foam production on most midpoint indicators (Fig. 5). Categories such as terrestrial acidification, climate change, and fossil depletion were logical beneficiaries of the reduced emission from the production and combustion of aviation fuel. However, even with this reduction in transportation requirements, chitosan production remained the key contributing process that required optimization.

3.3. Case C: use of recycled cellulose fibers

Interestingly, while the use of cellulose fibers from wastepaper reduced the impact on agricultural land occupation, other indicators showed similar impact levels to those of virgin cellulose fibers. Although general findings on the environmental advantage of recycling paper remain true, the present results agree with those of a previous study that stated that sulfate pulp production in Europe using renewable resources produces virgin pulp that has little overall environmental difference from recycled pulp (IVL Swedish Environmental Research Institute, 2019). Therefore, both options of cellulose fibers are valid environmental choices that FLAM producers can select in accordance with the accessibility of pulping industries and their respective technology levels.

3.4. Case D: lab produced chitosan

Finally, as BSF-sourced chitosan is not commercially available, primary data from a previous study were used to simulate a

Table 2
Chitosan production.

Chitosan production					
Input					
Flow	Amount	Unit	Basic uncertainty	Data quality entry	Provider (common across all cases unless specified)
Chitin	1.4	kg	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Chitin production, Table 3
Electricity, high voltage	1.06	kWh	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Case A: Market for electricity, high voltage | Electricity, high voltage | Cutoff, S – IN -western grid Cases B, C: Market for electricity, high voltage | Electricity, high voltage | Cutoff, S – VN
Heat, natural gas	31.3	MJ	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Market for heat, central or small-scale, natural gas | Heat, central or small-scale, natural gas | Cutoff, S - RoW
Sodium hydroxide, 50% solution state	5.18	kg	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Market for sodium hydroxide, without water, in 50% solution state | Sodium hydroxide, without water, in 50% solution state | Cutoff, S - GLO
Tap water	250	kg	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Market for tap water | Tap water | Cutoff, S - RoW
Wastewater	−0.25	m^3	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	Market for wastewater, average | Wastewater, average | Cutoff, S - RoW
Output					
Chitosan	1	kg	1.05	Case A: (1; 3;1; 1;2) Case B: (1; 4;1; 4;2) Case C: (1; 4;1; 4;2)	

scenario where chitosan can be produced locally in a circular economy. The results showed a drastic increase in impacts due to the significant use of sodium hydroxide and hydrochloric acid in the laboratory process without any initial consideration for the environmental impact. While impacts from transportation were slightly reduced, the unoptimized process of chitosan extraction

Table 3
Chitin production.

Chitin production					
Input					
Flow	Amount	Unit	Basic uncertainty	Data quality entry	Provider (common across all cases unless specified)
Diesel	0.91	MJ	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for diesel, burned in building machine | Diesel, burned in building machine | Cutoff, S - GLO
Electricity, high voltage	1.3	kWh	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Case A: Market for electricity, high voltage | Electricity, high voltage | Cutoff, S – IN - western grid Cases B, C: Market for electricity, high voltage | Electricity, high voltage | Cutoff, S – VN
Hydrochloric acid, 30% solution state	2.56	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for hydrochloric acid, without water, in 30% solution state | Hydrochloric acid, without water, in 30% solution state | Cutoff, S - RoW
Inert waste	−1.5	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Treatment of inert waste, inert material landfill | Inert waste, for final disposal | Cutoff, S - RoW
Sodium hydroxide, 50% solution state	1.3	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for sodium hydroxide, without water, in 50% solution state | Sodium hydroxide, without water, in 50% solution state | Cutoff, S - GLO
Tap water	167	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for tap water | Tap water | Cutoff, S - RoW
Material transport, tractor and trailer, agricultural	953.7	kg*km	2	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for transport, tractor and trailer, agricultural | Transport, tractor and trailer, agricultural | Cutoff, S - GLO
Waste shrimp shell	33	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Waste shrimp shell, recycled content, Cutoff
Wastewater	−0.167	m^3	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for wastewater, average | Wastewater, average | Cutoff, S - RoW
Deionized water	5.44	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	Market for water, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW
Output					
Carbon dioxide	0.7	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	
Chitin	1	kg	1.05	Case A: (1; 3;1; 1;2) Cases B, C: (1; 4;1; 4;2)	

Table 4
Chitosan production (laboratory), used in Case D only.

Chitosan production (laboratory)					
Input					
Flow	Amount	Unit	Basic uncertainty	Data quality entry	Provider
Chitin, laboratory	1	g	1.05	(2; 4;1; 1;5)	Chitin, laboratory, Table 5
Electricity, low voltage	1.80E-05	MWh	1.05	(4; 4;1; 1;5)	Market for electricity, low voltage | Electricity, low voltage | Cutoff, S - SG
Inert waste	0.36	g	1.05	(2; 4;1; 1;5)	Market for inert waste, for final disposal | Inert waste, for final disposal | Cutoff, S - RoW
Sodium hydroxide, 50% solution state	20	g	1.05	(2; 4;1; 1;5)	Market for sodium hydroxide, without water, in 50% solution state | Sodium hydroxide, without water, in 50% solution state | Cutoff, S - GLO
Deionized water	500	g	1.05	(2; 4;1; 1;5)	Water production, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW
Output					
Chitosan, laboratory	0.64	g	1.05	(2; 4;1; 1;5)	

Table 5
Chitin production (laboratory), using primary data on utilized chemical.

Chitin production (laboratory)					
Input					
Flow	Amount	Unit	Uncertainty	Data quality entry	Provider
BSF exuviae	50	g	1.05	(2; 4;1; 1;5)	
Electricity, low voltage	1.80E-05	MWh	1.05	(4; 4;1; 1;5)	Market for electricity, low voltage | Electricity, low voltage | Cutoff, S - SG
Hydrochloric acid, 30% solution state	121.5336	g	1.05	(2; 4;1; 1;5)	Market for hydrochloric acid, without water, in 30% solution state | Hydrochloric acid, without water, in 30% solution state | Cutoff, S - RoW
Inert waste	0	g	1.05	(2; 4;1; 1;5)	Market for inert waste, for final disposal | Inert waste, for final disposal | Cutoff, S - RoW
Sodium hydroxide, 50% solution state	320	g	1.05	(2; 4;1; 1;5)	Market for sodium hydroxide, without water, in 50% solution state | Sodium hydroxide, without water, in 50% solution state | Cutoff, S - GLO
Material transport	25 E^-4	t*km	2	(4; 4;1; 1;5)	Market for transport, freight, lorry, unspecified | Transport, freight, lorry, unspecified | Cutoff, S - RoW
Deionized water	0.947195	kg	1.05	(2; 4;1; 1;5)	Water production, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW
Deionized water	3.59488	kg	1.05	(2; 4;1; 1;5)	Water production, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW
Deionized water	5	kg	1.05	(2; 4;1; 1;5)	Water production, deionized, from tap water, at user | Water, deionized, from tap water, at user | Cutoff, S - RoW
Output					
Chitin, laboratory	9.29	g	1.05	(2; 4;1; 1;5)	

was a key challenge for the BSF-based FLAM.

3.5. Uncertainty dispersion

The calculations were iterated 10,000 times with the underlying numerical inputs sampled from their respective distributions (i.e., triangular or lognormal) through a Monte Carlo Analysis that calculates uncertainty propagation. These iterations produce a corresponding result, following a lognormal distribution when the results of all the 10,000 iterations are collated. It is noteworthy that compared to those studies not considering variability and relying on single calculations with absolute input values, the consideration of the uncertainty results in higher median values.

The results from the Monte Carlo Analysis were normalized to their respective distribution median impact level and is presented in Fig. 6 to visualize the resultant spread of calculated impacts between case scenarios. Interestingly, although uncertainty factors were adjusted for all cases, observable uncertainty emerged from the technological differences between the laboratory extraction of BSF-based chitosan and the upscaled commercial extraction of crustacean chitosan. Consequently, the choice of reducing the import distance and utilizing recycled cellulose fibers did not result in any observable difference. There was no evident change in the overall uncertainty in the midpoint impacts between using virgin cellulose fibers and using recycled cellulose fibers.

4. Conclusion

Since their main constituents are available from waste and can be decomposed without human intervention, FLAMs enable an unprecedented generalization of circular manufacturing models, where consumables are sourced, produced, and degraded in closed regional areas. In this study, we exhaustively evaluated the current state of the technology and determined areas where future improvements should be made. Our analysis of the four scenarios yielded the following results.

1. The current FLAM production practice, i.e., international material importation, results in lower environmental impacts on most midpoint indicators relative to the use of similar materials,

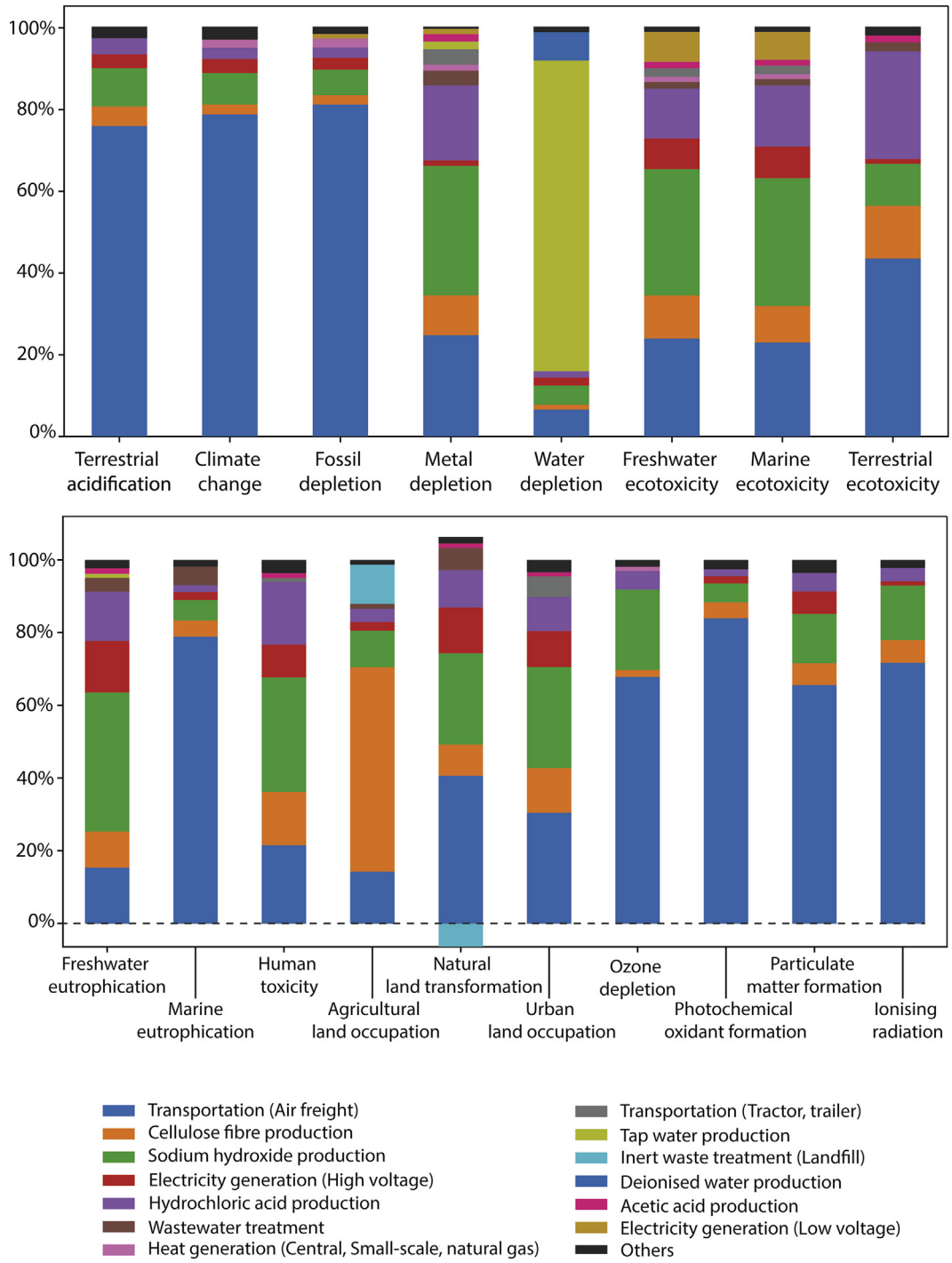


Fig. 4. Case A process contributions normalized to median of respective midpoint indicators. Key contributors, such as material transportation via air freight and use of chemicals (sodium hydroxide, hydrochloric acid), can be observed across all midpoint indicators.

such as rigid PUR foam. However, international material sourcing results in relatively high environmental impacts on indicators such as climate change and fossil depletion due to the great material import distances.

2. Reducing distances between suppliers and FLAM production facilities within a geographical radius of 3000 km substantially reduces environmental impacts. Current approaches that link bioinspired materials with circular and regional production paradigms are critical.
3. Considering environmental impacts, there is flexibility in terms of using virgin cellulose fibers or recycled cellulose fibers when state-of-the-art pulping technology is available. For cities that do not have substantial renewable energy sources in their electricity grids or well-established pulping facilities, recycled cellulose fibers from wastepaper would be suitable for FLAMs as well.
4. Unoptimized chitosan production from arthropods is environmentally unsustainable due to its excessive chemical waste

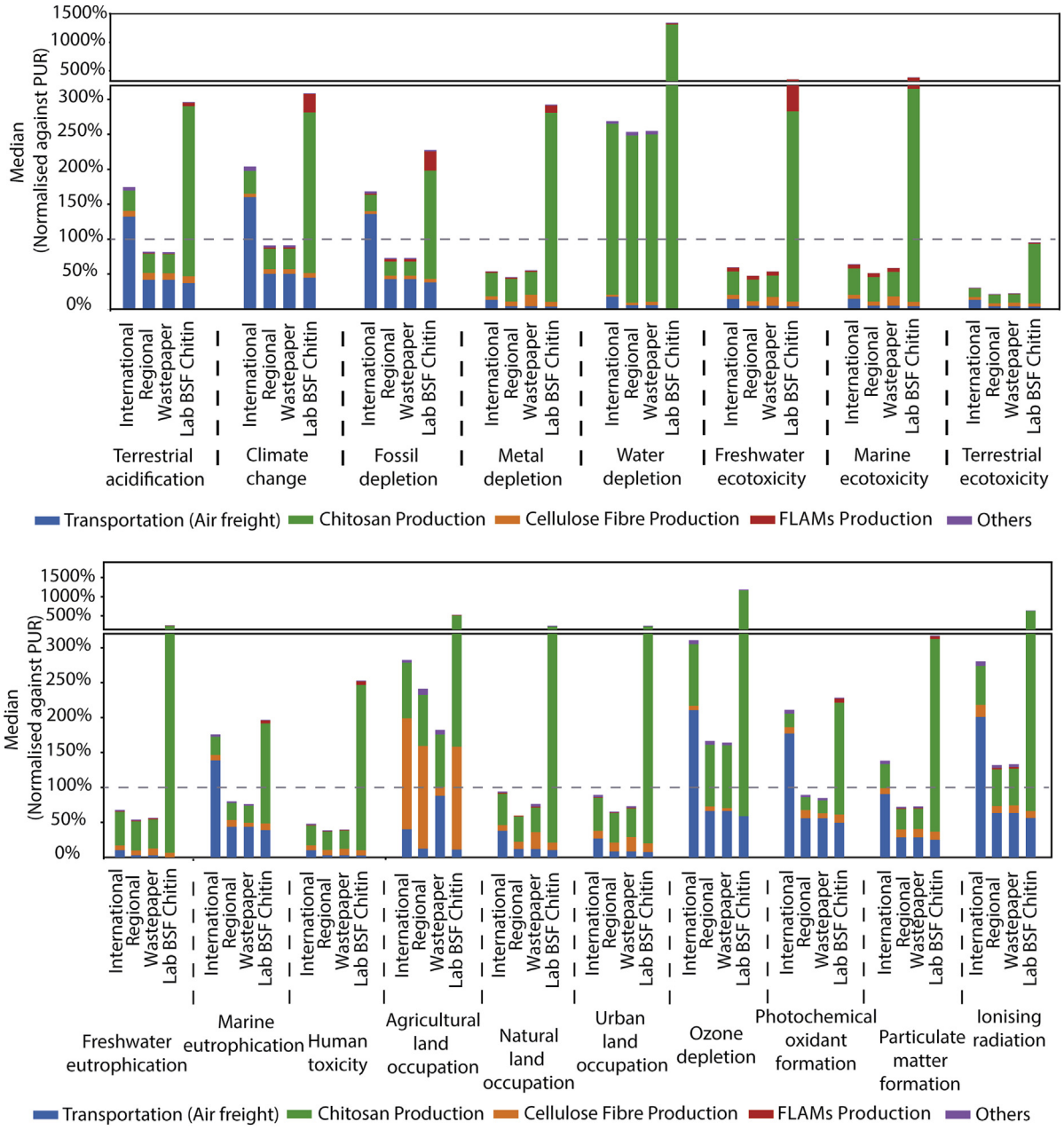


Fig. 5. Normalized median (to PUR production process) for four scenarios across all midpoint indicators. Case scenario contributions less than 100% signify a lesser environmental impact compared with that of rigid PUR foam.

production. This impact significantly decreases when the current industry standards of crustacean-based chitosan processing are considered. Relying on crustacean waste conflicts with the regionalization of production, which directly affects transportation. Thus, the industry should adopt more efficient, updated methods of chitinous composite fractionation. Moreover, alternative sources of chitin, such as BSFs or fungi from organic waste bioconversion, can be implemented anywhere.

5. Bioinspired manufacturing enables the use of ubiquitous materials and therefore constitutes an unprecedented opportunity to regionalize the production of consumables. This regionalization is not only a possibility but also a necessity. Bioinspired manufacturing within the current globalized paradigm results in limited improvements compared with the full potential of the technology within a circular and regionalized paradigm.

To further develop FLAMs as materials that enable substantial changes in production patterns, we recommend the following.

1. More efficient fractionation methods of chitinous organisms should be assessed. Alternative sources of chitinous polymers could reduce the need for chemical extraction. Fungal chitosan is commercially available and can be produced by bioconversion of organic waste; it does not require demineralization or further deacetylation (Abo Elsoud and El Kady, 2019). Broadening chitin sources can also directly impact its adaptability to different ecosystems.
2. Fermentation processes and enzymatic treatment in crustacean-based chitosan production are a promising alternative to the current chemical extraction method of chitosan (Yadav et al., 2019). Future research should focus on sustainable, large-scale

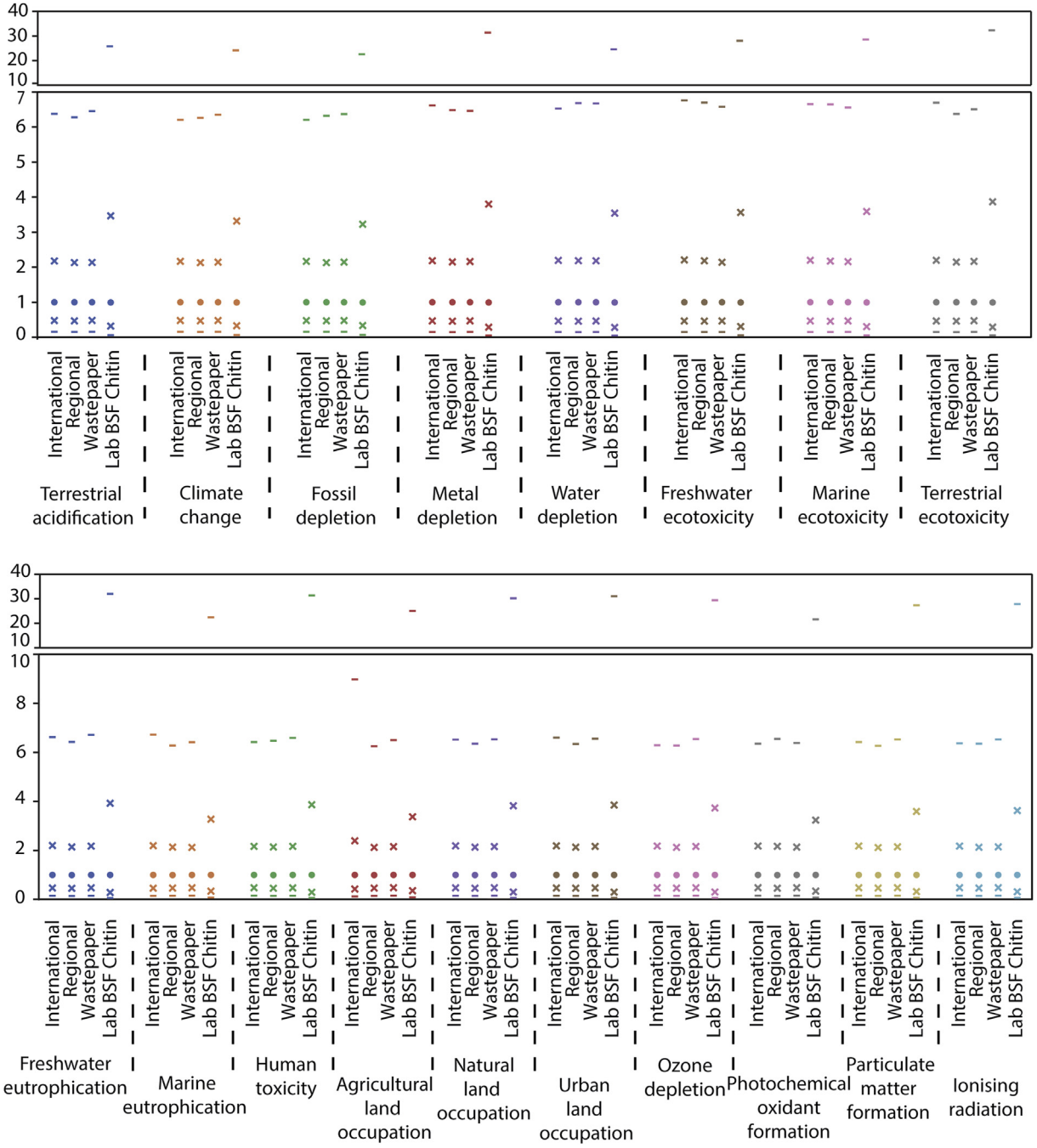


Fig. 6. Distribution of each indicator median due to propagation of uncertainty during aggregation of respective case scenario. The cross markers indicate the 25% and 75% percentiles, while the line markers indicate the 5% and 95% percentiles of data resulting from over 10000 iterations of Monte Carlo simulations.

production of chitosan to maximize the potential environmental benefits of chitinous products and biocomposites.

3. In addition to novel products, the use of FLAMs in consumable/disposable applications must continue to generate market demand that can ultimately drive technological improvements and incentivize industries to aim to lower costs and increase efficiency. FLAMs' ability to meet society's commercial and cultural needs will be the key driver of change in terms of production patterns and consumer behavior, which are the fundamental enablers of sustainable development (Delano, 2017).

CRediT authorship contribution statement

Shiwei Ng: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft, Visualization, Data curation. **Bin Song:** Methodology, Data curation, Supervision. **Javier G. Fernandez:** Conceptualization, Resources, Writing - review & editing, Supervision, Project administration, Funding acquisition.

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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