

Life cycle assessment of novel aerogels based on lignocellulosic residues from agriculture and forestry

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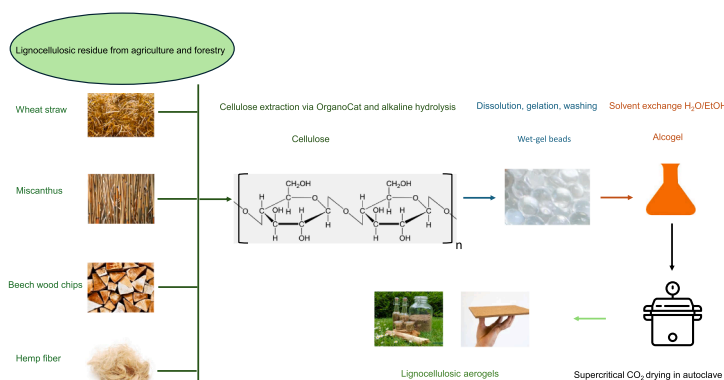
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HIGHLIGHTS

- LCA compares different lignocellulosic waste for lignocellulosic aerogel synthesis.
- Environmental impacts of alkali hydrolysis and OrganoCat are compared.
- Ethanol and acetic acid contribute more to environmental impacts than scCO₂ drying.
- Upscaling reduces environmental impact by 20–94% depending on the environmental impact category.

GRAPHICAL ABSTRACT



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ABSTRACT

The environmental impacts of aerogel synthesis from lignocellulosic residues are analyzed using the Life Cycle Assessment (LCA) approach. All steps in the process chain, cradle-to-gate, are considered: from agricultural processes to cellulose extraction, the preparation of wet-gel beads and alcogels, and aerogels obtained by drying with supercritical carbon dioxide. Two methods of cellulose extraction are discussed. Life cycle inventory was built using primary data from the laboratory and with support of the Sphera LCA for Experts software. Environmental impacts are quantified using the functional unit of 1 kg of lignocellulosic-based aerogel. The focus is to assess the contribution of each process to the overall environmental performance and to identify hotspots during production. A comparison of the individual unit operations of the process chain shows that the alcogel

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preparation, followed by cellulose extraction, contributes most to the overall impact. This is predominantly caused by the supply of ethanol, acetic acid and 2-methyltetrahydrofuran. As aerogel preparation was carried out on a laboratory scale, these results can be regarded as a conservative estimate and thus serve as a basis for process optimization during scale-up. With the assumed improvements during scale-up and in a scenario for 2050, significant reductions in environmental impacts can be achieved, e.g., by 87 – 92% in terms of the greenhouse gas balance, by 75 – 87% in terms of acidification and by 92 – 94% in terms of resource availability. However, a comparative LCA with chosen benchmarks shows that lignocellulosic-based aerogels are not yet fully competitive.

1. Introduction

Besides sodium-ion batteries, liquid solar fuels and vaccines with spherical nucleic acids, aerogels were listed among the ten most important emerging technologies by the International Union of Pure and Applied Chemistry (IUPAC) in 2022 [1]. Aerogels are nanostructured open-porous materials that are synthesized from various inorganic and organic sources [2]. They are characterized by high porosity, large specific surface area, low density, low thermal conductivity, and low acoustic velocities [3]. This makes them highly thermal insulating materials with excellent sound-absorbing properties that are also easy to functionalize [4]. The areas of application are very diverse [5–8] and range from thermal insulation of buildings [9,10], pipelines [11], and textiles, automotive and space applications, cosmetics [12], drug delivery [13], catalysts [14], batteries [15], sorbents for oil spills [16], filter [17] to food packaging [18].

A special class of aerogels is synthesized from bio-based sources and therefore offers sustainable, biocompatible, and recyclable properties [19,20]. Among these, cellulose-based materials are the most well-studied ones [2]. Agricultural residues such as wheat straw, rapeseed straw, rice straw, corn stover, flax shives, sugarcane bagasse, pineapple fibers and other natural resources such as wood, hemp fibers and energy crops like miscanthus have been investigated as sources not only for cellulose, but also for hemicellulose and lignin [21,22]. Due to their abundance, renewability, and lack of competition with food applications, these bio-based residue sources have high potential as substitutes for chemicals and materials derived from fossil-based raw materials [17].

Pre-treatment and fractionation are essential steps in utilizing the recalcitrant and heterogeneous lignocellulosic biomass [17]. Different technologies, such as steam explosion, hydrothermal, alkali, and acid treatments have been developed [23]. Organosolv-like technologies [24, 25] enable targeted lignocellulose fractionation and recovery of value streams beyond the cellulose fraction. The OrganoCat technology – being one of the organosolv-like treatments - provides good component separation into a cellulose-rich pulp, a hemicellulose-derived fermentable sugar solution in the aqueous phase and a lignin fraction in a

separate organic phase [26–28]. The mild reaction conditions of OrganoCat hydrolyze the amorphous non-cellulosic polysaccharides, and lignin is extracted in 2-methyltetrahydrofuran (2-MTHF) [23]. Alkaline hydrolysis has also mild process conditions and requires less energy. Common laboratory chemicals such as sodium hydroxide, potassium hydroxide, hydrogen peroxide, and acetic acid are used for cellulose extraction. The use of chlorinated chemicals is avoided, resulting in less environmentally harmful waste [29–32].

The current study compares two cellulose extraction processes, alkali hydrolysis and the OrganoCat process, using Life Cycle Assessment (LCA). The remaining cellulose-rich pulp is further processed into aerogels via the intermediate products wet-gels and alcogels and subsequent drying with supercritical CO₂ (scCO₂) (Fig. 1).

A recent review found that between 2012 and 2024, only 20 articles on aerogels had been investigated using LCA [33], and two articles were based on Environmental Product Declaration (EPD) [34,35]. Ten of these articles focus on silica-based aerogels, six are based on starch, two on alginates and three others are organic-based (protein, polymer). Five and four studies were conducted by two research groups investigating starch-based [36] and silica-based aerogels [37], respectively. Only one LCA study has examined a lignocellulose-based aerogel [17]. Unfortunately, Sun et al. [17] conducted the Life Cycle Impact Assessment (LCIA) using endpoint categories and is therefore not comparable with all other studies that used the more common midpoint categories. To the best of the authors' knowledge, no further LCA study on lignocellulose-based aerogel has been conducted to date.

Drying methods for aerogels are a key process step in many LCA studies and vary depending on the drying conditions. Xerogels are produced under ambient conditions, aerogels under supercritical conditions, and cryogels under low pressure through freeze-drying. The share of drying processes examined in LCA studies on aerogels is 30% scCO₂ drying, 37% freeze drying, 30% ambient drying, and 3% vacuum drying [33]. According to Kara et al., drying with scCO₂ is a safer and more environmentally friendly option than other drying methods. A recent review of LCAs of supercritical fluids processes shows the wide range of inputs and the resulting environmental impacts. However, only 7% of the 70 LCA studies examine scCO₂ drying, and only two of these

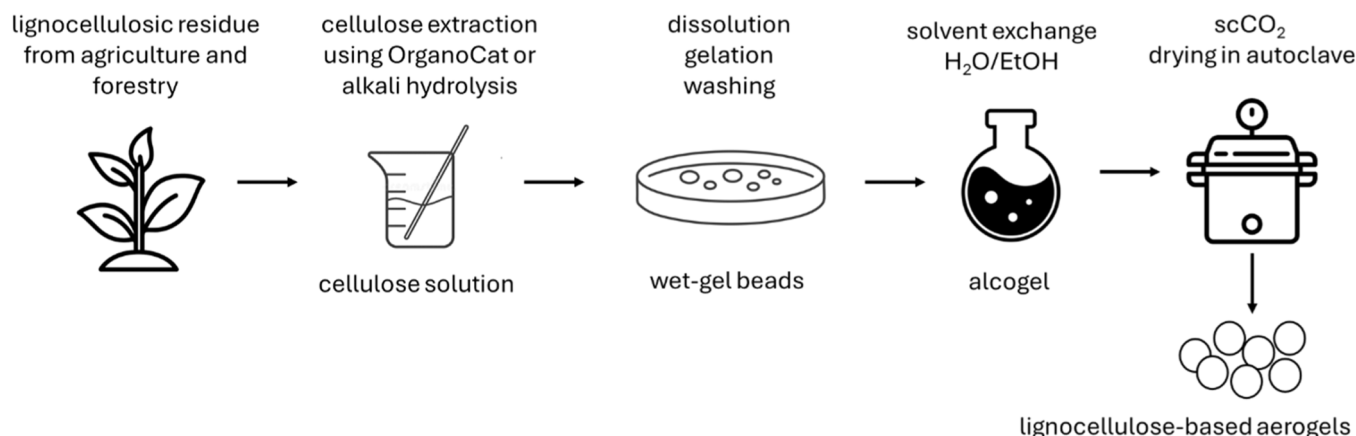


Fig. 1. General process diagram mapping the transition of lignocellulosic residue to aerogels.

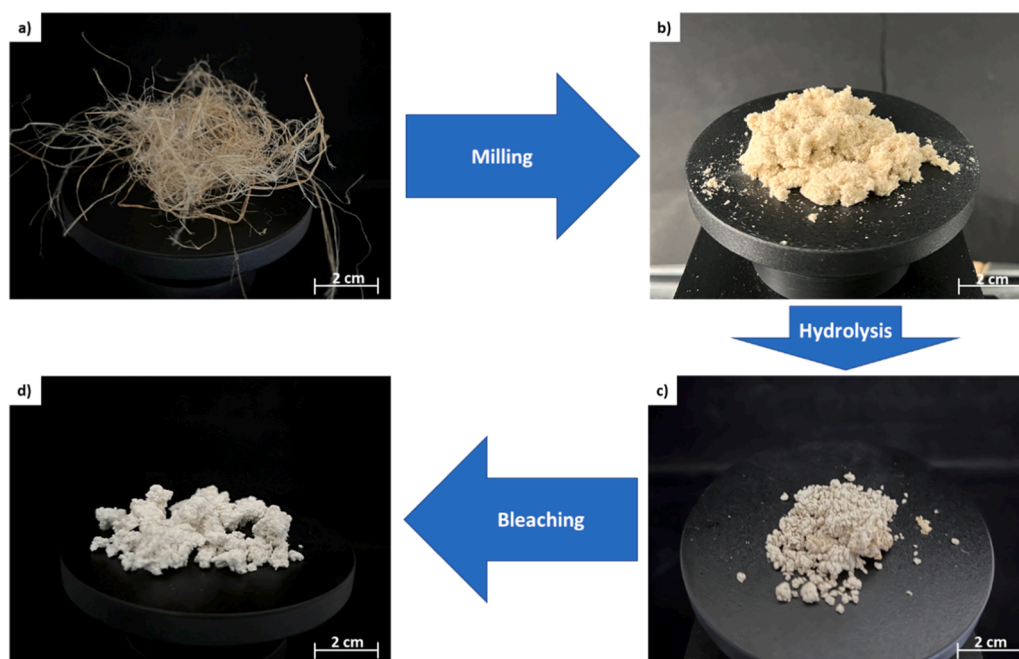


Fig. 2. Stages of cellulose extraction procedure for hemp: a) hemp fibers; b) milled hemp fibers; c) alkali hydrolyzed hemp; d) hemp cellulose.

studies analyze scCO_2 drying in connection with aerogels [13,36]. The other LCA studies mainly use supercritical fluids (e.g., methanol, water, CO_2) for extraction, gasification/reforming, recycling, and waste treatment processes [38]. A more sustainable production of aerogels is one of the motivations behind this study. LCA is a powerful tool for investigating whether lignocellulose-based aerogels are more sustainable than their conventional (fossil-based) benchmarks. As many questions remain unanswered regarding the efficiency and yield of the manufacturing process, as well as recycling of solvents and acids, the superiority of bio-based aerogels is by no means guaranteed [20]. Furthermore, the synthesis of lignocellulosic aerogels is currently at a low technology readiness level (TRL), which makes it difficult to perform an LCA based on accurate data. However, given the rapid progress in research [39–41], the time is right for initial efforts in this direction [20].

Therefore, this LCA study focuses on the environmental impacts of manufacturing lignocellulosic aerogels from different agricultural and forestry residues (wheat straw, hemp fibers, beech wood chips, miscanthus) which were produced within the framework of the GelSus project [42]. The inventory is first compiled using the manufacturing data from the laboratory scale (hereinafter referred to as lab scale) before a theoretical upscaling to an industrial scale is carried out. A scenario for 2050 and a comparison of the lignocellulosic aerogels with benchmark products complete the LCA. Fig. 1 shows the general process diagram.

2. Materials and Methods

The biomass feedstocks wheat straw, miscanthus, and beech wood were harvested in the area around the Forschungszentrum Jülich (Germany), hemp fibers were cultivated in Canada, near Langley (British Columbia) and provided by Canadian project partners from the University of British Columbia. As part of the GELSUS project, cellulose was extracted from all four biomass sources using the OrganoCat process; however, only hemp fiber and beech wood chips were treated with alkaline hydrolysis. Cellulose from the latter two sources was then used for further processing into aerogels (see also 3.3.1).

2.1. Cellulose extraction via OrganoCat process and alkali hydrolysis

2.1.1. OrganoCat process

The biomass is dried until weight constancy for 24 h at 50 °C. This results in a water content of approx. 10%. An OrganoCat fractionation has been conducted as described by Weidener et al. [21]. Approximately 1 g of each biomass sample are ground to a fine powder using a ball mill M 400 (Retsch, Haan, Germany) with stainless steel grinding balls (14.7 mm) in a 50 mL metal beaker (30 s^{-1} , 2 min) [21]. Then, 500 mg of biomass is suspended in 5 mL of aqueous phase (0.1 M oxalic acid) and 5 mL of organic phase (2-MTHF) before it is placed in a 25 mL stainless-steel reactor (Büchi AG, Uster, Switzerland). The reactor is pressurized with 10 bar argon to prevent evaporation of the solvent and heated to 140 °C, maintaining this temperature for 3 h while stirring at 1500 rpm. After cooling and depressurization, the reaction mixture is transferred to a 50 mL Falcon tube (Merck KGaA, Darmstadt, Germany) and centrifuged at 1880 g for 5 min to achieve improved phase separation. The organic phase is then collected using a syringe, and 2-MTHF is evaporated to obtain the lignin fraction. The aqueous phase is filtered with a paper filter (pore size 5 – 8 μm) and stored at –4 °C for subsequent analysis. The residual cellulose pulp is washed with distilled water until neutral pH is reached and then air-dried to constant weight.

2.2. Alkali hydrolysis

A three-step procedure was used to extract cellulose from milled hemp fibers and beech wood chips (Fig. 2): (i) Milling; (ii) Alkali hydrolysis; (iii) Bleaching.

- (i) Milling: Biomass fibers are cut into fragments of 1–2 cm and then milled with a rotor beater mill (SR200, Retsch GmbH, Haan, Germany) with a 0.2 mm sieve. Milling is done to achieve a higher yield of extracted cellulose due to the generation of a higher surface area [43]. Liquid nitrogen is poured every 5 min into the mill to refrigerate the fibers, keeping them stiff and avoiding overheating.
- (ii) Alkali hydrolysis: The alkali treatment is carried out in a round bottom flask containing a mixture of 900 g of sodium hydroxide (NaOH; 8 wt.-%) solution and 50 g of the milled biomass fibers.

Table 1
Cellulose yield as a function of lignocellulosic residue and extraction method.

Cellulose source	Extraction method	Yield (%)	Concentration (g/100 g)*
Hemp fibers	Alkali hydrolysis and bleaching	65	2.5
	OrganoCat	87	5.0
Beech wood chips	Alkali hydrolysis and bleaching	55	4.5
	OrganoCat	69	3.5

* Cellulose concentration is reported in g/100 g NaOH-urea-water mixture.

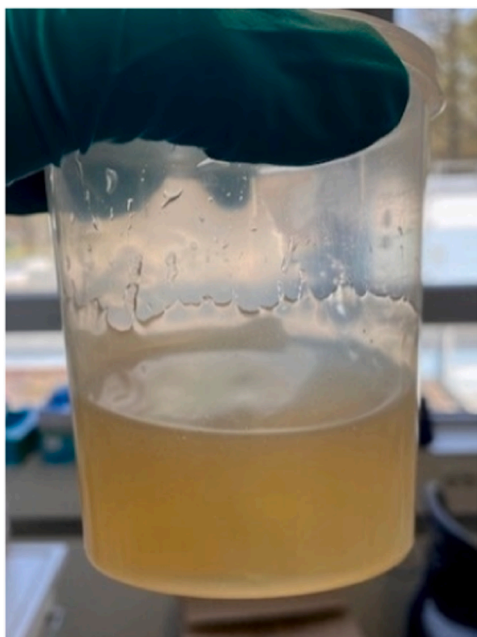


Fig. 3. Cellulose solution (example 2.5 g of the extracted hemp cellulose (alkali hydrolysis process) in 100 g of NaOH-urea-water mixture).

After stirring for 18 h at room temperature, the mixture is heated at 60 °C for three h. After cooling down to room temperature (23 °C) the mixture is neutralized using glacial acetic acid. Next, the fibers are collected by filtration (mesh 200 μm) and washed five times with 500 mL of distilled water. Subsequent washing with ethanol and acetone removes any remaining organic residues and water. Finally, the fibers are dried in a vacuum oven at 50 °C overnight. After alkali hydrolysis the yield is 95% for hemp fibers.

- (iii) Bleaching: The fibers obtained from the alkali hydrolysis process are employed for the bleaching process. The fibers are dispersed in a round bottom flask containing an aqueous solution (900 g) of potassium hydroxide (KOH; 5 wt.-%). To this mixture, 45 mL of hydrogen peroxide in water (from stock solution; 30 wt.-%) is added and stirred for 18 h at room temperature. Then the mixture

is heated at 60 °C for three h, cooled down to room temperature and neutralized using glacial acetic acid. The pale white fibers are collected by filtration and washed five times with 500 mL of distilled water. Then the mixture is washed with ethanol and acetone, and the vacuum drying is performed at 50 °C. After the bleaching process a yield of 65% compared to the raw hemp fibers is generated.

2.3. Preparation of cellulose solution

The cellulose solution is prepared according to the procedure reported in literature [44]. While stirring 7 g of NaOH is added to 81 g of distilled water. The solution is stirred until NaOH is fully dissolved. At room temperature, the purified cellulose fibers from hemp or beech wood are added to the solution at a previously defined concentration (g/100 g of NaOH-urea-water, see Table 1). The solution is stirred for an additional 15 min to achieve a good dispersion of the cellulose. Then, the solution is stirred in a pre-cooled (at -10 °C) glycerol-water bath for one h in order to initiate the dissolution process. Next, 12 g of urea is added to the cold cellulose solution and stirred for an additional hour at the same conditions, followed by stirring 15 min at room temperature before it is stored in a freezer (at -20 °C) overnight. Finally, the solution is defrosted to room temperature and stirred for 30 min to get a highly viscous homogeneous liquid (Fig. 3). The quality of extracted cellulose changes depending upon the extraction conditions. Therefore, in order to prepare the cellulose solution, which is good for building the spherical beads shape, different quantity of cellulose is employed for the solution preparation (see Table 1).

2.4. Preparation of cellulose aerogel beads

Wet-gel beads are produced by a conventional dropping technique, using a multi-nozzle dropping setup with a nozzle diameter of 3 mm (hemp) or 0.8 mm (beech wood). The cellulose solution is dropped into a gelation bath containing 2 M of aqueous acetic acid. The wet-gel beads are formed after the complete diffusion of acid through the cellulose droplet. After 30 min stirring in gelation bath, the wet-gel beads are collected and washed several times with water in order to neutralize the beads. Next, a stepwise solvent exchange with ethanol is performed. The ethanol to water volume ratio is increased in each step by 20% of ethanol (about three h aging time), followed by an additional exchange with 100% ethanol. In the ethanol bath, the gels were aged for 18 h. After the solvent exchange, the alcogel beads are dried under scCO₂ conditions to obtain the aerogel beads (Fig. 4). The drying process is conducted with a HTPe-150p extractor (Eurotechnica GmbH, Bargteheide, Germany) at 115 bar and 60 °C with an average CO₂ mass flow of 22.5 kg·h⁻¹ over a period of 4 h. All cellulose solutions can be used to synthesize aerogel beads successfully, except the beech wood cellulose from alkali hydrolysis process. The reason for this is that the cellulose solution from beech wood (alkali hydrolysis) is a suspension with more undissolved cellulose. In this case, a stable colloidal suspension is not possible.

The aerogels were characterized by envelope density measurement (Micromeritics GmbH, Unterschleißheim, Germany; GeoPyc 1360; 35 N;



Fig. 4. Stereomicroscope images of cellulose aerogel beads from hemp (left, cellulose obtained from alkali hydrolysis, middle: cellulose obtained from OrganoCat) and beech wood (right, cellulose obtained from OrganoCat).

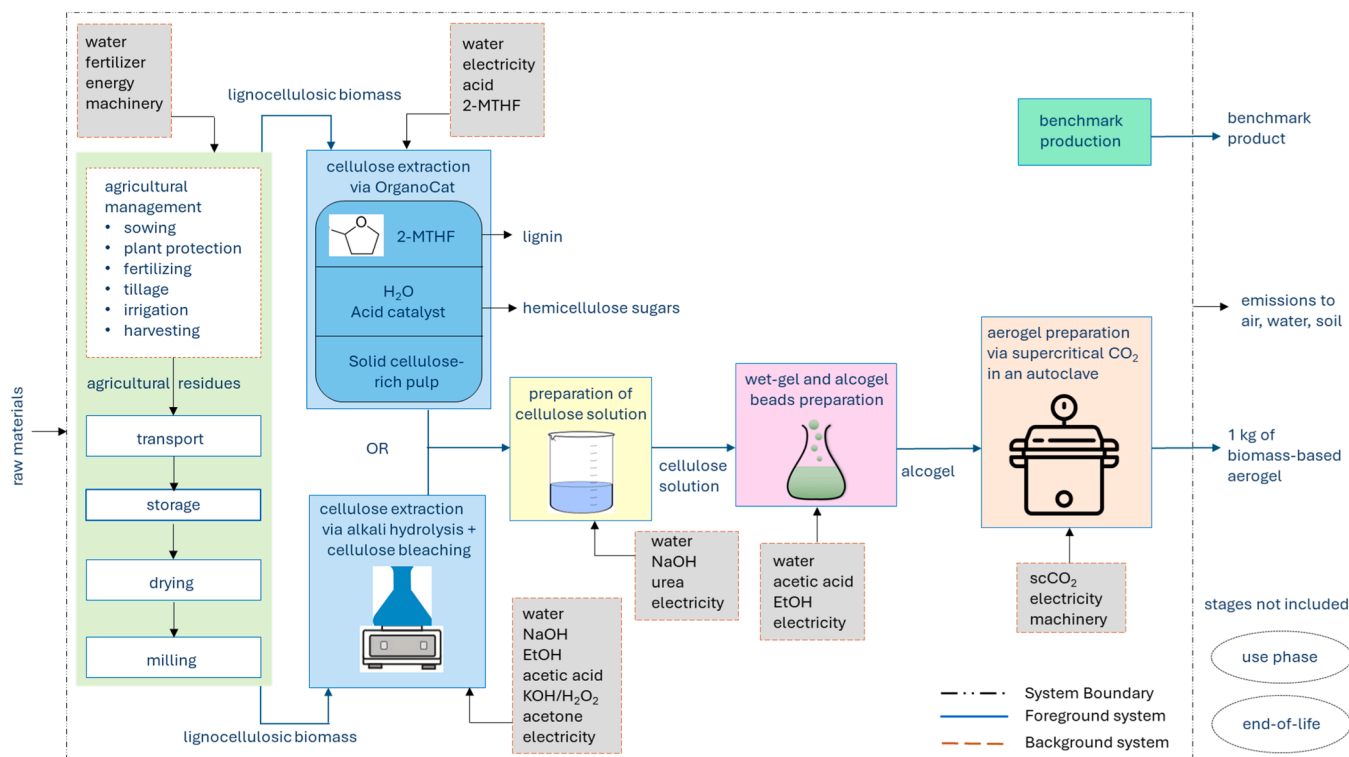


Fig. 5. LCA system boundaries of biomass-based aerogel production.

Table 2
Chosen impact categories of EF3.1 method.

Impact category	Unit	Abbreviation
Acidification	Mol H ⁺ eq.	AP
Climate Change – total	kg CO ₂ eq.	CC
Eutrophication, freshwater	kg P eq.	EP _{fw}
Ecotoxicity, freshwater	CTU _e	ETP
Land use	Pt	LU
Resource use, fossils	MJ	ADP _f
Resource use, mineral and metals	Sb eq.	ADP _{mf}
Water use	m ³ world eq.	WU

using a dry-flow medium), BET nitrogen adsorption-desorption isotherm analysis (Micromeritics GmbH, Unterschleißheim Germany; Tristar II 3020). The thermal conductivity of the cellulose beads was measured using the HFM 436 Lambda (NETZSCH-Gerätebau GmbH, Selb/Bayern, Germany). The beads were filled in a 10 × 10 × 1 cm mold and then the analysis was conducted at 25 °C.

3. LCA Methodology

This study uses the LCA approach in accordance to the ISO standards 14040 and 14044 [45,46] to evaluate the environmental impacts associated with biomass-based aerogel production in an attributional cradle-to-gate approach. The software package utilized for modelling process chains and for calculation of impacts is LCA for experts (version 10.9.3.0, previously known as GaBi software) [47].

3.1. Goal and scope definition, functional unit, system boundaries

The goal and scope of the current study is to evaluate the environmental performance of lignocellulosic aerogels by quantifying various ecological impacts associated with their lab scale manufacturing (TRL < 5). The processes with the highest environmental implications are to be identified and potential for improvement are highlighted. Two different

process paths for cellulose extraction from the biomass (alkali hydrolysis and OrganoCat process) are examined and compared. Additionally, a theoretical upscaling to industrial scale (TRL 7–8) is carried out using well-founded assumptions and key parameters that have been discussed and applied in previous studies [48,49]. The equipment extrapolation was performed by using the study from Pushpendra et al. [49]. The scaling of processes, especially for biorefinery processes, is still associated with considerable uncertainty, although individual studies have attempted to provide a framework for data estimation for upscaling from lab scale to industrial scale [50–52]. However, the studies are mostly limited to energy or chemical processes. Furthermore, a scenario for 2050 is drawn up to estimate the best-case scenario from today's perspective, knowing that this involves a high degree of (data) uncertainty. Finally, the lignocellulosic aerogels are compared with appropriate benchmarks for different application fields.

The analysis is carried out “cradle-to-gate” with the aim of assessing the environmental impact of the manufacturing of lignocellulosic aerogels. Use and end-of-life (EoL) processes are not included in the analysis, as it is assumed that these are identical for both lignocellulosic aerogels and benchmark materials. The functional unit (FU) selected for identifying hotspots and comparing lignocellulosic aerogels is initially 1 kg of the lignocellulosic aerogels. For benchmarking with other materials additional product specific functionalities, such as thermal isolation potential, are considered in the FU (see Section 4.4).

The system boundary drawn for this study focuses on the manufacturing stage in Germany consisting of five major sub-stages (Fig. 5): (i) lignocellulosic biomass supply, (ii) cellulose extraction/bleaching, (iii) preparation of cellulose solution, (iv) wet-gel and alcogel preparation, (v) aerogel preparation by scCO₂ drying. Transportation processes are also considered. Except for hemp, which was supplied by the Canadian project partners, the three other biomasses (wheat straw, miscanthus, beech wood chips) are cultivated in Germany.

The environmental impacts are assessed by applying the Environmental footprint method (EF) 3.1, following the recommendation of the EU and its wide recognition [53–55]. Eight impact categories are chosen based on the most frequently used categories in aerogel LCAs (Table 2)

Table 3
Summary of data origin.

Process step	Data origin			
	Wheat straw	Hemp fiber	Miscanthus	Beech wood chips
Biomass supply	database	database	database	database
Cellulose extraction: by OrganoCat	lab	lab	lab	lab
by alkali hydrolysis	not considered	lab	not considered	lab
Preparation of cellulose solution	not considered	lab*	not considered	lab**
Wet-gel and alcogel preparation	not considered	lab*	not considered	lab**
scCO ₂ drying	not considered	lab*	not considered	lab**

* each performed with cellulose from the OrganoCat process and from alkali hydrolysis

** only carried out with cellulose from the OrganoCat process

[33].

3.2. Data collection

Most of the primary life cycle inventory (LCI) data of the foreground system for lab scale were determined experimentally by the authors (Section 2). Data of the background systems (e.g., supply of water, chemicals, auxiliary materials, etc.) has been taken mainly from the Sphera Professional database [47], from the Ecoinvent 3.10 database [56,57], and from literature. Some background process data was also modeled, e.g., for electricity, hydrogen and 2-MTHF supply as well as bio-based methanol for future truck transport. Assumptions and parameters for upscaling to industrial scale and for the 2050 scenario are identified from the lab scale, expert opinions, and published papers on similar technologies [48–52]. They are specified in the description of the individual process steps and in the LCI.

3.3. Life cycle inventory

3.3.1. General assumptions for lab scale, industrial scale and scenario 2050

The manufacturing process for lignocellulosic-based aerogels

Table 4
Parameter assumption for lab scale, industrial scale and scenario 2050.

Parameter	Lab scale	Industrial scale	Scenario 2050	Source for the upscaling assumption
Biomass-water ratio	1:10	1:6.5	1:6.5	[58], [24], [49]
2-MTHF recycling	-	99%	99%	[48]
Heat integration for 2-MTHF supply	-	+	+	[59]
Use of acetone during alkali hydrolysis	+	-	-	expert opinion*, [60]
EtOH supply	chemically obtained (hydration of ethylene)	bio-based EtOH (fermentation)	bio-based EtOH (fermentation)	own assumption
EtOH recycling (from autoclave)	-	98%	98%	expert opinion*
Recycling of acids, alkalis, water, other chemicals	-	20%	20%	[50]
Transport distance for biomass	25 km	50 km	50 km	[49]
Transport	light commercial vehicle (mix of diesel and petrol)	truck, EURO 6 (diesel)	truck fueled by biobased methanol	own assumption
Electricity supply	mix 2030	av. mix 2030–2055	av. mix 2030–2070	own models
Heat supply	natural gas	waste biomass incineration	wood pellets	own assumption
Electricity and heat demand	-	10% of lab scale demand	10% of lab scale demand	[50], [13]
Hydrogen generation for 2-MTHF supply	steam methane reforming	steam methane reforming	water electrolysis	own assumption
Steel production	conventional coal blast furnaces	conventional coal blast furnaces	electric arc furnaces with renewable electricity	own assumption

* expert opinion refers to the evaluation of the project partners and co-authors from the German Aerospace Center (DLR), Institute for Frontier Materials on Earth and in Space, Department of Aerogels and Highly Porous Materials

consists of five main steps shown in Fig. 5. Table 3 provides an overview of these five main processes, which were carried out experimentally as part of the GelSus project (see Section 2). Cellulose extraction using the OrganoCat process was successfully applied to all types of biomasses. The alkali hydrolysis extraction method was carried out with hemp fibers and beech wood chips.

Table 3 shows that three aerogels were produced for which three LCAs were performed: one based on beech wood and two based on hemp fibers. Data from the Ecoinvent 3.10 database is used for biomass supply and adjusted where necessary. In the case of hemp fibers, the cellulose required for aerogel production was obtained using both possible extraction methods (OrganoCat and alkali hydrolysis). In the case of aerogel made from beech wood chips, only the cellulose obtained using the OrganoCat process could be used (see reason in Section 2.3). The cellulose extracted from wheat straw and miscanthus using the OrganoCat process has not yet been processed into aerogels as part of the GelSus project presented here. It is currently unclear how much cellulose extracted from miscanthus and wheat straw is required to produce a stable aerogel that retains its spherical shape and does not lose its gel structure. This depends on the degree of polymerization of the extracted cellulose. Therefore, no final LCAs can currently be created for aerogels based on miscanthus and wheat straw. These two process lines are therefore only monitored with LCA up to the extracted cellulose using OrganoCat process.

The key parameters chosen for lab scale, industrial scale and the scenario 2050 are listed in Table 4. Based on relevant references, the biomass-water ratio was reduced during upscaling, 2-MTHF recycling was assumed, and chemical and energy consumption was reduced (Table 4). Detailed electricity mixes (Table S 1 - Table S 4), models for transport (Table S 9 - Table S 11) and hydrogen supply (Table S 4) and further specifications are described in the Supplementary Material. Used

Table 5
Allocation factors used for OrganoCat process.

Products	Wheat straw	Hemp fiber	Miscanthus	Beech wood chips
Cellulose	82	91	88	85
Hemicellulose sugars	7	5	3	5
Lignin	11	4	9	10

Table 6
Parameter for benchmarking.

Application	Product	Characteristics	FU quantities for comparison [kg]
Filter	Activated carbon	Specific surface area [m ² /g] 300 ^a	1
	Expanded clay	1.4 ^b	214
	Lava granulates	1.4 ^b	214
	Zeolite powder	500 ^c	0.6
	lignocellulosic aerogel	336 ^d (323 ^e , 297 ^f)	0.893
1 m ² Insulation panel	PU	Thermal conductivity [mW/m ² K]	Density [g/cm ³]
		0.025	0.035
	PS	0.035	0.030
		0.09	0.400
lignocellulosic aerogel	0.037 ^d	0.070 ^{d,e} (0.12 ^f)	2.59

^a max. 2000 m²/kg [61]

^b data based on Angerer [62]

^c data based on Prechel GmbH Mineralien- und Chemikalienhandel [63]

^d hemp-based aerogel (using alkali hydrolysis)

^e hemp-based aerogel (using OrganoCat)

^f beech wood-based aerogel

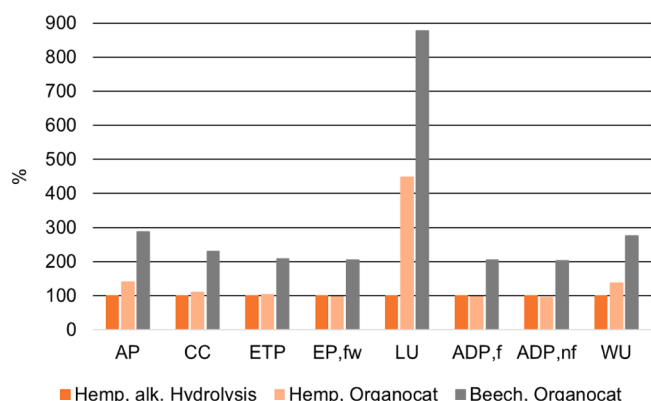


Fig. 6. Relative environmental impacts of lab scale manufacture of 1 kg aerogel based on hemp fibers and beech wood chips (aerogel from hemp fibers using alkali hydrolysis = 100%). AP: Acidification, CC: Climate Change – total, ETP: Ecotoxicity, freshwater, EP_{fw}: Eutrophication, freshwater, LU: Land use, ADP_f: Resource use, fossils, ADP_{nf}: Resource use, mineral and metals, WU: Water use.

EtOH obtained from the autoclave can be reused without further purification processes. For other EtOH recycling 3 MJ/kg is required.

The basic data for the equipment required for biomass supply, cellulose extraction, and aerogel preparation were taken from Pushpendra et al. [49] and, where necessary, adapted to the input quantities used in this study. The quantities of biomass processed on a lab scale range from 0.5 g (OrganoCat) to 50 g (alkali hydrolysis). On an industrial scale, this

was scaled up to approximately 3000 kg.

3.3.2. Biomass supply

The LCIs of the supply of hemp fiber, wheat straw, beech wood chips, and miscanthus are presented in the [Supplementary Material](#) (Section S 2.1, Table S 5 - Table S 8). As the cultivation of hemp, wheat and beech wood delivers valuable residuals (hemp fibers, wheat straw, beech wood chips), allocation between the actual biomass and the residuals is necessary. The allocation procedures are also described in detail in the [Supplementary Material](#) (Section S 2.1). Since no co-product is obtained from the cultivation of miscanthus and the entire plant can be used for cellulose extraction, no allocation is necessary. After cultivation, harvesting and chipping if needed, the biomass is transported (Table S 9 – Table S 12), dried (Table S 13), and milled (Table S 14) to a particle size of approx. 180 μm. The LCIs for individual transport, drying, and milling processes are listed in the [Supplementary Material](#) (Section S 2.2–2.4).

3.3.3. Cellulose extraction

As described in Section 2.1, two possible variants of cellulose extraction are investigated: the OrganoCat process and alkali hydrolysis. Since the OrganoCat process produces three usable outputs (cellulose, hemicellulose sugars, lignin), economic allocation is carried out. [Table 5](#) presents the calculated allocation factors. The experimentally analyzed biomass compositions and assumed product prices used for allocation are provided in the [Supplementary Material](#) (Section S 3.1, Table S 15).

OrganoCat fractionation requires 2-MTHF as organic phase, which can be produced via co-production of 2-MTHF and 1,4-pentanediol from corn stover [59]. Mass allocation is applied for production of these two

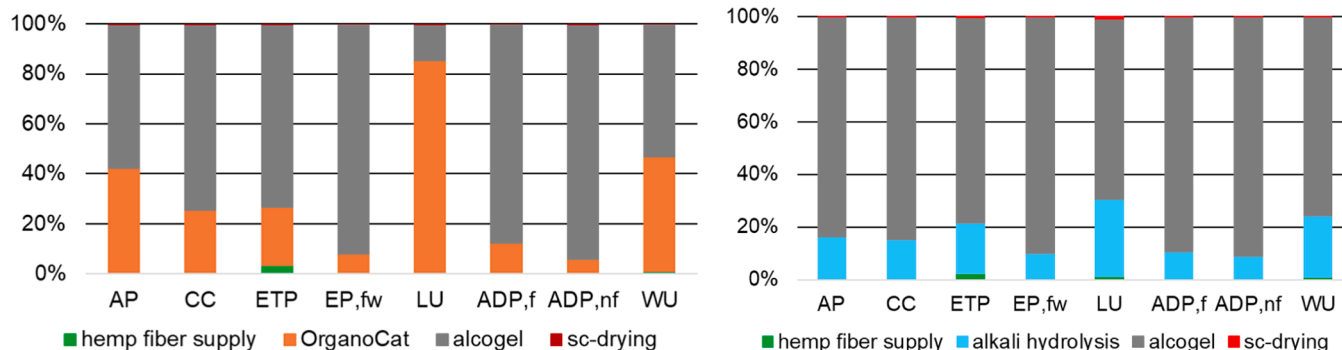


Fig. 7. Contribution analysis of lab scale manufacture of 1 kg aerogel based on hemp fibers using OrganoCat (left diagram) and alkali hydrolysis (right diagram) for cellulose extraction. AP: Acidification, CC: Climate Change – total, ETP: Ecotoxicity, freshwater, EP_{fw}: Eutrophication, freshwater, LU: Land use, ADP_f: Resource use, fossils, ADP_{nf}: Resource use, mineral and metals, WU: Water use.

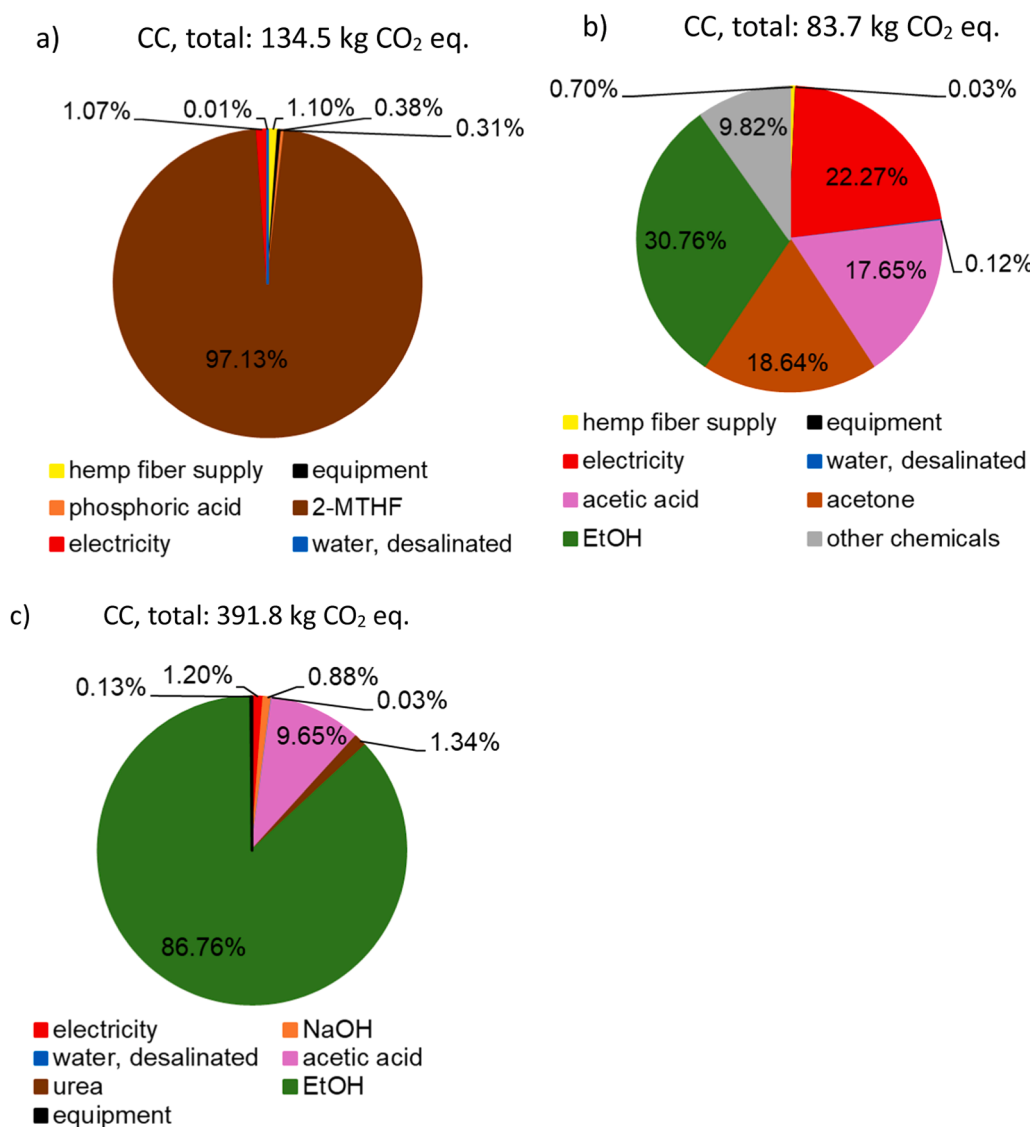


Fig. 8. Contribution analysis of cellulose extraction intermediate step from hemp fiber using (a) OrganoCat and (b) alkali hydrolysis, as well as c) contribution analysis of the alcogel intermediate step on lab scale for the example of the climate change (CC) impact category.

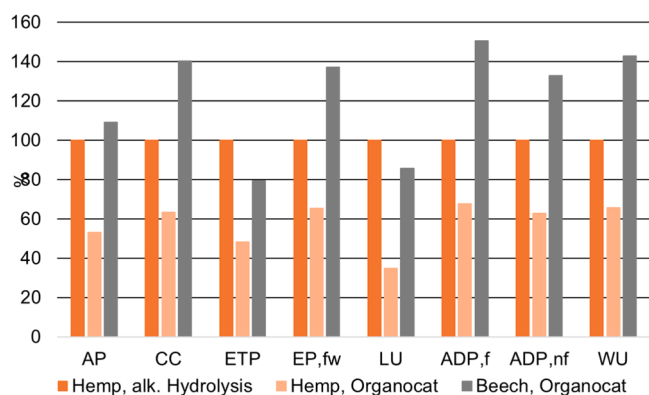


Fig. 9. Relative environmental impacts of industrial scale manufacture of 1 kg aerogel based on different lignocellulosic residues (aerogel from hemp fibers using alkali hydrolysis = 100%).

products. Additionally, a base case without process optimization and a best case with heat integration and utilization of lignin and humins for

heat and power generation are considered for 2-MTHF supply (Table S 16).

The detailed LCI for OrganoCat and alkali hydrolysis at lab scale (Table S 17, Table S 19) and industrial scale/scenario 2050 (Table S 18, Table S 20) are described in the [Supplementary Material](#), Section S 3.1.

3.3.4. Preparation of cellulose solution, wet-gel beads and alcogel beads

The LCI is based on experimental data presented in Sections 2.2 and 2.3. It includes the preparation of the cellulose solution as well as the preparation of wet-gel beads and alcogel beads. Table S 21 and Table S 22 in the [Supplementary Material](#), Section S 4, present the total inventories on lab scale and on industrial scale, respectively. At this point, a difference between the biomasses should be noted, which has a significant impact on the LCIA results. Approximately 40% more cellulose is required to produce aerogel from beech wood chips than when using other lignocellulosic materials. The reasons for this are the lower degree of polymerization of cellulose molecules and the lower purity of the extracted cellulose.

3.3.5. Aerogel preparation

As described in Section 2.3, scCO₂ drying is carried out by placing the alcogels in a 60 L stainless-steel autoclave containing pure CO₂. The

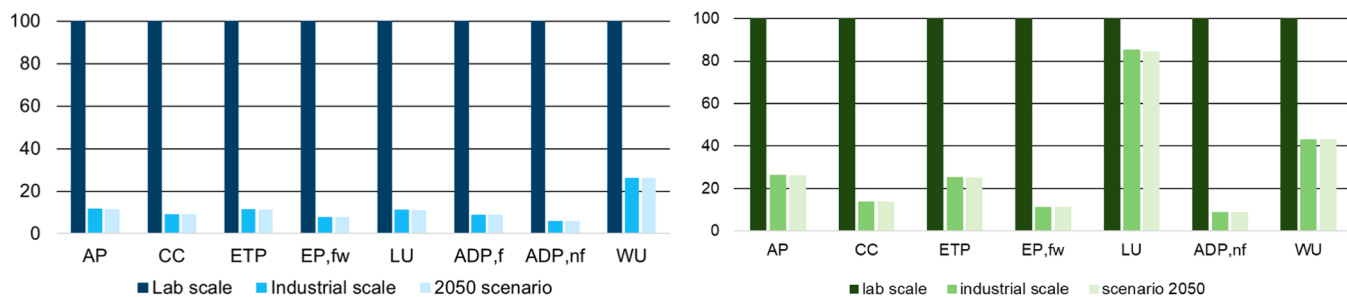


Fig. 10. Relative comparison of environmental impacts of lab scale, industrial scale and 2050 scenario aerogel manufacture based on hemp fiber using OrganoCat process (left diagram) and alkali hydrolysis (right diagram) (lab scale = 100%).

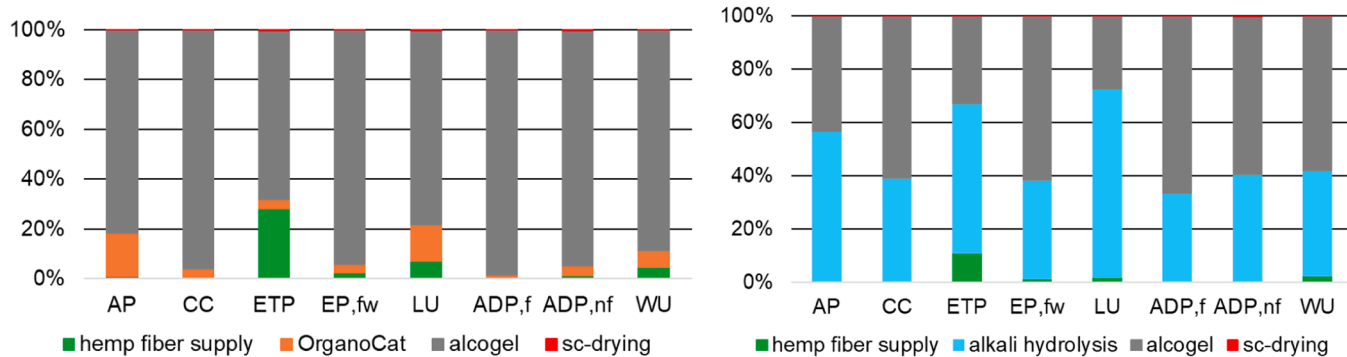


Fig. 11. Contribution analysis of industrial scale manufacture of 1 kg aerogel based on hemp fibers using OrganoCat (left diagram) and alkali hydrolysis (right diagram) for cellulose extraction.

detailed calculations of the steel amount for the autoclave can be found in the [Supplementary Material](#), Section S 5. The 60 L autoclave is filled with a maximum of 45 L alcogel beads (density 0.84 g/cm^3) and 1.8 L of ethanol. The energy requirement of the autoclave on lab scale is 6 kW, and the drying time is 4 h. The total energy requirement of 24 kWh per batch already includes the energy required to supply scCO_2 . Table S 23 and Table S 24 present the detailed LCIs of the aerogel beads preparation on lab scale and on industrial scale, respectively.

3.4. Benchmarks

Bio-based aerogels account for < 1% of the total aerogel market. Niche applications are primarily found in the areas of building insulation (especially for renovation of listed buildings), aerospace, special filters, pharmaceutical and medical applications (controlled release of active ingredients), and the food industry (nutrient carriers). We selected two applications for benchmarking: (i) filter material and (ii) an insulation board. The identified benchmark products are activated carbon, expanded clay, lava granulates, and zeolite powder to compare the filter effect. Some well-known insulation materials, such as polyurethane (PU), polystyrene (PS), and stone wool, serve as benchmarks for thermal insulation. For a fair comparison, the products must fulfil the same function. Therefore, it is necessary to calculate the quantities of material required to fulfill the function. Properties such as specific surface area for filter materials as well as thermal conductivity and density for insulation panels are used to define the FU. For example, 0.893 kg of aerogel is compared with less or more material to achieve the same filtering effect based on different specific surface areas (Table 6). In order to achieve the same thermal insulation for a 1 m^2 panel, different materials need different thicknesses due to their different thermal conductivities (Table 6). To calculate the quantities required for the panels, specific densities of the materials are also needed (Table 6).

The benchmark comparison is based on aerogel made from hemp fibers using alkali hydrolysis due to its more favorable properties in

terms of specific surface area and density compared to aerogel made from beech wood chips.

4. Results of Life Cycle Impact Assessment (LCIA)

4.1. Environmental analysis of aerogel production at laboratory scale

Fig. 6 shows the relative results of three lab scale aerogel productions for different impact categories. All absolute values can be found in the [Supplementary Material](#), Section S 6 (Table S 26 – S 27). The environmental impact of hemp aerogel made from cellulose obtained using OrganoCat is higher than that of aerogel made from cellulose obtained using alkaline hydrolysis. The reason for this is that more cellulose is required for aerogel synthesis if the cellulose has been extracted using the OrganoCat (Table 1). Aerogel made from beech wood chips has a considerably higher impact, as the yield of alcogel achieved is approx. 50% lower than of alcogel made from hemp fibers. The high land use impact of aerogels produced using the OrganoCat process is due to the land used for corn cultivation, which is required for 2-MTHF production.

Fig. 7 shows the contribution of individual steps during lab scale manufacturing on the example of hemp fibers. The alcogel intermediate step is the main contributor of almost all environmental impacts, followed by the cellulose extraction using OrganoCat or alkali hydrolysis. Biomass supply and scCO_2 drying are negligible. Further contribution analyses of the other aerogels can be found in the [Supplementary Material](#) Section S 6.1 (Table S26 – S27).

A deeper contribution analysis of cellulose extraction using OrganoCat and alkali hydrolysis, as well as the alcogel intermediate step, using the example of CC impact, is shown in Fig. 8. The contribution analyses of the other environmental impacts are shown in the [Supplementary Material](#) Section S 6.1 Figure S 1, S 3, and S 4. The supply of 2-MTHF required for the cellulose extraction using OrganoCat process (orange share of the bars in the left diagram of Fig. 7) dominates nearly exclusively the CC impacts considered for that step. A significant

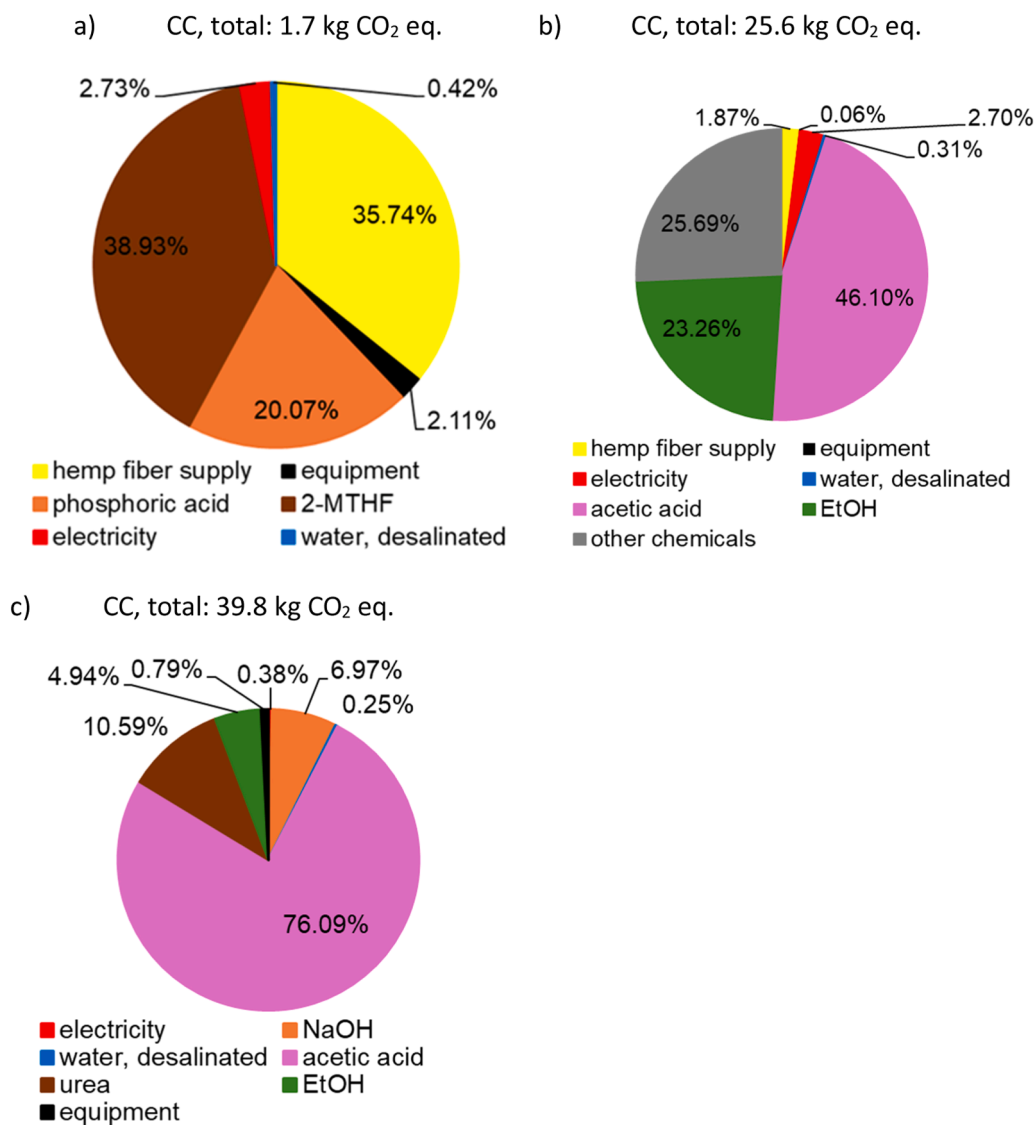


Fig. 12. Contribution analysis of cellulose extraction intermediate step from hemp fiber using (a) OrganoCat and (b) alkali hydrolysis, as well as (c) contribution analysis of the alcogel intermediate step on industrial scale for the example of the climate change impact (CC) category.

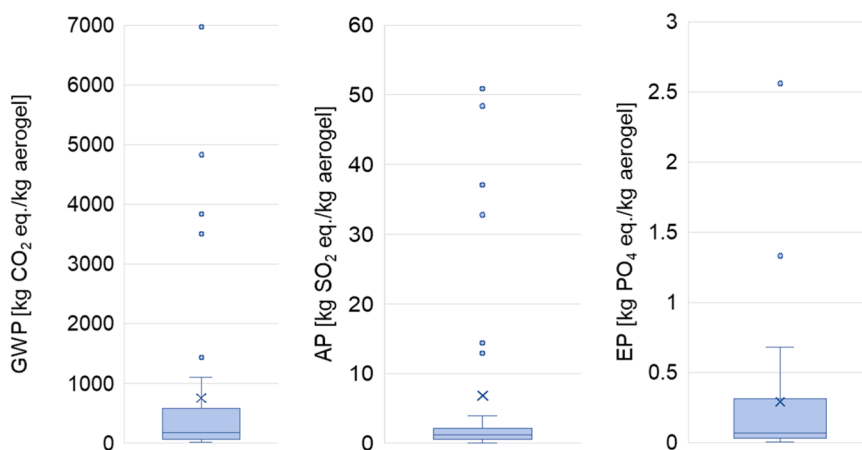


Fig. 13. Environmental impacts of 1 kg of different aerogels, extracted from Kara et al. [33] regardless of LCIA method and process scale.

contribution of EtOH supply, followed by the supply of acetic acid on CC can be seen in Fig. 8b and Fig. 8c. The significant contribution of EtOH

supply for aerogel manufacture has also been addressed in Kara et al. [33] and Antypa et al. [64] for silica aerogels. The latter even quantified

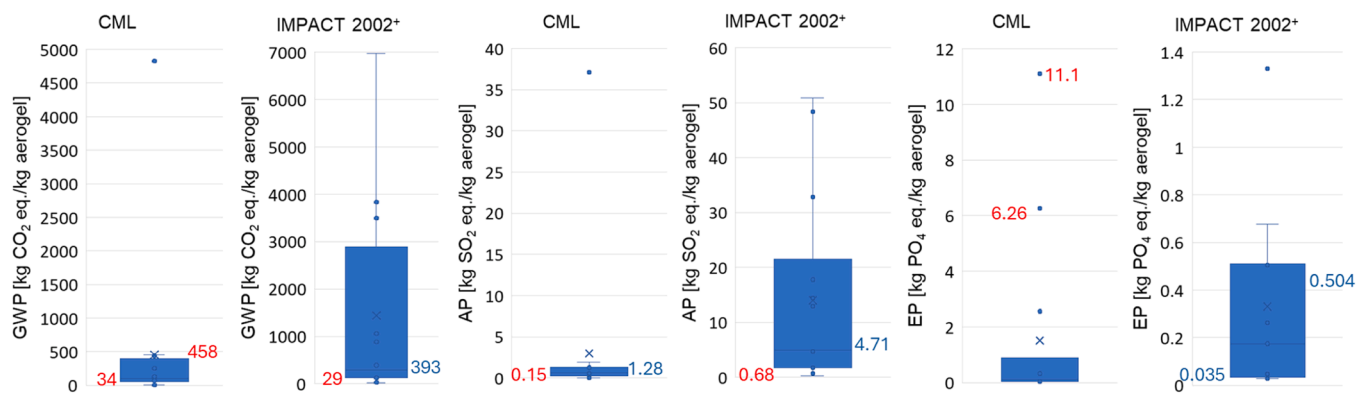


Fig. 14. Environmental impacts of 1 kg of different aerogels depending on the LCIA method (results extracted from Kara et al. [33]); own results based on the example of hemp fiber are shown with data values, whereby the higher values refer to lab scale and the lower values to industrial scale.

the share of EtOH in total CC to approx. 50%, although a recycling rate of 95% was assumed.

The influence of different types of lignocellulosic residues on the overall effects of cellulose extraction with OrganoCat is minor, as shown in the [Supplementary Material](#), Figure S 2.

4.2. Scale-up and 2050 scenario

Improvement potential due to recycling chemicals, energy saving and heat integration are considered when upscaling to an industrial scale. Fig. 9 shows the relative results of industrial scale aerogel production. All absolute values can be found in the [Supplementary Material](#), Section S 6.2 (Table S 27 – S 28). The aerogel synthesized from beech wood chips performs significantly worse in most impact categories, as was already the case at lab scale for the same reasons. In contrast to lab scale, the aerogels perform significantly better in terms of their environmental impact when the OrganoCat process is used for cellulose extraction. The reason for this is that 99% 2-MTHF recycling in the case of OrganoCat contributes more to reducing environmental impacts than 20% acids and EtOH savings in the case of alkali hydrolysis. Both paths benefit equally from 98% EtOH recycling during scCO₂ drying.

Fig. 10 shows the relative results of aerogel production on a lab scale, industrial scale, and using the 2050 scenario in comparison. Upscaling from a lab scale to an industrial scale, and to a 2050 scenario according to the parameter presented in Table 4 achieves significant reductions of environmental impacts up to 94%. The main cause is the assumed recycling of EtOH during the scCO₂ drying process of alcogels. In addition, the recycling of 2-MTHF during the OrganoCat process (left diagram) and the recycling of EtOH, acetic acid and other chemicals (right diagram) during alkali hydrolysis are crucial. Again, the OrganoCat process benefits more from upscaling than alkali hydrolysis for the same reasons as can be seen in Fig. 10 and Fig. 11 (left). The differences between the industrial scale and the 2050 scenario are marginal, even though some significant improvements were assumed for the 2050 scenario (Table 4). For example, transport is carried out using trucks powered by bio-based MeOH, steel for equipment is produced purely electric using renewable energy, heat is supplied using wood, hydrogen is provided using renewable-powered water electrolysis, and the electricity mix is even more environmentally friendly. However, since these are all background processes, they have little impact on overall performance.

The dominance of the alcogel intermediate step in the OrganoCat line becomes even more pronounced on an industrial scale compared to lab scale (Fig. 11, left diagram). The large share of the OrganoCat process in LU, which can be seen on a lab scale (Fig. 7, left diagram), has disappeared. The reason for this is recycling of 2-MTHF, as significantly less corn is required for 2-MTHF production. The opposite can be observed for industrial scale aerogel synthesis using alkali hydrolysis

(Fig. 11, right diagram). Here, the relative share of alkali hydrolysis increases compared to the lab scale (Fig. 7, right diagram), as the reducing effect induced by recycling solvents and chemicals is smaller than that caused by 2-MTHF recycling in the OrganoCat process. After upscaling, biomass supply and scCO₂ drying remain negligible. A share of the drying process of approx. 28% on the total CC as described in Antypa et al. [64] cannot be confirmed here. Since the LCI data from Antypa et al. [64] has not been published for reasons of confidentiality, the cause for the difference cannot be clarified.

The contribution analysis of the intermediate steps cellulose extraction (orange and blue shares of the bars in Fig. 11) and alcogel synthesis (gray share of the bars in Fig. 11) at an industrial scale (Fig. 12) shows a significant shift in the relative shares compared to lab scale (Fig. 8). The contribution of 2-MTHF and EtOH is no longer as significant as it is on a lab scale due to the assumed recycling. In contrast, the relative contribution of acetic acid is increasing. In the case of alkali hydrolysis, the share of acetone is omitted, as it is not used on an industrial scale for cost reasons. The contribution analyses of the other environmental impacts are shown in the [Supplementary Material](#) Section S 6.2, Figure S 5 – S 7.

Identifying environmental hotspots is a crucial task in LCA. Analysis of both the lab scale and industrial scale has shown a large proportion of chemicals such as EtOH, 2-MTHF, and acetic acid in all impact categories. At the same time, the analysis of the industrial scale/scenario 2050 highlighted the key role of recycling chemicals and solvents. While the substances mentioned dominate aerogel manufacturing, the supply of biomass, electricity, and desalinated water, as well as transportation and other substances such as caustic soda, urea, and potassium hydroxide, had hardly any impact on aerogel manufacturing.

4.3. Comparison with other studies

In order to evaluate the environmental performance of the aerogel production process developed in the GelSus project, it will be compared with other LCA studies on aerogels. A recent review of Kara et al. [33] has already done some preliminary work in this context. The authors investigated the sustainability of aerogel production by analyzing the environmental impact of 20 LCAs and two EPDs. They compared 40 CC values, 33 AP values, and 24 EP values for a wide variety of aerogels. However, they did not distinguish between lab, pilot, and industrial scale, nor between the different LCIA methods. Box plot diagrams show the wide range of the analyzed data and their distribution (Fig. 13). Most of the CC values range between approx. 60 and 600 kg CO₂ eq./kg aerogel, with a median of 170 kg CO₂ eq./kg aerogel; most of the AP values range between 0.55 and 2.16 kg SO₂ eq./kg aerogel, with a median of 1.22 kg SO₂ eq./kg aerogel; most of the EP values range between 0.03 and 0.31 kg PO₄ eq./kg aerogel, with a median of 0.06 kg PO₄ eq./kg aerogel. In general, silica-based and algae/alginate-based

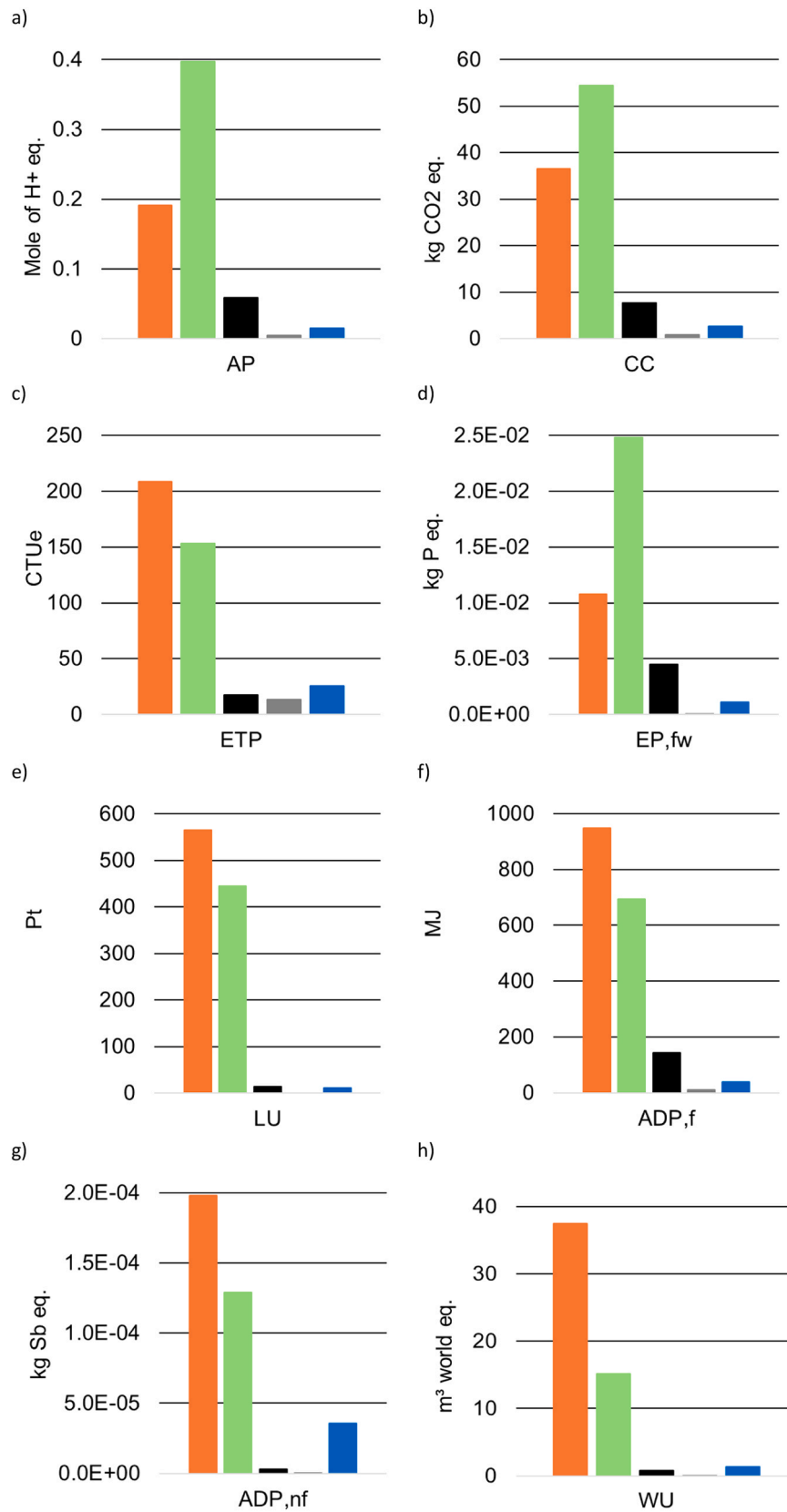


Fig. 15. LCIA results (a-h) for hemp-based aerogel (OrganoCat, scenario 20250) and selected reference products to compare the filtering effect. ■ 0.893 kg GelSus aerogel, ■ 214 kg expanded clay, ■ 1.0 kg activated carbon, ■ 214 kg lava granulate, ■ 0.6 kg zeolite powder.

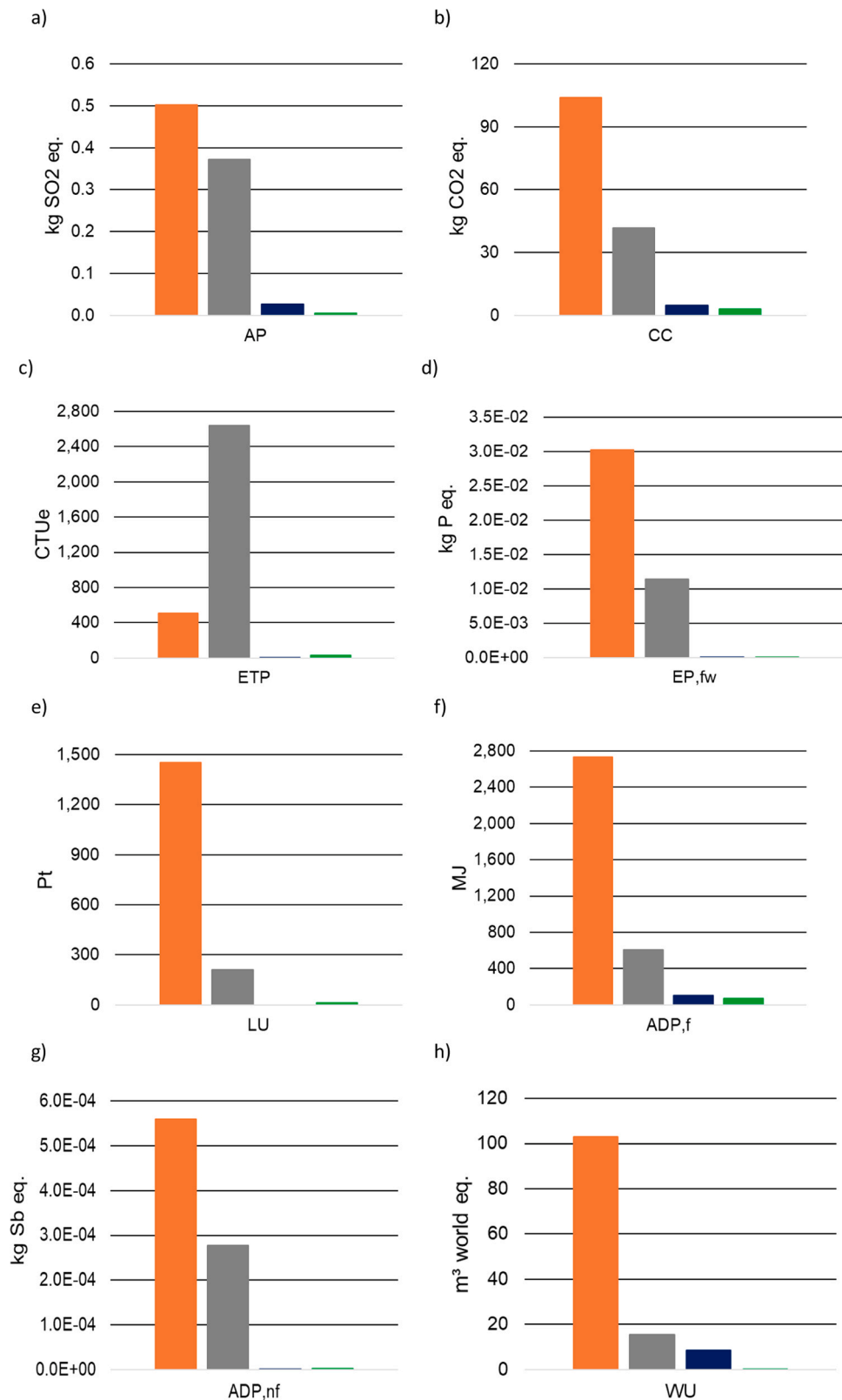


Fig. 16. LCIA results (a – h) for hemp-based aerogel (OrganoCat, scenario 2050) and selected reference products to compare the isolation effect. ■ 2.6 kg GelSus aerogel, ■ 36 kg stone wool, ■ 1.05 kg PS, ■ 0.875 kg PU.

aerogels perform significantly better than organic-based and bio-based aerogels.

Since the individual LCIA methods use different effect mechanism models and characterization factors and also differ in terms of regional

focus and actuality, only impact results that were obtained using the same LCIA method should be compared with each other. For this reason, we have broken down Fig. 13 into the individual LCIA methods CML, ReCiPe, IMPACT2002⁺, and Tracy 2.1. Since box plots can only provide

meaningful information if there is a sufficient amount of data, the studies used TRACY and ReCiPe are not considered further. Due to the separation into the CML (14–16 values) and IMPACT2002⁺ (11–14 values) methods, the interquartile range narrowed for CML and widened for IMPACT2002⁺ (Fig. 14). This means that the CML results show less variation within the middle 50% of the data set. In contrast, the IMPACT2002⁺ results are less consistent and differ more significantly from the median. For comparison, the results of the GelSus hemp fiber-based aerogel are also interpreted using CML and IMPACT2002⁺ and integrated into the box plots of Fig. 14. The environmental impact of producing a hemp-based aerogel on lab scale is four times within the box (higher values in blue in Fig. 14) and twice above it (higher values in red). For the upscaled aerogel (lower values in blue and red), only one EP value is within the box, four values are below it, and one is above it. This indicates firstly that GelSus lab scale aerogel manufacturing is consistent with other lab scale manufacturing processes. If the upscaled values are looked at in more detail, most of them are not within the box, but are comparable to industrial scale values of other studies, which are also not within the box. The reason for this is that there are more lab scale values than values for industrial scale, but not enough to separate them according to LCIA method and scale at the same time. It can therefore be stated as a second finding that the upscaled values are also comparable with those of other studies. The impacts of the upscaled GelSus aerogels are higher in four cases and lower in two cases than that of other industrial scale aerogels. All data used for boxplots can be found in the [Supplementary Material](#), Section S 6.3 (Table S 29).

4.4. Benchmark

For benchmarking the GelSus aerogels with other products, the amounts of aerogels were adopted to fulfill the same function for the different applications (see also Section 3.4, Table 6). Although the comparison with other aerogels' LCAs shows comparable performance of the GelSus aerogels at both lab and industrial scale, Fig. 15 and Fig. 16 indicate that the selected conventional benchmark products (Table 6) often perform significantly better. This is even the case despite selecting the most environmentally friendly scenario for 2050 using the OrganoCat approach for benchmarking.

In the case of filter materials, the new aerogel is significantly inferior to activated carbon, lava granulate, and zeolite powder in the most impact categories. Surprisingly, aerogel performs significantly better than expanded clay in AP, EP_{fw}, and in the important CC category.

Also, compared with conventional insulation materials such as stone wool, PS, and PU, the GelSus aerogel in its form and under considered manufacturing conditions is not competitive in terms of its environmental footprint. Only in the case of ETP is stone wool worse than aerogel.

The results of the benchmark are hardly surprising, as both conventional filter materials and insulation materials are produced in a less complex manner than aerogels. PS and PU, as bulk products, benefit significantly from optimized production conditions over many years.

All data and processes used for comparison can be found in Table S 30 and S 31 in the [Supplementary Material](#), Section S 6.4.

5. Conclusion

This study uses LCA methodology to quantify the environmental impacts associated with aerogel manufacturing based on various lignocellulosic residues. Since LCAs for aerogels have only been conducted for about ten years, there are still many gaps in research, and comparative LCA studies are difficult due to the small number of studies available. In addition, there are differences in the choice of FUs, which often cannot be compared due to a lack of information (e.g., density, specific surface area, thermal conductivity). Although (bio-based) aerogels have been known for almost 100 years, they have only gained increasing attention in the last two decades. However, due to the still

limited number of industrial applications and the confidentiality of LCI data required for market competition, data information remains very limited. The environmental impacts associated with aerogels vary widely due to different raw materials, solvents, manufacturing, and drying processes. The results of this study are in line with this range, especially for the lab scale. The upscaling of mass and energy flows was performed using many assumptions. Therefore, the upscaled results are subject to many uncertainties, which can be improved by using simulation-based tools such as Aspen.¹ As mentioned above, aerogels are rarely manufactured on an industrial scale, so these and other LCA studies offer an opportunity to mitigate hotspots in the production chain in order to ensure the environmental performance of these promising materials.

CRedit authorship contribution statement

Kathirvel Ganesan: Writing – review & editing, Supervision, Project administration, Investigation. **Philipp Michael Grande:** Writing – review & editing, Writing – original draft, Investigation. **Barbara Milow:** Supervision, Project administration, Funding acquisition. **Holger Klose:** Supervision, Project administration, Funding acquisition. **Ameya Rege:** Supervision, Project administration, Funding acquisition. **Sepideh Pakpour:** Supervision, Project administration, Funding acquisition. **Petra Zapp:** Writing – review & editing, Project administration, Funding acquisition. **Bruno Goncalves:** Writing – review & editing, Writing – original draft, Visualization, Investigation. **Andrea Schreiber:** Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization.

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. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.supflu.2026.107038](https://doi.org/10.1016/j.supflu.2026.107038).

Data Availability

All data is included in the supplementary material

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¹ Not to be confused with Aspen Aerogels, Inc., which is among the largest manufacturers of aerogel materials

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