

Prospective Life Cycle Assessment of Epitaxial Graphene Production at Different Manufacturing Scales and Maturity

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Summary

Epitaxial growth is a potential production process for the new material graphene, where it is grown on silicon carbide (SiC) wafers at high temperatures. We provide first estimates of the life cycle cumulative energy demand, climate change, terrestrial acidification, and ecotoxicity of this production. For this purpose, we applied prospective life cycle assessment (LCA) for three production scenarios (lab, pilot, and an industrial scenario), which reflect different production scales and technological maturity. The functional unit was one square centimeter of graphene. Results show that the three scenarios have similar impacts, which goes against previous studies that have suggested a decrease with larger production scale and technological maturity. The reason for this result is the dominance of electricity use in the SiC wafer production for all impacts (>99% in the worst case, >76% in the best case). Only when assuming thinner SiC wafers in the industrial scenario is there a reduction in impacts by around a factor of 10. A surface-area-based comparison to the life cycle energy use of graphene produced by chemical vapor deposition showed that epitaxial graphene was considerably more energy intensive—approximately a factor of 1,000. We recommend producers of epitaxial graphene to investigate the feasibility of thinner SiC wafers and use electricity based on wind, solar, or hydropower. The main methodological recommendation from the study is to achieve a temporal robustness of LCA studies of emerging technologies, which includes the consideration of different background systems and differences in production scale and technological maturity.

Introduction

Graphene is a one-carbon-atom-thin material that has been described as a “wonder material” and as a “rising star on the horizon of materials science” (Geim 2009, 1530; Geim and Novoselov 2007, 183). Although commercialization is yet limited, graphene has been suggested for use in many different areas, such as energy production and storage, including batteries and fuel cells (Brownson et al. 2011); transparent electrodes in computer screens (Blake et al. 2008); semiconductors in electronics

(Van Noorden 2006); reinforcement in composite materials (Li and Zhong 2011); and environmental applications, such as water purification (Shen et al. 2015). In order for graphene to be able to fulfill these promises, feasible and environmentally benign production processes are required. According to a patent analysis, there are currently three main production processes being developed toward large-scale production, namely, exfoliation, chemical vapor deposition (CVD), and epitaxial growth (Sivudu and Mahajan 2012).

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In exfoliation processes, graphene sheets can be exfoliated from graphite by ultrasound or thermal treatment. Another type of exfoliation is by oxidation of graphite, during which electrostatic repulsion facilitates separation of the graphene sheets, followed by reduction to remove the oxygen. Exfoliation processes provide graphene sheets suspended in wet solution. Environmental impacts from ultrasound and oxidation processes have been assessed in a life cycle assessment (LCA) study by Arvidsson and colleagues (2014), who concluded that the ultrasound process was environmentally preferable provided that the solvent could be efficiently recovered and reused. Such efficient production by ultrasound may have as low an energy use as 70 megajoules per kilogram (MJ/kg), whereas the oxidation process required at least 800 MJ/kg. Pizza and colleagues (2014) assessed life cycle impacts from a graphene-containing composite material, with graphene produced by thermal treatment. They found graphene produced by this process to be relatively energy intensive—almost 2,000 MJ/kg. The mass-based functional units of these studies are reflections of the form of graphene obtained from exfoliation processes: powders or sheets suspended in liquid. The currently most likely future application for exfoliated graphene is in composite materials (Sivudu and Mahajan 2012).

In CVD, graphene is obtained by depositing carbon-containing gases onto a metal catalyst at high temperature. Contrary to exfoliated graphene, CVD-grown graphene is not obtained as a suspension, but as a thin surface layer. Graphene produced by CVD is primarily suggested for use in transparent electrodes that can be used in displays (Blake et al. 2008). The production of graphene-based electrodes by CVD was assessed by Arvidsson and colleagues (2016), who found that such electrodes could be preferable to the currently used material (indium tin oxide) from a life cycle energy use perspective, given that the input of the raw material, methane, is low. Such low-input CVD of graphene may require as little as 7 kilojoules per square centimeter (kJ/cm^2), whereas indium tin oxide production requires 20 to 70 kJ/cm^2 . In addition, by using graphene instead of indium tin oxide, the scarce material, indium, can be substituted by carbon, which is beneficial from an abiotic resource depletion perspective (Arvidsson et al. 2016).

Similarly to CVD, epitaxial growth provides graphene in the form of a thin surface layer. Graphene produced by epitaxial growth has been suggested as a potential substitute for silicon as semiconductor material in microchips (Van Noorden 2006; Zhou et al. 2007) and as a biosensor, which could be used to indicate cancer and other diseases (Tehrani et al. 2014). Epitaxial graphene is suggested as suitable for these applications because of its high quality and because it can be grown directly on silicon carbide (SiC) semiconductor substrates without any need for transfer (Novoselov et al. 2012; Sivudu and Mahajan 2012; Hertel et al. 2012). To our knowledge, this article represents the first LCA of epitaxial graphene production.

The main challenge when conducting an assessment of this process is that no large-scale production yet exists. This implies the need to use prospective LCA methodology in order to assess future potential environmental impacts of epitaxial

graphene production. In the LCA field, the term prospective was first used to denote what is today referred to as consequential (change-oriented) LCA (Tillman 2000). However, it is now acknowledged that prospective LCA is not related to the distinction between consequential and attributional (accounting) LCA (Hillman and Sandén 2008; Sandén and Karlström 2007; Herrmann et al. 2014). Prospective LCA is used to assess products at an early stage of technological development, but with the aim of encompassing future environmental impacts.

Several researchers have highlighted challenges that arise when conducting prospective LCA studies. Hillman and Sandén (2008) discussed the implications of scale and changes in background systems when assessing emerging technologies. They showed that these aspects could alter the environmental impact of ethanol and rapeseed methyl ester fuel considerably. Hetherington and colleagues (2014) identified several challenges for LCA studies of emerging technologies, including scale of production, data availability, unknown future applications, and challenges for comparability, given that emerging technologies may not be functionally equivalent to those they replace. Although this last challenge relating to comparability also exists in conventional LCA, it becomes more fundamental in prospective LCA (Hetherington et al. 2014). Gavankar and colleagues (2014) highlighted the importance of considering production scale and technological maturity when assessing emerging technologies. They suggested manufacturing readiness level (MRL) as an indicator of the level of technological maturity. MRL is indicated on an ordinal scale from 1 (research identified for material properties, manufacturing, cost implications, and investments) to 10 (large-scale production) (US DoD 2015). Wender and colleagues (2014) criticized point-value estimations of environmental impacts for emerging technologies considering the inherent data uncertainty and instead recommended uncertainty analysis strategies. In their prospective LCA study of graphene produced by ultrasonication and chemical reduction, Arvidsson and colleagues (2014) further highlighted the influence of background system life cycle impacts, which can vary considerably. Some of these aspects, generic to LCA studies of immature products in general, are also mentioned in some reviews of LCA studies of nanomaterials (Gavankar et al. 2012; Arvidsson 2015).

In this article, the main recommendations from these studies will be considered in order to conduct a prospective LCA of epitaxial graphene production. These recommendations are to consider: (1) parameter uncertainties and avoid point-value estimations (Wender et al. 2014; Arvidsson 2015); (2) the increased efficiency of production scale and technological maturity (Gavankar et al. 2014; Hillman and Sandén 2008; Hetherington et al. 2014; Gavankar et al. 2012); and (3) different background systems, such as electricity production (Hillman and Sandén 2008; Arvidsson et al. 2014). Accordingly, the article aims to assess environmental impacts from epitaxial growth of graphene in a way that (1) includes a thorough sensitivity analysis of parameter uncertainties, (2) takes into account production scale and technological maturity, and (3) includes a thorough investigation of the impacts of different background systems.

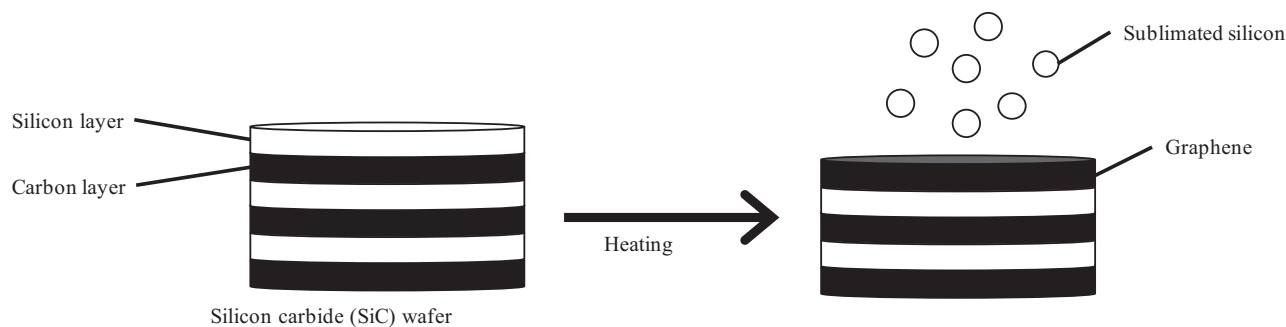


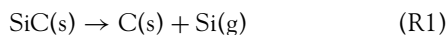
Figure 1 Schematic representation of the epitaxial graphene growth process.

This is performed by using three production scenarios for epitaxial graphene production (lab, pilot, and industrial) as well as best and worst cases for parameter values and background systems.

Method

System Description and Functional Unit

Epitaxial growth of graphene is the foreground system of this study. As described in a number of studies (de Heer et al. 2011; Virojanadara et al. 2008; Emtsev et al. 2009), epitaxial growth of graphene takes place in a reactor where cylindrical SiC wafers are placed. SiC wafers consist of one-atom-thick alternating layers of silicon and carbide. The epitaxial process occurs under a high temperature that makes the silicon sublimate, and follows this principal reaction:



The solid carbon in R1, which is left after the silicon sublimation, forms carbon-carbon bonds and becomes graphene. The entire topside of the SiC wafer becomes covered by one or perhaps a few graphene layers. Figure 1 illustrates the epitaxial growth process. The formed graphene layer prevents further silicon sublimation, and the reaction is thus self-limiting (de Heer et al. 2007). Given that the output of the epitaxial growth is a surface area of graphene, the functional unit of the study is 1 cm² of epitaxial graphene.

In preference, the epitaxial growth process is run in batches. With a continuous process, convection at the SiC-graphene surface could occur, leading to increased silicon sublimation rates. High silicon sublimation rates have been shown to adversely affect graphene quality (Yakimova et al. 2013; Virojanadara et al. 2008; Emtsev et al. 2009). There are three possible inputs to the epitaxial growth process: the SiC wafer, electricity for heating, and argon gas (figure 2). The SiC wafer is the source of carbon, heat is required for the silicon to sublimate, and the argon can be applied in order to create a counter pressure that reduces silicon sublimation rates and thereby ensures higher graphene quality.

Scenario Analysis

Three different foreground system scenarios have been considered. These represent different production scales and levels of technological maturity for the epitaxial growth process. The

first scenario is the lab scenario, which represents current lab production. The second scenario is the pilot scenario, which represents current small-scale production. The third scenario is the industrial scenario, where assumptions about future process settings are used in order to obtain an estimate of an efficient large-scale production. Process settings for the three production scenarios are shown in table 1 and are further described in detail in sections titled *Lab Scenario*, *Pilot Scenario*, and *Industrial Scenario*.

Rather than providing point-value estimations, we use best- and worst-case estimates for the three production scenarios. This is motivated by the current immature state of epitaxial graphene technology and the associated inherent uncertainties regarding its future production system. To ensure transparency (Pauliuk et al. 2015), we provide equations that show how all inputs to the epitaxial graphene growth were calculated and sources for all input data, thus enabling the reproduction and future updating of our results.

Impact Categories

It was known beforehand that high temperatures are required in epitaxial growth, and that SiC wafers are energy-intensive materials. Therefore, three impact categories related to energy use and impacts from energy use were chosen. The first was the cumulative energy demand (CED), as described by Hischer and colleagues (2010), and reported in MJ. This indicator includes all types of energy, both renewable and nonrenewable, as they are extracted from nature. The second was climate change as described in the ReCiPe impact assessment method (Goedkoop et al. 2013). Climate change was estimated in kilograms (kg) carbon dioxide (CO₂) equivalents (eq). The third chosen was terrestrial acidification, again according to the ReCiPe impact assessment method (Goedkoop et al. 2013), and reported in kg sulfur dioxide (SO₂) eq. In addition, we include eco-toxicity calculated by two different methods. The first method is the freshwater eco-toxicity included in the ReCiPe impact assessment method (Goedkoop et al. 2013), which is based on the USES-LCA method (Huijbregts et al. 2000). The second is the eco-toxicity included in the USEtox method (version 1) (Rosenbaum et al. 2008; Hauschild et al. 2008). The impact is estimated in kg 1,4-dichlorobenzene (1,4-DCB) and cumulative toxic units for eco-toxicity (CTUe) in these methods,

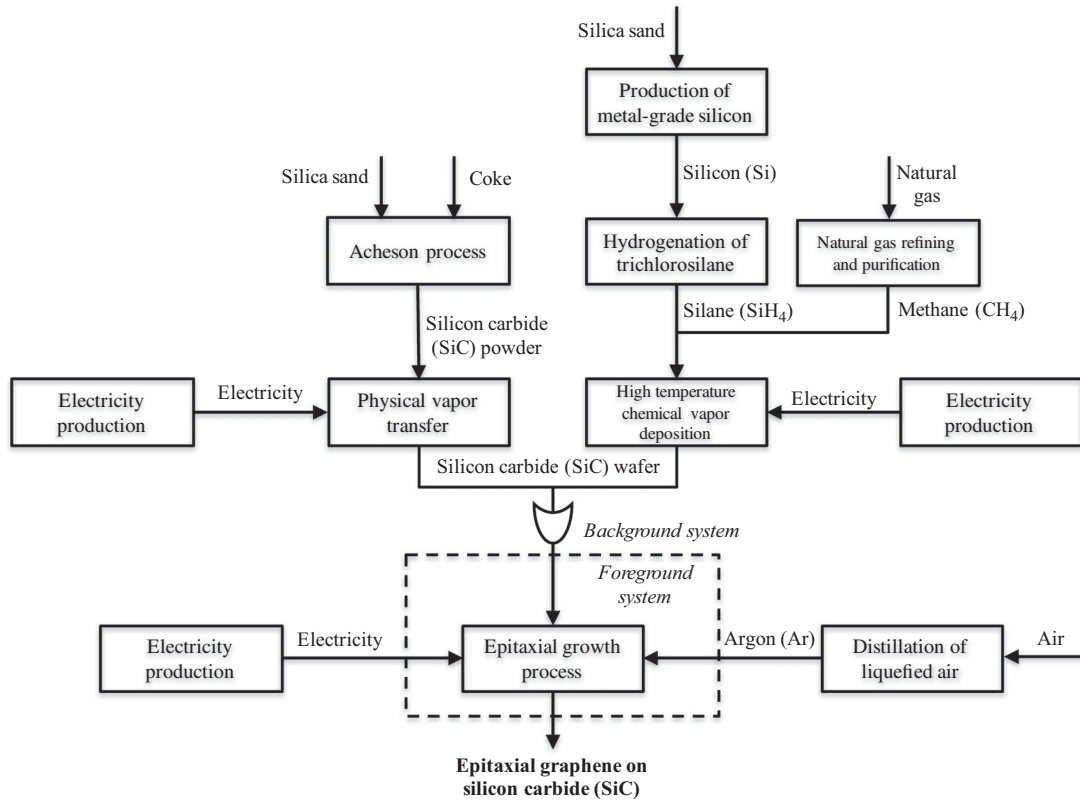


Figure 2 Flow chart describing the main processes of the cradle-to-gate life cycle of epitaxial graphene. The dashed line shows the boundary between the foreground and background systems. Note: An OR gate is used to illustrate that either of the two silicon carbide wafer production routes can be chosen. Further, note that argon is not an input in all scenarios (table 1).

Table 1 Description of the three considered production scenarios for the epitaxial growth process

| Parameter | Symbol | Lab | | Pilot | | Industrial | |
|-----------------------------------|--------|--------|---------|--------|---------|----------------|----------------|
| | | BC | WC | BC | WC | BC | WC |
| Wafers per batch | N | 1 | 1 | 5 | 15 | — | — |
| Wafer diameter (m) | d | 0.1 | 0.2 | 0.1 | 0.2 | — | — |
| Wafer thickness (μm) | h | 350 | 500 | 350 | 500 | 50 | 500 |
| Reactor volume (m^3) | V | 0.0005 | 0.002 | 0.005 | 0.02 | — | — |
| Reaction temperature (K) | T | 1,870 | 1,270 | 2,270 | 1,670 | 1,670 | 2,270 |
| Power requirement (kW) | P | 1 | 3 | 5 | 10 | — ^a | — ^b |
| Reaction time (min) | t | 10 | 20 | 10 | 20 | — | — |
| Reactor pressure (Pa) | p | 80,000 | 100,000 | 80,000 | 100,000 | — | — |
| Argon gas included? | — | No | Yes | No | Yes | No | No |

Note: BC stands for best case and WC for worst case. See main text for references and motivations. The industrial scenario is modeled in a less-specific manner and thus does not require as many input parameters to be specified. Note that the lowest temperature is actually worst case for the lab and pilot scenarios, because that gives the largest amount of argon input according to equation (3).

^aThe industrial-scale best-case power requirement cannot be estimated in terms of kW, but is instead estimated as the thermodynamically lowest possible energy use per square centimeter (cm^2) based on the reaction temperature.

^bThe industrial-scale worst-case power requirement cannot be estimated in terms of kW, but is instead estimated as the highest power requirements per cm^2 from the pilot-scale scenario.

BC = best case; WC = worst case; m = meters; μm = micrometers; m^3 = cubic meters; K = Kelvin; kW = kilowatts; min = minutes; Pa = pascals.

respectively. For climate change, terrestrial acidification, and freshwater eco-toxicity, a so-called hierarchical perspective was chosen in the ReCiPe method. This perspective is based on the currently most common policy principles and uses a 100-year time perspective.

Given that no agricultural land use occurs in the studied life cycle, there would be little relevance of including impact categories such as land use and biodiversity. The same is true for the use of scarce metals. The raw materials, natural gas and coke (figure 2), are energy carriers, so their extracted energy

content is accounted for by the CED indicator. However, silica sand is also an important raw material in epitaxial graphene production (figure 2), and the scarcity of silica sand is of growing concern (Peduzzi 2014). Moreover, there is a lack of consensus on nonenergy abiotic resource impact assessment methods in LCA (Klinglmair et al. 2014; Drielsma et al. 2016) and a lack of characterization factors for silica sand in contemporary resource impact assessment methods. Resource aspects related to the use of silica sand are therefore discussed qualitatively in the *Potential Silica Sand Scarcity* section instead, following the recommendation of the International Reference Life Cycle Data System (ILCD) handbook (EC-JRC 2011).

Inventory Analysis

Lab Scenario

The lab scenario is based on descriptions and data from a number of publications from the Epitaxial Graphene Lab at Georgia Institute of Technology in the United States. In their studies, this pioneering group refers to their process as confined compartment sublimation (de Heer et al. 2011; Ruan et al. 2012; Ruan 2012; Li 2008). They typically produce one wafer per batch. The SiC wafer is reported to be bought from Cree Inc., and this company sells SiC wafers with thicknesses between 350 and 500 micrometers (μm) (Cree Inc. 2015). Diameters of wafers sold are 0.15 meters (m) (Cree Inc. 2015), so a range of 0.1 to 0.2 m is used here. Photographs of the equipment in the studies indicate a reactor volume of approximately 0.001 cubic meters (m^3), so a range of 0.0005 to 0.002 m^3 is used in this study. Temperatures between 1,270 and 1,870 Kelvin (K) are reported. Induction heating is used for heat production, and they have used power requirements of 2 kilowatts (kW). A range of 1 to 3 kW is therefore used. Reaction times vary between 10 and 20 minutes, and the reaction takes place at approximately atmospheric pressure (80,000 to 100,000 pascals [Pa]).

The electricity required for heating was calculated as shown by equation (1):

$$E_{el} = \frac{Pt}{N\pi\left(\frac{d}{2}\right)^2} \quad [\text{J}/\text{cm}^2] \quad (1)$$

where E_{el} is the electric energy for heating, P is the power requirement of the induction heating, t is the reaction time, N is the number of wafers per batch, and d is the wafer diameter. The amount of SiC wafer required was calculated as shown by equation (2):

$$m_{\text{SiC}} = \frac{N\pi\left(\frac{d}{2}\right)^2 h \rho}{N\pi\left(\frac{d}{2}\right)^2} \quad [\text{kg}/\text{cm}^2] \quad (2)$$

where h is the thickness of the wafer, and ρ is the density of SiC (3,210 kg/m^3). The amount of argon required is calculated with the aid of the ideal gas law, which works well for inert gases such as argon (equation 3):

$$m_{\text{Ar}} = \left(\frac{MpV}{RT}\right) / \left(N\pi\left(\frac{d}{2}\right)^2\right) \quad [\text{kg}/\text{cm}^2] \quad (3)$$

where m_{Ar} is the mass of argon required, M is the molar mass of argon (0.040 kg/mol), p is the reactor pressure, V is the reactor volume, R is the ideal gas law constant (8.31 joules $[\text{J}]/\text{K}\cdot\text{mol}$), and T is the reactor temperature. In one variant of this production scenario, no argon is used to limit sublimation rates, but instead the sublimation rate of silicon is controlled by a reusable graphite enclosure (de Heer et al. 2011; Ruan 2012). This is considered in the sensitivity analysis of this production scenario by excluding the environmental impact of argon in the best case.

In some early studies from this group, hydrogen was used to smooth the SiC wafer surface before the reaction in order to obtain high-quality graphene. However, very smooth SiC wafers are now commercially available (Hass et al. 2008), so no hydrogen input is assumed in the lab scenario (nor in any other scenario).

There should be no process emissions from the epitaxial growth, neither of graphene nor of other materials (Ali 2015). The process takes place in a confined reactor and the graphene is bound to the SiC wafer. Silicon-containing gases leave the wafer surface during the reaction, but if the reactor is not opened until it regains room temperature, they will return to solid form. This applies for all three production scenarios.

Pilot Scenario

The recently installed production equipment at the company Graphensic in Linköping, Sweden, represents the pilot scenario. Information about this has been obtained from interviews with representatives from Graphensic by telephone and e-mail (Ali 2015; Syväjärvi 2015; Nilsson 2015), one study of theirs (Virojanadara et al. 2008), and from the patent describing their process (Yakimova et al. 2013). The pilot production process produces around 10 wafers per batch, so we use a range of 5 to 15 wafers per batch. The wafers used have a diameter of 0.15 m, so the same range as in the lab scenario is used (0.1 to 0.2 m). The thickness of the SiC wafers used is 500 μm , but a size of 350 μm is considered as a potential best case. The reactor has a volume of approximately 0.01 m^3 , so a range of 0.005 to 0.02 m^3 is used here. The temperature is between 1,670 and 2,270 K, and an induction furnace is used to provide heat at 5 to 10 kW. The reaction time is similar to that of the lab scenario (10 to 20 minutes). Argon is used to limit silicon sublimation rates, and the reactor pressure is reported to be 80,000 to 100,000 Pa (close to atmospheric pressure). However, we also test the exclusion of the argon as a best case as we did for the lab scenario. The amount of electricity, SiC wafer, and argon required are calculated by equations (1), (2), and (3), respectively, in the same manner as for the lab scenario.

Industrial Scenario

The industrial scenario is hypothetical and does not yet exist. It should be seen as a what-if scenario (Börjesson et al. 2006) of a future potential production. With regard to SiC wafer thickness, it is possible to produce layers as thin as

50 μm , although this is not currently the standard wafer sold by producers (Syväjärvi 2015). For the best case, we assume here that increased production of epitaxial graphene would spur the development toward thinner SiC wafers so that a thickness of 50 μm is obtained. Such co-evolution of interdependent technologies is not uncommon (Grübler 1998; Unruh 2000). For the worst case, we assume a thickness of 500 μm , which represents the possibility that no such co-evolution takes place. The same temperature range of 1,670 to 2,270 K as for the pilot scenario is assumed.

The amount of SiC wafer required was calculated by equation (2) in the same manner as for the lab and pilot scenarios. Regarding heating, we assumed that no improvements in electricity use take place with increased technological maturity for the worst case of the industrial scenario and thus applied the highest electricity use of the pilot-scale scenario (30 kJ/cm^2). For the best case, we assumed the lowest possible amount of energy that could be required. This energy is modeled by the heat equation as the heat required for increasing the temperature of the SiC wafer's mass from room temperature to the required reaction temperature (equation 4):

$$E_{\text{heat}} = C_p m_{\text{SiC}} \Delta T \quad [\text{J}/\text{cm}^2] \quad (4)$$

where E_{heat} is the required heat for the temperature change, c_p is the heat capacity of the SiC (750 $\text{J}/\text{kg}\cdot\text{K}$), m_{SiC} is the mass of the SiC wafer required per functional unit (calculated by equation 2), and ΔT is the temperature change. In addition to this energy required to increase the temperature, some energy for sublimating the silicon would be required. However, because only one or a few top layers of silicon are sublimated, this contribution to the energy use is minor and therefore omitted. By this modeling, it is assumed that the electric heating is 100% efficient even for such high temperatures (i.e., no heat losses when converting electricity to heat are assumed). It is further assumed that the reactor will be so well isolated and the reaction time shortened so that energy losses will effectively be none. In effect, this means a >95% electricity reduction per functional unit compared to the lab and pilot scenarios. This is of similar magnitude as some other energy efficiency improvements suggested in the LCA literature. For example, Li and colleagues (2013) assumed that the electricity use of nanofibrillated cellulose production would be reduced by 92% from lab scale to industrial production, and also note that this is actually a lower degree of reduction than some other estimations. Moreover, for carbon nanotube production, a review by Gavankar and colleagues (2014) revealed energy-use reductions of >99.9% when going from MRL 4 to 7 to MRL 10.

Given that it is technically possible to prevent rapid silicon gas sublimation without the continuous addition of materials (de Heer et al. 2011), the argon gas is excluded in the industrial scenario.

Silicon Carbide Wafer Production

SiC wafers are the substrate upon which the epitaxial graphene grows, and their production is part of the background

system. The two main industrial processes currently used to produce high-quality SiC wafers are physical vapor transport (PVT; also called seeded sublimation or modified Lely process) and high-temperature chemical vapor deposition (HTCVD). PVT is the standard process in industry today, although the newer HTCVD has been gaining interest from industry (Wijesundara and Azevedo 2011; Yakimova and Janzén 2000). In several studies describing lab-scale production of graphene, SiC wafers were obtained from Cree Inc. (Berger et al. 2004; Hass et al. 2006; Song 2006; Sprinkle 2010; Ruan 2012), and this company has been reported to use the PVT process (Yakimova and Janzén 2000). The company Norstel AB is providing SiC wafers for the pilot-scale production by Graphensic, and they produce SiC wafers by both of these two processes. Consequently, both of these SiC wafer production processes were tested for the three production scenarios.

Physical Vapor Transport

In standard PVT, a crude SiC source material is vaporized at high temperature and the formed silicon and carbon gases are redeposited onto a high-quality SiC seed material (Wijesundara and Azevedo 2011). Given the right conditions in terms of temperature and pressure, and a high-quality seed crystal, SiC will expand on the addition of the gases. The quality of the expanding crystal will be that of the high-quality seed crystal.

Warren and colleagues (2015) estimated the electricity required for producing a SiC substrate with the PVT process to be 4.0 kilowatt-hours (kWh/cm^2) (15 MJ/cm^2). This applies for a 350- μm wafer thickness and was linearly extrapolated for other thicknesses in this study. In addition to electricity, there are two input materials for the PVT process—argon and the source material—which is typically a low-quality SiC powder (Yakimova and Janzén 2000). Although an excess amount of SiC powder is generally used to create an oversaturated environment, unused powder could be reused. Losses attributed to discarding of poor-quality parts, slicing and polishing of SiC wafers are considered, giving a yield of 0.35 according to Warren and colleagues (2015). Data for production of low-quality SiC powder by the standard Acheson process was obtained from theecoinvent version 2.2 database (ecoinvent 2010) (data post number 321). No data on how much argon is required have been found. We consider it likely that the production of argon has much lower impacts than do the production of electricity and SiC powder. For example, the argon production's CED per kg is only 4% of that of SiC powder production according to theecoinvent version 2.2 database (ecoinvent 2010) (data post numbers 321 and 251, respectively). Because of the lack of data and presumed minor influence, we have therefore excluded the argon in the PVT process. Because no data on process emissions from SiC production by PVT were found, it had to be assumed that such emissions are negligible.

High Temperature Chemical Vapor Deposition

Similar to the PVT process, the HTCVD process includes the growth of a seed crystal (Wijesundara and Azevedo 2011). The two precursors are the gases silane (SiH_4) and hydrocarbon.

The hydrocarbon could be either methane (CH₄), ethane, or propane (Wijesundara and Azevedo 2011), but is here assumed to be:



These gases are also used in excess, but could be recirculated, and the yield is equal to 0.5 attributable to slicing and polishing of the SiC wafer according to Warren and colleagues (2015). The hydrogen gas produced according to R2 is diluted and cannot be used for other purposes and therefore is assumed to be emitted to air. Data on production of methane in the form of natural gas is available in the ecoinvent version 2.2 database (ecoinvent 2010) (data post number 11053). Further electricity requirement for purifying the methane was estimated to require 0.21 to 2.5 MJ of electricity per kg by Arvidsson and colleagues (2016). The two extremes of this range are used in the best and worst cases, respectively. Silane can be produced by hydrogenation of trichlorosilane, and impacts from this process are available in the ecoinvent version 2.2 database (ecoinvent 2010) (data post number 7210). In addition, Warren and colleagues (2015) estimated the electricity use of the HTCVD process itself to be 2.8 kWh/cm² (10 MJ/cm²) for a 350- μm wafer, which was again scaled linearly for other wafer thicknesses. Because no data on process emissions from SiC production by HTCVD were found, it again had to be assumed that such emissions are negligible.

Argon Gas Production

Argon gas production is part of the background system and typically occurs by liquefying air and then separating the gases (nitrogen, oxygen, and argon) by distillation based on their differing boiling points. Impacts from its production are reported in the ecoinvent version 2.2 database (ecoinvent 2010) (data post number 251). Allocation between the gases in air was based on the extent to which the specific gases contributed to the energy requirement of air liquefaction.

Electricity Production

Epitaxial growth, SiC wafer production, and methane purification require electricity, of which production is part of this study's background system. In a prospective LCA study, it is important to not only consider current electricity production, but also potential future electricity production systems, given that large differences in environmental impacts can occur between different systems (Hillman and Sandén 2008). This is especially important because the locations of future production facilities for graphene, SiC wafers, and methane are inherently unknown. In line with the general setup of the study, best and worst cases were considered.

For the three impact categories considered, electricity from wind, solar, and hydropower typically have low impacts, whereas coal power has high impacts (Hertwich et al. 2015). The best case was thus taken to be the electricity mix of Norway, which has more than a 95% share of hydropower. Note

that electricity mixes based on solar and wind power, and combinations of these three power sources, give similar impacts per MJ of electricity. The worst case is taken to be the electricity mix of Poland, which consists of more than 90% coal power. Similarly, data for other electricity mixes primarily based on coal power give similar impacts per MJ of electricity. This includes China, whose mix has around 80% coal power. Data for these electricity mixes were obtained from the ecoinvent version 2.2 database (ecoinvent 2010) (data post numbers 657 and 659, respectively). Notably, the CED of Norwegian electricity is lower than that of the Polish electricity by around a factor of 3, whereas climate change is approximately 90 times lower, terrestrial acidification is approximately 200 times lower, freshwater eco-toxicity based on ReCiPe is approximately 100 times lower, and eco-toxicity based on USEtox is approximately 30 times lower.

Transport

Including impacts from transport in prospective LCA is difficult, given that the location or future production processes and modes of transport are inherently unknown (Arvidsson et al. 2014, 2015, 2016). The mass that could potentially be transported is the combined mass of the SiC wafer, argon, and materials for SiC production (SiC powder, or silane and methane). Again, in this study, best and worst cases were used.

In the best case, transport impacts were assumed to be effectively nonexistent. This would correspond to production within a local industrial cluster or eco-industrial park.

Savy and Burnham (2013) provided average transport distances for product groups in the United States. These range from crushed stone, which is transported approximately 60 kilometers (km), on average, to textiles and leather, which are transported approximately 2,000 km on average. To represent a worst case, the same average transport distance as for textiles and leather was assumed for the combined mass of the above-mentioned input materials. Transport was assumed to be one way by truck. The truck was assumed to be a 2005 fleet average capable of carrying more than 28 metric tonnes, as described in the ecoinvent version 2.2 database (ecoinvent 2010) (data post number 1944). This corresponds to a relatively polluting mode of transport, which is appropriate for a worst case.

Results and Discussions

Impact Category Results and Discussion

The results of this study are presented as best-case and worst-case environmental impacts for each of the production scenarios along a technological scale and maturity axis, forming an environmental impact area suggested to contain a wide range of outcomes. Figure 3 shows the results for CED, climate change, and terrestrial acidification, respectively. The graphs have similar shapes for all four impact categories. Between the lab and pilot scenarios, impacts are of the same order of magnitude. Between the pilot and industrial scenarios, impacts decrease by

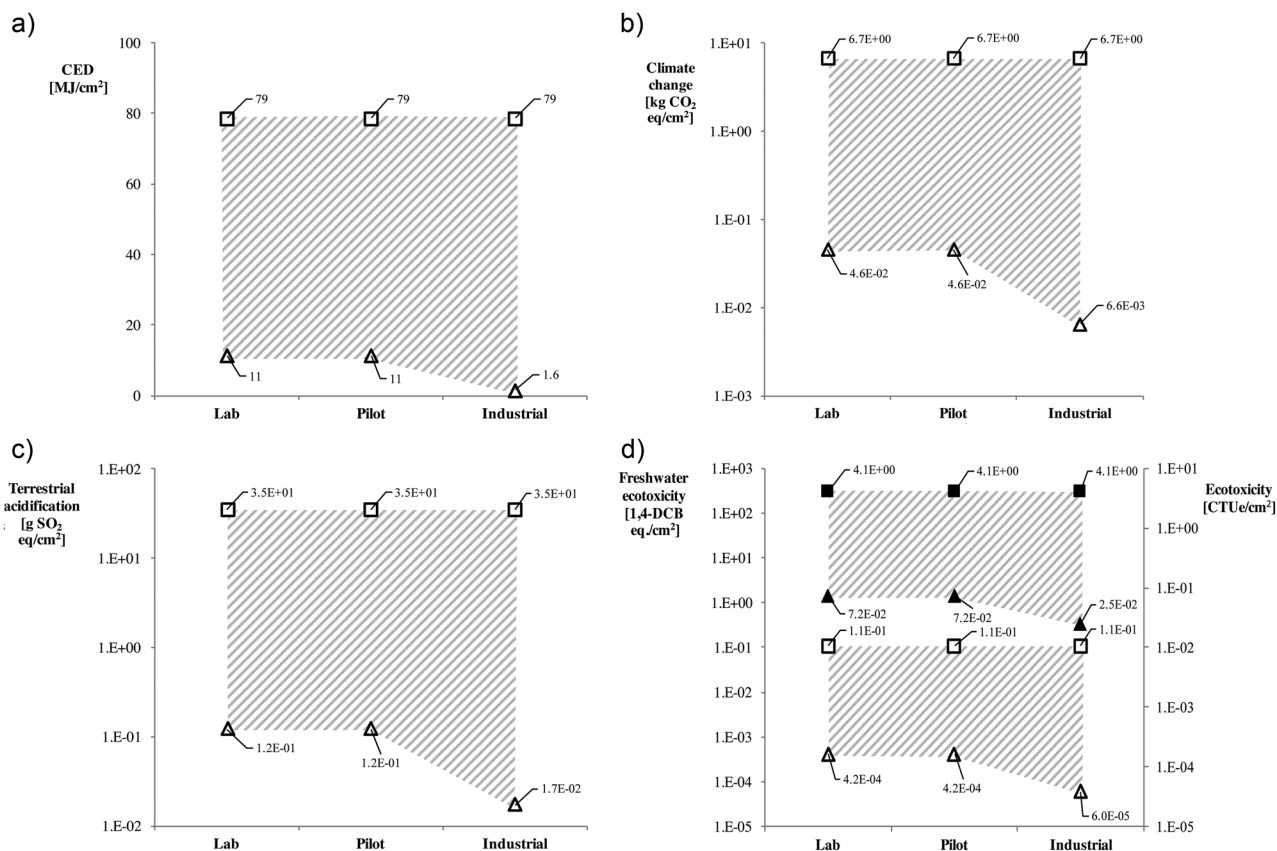


Figure 3 Results for the (a) cumulative energy demand (CED), (b) climate change, (c) terrestrial acidification, and (d) eco-toxicity impact categories. Squares show worst cases, and triangles show best cases. Note the logarithmic scales on (b), (c), and (d). In (d), white markers show results for freshwater eco-toxicity based on ReCiPe (left axis), and black markers show results for eco-toxicity based on USEtox (right axis). 1,4-DCB = 1,4-dichlorobenzene; CTUe = cumulative toxic units for eco-toxicity; g SO₂-eq/cm² = grams sulfur dioxide equivalents per square centimeter; kg CO₂-eq/cm² = kilograms carbon dioxide equivalents per square centimeter; MJ/cm² = megajoules per square centimeter.

approximately 1 order of magnitude for the best case. This is partly in accord with the findings of Gavankar and colleagues (2014), which suggested a clear decrease in environmental impacts as the production scale and technological maturity increased. The reduction in energy use in that study was shown to be several orders of magnitude for carbon nanotubes when going from an MRL corresponding to pilot scale to an MRL corresponding to mass production. In this study, such reductions in impact did not take place between the lab and pilot scenarios, and not between pilot and industrial scenarios for the worst cases. The reason for this behavior is the dominance of one factor in the background system for the environmental impacts: the electricity required to produce the SiC wafer. This accounts for more than 99% of the impacts in the worst case and more than 76% in the best case (see figures S1 to S5 in the supporting information available on the Journal's website). In the best case, silane production accounted for the other <24% of the impacts. Both these two major contributions are related to production of the SiC wafer.

The dominance of SiC wafer production for the impacts explains the behavior of the graphs in figure 3. For the lab and

pilot scenarios, the SiC wafer thickness was 350 μm for the best case and 500 μm for the worst case. The reason for the approximately 10 times lower impacts of the industrial best-case scenario is that the SiC wafer thickness was reduced approximately 10 times, from 350 to 500 to 50 μm .

This illustrates that the relationship outlined by Gavankar and colleagues (2014), suggesting that scaling up production will lead to reductions in impacts, may not apply in all cases. In the case of epitaxial graphene, there is a dominating contribution in the form of the SiC wafer. A scale-up will not lead to any notable reductions in impacts unless this dominating contribution is reduced during scale-up. In cases of products with such dominating contributions, the relationship between technological maturity and environmental impacts may not follow the suggested trajectory.

Given that electricity use during SiC wafer production was dominating, it is interesting to look closer at the original sources for this data provided by Warren and colleagues (2015). For the PVT process, data on process steps were obtained from personal communication with an industry consultant and energy-use data for each step were obtained from the article by Boyd and

colleagues (2009). Data for the HTCVD process were based on lab-scale production equipment described by Elhaddad (2010). Considering that the HTCVD process is still in an early stage of technical development (Wijesundara and Azevedo 2011), it is not surprising that no large-scale production data were available. The data used for the two processes thus seem to have been the best available, although additional data would be valuable for validation, especially for the early-stage HTCVD process. As partial validation of the PVT process' high contribution to the life cycle impacts, it has been described as "energy-intensive" (Warren et al. 2015, 10297) and "requires very high energy for the sublimation of the SiC powder at very high temperatures up to 2600°C" (Elhaddad 2010, 4). The HTCVD process is described as less energy intensive than the PVT process (Elhaddad 2010), which is in accord with the data from Warren and colleagues (2015). However, the HTCVD is typically performed at high temperatures, typically above 1,000°C (Yakimova and Janzén 2000), which suggests a high energy use.

Considering the dominance of electricity for the SiC wafer, the source of the electricity becomes important. The differences between the best and worst cases in figure 3 are primarily results of different SiC wafer thicknesses and the differences between the two electricity mixes.

Unfortunately, Warren and colleagues (2015) only provide data on energy use and input materials, and not on emissions from the SiC wafer production process. The investigation into such emissions is an important area for future research. However, the inclusion of such emissions would only increase the dominance of the SiC wafer production for the environmental impact of epitaxial graphene and would thus not change the main result of this study.

Comparison with Graphene from Chemical Vapor Deposition

As mentioned in the *Introduction*, both epitaxial growth and CVD produce surface layers of graphene (contrary to exfoliated graphene, which is obtained as a liquid suspension). It is therefore possible to compare these two processes on a surface area basis. However, it should be noted that whether the two types of graphene are functionally equivalent for subsequent uses is difficult to say at present. Material properties that could affect the usefulness of graphene in different applications include its electron mobility (measured in square meters [m²] per volt-second), sheet resistance (Ω/\square), thermal conductivity (W/m-K), and sheet surface area (m²). A comparison between CVD-grown and epitaxial graphene by Syväjärvi and Yakimova (2012) suggested that the quality of CVD-grown graphene could be lower, mainly because of imperfections caused during the transfer process from the metal catalyst onto other substrates. Contrarily, in a screening comparison done by Novoselov and colleagues (2012), CVD was reported to give higher-quality graphene than epitaxial growth. Although epitaxial growth is the main production process envisioned for transistors and other integrated circuit components, graphene made by CVD has also been used in laboratory studies for this purpose (Smith et al. 2015;

Nakaharai et al. 2015; Asadi et al. 2015). It thus seems possible that graphene from these two production routes may compete for use in the same applications in the future. We here compare the two routes on a surface area basis, but it should be noted that more-specific functional units may need to be defined in order to make relevant comparisons for specific subsequent applications.

In a recent study by Arvidsson and colleagues (2016), the energy use of graphene produced by CVD was assessed for a modeled continuous large-scale production system. The energy use in that study was calculated into primary energy and included both renewable and nonrenewable energy; thus, it is effectively the same as the CED indicator used in this study. In that production process, graphene was grown at high temperature on a copper substrate. The copper was then etched away, and the graphene layer was transferred to a quartz substrate in order to act as an electrode material. Energy-use results for this CVD process obtained by Arvidsson and colleagues (2016) were 7 to 160 kJ/cm². In figure 3, it can be seen that this is approximately 3 orders of magnitude lower than for the epitaxial graphene, which is in the range of 2 to 80 MJ/cm². This suggests that, when compared on a surface area basis, epitaxial graphene is considerably more energy intensive to produce from a life cycle perspective.

Potential Silica Sand Scarcity

Silica sand is an important raw material for SiC wafer production and thus for epitaxial graphene. High-quality silica sand, sometimes called quartz sand or industrial sand, has a high content of silica and is used for production of silane and silicon (Dolley 2015a; Bloodworth et al. 2009), which, in turn, are used for SiC wafer production (figure 2). In contrast, low-quality silica sand is used to produce cement for construction (Peduzzi 2014; Gibbs 2011). Recent concerns about the scarcity of high-quality silica sand have been raised (Peduzzi 2014; Gibbs 2011; Bloodworth et al. 2009).

At the same time, silicon is one of the most common elements in the earth's crust (Tao et al. 2011; Ljunggren Söderman et al. 2014). Dolley (2015b, 139) writes that "[industrial] sand and gravel resources of the world are large. However, because of their geographic distribution, environmental restrictions, and quality requirements for some uses, extraction of these resources is sometimes uneconomic." Therefore, even if there seem to be short-term concerns related to the use of high-quality silica sand, it is likely that the production of epitaxial graphene can be sustained on a long-term basis.

Conclusions

This study provides further support for two important aspects that have previously been highlighted for prospective LCA. The first is the importance of production scale and technological maturity (Gavankar et al. 2014; Hillman and Sandén 2008; Hetherington et al. 2014; Gavankar et al. 2012). We

have conducted a prospective LCA study of epitaxial graphene production for three production scenarios: lab, pilot, and industrial. Results were only partly in agreement with the previously established relationship of reduced environmental impact with increased production scale and technological maturity. This suggests that the scale-up behavior of emerging technologies may be case specific. The second aspect is the importance of considering different background systems (Hillman and Sandén 2008; Arvidsson et al. 2014). We noted that changes in the electricity production background system had a considerable influence on the impacts of epitaxial graphene production.

These two aspects belong to the more general concept of temporal robustness. For emerging technologies assessed in prospective LCA, there is a possibility that both foreground and background systems will change with time. Although such changes can be difficult to anticipate, the assumption that change will not take place can be as difficult to validate as any other assumption. The main methodological recommendation from this study is thus to strive for temporal robustness when conducting prospective LCA studies. This was also recommended by Nordelöf and colleagues (2014) following a review of LCA studies of hybrid, plug-in hybrid, and battery electric vehicles.

We have also shown that the impacts of epitaxial graphene production are dominated by electricity for production of SiC wafers. Based on this, we recommend that producers of epitaxial graphene investigate the feasibility of thinner SiC wafers, preferably produced by the less energy-intensive HTCVD process. We further recommend them to use electricity from wind, solar, or hydropower.

When compared to graphene made by the CVD process on a surface area basis, it was shown that epitaxial graphene production was more energy intensive by approximately a factor of 1,000. Continued studies of the environmental impacts of these two potential production processes for surface-bound graphene are recommended in order to refine this preliminary comparison, for example, by taking into account material properties relevant for specific subsequent applications.

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Supporting Information

Supporting information is linked to this article on the *JIE* website:

Supporting Information S1: This supporting information presents the contribution of different processes to the cumulative energy demand (figure S1), climate change (figure S2), terrestrial acidification (figure S3), freshwater ecotoxicity (based on ReCiPe; figure S4), and eco-toxicity (based on USEtox; figure S5) of epitaxial graphene production as described in the main article.