Resources, Conservation & Recycling 180 (2022) 106171

Contents lists available at ScienceDirect

Resources, Conservation & Recycling

journal homepage: www.elsevier.com/locate/resconrec

Country-specific carbon footprint and cumulative energy demand of metallurgical grade silicon production for silicon photovoltaics

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ARTICLE INFO

Keywords:
Quartz mines
Life-cycle assessment
Silica sand
Carbon footprint
Cumulative energy demand

ABSTRACT

The photovoltaic (PV) industry requires high-quality silica sand to produce metallurgical-grade silicon (MG-Si) for silicon PV (Si PV). However, high-quality deposits are scarce, and using lower-quality resources may increase the carbon footprint and cumulative energy demand (CED) of Si PV modules. The environmental impact of quartz mining and silica sand extraction for PV has not been updated in over 15 years. It's not representative of current methods used for low-quality sand and is not country-specific. As a result, PV production's environmental impact might be underestimated. We used life-cycle assessment to evaluate the carbon footprint and CED of quartz mining, silica sand extraction, and MG-Si production for high-quality (> 98% silica), industrial-grade (95% silica), and low-quality (65% silica) quartz deposits, which are necessary to calculate Si PV's energy and carbon payback time. The carbon footprint per metric ton of silica sand extraction increased from 22.7 kg CO_{2eq} for high-quality quartz to 47.9 for industrial-grade and 86.7 for low-quality. China currently uses foreign industrial-grade quartz but plans to use low-quality domestic resources, which could increase the carbon footprint of producing one kg MG-Si from 12.1 to 16.5 kg CO_{2eq}. The CED could increase from 188 to 286 MJ. We also investigated illegal mining's environmental impact. The carbon footprint and CED of MG-Si production decreased by 26 to 60% for illegal mines compared to legal mines. Illegal MG-Si production can be cheaper and have a lower carbon footprint, and therefore, third-party-supply-chain verifications are essential to ensure that PV doesn't contribute to the problem.

1. Introduction

Photovoltaics (PV) is a promising energy technology to reduce the carbon footprint of electricity production (Shahsavari and Akbari, 2018). Cumulative PV installations have increased from one GW in 2000 to 480 GW in 2018 and are expected to reach 8519 GW by 2050 (International Renewable Energy Agency (IRENA), 2019). Silicon PV (Si PV) represents 97% of the current PV market and should remain the dominant technology until 2040, but raw material shortage could reduce the market share of Si PV (Masson and Kaizuka, 2020). It is estimated that more than three million metric tons of metallurgical grade silicon (MG-Si) will be required to meet the demand for Si PV manufacturing in the next ten years (Heidari and Anctil, 2021). The raw material for Si PV is quartz which is mined to extract silica sand and purified to produce MG-Si (R. Frischknecht et al., 2020). Further purification is necessary to remove impurities such as Fe, Al, B, and P to produce solar-grade silicon (Ramírez-Márquez et al., 2018) used in photovoltaic cells. The increase in PV installations will increase the demand for MG-Si and, ultimately, the demand for quartz. Evaluating the potential environmental and social impact of mining quartz for PV is necessary to ensure that it remains a sustainable energy source.

Life-cycle assessment (LCA) is widely used to evaluate the carbon footprint and cumulative energy demand (CED) of Si PV. We reviewed 31 Si PV LCA published in the last 20 years, and the quartz purity was not considered in any of them (Supplementary Material (SM) Fig. S1-S4). For all the Si PV LCAs, the same reference process for MG-Si production was used, which is based on sand production in Germany (Bach et al., 1999), which is not a main producer of silica sand used in PV. To model high-quality silica sand extraction rather than sand production, a drying step was added to the process. Based on those assumptions, the carbon footprint is 22.7 kg CO_{2eq} and CED 339 MJ per metric ton of silica sand. The drying process is only a valid assumption for quartz containing more than 98% silica. However, only a few places in the world have quartz deposits with that purity level. The rest of the available quartz resources are industrial (95% purity) or low-quality grade, which need additional steps to remove impurities (Unimin

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https://doi.org/10.1016/j.resconrec.2022.106171
Received 3 August 2021; Received in revised form 6 January 2022; Accepted 11 January 2022
Available online 20 January 2022
0921-3449/

Corporation, 2019). We found two studies that evaluated the environmental impact of silica sand production from industrial-grade quartz for other applications than PV, but none for low-quality quartz mining. The carbon footprint of extracting industrial-grade quartz for glass in Croatia was 43 kg CO_{2eq} per metric ton of silica sand (Grbeš, 2015), but the CED was not reported. In the other study, the natural resource consumption, ecosystem quality, and human health associated with mining industrial-grade quartz for foundry application in Poland was calculated but not the carbon footprint and CED (Mitterpach et al., 2015).

To properly evaluate the environmental impact of PV manufacturing, it is necessary to evaluate the impact of producing quartz for MG-Si in various locations from various quality. This analysis is timely since China is the largest MG-Si producer in the world (*U.S. Geological Survey (USGS)* Mineral Commodity Summaries, 2020), but does not have enough domestic high-quality quartz resources (Zhou and Yang, 2018) and therefore either produce MG-Si from lower-quality resource or import high-quality sands from other countries.

In addition to legal mines, illegal sand mining is an increasing concern globally as quartz is the second most illegally traded product (The Global Rise of Illegal Sand Mining, 2020). Illegal silica sand mining is happening in more than 70 countries (Rege and Lavorgna, 2017). Countries such as China, India, and Singapore with high growth rates need a large amount of silica sand to expand their high-tech industries (Ioannidou et al., 2020). This demand has increased silica sand price, making illegal trades more attractive. For example, the average price of imported silica sand by Singapore has increased from 3 to190 $/metric ton in the last ten years (UNEP, 2013). It is difficult to trace the origin of the raw material, and some silica sand used for PV might be from illegal mines. Therefore, current PV LCA assessments are likely underestimating the carbon and CED from manufacturing PV modules that contain MG-Si from China.

The primary purpose of this study was to evaluate the carbon footprint and CED of producing MG-Si from various locations and purity to meet the increasing global demand for Si PV manufacturing. We compiled the availability and the purity of quartz deposits in the USA (the leading silica sand producer), China (the primary silica sand consumer), and the rest of the world. The main MG-Si producers in China and the rest of the world were identified as well. Second, the amount of silica sand and MG-Si production were compiled for international key players, including the US, China, Australia, Cambodia, Malaysia, North Korea, and Pakistan. Third, we modeled quartz mining, silica sand extraction, and MG-Si production processes for the high-quality (>98% purity), industrial-grade (95% purity), and low-quality (65% purity) deposits to quantify the associated carbon footprint and CED. Global warming potential (GWP) and CED were calculated for various scenarios to estimate the environmental impacts of silica sand production and MG-Si production from various domestic and foreign quartz deposits. This work also identified the location of illegal mines that might be used by MG-Si producers and quantified the carbon footprint and the CED of extracting silica sand and producing MG-Si from illegal quartz.

2. Methods

The first step was to collect data on the location of quartz deposits and purification, the annual silica sand production per country, and the annual import/export amount of silica sand between leading producers and primary consumers. In the second part, we used life-cycle assessment to calculate the carbon footprint and CED for quartz mining, silica sand extraction, and MG-Si production used in Si PV manufacturing.

2.1. Location of legal and illegal mines and MG-Si production

Data about the location, purity, annual production of active legal mines in the USA, China, and other countries was collected from the 2020 U.S. Geological Survey (USGS) report (*USGS Report,* 2020), UN Comtrade, 2020, companies' annual report (Diatreme, 2019; Unimin Corporation, 2019), and literature (Allen and Voiland, 2016; Benson and Wilson, 2015; Bide et al., 2016; Hackney et al., 2020; Hamidullah et al., 1996; JIA et al., 2014; Koehnken and Rintoul, 2018; Lai et al., 2014; Lines et al., 2004; Liu et al., 2019; Lumpur, 2010; ***U.S. International Trade Commission***, 2018, *R. UNEP 2019;* Mitchell, 2012; Piman and Shrestha, 2017; dos Santos et al., 2014; van der Meulen et al., 2009; Vatalis et al., 2015; Zhao et al., 2017; Zhou and Yang, 2018). Data about the location of illegal mines was obtained from articles published in local and international news agencies from 2017 to 2020 (Beiser, 2017, 2015; Berlinger, 2020; Menon, 2018; Meynen, 2017; Salopek, 2019), NGO reports (F. Pearce, 2019), and published papers (Bendixen et al., 2019, 2017; Ioannidou et al., 2020). The annual production and location of MG-Si producers were compiled from 2020 USGS reports (Schnebele, 2020; *USGS*, 2020).

2.2. Environmental impact assessments

2.2.1. Life-cycle assessment (LCA)

The objective of the LCA was to evaluate the carbon footprint and CED of 1 kg of >99% purity MG-Si for Si-PV. The stages within the system boundaries were quartz mining, silica sand extraction, and MG-Si production (Fig. 1). The life cycle inventories for this study were taken from published papers (Table S1), the International Energy Agency (IEA) PVPS Task 12 (R. Frischknecht et al., 2020), Ecoinvent 3.6 Database (Wernet et al., 2016), and DATASMART LCI (US-EI 2.2)(PRe--Sustainability, 2019). Life cycle impact assessment was conducted using SimaPro (PRe'-Sustainability, 2018). ReCipe2016 method was used for global warming potential (GWP) analysis, and Cumulative Energy Demand V1.1 was used for energy assessment. Carbon footprint and CED are the required impact categories from the International Energy Agency Photovoltaics Power Systems Programme (IEA-PVPS) methodologies to conduct LCA of photovoltaics systems (R. Frischknecht et al., 2020). The CED is required to calculate the energy payback time (EPBT) and energy return on investment (EROI) (Raugei et al., 2021), which are used to compare different types of PV modules or other types of energy technologies.

2.2.2. Quartz mining, silica sand extraction, and MG-Si production

The process for silica sand extraction from high-quality (>98%), industrial-grade (95%), and low-quality (65%) quartz deposits was based on existing industrial processes. The Unimin Co process was selected to model the silica sand extraction process since they are the leading global silica sand producer and transform quartz of various purity. We divided the silica sand production into two main steps (Fig. 1) (Unimin Corporation, 2019). The first step (quartz mining) begins with removing the topsoil with loaders and bulldozers, excavation with excavators, transportation with lorry and conveyor belts, sieving, washing, and dewatering. This step is similar for high-quality, industrial-grade, and low-quality deposits. The second step (silica sand extraction) depends on quartz purity and includes beneficiation processes such as magnetic separation, flotation, and gravity separation to remove impurities physically or chemically. There is no need to use the beneficiation process for mines with the highest purity except for drying silica sand before transportation to MG-Si facilities. In contrast, in mines with industrial-grade and lower quality, beneficiation processes are required to remove impurities.

Refining is required to increase the purity of silica sand to produce MG-Si with more than 99% purity. At this stage, purified silica sand is called MG-Si. Further purification is necessary to provide solar grade silicon with 99.99999% purity that can be used for Si PV module manufacturing. We modeled only MG-Si production for this study since the silica sand quality does not affect the rest of the manufacturing processes if the produced MG-Si reaches more than 99% purity. The MG-Si production process was modeled based on IEA PVPS Task 12 and Ecoinvent (Table S1 and Fig. S11).

Input data for the LCA, including required electricity and fuel at each

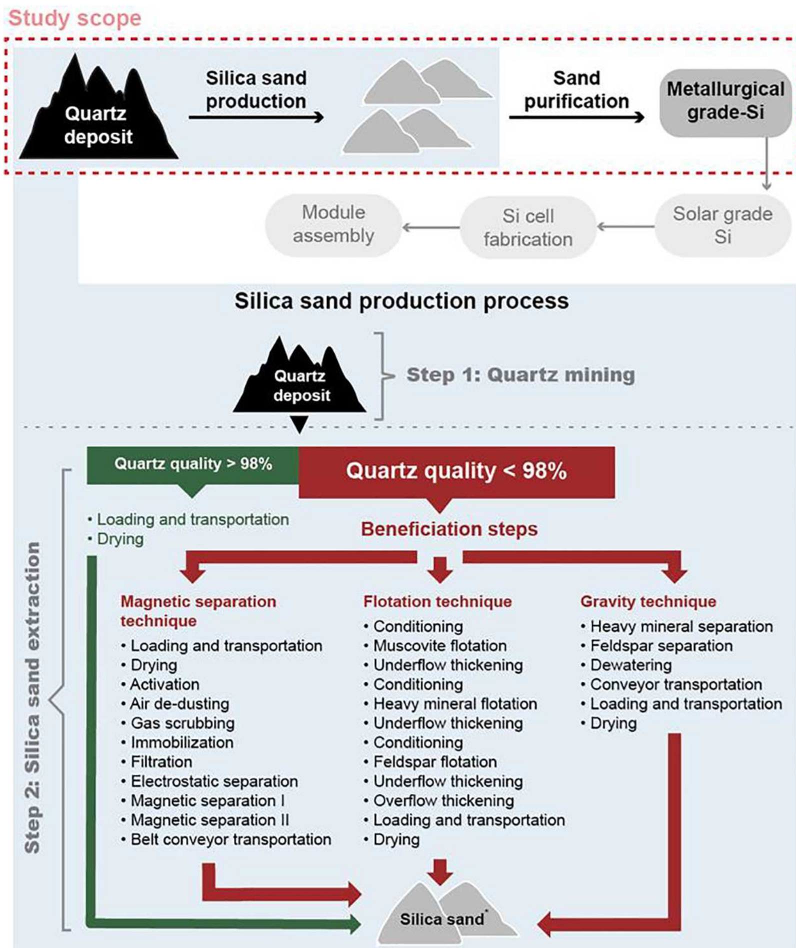


Fig. 1. Study scope including silica sand and MG-Si production for manufacturing Si PV modules. Silica sand production entails two stages. The 1st step (quartz mining) is similar for all types of quartz, and the 2nd step (silica sand extraction) depends on quartz purity. *Silica sand purity depends on initial quartz quality, mining process, and extraction chosen.

stage, were estimated based on existing similar processes in literature (Fig S5-S11 and Table S1). For the USA, we used the national average electricity mix because quartz deposits are distributed all around the country. The regional electricity was selected for China since potential quartz deposits are distributed in some regions, primarily east and southeast. A significant regional difference was reported for electricity in China (Shen et al., 2019). China's electricity was divided into six regions (Northwest, North, Northeast, East, South, and Center) based on energy resources for electricity in earlier research (Fig. S13) (Shen et al., 2019). We selected a representative province for each region based on the highest demand for silica sand. For example, Xinjiang had the highest demand for silica sand and was chosen as a Northwest region representative. Beijing, Liaoning, Fujian, Yunnan, and Sichuan were selected for North, Northeast, East, South, and Center, respectively. The carbon footprint and the CED of silica sand production and MG-Si production in China were calculated based on Eq. (1) and Eq. (2).

$$GWP_t = \sum_{i=1}^{n}\left(EF_i \times \frac{IM_i}{\sum_{i=1}^{n} IM_i}\right) \quad (1)$$

$$CED_t = \sum_{i=1}^{n}\left(ED_i \times \frac{IM_i}{\sum_{i=1}^{n} IM_i}\right) \quad (2)$$

Where t represents the type of product (e.g., silica sand or MG-Si), EF represents the emission factor (kg CO_{2eq}), ED represents the energy demand factor (MJ), IM represents the amount of imported product, i represents the countries, and n represents the number of countries. Emission and energy demand factors were quantified based on the impact category analysis of the target product in SimaPro software.

3. Results and discussion

3.1. Location of legal and illegal quartz mines and MG-Si production

3.1.1. Legal mines

The annual production of industrial-grade silica sand was compiled from 1994 to 2019 (Fig. 2A and S14). The global production in 2019 was 329 million metric tons, with 35% of the production in North America, 46% in Europe, and 12% in the Asia-Pacific region (APAC). The USA was the main industrial-grade silica sand producer, and 73% of its annual production was used for hydraulic fracturing (USGS, 2020). The Netherlands produced 37% and Spain 24% of the European production. India (30%) and Malaysia (25%) were the leading producers in APAC.

Fig. 2B shows the mine locations of the top ten high-quality and industrial-grade silica sand producers in the USA. Most of the US quartz mines are located in Illinois, Ohio, Michigan, Texas, and North Carolina. Unimin Corporation is the largest producer with an annual capacity production of more than 41 million metric tons and 21 active quartz deposits in the USA (Dolley, 2004; Unimin Corporation, 2019; Vatalis et al., 2015). US Silica Inc. is the second-largest silica sand producer in the USA with an annual production of 19 million metric tons and 15 mines (Silica Holdings, 2020; USGS, 2020). A small portion of industrial-grade silica sand production in the US is currently used for MG-Si production, and the majority is used for fracking in natural gas and oil wells. However, due to the expected transition from fossil fuels to

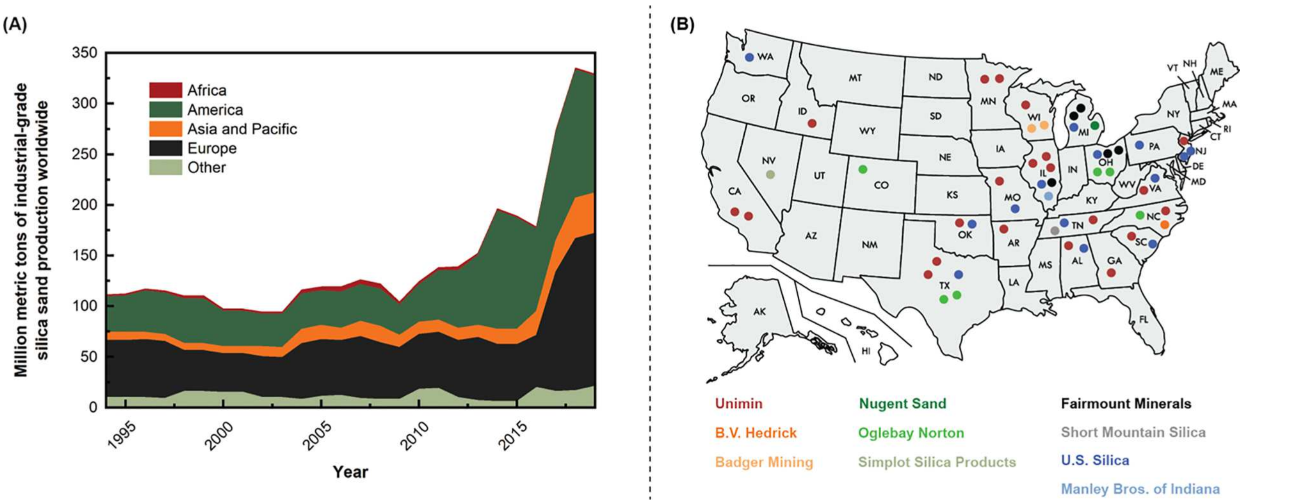


Fig. 2. (A) Annual industrial-grade silica sand production in the world from 1994 to 2019 and (B) The mines distribution of 10 major high-quality and industrial-grade silica sand producers in the USA. Data was compiled from USGS annual reports (*U.S. Geological Survey* (USGS) Mineral Commodity Summaries *2019*, 2020).

renewables in the next couple of years and the expected increasing demand for high-quality sand, the production could shift to MG-Si production.

High-quality silica sand is scarce, and only a few high-quality deposits, such as the Spruce Pine in North Carolina, USA, exist in the world (Müller et al., 2007; Vatalis et al., 2015). The location and amount of global industrial-grade quartz deposits in 2019 were compiled (Fig. 3A). Although typical sand is found everywhere, high-quality and industrial-grade silica sand resources are not equally distributed, and some countries suffer from a high-quality deposits' shortage. China has no domestic high-quality and industrial-grade deposits and is highly dependent on silica sand imports (Zhou and Yang, 2018). The main silica sand suppliers for China MG-Si production were the USA and Vietnam until 2012 and are now Cambodia, Australia, Malaysia, and Pakistan (Fig. 3B). Other countries like India and the United Arab Emirates also do not have enough high-quality resources to meet their growing demand. A portion of this global demand was supplied via documented international trades between producers and consumers, as

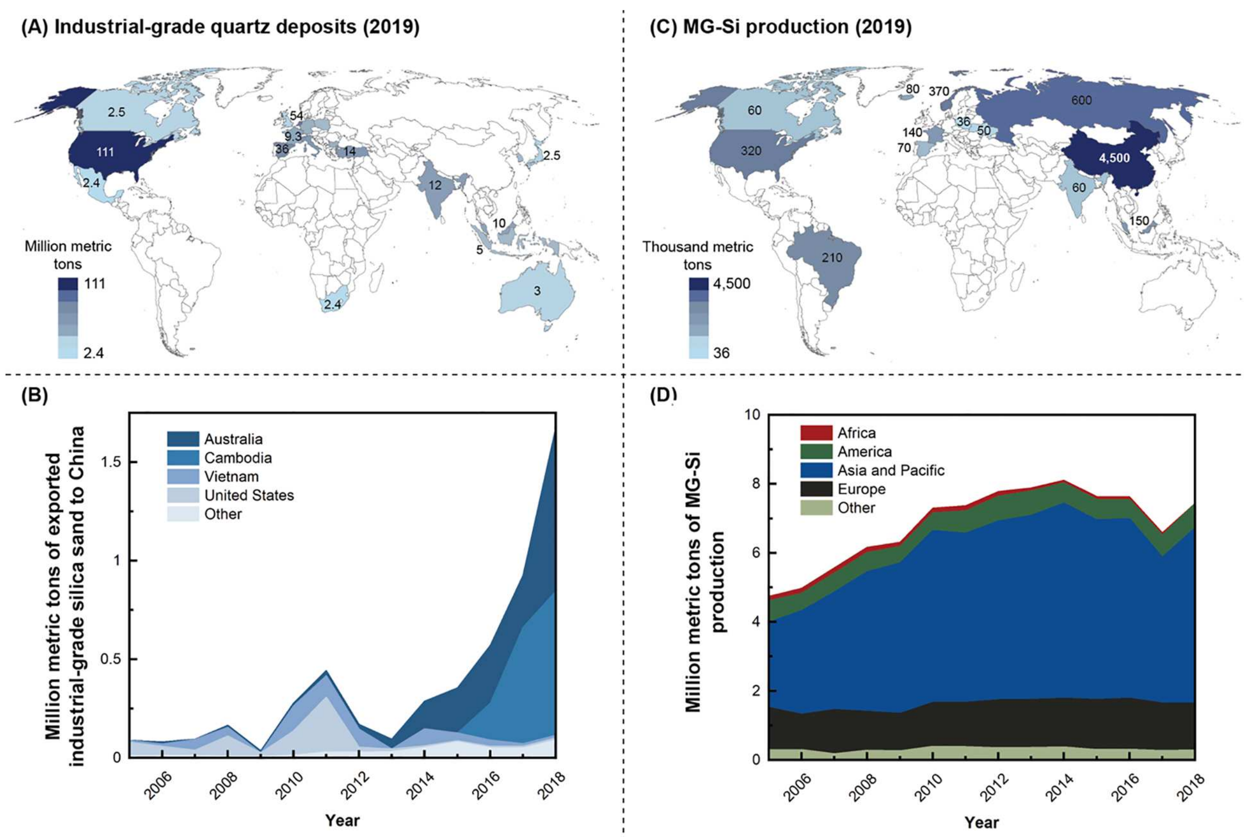


Fig. 3. (A) Country-specific silica sand production from active industrial-grade quartz (95% SiO_2) mines in 2019. (B) Annual industrial-grade silica sand export to China. Data was compiled from the annual UN Comtrade reports (*United Nations Comtrade annual reports; International Trade Statistics Database*). (C) Country-specific MG-Si production in 2019. (D) annual MG-Si production from 2005 to 2019. Data was collected from USGS annual reports for A, C, & D (*USGS* 2020).

reported by the UN. The lack of high purity quartz resources may lead to the use of quartz resources with a lower purity which can consequently increase MG-SI production's carbon footprint.

3.1.2. Illegal mines

A review of news articles and NGO's reports was conducted to identify illegal quartz mines that might be used in the PV industry (Table 1). We found that illegal quartz has been used in various industries, such as construction, glass, and PV. The compiled data shows that illegal quartz might have been mined from illegal mines in Cambodia and North Korea for the PV industry.

Many illegal mining activities were reported in countries close to China. China itself does not have domestic high-quality quartz deposits, and illegal domestic mines are mainly used in the construction industry. To produce one kg of MG-Si (>99% purity), 2.7 kg of industrial-grade quartz (95% purity) is required (R. Frischknecht et al., 2020). In 2019, China needed to import 12.15 million metric tons of industrial-grade silica sand from foreign resources for MG-Si production. However, only 14% of this amount can be accounted through documented trades. Meanwhile, multiple reports from NGOs and news agencies have identified illegal mining in Cambodia and North Korea that supplies silica sand to China. Table 1 and Fig. S16 show the location of illegal mining in North Korea, Cambodia, and the rest of the world (Berlinger, 2020; Hackney et al., 2020; F. Pearce, 2019). Since there is no supply chain tracing for silica, without knowing it, some Chinese MG-Si producers might have used silica from those illegal mines.

3.1.3. MG-Si producers

The location of MG-Si producers (Fig. 3C) and the amount of annual MG-Si production (Fig 3D and S17) were compiled. Since 2005, the global MG-Si production has increased by about 60% and in 2019, China, Russia, Norway, and the USA were the main producers. China produced 64% of the global MG-Si production in 2019, and 85% of it was produced in the Yunnan, Xinjiang, Sichuan, Guizhou, Hunan, and Fujian provinces (Fig 4). Since 2012, China production has increased by almost 500% and is expected to continue to grow to meet their demand for PV cells and module production (International Energy Agency IEA, 2013; Masson and Kaizuka, 2020); however, this can change in the near future due to policy favoring local manufacturing. The USA, the third-largest PV market after China and Europe, implemented restrictions on Si PV modules imports from other countries, which can affect the upstream process (e.g., MG-Si production) of Si PV module manufacturing. For example, under the Section 201 of the US Trade Act of 1974 in 2018, the USA has implemented a safeguard tariff (up to 30%) on Si PV modules made by countries, such as China and Korea, that contribute more than 3% of total PV imports to the USA (Pickerel, 2018; Shum, 2018). The section 201 encourages some Chinese and Korean PV manufacturers to move the manufacturing operation to the USA to avoid the tariff. The other example is the US sanction against China under the section 301 of the US Trade Act of 1974, where a 25% tariff was imposed on Chinese PV modules in 2019. This pushed Chinese PV manufacturers to shift their manufacturing bases to other countries like Malaysia and Thailand (See-Yan, 2019; Masson and Kaizuka, 2020). The US sanction against Chinese materials has encouraged REC Silicon, a major silicon producer, to use investments to restart its US silicon production at Moose Lake in Washington to fortify its position in the US PV market and be considered under the Solar Energy Manufacturing for America (SEMA) Act, a source of support US policymakers consider (Rai-Roche, 2021; Stoker, 2021). On the other hand, China has imposed various tariffs, such as antidumping duties and countervailing duties on imported PV modules from the USA, Korea, and Europe since 2014. European countries were exempt after 2018, which for example, encouraged the European companies, REC Silicon, to establish a joint company with Chinese PV manufacturers in China. The current policies in China, a major Si PV manufacturer, and in the USA, a major consumer, can lead to setting up new PV manufacturing bases in other

Table 1
Summary of illegal silica sand mines and potential usages.

Location of illegal mines	Purity	Location of consumers	Applications	Ref
Bangladesh	NA*	NA	Construction	(Anthony et al., 2015); (Koehnken and Rintoul, 2018)
Cambodia	Industrial grade	China, Malaysia, and Singapore	PV and construction	(Mother Nature Cambodia Inc, 2021); (Anthony et al., 2015); (Koehnken and Rintoul, 2018); (F. Pearce, 2019)
China	NA	China	Glass and construction	(Allen and Voiland, 2016); (Koehnken and Rintoul, 2018); (Sharma and Scarr, 2021)
Colombia	NA	NA	Mining for gold extraction	(Tobella, 2015)
Hungary	NA	Hungary	Construction	(Koehnken and Rintoul, 2018)
India	Low quality and industrial grade	India & Malaysia	PV and construction	(Anthony et al., 2015); (IFEX, 2017); (Rege and Lavorgna, 2017); (Koehnken and Rintoul, 2018); (The Times of India, 2019); (F. Pearce, 2019)
Indonesia	NA	China, Hong Kong, Singapore, Thailand,	Construction	(The New York Times, 2010); (Anthony et al., 2015); (Beiser, 2015); (Koehnken and Rintoul, 2018); (F. Pearce, 2019)
Iran	NA	Iran		(Farahani and Bayazidi, 2018)
Israel	NA	Israel		(Beiser, 2015)
Italy	NA	Italy		(Rege and Lavorgna, 2017)
Jamaica	NA	NA	NA	(Beiser, 2015)
Kenya	NA	NA	Construction	(Ekin, 2017); (Koehnken and Rintoul, 2018)
Malaysia	Industrial grade	Singapore		(Anthony et al., 2015); (Beiser, 2015); (Koehnken and Rintoul, 2018)
Morocco	NA	Morocco		(Beiser, 2015); (Quérouil and de Viguerie, 2015)
Mozambique	NA	NA		(Koehnken and Rintoul, 2018)
Nigeria	NA	NA		(Beiser, 2015)
North Korea	Industrial grade	China	PV and construction	(Berlinger, 2020)
Russia	NA	Russia	NA	(Koehnken and Rintoul, 2018)
Sierra Leon	NA	NA	Construction	(Koehnken and Rintoul, 2018)
South Africa	NA	South Africa		(Koehnken and Rintoul, 2018)
Tanzania	NA	NA		

(continued on next page)

Table 1 (*continued*)

Location of illegal mines	Purity	Location of consumers	Applications	Ref
Thailand	NA	Foreign countries		(Koehnken and Rintoul, 2018) (Rege and Lavorgna, 2017)
Ukraine	Industrial grade	NA	NA	(Koehnken and Rintoul, 2018)
Vietnam	NA	Singapore	Construction	(The New York Times, 2010); (Beiser, 2015); (Koehnken and Rintoul, 2018)

*NA: Not Available.

regions like Europe.

3.2. Environmental impact assessments

3.2.1. Modeling silica sand extraction

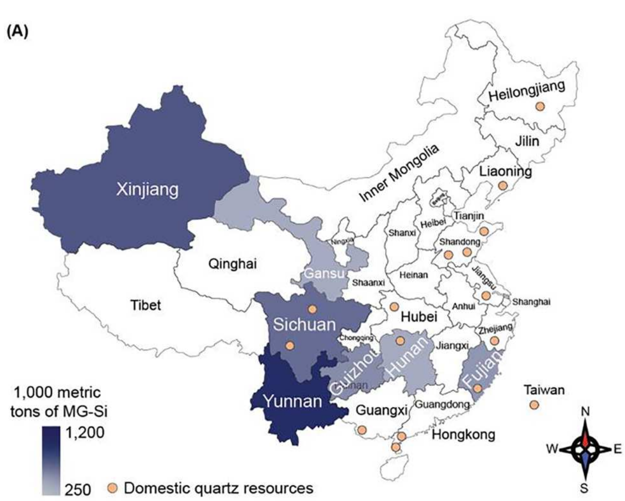
When silica sand is produced from low-quality quartz, extra beneficiation steps are required to remove impurities. The beneficiation processes were selected based on the types and locations of mines. For mines with access to natural water resources like rivers, the gravity method, which requires a large amount of water, is possible, but residual sediments might affect the downstream river ecosystem (Grbeš, 2015). The high purity quartz resources in China are shown in Fig. 4A. They are mostly located in the east, south, and center of China and have an average purity of 65% (JIA et al., 2014; Zhou and Yang, 2018). Fig. 4B shows available foreign silica sand resources for China. We modeled quartz mining and silica sand extraction process for various scenarios to evaluate the carbon footprint of supplying silica sand needed for MG-Si production in China now and in the future. Fig. 4B describes the scenarios considered. The assumptions and modeling inputs for quartz mining and silica sand extraction (e.g., transportation distances, energy types, energy resources in various regions, required energy and materials for equipment used in each step of mining and extraction, etc.) are summarized in Table S1 and Fig. S5-S10.

The GWP and CED of extracting silica sand from high-quality (>98% silica) and industrial-grade quartz (95% silica) were calculated for the USA (Fig. 5A and D). The average GWP of extracting one metric ton of silica sand from high-quality deposits in the USA was 22.7 kg of CO_{2eq}, while it was more than double (47.9 kg of CO_{2eq}) for industrial-grade deposits due to the beneficiation processes needed to remove impurities from industrial-grade quartz. In the only paper we found on industrial-grade silica sand production, which was modeled for glass production in Croatia, the GWP was 43 kg CO_{2eq} for the required silica sand needed for glass production in Croatia (Grbeš, 2015). The difference is due to calculating the carbon footprint based on various silica sand production procedures and the source of electricity used in Croatia and the US. We modeled the separation stages, such as magnetic and flotation, which are necessary to remove impurities from industrial-grade quartz. In our study, the required energy for extracting silica sand from high-quality deposits was 339 MJ/metric ton of silica sand and increased to 1010 MJ for industrial-grade quartz. Compared to the current process commonly used in PV LCA, this number is almost three times higher than high-quality quartz. We also analyzed the carbon footprint of silica sand extraction in leading silica sand producers in Europe (Fig. S18). The highest carbon footprint was for producing one metric ton of silica sand in the Netherlands (44.8 kg of CO_{2eq}), and the lowest carbon footprint was for France (41.8 kg of CO_{2eq}).

Fig. 5B and C show the GWP of silica sand for MG-Si production in China. The baseline scenario for China was based on high-quality quartz. The GWP of imported industrial-grade silica sand from legal mines was 117 kg CO_{2eq} for Australia, 78.3 kg CO_{2eq} for Pakistan, 69.3 kg CO_{2eq} for Malaysia, and 58.3 kg CO_{2eq} for Cambodia Fig. 5B). For illegal mines in North Korea, the GWP was 46.8 kg CO_{2eq}. The difference by country is due to differences in electricity mix and fuel types necessary for mining operations and distance to China. When considering silica sand imports reported by the UN (51% Australia, 45%, Cambodia, 1% Malaysia, and 3% Pakistan), the GWP was 88.9 kg CO_{2eq} (Eq. (1) & (2). For domestic low-quality quartz, the GWP depends on the type of beneficiation process (Fig. 5C) and was 74.6 kg CO_{2eq} for the magnetic separation, 75.1 kg CO_{2eq} for the gravity separation, and 86.7 kg CO_{2eq} for the flotation per metric ton of silica sand, The flotation technique had a higher GWP due to higher fuel requirements. For foreign sand supply, the important parameter that affects the final product's carbon footprint is quartz purity, which is decreasing over time (Calvo et al., 2016) and could increase the carbon footprints of future Si PV products. The associated carbon footprint of low-quality silica sand can increase up to three times (Fig. 5B and 5C).

The lowest CED was for importing industrial silica sand from illegal mines in North Korea, which was 875 MJ per metric ton of industrial silica sand (Fig. 5E). The highest CED was 1890 MJ for Australia. The CED of domestic low-quality quartz was 1120 to 1580 MJ (Fig. 5F). This

(A)



(B)

	LCA Scenario		Description
US	Extracting silica sand from domestic quartz		High-quality deposits (>98% SiO_2) Industrial-grade deposits (95% SiO_2)
China	Baseline		Extracting silica sand from high-quality deposits
	Imported industrial-grade silica sand	Legal mines	Australia (AU) Cambodia (KH) Malaysia (MY) Pakistan (PK) 51% AU, 45% KH, 15 MY, and 3%PK
		Illegal mines	Cambodia (KH) North Korea (NK)
	Extracting silica sand from domestic quartz		Low-quality quartz (65% SiO_2) using gravity separation technique Low-quality quartz using flotation technique Low-quality quartz using magnetic separation technique

Fig. 4. (A) The regional production of MG-Si (colored provinces) (adopted from (Liu, 2015; *U.S. Geological Survey (USGS)* Mineral Commodity Summaries *2019*, 2020)) and potential domestic quartz deposits (orange dots) for China. (B) LCA scenarios for quantifying the carbon footprint and the CED of supplying silica sand for the US and China (The purity of deposits is >98% for high quality, 95% for industrial-grade, and 65% for low quality).

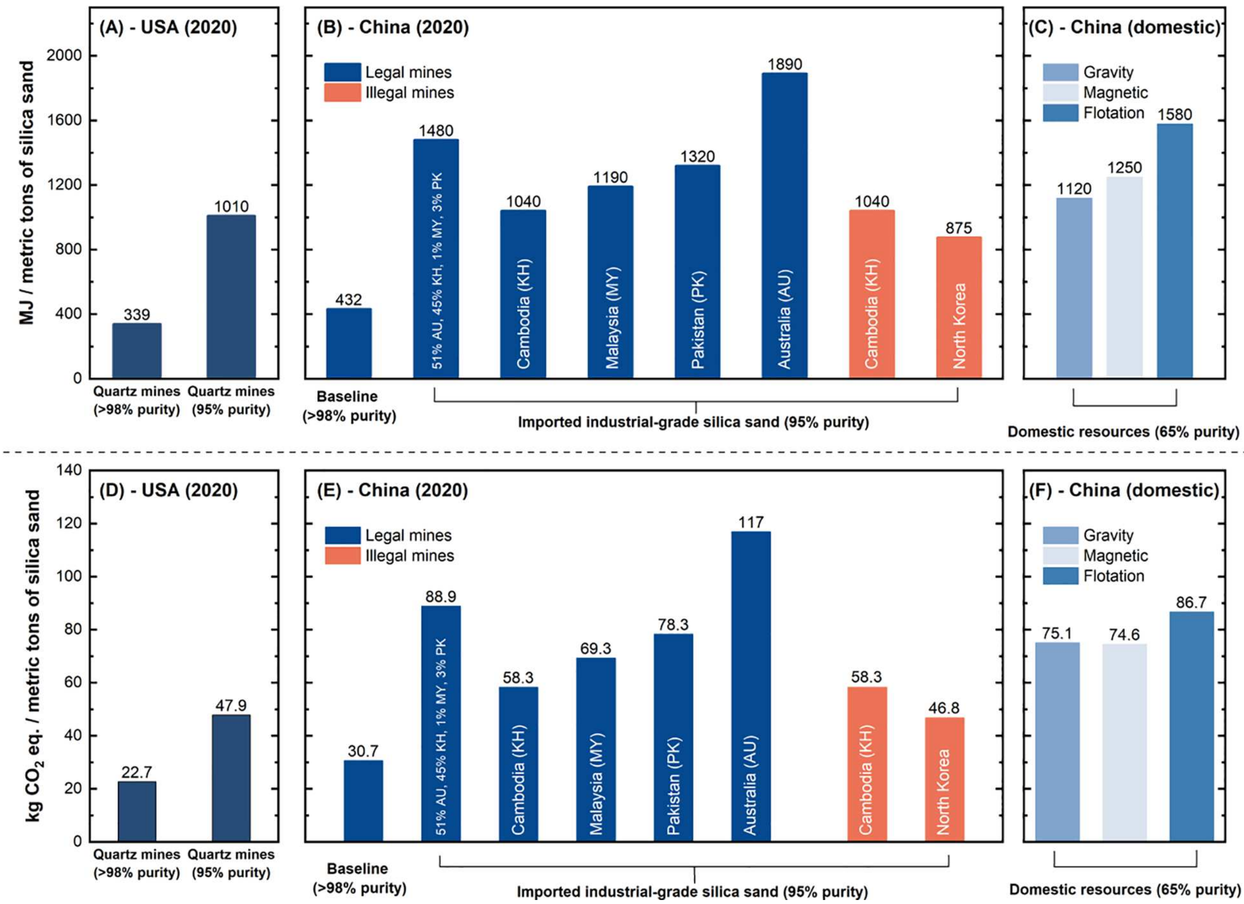


Fig. 5. The GWP (A, B, C) and CED (D, E, F) of silica sand production for MG-Si production in the US and China for the scenarios mentioned in Fig. 4B.

value was 16–40% lower than importing industrial silica sand from Australia but 28–81% higher than importing silica sand from North Korea and 8–52% from Cambodia. The CED increased for extracting silica sand from quartz with low purity, which needs to be considered for calculating the EPBT and EROI of Si PV. CED is also an indicator of the product cost (Anctil et al., 2011). Therefore, importing industrial-grade silica sand from Cambodia and North Korea would be cheaper than using domestic low-quality resources or importing from Australia, Pakistan, and Malaysia.

One way of reducing the use of illegal quartz for Si manufacturing would be through PV supply chain tracking and third-party certification. One existing effort to improve supply chain transparency and PV sustainability is the Electronic Product Environmental Assessment Tool (EPEAT) ecolabel from the Green Electronics Council for Photovoltaic Modules and Inverters (*EPEAT Program, GEC, PVMI-2021*). The product criteria used the NSF International standard #457: Sustainability Leadership for Photovoltaic Modules and Photovoltaic Inverters (NSF/ANSI 457, 2019), which include disclosure and social criteria for suppliers. There is an increasing concern about the use of forced labor in the PV supply chain. In February 2021, the Solar Energy Industries Association (SEIA) and about 175 solar companies signed an agreement opposing the use of forced workers in PV supply chains (Wagman, 2021). In June 2021, the US banned the import of Chinese polysilicon from Xinjiang due to concerns of forced labor (The White House, 2021). Overall, the PV industry supply chain is facing increasing scrutiny, but most of the actions so far have been limited to forced labor, while there are other potential concerns with illegal mining. Quartz mining is becoming a global socio-environmental challenge. The mining industry may positively impact local communities' developments by creating direct and indirect jobs. However, the possibility of excessive excavation in illegal mining may have adverse effects in the long term, even though there might be short-term financial benefits for local communities. Mining silica sand from oceans and riverbeds is more interesting since they are naturally crushed and ready to use. Riverbank sand is cheaper since removing the salt is unnecessary (R. UNEP, 2019). But illegal and excessive mining in rivers and oceans could create severe social and environmental problems.

In Indonesia, 24 small islands and their ecosystems disappeared between 2005 and 2010 due to excessive quartz mining and silica sand extraction to export to Singapore (The New York Times, 2010). The Mekong River is another example that silica sand mining threatens the ecosystem. The Mekong River is the 10th longest river globally and starts from China and passes through Laos, Thailand, Vietnam, and Cambodia. The largest extractors are located in Cambodia, where they extract 33 million metric tons of silica sand per year (Hackney et al., 2020). Excessive silica sand mining may change the river morphology and erosion pattern, affecting fisheries and, consequently, threatening the main food source of 60 million people living in that region, as is stated in the literature (Koehnken and Rintoul, 2018; Piman and Shrestha, 2017). Farmers' incomes can also be affected due to excessive mining activities. A 20% reduction in Cambodia was reported due to a lack of agricultural lands (*DHI*, 2015).

Quartz mining may also affect communities' livelihood. In a survey study in a village in Tangail District in Bangladesh, the local community was concerned about potential disasters and the black market due to quartz mining activities near their livelihoods (Khan and Sugie, 2015). Depletion of groundwater, reduction of soil ability to provide nutrients, increase the pH water and turbidity of the river, destruction of infrastructures, riverbank collapse, and social collapse are only some examples of mining silica sand in rivers that are also reported in the literature for mining activities (Koehnken and Rintoul, 2018; Lumpur, 2010; M. Naveen Saviour, 2012; Musah, 2009; Qin et al., 2020; *R. UNEP 2019*). Those socio-environmental impacts can be more severe for illegal mines due to a lack of legislation and public awareness.

Recent changes in legislation in Cambodia have limited silica sand extraction in the Mekong River and may affect export to China, which could affect Si PV manufacturing. However, it is reported that China has already started developing new low-cost purification methods (JIA et al., 2014; Wanfang Data, 2021; Zhou and Yang, 2018) and will likely start using domestic low-quality quartz resources.

3.2.2. Producing MG-Si from silica sand in the USA and China

The quartz purity affects the carbon footprint and CED of producing MG-Si. Fig. 6A shows the LCA scenarios considered for MG-Si production in the US and China. Table S1 and Fig. S11 provide additional details of the MG-Si production model. The GWP to produce one kg of MG-Si from imported industrial-grade silica sand was 12.1 kg CO_{2eq} (Fig. 6B). In comparison, using domestic low-quality quartz produced 16.5 kg CO_{2eq}, which is a 36% increase compared to imported industrial-grade silica sand. The GWP of producing one kg of MG-Si in the USA was 12.0 kg CO_{2eq}. The CED of producing one kg of MG-Si from current resources (imported industrial-grade silica sand from 51% AU, 45% KH, 1% MY, and 3% PK, (UN Comtrade)) was 188 MJ and increased by 53% (286 MJ) if domestic low-quality quartz resources were used instead (Fig. 6C). Silica sand purification requires a large amount of energy due to the required high temperature (about 1800 °C) to remove impurities from silica sand. Low-quality quartz contains more impurities and consequently requires more energy. The CED was 186 MJ for producing one kg of MG-Si in the USA.

The increase in Si PV installations will increase the demand for MG-Si production and likely increase the use of lower quality quartz due to the limited availability of high-quality resources globally. Unless significant progress is made in the process or either reducing the electricity intensity, MG-Si production carbon footprint and CED could increase over time and increase the carbon footprint and CED of Si PV products. For silica sand production, the CED and carbon footprint are influenced mainly by the transportation distance and the purity of quartz deposits. For MG-Si production, the required electricity to remove impurities from low-quality quartz plays a more significant role in increasing carbon footprint and CED compared to the transportation distance, showing a need to improve the MG-Si production technique before using low-quality quartz deposits.

4. Conclusion

Manufacturing Si PV requires a large amount of quartz. Quartz purity affects the carbon footprint and CED of producing MG-Si used in Si PV. Previous studies focused only on scarce material and ignored quartz, probably since quartz deposits are perceived as abundant and available everywhere. Some countries, such as China, the global leading Si PV

(A)

	LCA Scenario		Description
US	MG-Si production from domestic quartz		83.35% high-quality deposits (>98% purity) and 16.65% industrial-grade deposits (95% purity)
China	MG-Si production from industrial-grade quartz	Legal mines	51% AU, 45% KH, 15 MY, and 3%PK
		Illegal mines	50% KH and 50% NK
	MG-Si production from domestic quartz		Low-quality deposits (65% purity, separation techniques: 1/3 gravity, 1/3 flotation, and 1/3 magnetic)

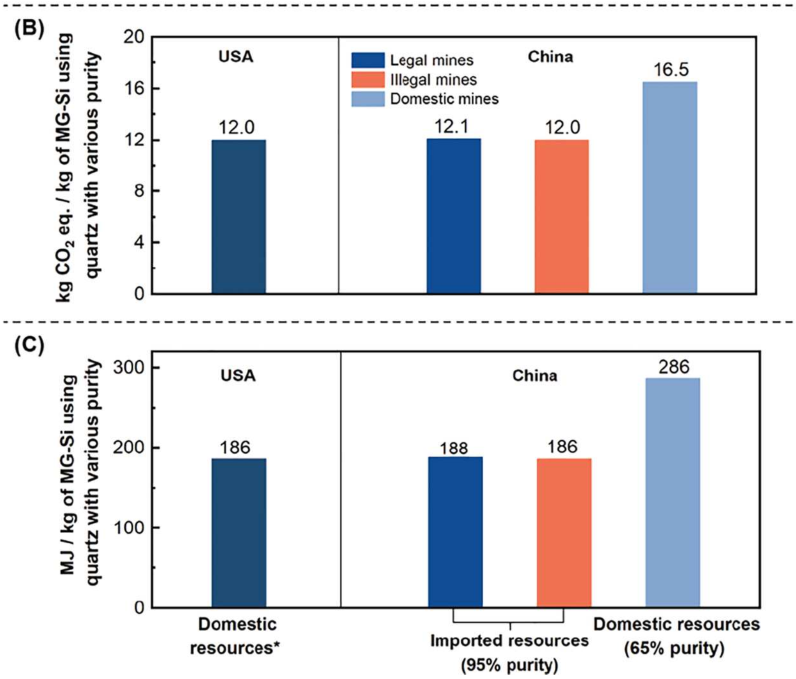


Fig. 6. (A) The LCA scenarios of producing one kg MG-Si from high- and low-quality silica sand and associated carbon footprint (B) and CED (C).

producer, do not have high-quality (>98% purity) and industrial-grade (95% purity) quartz deposits and are highly dependent on foreign resources. Regulations restricting silica sand export to China may force Chinese MG-Si producers to use domestic low-quality (65% purity) quartz. China produced about 4.5 million metric tons MG-Si (64% of global production) in 2019 (Fig. 3D). The transition from importing high-quality or industrial-grade silica sand to mining low-quality domestic quartz can increase MG-Si production's annual carbon footprint by at least 23% (Fig. 6B). This corresponds to the annual greenhouse gas emissions from about nine million passenger cars. Currently, most MG-Si is produced in China, but the USA production might increase since the Biden administration plans to ensure the future is "Made in All of America," which would also have environmental benefits since China does not have high-quality quartz.

Si PV companies should improve the efficiency of existing MG-Si production before using low-quality quartz resources and increasing the carbon footprint of PV. Upgraded MG-Si is a new technology that can be a replacement for existing MG-Si production processes in terms of cost and carbon footprint (Méndez et al., 2021). Other sources of silica should be considered for MG-Si production, including secondary resources such as rice husk ash (Azet et al., 2019; Joglekar et al., 2019) and gold mining tails to reduce the pressure on natural resources (Okereafor et al., 2020). We analyzed domestic scenarios based on today's technology, which was assumed to be used in the future. It is recommended to investigate the future mining technology in more detail and explore how the technology changes can affect the silica sand production from low-quality quartz. The evaluation of CED of quartz mining, silica sand extraction, and MG-Si production can be used for calculating the energy return on investment (EROI) and the energy payback time (EPBT) of silicon-based photovoltaics.

In addition to the environmental impact of MG-Si production, the social impacts associated with quartz mining are not often discussed in the literature. Global silica sand production has tripled in the last decade. Even though the implication of quartz mining projects can have short positive effects such as creating temporary jobs for locals, it may have severe long impacts on local livelihood, particularly in regions with limited regulations. The New York Times has recently reported that some countries have regulations that may let MG-Si producers hire forced workers from ethnic minorities in an unacceptable condition (Swanson and Buckley, 2021). As previously discussed, there are efforts to the recent ban on importing silicon from some producers based on forced workers, which hire forced workers, is a proper effort from the US government, but it should not be limited to forced labor. Apart from inefficient rules for quartz mining, illegal mines can have more severe impacts on the environment and local communities, which are ignored in the literature. The PV installation is increasing and can result in a more serious silica sand shortage. This would create more space for "Sand Mafias" to fill the gap between market demand and supply as it has been happening for providing the required silica sand for construction in more than 70 countries (Rege and Lavorgna, 2017). So, it is essential to pay attention to potential environmental and social issues of illegal mining as well. Recent efforts such as EPEAT ecolabel, as formerly mentioned, are helpful to increase the transparency of the PV supply chain, which needs to be expanded to reduce the risk of using illegal raw materials for PV. Solar PV is a greener alternative to current electricity production, but its image could be tarnished by an increasing manufacturing carbon footprint, or even worst, using quartz from illegal mines and forced labor. To ensure PV remains a sustainable energy option, we must ensure that PV supply chains are free from unethical activities such as using illegal mining and forced labor.

CRediT authorship contribution statement

Seyed M. Heidari: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Visualization. **Annick Anctil:** Conceptualization, Methodology, Validation, Resources, Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

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

Acknowledgement

This work was supported by the National Science Foundation grant number NSF-1801785.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.resconrec.2022.106171.

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