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Wickerts, S., Arvidsson, R., Peters, G. et al (2021)

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ACS Sustainable Chemistry & Engineering, 9(14): 5187-5195

<http://dx.doi.org/10.1021/acssuschemeng.1c00376>

N.B. When citing this work, cite the original published paper.

Prospective Life-Cycle Modeling of Quantum Dot Nanoparticles for Use in Photon Upconversion Devices

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Cite This: <https://doi.org/10.1021/acssuschemeng.1c00376>

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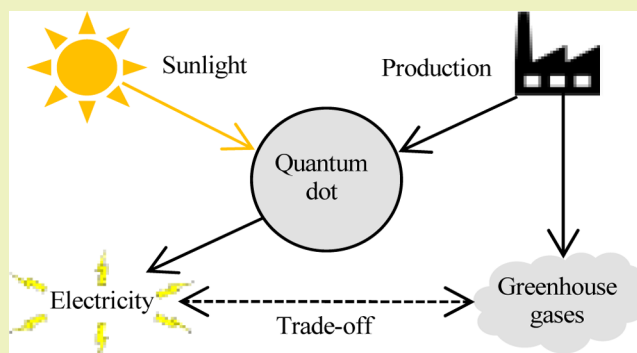
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Supporting Information

ABSTRACT: Quantum dot nanoparticles (NPs) can be used in several applications, for example, photon upconversion devices that increase the electricity output of solar modules. In order to facilitate life cycle assessment (LCA) studies of such applications, this study provides ready-to-use LCA unit process data for four NPs suitable for photon upconversion applications: cadmium selenide, cadmium sulfide, lead selenide, and lead sulfide. The data is provided for two prospective scenarios: one optimistic and one pessimistic. An impact assessment is conducted in order to assess the NPs' climate change performance, where solvent-related processes such as steam production for recycling and hazardous waste treatment are shown to be hotspots. To show the applicability of the data, a prospective assessment of a solar module with an upconversion layer is conducted to investigate whether it is preferable from a climate perspective to install more solar modules or equip existing ones with upconversion devices, leading to more electricity produced in both cases. The assessment shows that solar modules need to become 0.05–2 percentage points more efficient per gram of NPs applied, depending on the scenario, in order for the upconversion layer to be preferable.

KEYWORDS: LCA, nanomaterials, solar energy, cadmium selenide, cadmium sulfide, lead selenide, lead sulfide



INTRODUCTION

Engineered nanomaterials (ENMs), often defined as deliberately manufactured materials with at least one dimension in the 1–100 nm size range, are increasingly being used in consumer products.^{1,2} Estimates suggest that ENMs are produced at tens of thousands metric tonnes per year globally, or even millions of tonnes per year if historically well-established ENMs, such as carbon black, are included.³ There are also ENMs which are not yet used in products to a large extent but have promising applications that might become extensively used in the future. Following the increase in both realized and envisioned applications, ENMs have increasingly become the object of study in life cycle assessment (LCA).⁴ Examples of ENMs assessed so far include fullerenes,⁵ carbon nanotubes,^{6–8} nanocellulose,^{9–12} silver nanoparticles (NPs),¹³ titanium dioxide NPs,^{14,15} and graphene.^{16–18} Most LCA studies of ENMs are cradle-to-gate studies, where only the production and raw material extraction are included. Such studies have the main purposes of improving the environmental performance of production processes and enabling future LCA studies of products containing the ENMs. The latter is important since each practitioner faced with the task of conducting an LCA of a product containing an ENM would otherwise need to conduct an assessment of the often complex synthesis route for that ENM, which is not always feasible given the time and

monetary constraints of projects. Providing ready-to-use data for ENMs can thus facilitate LCA studies of products containing those materials. This study provides ready-to-use data for four quantum dot ENMs: cadmium sulfide NPs (CdS NPs), cadmium selenide NPs (CdSe NPs), lead sulfide NPs (PbS NPs), and lead selenide NPs (PbSe NPs). In addition, an impact assessment is conducted in order to provide information about the NPs' climate change performance. To the best of our knowledge, no previous LCA studies have assessed CdS, PbS, or PbSe NPs. For CdSe NPs, two previous LCA studies exist,^{19,20} but they were based on different synthesis routes than in the present study.

The main reason for our interest in these NPs is their potential use in a future application called photon upconversion.²¹ For certain applications, photons of sufficient energy are required. For example, current silicon solar modules require photons with a minimum energy to overcome the bandgap of silicon, $E_{bg} = 1.1$ eV. Combining two low-energy

Received: January 18, 2021

Revised: March 8, 2021

photons in the near-infrared part of the solar spectrum through upconversion into one higher-energy (>1.1 eV) photon in the visible range can potentially be utilized in a silicon solar module.²² Another application is to upconvert visible light to ultraviolet light, which is required to split water into hydrogen and oxygen on simple and stable wide-bandgap materials, thereby enabling direct production of hydrogen fuel from water.²³ Both applications utilize a specific variant called triplet–triplet annihilation photon upconversion (TTA-UC), which uses two different molecular species called sensitizers and annihilators to produce higher-energy photons.²⁴ The sensitizer absorbs incident light, which creates excitations to higher energy states. An annihilator will then receive this energy, resulting in an excited annihilator and a ground-state sensitizer. Two excited annihilators will together undergo TTA, during which one of them returns to its ground state by transferring its excess energy to the other one, which emits a higher-energy photon to be utilized in the application. The NPs assessed in this study can be used in TTA-UC, functioning as sensitizers. Due to their respective bandgaps, CdS and CdSe NPs are more suitable for converting visible light to ultraviolet (e.g. for water splitting), whereas PbS and PbSe NPs are more suitable for converting near-infrared light to visible light (e.g. for improving efficiency of silicon solar modules). To show the applicability of the derived cradle-to-gate LCA data on the NPs for assessing the environmental performance of such applications, we conduct a simplified, prospective environmental assessment of PbS and PbSe NPs for upconversion of near-infrared light to light utilizable by a silicon solar module.

The aim of this study is thus twofold: (i) to provide ready-to-use LCA unit process data and climate change impact assessment results for CdS, CdSe, PbS, and PbSe NPs, as well as (ii) to apply the data for PbS and PbSe NPs in a simplified assessment of a photon upconversion device used in conjunction with a silicon solar module. Considering that the NPs are not yet produced at a large scale and the photon upconversion has only been proven at the concept level, this study utilizes insights and concepts from the LCA of emerging technologies, so-called the prospective LCA.²⁵

METHODS

In a prospective LCA, the studied emerging technology is placed in a future state where the technology has matured and is produced at a large scale.²⁵ This means that higher technology readiness levels (TRLs)²⁶ and/or manufacturing readiness levels (MRLs)²⁷ should be considered in the inventory modeling of the emerging technology. The NPs studied were in 2020 produced at a small scale and low rates, corresponding approximately to MRL = 8 or 9 out of 10. Photon upconversion has so far only been shown in terms of experimental proof of concept at the laboratory scale, thus having approximately TRL = 3 out of 9. The prospective modeling conducted here is based on two prospective scenarios with MRL = 10 for the NPs and TRL = 9 for the photon upconversion: (i) an optimistic production scenario (OS) with lower environmental impacts and (ii) a pessimistic scenario (PS) with higher environmental impacts. To model a mature state production (MRL = 10), some parameters are derived from current fine chemical production, with the two scenarios differing in terms of production yields and requirements for auxiliary input flows such as solvents, energy, and water (Table 1). The two scenarios also differ regarding electricity supply to the foreground system. Unfortunately, changing the electricity mixes also throughout the entire background system is challenging given the current structure of LCI databases, which rely on specific process–process linking and are therefore not well

Table 1. Overview of the Prospective Modeling of Quantum Dot NPs Conducted in This Study^a

parameter	OS	PS
photon upconversion TRL	actual system proven successful (TRL = 9)	actual system proven successful (TRL = 9)
quantum dot NP production MRL	full rate production (MRL = 10)	full rate production (MRL = 10)
production yields	best-case fine chemical production yield (97%)	current experimental yields (50%–80%)
production input amounts ^b	like current fine chemical production, best case	like current fine chemical production, worst case
production input impacts	current impacts (status quo)	current impacts (status quo)
electricity supply to the foreground system	solar power	European average electricity mix

^aTRL = technology readiness level, MRL = manufacturing readiness level. ^bSolvents, steam, electricity, cooling water, and inert gas.

designed for scenario analyses throughout whole product systems,²⁸ although this would have been preferable in a prospective LCA. In both scenarios, the status quo is assumed for chemicals produced in mature markets, assuming no changes in current production processes. Environmental impacts from building production facilities needed for the future manufacturing of the NPs are excluded in both scenarios since they are believed to be negligible.

Prospective Unit Process Modeling. For the prospective unit process modeling of the NPs at MRL = 10, we follow a stepwise procedure outlined by Arvidsson et al.,²⁹ with some modifications:

- (i) identify a technically plausible future production process;
- (ii) estimate reactant and product amounts;
- (iii) estimate auxiliary input amounts, such as solvents and energy requirements;
- (iv) estimate process emissions;
- (v) identify likely by-products and their handling; and
- (vi) estimate amounts of waste.

Step (i) is conducted by identifying NP synthesis routes used in successful photon upconversion studies, which proved to be generally established synthesis procedures for the NPs. Thus, the cradle-to-gate data for the NPs in this study is not only applicable to photon upconversion. Steps (ii)–(iv) are conducted by means of process calculations,³⁰ preferably relying on process-specific data provided in the NP synthesis descriptions if available. Process synergies, such as recovery and reuse of heat suggested by van der Hulst et al.,³¹ are not explicitly assumed besides the recycling of solvents but implicitly in terms of lower energy requirements in the OS. The solvent recycling is modeled as a distillation process (requiring electricity, steam, nitrogen gas, and cooling water) with a 95% recovery rate in the OS and 0% in the PS.³² All cooling water is assumed to be lake water.

In step (v), by-products would have been identified as output flows feasible to separate and utilize commercially. However, since all non-solvent output flows are present in low concentrations, they are likely not valuable enough to separate. However, partitioning of the environmental burden between products and by-products is performed by mass-based allocation in the modeling of the trimethylchlorosilane and naphthalene production processes in the PbS NP production system (Figure 4). No subdivision of the system to avoid allocation is possible since all products are formed jointly and no economic allocation is possible since relevant prices could not be obtained for some by-products. To check the sensitivity of this choice, the calculations were also conducted by allocating all burdens to the PbS NP production system, that is, using the “main product bears all burden” approach,³³ which constitutes a worst-case scenario with regard to allocation.³⁴

In step (vi), the amount of waste is estimated from a mass balance over the entire unit process, subtracting the amounts of products, by-products, and emissions from the total amount of input materials. The

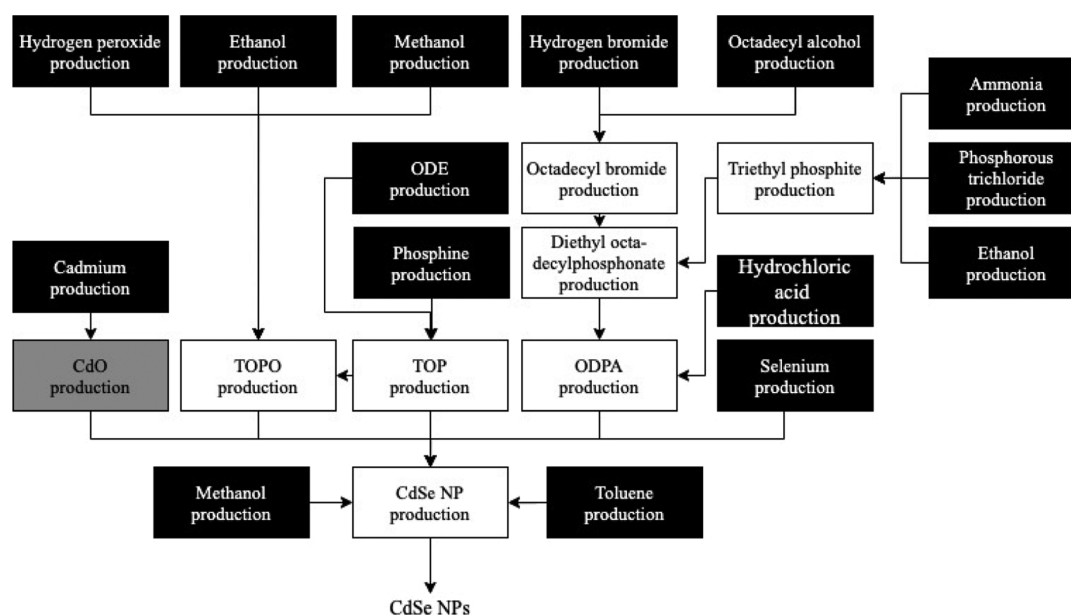


Figure 1. Flowchart showing the cradle-to-gate life cycle of CdSe NPs. Fine chemical production processes modeled in this study are white, bulk chemical production processes modeled in this study are gray, and production processes for which data was obtained from the ecoinvent database or other sources are black. The gray and white boxes constitute the foreground system, while the black boxes constitute the background system. CdO = cadmium oxide, ODE = 1-octadecene, ODPa = octadecylphosphonic acid, TOP = trioctylphosphine, and TOPO = trioctylphosphine oxide.

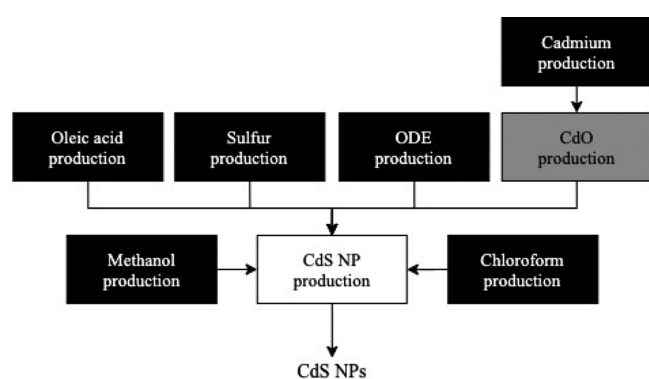


Figure 2. Flowchart showing the cradle-to-gate life cycle of CdS NPs. Fine chemical production processes modeled in this study are white, bulk chemical production processes modeled in this study are gray, and production processes for which data was obtained from the ecoinvent database or other sources are black. The gray and white boxes constitute the foreground system, while the black boxes constitute the background system. CdO = cadmium oxide and ODE = 1-octadecene.

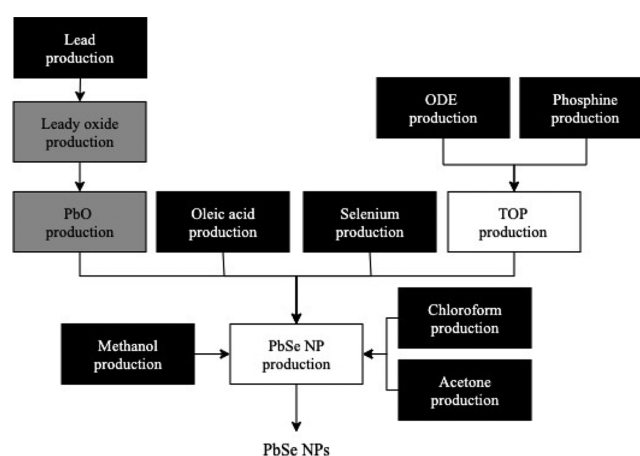


Figure 3. Flowchart showing the cradle-to-gate life cycle of PbSe NPs. Fine chemical production processes modeled in this study are white, bulk chemical production processes modeled in this study are gray, and production processes for which data was obtained from the ecoinvent database or other sources are black. The gray and white boxes constitute the foreground system, while the black boxes constitute the background system. ODE = 1-octadecene, PbO = lead oxide, TOP = trioctylphosphine.

residual is assumed to become liquid hazardous waste sent for incineration, since it mainly consists of non-recycled solvents with at least one toxic component.

In addition to the NPs, materials for which production data was unavailable in the LCA databases and the scientific literature require specific modeling, mainly fine chemicals required in the NP production [bis(trimethylsilyl)sulfide (TMS) and octadecylphosphonic acid (ODPA) and most of its precursors, trioctylphosphine (TOP) and trioctylphosphine oxide (TOPO)] but also some bulk chemicals [lead oxide (PbO), cadmium oxide (CdO) and hydrogen bromide]. For these additional materials, in order to have a consistent modeling throughout the study, we apply the same steps (i)–(vi) above. For a few processes producing hazardous solid waste (lead and cadmium oxide production), the waste is assumed to be sent to underground deposits.

For the NPs and other material production, there are some data gaps regarding yields, inputs, and emissions. In order to fill data gaps, we distinguish between three categories of materials: the NPs, fine chemicals without data, and bulk chemicals without data. Fine and bulk chemicals are distinguished based on Pollak,³⁵ with bulk (or basic) chemicals being produced in large volumes at low prices, whereas fine chemicals are produced in small volumes at high prices. Table S1 in the Supporting Information describes how data gaps were filled using proxy data³⁰ for these three categories of materials in the PS and OS.

In the NP production processes, precipitation and washing chemicals (PWCs) are generally used, but they are not quantified in any of the experimental studies nor in the proxy data sources in

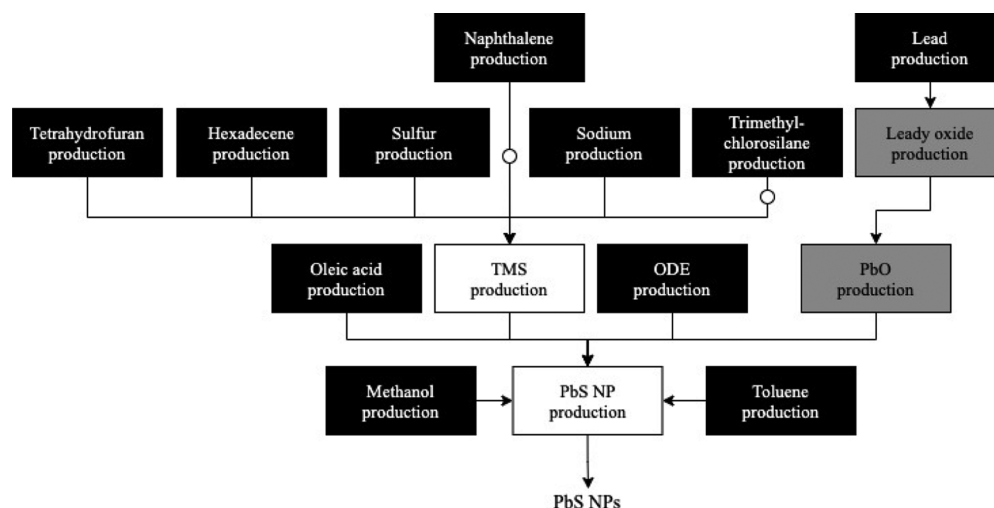
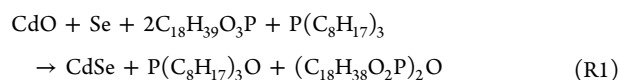


Figure 4. Flowchart showing the cradle-to-gate life cycle of PbS NPs. Fine chemical production processes modeled in this study are white, bulk chemical production processes modeled in this study are gray, and production processes for which data was obtained from the ecoinvent database or other sources are black. The gray and white boxes constitute the foreground system, while the black boxes constitute the background system. Flows for which inputs and emissions are allocated are marked with a circle. TMS = bis(trimethylsilyl)sulfide, TOP = trioctylphosphine, ODE = 1-octadecene, and PbO = lead oxide.

Table S1 (Supporting Information). To fill data gaps regarding PWCs, we create proxy data (optimistic and pessimistic) based on a literature review (Table S2, Supporting Information).

The four subsections below describe the modeling of the NP production with corresponding flowcharts presented in Figures 1–4. The descriptions in these sections only include the process-specific data from the NP synthesis descriptions, while data gaps are all filled using proxy data, as shown in Tables S1 and S2 (Supporting Information). ecoinvent (version 3.6, cutoff) datasets used for background system modeling are provided in Table S21 in the Supporting Information.

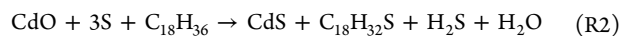
CdSe NP Production. Several photon upconversion studies, for example, Huang et al.³⁶ and Mongin et al.,³⁷ used the synthesis described by Carbone et al.,³⁸ where CdO is reacted with selenium, ODPa ($C_{18}H_{39}O_3P$), and TOP ($P(C_8H_{17})_3$) to produce CdSe NPs. This synthesis was selected in step (i), with the following reaction^{39,40}



In step (ii), the reactant amounts are provided,³⁸ but the output amounts had to be calculated stoichiometrically with ODPa as the limiting reactant and modified using a yield of 80%⁴¹ in the PS and the proxy yield (97%) shown in Table S1 (Supporting Information) in the OS. In step (iii), the amounts of the TOP and TOPO are provided.³⁸ Methanol and toluene are used as PWCs in this synthesis.³⁸ Since TOP serves as both a reactant and solvent, the solvent recycling factor is only applied to the solvent amount of TOP in the OS. In steps (v and vi), no by-products are likely to be obtained from the hazardous mixture washed away from the CdSe NPs.

Note that in previous LCA studies of CdSe NPs,^{19,20} the CdSe NP production was modeled based on other syntheses.^{42,43} These syntheses are similar but use, for example, different cadmium precursors (methyl cadmium and cadmium acetate).

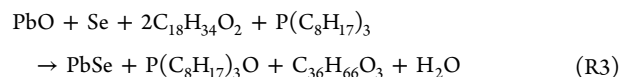
CdS NP Production. Several photon upconversion studies, for example, Gray et al.,²³ prepared CdS NPs by reacting CdO with sulfur and 1-octadecene (ODE, $C_{18}H_{36}$) according to the synthesis described by Yu and Peng.⁴⁴ This synthesis is selected in step (i), and the overall reaction is^{45,46}



In step (ii), the output amount is calculated stoichiometrically from R2 based on the limiting reactant (sulfur) with a yield of 67% in the PS⁴⁵ and the proxy yield (97%) from Table S1 (Supporting

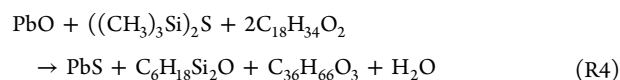
Information) in the OS. In step (iii), only the amount of the ODE solvent is provided.⁴⁴ Since ODE serves both as a reactant and a solvent, a solvent recycling factor of 95% is applied to the solvent part of the ODE amount only in the OS. Methanol and chloroform are reported as PWCs.⁴⁴ In steps (v and vi), it is assumed that all reaction products other than CdS NPs become liquid hazardous waste along with used solvents and unreacted reactants.

PbSe NP Production. For the production of PbSe NPs, several upconversion studies, such as Bonati et al.⁴⁷ and Marques-Hueso,⁴⁸ used the synthesis described by Yu et al.,⁴⁹ where PbO is reacted with selenium, oleic acid ($C_{18}H_{34}O_2$), and TOP ($P(C_8H_{17})_3$). This synthesis is selected in step (i) and occurs as follows^{40,46}



In step (ii), the output amounts are calculated stoichiometrically based on a proxy yield of 97% in the OS. Regarding the current experimental yields, values from about 2%^{50,51} to almost 100%⁵² are reported. In the PS, we therefore assume that at least 50% will be required for a future large-scale production of PbSe NPs. In step (iii), only the amount of solvents (TOP and ODE) is reported.⁴⁹ As TOP serves as both a reactant and solvent, a solvent regeneration factor (95%) is applied only to the solvent part of the total TOP amount in the OS. Methanol, chloroform, and acetone are reported as PWCs.⁴⁹ In steps (v and vi), reaction products besides PbSe NPs are assumed to become liquid hazardous waste.

PbS NP Production. The production of PbS NPs is based on the synthesis described by Hines and Scholes,⁵³ used in several photon upconversion studies,^{22,54} where PbO reacts with the sulfur carrier TMS and oleic acid. This synthesis is selected in step (i)⁴⁶



In step (ii), the output of PbS NPs is calculated stoichiometrically and inputs are modified using a yield of 50%⁵⁵ in the PS and an optimistic yield (97%) in the OS. In step (iii), the input amounts of the two solvents used (ODE and oleic acid) are provided.⁵³ Since oleic acid is also a reactant, the recycling factor is only applied to the solvent share. Regarding PWCs, methanol and toluene are reported to be used.⁵³ In steps (v and vi), reaction products besides PbS NPs are considered to become hazardous liquid waste.

Impact Assessment. In order to illustrate the use of the prospective inventory data for the NPs and identify hotspots, an impact assessment is conducted. Climate change is chosen as an impact category considering its importance as an environmental issue⁵⁶ and since it captures, for example, the use of fossil solvents in NP production as well as the different carbon intensities of the electricity supplies in the PS and OS (Table 1). It is modeled by characterization factors from IPCC AR5 with a 100 year time horizon.⁵⁶ In addition, when assessing the NPs with a broader range of impact categories from ReCiPe 2016,⁵⁷ it was noted that most correlate to a high degree with climate change (both in the PS and the OS), including fossil resource scarcity, water use, freshwater ecotoxicity, and terrestrial acidification. Results of the latter three impact categories can be found in the Supporting Information. The modeling was conducted in the open-source software OpenLCA.

Photon Upconversion Device Modeling. Photon upconversion devices are often envisioned to be mounted onto the rear side of a solar module,^{58,59} thereby not affecting the opacity of the absorption area of the module. Such devices must contain at least a sensitizer material that captures the photons and an annihilator material that converts into high-energy photons. In addition, quantum dot NP sensitizers are sometimes coated with thin shells.^{2,3} Small organic molecules called transmitters must also be attached to the particles' surfaces to prolong the lifetime of certain intermediate states.⁶⁰ Solvents for the sensitizer and annihilator materials, as well as back mirror reflectors to increase light capturing, can also be applied.^{24,58} An LCA of a solar module-upconversion layer device would ideally include all these upconversion components and possible alterations of the solar module itself. Due to the currently unknown configuration of such devices, most of these considerations are not included in this assessment. Instead, a simplified LCA is conducted, where it is anticipated that in future upconversion devices, the NPs will constitute the largest share both in terms of mass and environmental impact. The annihilators and transmitters, although technically required for TTA-UC, are thus omitted in this assessment. It also seems reasonable to assume that future upconversion devices will seek to minimize solvent use, perhaps even become all-solid devices. The technology is therefore modeled in terms of two parameters only: the amount of NPs of type *n* applied per solar module active surface (m_n [g/m²]) and the change in solar module efficiency in terms of percentage points ($\Delta\eta$). Unfortunately, there is yet no established relationship between m_n and $\Delta\eta$. We therefore investigate for which values of m_n and $\Delta\eta$ the environmental impact of the photon upconversion device could break even with that of a silicon solar module without the upconversion layer. The setup of the upconversion assessment is illustrated in Figure 5.

Starting from the lifetime electricity production of a solar module, it is given by

$$E = S\eta\tau \quad (1)$$

where E (kW h/m²) is the electricity production per m² for a certain location, S is the solar insolation (kW h/m² year), η is the module

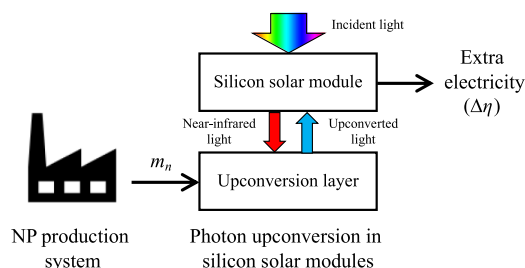


Figure 5. Illustration of the simplified assessment of photon upconversion with PbS NPs or PbSe NPs to enhance a silicon solar module by utilizing a larger fraction of the incident light. m_n = amount of NP (kg/m²) and $\Delta\eta$ = increase in energy efficiency (percentage points).

efficiency, and τ is the module lifetime (year). The impact for a certain category *j* per electricity produced (impact/kW h) can be expressed as

$$u_j = \frac{I_j}{E} = \frac{I_j}{S\eta\tau} \quad (2)$$

where I (impact/m²) is the life cycle environmental impact per m² of the solar module. The basic identity at the point of break-even is that the impact per kW h is equal for the product system for solar electricity without upconversion ($u_{j,s}$) and with upconversion ($u_{j,n}$)

$$u_{j,s} = u_{j,n} \quad (3)$$

Assuming that the lifetime of the solar module is not affected by the upconversion layer, this results in the following expression, where the increased impact from producing the NPs of type *n* ($I_{j,n}$) must be compensated by an increased efficiency ($\Delta\eta$)

$$\frac{I_{j,s}}{S\tau\eta_s} = \frac{I_{j,s} + I_{j,n}}{S\tau_s(\eta_s + \Delta\eta)} \quad (4)$$

Rearranging eq 4 leads to

$$\Delta\eta I_{j,s} = \eta_s I_{j,n} \Rightarrow \Delta\eta = \frac{\eta_s I_{j,n}}{I_{j,s}} \quad (5)$$

$I_{j,n}$ in eq 5 is calculated as

$$I_{j,n} = m_n v_{j,n} \quad (6)$$

where m_n is the amount of NPs of type *n* (kg/m²) and $v_{j,n}$ is the calculated cradle-to-gate environmental impact (impact/kg) from the present study. The analysis was conducted with *j* being climate change and *n* being the OS and PS of PbS and PbSe production, respectively, yielding four separate break-even assessments. Potential recycling of the NPs is not included in this break-even analysis. We assume an η_s of 20%, a reasonable average for modules in the decades to come,⁶¹ and an $u_{j,s}$ from the ecoinvent database (version 3.6, cutoff allocation), specifically the dataset "electricity production, photovoltaic, 3 kWp flat-roof installation, multi-Silelectricity, low voltage/cutoff, U, rest of the world".⁶² To calculate $I_{j,s}$ as per eq 2, E is needed, which is calculated based on data from Jungbluth et al.⁶²

RESULTS AND DISCUSSION

Ready-to-Use Unit Process Data. Ready-to-use unit process datasets for CdSe, CdS, PbSe, and PbS NP production can be found in the Supporting Information (Tables S5–S8). Considering the generality of the syntheses, these unit processes can be used as building blocks in prospective LCA studies involving these NPs as input materials. Most notably, there is a large use of solvents during the production of the NPs, in particular in the PS, where solvent inputs range from about 15 (for PbSe NPs) to 1800 (for CdS NPs) kg/kg NP. The high solvent use is in the same order of magnitude as the solvent use for pharmaceutical production, which can typically range from 25 to more than 100 kg/kg active pharmaceutical ingredient.⁶³ The PWCs also constitute large inputs in the NP production unit processes in the PS, ranging from about 60 to 800 kg/kg NPs. In the OS, less solvent is required due to a 95% solvent recycling factor. Less PWCs are also used, and the amount used is also recycled at 95%. However, in the OS, considerable inputs of steam, cooling water, and electricity are instead required to recirculate 95% of the solvents and PWCs. A conclusion from this study is thus that the studied NP syntheses require large amounts of input materials, in particular in the PS. Efforts should therefore be put into developing less material demanding synthesis routes than those considered in this study, for example, by considering the 12 principles of green chemistry.⁶⁴ In particular, principle 2 about the "atom

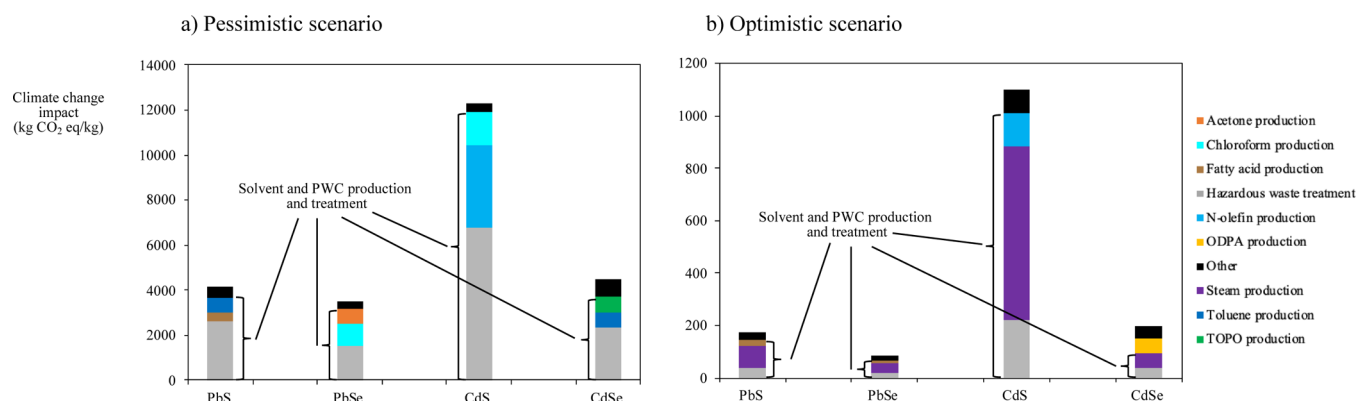


Figure 6. Cradle-to-gate impact assessment results for climate change given (a) a PS and (b) an OS of NP production. PbS = lead sulfide, PbSe = lead selenide, CdS = cadmium sulfide, CdSe = cadmium selenide, PWC = precipitation and washing chemicals, ODPa = octadecylphosphonic acid, and TOPO = trioctylphosphine oxide.

Break-even analysis

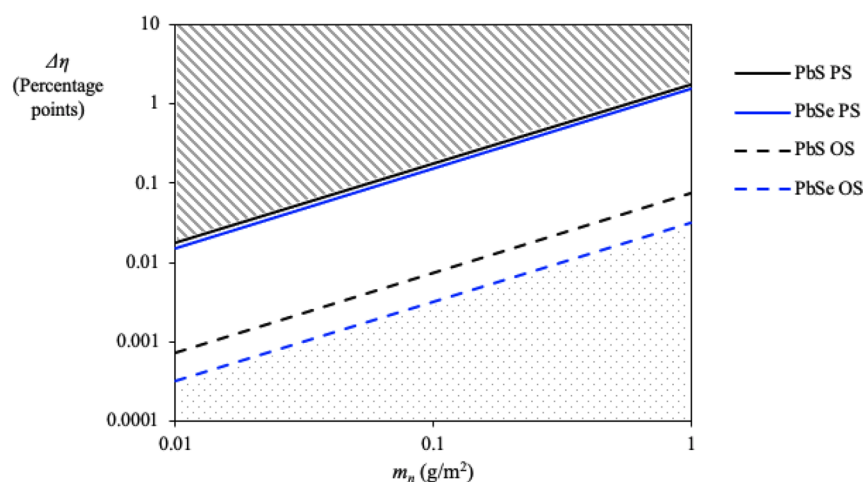


Figure 7. Break-even analysis for the climate change impact of a photon upconversion device with a certain mass of PbS or PbSe NPs per surface area (m_n), requiring a certain efficiency improvement ($\Delta\eta$) to break even with a current silicon solar module. The striped area represents values of m_n and $\Delta\eta$ where the upconversion layer is more preferable from a climate change perspective compared to installing additional solar modules for both scenarios of PbS and PbSe NP production, while the dotted area represents values where installation of additional solar modules is preferable in all cases. In the white area, the preferable option depends on the choice of NP and its production system. PbS = lead sulfide, PbSe = lead selenide, PS = pessimistic scenario, and OS = optimistic scenario.

economy”, stating that an as large share of the input materials as possible should be incorporated into the final product, is important to consider, since this share is low for the NP syntheses.

In addition, ready-to-use unit process datasets for the precursor materials without data in databases are also provided, that is, CdO, PbO, TMS, ODPa, TOP, and TOPO (Tables S9–S20, Supporting Information).

Climate Change Impacts. Looking first at the total results, CdS NPs have the largest climate change impact (Figure 6), causing about 10,000 kg CO₂ equiv/kg in the PS and 1000 kg CO₂ equiv/kg in the OS. The higher impact for CdS NPs depends on higher inputs of solvents and PWCs. The CdSe, PbS, and PbSe NPs have climate change impacts of similar order of magnitude to one another: about 4000–5000 kg CO₂ equiv/kg in the PS and 80–200 kg CO₂ equiv/kg in the OS. When applying the “main product bears all burden” allocation approach to the PbS NP system, the impacts are not significantly affected in the PS (from 4200 to 4700 kg CO₂ equiv/kg) but more so in the OS (from 170 to 430 kg CO₂

equiv/kg). Sengül and Theis¹⁹ reported life-cycle emissions of greenhouse gases (CO₂, CH₄, and N₂O) for CdSe NP production, amounting to about 5000 kg CO₂ equiv/kg CdSe NPs. Thus, their result is in the same order of magnitude as that of the PS for CdSe NP production in the present study, even though a somewhat different synthesis is modeled.

Looking then at the prospective hotspots, the vast majority of the climate change impacts are related to the waste treatment and recycling of solvents and PWCs. In the PS, treatment of hazardous waste is the largest hotspot for all NPs, and this waste comes mainly from spent solvents and PWCs (Figure 6a). In the OS, the use of steam, produced from natural gas, as a heat carrier for solvents and PWC recovery by distillation is the largest hotspot (Figure 6b). In addition, the production of some input materials (TOPO, acetone, chloroform, *n*-olefin, toluene, fatty acids, and ODPa) is also among the main contributors for certain NPs, where all except ODPa are either solvents or PWCs.

Changing the electricity mix in the foreground system, between solar power in the OS and average European

electricity in the PS, has a negligible influence on the results. This is because electricity is not a major input to the NP syntheses, for which most energy use rather originates from fossil-based materials with generally low electricity demand (in both scenarios) and steam from natural gas (in the OS). Since the main inputs are fossil-based materials with generally low electricity demand, we hypothesize that changes in background system electricity mixes would not have a profound influence on the results either.

Break-Even Analysis. In Figure 7, results from the break-even analysis are shown. Based on an approximate calculation of a plausible amount of NPs per upconversion layer area, as provided in the Supporting Information, resulting in 0.1–0.7 g/m², the range on the horizontal axis (0.01–1 g/m²) can be considered broad but not unreasonable. The four lines in the figure represent the values of m_n and $\Delta\eta$ for which the photon upconversion layer and the silicon solar module break even regarding climate change impacts. To make the upconversion device preferable, roughly 2 percentage points improved efficiency per gram of NP is required in the PS. For the OS, an efficiency improvement of about 0.05% points/g of NP is sufficient. It can furthermore be seen in Figure 7 that given the same m_n , a slightly higher $\Delta\eta$ is required for PbS NPs compared to PbSe NPs in order to break even as per the slightly higher climate change impacts of PbS NPs (Figure 6). Observe also that I_s (in eqs 4 and 5) will likely be lower for future modules as solar module production improves, implying that a higher $\Delta\eta$ will be needed to break even.

Limitations and Further Studies. From a prospective perspective, there might be future changes in processes which are not accounted for in this study. For example, while distillation is the conventional choice for recovering organic solvents in the industry today, other separation processes might be used in the future, such as membrane filtration.^{65,66} Considering the high use of solvents in the NP syntheses, the influence of other separation processes constitutes an interesting aspect to investigate in future prospective LCA studies of the NPs. Future studies investigating alternative heat and materials production (e.g., bio-based instead of fossil) would also be interesting, considering the high use of fossil materials (especially in the PS) and natural gas-based heat (especially in the OS). In addition, the production and technical data for solar electricity supply is from the 2010's at best. This effectively means that we are investigating the break-even point between a future upconversion device and silicon solar modules as produced in the 2010's, only corrected for by assuming a higher module efficiency. Although beyond the scope of this study, considering future developments of solar module production would be an interesting elaboration in further studies.

As stated in the Methods section, all upconversion components besides the sensitizers (the NPs) are omitted from the break-even analysis because NPs are expected to constitute the largest share of future upconversion devices both in terms of mass and environmental impact. Regarding the exclusion of transmitters, a PbS or PbSe NP weighs more than 300 times as much as a transmitter molecule in some current upconversion devices, see Table S4 in the Supporting Information for a quantification. Since the number of transmitters per NP in such devices is preferably 5–30,²² the omission of transmitters is not likely to have a large influence on the results. Regarding the exclusion of annihilators, a PbS or PbSe NP weighs 160–180 times as much as the commonly

used annihilator rubrene (Table S4, Supporting Information). Although some current experimental upconversion devices use a large excess of annihilators,²² we anticipate that future upconversion devices will strive to reduce the amount of a liquid annihilator. Potential solvents in the upconversion device are excluded due to the relatively low environmental impact of solvents. For example, a commonly used solvent is toluene,^{22,36,54} which emits around 1.6 kg CO₂ equiv/kg during its production as per the ecoinvent database (version 3.6, cutoff). The NP with the lowest climate impact emits approximately 90 kg CO₂ equiv/kg, which means that the omission of solvents is likely to have a negligible influence on the results as long as the mass of the solvent in the device is less than about 50 times the mass of the NPs.

In addition, the end of life of solar modules equipped with upconversion layers and their components has not been considered in the break-even analysis. If upconversion devices will be mounted onto solar modules on a large scale, it would be important to also assess the difference in end-of-life scenarios between upconversion-enhanced solar modules and conventional ones, in particular due to the toxic nature of cadmium and lead.

CONCLUSIONS

Improving synthesis routes for quantum dot NPs, for example, by improving their overall atom economy and decreasing the waste-to-product ratio by reducing or recovering input materials, is the key to improve their climate performance as well as the climate performance of applications containing these ENMs. Given such improvements, the efficiency improvement of a silicon solar module with an upconversion layer required to break even with current silicon solar module electricity from a climate change perspective can be modest—only 0.05 percentage points/g of PbS or PbSe NPs in the most OS. A next step for developers of upconversion devices could be to monitor the amount of NPs applied per square meter (m_n) and the efficiency increased achieved ($\Delta\eta$) in order to compare with the break-even analysis results from this study. This would give them an early indication of whether their devices have the potential to be beneficial from a climate change point of view.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.1c00376>.

Proxy data, modeling of precursor materials, PWC data, estimation of NP surface concentration, additional cradle-to-gate results, unit process data tables, and a list of background system processes used (PDF)

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We would like to thank the Swedish Energy Agency for financially supporting this study (grant number 36436-2).

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