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Abstract:

The Life Cycle Assessment (LCA) methodology was applied to assess the environmental feasibility of a novel wastewater treatment technology based on oxygenic photogranules (OPG) biomass in comparison to a conventional activated sludge (CAS) system. LCA using laboratory scale experimental data allowed for eco-design of the process during the early stage of process development at laboratory scale. Electricity consumption related to artificial lighting, the fate of the generated biomass (renewable energy and replacement of mineral fertilizer), and the nitrogen flows in the OPG system were identified as major contributors to the potential environmental impact of the OPG treatment system. These factors require optimization in order to reduce the environmental impact of the overall OPG system. Nonetheless, the environmental impact of a non-optimized OPG scenario was generally lower than for a CAS reference system. With an optimization of the artificial lighting system, an energy neutral treatment system may be within reach.

Key words: oxygenic photogranules, life cycle assessment, cyanobacteria, anaerobic digestion, granular biomass

1 Introduction

Recently, the development of oxygenic photogranules (OPG) from activated sludge has been reported (e.g. Milferstedt et al., 2017b; Quijano et al., 2017). OPG are roughly spherical biological aggregates with a diameter of up to 5 mm (Abouhend et al., 2018). In OPG, the phototrophic biomass produces oxygen through photosynthesis and uses carbon dioxide for growth. Conversely, the heterotrophic biomass produces carbon dioxide and consumes the produced oxygen. The close vicinity of phototrophs and heterotrophs engaged in this syntrophy makes mass transfer more efficient than between dispersed cells found in high rate algal pond (Judd et al., 2015). It has been shown that OPG can perform simultaneous oxidation of organic carbon and nitrogen removal (Abouhend et al., 2018; Arcila & Buitrón, 2017; Arcila & Buitrón, 2016). A novel OPG-based wastewater treatment technology was proposed, considering carbon and nitrogen removal by OPG coupled to anaerobic digestion of harvested biomass for the production of renewable energy (Milferstedt et al., 2017b; Park & Dolan, 2015). Internally produced oxygen by OPG entirely replaces the energy intensive external supply of oxygen for wastewater treatment. External aeration in conventional wastewater treatment accounts for 45-75% of the overall electric energy consumption of the wastewater treatment plant (WWTP; or water resource recovery facility) (Longo et al., 2016). The overall biomass yield (per gram of removed organic carbon) of OPG treating wastewater is up to three times higher than for activated sludge (Abouhend et al., 2018). The biological methane potential (BMP) of the produced OPG is about 15-20% higher compared to activated sludge (Park et al., 2015), which is in line with other works on digestion of an algal-sludge biomass (Shoener et al., 2014; Ward et al., 2014). The differences in aeration requirements, sludge production and BMP, compared to activated sludge systems, may result in significant net energy savings when using the OPG process. However, a closer look at environmental performances of this innovative process combining OPG production and anaerobic digestion has not yet been done and therefore is required.

The aim of the present study was to evaluate the environmental performance of a putative OPG process compared to conventional technology. For this, Life Cycle Assessment (LCA) will be used, a standardized methodology (ISO 14040, 2006; ISO 14044, 2006) to assess potential environmental impacts considering the entire life cycle of products, processes or services based on the function they fulfill, in this case the treatment of municipal wastewater. A product, process or service is modeled along its life cycle (from raw material extraction to its end of life). Its impact is assessed with regard to different impact categories (e.g., global warming, ozone depletion, eutrophication, and acidification). LCA has been applied in the field of wastewater treatment to assess the environmental impact of both conventional activated sludge (CAS) wastewater treatment systems (e.g. Foley et al., 2010; Hospido et al., 2008), and advanced wastewater treatment technologies (e.g. advanced oxidation (Muñoz et al., 2006) and high rate algal ponds (Colzi Lopes et al., 2018)). While the use of LCA for assessing the environmental impact of existing wastewater treatment technologies increases, its use for the eco-design of emerging treatment processes under development is rare.

The present study aimed at evaluating the environmental performances of the novel OPG treatment system using LCA in order to determine whether the OPG system is a feasible technology from an environmental perspective in comparison to a CAS system. The study was based on a hypothetical model of an average, midsize treatment plant and data derived from laboratory scale process operation, having in mind that many technological problems are still unsolved before considering industrialization. The goal of the assessment at this early stage was to identify potential bottlenecks and critical parameters with high environmental impact. Future research can then target these areas to improve within an eco-design approach the process design prior to detailed process engineering studies. In particular, it was considered how far artificial lighting can be used for the OPG system without substantially deteriorating the environmental performance.

2 Materials and methods

2.1 LCA - Goal and scope

The goal of the LCA was to evaluate the environmental feasibility of the OPG system with regard to a reference CAS system and to assess the impact of different operating scenarios of the OPG system during early process development allowing the identification of design considerations with highest environmental impact. Those will then be targeted in future research to eco-design an up-scaled system. The system under study included in addition to the operation phase, the construction of the infrastructure and nutrient recycling from the sludge by agricultural use. Sludge utilization is considered an integral part of wastewater treatment and the basis of a water resource recovery facility.

The functional unit, based on which the processes are compared, is “the treatment of 1 m^3 of urban wastewater in an average, midsize WWTP”. Environmental impacts were computed in SimaPro (version 9.0.0.35, PRé Consultants) using the Ecoinvent database version 3.5 (Wernet et al., 2016) and the Environmental Footprint life cycle impact assessment method (Fazio et al., 2018).

A challenge when assessing the impact of a novel process is the availability of process operation data. Here, the evaluation of the OPG system was based on experimental data from the authors’ laboratory scale OPG experiments (Abouhend et al., 2018; Milferstedt et al., 2017b). These first experimental studies were mostly conducted to demonstrate a proof of concept and were not dedicated towards optimization and intensification of the process. A large margin of progress is to be expected in the future. Data from the authors’ own experimental studies were complemented with laboratory scale data from another research group working on municipal wastewater treatment using microalgae-bacteria aggregates (Arcila & Buitrón, 2017; Arcila & Buitrón, 2016) that resemble the OPGs used in the authors’ works. Nonetheless, several assumptions were made, that are laid out in detail later, regarding reactor configuration, operating conditions, and treatment performance in a full-scale implementation as the process currently exists only at the laboratory scale. Given the low technical readiness level of the innovative OPG process, some variables remain unknown and need to be estimated from sparse data. The present work, therefore, displays LCA results depending on the estimated value of these variables, such as energy balance and nitrogen flow through the process.

2.2 Life Cycle Inventory

The life cycle inventories for both OPG and CAS systems were estimated for an hypothetical average midsize WWTP (10,000 to 50,000 people equivalent), which is equivalent to a WWTP of the Swiss capacity class 3, inventoried in the Ecoinvent 3.5 database (Doka, 2009). A low loaded wastewater was considered here based on experiments carried out using primary effluent from a US WWTP (Abouhend et al., 2018). Its raw wastewater characteristics are presented in Table 1.

Table 1

Figure 1

<u>Reference system: Conventional activated sludge system</u>

The reference scenario for this comparative LCA is a conventional midsize wastewater treatment plant as described in Ecoinvent (Doka, 2009) and presented in Figure 1 a. Grit removal is followed by primary settlers (not shown in the figure). Organic carbon and nitrogen are removed in an activated sludge process. Phosphorus is removed by precipitation using $Fe(III)Cl_3$. The sludge is separated from the treated water in secondary settlers. Primary and secondary sludge are treated together by anaerobic digestion. The digester supernatant is returned to the influent of the WWTP. Digested and dewatered sludge is assumed to be spread on land as organic fertilizer.

The Ecoinvent datasets for WWTPs of different sizes are based on average treatment efficiencies of Swiss WWTPs dating from 1993-2002 (Doka, 2009) and do not reflect current treatment efficiencies. Consequently, only infrastructure data for the sewer grid and the WWTP were used from the dataset for the midsize treatment plant. Mass balances for chemical oxygen demand (COD), nitrogen, phosphorus, total suspended solids (TSS), and volatile suspended solids (VSS) for the CAS plant were calculated following textbook mass balance equations for biological wastewater treatment (Henze et al. 2008). A sludge retention time (SRT) of 15 days, a mixed liquor suspended solids (MLSS) concentration of 4 g/L, and a design wastewater temperature of 10°C were considered. N_2O emissions from wastewater treatment were estimated using an emission factor of 0.03 g N_2O-N/g $N_{denitrified}$ (Foley et al., 2010; Kampschreur et al., 2009). Treatment efficiencies for aerobic carbon oxidation, nitrification, and nitrogen removal calculated from mass balance equations were 90%, 84% and 70%, respectively. Phosphorus (P) not incorporated into biomass is precipitated, assuming that 2.5 g $Fe(III)Cl_3$/g $P_{removed}$ are needed and that 4.9 g $FePO_4$/g $P_{removed}$ are produced (Rittmann & McCarty, 2001).

Mass balances for COD, nitrogen, TSS, VSS, biogas and flow rates for the anaerobic digester were calculated using the steady state model for anaerobic digestion developed by Sötemann (2005), considering a hydraulic retention time of 10 days based on Ekama (2009). Kinetic parameters for anaerobic digestion of primary and secondary sludge were taken from Ekama (2009) and Sötemann (2005). BMPs of primary and secondary sludge vary widely (e.g. Mottet et al., 2010). For primary sludge, a methane production of 230 mL CH_4/g VSS was assumed based on a hydrolysis rate of 2 g COD/L/d (Sötemann, 2005). This is in the range of methane productions of primary sludge reported by Elbeshbishy et al. (2012). For secondary sludge, a methane production of 195 mL CH_4/g VSS was assumed (Park et al., 2015). A lower methane production of secondary sludge compared to primary sludge is in accordance with literature (e.g. Mahdy et al., 2015). A biogas composition of 65% CH_4 and 35% CO_2 was considered. All CH_4 was assumed to be oxidized to CO_2 during biogas combustion, producing 3 kWh electricity per m^3 CH_4 (Nowak, 2003). The solids concentration the digested and dewatered sludge was assumed to be 200 kg TSS/m^3.

A mean electricity consumption of the WWTP of 0.4 kWh/m^3 was considered based on average data for European WWTPs (Jonasson, 2007). Half of the overall electricity consumption was assumed to be spent for aeration (Longo et al., 2016). It was considered that electricity produced from biogas combustion is used onsite, covering a part of the needed electricity. For the remainder of needed electricity, an average European electricity mix was considered produced from fossil fuels (50%), nuclear power (27%), hydropower (17%), renewable sources and others (6%) (Itten et al., 2014).

Digested and dewatered sludge was assumed to be applied to farmland as organic fertilizer, replacing ammonium nitrate and triple superphosphate application at equivalent nutrient rates. The long-term nitrogen mineral fertilizer equivalent (MFE) of the sludge, nitrogen field emissions from sludge spreading, and avoided field emissions from mineral fertilizers were calculated following Brockmann et al. (2018), considering a nitrogen content of 9.7 kg N per m^3 sludge (6.4% total ammonia nitrogen (TAN) and 93.6% organic nitrogen (N_{org})) as calculated from the mass balance equations. For phosphorus, a MFE value of 0.95 was assumed. Phosphorus field emissions were estimated following Brockmann et al. (2014). Sludge application by broadcaster without incorporation and mean French soil and climate conditions were assumed. Tables 2 and 3 summarize characteristics of the CAS system and calculated emissions from wastewater treatment and sludge spreading. Parameters and emission factors used for calculating field emissions were taken from Brockmann et al. (2014) and Brockmann et al. (2018).

Differences in the oxygenic photogranule (OPG) system

The only difference between the considered reference CAS and the OPG plant layouts is the basin for secondary treatment and the absence of secondary settlers (Figure 1 b). In the OPG system, secondary treatment took place in sequencing batch reactors (SBR). As settling of the OPG was carried out in the SBRs during a settling phase, secondary settlers were not needed. The model of the full-scale OPG system was based on data from the authors' laboratory scale OPG studies (Abouhend et al., 2018; Milferstedt et al., 2017b) complemented with data from laboratory scale experiments with microalgae-bacteria aggregates treating municipal wastewater (Arcila & Buitrón, 2017; Arcila & Buitrón, 2016). Based on the authors' laboratory scale OPG experiments, operation in

SBRs at a hydraulic retention time (HRT) of 0.5 days and a MLSS concentration of 4 g/L was assumed. For these operating conditions, the overall SBR reactor volume needed is similar to that of the activated sludge reactor plus the secondary settlers. Therefore, the same environmental impact for the required infrastructure was assumed for both systems.

Sludge production was estimated based on a biomass yield of 0.7 g VSS/g $COD_{consumed}$ (1.26 g COD/g $COD_{consumed}$) and a COD/VSS ratio of the OPG of 1.8 g COD/g VSS (Abouhend et al., 2018). An average organic carbon removal efficiency of 85% was considered (Abouhend et al., 2018; Arcila & Buitrón, 2016), assuming that 50% of the removed organic carbon was incorporated into the heterotrophic part of OPG biomass. The remaining 50% of the removed organic carbon were assumed to be oxidized and then available for phototrophic uptake. An average ammonia transformation efficiency of 90% was considered (Abouhend et al., 2018), including nitrogen incorporation into biomass and nitrification. Nitrogen incorporation into biomass was estimated using a nitrogen content of the OPG of 10% (as for CAS), resulting in 54% of the organic and ammonia nitrogen being incorporated into OPG (vs. 22% calculated for CAS). The remaining unaccounted transformed ammonia was assumed to be nitrified. Based on this assumption, 33% of the NH_4-N transformed was converted to NO_3-N, which was below an observed NO_3-N production rate of 50% (Abouhend et al., 2018; Arcila & Buitrón, 2016), but of the same order of magnitude as reported by Arcila and Buitrón (2017). Denitrification by OPG has, so far, not been reported, but the presence of genes (Stauch-White et al., 2017) and of 16S rRNA bacterial sequences (Milferstedt et al., 2017b) associated with denitrification was revealed. It was assumed that the OPG system includes a denitrification step with a denitrification efficiency similar to the reference system (80%). Overall, a total nitrogen removal efficiency of 71% was obtained based on mass balances, which was equivalent to the CAS system. A methane production of 290 mL CH_4/g VSS for OPG (Arcila & Buitrón, 2016) and a biogas composition of 65% CH_4 and 35% CO_2 were considered.

In contrast to the CAS system, the OPG system does not require aeration since OPG produces oxygen under light. Based on laboratory scale experiments with 3.5 hours illumination per 6 hours cycle (Milferstedt et al., 2017b), illumination during 14 hours for the four daily cycles is considered. With an oxygen production rate of the OPG under light of 12.6 mg O_2/(g VSS·h) (Abouhend et al., 2018), oxygen production by OPG with 14 h/day illumination exceeds the oxygen requirements for organic carbon oxidation and nitrification. To ensure adequate suspension of OPG, mechanical mixing is needed. Electricity consumption for mixing of OPG was estimated following Walas (1990) considering a turbulent regime and a reactor geometry similar to that of activated sludge reactors. Considering an anchor or gate paddle per reactor and a mixing time of 22 h/day, mixing of OPG consumes 26 Wh/m^3 electricity. Electricity consumption for lighting was estimated based on a photosynthetically active radiation (PAR) of 150 μmol/m^2/s (Abouhend et al., 2018) and a PAR efficiency of 2 μmol/s/W (Blanken et al., 2013), yielding an electricity consumption of 75 W/m^2. The enlightened surface area was assumed twice the footprint of the reactors without further detailed design of the artificial lighting system, resulting in an electricity consumption of 134 Wh/m^3 wastewater. This way of operation represents a generic, non-optimized solution. An electricity consumption for pumping, valves, sensors etc. of 200 Wh/m^3 was estimated based on the electricity consumption of the CAS system without aeration. Thus, the overall electricity consumption of the OPG system consists of 200 Wh/m^3 for pumping, valves etc., 26 Wh/m^3 for 22 hours of mixing, and 134 Wh/m^3 for lighting.

For land application of the OPG, a long-term MFE of 0.562 was calculated, based on a nitrogen content of 13.4 kg N/m^3 OPG (15.4% TAN and 84.6% N_{org}), as calculated from mass balance equations.

Table 2

Table 3

Operating scenarios

OPG is exposed to sunlight during the day, enabling oxygen production by photosynthesis. For providing oxygen during night-time, when OPG is not exposed to sunlight and thus not photosynthetically active, different operating scenarios were considered:

- Scenario 1: Artificial lighting is provided during the night. Considering overall 14 h/day of illumination and assuming that natural light covers on average 7 h/day, the same duration of artificial lighting is needed.
- Scenario 2: Aeration replaces the 7 h/day of artificial lighting. Thus, aeration is needed for 7 h/day and mixing for 15 h/day.
- Scenario 3: OPG is not exposed to sunlight at all and artificial lighting is provided 14 h/day.
- Scenario 4: As scenario 1 but with a 50% reduction in light energy provided.

Scenario 1 was used as the default scenario for the OPG system. The impact of the other operating scenarios was assessed in a sensitivity analysis.

3 Results and discussion

3.1 Comparison of the two treatment systems

The environmental impact of treating 1 m^3 municipal wastewater and applying the resulting sludge on farm land was compared using a CAS system and the OPG system (scenario 1). A graphical representation of this comparison is shown as radar plot in

Figure 2 for all evaluated impact categories. The black line signifies the CAS system as reference, and the blue line the OPG system. For most impact categories, the OPG system's environmental impact is inferior of the CAS impact ranging from a 4% difference for freshwater eutrophication to 61% for ionizing radiation. The two notable exceptions are for the impact categories terrestrial eutrophication and acidification being 2 and 3 times higher, respectively, compared to the CAS system.

Figure 2

In Figure 3, the environmental impacts in the various impact categories are related to where they are generated, i.e., treatment infrastructure, emissions from treatment, electricity consumption, Fe(III)Cl_3 use for precipitation, sludge spreading and the substitution of mineral fertilizer. Contributions that are identical for both systems are not displayed even though considered in the overall calculations (e.g., sewer grid infrastructure, emissions from overload discharge, grit removal, and treatment plant infrastructure). It should be noted that sewer grid and treatment plant infrastructure are major contributors to the environmental impact. But in this comparison, this impact is not considered relevant as it is assumed to be independent of the biological configuration of the treatment plant. Figure 3 reveals that the significantly higher impacts of the OPG system in the impact categories acidification, terrestrial eutrophication, and respiratory inorganics can be considered a mass effect, as significantly larger quantities of digested sludge are land-applied. The impact is not specific to OPG. In particular, most of the increased impact on terrestrial eutrophication, acidification, and respiratory inorganics from sludge spreading is caused by higher ammonia emissions from land application. Ammonia emissions from sludge spreading contributed 114%, 107%, and 64%, respectively, to the mentioned impact categories. Twice as much nitrogen was recovered in the digested OPG and spread on land than for the CAS system (203 kg N/d vs. 110 kg N/d). The amount of nitrogen emissions from land application does not only depend on the amount of nitrogen applied, but also on the method of sludge application. Surface application of sludge without subsequent incorporation into the soil is the worst case with respect to ammonia emissions. The significantly lower impact of the OPG system on ionizing radiation is due to a much lower electricity consumption from the grid compared to the CAS system. The OPG system has an overall electricity consumption of 359 Wh/m^3, of which 269 Wh/m^3 are covered by combustion of the produced biogas and 90 Wh/m^3 by electricity from the grid. In contrast, to cover the overall electricity consumption of the CAS system of 400 Wh/m^3, 263 Wh/m^3 are needed from the grid as only 137 Wh/m^3 are produced from biogas combustion. Lower impacts on ozone depletion, photochemical ozone formation, human health effects, freshwater ecotoxicity, and resource use are due to lower electricity consumption and higher amounts of mineral fertilizers replaced by land-applied digested biomass. The reduced impact on climate change, compared to the CAS system, result from lower N_2O emissions from wastewater treatment and lower electricity consumption of the OPG system. As for the CAS system, N_2O emissions from wastewater treatment with OPG were estimated based on the amount of nitrogen denitrified and may change using real data. Nonetheless, climate change impacts of the OPG system remain inferior to the ones of the CAS system even when the contribution of N_2O emissions is excluded.

Figure 3

3.2 Effects of alternative illumination scenarios on the environmental impact of an OPG treatment plant

The default OPG system (scenario 1) operated with 7 h/day artificial lighting, even in its immature design state, outperforms the CAS system. In the following, the sensitivity of the LCA results is assessed using three envisioned operating scenarios, which affect the electricity consumption of the OPG treatment plant: 7 h/day aeration (scenario 2), 14 h/day artificial lighting (scenario 3) and a modified scenario 1 with a 50% cut-off in light energy provided to the system (scenario 4). It is acknowledged that scenarios 2 and 4 could affect the stability and the performance of the OPG system. Despite these potentially important unknowns, the suggested scenarios allow to assess whether these or similar approaches are environmentally feasible, before even starting the appropriate experiments. The LCA methodology should be considered here a modeling approach that evaluates the environmental impact of putative potential alternatives. For this evaluation, it was assumed that operating conditions for scenarios 3 and 4 impact only electricity consumption, while biochemical conversion rates and overall treatment plant performance remained unchanged. For scenario 2 (7 h/day aeration), the phototrophic biomass yield was reduced, assuming that with only half of the light, half of the phototrophic growth will occur. The reduced phototrophic biomass yield entails lower biomass and, thus, biogas and electricity production. Replacing artificial lighting during night by aeration reduced electricity consumption by about 25% and electricity production by about 15%, requiring only 48 Wh/m^3 from the grid. It may be surprising that the partially aerated system is less energy consuming than the fully photosynthetic system. This is possibly because of the substantial increase of energy recovered from the digested phototrophic sludge. A lower biomass production also implicates lower ammonia emissions from field application of the digested biomass, and lower amounts of mineral fertilizers replaced. Figure 4 shows that the reduced electricity consumption from the grid decreased environmental impacts of the OPG system on ionizing radiation by 35% and on resource use of energy carriers by 11%, compared to scenario 1. Furthermore, the reduced amount of land-applied digested OPG reduced environmental impacts on acidification and terrestrial eutrophication by 24% and on respiratory inorganics by 13%, but increased the environmental impact on resource use of minerals and metals by 14%, compared to scenario 1. Impacts in other impact categories changed by less than 7%. For scenario 3, where artificial lighting needed to be provided 14 h/day, electricity consumption from the grid more than doubled compared to scenario 1, but remained below the electricity consumption from the grid of the CAS system. Due to increased electricity consumption from the grid, environmental impacts on ionizing radiation doubled and increased by 46% for resource use of energy carriers and land use, and by 31% ozone depletion (Figure 4, orange line). Impacts on other impact categories increased by 1 to 17%.

The artificial lighting system considered here represents a largely non-optimized solution. The efficiency of LED lighting is drastically developing and will likely improve significantly over the next years (Zhang et al., 2018), reducing the energy requirements to yield a given photoactive radiation. This kind of development is not expected for the aeration in the CAS system, which is already a mature technology. In addition, the use of white light LEDs was assumed here of which a considerable bandwidth is not suitable for photosynthesis. The use of LEDs with adapted spectra for phototrophic light alone could yield significant energy savings at a similar biological activity (Abomohra et al., 2019). In addition, the use of suspended, free-moving LED (Murray et al., 2017) may further reduce energy consumption for artificial lighting. Therefore, an optimized artificial lighting system with an assumed improved energy efficiency of LED lighting of 50% (scenario 4) was evaluated here and compared to scenario 1. All other parameters of scenario 1 were kept. With optimized artificial lighting, the electricity consumption of scenario 1 decreased to 293 Wh/m^3, requiring only 23 Wh/m^3 from the grid. Thus, with an optimized artificial lighting system, an energy neutral treatment system may be within reach.

Figure 4

3.3 Operation of the OPG system at higher biomass concentrations

The presented impact assessment was computed for operating the OPG system at the same biomass concentration as the laboratory scale reactor (4 g/L). This is a typical biomass concentration for CAS systems and a relatively low MLSS concentration compared to other granular biomass systems (Milferstedt et al., 2017a). For example, aerobic granular sludge systems can be operated at biomass concentrations up to 10 g/L (Keller & Giesen, 2010). It is worthwhile investigating the OPG treatment performance at higher biomass concentrations and lower HRT, while maintaining the SRT in the system, to reduce the overall reactor volume of the treatment system. This will decrease treatment plant infrastructure needs, which significantly contribute to environmental impacts on land use, human health effects, freshwater ecotoxicity, resource use, photochemical ozone formation, and respiratory inorganics. Assuming operation of the OPG system at a biomass concentration of 6 g/L and keeping the sludge retention time unchanged, reduces the HRT to 0.33 days and the required reactor volume by 30%. Energy requirements for mixing need to be considered in a putative OPG system. Several alternative modes of mixing could be envisioned, e.g., intermittent gas sparging or mixing by pulse-like waves (e.g., oloid.ch). Here, a traditional mixing approach using constantly turning impellers was considered. Energy needs for mixing will decrease with reduced reactor diameter, as the impeller diameter decreases, but energy requirements for artificial light supply may increase with a higher biomass concentration. It is assumed here that the potentially higher energy requirements for artificial light supply compensate the energy savings from reduced energy needs for mixing. This means that the reduction of the reactor volume does not significantly affect the overall electricity consumption of the OPG system. It is further considered that operation at higher biomass concentration and shorter HRT does not result in a considerable loss of treatment performance. Based on the aforementioned assumptions, the reduction of the required reactor volume decreases the environmental footprint with regard to resource use of minerals and metals by 50%, to land use by 37%, to ozone depletion and non-cancer human health effects by 17%, and to photochemical ozone formation by 16%. Reductions on other impact categories range from 0.7 to 13%. These results are based on assumptions that still need to be validated experimentally, but show the interest (from an environmental perspective) of a more compact treatment system.

3.4 Nitrogen flows in the OPG system

In the LCA model, nitrogen emissions in the forms of ammonia, nitrite/nitrate, and organic nitrogen from wastewater treatment with OPG were on the same order of magnitude as for the reference CAS system. N_2O emissions from biological treatment, estimated based on the amount of nitrogen denitrified, were lower for the OPG system, because more nitrogen was incorporated into biomass, requiring less denitrification. The same emission factor for estimating N_2O emissions from a wastewater treatment with OPG and CAS system was used. The factor is based on Foley et al. (2010) and Kampschreur et al. (2009) and remains to be estimated using experimental data from the OPG system. It was demonstrated that major nitrogen emissions occurred also downstream of the treatment plant, at the stage of land application of the digested biomass (Figure 3), affecting largely the impact categories terrestrial eutrophication, acidification, and respiratory inorganics. This was caused by significantly higher biomass produced by the OPG system, resulting in twice as much nitrogen spread on land with the digested OPG than with the activated sludge produced by the CAS system. Consequently, significantly higher nitrogen field emissions for the same volume of wastewater treated can be expected (23 kg NH_3/d for OPG system vs. 5.1 kg NH_3/d for CAS system). Optimization of the environmental impact of the assessed OPG system should, therefore, also consider the fate of the generated biomass in applications downstream of the treatment plant. The observed large contribution of land application of the digested biomass to the environmental burdens of the treatment systems is in line with other works considering land application of sludge and replacement of mineral fertilizers (e.g. Brockmann et al., 2014; Pasqualino et al., 2009).

In the default scenario, land application of digested sludge by a broadcaster, without incorporation into the soil was assumed. This is common agricultural practice (Loyon, 2018) and known to be the worst case scenario with regard to ammonia field emissions (Bittman et al., 2014). Land application of digested biomass by deep injection, considered the best case scenario with regard to ammonia field emissions (Bittman et al., 2014), significantly reduced ammonia field emissions in the LCA model from 1504 g NH_3/m^3 sludge to 301 g NH_3/m^3 sludge for OPG, and from 452 g NH_3/m^3 sludge to 91 g NH_3/m^3 sludge for activated sludge. The decreased ammonia field emissions yielded in larger amounts of ammonia nitrogen available to the plants, increasing the nitrogen MFE values for OPG (0.64 vs. 0.56) and CAS (0.62 vs. 0.59). Consequently, more mineral fertilizers could be replaced by the digested biomass (10.2 kg N/m^3 sludge vs. 9 kg N/m^3 sludge from OPG, 7.2 kg N/m^3 sludge vs. 6.9 kg N/m^3 sludge from CAS). The effect of agricultural practice is illustrated in Figure 5. Optimizing the land application of digested biomass from broadcaster application to deep injection significantly decreased the environmental impact in three categories: terrestrial eutrophication, acidification, and respiratory inorganics. The double effect of reducing ammonia emissions from land application of digested biomass and increasing the amount of replaced mineral fertilizers and associated emissions yields in a lower environmental footprint of the OPG system for all 16 impact categories. Thus, in addition to modifications at the treatment plant, the fate of the excess sludge is a determining factor for the environmental impact of the OPG process, and as well for

CAS. Particularly, changes of the sludge disposal method can considerably drive the environmental impact and must be considered already in the process design.

Figure 5

3.5 Use of produced OPG biomass

For a water resource recovery facility, the produced biomass is a resource that can be transformed into bioenergy and other bioproducts. Even though the high yield in an OPG system and the high degree of uniform phototrophic biomass may make other transformations and valorization ways possible, a 'classical' transformation of the biomass into energy and organic fertilizer by anaerobic digestion was considered in this study. Considering the same biomass transformation for the OPG and the reference CAS system made it possible to show that wastewater treatment using the evaluated OPG system allows for higher energy and nitrogen recovery than the CAS system. The OPG system produced twice as much methane per m^3 of treated wastewater as the CAS system (0.090 vs. 0.046 Nm^3 CH_4/m^3 wastewater) and replaced 75% more mineral nitrogen fertilizer (per m^3 wastewater treated) through agricultural use of the digested biomass. This 'base' case of agricultural use was considered because it is current common agricultural practice in France, Spain, and Ireland (European Commission, 2019a) and commonly used when assessing the environmental impact of wastewater treatment (Foley et al., 2010; Hospido et al., 2008). The land application of digested sludge is probably the worst case with regard to the environmental impact/benefit of the biomass use. In addition, land application of digested sewage sludge will be more restricted in the EU due to more stringent limits for contaminants following the new Fertilizing Products Regulation of the EU (European Commission, 2019b). Electricity generation from biogas is only one potential use of the biogas. Other uses such as upgrading to biomethane as a substitute for natural gas or as a raw material to produce platform chemicals (Tsui & Wong, 2019) should be considered as well in the future.

The objective of this LCA was to guide research efforts to eco-design and improve the sustainability of the OPG process. It is therefore wise to envision alternative uses of the produced OPG, possibly even resulting in larger environmental benefits. One area of active research is the downstream use of extracellular polymeric substances from granular sludge (Quijano et al., 2017; van Loosdrecht & Brdjanovic, 2014). OPG may be a suitable candidate for this approach, as these granules are composed of a mat-like outer layer densely populated by filamentous cyanobacteria (Milferstedt et al., 2017b). This layer contains large amounts of extractible extracellular polymeric substance (EPS) produced by cyanobacteria (e.g. Ansari et al., 2019; Milferstedt et al., 2017b). The extracted EPS may serve, depending on the physico-chemical properties (e.g., molecular weight, charge, hydrophobicity) as hydrogels, biosurfactants or bioflocculants, as demonstrated in part already for aerobic granules (Lin et al., 2015). It is of great advantage that through the syntrophy between heterotrophs and phototrophs, heterotrophically produced CO_2 is apparently immediately fixed in OPG by the growing phototrophic biomass. Resource recovery and conservation of fixed nitrogen are intrinsic feature of this biomass and must be taken advantage of.

3.6 Research perspectives

Two major groups of bottlenecks in the development of a potential OPG-based bioprocess have been identified in the analysis: reducing the environmental costs of providing light to OPGs and increasing the environmental benefit of generating OPG biomass.

Light dependencies touch reactor configuration as much as they potentially affect the biological activity of the phototrophic biomass. Research on the activity of OPGs as a function of lighting, possibly even at the scale of individual photogranules, must be coupled to identifying actual lighting conditions in current or prospective bioreactor designs. This research must also consider effects of biomass density and mixing to successfully narrow down process engineering constraints.

While coupling of microbiological and engineering research has been successfully done for years, biomass valorization requires the establishment of new collaborations. Novel valorization approaches will be a compromise between constraints from wastewater treatment (e.g., treatment efficiency) and the target quality and quantity of potential bio-products. Developing a value chain that meets these requirements can only be done through the commitment of public and private entities across sectoral borders and disciplines.

4 Conclusions

The environmental impact of a novel OPG-based treatment process was evaluated using LCA and compared with the well-established conventional activated sludge (CAS) process. The environmental impact of a non-optimized OPG scenario was generally lower than for the reference CAS system. Electricity consumption related to artificial lighting, the fate of the generated biomass (renewable energy and replacement of mineral fertilizer), and the nitrogen flows in the OPG system were identified as the major contributors to the potential environmental impact of the OPG treatment system. With an optimized artificial lighting system, an energy neutral treatment system is within reach.

5 Acknowledgements

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7 Figure captions

Figure 1: Process schemes of the two compared wastewater treatment systems. a) Conventional activated sludge system (CAS), and b) oxygenic photogranules (OPG) system. In b), only changes with respect to a) are highlighted. Upstream unit processes identical for both processes (overload discharge, grit removal, and primary settling) are omitted from the figure for clarity. Line colors correspond to sludge (brown), gas and energy (green) and liquid phase (blue).

Figure 2: Comparison of environmental impacts of the treatment of 1 m^3 urban wastewater by the OPG system (scenario 1) and the CAS system as reference. Black: CAS system, blue: OPG system. All calculated environmental impacts were normalized by the impact obtained for the reference CAS system. Distances between circles are log2 scaled.

Figure 3: Environmental impacts by impact category and differentiated by their origin (colored stacks) for the treatment of 1 m^3 municipal wastewater in the OPG system (scenario 1) and the reference CAS system. The units of the different impact categories are given in the panel headers.

Figure 4: Environmental impact of different operating scenarios for the OPG system. Scenario 1 (as reference): 7 h/day artificial light (solid blue line). Scenario 2: 7 h/day aeration instead of lighting (short yellow dashes). Scenario 3: 14 h/day artificial lighting (orange dots). Scenario 4 (light-optimized scenario 1): 7 h/day artificial lighting with twice more efficient lighting system (dash-dots).

Figure 5: Effects of agricultural practice on the environmental impact of the OPG (scenario 1) and the reference CAS systems. Black, solid: default CAS system, including surface spreading of sludge; blue, solid: OPG system (scenario 1), including surface spreading of sludge; Black, dashed: CAS system with deep injection of digested biomass; blue, dash-dots: OPG system with deep injection of digested biomass. For each impact category, calculated environmental impacts were normalized by the impact obtained for the reference CAS system. The distance between circles is log2-scaled.

8 Tables

Table 1: Raw municipal wastewater characteristics (US)

Parameter	**Symbol**	**Unit**	**Values**
Flow rate	Q_{in}	m^3/d	15000
Total chemical oxygen demand	Total COD	g COD/m^3	500
Soluble chemical oxygen demand	Soluble COD	g COD/m^3	200
Total Kjeldahl Nitrogen ($N_{org.}$ + NH_4-N)	TKN	g N/m^3	30
Ammonia nitrogen	NH_4-N	g N/m^3	20
Nitrate nitrogen	NO_3-N	g N/m^3	0
Total phosphorus	Total P	g P/m^3	6
Orthophosphate	Ortho-P	g P/m^3	4
Total suspended solids	TSS	g TSS/m^3	250
Volatile suspended solids	VSS	g TSS/m^3	-

Table 2: Characteristics of the reference CAS system and the OPG system: Consumptions and productions, digested sludge characteristics, and avoided mineral fertilizer as calculated from mass balances. Units are per m^3 wastewater unless otherwise stated

	Unit	CAS	OPG
Consumptions			
Electricity	kWh/m^3	0.4	0.2+mixing+light$^{(*)}$
Iron(III)chloride	g/m^3	19.4	8.7
Productions			
Primary sludge	kg VSS/d	2184	2184
Secondary sludge	kg VSS/d	962	2916
Methane	Nm^3 CH_4/m^3	0.046	0.090
Electricity from CH_4 combustion	kWh/m^3	0.137	0.269
Digested sludge characteristics			
Dewatered sludge	m^3/d	11.3	15.2
TSS content of sludge	kg TSS/m^3_{sludge}	200	200
N content of sludge	kg N/m^3_{sludge}	9.7	13.4
TAN/N_{tot}	%	6.4	15.4
N_{org}/N_{tot}	%	93.6	84.6
P content of sludge	kg P/m^3_{sludge}	5.6	4.2
Nitrogen MFE (long-term)	-	0.594	0.562
Phosphorus MFE	-	0.95	0.95
Avoided mineral fertilizers			
Ammonium nitrate	kg N/m^3_{sludge}	6.9	9.0
Triple superphosphate	kg P_2O_5/m^3_{sludge}	24.2	17.1

$^{(*)}$ Mixing (22h/d): 0.026 kWh/m^3; light (7h/d): 0.134 kWh/m^3

Table 3: Emissions from wastewater treatment and sludge spreading, and avoided emissions from mineral fertilizers as calculated from mass balances. Units are per m^3 wastewater for wastewater treatment and per m^3 sludge for sludge spreading.

	Unit	CAS	OPG
Effluent characteristics			
COD	g COD/m^3	31.3	48.3
TKN	g N/m^3	3.8	4.6
NH_4-N	g N/m^3	1.2	2.0
NO_3-N	g N/m^3	3.7	2.3
Total P	g P/m^3	1.8	1.8
Emissions from wastewater treatment			
N_2O-N	g N/m^3	0.44	0.29
Biogenic CO_2 from carbon removal	g CO_2/m^3	283	202
Biogenic CO_2 from AD	g CO_2/m^3	45	89
Biogenic CO_2 from CH_4 combustion	g CO_2/m^3	84	165
Emissions from sludge spreading			
NH_3	g/m^3_{sludge}	452	1504
N_2O	g/m^3_{sludge}	173	250
NO_x	g/m^3_{sludge}	243	316
NO_3^-	g/m^3_{sludge}	4764	6581
P	g/m^3_{sludge}	47	33
Avoided emissions from mineral fertilizers			
NH_3	g/m^3_{sludge}	255	333
N_2O	g/m^3_{sludge}	122	159
NO_x	g/m^3_{sludge}	174	227
NO_3^-	g/m^3_{sludge}	3383.6	4424.8
P	g/m^3_{sludge}	11.6	8.2

9 Figures

Figure 1:

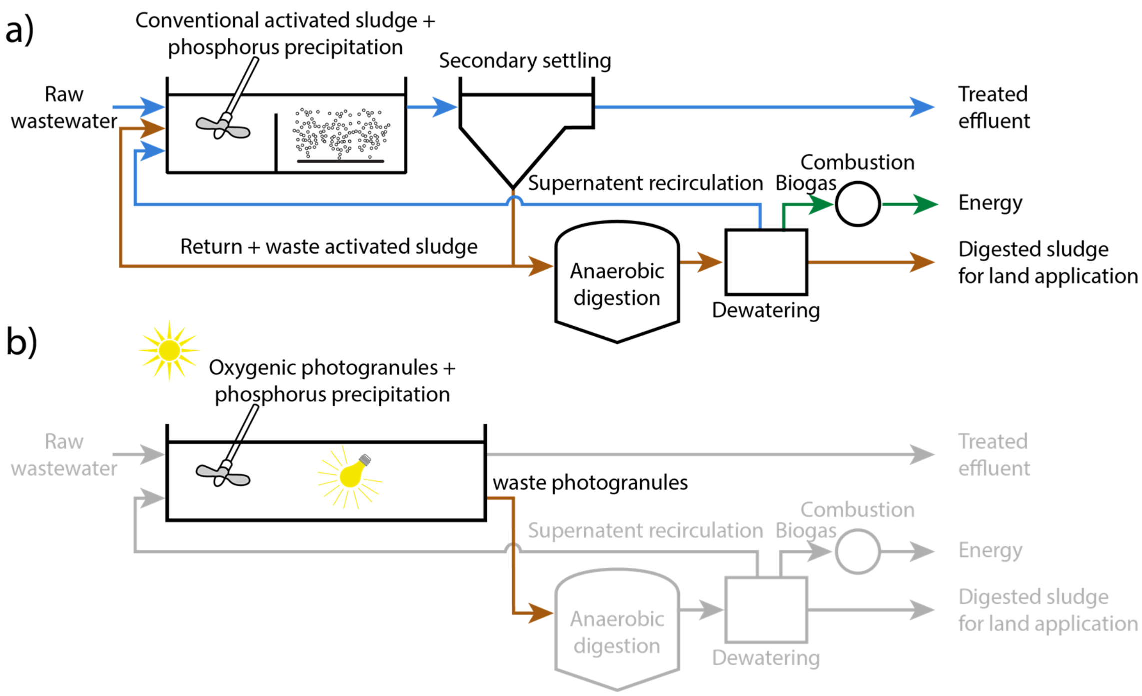
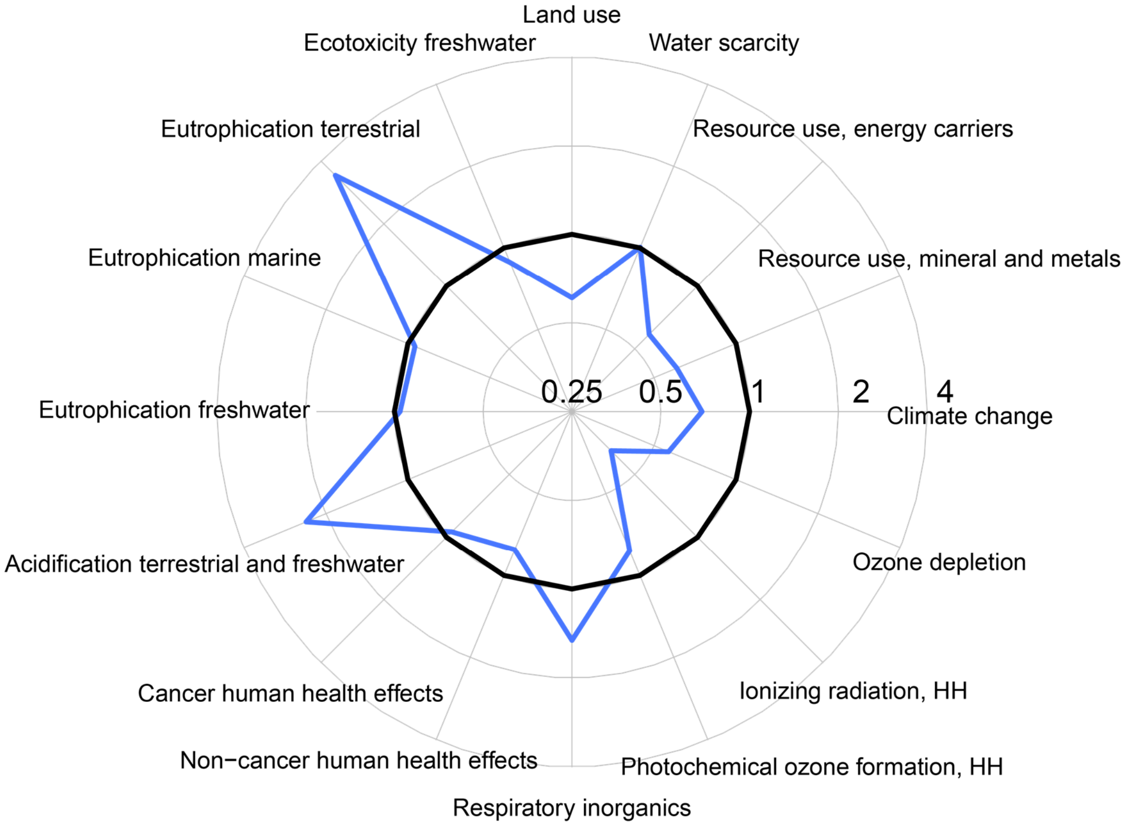


Figure 2:



Land use
Water scarcity
Resource use, energy carriers
Resource use, mineral and metals
Climate change
Ozone depletion
Ionizing radiation, HH
Photochemical ozone formation, HH
Respiratory inorganics
Non−cancer human health effects
Cancer human health effects
Acidification terrestrial and freshwater
Eutrophication freshwater
Eutrophication marine
Eutrophication terrestrial
Ecotoxicity freshwater
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0.5
1
2
4

Figure 3:

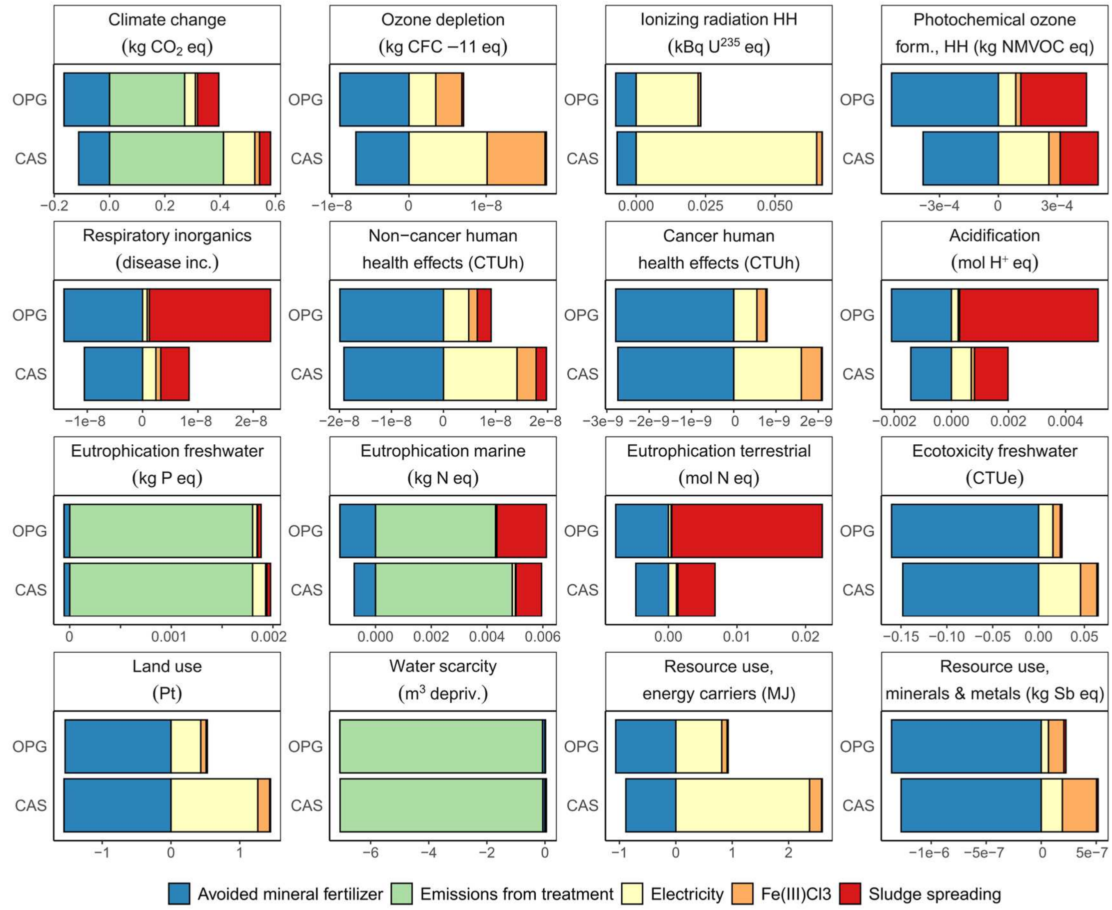
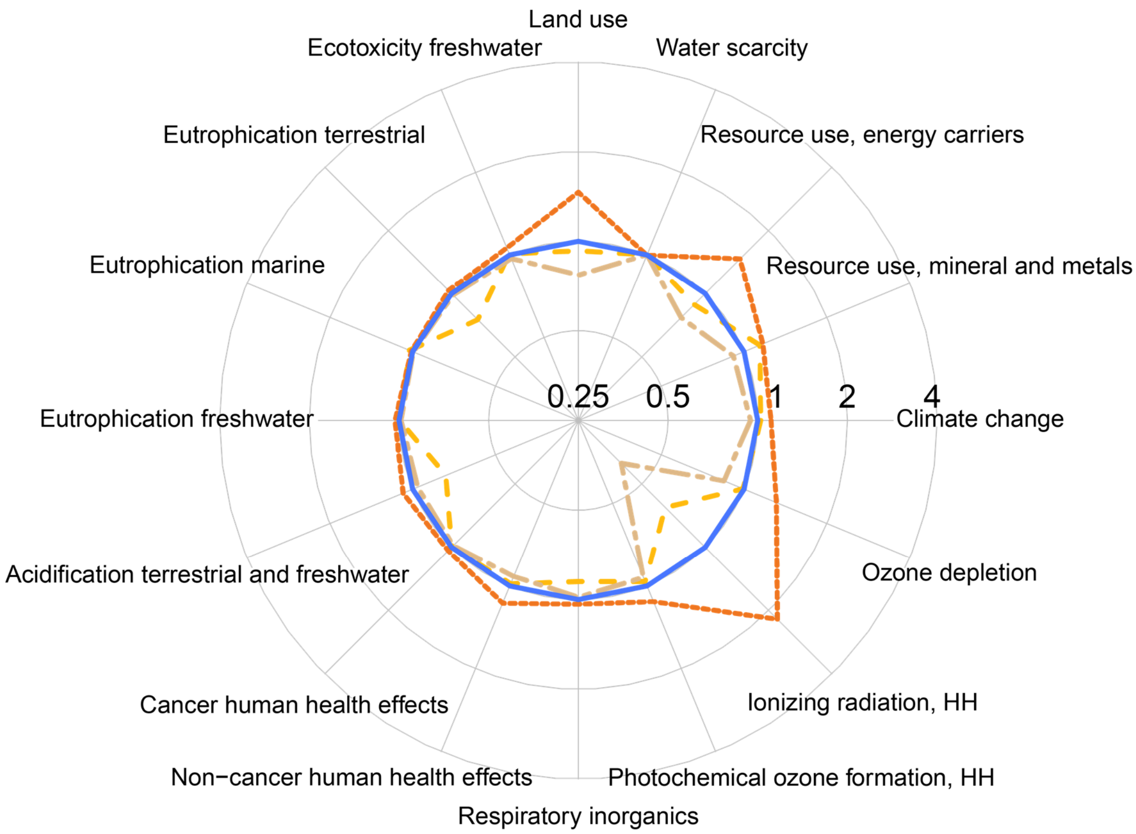

Climate change
(kg CO2 eq)
Ozone depletion
(kg CFC −11 eq)
Ionizing radiation HH
(kBq U235 eq)
Photochemical ozone
form., HH (kg NMVOC eq)
Respiratory inorganics
(disease inc.)
Non−cancer human
health effects (CTUh)
Cancer human
health effects (CTUh)
Acidification
(mol H+ eq)
Eutrophication freshwater
(kg P eq)
Eutrophication marine
(kg N eq)
Eutrophication terrestrial
(mol N eq)
Ecotoxicity freshwater
(CTUe)
Land use
(Pt)
Water scarcity
(m3 depriv.)
Resource use,
energy carriers (MJ)
Resource use,
minerals & metals (kg Sb eq)
OPG
CAS
Avoided mineral fertilizer
Emissions from treatment
Electricity
Fe(III)Cl3
Sludge spreading

Figure 4:


Land use
Ecotoxicity freshwater
Water scarcity
Eutrophication terrestrial
Resource use, energy carriers
Eutrophication marine
Resource use, mineral and metals
Eutrophication freshwater
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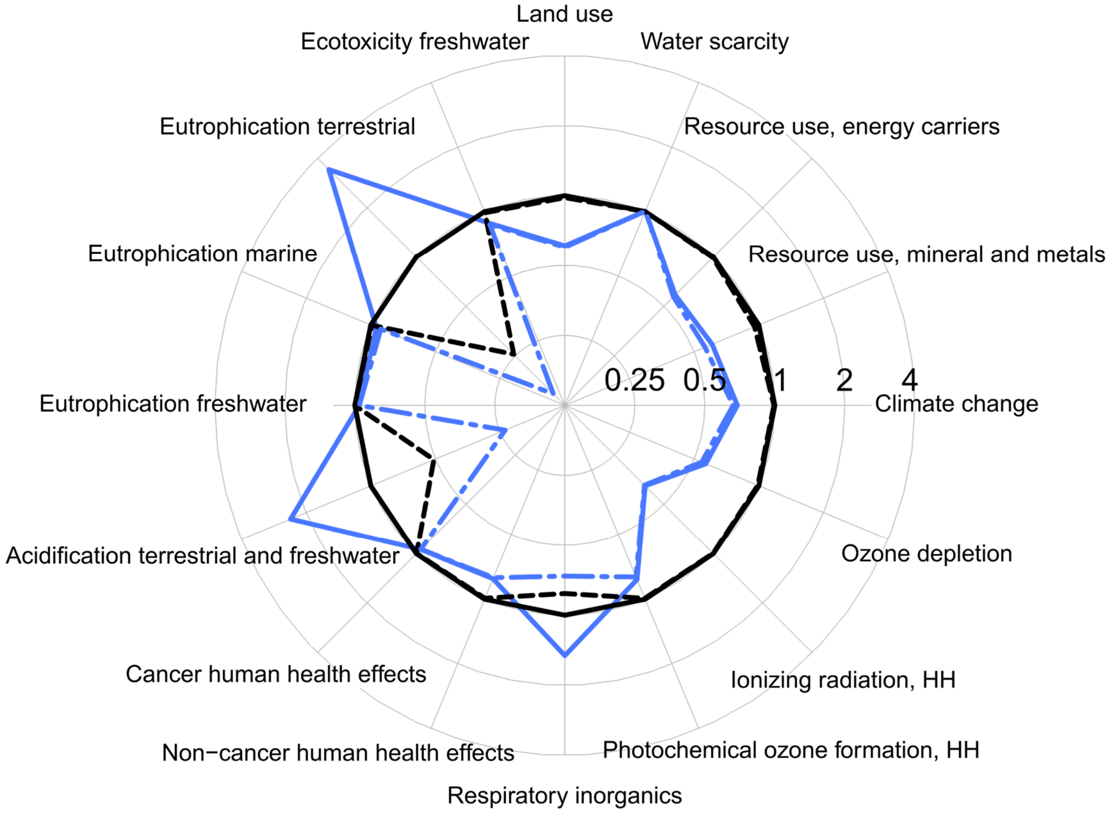
Climate change
Acidification terrestrial and freshwater
Ozone depletion
Cancer human health effects
Ionizing radiation, HH
Non−cancer human health effects
Photochemical ozone formation, HH
Respiratory inorganics

Figure 5:


Land use
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Resource use, energy carriers
Resource use, mineral and metals
Climate change
Ozone depletion
Ionizing radiation, HH
Photochemical ozone formation, HH
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