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To cite this article: Ben Joseph *et al* 2020 *Environ. Res. Lett.* **15** 064023

Environmental Research Letters

OPEN ACCESS

RECEIVED
29 January 2020

REVISED
15 March 2020

ACCEPTED FOR PUBLICATION
25 March 2020

PUBLISHED
29 May 2020

LETTER

Sustainability assessment of activated carbon from residual biomass used for micropollutant removal at a full-scale wastewater treatment plant

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Keywords: life cycle assessment (LCA), biochar, social risk assessment, global warming potential, sustainable resource management

Supplementary material for this article is available online

Abstract

Activated carbon (AC), used for removal of organic micropollutants in European wastewater treatment plants (WWTPs), is usually produced from non-renewable resources that need to be transported over long distances. Utilising local residual biomass as a raw material may be advantageous in terms of sustainability. This study investigated the environmental and energy balances of using biowaste and biomass from landscape management for micropollutant removal at a commercial scale WWTP. Both residual biomasses were processed using the integrated generation of solid fuel and biogas from biomass (IFBB) technique to obtain a press cake that was used as feedstock for AC production. The results showed a lower global warming potential (GWP) and cumulative energy demand in comparison to a fossil-based conventional AC Differences in GWP between residual and fossil ACs were enhanced when the end-of-life incineration step was considered, and residual AC had a lower social risk associated with its production. Energy efficiency of AC production was substantially increased by utilising waste heat generated in the pyrolysis process of biochar production and by using electricity generated in a combined heat and power plant using biogas from the methanation of IFBB press fluids. Converting residual biomass into activated carbon using IFBB and a state-of-the-art pyrolysis and activation unit along with energy recovery would improve WWTP sustainability and self-sufficiency in terms of the raw materials required.

List of abbreviations

AC-	Activated Carbon
BM-	Baden-Baden Mixture
BMC-	Baden-Baden Mixture Activated Carbon
BW-	Household Biowaste
BWC-	Household Biowaste Activated Carbon
CED-	Cummalative Energy Demand
GWP-	Global Warming Potential
IFBB-	Integrated Generation of Solid Fuel and Biogas from Biomass
PAC-	Powdered Activated Carbon
WWTP-	Wastewater Treatment Plant
OMP-	Organic Micropollutant
LCA-	Life Cycle Assessment
CC-	Conventional Activated Carbon
FU-	Functional Unit

1. Introduction

1.1. Background

Pharmaceuticals are used for therapeutic purposes and contain compounds that are classified into antibiotics, analgesics, beta-blockers and anti-inflammatories (Díaz-Cruz and Barceló 2004). Post human consumption, residues of these compounds, commonly known as organic micropollutants (OMPs) are released into the sewage system and eventually enter wastewater treatment plants (WWTPs). During treatment at WWTPs, most OMPs resist the primary and secondary treatment steps and hence are only partially eliminated (Joss *et al* 2004, Clara *et al* 2005). The detection of OMPs in surface waters indicates that the present treatment at waste water treatment plants (WWTPs) is insufficient to remove OMPs effectively (UBA 2015), hence legal regulations are already in force in Switzerland for the implementation of a fourth treatment stage at WWTPs (FOEN 2015). Similarly, the European Union water framework directive (2000/60/EC) and the German federal water act (Wasserhaushaltsgesetz, WHG) have established legal obligations to maintain the quality of water (UBA 2018).

Removal of OMPs can be achieved using advanced treatment methods such as physical adsorption, biological degradation and chemical oxidation (Wang and Wang 2016). Physical adsorption using activated carbon (AC) is one of the most established process used for OMP removal. Compared to chemical oxidation using ozonation, a broader range of OMPs can be removed using AC treatment (Margot *et al* 2013). However, ACs used in European WWTPs are either manufactured from bituminous coal (Bayer *et al* 2005), which is related to substantial environmental and social impacts associated with material extraction, or from cocunut shells, which frequently originate from monocultures with adverse environmental impacts due to land use change and the use of chemical fertilizers and pesticides (Hartemink 2005). A large share of these materials are produced in Asian countries and need to be transported over long distances.

Using local residual raw materials (e.g. green cut, wood chips, fruit stones) instead, which are available in large quantities across Europe (Scarlat *et al* 2019), may overcome the constraints mentioned above and provide a pathway to convert these residues into a valuable resource. The public enterprise Eigenbetrieb Umwelttechnik in Baden-Baden, Germany, operates a municipal WWTP and biomass recycling center on the same site. Among other things, landscape management residues, hereinafter referred to as Baden-Baden Mixture (BM), and household biowaste (BW), consisting mainly of kitchen and garden waste, are regularly delivered there (table S1.1). Previously, these inputs have mainly been used for biogas and compost production. In the present study, BM and BW were pre-treated using the integrated generation of solid fuel and biogas from biomass (IFBB) technique, which mechanically separates biomass into a solid and a liquid fraction (Wachendorf *et al* 2009, Richter *et al* 2010, Hensgen *et al* 2011). Ash and harmful elements (e.g. N, S, Cl and K) are removed from the solid fraction, and the liquid fraction can be co-digested to produce biogas that can be used as an energy source for the IFBB process. For carbonisation and steam activation, a Pyreg A500 plant (Pyreg GmbH, Dörth, Germany) was put into operation which can handle ca. 900 t dry matter (DM) per year.

1.2. Literature review

As an alternative to conventional raw materials used for AC production, raw materials derived from residual sources with high carbon and low inorgnic content are preferred (Tsai *et al* 1997). Lignin and cellulose content of the biomass also influences the final pore structure of the AC (Savova *et al* 2001). AC produced using agriculutural and wood industry residues have been found to have comparable contaminant removal capacities to conventional coal based AC (Dias *et al* 2007). In addition to the raw material properties, process parameters such as pyrolysis temparature and activation conditions have a large influence on the charecteristics of the AC produced (Minkova *et al* 2000, Haykiri-Acma *et al* 2006). The adsorption capacity of AC produced is influenced by the BET surface area and total pore volume, the interaction with the polar and non-polar adsorbates is dependent on the chemical structure of AC (Moreno-Castilla 2004, Çeçen and Aktaş 2012).

AC characteristics can be modified to a certain extent by varying the material and process parameters to broaden the range of OMPs that can be adsorbed (Crittenden *et al* 2012). However, predicting the OMP adsorption capacity of AC based on its physical and chemical characteristics does not provide reliable results. But the reduction in UV_{254} absorption rate was found to have a correlation to the OMP removal rate, therefore, it can be used for predicting the removal rate (Zietzschmann *et al* 2014, Altmann *et al* 2015). Powdered AC (PAC) and granular AC are widely used for wastewater treatment (Margot *et al* 2013), but PAC has been found to be a better alternative for the removal of most OMPs (Boehler *et al* 2012, Kårelid *et al* 2017). Addition of PAC to wastewater can be implemented at different points in the treatment process, PAC can be brought in contact with wastewater in a mixing tank in conjunction with a flocculant (Boehler *et al* 2012, Faust and Aly 2018). In addition to direct addition of the PAC, advanced filtration mechanisms including anaerobic biofilters (Kaetzl *et al* 2019) and membrane filters can be utilised together with PAC. The dosage of PAC and the contact time also influences the removal rate of OMPs (Nam *et al* 2014), considering the economic feasibility of the commercial scale application, PAC dosages normaly range from 10–20 mg l^{-1} and contact time of 30 mins is used (Ruhl *et al* 2014). Better OMP removal performance translates to a lower dosage, thereby resulting in a lower quantity of AC required at the WWTP. The quantity of raw material essential to produce the required amount of AC depends on the char yield, which varies between 18% and 32% for biomass-based AC and is influenced by the raw material and process parameters (Ioannidou and Zabaniotou 2007, Schröder *et al* 2011).

1.3. Gaps in knowledge

Previous studies were aimed at production and testing of ACs from residual biomass raw materials high carbon and low ash concentrations (Schröder *et al*

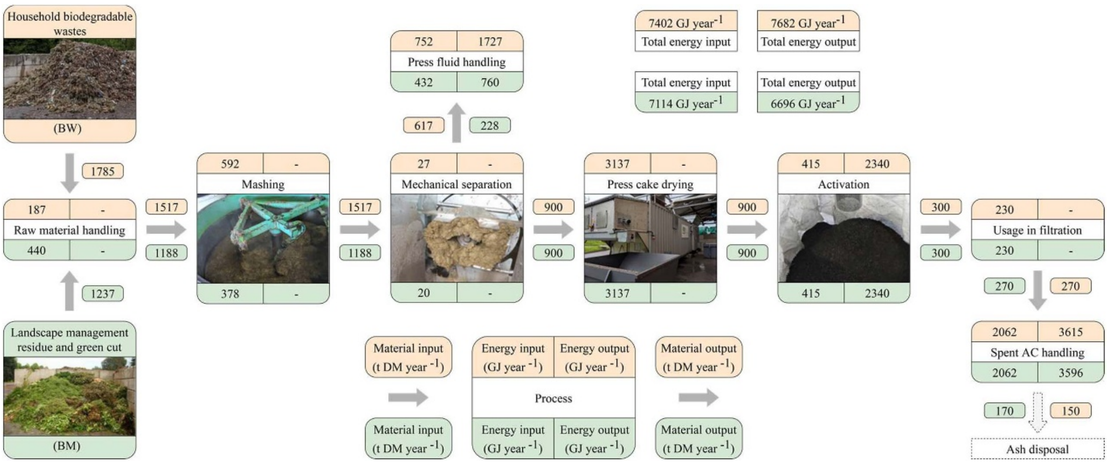

Figure 1. System boundary diagram elaborating the BM and BW AC production at the WWTP. The amount of input and output material and energy required for AC production at the annual capacity is indicated in the diagram. Ash disposal was not considered in this study.

2011, Margot *et al* 2013). More studies investigating the technical and environmental aspects of using residual biomasses consisting of a high ash content would enable a wider range of biomasses to be utilised for AC production. Studies assessing the environmental impacts of producing AC utilising residual biomasses were either carried out on a laboratory or protype scale (Hjaila *et al* 2013, Kim *et al* 2019) or using literature data (Arena *et al* 2016). Applying data obtained from a commercial scale AC production unit at a WWTP for modelling the environmental impacts would result in a more accurate assessment.The aims of this study were to assess the enviromental impact and energy demand of (i) production of AC from a biomass mixture (BMC) and biowaste (BWC) and (ii) the usage of this AC as an additional treatment step for micropollutant removal at a WWTP in comparison to conventional activated carbon (CC). Finally, a social risk assessment was conducted (iii) to provide deeper insight into the social effects of AC production. Although the production of AC from residual materials is well studied, most environmental impact studies were based on laboratory-scale data. Therefore, life cycle assessment (LCA) of AC production at a commercial scale WWTP, as in the present study, will increase the understanding of environmental and social impacts of this technical approach.

2. Methods

2.1. Life cycle inventory (LCI), system boundaries and impact categories

Samples of raw and IFBB pre-treated BM and BW used in this study were obtained from the WWTP Baden-Baden. Press cakes from IFBB were converted into ACs using a Pyreka (Pyreg GmbH, Dörth, Germany) laboratory-scale pyrolysis and activation reactor. The NORIT SAE Super (Boston, Massachusetts, USA) was used as a reference CC for benchmarking BMC and BWC. For the LCI, material flows and process data were calculated using data obtained from the WWTP. The datasets used for modelling the processes are elaborated in section S2, and compiling was done using OpenLCA 1.9.0.

A 'gate to grave' approach was used in this study, considering all processes involved, the 'gate' refers to the entry point raw material to the WWTP, from this point the processes involved are considered. 'Grave' refers to the final process in the system which is the end-of-life option for handling the AC produced (Martínez-Blanco and Finkbeiner 2018). Except for ash disposal, incineration of ACs with heat recovery was evaluated as an end-of-life option. The maximum annual production capacity of 900 t (dry weight) of BM and BW input material of the pyrolysis and activation unit was used for calculation of mass and energy balances (figure 1).

2.2. Activated carbon production and usage

Raw BM and BW was pre-treated according to the IFBB technology approach, as described in detail by (Joseph *et al* 2018). Ensiled BM and fresh BW was further processed according to the IFBB technique. Therefore, BM and BW were conveyed into a mashing unit and mashed with warm water (40 °C) for 15 min using stirrers forming a mash composing of 6%–7% dry matter content. The mash was then pumped into a screw press for mechanical separation of the solid and liquid fractions. Subsequently, the moisture content of the separated solid fraction (press cake) was reduced from 48% to almost 15% using a band dryer. The liquid fraction containing 3% DM was co-digested along with sewage sludge to generate biogas that was used to generate electricity and heat. Through the IFBB-process, mineral and ash content of BM and BW are significantly reduced, improving the feedstock quality for AC production (table 1). Energy and material balances for all the processes

Table 1. Characteristics of the Baden-Baden mix (BM) and biowaste (BW) at different steps in the production process of activated carbon.

	Unit	BM	BW
Raw material			
DM content	% FM	36.3	34.7
Ash	% DM	10.8	24.2
Volatile solids	% DM	89.3	75.8
C concentration	% DM	46.3	36.5
DM flow into press cake	%	75.8	59.3
DM flow into press fluid	%	24.2	40.7
Press cake			
DM content	% FM	55.3	52.7
Ash	% DM	6.5	10.8
Volatile solids	% DM	93.6	89.2
C concentration	% DM	47.9	43.1
Lower heating value	MJ DM^{-1}	18.0	16.1
DM content after drying	%FM	85.2	85.2
Activated carbon			
Ash	% DM	70.7	62.5
Volatile solids	% DM	29.3	37.5
C concentration	% DM	27.9	37.9
Lower heating value	MJ DM^{-1}	16.7	16.7
Press fluid			
DM content	% FM	1.8	2.9
Ash	% DM	23.2	54.6
Volatile solids	% DM	76.8	45.4
C concentration	% DM	41.5	26.0
Methane yield	L_n CH_4 kg^{-1} VS	275.0	395.0

involved in BM and BW production at the WWTP were calculated (tables S1.2–1.5).

For AC production using the Pyreka laboratory-scale reactor, BM and BW were pyrolyzed at 900 °C and physically activated by adding water vapour as an oxidation agent during pyrolysis. This ensured very good transferabillity of results to the full-scale Pyreg A500 reactor used at the WWTP Baden-Baden . Samples from raw biomass and IFBB press cake of BM and BW were obtained from the Eigenbetrieb Baden-Baden. As the press cake was used as feedstock for AC production, raw biomass was analysed for their physio-chemical composition and used for calculation of mass flows. BW was dried at 105 °C, ground in a cutting mill with a 20 mm sieve and used without further sieving. Due to its fluffy structure, which did not permit uniform pyrolysis and activation, the BM was pelletized before processing. For this purpose, the BM was dried at 60 °C, ground in a hammer mill and then pressed into pellets with a diameter of 6 mm. While the BW feedstock was subsequently pyrolyzed at 900 °C for 10 min, BM pellets required with 30 min a longer residence time in the reactor at the same temperature for complete carbonization. The activation was done by adding defined volumes of water vapour to the pyrolysis.

At the full-scale PYREG A500 reactor, the thermal energy required for the pyrolysis process is generated by burning the resulting pyrolysis gases. In addition, surplus heat energy from pyrolysis is recovered and used to supply the processes in the pre-treatment steps (figure S1.3(stacks.iop.org/ERL/15/064023/mmedia)). After its use for the removal of OMPs from wastewater, incineration of spent ACs was assumed to be the typical end-of-life procedure. The resulting heat was assumed to offset local heating requirements, which is considered in the LCI.

2.3. Functional unit

OMPs pose an ecotoxicological risk in aquatic environments (Oldenkamp *et al* 2019). This risk was calculated according to the USEtox model (Rosenbaum *et al* 2008) for selected pollutants measured in our study (table S2.1). By using the investigated ACs for wastewater treatment, OMPs were removed and, thus, the ecotoxicological risk in receiving water bodies was reduced. The factors defined in the USEtox model for OMPs were multiplied by their concentration in wastewater to calculate the mean ecotoxicity value for all OMPs. Based on these values, the functional unit for the LCA was fixed to the required quantity of AC to reduce the aquatic freshwater exotoxicity potential to a defined level in terms of the comparative toxicity unit, i.e. ecotoxicity (CTU_e L^{-1}) per 1000 m^3 of wastewater (figure S2.2) (Rosenbaum *et al* 2008).

Performance of AC in terms of OMP removal from wastewater was measured in laboratory experiments. For this, concentrations of 1.75 to 7 mg l^{-1} AC were added to wastewater to evaluate the influence of the dosage on the reduction of OMPs. The ecotoxicity reduction at a dosage of 5 mg l^{-1} of CC was used as a benchmark. The required amount of BMC and BC to achieve the same ecotoxicity reduction was calculated and used for the LCA (figure S2.2). Based on these data, multiplication factors for the LCA were determined by dividing the amounts of BM and BW AC by 5 mg l^{-1} of CC (figure S2.1).

2.4. Social risk identification

The social risks involved in the production chain were identified based on country-specific risk assessments adapted from (Saling *et al* 2020) considering all main foreground processes. The processes were classified into the 11 social impact categories selected from Goedkoop *et al* (2018) (table S4.1) according to the intensity of social risk in the country of production. These categories were weighted according to the severity of the social issues as estimated by INEF (Institut für Entwicklung und Frieden) (table S4.1) (Saling *et al* 2020). Depending on the data availability, social risk scores for a specific country or mean values for the ten largest producer countries were weighted by production volume in the LCA.

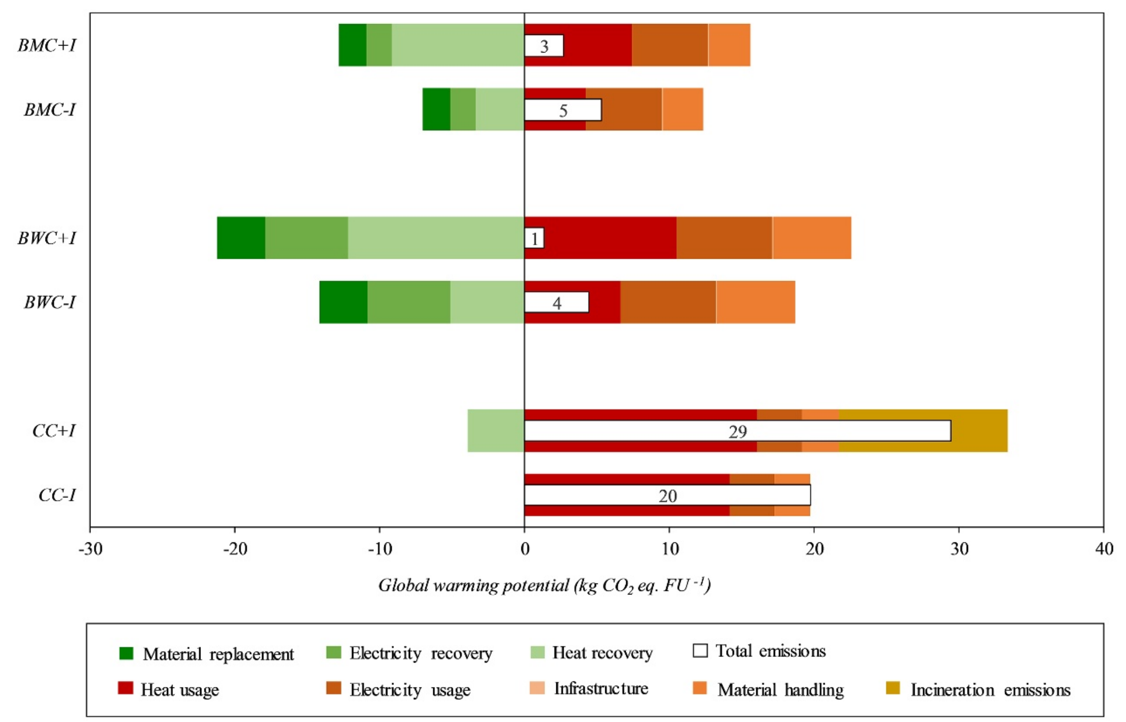

Figure 2. Global warming potential (GWP) generated during the production and usage of activated carbon derived from Baden-Baden mix (BM), biowaste (BW) and conventional coal (CC). Each section of the horizontal bars indicates the amount of GWP emitted or saved by the respective parameters as indicated in the legend. (+ I) denotes the inclusion of end-of-life incineration and related effects on the GWP, whereas (-I) indicates the case without incineration. Negative values indicate GWP savings and positive values indicate GWP emissions.

3. Results

3.1. Ecotoxicity

Removal efficiency of OMPs from wastewater was higher for CC compared to BMC and BWC (table S2.2 and S2.3). A dosage of 5 mg l^{-1} CC was able to attain an ecotoxicity of approximately 55 000 CTU_e L^{-1}. To achieve the same ecotoxicity, a dosage of 9.1 mg l^{-1} BWC was needed, which resulted in a 1.82-fold higher demand of BWC compared to CC. In contrast, performance of BMC was significantly better, requiring a dosage of 6.1 mg l^{-1} and thus an equivalence of 1.22 times CC (figure S2.2). Based on annual wastewater production of around 9 510 000 m^3 at the Baden-Baden WWTP to be treated and a dosage of 5 mg l^{-1} of CC, this results in a demand of approximately 48 t CC a^{-1}. Considering the multiplication factors of BMC (1.22) and BWC (1.82), 58 t and 87 t a^{-1}, respectively, would be needed to achieve the same ecotoxicity level.

3.2. Global warming potential

The GWP for BMC and BWC production (5.3 and 4.5 kg CO_2 eq. FU^{-1}, respectively) was significantly lower compared to CC (20 kg CO_2 eq. FU^{-1}) (figure 2). Considering incineration of the spent ACs and the recovered energy obtained, GWP for BMC and BWC was reduced to 2.7 and 1.3 kg CO_2 eq. FU^{-1}, respectively, whereas this resulted in an increased GWP of 29 kg CO_2 eq. FU^{-1} for CC. Considering the various production steps, the heat requirements for press cake drying had the highest GWP for BMC and BWC (7.1 and 4.5 kg CO_2 eq. FU^{-1}, respectively). For CC, heat usage for pyrolysis and activation was the highest contributor (14 kg CO_2 eq. FU^{-1}). For the production of the required 58 t of BMC and 87 t of BWC, 345 and 515 t DM of raw material would need to be processed, respectively. Hence, the GWP associated with electricity usage in the facility was higher for BWC than for BMC. Similarly, GWP related to the handling of press fluid digestates depended on the quantity that was transported and applied on agricultural land, leading to higher GWP for BWC versus BMC (6232 versus 4689 t FM, respectively). Mining and transportation of hard coal for further processing led to substantial GWP for CC. GWP related to infrastructure usage was negligible compared to the overall GWP for all three ACs. The total usage-related GWP was highest for CC, closely followed by BWC and much lower for BMC.

In contrast to CC, for which production only generated GWP emissions, the production of BMC and BWC also enabled GWP savings. Heat recovered from the activation unit saved 3.4 kg CO_2 eq. FU^{-1} for BMC compared to 5.1 kg CO_2 eq. FU^{-1} for BWC. Electricity recovered from combustion of press fluid-derived methane was higher for BWC, saving 5.7 kg CO_2 eq. FU^{-1} versus 1.7 kg CO_2 eq. FU^{-1} for BMC. Application of digestate on agricultural fields replaced the need for conventional mineral fertilisers,

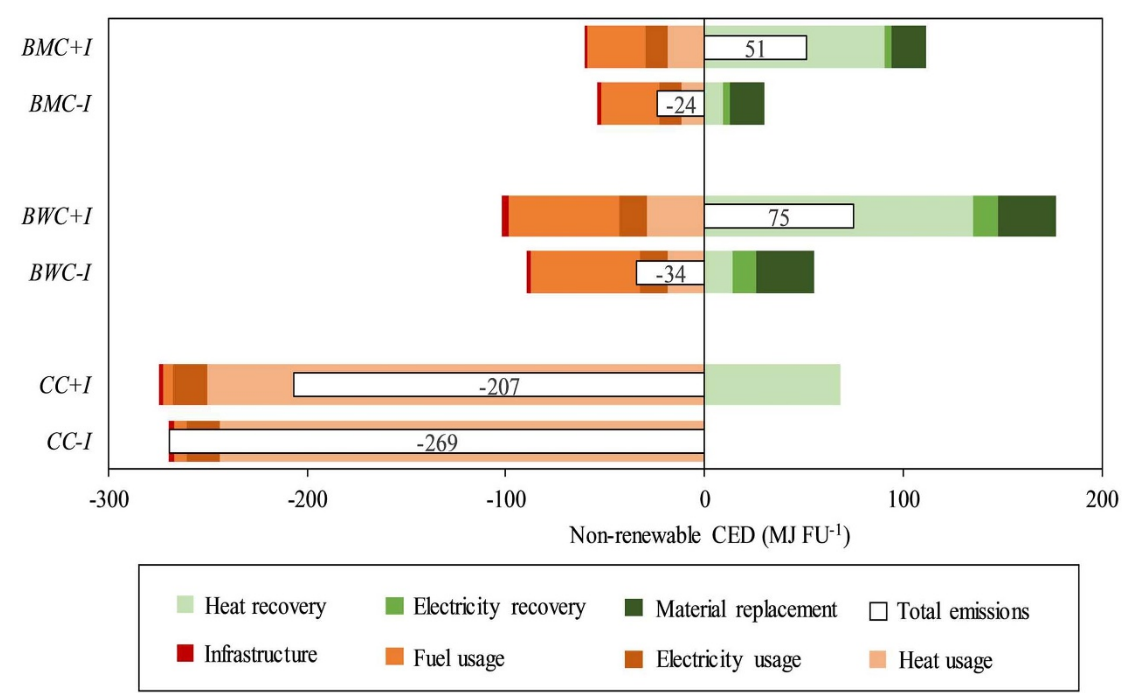

Figure 3. Cumulative energy demand (CED) for non-renewable sources generated during the production and usage of activated carbon derived from Baden-Baden mix (BM), biowaste (BW) and conventional coal (CC). Each section of the horizontal bars indicates the amount of non- renewable CED used or saved by the respective parameters as indicated in the legend. (+ I) denotes the inclusion of end-of-life incineration and related effects on CED, whereas (-I) indicates the case without incineration. Negative values indicate usage of energy and positive values indicate energy savings.

thereby saving 3.3 kg CO_2 eq. FU^{-1} for BMC and 1.9 kg CO_2 eq. FU^{-1} for BWC. Energy recovered from incineration of spent AC resulted in GWP savings of 5.8 and 7.1 kg CO_2 eq. FU^{-1} for BMC and BWC, respectively. However, incineration also caused additional emissions of 3.2 (BMC) and 3.9 (BWC) kg CO_2 eq. FU^{-1} from heat usage for de-watering and drying of spent AC sludge. For CC, there was an GWP savings of 4 kg CO_2 eq. FU^{-1} from the energy recovered by incineration; however, the benefits of incineration were overshadowed by additional emissions of 12 kg CO_2 eq. FU^{-1} resulting from the release of CO_2 during incineration and 2 kg CO_2 eq. FU^{-1} for de-watering and drying. Overall, GWP savings were highest for BWC, followed by BMC and CC.

3.3. Cumulative energy demand (CED)

Energy needed for operating the processes was estimated in terms of CED and classified into renewable and non-renewable CED according to the source of energy. Energy used to produce CC was almost completely based on non-renewable sources, whereas biomass was the primary source of energy for producing BMC and BWC (5% and 11% share of non-renewable CED, respectively). Total use of non-renewable energy was substantially higher for CC (269 MJ FU^{-1}) than for BMC and BWC (54 and 83 MJ FU^{-1}, respectively) (figure 3).

Due to energy recovery and material replacement, the energy balance for BMC and BWC further improved to 24 and 34 MJ FU^{-1}, respectively, and the highest non-renewable CED resulted from fuel usage for transportation of digestate (almost 60% of the total non-renewable CED both for BMC and BWC). While incineration as an end-of-life option improved the energy balance of all three ACs, the combustion of the spent BMC and BWC even generated excess energy. In contrast, the balance for renewable CED was higher for both BMC and BWC (291 and 222 MJ FU^{-1}, respectively) compared to 150 MJ FU^{-1} for CC (table S3.3). Heat usage for press cake drying represented almost 75% of the renewable CED from both BMC and BWC, and it increased to 81% when the heat required for drying AC sludge was considered. Though overall CED was higher for BMW and BWC, almost 90% of it was generated from a renewable biomass feedstock.

3.4. Additional environmental impacts

The acidification potential associated with the production and usage of bio-based AC was significantly higher than for CC (table S3.1). This was mainly caused by the SO_x and NO_x emissions from the activation process, which accounted for more than 60% of the total acidification potential, while NH_3 emissions from the press fluid digestate application on the field had a substantial contribution of 26 and 33% for BMC and BWC, respectively. Almost 42% of the acidification potential of CC was related to electricity and fuel demands during coal mining. Further emissions associated with heat and electricity usage during drying, pyrolysis and activation amounted to 53% for CC.

BWC had the highest eutrophication potential, followed by BMC and CC. NH_3 emissions from digestate application accounted for the largest share of total emissions both for BMC and BWC (38% and 50 %, respectively). For CC, nitrogen (N)-based emissions for heat and electricity generation were the highest contributors. Incinerating spent ACs reduced the acidification and eutrophication potential of all ACs (table S3.1 and S3.2).

Environmental and energy balances were also examined at a dosage of 20 mg AC L^{-1}, which is the highest standard dosage used at WWTPs, and it was found that this increased GWP balances of BMC, BWC and CC by a factor of 3.2, 3.0 and 3.55, respectively. Incineration resulted in a lower GWP for BMC and BWC (6 and −4 kg CO_2 FU^{-1}) but higher GWP for CC (107 kg CO_2 FU^{-1}) (figure S3.1).

3.5. Social life cycle assessment

Producing bio-based ACs at the WWTP was found to reduce social risks substantially compared to the conventionally produced AC, as 83% of all processes at Baden-Baden were categorised as 'low risk' in contrast to only 14% for the production of CC (figure S4.2). This was mainly due to the fact that processes involved in the extraction of bituminous coal take place in countries which usually perform poorly for almost all social impact categories (table S4.2). While processing, production and usage of CC took place at locations with somewhat lower social risks, the associated use of diesel and natural gas resulted in very high to high social risks, respectively.

4. Discussion

4.1. Ecotoxicity

Performance of BMC and BWC in terms of OMP removal from wastewater was significantly lower compared to CC (table S2.2 and S2.3). This may be explained by the significantly lower specific surface area and adsorption capacity of bio-based ACs. Hence, higher dosages of BMC and BWC were needed to achieve the same level of ecotoxicity reduction. Removal of OMPs from effluent water mainly depends on the adsorption characteristics of ACs used, such as specific surface area (Mailler *et al* 2016). These characteristics are highly affected by the properties of the raw materials (Erdoğan *et al* 2017) and the activation process (Bergna *et al* 2019). However, the precise influence of the raw materials on adsorption of OMP is not clear, although the process conditions for BMC and BWC production were comparable (Mailler *et al* 2016). For example, it is remarkable that the removal of OMP by BMC was higher than for BWC though the specific surface area for BMC was lower. The high ash and low carbon content for BMC and BWC may have affected the adsorption capacity, as these properties have an influence on the pore structure (Anisuzzaman *et al* 2015). The ash content of the raw biomass was significantly reduced by the IFBB pre-treatment, which improved the quality of the ACs (Joseph *et al* 2018; László *et al* 1997). Further improvements in biomass processing, pyrolysis and activation can be expected to increase the adsorption capacity of activated carbons. This, in turn, would result in a further reduction of GWP, CED and other environmental impacts through BMC and BWC production and usage.

4.2. Global warming potential

The combination of raw biomass collection, processing into ACs and their subsequent utilization at the investigated WWTP is unique. To facilitate the comparison with other studies, all life cycle stages including production, usage and end of life were examined individually and compared with similar studies.

Emissions during the production stage resulted in maximum contributions to GWP in the life cycle of all ACs. To allow a comparison with other studies at this stage, the GWP generated by producing 1 kg of the respective AC was used instead of the functional unit. The production of 1 kg BMC and BWC resulted in emissions of 0.43 and 0.20 kg CO_2 eq., respectively, which was significantly lower than those reported for AC from olive waste cake (11.1 kg CO_2 eq. kg^{-1}) (Hjaila *et al* 2013). GWP for ACs produced from conventional raw materials was 1.15 kg CO_2 eq. kg^{-1} for coconut shell (Kim *et al* 2019), 8.4 to 11.1 kg CO_2 eq. kg^{-1} for bituminous coal (Bayer *et al* 2005, Gabarrell *et al* 2012) and 2.45 kg CO_2 eq. kg^{-1} for regenerated AC (ROFA Carbon 2018), which are all notably higher compared to BMC and BWC. In our study, GWP for CC production from bituminous coal (3.41 kg CO_2 eq. kg^{-1}) was significantly lower than the range reported by (Bayer *et al* 2005) and (Gabarrell *et al* 2012), which can be explained by the fact that CO_2 emissions associated with removal of fossil carbon was not considered in the production stage for this study, but rather included in the end-of-life stage as CO_2 emitted during incineration.

The low GWP in our study can be attributed to the efficient pyrolysis and activation process, whereby the combustion of pyrolysis gas provided more heat than was required. While pyrolysis was reported to account for 30%–47% of the total GWP in other studies (Hjaila *et al* 2013, Gu *et al* 2018), it only contributed 7% in the present study. Furthermore, electricity generation from the IFBB press fluid reduced the energy demand for production and, consequently, the GWP.

The delivery of residual raw materials to the facility prevented CO_2 emissions related to transportation and extraction. Conversion factors from raw material to AC were slightly different for BM (11 %) and

BW (7 %) due to different physico-chemical properties, such as dry matter and carbon content, as well as differing mass flows during mechanical separation. The conversion efficiency from press cake to AC was equal for BM and BW (ca. 24%). However, the transfer of BM dry matter into the press cake (76 %) was considerably higher than for BW (59 %), which resulted in a lower contribution of AC production to the entire GWP for BM, as less raw material had to be processed. On the other side, the higher mass flow of BW dry matter into the press fluid increased GWP savings from electricity recovery. Though the type of raw material affected the overall GWP, the impact of the production process was much greater. Since the complete production and usage of BMC and BWC took place at the WWTP, material transport did not contribute to the GWP. However, emissions related to the transport of the reference CC were also negligible (0.003 kg CO_2 eq. FU^{-1}). Nevertheless, the GWP associated with the production did not consider the contrasting performances of ACs in terms of OMP removal. To this end, an appropriate functional unit (i.e. the quantity of AC to achieve a defined ecotoxicity level in wastewater) was introduced, which allowed determination of the amounts of ACs required and, thus, a true comparison between conventional and bio-based ACs.

Energy recovery through incineration of the spent BMC and BWC reduced the overall GWP for these ACs, whereas the combustion of CC resulted in an increased GWP, as the resulting CO_2 emissions originated from fossil sources. Hence, using AC produced from local residual biomass has an advantage during incineration, which is a commonly used end-of-life step for ACs. OMP removal using ozonation usually has a lower GWP (18 kg CO_2 eq. 1000 m^{-3}) (Kounina and Wencki 2015) compared to use of CC (20 kg CO_2 eq. 1000 m^{-3}) (Baresel *et al* 2019); however, the figures for BMC and BWC (4 and 5 kg CO_2 eq. 1000 m^{-3}, respectively) in our study were even much lower compared to ozonation.

4.3. Cumulative energy demand (CED)

The energy balance of AC production and use is decisive for the profitability of the concept (Blumenstein *et al* 2012). The CED for the production of BMC and BWC (30 and 15 MJ kg-1, respectively) was notably lower than for CC (53 MJ kg^{-1}). Our data differ widely from results of other studies, which found CED values between 158 and 241 MJ kg^{-1} for woodchip and coal-based AC, respectively (Bayer *et al* 2005, Gu *et al* 2018). The lower CED for BMC and BWC production in our study was mainly caused by the efficient pyrolysis and activation processes as well as by recovered electricity from anaerobic digestion of press liquids.

Methane yield from the press fluid was considerably higher for BWC (395 L_n CH_4 kg^{-1} VS) compared to BMC (264 L_n CH_4 kg^{-1} VS). Methane content and mass flow of volatile solids into the press fluid were highly influenced by material properties, such as dry matter content and IFBB process parameters (Richter *et al* 2009, Hensgen *et al* 2011). Although the overall CED was lower for production of BMC and BWC, the share of non-renewable CED was higher than for CC. As this energy comes from depletable resources (Arvidsson and Svanström 2016), additional research is needed to improve the sustainability of alternative AC production. Substitution of diesel used for transportation and processing by biofuels is one potential solution.

4.4. Additional environmental impacts

Emissions of NO_x, SO_x and NH_3 damage land ecosystems by affecting plant health and lowering soil pH (Rosenbaum *et al* 2018). Though the emission of NO_x and SO_x from the activation process were the largest contributors to acidification, they were still below the German limit of 350 mg m^{-3} (Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit 2002). Mass flow of N during mechanical separation into press cake and fluid caused N emissions in the following stages, i.e. NO_x emissions during pyrolysis of the press cake and activation of the resulting biochar (Zhan *et al* 2018) as well as NO_x and NH_3 emissions from the field application of digestates originating from the anaerobic digestion of press fluids (Eggleston 2006). Recent research showed that the flow of elements during mechanical separation is influenced by the raw material properties, process parameters (Richter *et al* 2011) and scale of the conversion process (Joseph *et al* 2018). Modification of these factors affects the flow of elements and, thus, the acidification and eutrophication potential. Reduction of these impacts is possible through the use of precision application techniques for digestates (Nicholson *et al* 2018) and advanced flue gas cleaning systems (Turconi *et al* 2013).

4.5. Social risk assessment

For estimating the social risks associated with AC production, available average values on the country level were used, as data from producers did not exist or were not representative (Saling *et al* 2020). Our results indicate that risks could be reduced when all processes take place in countries with a low risk level or when materials from countries with a lower associated social risk were used. To this end, materials need to be labelled appropriately.

The local production and use of AC from residual biomass in industrialized countries like Germany would affect economic activities in the current production countries. Due to a lack of revenue, this may lead to a further deterioration of the social situation in the developing countries. Possible losses of employment could, however, be offset by investments in alternative employment opportunities for people working in fossil-based sectors (Sparkes 2008). Socially responsible investments would help to improve the social conditions, and certification of socially-compatible production processes may contribute to a higher demand of products with lower social impact (Goedkoop *et al* 2018). Nevertheless, the conducted social risk assessment for AC production contained global and, therefore, inaccurate data. Hence, only potential hotspots in the production process on the country-level could be identified (Saling *et al* 2020). However, our findings may serve as a starting point for further investigations on the exact sources of social risk at a sector and company level.

5. Conclusion

Using ACs produced from local residual biomass for the removal of OMPs at a large-scale WWTP was found to have lower environmental impacts in terms of GWP and CED compared to conventional ACs. The novel concept was found to be flexible in terms of raw materials, though the BM and BW used in this study had distinctive properties. The IFBB step significantly reduced the ash content of feedstocks for AC production, and the generated press fluids further improved energy generation and material replacement. Thus, IFBB was a crucial step in utilising residual biomass for AC production. On-site production and use of the AC enabled a significant reduction of monetary and environmental costs compared to systems based on conventional ACs. The lower OMP adsorption performance of BMC and BWC could be overcome by using higher dosages of AC without risking GWP and CED advantages. Performance of BMC and BWC is expected to be increased by further process optimization, which would render the novel system even more attractive for WWTPs that currently use conventionally produced AC.

Acknowledgments

The authors would like to acknowledge the European Union for financing the Re-Direct project (NWE 294) through the INTERREG European regional development fund and the German Federal Ministry for Education and Research (BMBF) for financing the CoAct project within the research initiative Stadt-Land-Plus (Grant Number 033L206). We would also like to thank O. Hermann from Eigenbetrieb Umwelttechnik Baden-Baden for providing the material and information needed for the study. We are grateful to the support of K. Stenchly, T. Astor, J. Wijesingha, T. Fricke, R. Graß and A. Gerke. The authors also wish to thank the support by the Open Access Publication Fund of the Universität Kassel.

Data availability statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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