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Review article

THE DIFFERENT FACES OF BIOCHAR: CONTAMINATION RISK VERSUS REMEDIATION TOOL

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Abstract. This article reviews the different aspects of biochar as source and sink of organic and inorganic contaminants. Biochar can contain organic contaminants such as polycyclic aromatic hydrocarbons or heavy metals. As the distribution coefficients of the biochar especially for contaminants are high, the freely dissolved concentrations are low and with that also the bioavailability. The link between biochar's inherent contaminants and toxicity to soil meso– and macro–fauna remains unclear, with data being often contradictory and influenced by feedstock and pyrolysis conditions. The biochar's potential to remediate contaminated soils has mainly been addressed in lab studies, but rarely in the field. This far, results have been contradicting. Many studies reported successful immobilization of contaminants but some not. In summary, the ambivalent face of the biochar with regard to contaminants prevails. In future, long term field studies are needed to properly address the sustainability of biochar in this respect.

Keywords: biochar inherent organic and inorganic contaminants, bioavailability, bioaccessibility, ecotoxicity, environmental risks.

Introduction

Besides its potential agricultural benefits (Atkinson *et al.* 2010; DeLuca *et al.* 2015; Ippolito *et al.* 2015; Jeffery *et al.* 2015; Thies *et al.* 2015; Van Zwieten *et al.* 2015; Whitman *et al.* 2015), biochar can contribute to remediation and recovery of contaminated soils through sorption of heavy metals and/or organic contaminants (Beesley *et al.* 2011; Ahmad *et al.* 2016; Kupryianchyk *et al.* 2016; Li *et al.* 2016; Shen *et al.* 2016). This can be envisioned as the positive face of biochar. Nonetheless, biochar may contain contaminants itself, either introduced by its feedstock (e.g., heavy metals), or co-produced during (improper) pyrolysis (e.g., polycyclic aromatic hydrocarbons; PAHs). The over-time release of contaminants inherent to biochar and the potential consequences to soil and the wider environment represent the negative face. Due to the manifold agricultural and environmental management procedures in which biochar is involved, it is important to rigorously assess its potential hazards and benefits over the relevant timescales. This paper reviews the different aspects of

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biochar and discusses and contextualizes hazards and benefits of biochar in light of the latest scientific research. *Pro et contra* of contaminants in biochar and its suitability for remediation purposes will be summarized and denoted with a "smiley", a "frowny", and – where not clear – with an ambivalent emoticon. Furthermore, current knowledge gaps will be listed and future research needs identified.

1. Biochar – associated contaminants – total concentrations

1.1. Organic compounds

During pyrolysis, a multitude of (natural) organic compounds are involved either as precursor, intermediate or terminal products in any of the three physical states: syngas, bio-oil and biochar (Bucheli *et al.* 2015). These authors concluded that formation of polychlorinated aromatic compounds under fully pyrolytic conditions is not likely, whereas polychlorinated dibenzo-p-dioxins, -furans (PCDD/Fs) may be formed if suitable precursors, such as salt in food waste, are present in the feedstock. Hale *et al.* (2012) reported of up to 92 ng kg^{-1} PCDD/Fs (0.15 ng kg^{-1} toxic equivalents (TEQ)) from biochars whose feedstock was, amongst many, food waste. These concentrations are very low as the European Biochar Certificate (EBC 2012) set the TEQ of PCDD/Fs <20 ng kg^{-1}. However, during pyrolysis of the feedstock, PAHs are formed (Bucheli *et al.* 2015). The authors reviewed 21 studies that reported PAHs in biochars and contextualised the concentrations as determined by pyrolysis type and temperature, feedstock, and residence time of the feedstock in the reactor. These concentrations ranged from a low 100 μg kg^{-1} to 100 mg kg^{-1}, with outliers in the thousands of mg kg^{-1} for the sum (Σ) of the 16 US EPA PAHs. Generally, the evaluation was difficult, since different extraction methods were used to quantify the PAHs at that time. These are now standardized by the EBC and the International Biochar Initiative (IBI 2015b). Nevertheless, trends could be identified. For instance, elevated PAH concentrations were found at high (>800 °C) temperatures that are typical of gasification (Bucheli *et al.* 2015), which is due to the re–condensation of the PAHs on relatively small fractions of solid residues (Hale *et al.* 2012; Schimmelpfennig, Glaser 2012). In contrast, Yargicoglu *et al.* (2015) could not detect PAHs after the gasification of wood pellets. They hypothesize that this might be due to some oxygen during the gasification leading to a more complete combustion of the wood and less PAH formation. Additionally, the pelleting prior to pyrolysis might have removed the small particles where PAH preferably re-condensate on. Most recently, Buss *et al.* (2016) convincingly confirmed the hypothesis expressed in Bucheli *et al.* (2015) that recondensation of pyrosynthesized PAHs onto biochar is responsible for elevated PAH concentrations in biochar, and that effective separation of syngases from solid feedstock residues (biochar) during pyrolysis leads to biochar with PAHs in the low mg kg^{-1} concentration range. Irrespective of the pyrolysis type, recondensates also contain a large variety of (semi-)volatile organic compounds (VOC, (Spokas *et al.* 2011; Buss *et al.* 2015)). The most encountered VOC were acetic and formic acid, phenols, o-, m- and p-cresol, 2,4-dimethylphenol (Buss *et al.* 2015) and acetone, methyl ethyl ketone, methyl acetate, benzene, and trichloroethene (Spokas *et al.* 2011). Polycyclic aromatic hydrocarbons in biochar peaked at temperatures of 400–500 °C, without a clear influence of the feedstock type (Bucheli *et al.* 2015). The impact of residence time was also minor. Some individual studies reported a tendency of decreasing PAH concentrations with increasing residence time (Bucheli *et al.* 2015) but was not observed for slow pyrolysis in the study of Buss *et al.* (2016).

1.2. Heavy metals

Heavy metals (elements with a specific density >5 g mL^{-1}; (Morris 1992)) and metalloids that occur in the input material for pyrolysis become concentrated during carbonization, because of volatilization of gaseous carbon products (syngases), and a corresponding mass loss of the residual solid material (Brown *et al.* 2015). This concentration process has to be considered especially in the case of sewage sludge as input material (Agrafioti *et al.* 2013). Waste water originating from industrial areas may produce sewage sludge with elevated heavy metal concentrations that will further concentrate during pyrolysis. In such cases, a subsequent gasification step of the char product involving the addition of chlorides results in enhanced volatilization of metal chloride complexes (Luan *et al.* 2013) that can decrease the heavy metal content in the char residues (Dong *et al.* 2015; Li *et al.* 2015).

Besides sewage sludge also vegetation–derived feedstock may contain heavy metals that will not be lost during pyrolysis. Depending on the geological conditions and the resulting soil concentrations, plant root uptake may lead to elevated concentrations in the above-ground tissues. Atmospheric deposition may cause metal enrichment in plant tissues, too, but it will mainly remain on leaf surfaces. Green waste residues usually are separated into fine fractions, including leaf materials that are rather composted than carbonized and woody fractions that are preferably used for pyrolysis or gasification. Therefore vegetation-derived biochar is more likely to contain heavy metals from plant uptake if the plants are grown on soils with high geogenic metal concentrations or on anthropogenically contaminated land, or if the plant is a species that is able to translocate the contaminants into the shoots (Greger *et al.* 2007; Enell *et al.* 2016).

1.3. Guide/threshold values of contaminants in biochar

The two main guidelines, EBC and IBI, denote the organic and inorganic contaminant content threshold values, which are summarized in Table 1 (for further information on the IBI values we point to the references therein). The minimum threshold values of the IBI and the values for premium grade biochars of the EBC for inorganic and organic contaminants are alike. The maximum threshold values of the IBI are factors higher (2x for Pb, 60x for Cu, 25x for the ∑16 US EPA PAHs) than the concentrations set by the EBC for all contaminants. This difference reflects the management of the contaminants in different countries as the IBI based their maximum values mainly on Australian and US regulations, while the EBC based theirs on those of Germany and Switzerland. After all, national standards need to be met also. For instance Austria regulates the heavy metal contents of plant materials for quality control of pyrolysis feedstock (ÖNORM S 2211–1 2016). According to this standard, the biochar must not exceed certain threshold concentrations if agricultural applications are envisaged (in mg kg^{-1}_{dw}): Pb 100, Cd 3, Cu 150, Ni 100, Hg 1, Zn 500, Cr_{tot} 100. For use in organic agriculture there are lower thresholds than in conventional agriculture (in mg kg^{-1}_{dw}): Pb 45, Cd 0.7, Cu 70, Ni 25, Hg 0.4, Zn 200, Cr_{tot} 70. The EBC as well as the IBI also give guidelines for producers concerning biochar properties, quality assurance and control (QA/QC), analytical methods, among others. Also here, we refer to the recommendations and guidelines listed in the EBC and IBI for the specific analytical procedures.

The QA/QC in Europe is coordinated by the q.inspecta agency which awards certificates to producers according to the EBC guidelines. Q.inspecta is a governmental accredited company with inspections of production plants in individual countries carried out by independent national inspection agencies. Producers with a capacity <20 t y^{-1} are exempt from the yearly inspections of the production process. These small scale producers are to assure the quality of their biochar via self-declaration and a detailed description of the complete production process. The IBI Certification Programme (IBI 2015a) provides biochar manufacturers the opportunity to certify their biochar(s) as having met the minimum criteria established in the IBI Biochar Standards. The biochar producer shall apply online for the certification procedure, the sample will be tested by a laboratory according to the IBI Standards and the certificate shall be renewed, if obtained, after a year.

Table 1. List of inorganic and organic thresholds of contaminants recommended in biochars of the European Biochar Certificate (EBC 2012) and the International Biochar Initiative (IBI 2015b) guidelines. For details, see text

Contaminant	EBC	EBC	IBI	IBI
Heavy metals	Premium grade g	Basic grade	Minimum	Maximum
		gt^{-1}_{dw}		
Pb	<120	<150	121	300
Cd	<1.0	<1.5	1.4	39
Cu	<100	<100	143	6000
Ni	<30	<50	47	420
Hg	<1	<1	0.8	17
Zn	<400	<400	416	7400
Cr	<80	<90	93	1200
Co	–	–	34	100
Mo	–	–	5	75
Se	–	–	2	200
Organic compounds				
Sum of the 16 US EPA PAHs	<4	<12	6	300
Benzo[*a*] pyrene	–		3 g t^{-1} TEQ_{dw} [a]	
PCB	<0.2		0.2	1.0
Dioxins & furans	<20 ng kg^{-1} TEQ		17 ng kg^{-1} TEQ	

Note: [a] Toxicity equivalents TEQ.

1.4. Analytical aspects

The biochar community endeavours to develop and apply recommended methods for the quantification of organic and inorganic contaminants in biochar. Prior to the quantification, it is crucial to sample the biochar lot appropriately to obtain a representative sample that can be sent to the laboratory. In essence, a (biochar) sample is representative if its distributional heterogeneity is reduced. This can be achieved by a thorough mixing and splitting of the biochar lot for several times (Bucheli *et al.* 2014; Hilber *et al.* 2017). After that, the biochar lot shall be sampled several times and the subsamples joined to one bulk sample and further prepared (EBC) prior to be sent to the laboratory. The representative mass of the sample depends on its particle size in the way the bigger the particle the more of the sample (mass) has to be taken. The ISO norm (ISO 10381-8 2006 2006) provides, as a rule of thumb, a sample particle size to sample mass relation for the appropriate mass to be taken to the laboratory (Hilber *et al.* 2017).

To estimate the reliability of analytical results, 22 laboratories in 12 countries participated in an interlaboratory comparison within the COST Action TD1107 (Bachmann *et al.* 2016). Three biochars produced by slow pyrolysis from wood chips, paper sludge and wheat husks mixture, and from sewage sludge were analysed

on 38 physical-chemical parameters (macro- and micro-elements, heavy metals, PAHs, pH, electrical conductivity, and specific surface area) by the participants with their preferential methods. The data were evaluated with the PROLab Plus™ (Version 2015.11.3.0) software that compares Zu scores, and the performance was valued according to EBC. A Zu-score is a point-based system that identifies a result of a parameter measured by a laboratory as inconspicuous, conspicuous or as an outlier (Bachmann *et al.* 2016). In summary, while the intra-laboratory reproducibility was generally good, the inter-laboratory reproducibility was mostly not (Bachmann *et al.* 2016). Only the carbon content (C) and pH showed a mean reproducibility standard deviation (SD) <10%. Those of hydrogen (H), ash and phosphorous (P) were between 10 and 20%. The variability of all other parameters was too high to be reliable. This result seems to be the consequence of participant laboratories using their own methods, because the EBC guidelines are quite recent (first version published in 2012). The study showed an urgent need to improve and standardize methods for biochar characterization. Future QA/QC measures require the production of a set of representative biochar reference materials and professional round robin tests with laboratories that use biochar reference methods (Bachmann *et al.* 2016). A recent review of benefits and risk of biochar clearly identified the need to develop biochar quality standards (Kuppusamy *et al.* 2016).

The QA/QC in the above section covers the technical aspects of certification. However, it can extend far beyond a pure labelling for a high quality biochar product. The sustainability inherent to a certification scheme should also include the application component, for instance, in the form of an optimal biochar dose (OBD), a contamination control, a life cycle assessment, among other aspects (Verheijen *et al.* 2015a). The authors of this new and sustainable certification concept identified future challenges such as e.g., (i) trade–offs between soil functions, (ii) the longer mean residence time in soil compared to traditional soil amendments, and (iii) limited mechanistic understanding of the nature and extent of the effects of biochars in a soil-crop system. Although a considerable effort is required to achieve such an comprehensive and practical certification system, sustainable biochar production and application should be considered as two sides of the same coin (Verheijen *et al.* 2015a).

2. Biochar-associated contaminants – bioavailable concentrations

Previous studies suggested that biochar total concentrations of organic (Hale *et al.* 2012) and inorganic (Freddo *et al.* 2012) contaminants are often comparable to those in other soil additives or natural soils, including in natural reference substrates such as Lufa 2.2 soil (Bastos *et al.* 2014b). Despite the specific mechanisms having not yet been identified, it is likely that the potential to induce toxicity to exposed organisms can be attributed to water–extractable organic and inorganic contaminants in certain biochars, individually or interacting synergistically and/or antagonistically (e.g., Smith *et al.* 2013; Bastos *et al.* 2014a). According to the equilibrium partitioning theory, toxicity to soil and sediment dwelling organisms upon exposure to toxicants occur via the aqueous phase, this being the main uptake route for most representative soil organisms (Belfroid *et al.* 1995; Janssen *et al.* 1997).

2.1. The strong sorption capacity of biochar

While the definition of bioavailability in the section above refers to the effects of contaminants in biochar for the ecosystem, this chapter sheds light on the exposure assessment by means of traditional phase distribution models, assessed by chemical proxies that are used to evaluate the bioavailability and -accessibility of contaminants in biochar.

2.1.1. Sorption and desorption of organic compounds

As discussed in Section 1.1., PAHs are among the priority contaminants in biochar. Therefore, the following section focuses on PAHs in biochars. Nevertheless, the sorption behavior of a variety of organic contaminants and biochar has been tested, including personal care products, pesticides, and pharmaceuticals (Yao *et al.* 2012; Jung *et al.* 2013; Zheng *et al.* 2013a; Lian *et al.* 2014; Xie *et al.* 2014; Ahmed *et al.* 2015; Calisto *et al.* 2015). Similar to the aqueous phase that determines the uptake route for organisms in ecotoxicity, it is the freely dissolved (aqueous) concentration (also C_{free} or C_W), thus molecules that are not bound to particles or dissolved organic carbon (Reichenberg, Mayer 2006), that determines the bioavailability of an organic contaminant. The C_{free} over the total solid phase concentration (C_S) determines the distribution coefficient (K_D) of compounds in a matrix in relation to water. In a recent ring-trial, the binding of PAHs in 21 biochars was found to be strong (Cornelissen *et al.* 2017). Hence, the biochar-to-water distribution coefficient, $K_{biochar}$, of PAHs was compared with that of organic carbon (organic carbon-to-water partition coefficient, K_{OC}) or an examplary soil which contained 5% OC (Fig. 1). The K_{OC} example is taken to represent the average PAH sorption characteristics for amorphous OC not containing any black carbon (Cornelissen *et al.* 2005). The example soil is simply taken to represent a soil containing 5% of this "average" amorphous OC. It can be observed from Figure 1 that the 21 biochars sorbed PAHs on average 400±200 times more strongly than for the example "average" soil, and 20±10 times more strongly than "average" OC.

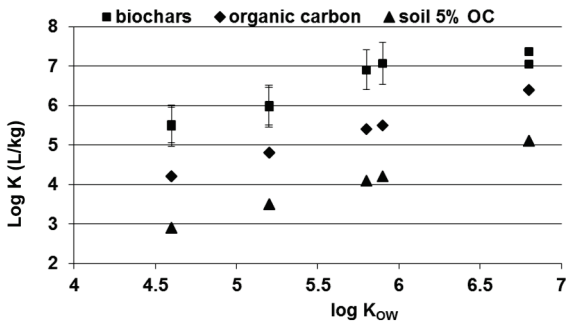

Fig. 1. Sorption (expressed as logarithmic solid-water distribution coefficients, K) to 21 biochars (average and standard deviation), organic carbon (without black carbon) and an examplary soil containing 5% of this organic carbon, as a function of PAH hydrophobicity expressed as octanol–water partition constant (log K_{OW}) (source: Cornelissen *et al.* 2017)

Table 2. PAH octanol-water (K_{OW}) partition constants (Jonker, Koelmans 2002) (numbers without footnote), organic carbon-water ($K_{OC\ amorphous}$) partition coefficients without black carbon calculated ($K_{OC} = 0.35K_{OW}$) from Seth *et al.* (1999), experimentally measured charcoal–water distribution coefficients ($K_{charcoal}$) of Jonker and Koelmans (2002), and biochar-water distribution coefficients ($K_{biochar}$) of medians of 59 biochars (Hale *et al.* 2012)

Compound	Log K_{OW}	Log $K_{OC\ amorphous}$	Log $K_{charcoal}$	Log $K_{biochar}$
Naphthalene	3.3[a]	2.8	n.d.	5.0
Acenaphthylene	4.0[a]	3.5	n.d.	n.d.
Acenaphthene	4.2[a]	3.7	n.d.	n.d.
Fluorene	4.3[a]	3.8	n.d.	5.0
Phenanthrene	4.6	4.1	6.2	5.4
Anthracene	4.6	4.1	7.1	5.5
Fluoranthene	5.2	4.7	6.3	5.7
Pyrene	5.2	4.7	6.4	5.7
Benzo[*a*] anthracene	5.9	5.4	7.5	6.1
Chrysene	5.8	5.3	7.2	5.9
Benzo[*e*]pyrene	6.4	5.9	7.9	n.d.
Benzo[*b*] fluoranthene	5.8	5.3	7.9	6.7
Benzo[*k*] fluoranthene	6.2	5.7	8.1	6.6
Benzo[*a*]pyrene	6.0	5.5	9.1	6.2
Indeno [1,2,3–*cd*] pyrene	n.d.	n.d.	n.d.	7.6
dibenzo[*a,h*] anthracene	n.d.	n.d.	n.d.	n.d.
Benzo[*ghi*] perylene	6.9	6.4	8.2	n.d.

Note: [a] values from Schwarzenbach *et al.* (2003), [b] n.d. = not detected.

Early evidence for limited bioavailability came from Jonker and Koelmans (2001). As shown in Table 2, the charcoal-to-water distribution coefficients ($K_{charcoal}$) from Jonker and Koelmans (2001) for PAHs were comparable to that for $K_{biochar}$. These were about 100–1000 times higher than octanol-water partition constants (K_{OW}), which were proportional to $K_{OC\ amorphous}$ (= $0.35K_{OW}$) from Seth *et al.* (1999) as indicated in Figure 1. By defining organic matter-bound PAHs as bioavailable indicates that only 0.1–1% of charcoal-bound PAHs are bioavailable.

As a consequence from the definition of the partition coefficient ($K_D = C_S/C_W$), either C_S is high and/or C_W is low to obtain such high $K_{biochar}$. Indeed, the C_{free} in a study of Hale *et al.* (2012) ranged from 0.2–10 ng/L, with the highest concentration of 162 ng/L listed for a biochar produced via fast pyrolysis (gasification). These C_{free} are lower than environmental background levels reported by Lang *et al.* (2015). Thus, the bioavailability of biochar-bound PAHs is probably low in the short-term after soil application.

From the chemical sorption point-of-view, a compound is bioaccessible when it has slow desorption rates due to physical obstruction and strong binding of the contaminant to the matrix (Reichenberg, Mayer 2006). It is technically challenging to quantify bioaccessibility of organic contaminants from highly sorptive materials as for instance biochar (Mayer *et al.* 2016). Several papers state the so called infinite sink conditions via depletive methods as a prerequisite to determine the bioaccessibility of hydrophobic organic compounds (i) in matrices that have high $K_{i,D}$s (>3 log units) (Cornelissen *et al.* 2001; Hilber *et al.* 2009; Mayer *et al.* 2011; Gouliarmou, Mayer 2012; Collins *et al.* 2013; Mayer *et al.* 2016).

Here, we define the sorption capacity ratio (SCR) for a given contaminant i as the ratio of the sink material m_{sink} (e.g., a polymer) times its $K_{i,D}$ to the sample material m_{sample} times its $K_{i,D}$ (eq. 1). The product of $K_{i,D}$ and m_{sink} must be bigger than the one of the sample so that the sink can act as such for a contaminant (i.e., $SCR > 1$). In practice, a method can only be depletive if the *SCR* for a contaminant used in a desorption experiment is factors or magnitudes greater than 1 (i.e., $SCR >> 1$).

$$\text{with } SCR = \frac{m_{sink}}{m_{sample}} \cdot \frac{K_{i,sink}}{K_{i,\,sample}}. \qquad (1)$$

Mayer *et al.* (2016) showed that evaluating the bioaccessibility in matrices with such high K_Ds as biochar (Table 1) is very challenging because the usual sink materials such as Tenax® or silicone rods do not provide a high enough SCR. The only matrix fulfilling the infinite sink condition (SCR > 1) in presence of biochar is charcoal (Table 1) or activated carbon (AC, similar K_Ds as charcoal), if used in excess (i.e., $m_{sink} > m_{sample}$). To this end, Mayer *et al.* (2016) used so called "contaminant traps", i.e., glass jars with a silicone/AC layer, as sinks for PAHs from biochars.

The mass ratio was 500 originating from 50 g AC in the trap layer and 0.1 g biochar. Although this method is appropriate because the infinite sink condition is given (SCR > 1), the bioaccessibility cannot be assessed directly because the trapped PAHs in the silicone/AC layer cannot be extracted. Instead, it is assessed indirectly by extraction of the desorption resistant PAHs after the incubation of biochar to the contaminant traps (Mayer *et al.* 2011), and in comparison with PAHs quantified in biochar controls in absence of the sink. Results by Mayer *et al.* (2016) revealed that the contaminants in two investigated biochars were highly desorption resistant. All PAHs scattered around the 1:1 line indicating equal concentrations in the control (–trap) and the container with the sink layer (+trap, Fig. 2). Thus, the bioaccessibility of PAHs in biochars was very limited (Mayer *et al.* 2016). Nonetheless, a range of biotic (e.g., biological activity) and abiotic (e.g. time scale, climate, natural organic matter) factors may act on biochar's physical and chemical properties over time, resulting in increased biochar mobility (Jaffé *et al.* 2013; Verheijen *et al.* 2014) and/or bioaccessibility/bioavailability of a contaminant fraction.

2.1.2. Sorption of heavy metals

Before any beneficial or adverse effect of biochar and/or contaminated soils appear, the elements have to become plant available for root uptake (Semple *et al.* 2004). Thus, the elements need to be dissolved in the pore water. Here the sorption ability of biochar plays an important role. The organic matrix of biochar may immobilize and thereby reduce the bioavailability of heavy metals either existing in biochar or in the surrounding soil solution (Park *et al.* 2011). The ability of immobilizing heavy metals is achieved by different physical and chemical binding mechanisms of the biochar and are direct sorption mechanisms. The high sorption capacity of biochar to metal elements is made possible by high specific surface area, high cation exchange capacity (CEC) and long residence time in soil (Beesley *et al.* 2015). Biochar surfaces may be modified during pyrolysis or by post-treatments with reagents inducing artificial aging to increase the binding capacity deliberately and for specific purposes (Chen *et al.* 2011; Fristak *et al.* 2014).

Biochar that has been freshly produced at high temperatures (>500 °C) contains relatively few functional groups (e.g., carbonyl, hydroxyl, carboxyl or phenolic groups). However, subsequently the reactions of biochar with oxidizing additives, atmospheric oxygen, microbial metabolism or enzymatic breakdown may create or modify functional groups. These provide a negative charge to the biochar surface and a high CEC. Biochar produced at lower temperature is characterized by higher CEC even as fresh material (Beesley, Marmiroli 2011). The sorption of metals frequently is accompanied by a release of protons (Uchimiya *et al.* 2010c).

Electrostatic interactions between positively charged metal cations and the π-electrons of the ligands of the aromatic structures of biochars may also contribute to immobilization of heavy metals (Gomez-Eyles *et al.* 2013). As the aromatic condensation of biochar increases with higher pyrolysis temperature and the oxygenated functional groups decrease (McBeath *et al.* 2015), the proportion of cations sorbed by electrostatic binding increases. Biochar produced at lower temperatures conserves more functional groups because of incomplete pyrolysis, but these are less resistant to modification or degradation when deployed as soil

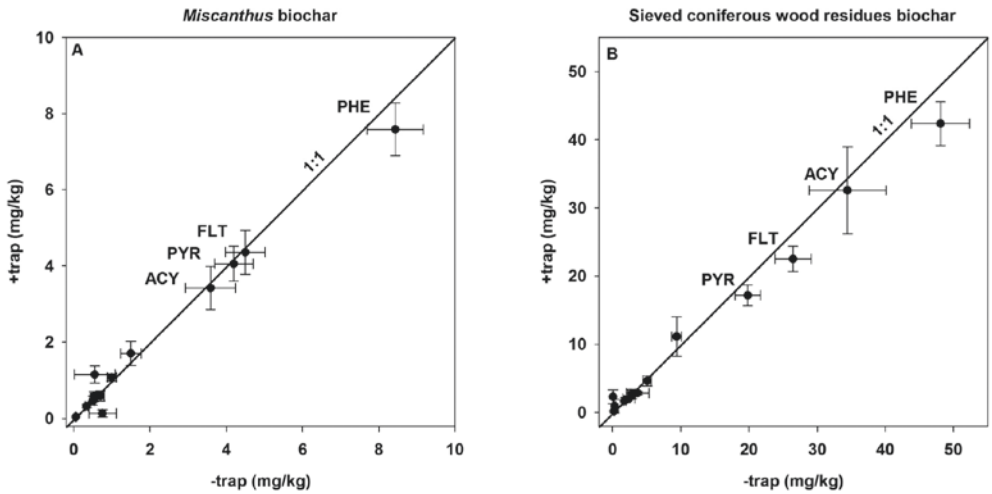

Fig. 2. Scatter plots of remaining PAH contents of two biochars (panel A and B) after their incubation in either contaminant traps (+trap) or similar controls without the infinite sink (–trap). Error bars indicate the standard deviation of duplicates and fall within the symbol if not visible. Individual PAH that showed the highest concentrations are phenanthrene (PHE), fluoranthene (FLT), acenaphthylene (ACY), and pyrene (PYR). (source: Mayer *et al.* (2016) modified. "This is an unofficial adaptation of an article that appeared in an ACS publication. ACS has not endorsed the content of this adaptation or the context of its use.")

additive, thereby releasing again previously sorbed metals (Mukherjee *et al.* 2011).

Indirect mechanisms rely on the effects of biochar on soil characteristics, which subsequently affect the release or binding of heavy metals or metalloids. The mixing of biochar with soil frequently results in pH-changes (Kloss *et al.* 2014c). Minerals in the feedstock become constituents of the ash fraction of biochar. They cause a high pH of biochar and facilitate the precipitation of Cu, Cd, Pb or Zn as phosphates or carbonates at the biochar surface (Beesley *et al.* 2015). Non-modified plant-based biochars usually have pH-values in the range of 8–9.5 and reduce the solubility of metals by the formation of metal hydroxides (e.g. $Zn(OH)_2$) (Brümmer 1986). The situation is quite different for metalloids as, e.g., As. Its solubility and availability increases at higher pH (Moreno-Jiménez *et al.* 2012). Antimony and Mo, too, occur as anionic species in the soil and behave similarly to As. Cationic metals that are bound to negatively charged surfaces in soil increase their solubility at low pH as CEC and high proton density are positively related. This example shows that biochar may act positively for cationic and negatively for anionic metal species in the soil. However, there exist modification options for biochar that may reverse the negative face of biochar for anionic forms (Pan *et al.* 2015).

Biochar in soil may (at least in the short term and if the pyrolysis temperature was not high) increase dissolved OC. Some metals and metalloids like Cu and As preferably form complexes with organic materials and thereby enhance their mobility (Kloss *et al.* 2014a). Biochar may also become a source for phosphorus that is bioavailable, depending on the feedstock and pyrolysis conditions. Enhanced P-supply of plants may decrease the uptake of As in the plant roots as both elements use the same uptake system in the roots (Moreno-Jiménez *et al.* 2012; Beesley *et al.* 2013). Increased P supply may also enhance the precipitation of Pb-phosphate if soil pore water is contaminated with Pb, e.g., by mining activities (Karami *et al.* 2011).

The oxygenated functional groups at biochar surfaces may modify the oxidation status of redox-sensitive metals and thus also their speciation. As an example, the ecotoxicologically undesired Cr(VI) may be modified to the less harmful Cr(III) by sorption to biochar surfaces (Bolan *et al.* 2012).

2.2. Contaminant bioavailability and potential ecotoxicity of biochar

Microbial biomass and activity have shown to increase upon biochar amendment (Van Zwieten *et al.* 2010; Domene *et al.* 2014; Domene *et al.* 2015b), as it provides refuge (Jaafar *et al.* 2014, 2015) and nutrients in the case of fresh biochars from nutrient-rich feedstocks (Jeffery *et al.* 2011) or reduces bioavailability of toxic compounds in their environment (e.g., Elad *et al.* 2011; Graber *et al.* 2014). Short-term effects have also been reported on avoidance behaviour and survival of earthworms (*Eisenia fetida*, *Aporrectodea caliginosa*), collembolans (*Folsomia candida*) and enchytraeids (*Enchytraeus crypticus*) as well as on earthworm weight loss and abundance of ant communities, at biochar application rates of 0–14% (w/w) (Liesch *et al.* 2010; Li *et al.* 2011; Busch *et al.* 2012; Hale *et al.* 2013; Tammeorg *et al.* 2014; Castracani *et al.* 2015; Domene *et al.* 2015a, 2015b). Chronic toxicity studies have been performed to a much lesser extent, focusing primarily on reproduction of collembolans and earthworms, for up to 28 and 56 days, respectively (Liesch *et al.* 2010; Li *et al.* 2011; Marks *et al.* 2014; Domene *et al.* 2015a). However, the link between biochar's inherent contaminants and toxicity to soil meso- and macro-fauna remains unclear, with data being often contradictory and influenced by feedstock, pyrolysis conditions, application rates, soil type, pH and the exposed organism (Lehmann *et al.* 2011; Ameloot *et al.* 2013). Yet, further to the aforementioned direct effects, biochar amendment can also impact soil biological activity indirectly. For instance, biochar (10% v:v) applied to contaminated mine soil increased soil toxicity (based on *Vibrio fischeri* luminescence inhibition and *Lolium perenne* germination), due to solubilisation of As in pore water (>2500 µg/L) linked to changes in pH and soluble phosphate (Beesley *et al.* 2014). However, by combining biochar with compost, the amended soil toxicity was effectively reduced, due to decreased metal extractability and increased soluble nutrients (e.g. P; Beesley *et al.* 2014).

Indirect effects from biochar amendment also include the potential risk to aquatic communities by run-off or leaching from amended soils, due to the inherent mobility of soil (Verheijen *et al.* 2015b) and black carbon (Jaffé *et al.* 2013) particles, as well as evidence of biochar dust subjected to wind erosion (Silva *et al.* 2015) and emitted as aerosols (Genesio *et al.* 2016). Recently, Bastos *et al.* (2014a) assessed the effects on aquatic organisms upon exposure to water-extracts of biochar-amended soil at 80 t/ha (mid-point of the biochar concentration range (Jeffery *et al.* 2011)). Although aqueous extracts contained metals (∑metals 96.3 µg/L) and PAHs (∑16 EPA PAHs 106 ng/L) within EU surface water quality targets (2008/105/EC), there was reduced bioluminescence by the marine bacterium *V. fischeri* and 20–25% acute mobility impairment of the invertebrate *Daphnia magna* (Bastos *et al.* 2014a). In contrast, no effects were observed on the growth of the microalgae *Raphidocellis subcapitata* (Bastos *et al.* 2014a).

Interestingly, toxicity to *V. fischeri* exposed to biochar leachates was reduced, by decreasing biochar's inherent PAHs concentrations (and possibly other dissolvable organic contaminants) after pyrolysis, by thermal

post-treatment (Kołtowski, Oleszczuk 2015). In contrast, biochar post-treatment did not have a substantial effect on phytotoxicity or *D. magna's* survival (Kołtowski, Oleszczuk 2015). A recent study by Buss *et al.* (2015) revealed that the presence of VOCs in biochar, such as organic acids and phenols, caused germination inhibition of cress, due to phytotoxic leachates in amended soil, as they are more water soluble and therefore, more mobile than PAHs. Ecotoxicological responses to water or solvent-based extracts in the above studies were also dependent on biochar characteristics and concentration of the extract (Oleszczuk *et al.* 2013; Smith *et al.* 2013). Potential impacts associated to biochar ageing in pristine (i.e. non-contaminated) soil on biota-mediated ecosystem functions, at the relevant time scales, can only be speculated at present, where specific effects and mechanisms are expected to depend on a combination of biochar characteristics, soil type and on-site environmental factors. For instance, various reports suggest that ageing of pyrogenic organic matter may further improve some of its attributed benefits to soil, including the provision of microbial habitat (e.g. Glaser 2007; Mukherjee *et al.* 2014). Yet, it is also possible that physical, chemical and biological weathering of pyrogenic organic matter (e.g. Hamer *et al.* 2004; Mukome *et al.* 2004; Brodowski *et al.* 2005; Cheng *et al.* 2006; Brodowski *et al.* 2007; Hale *et al.* 2011; Sorrenti *et al.* 2016) overtime, may contribute to enhanced desorption and/or bioavailability of some of its inherent toxic elements in amended soils, in the long-term. According to the recently assembled thematic task force on ecotoxicology and functional biodiversity within COST Action 1107 (Verheijen *et al.,* this issue; Tammeorg *et al*., this issue), achieving an acceptable level of scientific understanding of biochar effects on terrestrial and aquatic ecosystems involves further testing of biochar properties and combinations of biochar-soil type, climatic conditions, and a functionally diverse range of representative organisms (Tammeorg *et al.,* this issue). It is also necessary to consider the relevant spatial and temporal scales to biochar, as a soil amendment and an environmental management tool (Verheijen *et al.* 2012; Verheijen *et al.* 2014). Specifically, biochar ecotoxicological studies will need to include representative biochar ageing procedures and modeling approaches that account for the relevant biochar characteristics and environmental factors, to more easily derive and extrapolate long-term effects on biota, in natural systems.

3. Biochar for remediation of contaminated soils

Biochar can be used for the remediation of soils (or sediments) that are contaminated by inorganic and organic compounds. Its high sorption capacity in combination with its high surface area are appropriate to immobilize contaminants. Thus, the contaminants will not be removed from the matrix but sequestered. It is assumed that the contaminants will not be released into the matrix until the biochar is degraded. A detailed discussion about the stability of biochar over time is beyond the focus of this paper. However, mean residence times (MRT) of pyrogenic organic matter (PCM) studied under different field conditions varied widely and ranged from 6 to 5448 years (Lehmann *et al.* 2015). Part of this variability owes to the fact that the models to calculate the MRT differed, or that PCM properties or experimental approaches were different (Lehmann *et al.* 2015).

3.1. Biochar for remediation of soils contaminated with organic compounds

While a considerable number of studies and reviews have been published on the suitability of AC (Hale *et al.* 2015) and references therein) for the reclamation of organically contaminated soils, Hale *et al.* (2015) pointed out in their overview that relatively few laboratory studies were carried out with biochar and, at that time, no field study results were published. Another review by Xie *et al.* (2015) confirmed that the majority of the studies with biochar were conducted in the laboratory. Biochars used for remediation were better suited to immobilize organic than inorganic contaminants (Xie *et al.* 2015) and AC reduced organic contaminants more effectively than biochar (Hale *et al.* 2015). The modest effect was due to the relatively low K_D values of the biochars in comparison to *in situ* K_{OC}. In comparison to these, the K_{OC} values listed in Table 2 are not containing carbonaceous geosorbents such as black carbon, coal or kerogen that naturally occur in organic matter and exhibit a 10–100 times higher sorption capacity than amorphous OC (Cornelissen *et al.* 2005).

A recent field study over two consecutive years where biochar and granulated AC (GAC) were added at 2.5% each in different plots to a field soil that was contaminated with 39 mg kg^{-1} dichlorodiphenyltrichloroethane (DDT) showed that biochar could significantly reduce DDT accumulation in earthworms (49%), whereas the GAC did not and the invertebrates showed toxic effects (Denyes *et al.* 2016). None of the char amendments reduced DDT uptake in plants (*Cucurbita pepo spp. pepo*). The C_{free} was assessed by the polyoxymethylene (POM) passive sampler proxy and correlated well with the availability of the earthworms, but not with the phytoavailability (Denyes *et al.* 2016). So, different endpoints (earthworms, plants, proxy) under real world conditions indicated contradicting results and the form and type of char influences the outcome. In summary, the limited data of only a recent field study with biochar leads to inconclusive results.

Equilibrium times are, besides the weathering of biochars in soils or sediments, important and need to be taken into account before judging over success or failure of remediation of contaminated sites with biochar (or AC).

Positive or immobilization effects cannot be expected to be reached immediately, as equilibration times may be very long for biochar or AC amendment. A polychlorinated biphenyl-contaminated sediment that was amended with 2% AC reached equilibrium after about 2000 to 2500 days (Werner *et al.* 2006). When the batch experiment was mixed, the C_{free} was reduced to a tenth of its original concentration after only 50 days, but when it was left unmixed the same result was reached not until about 900 days (Werner *et al.* 2006).

3.2. Biochar for remediation of soil contaminated with heavy metals

The sorption characteristics of biochar may reduce the bioavailability of undesired elements and reduce ecotoxic effects (Karer *et al.* 2015). Nutrients from organic soil additives in combination with biochar may enhance the productivity of marginal lands with contaminated soils. As for other soil remediation technologies, it is important to survey the site-specific soil and contaminants conditions and to check the efficacy of the intended remediation technology at small scale. Different mixtures of contaminants might require bespoke biochars or combinations of biochars with different sorption characteristics. As shown in Figure 3, that contains results of mostly lab studies, the dominance of the positive or the negative face of biochar for remediation purposes depends on the element and its speciation. Whereas metals occurring predominantly in cationic form will be bound by biochar and therefore show a smiley, anionic metals and metalloids will not be immobilized by biochar and show a frowny. Elements like As, B and Mo that predominantly occur in anionic form are rather mobilized because of negative surface charges of biochar, pH effects and competition with DOC for binding sites (Beesley *et al.* 2014).

In the following we focus on recent field studies about remediation of heavy metal contaminated sites. Similar to Figure 3, the most investigated heavy metals are Cd and Pb in paddy rice soils (Cui e*t al.* 2011; Chen *et al.* 2016) or Zn (and Ni) (Shen *et al.* 2016). The uptake of Cd into rice grain was reduced by 57–86% over two seasons (Chen *et al.* 2016) and by 17–62% over two years (Cui *et al.* 2011). The study of Shen *et al.* (2016) revealed that 0.5–2% biochar amendment showed reduced carbonic acid leachates of Ni and Zn by 83–98% over three years, which was comparable with the cement based treatment carried out at the same site in parallel. Despite the positive aspect that the contaminant uptake into plants was reduced in the above studies, plants react differently to the biochar amendment e.g., different rice cultivars reacted differently on the Cd uptake (Chen *et al.* 2016) or no grass could be planted on the field (Shen *et al.* 2016). These outcomes indicate that though biochar has excellent sorption capacities for heavy metals, other factors like the plant species, the contaminant itself, and the influence of the biochar on soil conditions such as pH, available nutrients, earthworm habitats, etc. (Beesley *et al.* 2015) are important for a successful reclamation of a site.

Finally, Shaheen and Rinklebe (2015) compared different emerging amendments as, e.g., biochar, chitosan, nano-hydroxyapatite and organo-clay and low cost alternative material such as limestone, zeolite, bentonite and others in their immobilizing effects of a Cd and Pb floodplain soil as expressed by rapeseed (*Brassica napus*)

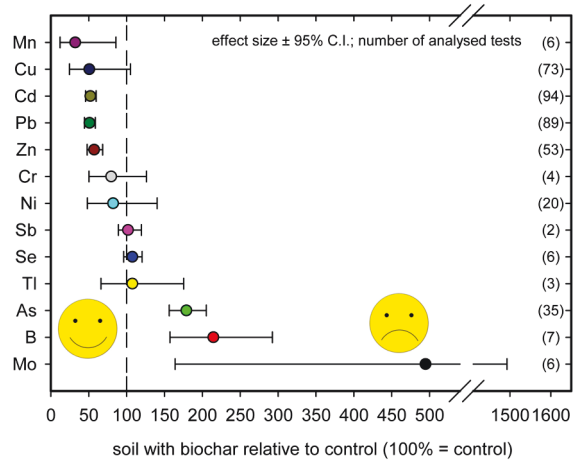

Fig. 3. Changes in the availability of elements from soil to soil solutions (studied with extractants simulating bioavailability for plant roots) in response to biochar additions in lab and field experiments. 100% means identical elemental availability in soils with and without biochar (= treatment and control). 50% means that in soils with biochar the elements were only half as available as in soils without biochar. 200% means that in soils with biochar the elements had double the availability compared to soil without biochar. The reduced availability (up to max. 100%) is a positive effect and therefore marked with a smiley. All studies that showed enhanced availabilities of heavy metals (>100%) after biochar amendments were marked with a frowny. Effect sizes (±95% confidence interval, C.I.) for different elements were calculated with MetaWin 2.1 software. Numbers in brackets give the numbers of control-treatment-comparisons in peer-reviewed literature that fulfilled the data quality requirement for meta-analyses. Data were taken from the following references (Hartley *et al.* 2009; Namgay *et al.* 2010; Uchimiya *et al.* 2010a; Uchimiya *et al.* 2010b; Beesley, Dickinson 2011; Beesley, Marmiroli 2011; Beesley *et al.* 2011; Cao *et al.* 2011; Fellet *et al.* 2011; Karami *et al.* 2011; Park *et al.* 2011; Sizmur *et al.* 2011; Trakal *et al.* 2011; Uchimiya *et al.* 2011a; Uchimiya *et al.* 2011c, 2011b, 2011d, 2012a, 2012b; Buss *et al.* 2012; Choppala *et al.* 2012; Cui *et al.* 2012; Debela *et al.* 2012; Jiang *et al.* 2012a, 2012b; Mendez *et al.* 2012; Zheng *et al.* 2012; Beesley *et al.* 2013; Bian *et al.* 2013; Bolan *et al.* 2013; Chang *et al.* 2013; Gartler *et al.* 2013; Houben *et al.* 2013a, 2013b; Jiang, Xu 2013; Khan *et al.* 2013; Melo *et al.* 2013; Moon *et al.* 2013; Park *et al.* 2013; Sneath *et al.* 2013; Uchimiya, Bannon 2013; Xu, Zhao 2013; Zheng *et al.* 2013b; Ahmad *et al.* 2014; Beesley *et al.* 2014; Bian *et al.* 2014; Brennan *et al.* 2014; Kelly *et al.* 2014; Kloss *et al.* 2014b; Liang *et al.* 2014; Lucchini *et al.* 2014a, 2014b; Rees *et al.* 2014; Wagner, Kaupenjohann 2014; Waqas *et al.* 2014)

uptake in a pot experiment. The biochar and chitosan amendments decreased the Cd concentrations in the plant, but so did zeolite and bentonite. In contrast, the amendment with limestone increased the Cd concentration in *B. napus*. The authors suggested that limestone increased the carbonate fraction ($CaCO_3$ content in limestone is 98.5%), which induced Cd precipitation and complexation with carbonates, and was then mobilized in the acid rhizosphere zone and taken up into the plant (Shaheen, Rinklebe 2015). The researchers proposed to profit from this enhanced phytoextraction for Cd for the use of rapeseed as bioenergy crop. Biochar would indirectly remediate Cd-contaminated soils by phytoremediation. The results of the emerging amendments, except organo-clay, showed also a decreased uptake of Pb into the plant, and so did all low cost amendments except zeolite.

It can be concluded that field– and long–term studies with biochar and contaminated soils are very scarce for both organic contaminants and heavy metals and represent future research needs (Zhang *et al.* 2013). Another aspect of biochar amendment might be the reduced mineralization versus the immobilization and low bioavailability and -accessibility of the contaminants. These effects of opposite directions, immobilization and microbial degradation of biochar or AC amendment need to be carefully and systematically evaluated. Furthermore, the oral exposure of biochar contaminants, in other research field called bioavailability, needs to be assessed in future, similar to works published with soot (Gouliarmou *et al.* 2013; Zhang *et al.* 2015).

Conclusions

A qualitative overall assessment of biochar (mainly) in relation to contaminants, is compiled in Table 3. The positive, negative, and unclear aspects of biochar in these respects are depicted with corresponding emoticons. If the assessment was positive a smiley is put, if negative a frowny and if unclear an indifferent emoticon is placed. The indifferent face prevails while the smiley and frowny are both two times encountered. The indifferent expression reflects outcomes of biochar that are not yet clear as for instance the consequences of the interplay of biochar with the environment over time or for remediation purposes. The behavior of soil, sediment dwelling, or aquatic organisms to biochar in their habitat is species dependent. Effects to biochar might be positive, adverse, or the organism might avoid a biochar amended area. The same holds for plants that root in biochar amended soils. Yield might increase or decrease according to different plant species and in some studies even plant subspecies reacted differently to the same amendment in the same soil.

According to the overall assessment of biochar as a sink or source of contaminants and its suitability for pollutant remediation purposes, the following questions reflecting knowledge gaps and future research directions were identified:

1. How can high quality biochar and a robust contaminant quantification in biochar be assured?
2. How can robust and effect-based assessments of contaminants and contaminant mixtures to biota (plants, soil/sediment dwelling organisms) be achieved?
3. How can the above two points be harmonized towards an international protocol?
4. Long term impacts: What is the influence of time (years, decades) in biochar amended soils/sediments to:
   a. Biota (habitat, biodiversity changes, etc.)?
   b. Biochar itself and its immobilization of organic and inorganic contaminants?
   c. The contaminant's bioavailability and –accessibility and to their ecotoxicological effects?

The first three points address the QA/QC where the analytical methods in the lab (Bachmann *et al.* 2016) including as well as sustainable certification procedures (Verheijen *et al.* 2015a) need to be improved and standardized or, in the latter two cases, even included. Here, we would like to open up the radar to other contaminants such as VOC as pointed out by Buss *et al.* (2015).

The long-term effects of biochar in ecosystems need to be addressed in future that require especially long-term field experiments as also already highlighted in the review of Kuppusamy *et al.* (2016). Ecological effects of biota in soils or sediments can be studied as well as the attenuation effect of the biochar's sorption capacity, the weathering and degradation of biochar and with it the fate of the contaminants inherent in the biochar and the matrix. Researchers from different disciplines would need to collaborate to address as completely as possible the whole topic. Thus, we propose more international research programs as the one of COST Action TD1107 (ref of this volume of JEELM) that help to decide over the positive or negative face of biochar, its inherent contaminants and appropriateness for remediation.

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Table 3. Qualitative overall assessment of contaminant-related aspects of biochar, of biochar itself, and its suitability for remediation of contaminated sites. Emoticons indicate whether the outcome per topic is positive, negative or unclear

Topic	Organic compounds	Heavy metals
Total concentrations inherent in biochars	PAHs can easily exceed the EBC guideline ☹ but less the maximum value of IBI; mechanisms of PAH formation are about to be understood and recommendations to minimize PAHs are communicated 😐	Not a problem if the feedstock is not polluted by heavy metals 🙂 → increase of heavy metals due to concentration in the pyrolysed material ☹
Bioavailability and -accessibility of inherent contaminants	Generally low 🙂	Low due to low total concentrations 🙂
Sorption capacities	Very high (1–100000 times stronger than organic carbon) 🙂 but – 1–100 times weaker if pores are blocked by organic matter 😐 – 1–10 times weaker if the mass transfer of the contaminants to the biochar is incomplete 😐 – 10–100 times weaker due to strong sorption to carbonaceous geosorbents in the soil or sediment 😐	High for cationic heavy metals 🙂 but low for anionic metals and metalloids such as As, Sb, Mo. ☹
Remediation	Can be an attractive alternative to activated carbon 🙂. However, negative sides can be: – Long equilibrium time ☹ – Reduced sorption due to fouling and pore blockage ☹ – pH increase → overliming effect ☹ – Anionic metals and metalloids are mobilized ☹ – Success contaminant and crop species/cultivar dependent ☹ – Ecological biodiversity altered and remediated areas avoided by some species 🙂 The remediation with biochar must be tailored for every situation as there does not seem to exist an allin-one solution 😐. What is therefore the… – Correct dose? – Appropriate particle size? – Appropriate soil incorporation technique? – Appropriate feedstock for the least ecotoxicological effects? – Appropriate pre- or post-pyrolysis modification of the feedstock or biochar? – Appropriate contaminant?	

COST Action TD1107 Biochar as option for sustainable resource management.

Disclosure statement

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