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Organochlorine POPs sequestration strategy by carbonaceous amendments: toward a better understanding of the transfer reduction to laying hens.

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Keywords

Bioavailability ; Exposure ; POPs ; Biochar ; Activated carbon ; Chlordecone ; PCBs ; PCDD/Fs

Abstract

PCBs, PCDD/Fs, and Chlordecone (CLD) are POPs found in soils and transferred to animals through involuntary soil ingestion. In this frame, the amendment of contaminated soil with porous matrices, like Biochars (BCs) and Activated Carbons (ACs), is a promising technique for reducing this transfer. In this study, the efficiency of 3 biochars and 3 activated carbons was assessed by amending 2% (by weight) of these matrices on (i) CLD or (ii) PCBs and PCDD/Fs contaminated artificial soils. Porosity of the carbon-based materials and molecules physico-chemical characteristics were then linked to the obtained results. The concentrations of pollutants were then measured in the egg yolks of laying hens (n = 3), which were fed on a daily basis pellets containing 10% of soil for 20 days. Overall, no significant transfer reduction was observed with the biochar and the granular AC amendments for all the compounds. However, significant reductions were obtained with the two efficient activated carbons for PCDD/Fs and DL-PCB up to 79–82% (TEQ basis), whereas only a slight reduction of concentrations was obtained with these activated carbons for CLD and NDL-PCBs. Thus, (i) biochars were not proven efficient to reduce halogenated pollutants transfer to animals, (ii) powdered AC amendments resulted in reducing the bioavailability of soil POPs, and (iii) the effectiveness of such strategy depended on both characteristics of the matrix and of the pollutants.

1. Introduction

Pollution caused by anthropogenic activities is of growing concern in the past decades, and Persistent Organic Pollutants (POPs), as laid down in Stockholm Convention (UNEP, 2011), represent the most concerning group due to their proven ecotoxicological and toxicological harmful effects and their persistence in the environment. Among these POPs, Polychlorinated biphenyls (PCBs) (Doick et al., 2005), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) (Terzaghi et al., 2020), and Chlordecone (CLD) (Cabidoche et al., 2009) are recognized to persist for decades to centuries in soil due to their retention by the soil organic matter. PCBs and PCDD/Fs are found ubiquitously at background concentrations in soils (Gis Sol, 2011; Meijer et al., 2003) whereas soil of the French West Indies are extensively contaminated by CLD (Cabidoche et al., 2009, Collas et al., 2019, Le Déaut and Procaccia, 2009). Animals may ingest significant amounts of soil (Collas et al., 2019, Jurjanz et al., 2012) and thus be exposed to contaminants accumulated in soil. Laying hens are a particularly exposed species for two reasons: they may ingest up to 25% of soil in nonoptimal rearing conditions (Jondreville et al., 2010, Waegeneers et al., 2009) and they are able to efficiently absorb organochlorine compounds bound to soil particles they ingest (Fournier et al., 2012, Jondreville et al., 2013, Stephens et al., 1995). Such exposure leads to PCBs (Fournier et al., 2015, Fournier et al., 2012), PCDD/Fs (Schuler et al., 1997, Stephens et al., 1995), and CLD (Jondreville et al., 2014) bioaccumulation in egg yolk, adipose tissue, and liver. Moreover, eggs originated from contaminated areas and free-range laying hens may exceed the Maximum Residues Limits (MRLs) as laid down in EU regulation for PCBs or PCDD/Fs (Knutsen et al., 2018) as well as for CLD (Jurjanz et al., 2020). Indeed for eggs, even relatively low concentrations in soil may lead to contamination of eggs above these MRLs for PCBs and PCDD/Fs (Weber et al., 2018) as for CLD (Jurjanz et al., 2020). Soil ingestion constitute the most sensitive exposure pathway to POPs (Weber et al., 2019).

This emphasizes the urgent need to reduce the exposure of laying hens to preserve poultry production in these contaminated areas. Waegeneers et al. (2009) proposed preventive measures to reduce contamination exclusively based on adapted husbandry practices. We propose another strategy based on sequestration of these POPs in order to

reduce their bioavailability and subsequently the contamination of animal foodstuffs. The use of porous matrices, like biochars and activated carbons (ACs), was extensively studied in the past decade. AC was successfully used to eliminate the bioavailability of 2,3,7,8-TCDD in mice (Boyd et al., 2017, Sallach et al., 2019), or of PCDD/Fs and PCBs in laying hens (Fujita et al., 2012). These experiments were conducted using contaminated feed and not soil. The use of biochars or ACs was also demonstrated to be efficient in order to reduce the bioavailability of NDL-PCBs in swine (Delannoy et al., 2014a) or chlordecone in swine (Delannoy et al., 2019, Delannoy et al., 2018) and goats (Yehya et al., 2017) following ingestion of contaminated soils.

The aim of this study was to assess the capacity of several carbonaceous matrices (ACs, Biochars) presenting contrasted porosity characteristics to limit the transfer of organochlorine compounds from contaminated soils to laying hens.

2. Material and Methods

2.1. Production and acquisition of condensed materials

A set of 6 distinct highly carbonaceous materials was obtained: (i) 3 commercial ACs (ROTH Sochiel E.U.R.L., Lauterbourg, France) and (ii) 3 biochars produced by CARBOFRANCE (Montier-sur-Saulx, Lorraine, France) by pyrolysing oak (500 °C or 700 °C) or Japanese knotweed (700 °C). After the pyrolysis process, biochars samples were ground and sieved to < 500 µm.

2.2. Characterization of carbonaceous matrices

BET Specific Surface Areas (SSA) of all ACs and BCs were determined at PrimeVerre Montpellier. The measurements were performed using a "MICROMERITRICS ASAP 2010″ device, based on nitrogen (N_2) adsorption.

Samples were weighed at 10^{-4} g, outgassed at 200 °C for 4 h, then cooled under vacuum at − 196 °C using liquid nitrogen. Nitrogen gas was dosed in controlled increments and, after each dose, the amount of adsorbed gas was determined after reaching the equilibrium pressure. The quantity of adsorbed gas is plotted as a function of the pressure (P/P_0 extending from 0.001 to 0.3, where P is the equilibrium pressure of the adsorbing gas and P_0 is the saturation pressure). The specific surface area was calculated from the amount of gas required to form a monolayer, using the BET equation (Brunauer, Emmett and Teller) (Brunauer et al., 1938).

The Barret-Joyner-Halenda method (BJH) was carried out to determine the volume and pore size distribution (Barrett et al., 1951). The measurements were performed on the basis of nitrogen adsorption-desorption isotherms using a MICROMERITRICS ASAP 2010 device. Gas pressure was gradually increased until all pores were filled with liquid. Then the gas pressure was gradually reduced, evaporating the condensed gas from the system ($0.06 < P/Po < 1$). Evaluation of adsorption and desorption isotherms reveals information about the volume and pore size distribution. The volume and pore size distribution were then determined using the BJH calculation.

2.3. OECD soils preparation

The preparation of artificial soils was carried out at GISFI (Groupement d'Intérêt Scientifique sur les Friches Industrielles, Homécourt - France). Three distinct contaminated sets of artificial soils were produced: (1) the first contaminated by CLD only (2) the second contaminated by a mix of PCBs and PCDD/Fs, and (3) the third remained uncontaminated. 5% of the total mass of sand for each set of contamination was subsampled. The subsamples were contaminated by one of the 2 spiking solutions containing either (1) one mix of NDL-PCBs and PCDD/Fs (2) or CLD. Briefly, those solutions were spread over one of the 2 subsamples of sand using a hexane-methanol solution (50:50; vol:vol) and hand-mixed with a spatula for 10 min. Solvent traces were then evaporated under an extractor hood overnight. Then, the contaminated sand was thoroughly mixed during 24 h using an end-over-end agitator. At last, the sand was mixed with the other constituents according to the OECD guideline 207. In general, the common portion present in each artificial soil contains sand and kaolin (77:22, m:m dry basis) (Sigma-Aldrich, St Louis, USA). A portion of Sphagnum Peat (Tourbe de Sphaigne, Florentaise, Truffaut, Paris) was introduced (10% mass basis of the kaolin:sand mix). Soils were left to age for 1 month prior to subsampling in 7 aliquots for each set. Finally, 6 out the 7 soils were amended by 2% of one of the highly condensed matrices, thoroughly mixed and allowed to age for three additional months. Final soil concentrations were chosen in order to have pollutant levels close to those found in contaminated sites as presented in Table 1. The composition of each artificial soil is presented in Supplemental data Table S1.

2.4. Determination of CLD relative bioavailability

2.4.1. Animals

This study was performed in strict accordance with the recommendations laid down in the Guide for the Care and Use of Laboratory Animals of the French Ministry of Agriculture and Food for Animal Research and European Council Directive (European directive 2010/63/EU) and was carried out in the animal facility of the platform Bio-DA (Université de Lorraine, Vandœuvre-les-Nancy France).

The experimental design was approved by the Ethical Committee of Lorraine (HC-2019–001). Forty-eight 20-week-old laying hens (n = 3 in 14 distinct groups of artificial soils and 2 of control feed without any soil) from Couvoir de la Solitude (Bréchaumont, France) were involved in this experiment. A 10-day acclimation period was applied prior to the start of the 20-day exposure one (Day 1 to Day 20). During the exposure

period, animals were individually weighed once a week and fed daily 47 g of pellets per Kg of BW containing 10% of the soil that corresponds to their treatment. The animals were afterwards kept in individual compartments for 3 h in order to ensure the correctness of the exposure dose, prior to the distribution of a 40 g standard uncontaminated ration provided from C.A.L (Cooperative Agricole Lorraine, Ville-en-Vermois, France). The temperature was kept at 22–24 °C all along this experimental part. Egg production was monitored. Laying rate was above 85% for all hens during the exposure period.

2.4.2. Sampling

From the 18th to the 20th day of exposure, eggs were collected and yolks were pooled by laying hen. Samples were homogenized and stored at − 20 °C before analyses.

2.4.3. Analytical processes

Quantification of PCBs and PCDD/Fs was performed on a purified extract of the biological matrices using Accelerated Solvent Extraction (ASE) and acidified silica columns followed by Gas Chromatography - High Resolution Mass Spectrometry (GC-HRMS) by Departmental Analytical Laboratory of Vendée (Laboratoire de l'Environnement et de l'Alimentation de la Vendée, La Roche-sur-Yon, France). Methodologies are fully described in the document LABERAC/DGAL/DPCB-tma.2 and approved by the French services (accreditation number: 1–1064) as the official reference methodology for determining PCBs, Dioxins, and Furans contents in animal foodstuff in France.

Quantification of CLD was performed on a purified extract of the two biological matrices using Liquid Chromatography-tandem Mass Spectrometry (LC-MS/MS). For the biological matrices, the validated method LSA-INS-016 was used in the Departmental Analytical Laboratory of Morbihan (LABOCEA, Quimper, France). Protocol was extensively described elsewhere (Yehya et al., 2017). Values below LOQ were replaced by LOQ values in the data set.

2.5. Data analysis

2.5.1. Part 3

All analyses were carried out using R software (version 4.0.2, R Foundation for Statistical Computing, Vienna, Austria). In order to assess the impact of BCs and ACs on pollutant concentrations in tissues, a Dunnett test (using the package multcomp, Hothorn et al., 2008, version 1.4.17) was used in order to compare reductions of concentrations with the control SS. Then, the ANOVA procedure followed by the Tukey–Kramer post-hoc test (using the package agricolae, Mendiburu, 2021, version 1.3.5) were used in order to compare the efficiency of matrices. Differences were considered significant at $P < 0.05$.

2.5.2. Relative bioavailability (RBA) estimates calculation

For each pollutant, RBA factors were calculated as the ratio of each pollutant's concentration in eggs from each soil compared to the control: the SS soil. This method was adapted from a previously described method (Delannoy et al., 2014a, Wittsiepe et al., 2007). A linear dose response is a prerequisite for using this method (Littell et al., 1997). This linearity was previously proven for CLD (Jondreville et al., 2013) and PCBs (Fournier et al., 2012) in laying hens. Each laying hen was considered as an experimental unit. Statistical analyses were carried out using R (version 4.0.2, R Foundation for Statistical Computing) on RBA values.

Then, a 90% confidence interval of RBA was calculated according to Student's Law based on our experimental conditions.

2.5.3. Assessing the impact of porosity and characteristics of the molecules to pollutants sequestration

To further analyze porosity characteristics along with molecule properties and RBA factors, a complementary step was performed using a Factor Analysis of Mixed Data (FAMD). Details of the variables and data used to perform this analysis are presented in Table 3 (for porosity characteristics of ACs and BCs) and in Supporting information Table S 2**.** (for molecules characteristics).

Briefly, pollutant characteristics and porosity properties of the different matrices were set as active variables whereas RBA factor and Toxicity Equivalent Factor (TEF) were set as illustrative ones, aiming to derive respectively the influence of each characteristic on sequestration potential and toxicity.

Then, a hierarchical clustering was performed to allow the non-supervised discrimination of 10 clusters on the basis of active variable of FAMD, as no inertia was substantially explained above, using characteristic data only. Each cluster was characterized by illustrative variables. Data analyses were carried out using R (version 4.0.2, R Foundation for Statistical Computing) using the packages FactoMineR (version 2.4) and flashClust (version 1.1.2).

3. Results and Discussion

3.1. Part 1

Overall, a large panel of different textural properties was observed from the set of condensed materials as shown by the specific surface areas ranging from 31 to 1142 $m^2 g^{-1}$ (Table 3). As previously described, biochars displayed the lowest specific surface area (between 31 and 281 $m^2 g^{-1}$) and ACs the highest (between 566 and 1142 $m^2 g^{-1}$). Regarding biochars, BC2 displayed a higher surface area than BC1 (281 ± 11 $m^2 g^{-1}$ vs 31 ± 2 $m^2 g^{-1}$ respectively), confirming that

higher pyrolysis temperature produces biochars with higher surface areas (Chen et al., 2008, Delannoy et al., 2018, Khalid et al., 2020, Park et al., 2015, Yang et al., 2019). In addition, Oak 700 and Knotweed 700 displayed different characteristics such as specific surface area (281 ± 11 m^2 g^{-1} vs 196 ± 6 m^2 g^{-1} respectively), mesoporosity (absence of mesoporosity vs 0.03 cm^3 g^{-1} respectively) and pore width (100% of relative pore distribution between 20 nm and 300 nm vs >97% lower than 20 nm), indicating the role of raw material in generating different porous characteristics and confirming that BC originating from wood material present higher porosities than those originating from herbaceous material (Behazin et al., 2015, Keiluweit et al., 2010, Wang et al., 2015).

Regarding ACs, the highest specific surface area was obtained from both powdered ACs, AC2 and AC3 (1142 ± 31 and 776 ± 21 m^2 g^{-1} respectively). They also present the highest mesopore volume (0.6 and 0.4 cm^3 g^{-1} respectively, Table 4) as demonstrated by the distribution of their corresponding pore width (80% and 76% relative pore distribution between 3 and 20 nm respectively), a high microporosity (18% and 23% relative pore distribution between 0 and 3 nm respectively) and a low macroporosity (2% for both ACs relative pore distribution between 20 and 300 nm). In contrast, the granular AC (AC1) displays the lowest BET surface area (566 ± 5 m^2 g^{-1}), the lowest microporisity (13%) and the highest macroporisity (4%) among ACs.

3.2. Organochlorine POPs relative bioavailability

As expected, highest concentrations of POPs in egg yolks were observed from non-amended soil groups. Quantifiable levels of PCBs and some PCDD/Fs were found in egg yolks from control groups, as they are ubiquitous in the environment including feed.

Within each molecule group (i) NDL-PCBs and (ii) Dioxins and DL-PCBs similar trends in terms of concentrations were observed (Fig. 1). Thus, RBA factors were calculated in order to estimate the pollutant bioavailable fraction for each group (Table 2). Overall, three different levels of concentrations and bioavailabilities were obtained showing the different potential of the carbonaceous material to sequester the POPs. (1) Those presenting the highest levels of bioavailability when exposed to biochar amended soil (41%−96% for PCDD/Fs, 73%−77% for PCB77, 53%−93% for NDL-PCBs). For most of the studied molecules, no significant reduction was observed for biochars amended soils against SS (Fig. 1). (2) Groups exposed to soils amended with AC1 showed lower POPs eggs yolks concentrations and RBA (18%−37% for PCDD/Fs, 51% for PCB77, 45%−88% for NDL-PCBs). At last, soils amended with AC2 and AC3 presented the lowest RBA (67%−74% for CLD, 10%−27% for PCDD/Fs, 34%−47% for PCB77, 69%−80% for NDL-PCBs). Overall, these results illustrate a first hierarchy order to discriminate the carbonaceous materials along their sequestering potential: AC3 =AC2 >AC1 ≥BC3 ≥BC2 =BC1. Another hierarchization may also be derived as molecules appeared to be differently sequestered by carbonaceous material: ∑PCDD/F>PCB77 >CLD> ∑NDL-PCB.

Currently, few data exist in the literature concerning halogenated POPs transfer reduction to animals using such carbonaceous material as soil amendment. Similar reduction levels were reported when using biochars and ACs amendment of a CLD contaminated soil for piglets (36% reduction using biochar, 54–89% reduction using ACs) (Feidt et al., 2021). For PCBs, PCDD/Fs, direct adsorption prior ingestion of biochars or ACs (without soil) was proven to limit drastically the bioavailability of these compounds to rodents (95–99% reduction) (Boyd et al., 2017, Sallach et al., 2019), far more than found in the present study. Indeed, when applied on soil, soil constituents (like soil organic matter) are known to compete with the amended material and limit their transfer and adsorption to the biochars. Koelmans et al. (2009) provided evidence that humic acid, a part of soil organic matter (SOM), could attenuate the POP sorption by blockage of micropores. This attenuation was particularly important for NDL-PCBs, reaching almost one order of magnitude (Koelmans et al., 2009, Pignatello, 2013, Quinlivan et al., 2005). Such competition may explain the differences between congeners as observed in the present study, as OECD artificial soil present an important portion of *Sphagnum* peat, a condensed organic material presenting high amounts of humic acids (Chiou et al., 1997).

Such discrepancies with current literature data concerning efficiency of POPs sequestration may also be caused by the model used to assess the POP transfer. In soil, most of the current data were obtained from plant assays emphasizing greater reduction factors than observed in the present study for PCBs when using ACs: 67%− 96% (Vasilyeva et al., 2010, Vasilyeva et al., 2006) and BCs: 89% (Yu et al., 2009) as well as for pesticides: 80–98% (Guo et al., 1991, Hilber et al., 2009, Hilber and Bucheli, 2010, Rydrych, 1985). Such differences from plants and animal assays may be explained by distinct transfer mechanisms involved and notably the particular ability of monogastric animals to solubilize lipophilic compounds (Chen et al., 2020, Delannoy et al., 2018). Two main processes were previously reported to support the high absorption efficiencies of lipophilic POPs like PCBs: a mediated solubilization through lipidic micelles and POPs concentration-gradient between intestinal lumen and intestinal blood and lymph (Delannoy et al., 2014b). Such processes may also lead to a mobilization of a greater part of the pollutants adsorbed on amended ACs or BCs than through plants assays. Thus, these findings emphasize that soil remediation strategy should be chosen and assessed upon their predicable future uses by a representative model.

3.3. Characteristics of POPs and carbonaceous material driving the sequestration

The effectiveness of the sequestration strategy implemented is known to depend on the adsorption of these pollutants onto the amended material in order to prevent their subsequent physiological absorption. The key step, the adsorption of these pollutants on the carbonaceous material, results from the adequacy of the molecular properties with those of the carbonaceous matrices (Boyd et al., 2017, Johnston et al., 2012, Khalid et al., 2020). To discriminate the most appropriate characteristics of this two-element system (i.e. the

carbonaceous matrices and the molecule), we performed a Factor Analysis of Mixed Data (FAMD) targeted on the descriptive characteristics of these matrices (surface porosity data) and of these molecules (physicochemical properties) (Fig. 2).

Thus, it appears that the characteristics chosen described and discriminated adequately the biochars and molecules used (sum of dimensions 1 and 2 explaining more than 50% of the observed differences). The first dimension represents the data of porosity of carbonaceous matrices and the second dimension the properties of the molecules. We observed that the minimum projection radius, the molar mass and the log K_{ow} are the parameters that discriminate the most of the selected pollutants, as well as the substitution status and the coplanar character of the molecule. For carbonaceous materials, two opposite groups of characteristics discriminate the most of the studied materials: the total porosity, the micro-porosity (0–3 nm), and mesoporosity (4.4–20 nm), on the one hand, and the macroporosity (20–300 nm), on the other hand. Finally, an intermediate porosity between 3 and 4.4 nm appears to be the least discriminating for the studied biochars.

The illustrative variables highlight that the RBA is mainly explained by the effect of the carbonaceous matrices and particularly by the porosity distribution of the latter. Thus, an important nanoporosity or mesoporosity, is negatively related to the levels of bioavailability observed. In contrast, matrices mainly macroporous do not present any RBA reduction. Finally, differences related to the characteristics of the molecules are secondary explanatory factors: planarity, molar mass, log K_{ow} or the minimal projection radius could be observed as factors negatively related to the levels of bioavailability, a substitution presenting higher levels of bioavailability for non-coplanar molecules.

Such results confirm the few available data of the literature concerning the reduction of POP transfer using such carbonaceous material through a soil-amendment strategy.

In the present study, Log K_{ow} was reported as a main characteristics positively correlated to sorption of contaminants onto carbonaceous material (Bacci et al., 1994, Jonker and Smedes, 2000) and is a well-known explaining factor of POP transfer intensity to animals (Amutova et al., 2021, Drouillard, 2001). However, due to similar trends between Log K_{ow} and different characteristics (molar mass and minimal projection radius), the respective importance of these three characteristics could not be determined.Fig. 3.

Planarity effect is also a known explaining characteristic of this transfer. Present data pointed out that transfer of planar molecules (PCDD/Fs and planar DL-PCBs) is far lesser than non-planar ones (NDL-PCBs and CLD). Such trend was depicted also for carbonaceous natural geosorbents proven to sorb far more planar compounds than non-planar ones (Brändli et al., 2008, Cornelissen et al., 2005, Jonker and Smedes, 2000).

3.4. Explanation of RBAs for each pair of matrix-pollutant

Finally, a hierarchical classification was carried out in order to discriminate the pairs of matrix-pollutant and to derive exposure and risk considerations accordingly. It appeared in a first approach that the biochar from oak pyrolyzed at 500 °C (BC1) is the most particular matrix (cf Fig. S19). Indeed, BC1 showed a particular absence of micro- and mesopores. Its average pore diameter was the largest among the studied materials (>50 nm), explaining its low specific surface area compared to the other matrices ($31 \pm 2\ m^2\ g^{-1}$). It showed also no efficiency to limit POPs bioavailability whatever the molecules. A first intent of classification confirmed the particular distinctiveness of this material, limiting comparisons with the others (cf Fig. S19). Thus, in a second step, this material was suppressed from the dataset to compare micro and mesoporous materials between them.

A first level of distinction in the classification discriminates the first 3 clusters (1−3) comprising BC2, BC3 and AC1 from the last two (4 and 5) including AC2 and AC3. Interestingly, this distinction best explained the most the bioavailability results as the first three clusters present the highest bioavailability (cluster 1: RBA of 54 ± 13%, 2: 99 ± 6% and 3: 65 ± 16%), whereas the last two were the most efficient to reduce POPs transfer to piglets (cluster 4: RBA of 49 ± 16%; 5: 15 ± 2%). The obtained dendrogram also highlights that AC or BC porosity properties are the most cleaving characteristics of all to classify pairs of carbonaceous matrices and pollutants. Such results confirm on a larger set of POPs first investigations which evidenced that micro and mesoporosity volumes are of first importance to limit POP transfer as assess for CLD contaminated soil through in vitro (Ranguin et al., 2021, Ranguin et al., 2020) and in vivo (Feidt et al., 2021) assays. Then, it appears that the chemical characteristics of the studied molecules explain the following obtained classification levels by discriminating: (i) Light PCDD/Fs (L-DFs); (ii) heavy PCDD/Fs (H-DFs); (iii) PCB 28 as Light NDL-PCB- (L-NDL-PCB) (iv) CLD along with heavy NDL-PCBs (H-NDL-PCBs) and PCB 77. This distinction further discriminates the groups of molecules most susceptible to be sequestered by such strategy. Thus, CLD and H-NDL-PCBs appeared to be the compounds the less efficiently retained by carbonaceous media as high RBA factors were observed for BC2, BC3 and AC1 (cluster 2: RBA of 99 ± 6%), and few reductions for all molecules except H-DFs for AC2 and AC3 (cluster 4: 49 ± 16%). Such reduction of transfer appeared to be quite insufficient to preserve egg production on contaminated soils. In contrast, for L-DFs and L-NDL-PCBs compounds, higher retentions were obtained, even for BC2, BC3 and AC1 (cluster 1: RBA of 54 ± 13%). Lastly, H-DFs appeared particularly retained through this strategy as they present the lowest values of RBA for AC2 and AC3 (cluster 5: RBA of 15 ± 2%). In this frame, such a strategy may be of help to preserve poultry production on mildly PCDD/Fs contaminated soils up to a TEQ concentration of 5.4 $ng.kg^{-1}$ (Van der Meulen et al., 2008), assuming a 10 g of soil daily ingestion. Indeed, such reduction levels appear of particular interest in the context of home rearing of free-range chickens as high rates of home-produced eggs appeared to exceed EU MRLs for NDL-PCBs and PCDD/Fs (Hoogenboom et al., 2016). Such an extent of reduction may not be sufficient in CLD contaminated areas as even on a mildly contaminated soil (100 µg CLD per kg of soil) concentrations in eggs should exceed French MRLs (Jurjanz et al., 2020).

4. Conclusion

Sequestration strategy reduces significantly the transfer of halogenated POPs, particularly for PCDD/Fs and PCB 77 (up to 82% TEQ basis). However, almost no significant reduction was observed when using the 500 °C biochar, few for the other BCs whereas ACs were the most efficient to limit the bioavailability of these halogenated compounds. Thus, micro- and mesoporous surfaces appeared as the characteristics explaining the most these results.

In addition, the efficiency of the methodology appeared to be compound-dependent as Heavy PCDD/Fs appeared to be the most susceptible compounds to this strategy. This efficiency could be ranked as: H-DF > L-DF > PCB 77 >NDL-PCBs = CLD. Hence, interestingly, risk of PCDD/Fs exposure should be strongly reduced by implementing this strategy.

Further investigations are needed to implement this strategy at field scale and to provide solutions to improve its cost-efficiency. In this frame, the use of local carbonaceous biomass and improvement of the energy efficiency of the pyrolysis could be of particular importance. Such investigations are in progress in French West Indies context to use Sargasso washed ashore and a solar-based pyrolysis in the frame of the project PYROSAR (end in 2025).

Table 1 : Composition of the different artificial soils and treatment of the experiment.

Soil SET		Sand	Kaolin	Sphagnum peat	Activated carbon	Biochar
CLD	Standard Soil (SS)	70%	20%	10%		
	S with oak tree 500 (BC1)	68.6%	19.6%	9.8%		2%
	S with oak tree 700 (BC2)	68.6%	19.6%	9.8%		2%
	S with Japanese knotweed (BC3)	68.6%	19.6%	9.8%		2%
	S with AC1(AC1)	68.6%	19.6%	9.8%	2%	
	S with AC2 (AC2)	68.6%	19.6%	9.8%	2%	
	S with AC3 (AC3)	68.6%	19.6%	9.8%	2%	
PCB, PCDD/Fs	Standard Soil (SS)	70%	20%	10%		
	S with oak tree 500 ($BC1SOT_{500}$)	68.6%	19.6%	9.8%		2%
	S with oak tree 700 ($BC2SOT_{700}$)	68.6%	19.6%	9.8%		2%
	S with Japanese knotweed (BC3)	68.6%	19.6%	9.8%		2%
	S with AC1 (AC1)	68.6%	19.6%	9.8%	2%	
	S with AC2 (AC2)	68.6%	19.6%	9.8%	2%	
	S with AC3 (AC3)	68.6%	19.6%	9.8%	2%	
Without contamination	Standard Soil (SS)	70%	20%	10%		
	S with oak tree 500 ($BC1SOT_{500}$)	68.6%	19.6%	9.8%		2%
	S with oak tree 700 ($BC2SOT_{700}$)	68.6%	19.6%	9.8%		2%
	S with Japanese knotweed (BC3)	68.6%	19.6%	9.8%		2%
	S with AC1 (AC1)	68.6%	19.6%	9.8%	2%	
	S with AC2 (AC2)	68.6%	19.6%	9.8%	2%	
	S with AC3 (AC3)	68.6%	19.6%	9.8%	2%	

	BC1	BC2	BC3	AC1	AC2	AC3
CLD	ND	ND	ND	ND	67% [54 ;81]	74% [53 ;96]
2,3,7,8-TCDD	71% [48 ;93]	44% [41 ;48]	41% [30 ;51]	18% [14 ;22]	14% [9 ;18]	11% [7 ;16]
1,2,3,7,8-PCDD	ND	74% [65 ;83]	68% [55 ;81]	28% [24 ;33]	21% [19 ;22]	17% [15 ;20]
1,2,3,6,7,8-HxCDD	ND	79% [76 ;82]	76% [72 ;80]	32% [27 ;36]	17% [17 ;17]	17% [16 ;19]
1,2,3,4,7,8-HxCDD	ND	78% [74 ;83]	76% [69 ;82]	31% [27 ;36]	17% [15 ;18]	17% [15 ;19]
1,2,3,4,6,7,8-HpCDD	ND	81% [74 ;89]	83% [80 ;85]	27% [23 ;31]	12% [11 ;13]	13% [12 ;15]
1,2,3,4,6,7,8,9-OCDD	94% [91 ;97]	ND	85% [81 ;90]	25% [21 ;28]	10% [9 ;11]	11% [11 ;12]
2,3,7,8-TCDF	ND	66% [63 ;69]	61% [47 ;75]	31% [25 ;38]	27% [24 ;30]	23% [19 ;27]
2,3,4,7,8-PCDF	ND	79% [77 ;81]	73% [60 ;85]	37% [30 ;44]	22% [22 ;23]	22% [19 ;25]
1,2,3,6,7,8-HxCDF	ND	77% [75 ;79]	83% [78 ;87]	33% [27 ;38]	17% [15 ;18]	17% [16 ;18]
1,2,3,4,6,7,8-HpCDF	ND	87% [74 ;99]	84% [82 ;87]	33% [28 ;38]	12% [11 ;13]	14% [13 ;15]
1,2,3,4,6,7,8,9-OCDF	ND	ND	ND	30% [25 ;35]	11% [8 ;14]	13% [11 ;15]
PCB77	ND	77% [63 ;90]	73% [52 ;93]	51% [38 ;64]	47% [40 ;54]	34% [27 ;41]
PCB28	53% [28 ;78]	45% [28 ;63]	ND	45% [22 ;68]	50% [42 ;57]	28% [23 ;33]
PCB138	ND	ND	93% [88 ;98]	88% [82 ;95]	80% [65 ;95]	69% [65 ;72]
PCB153	ND	ND	ND	ND	ND	77% [74 ;79]
PCB180	ND	ND	ND	ND	76% [66 ;86]	74% [68 ;81]

Table 2 : Computation of relative bioavailability factors.

Relative bioavailability factors were calculated as the ratio of the mean of each pollutant concentration in egg yolks over the corresponding concentration mean of the non-amended group (SS). Then, the 90th confidence interval was calculated according to the Student law.

Table 3 : Origin and physico-chemical characterization of ACs and BCs

Sample	Furnisher	Form	Origin	Activation	BET surface area (m^2/g)	Volume mesopore (BJH) (cm^3/g)	Relative pore width distribution			
							[0-3] nm	]3-4.4] nm	]4.4-20] nm	]20-300] nm
AC1	Roth (ref: 5966.2)	Granular	Peat	Steam	566 ± 5	0.2	13%	47%	35%	4%
AC2	Roth (ref: 5963.2)	Powder	Peat	Phosphoric acid	1142 ± 31	0.6	18%	32%	48%	2%
AC3	Roth (ref: 865.3)	Powder	Pine	Steam	776 ± 21	0.4	23%	33%	43%	2%
BC1	CarboFrance	Powder	Oak	-	28 ± 2	-	0 %	0%	0%	100%
BC2	CarboFrance	Powder	Oak	-	281 ± 11	-	12%	86%	0.1%	2%
BC3	CarboFrance	Powder	Knotweed	-	196± 6	0.03	15%	68%	14%	3%

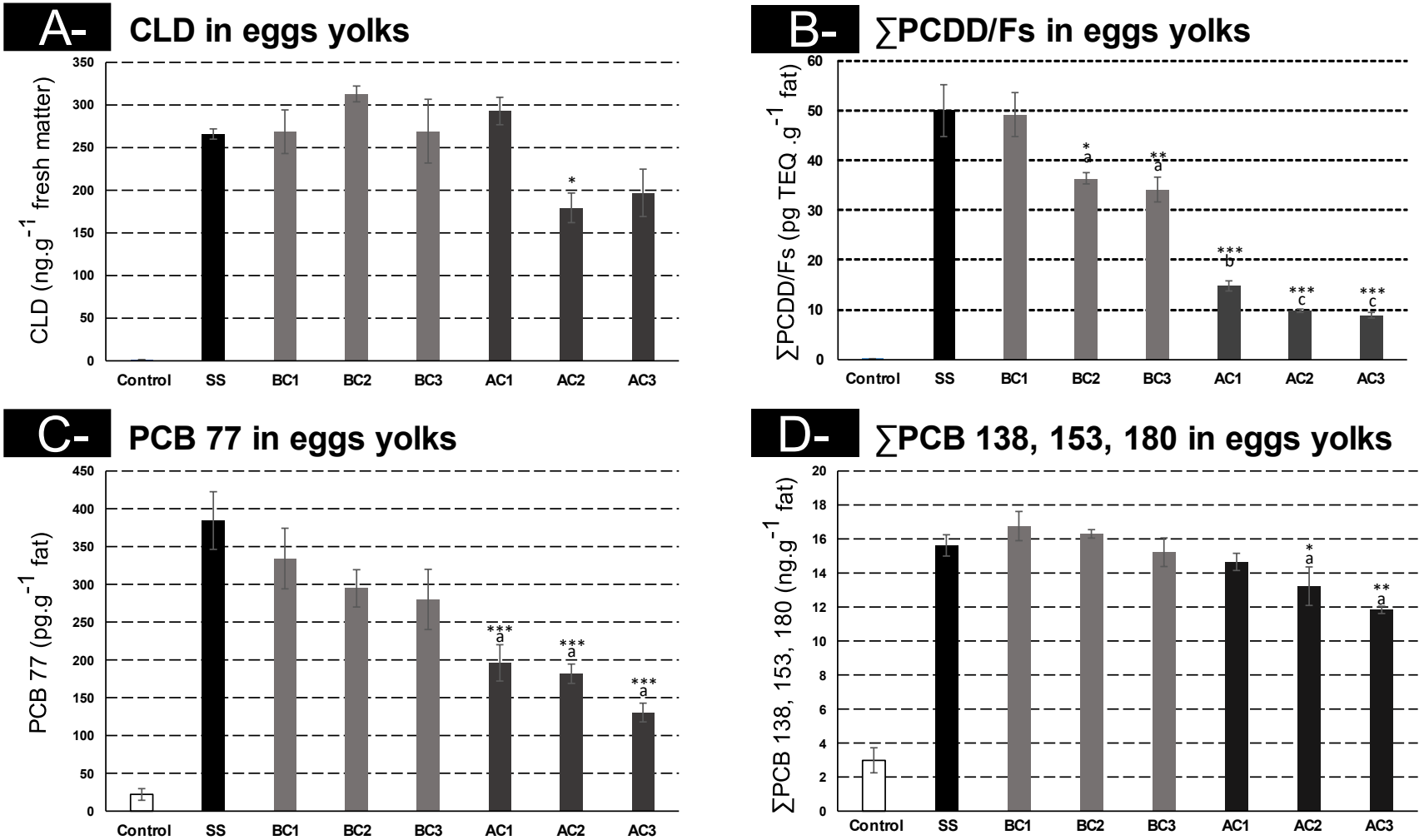


Figure 1: Concentrations of CLD, PCDD/F, PCB 77 and NDL-PCBs in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of POPs in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (*: 0.05>P>0.01; **: 0.01>P>0.001; ***: P< 0.001) using a variance analysis and a Dunnett post-hoc test. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on groups significantly different from Control from previous test.

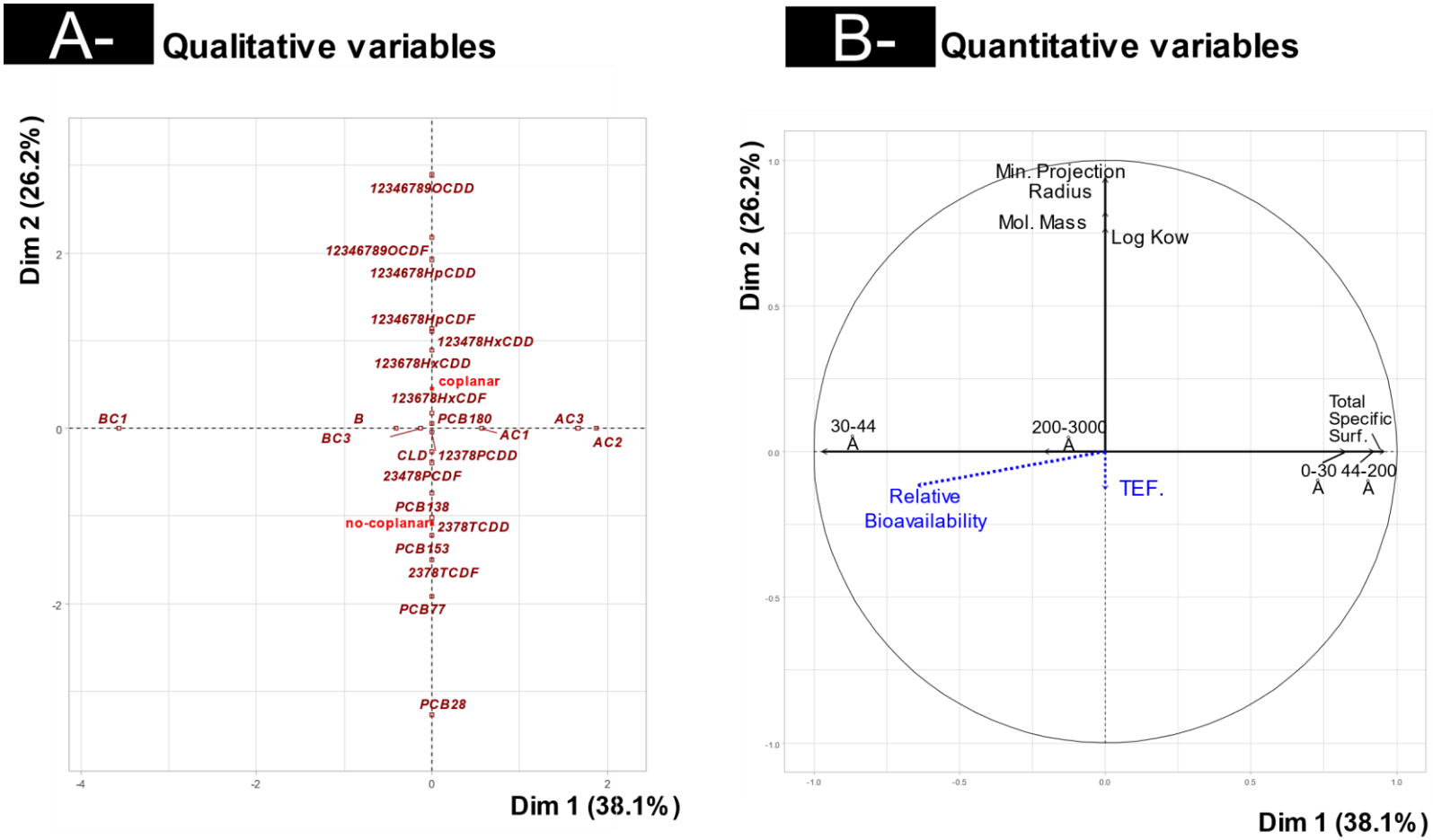
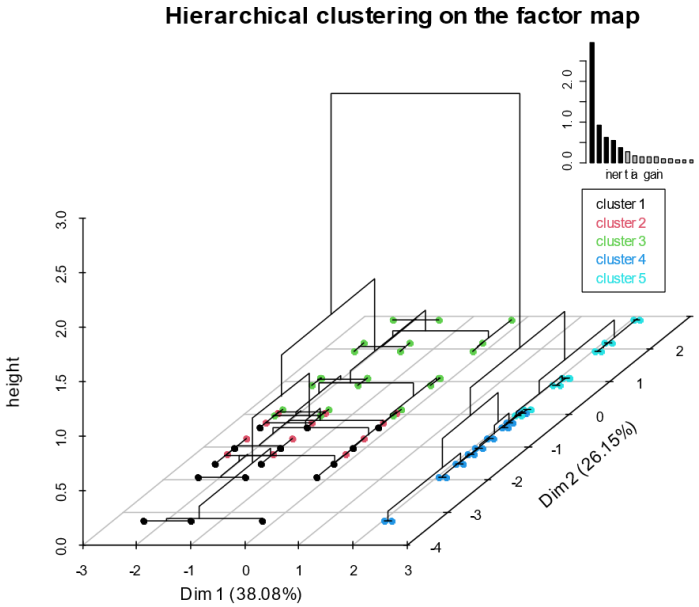


Figure 2: Dispersion of A-qualitative and B-quantitative variables describing carbonaceous materials (Dim1) and molecules (Dim2)
A- the qualitative descriptors used to qualify the molecules are presented in red ordinates (substitution and coplanarity). The congeners, the families were arranged as illustrative variables presented in brown. The different biochars and activated carbons used are shown on the x-axis and as illustrative data.
B- the quantitative descriptors of the materials are presented as follows:
on the abscissa, the explanatory variables describing the porosity of the materials (of the classes 0-30 Å; 30-44 Å; 44-200 Å; and 200-3000 Å) and the total surface area of the material.
on the ordinate, the explanatory variables characterizing the molecules: minimum projection, molecular mass, log Kow.
Finally the TEF, the values obtained of relative bioavailability and the concentrations were arranged as explanatory variables.
The FAMD was performed by using the FAMD function of the R software (v. 4.0.3) using FactoMineR (v. 2.4).



Cluster #		Relative Bioavailability	TEF	BCs /Acs	Compounds
1		54±13% (9)	0.28	BC2, BC3, AC1	L-DF ; L-NDL-PCB; PCB 77
2		99±6% (18)	-	BC2, BC3, AC1	CLD ; H-NDL-PCB
3		65±16% (9)	0.17	BC2, BC3, AC1	H-DF
4		49±16% (12)	0.16	AC2, AC3	L-DF; L-NDL-PCB; H-NDL-PCB; PCB 77; CLD
5		15±2% (6)	0.17	AC2, AC3	H-DF

Figure 3: Unsupervised hierarchical classification of characterization data and cluster composition

For each cluster are presented:

Relative bioavailability factor (%) (mean ± p90), in parenthesis the number of individuals associated with this value.

Average TEF: the average TEF of the cluster

BCs /ACs: matrices present in the cluster

Compounds: compounds present in the cluster:

L-DF (low weight dioxins: 2378TCDF, 2378TCDD, 23478PCDF); H-DF (high weight dioxins: 12378PCDD, 123678HxCDF 123678HxCDD, 123478HxCDD, 1234678HpCDF, 1234678HpCDD, 12346789OCDF, 12346789OCDD); H-NDL-PCB (High weight Non dioxin like PCBs): PCB 138, PCB 153, PCB 180); PCB 77; L-NDL-PCBs (PCB 28)

- : value not computable.

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6. Acknowledgements

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7. Acknowledgements

Nadine El Wanny: Conceptualization, Methodology, Investigation, Data curation, Formal analysis (supporting) Writing – original draft, Visualization (supporting), **Yves Le Roux**: Conceptualization(supporting), Methodology, Data curation, Formal analysis,Writing – review & editing, Visualization (supporting), **Agnès Fournier**: Conceptualization(supporting), Methodology, Writing – review & editing, Supervision (supporting), Project administration, **Moomen Baroudi**: Writing – review & editing (supporting), **Thierry Woignier:** Investigation, Formal analysis, Writing – original draft (supporting), Writing – review & editing, **Cyril Feidt:** Conceptualization, Writing – review & editing, Data curation, Funding acquisition, Visualization (supporting), **Matthieu Delannoy**: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Writing – original draft, Writing – review & editing, Project administration, Visualization.

8. Declaration of Competing Interest

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

Supplemental Data to :

Organochlorine POPs sequestration strategy by carbonaceous amendments: toward a better understanding of the transfer reduction to laying hens.

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Table S 1: Concentration of pollutants of the two different artificial soil sets.

Soil set		POPs massic concentrations in soil (µg/Kg DM for CLD and PCBs; ng/Kg DM for Dioxins and Furans)
CLD	Kepone	1000
PCB, Dioxin Furans	PCB 77	2.5
	PCB 28	2.5
	PCB 52	4.8
	PCB 101	5.8
	PCB 138	13.5
	PCB 153	13.2
	PCB 180	10.8
	1,2,3,7,8-PeCDD	97.0
	OCDF	242
	OCDD	345
	1,2,3,4,6,7,8-HpCDD	124
	1,2,3,6,7,8-HxCDF	89.0
	2,3,4,7,8-PeCDF	99.02
	1,2,3,4,7,8-HxCDD	79.21
	2,3,7,8-TCDF	43.0
	1,2,3,6,7,8-HxCDD	149
	1,2,3,4,6,7,8-HpCDF	98
	2,3,78-TCDD	34

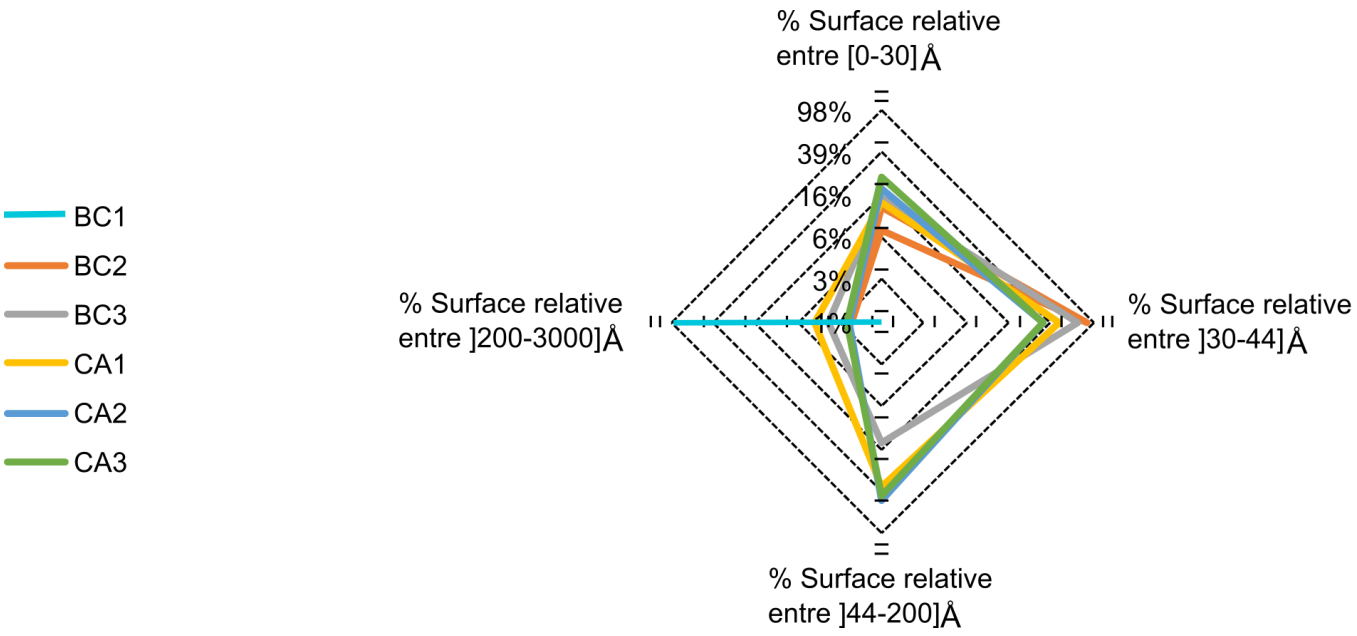

Figure S1: Porosity distribution of carbonaceous matrices

Table S 2: Compounds characteristics used during the data analyses

Compound	POP Family	coplanarity	Molecular mass ($g.mol^{-1}$)	$LogK_{ow}$	TEF (WHO_{2005})	Minimal projection radius (Å)	Position
1,2,3,4,6,7,8,9-OCDD	**Dioxins**	**Y**	459.73	8.4	0.0003	5.35	/
1,2,3,4,6,7,8,9-OCDF	**Furans**	**Y**	443.73	7.9	0.0003	5.13	/
1,2,3,4,6,7,8-HpCDD	**Dioxins**	**Y**	425.29	8.1	0.01	5.05	/
1,2,3,4,6,7,8-HpCDF	**Furans**	**Y**	409.29	7.5	0.01	4.82	/
1,2,3,4,7,8,-HxCDD	**Dioxins**	**Y**	390.85	7.6	0.1	4.86	/
1,2,3,6,7,8-HxCDD	**Dioxins**	**Y**	390.85	7.6	0.1	4.71	/
1,2,3,6,7,8-HxCDF	**Furans**	**Y**	374.85	7.2	0.1	4.48	/
1,2,3,7,8,-PCDD	**Dioxins**	**Y**	356.4	7.2	1	4.44	/
2,3,4,7,8-PCDF	**Furans**	**Y**	340.41	6.8	0.3	4.47	/
2,3,7,8-TCDD	**Dioxins**	**Y**	321.96	6.7	1	4.19	/
2,3,7,8-TCDF	**Furans**	**Y**	305.96	6.2	0.1	4.12	/
CLD	**Kepone**	**N**	490.61	4.5	0	4.9	/
PCB 101	**PCB**	**N**	326.42	6.3	0	4.21	di-ortho
PCB 138	**PCB**	**N**	360.86	6.7	0	4.57	di-ortho
PCB 153	**PCB**	**N**	360.86	6.8	0	4.21	di-ortho
PCB 180	**PCB**	**N**	395.31	7.2	0	4.72	di-ortho
PCB 28	**PCB**	**N**	257.54	5.6	0	3.89	mono-ortho
PCB 52	**PCB**	**N**	291.98	5.8	0	4.31	di-ortho
PCB 77	**PCB**	**Y**	291.98	6.4	0.0001	3.86	non-ortho

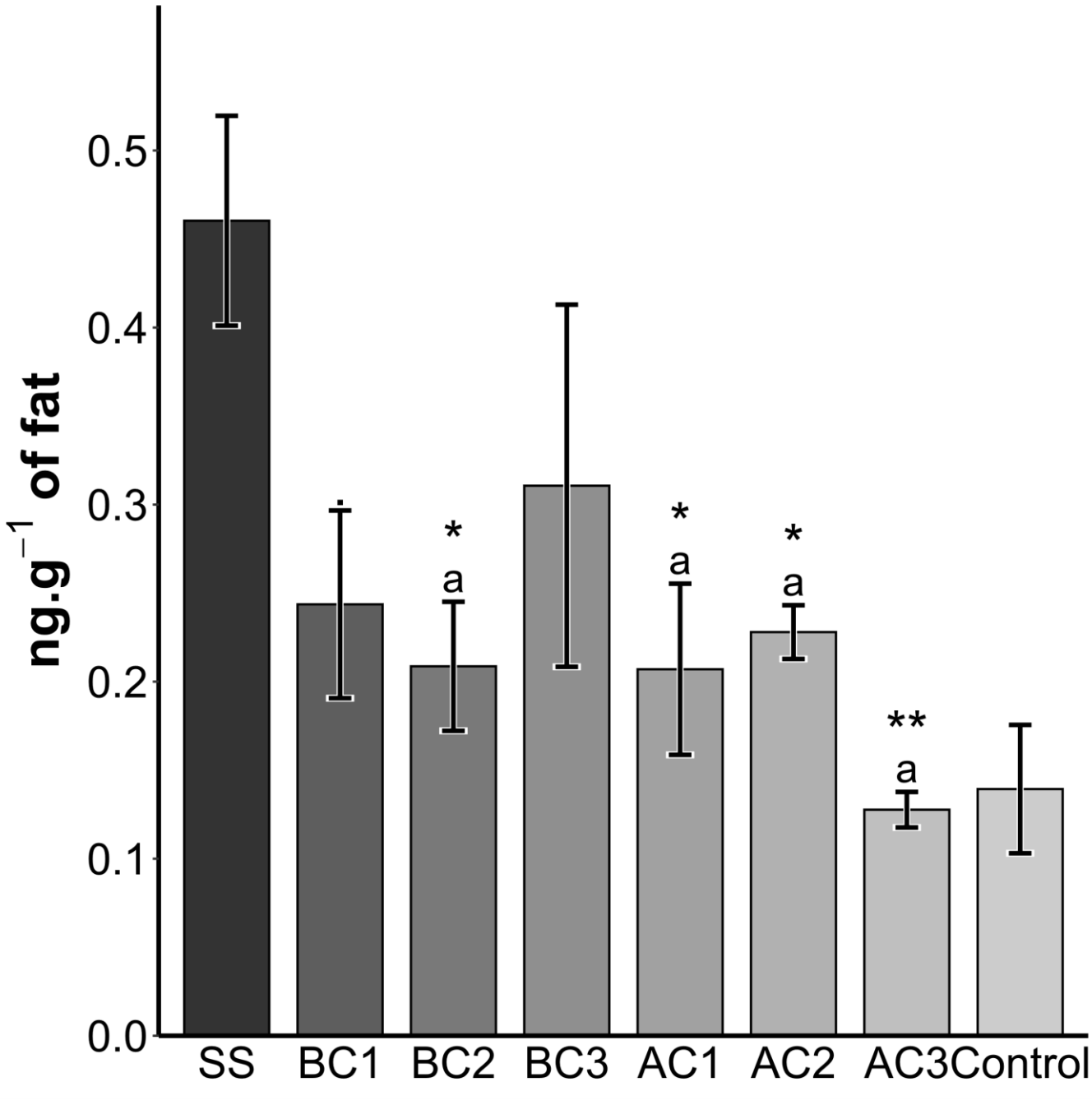

Figure S2: Concentrations of PCB 28 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 28 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using Dunnett(p<0.05).
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

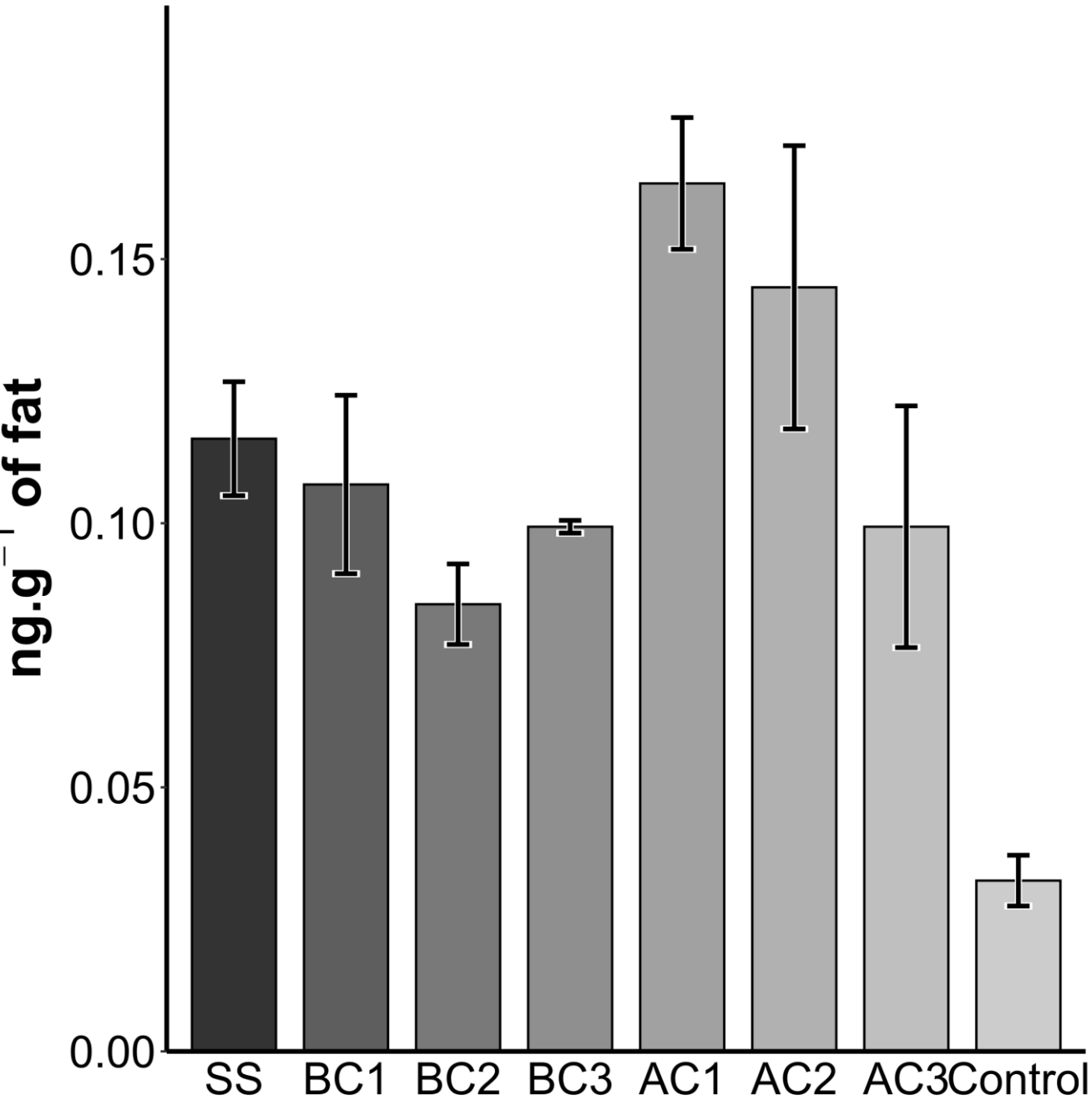

Figure S3: Concentrations of PCB 52 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 52 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

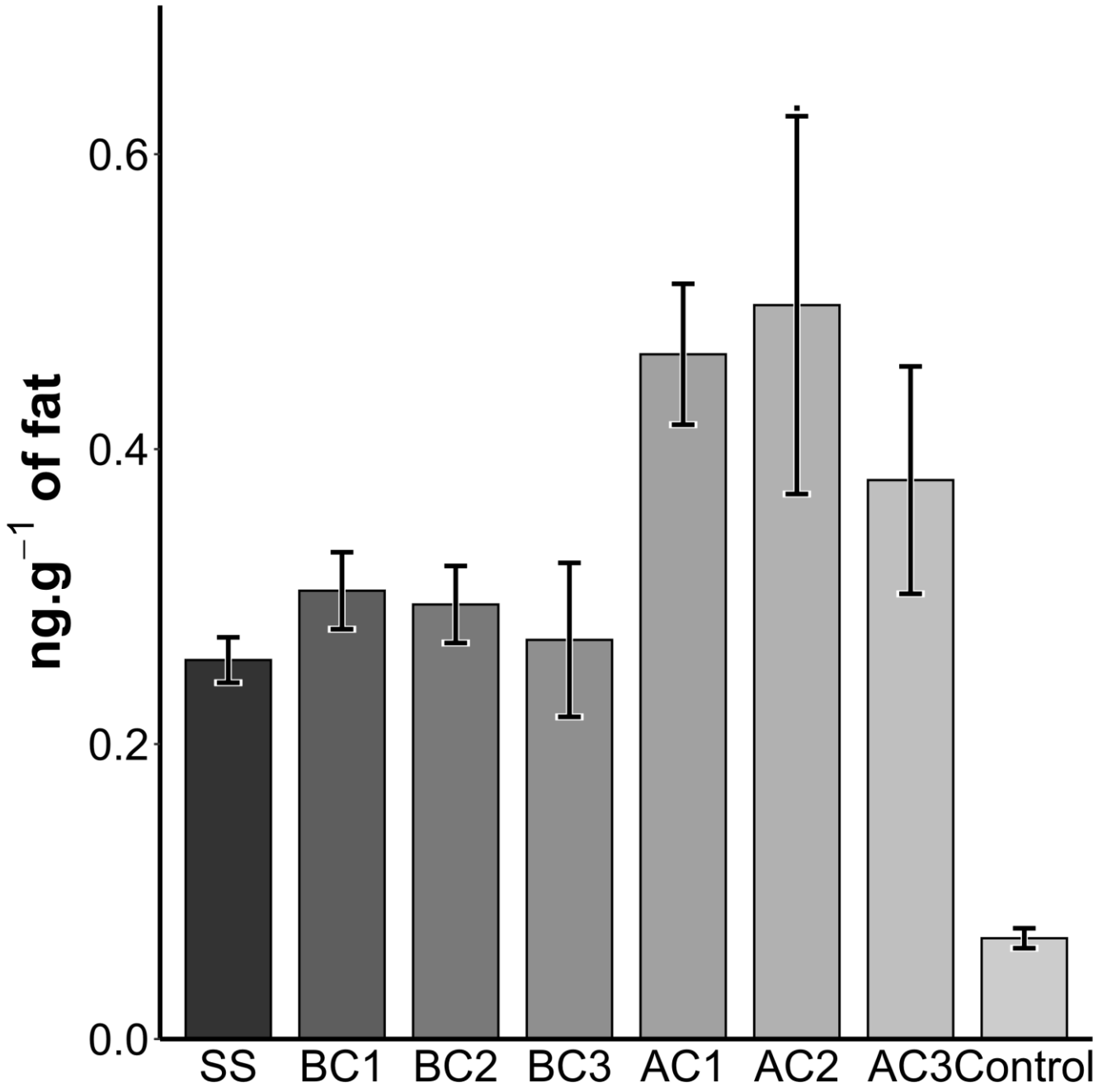

Figure S4: Concentrations of PCB 101 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 101 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

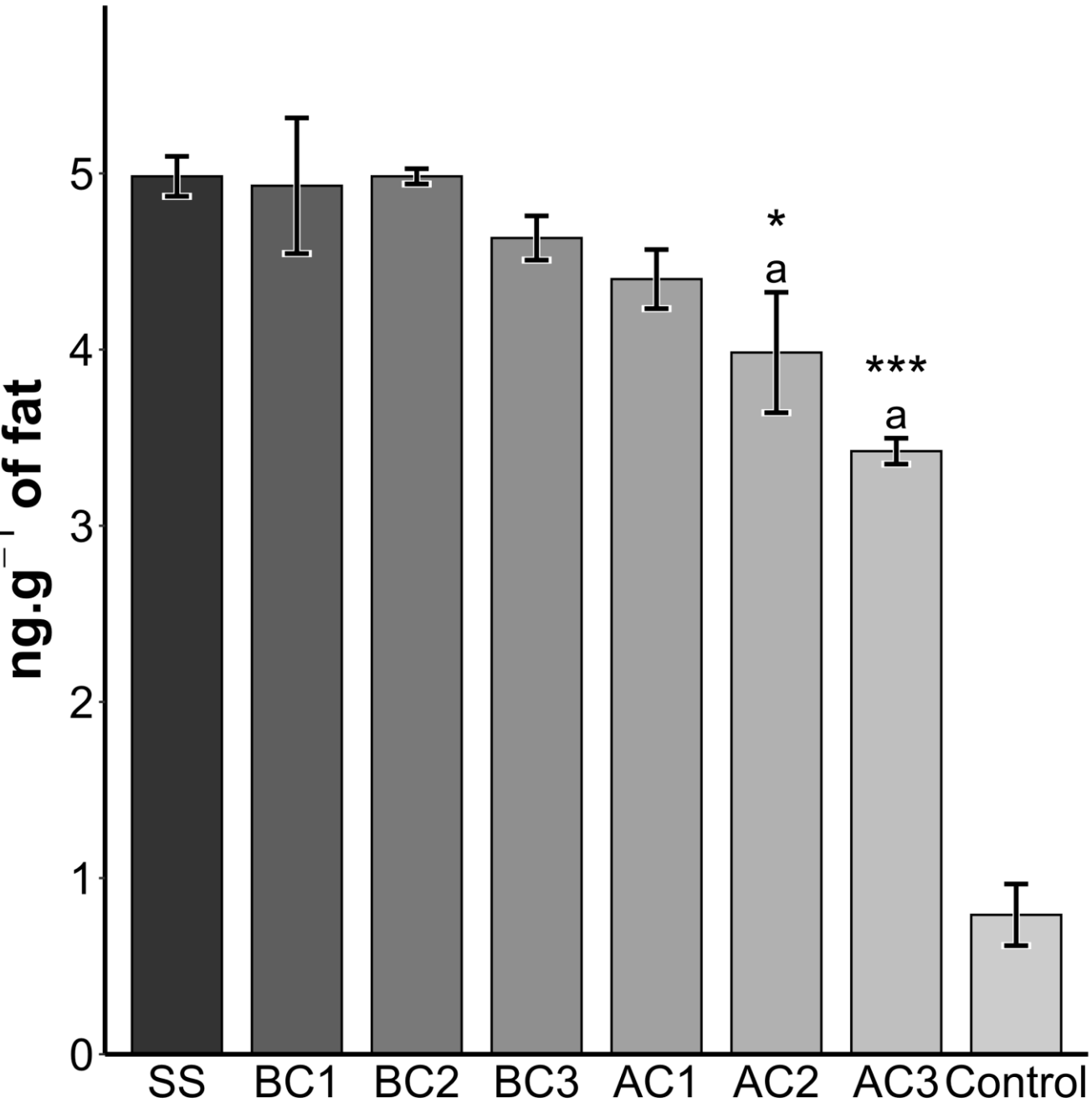

Figure S5: Concentrations of PCB 138 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 138 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

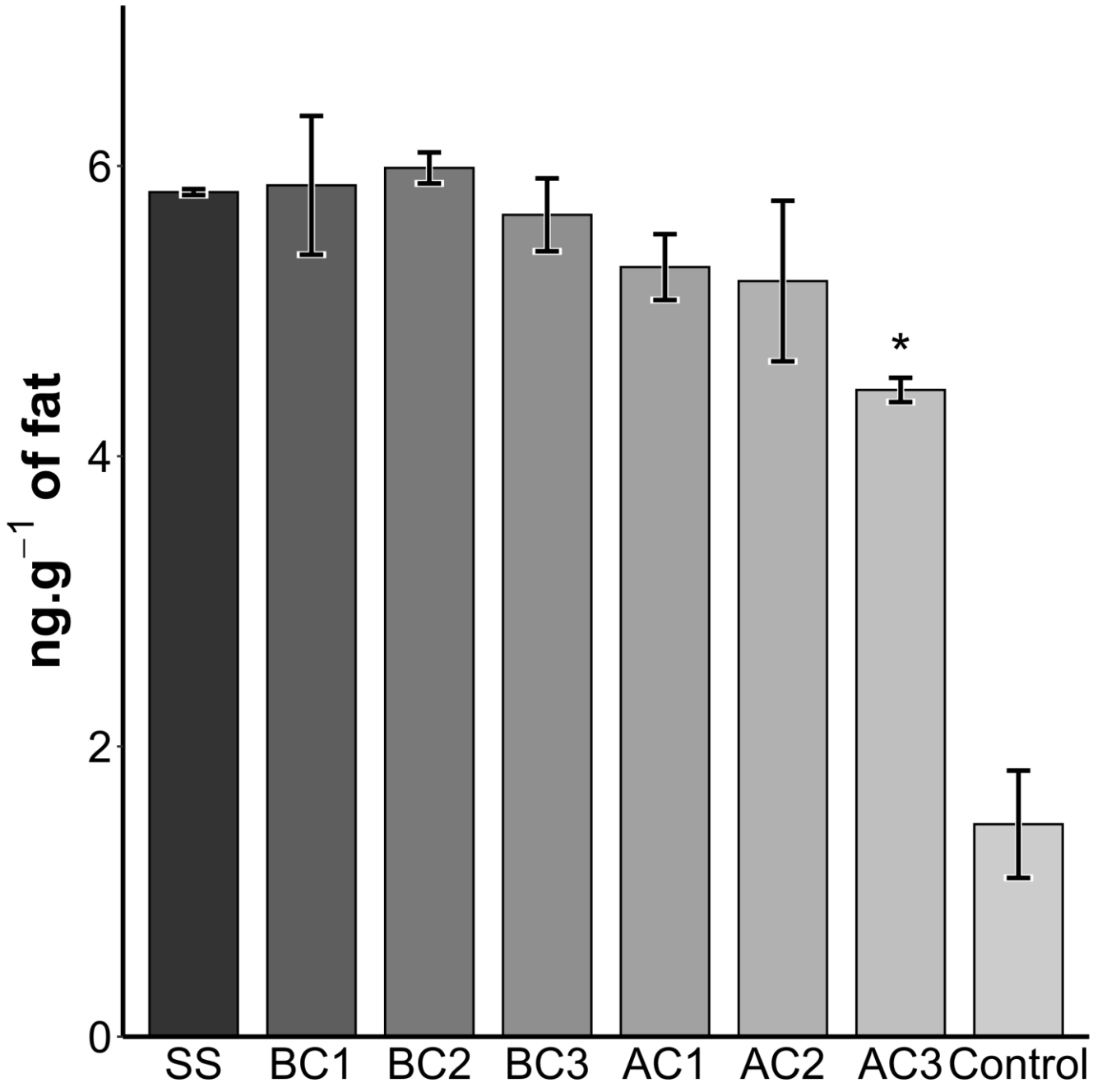

Figure S6: Concentrations of PCB 153 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 153 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

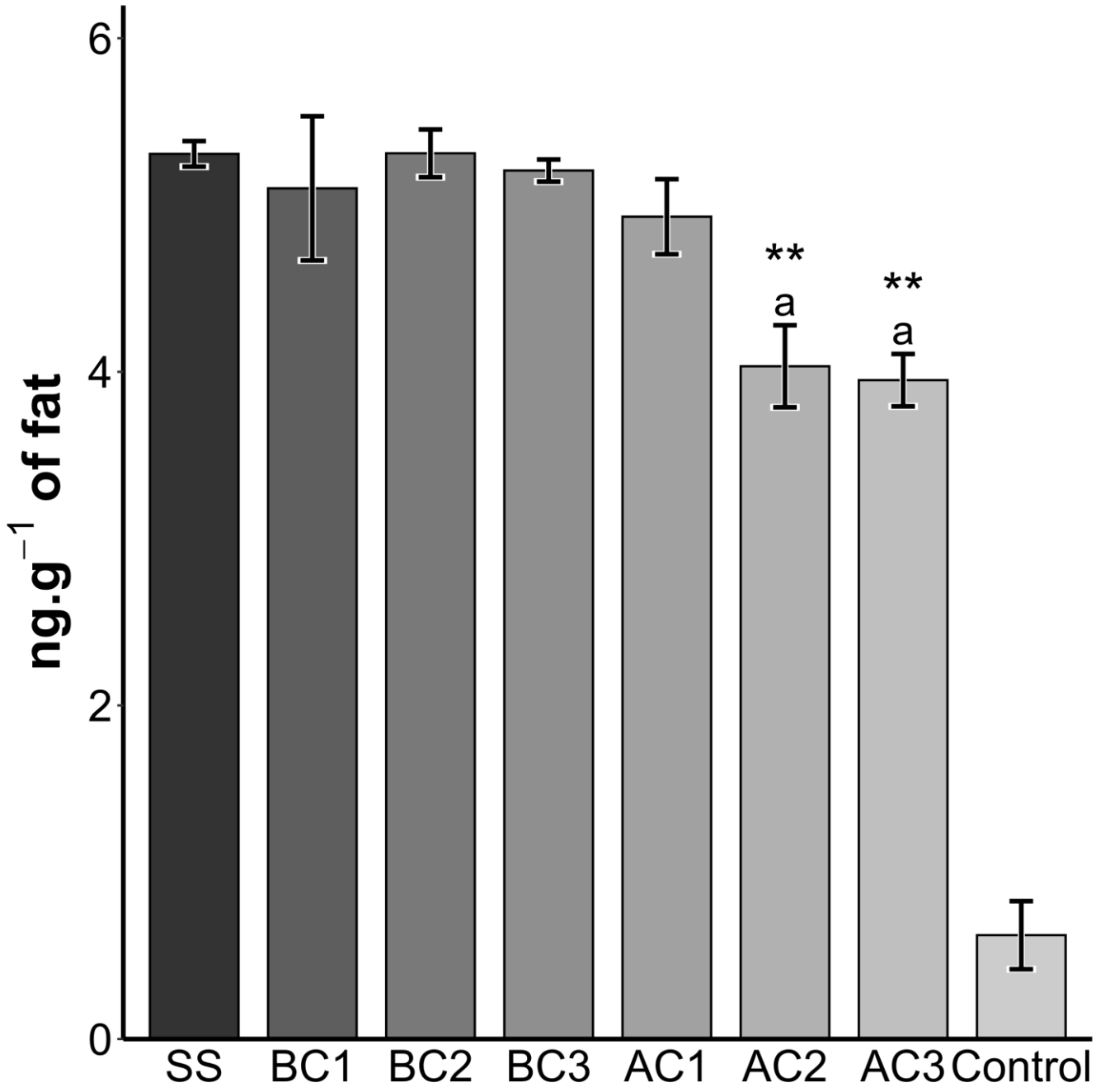

Figure S7: Concentrations of PCB 180 in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of PCB 180 in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

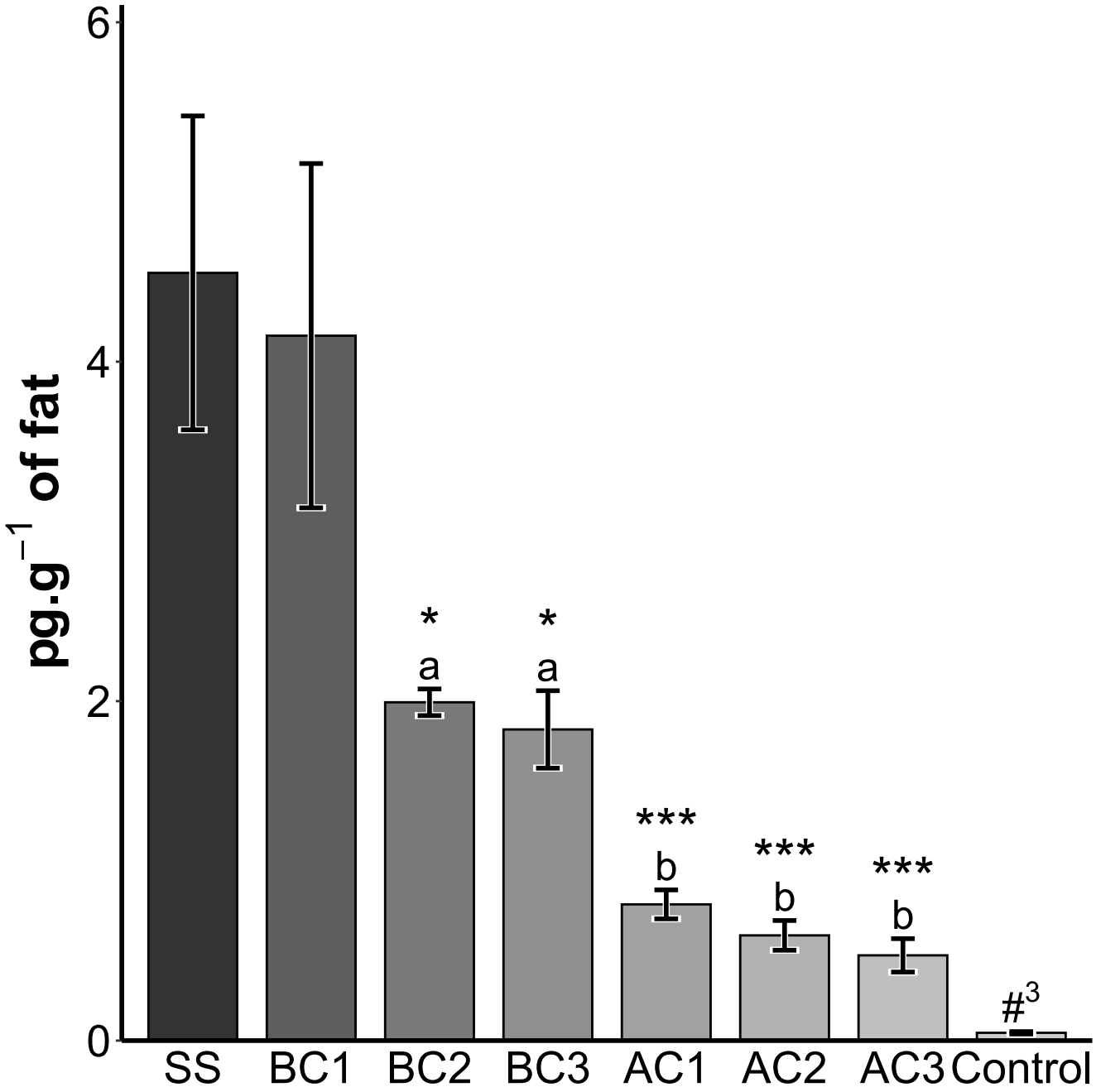

Figure S8: Concentrations of 2,3,7,8-TCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 2,3,7,8-TCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

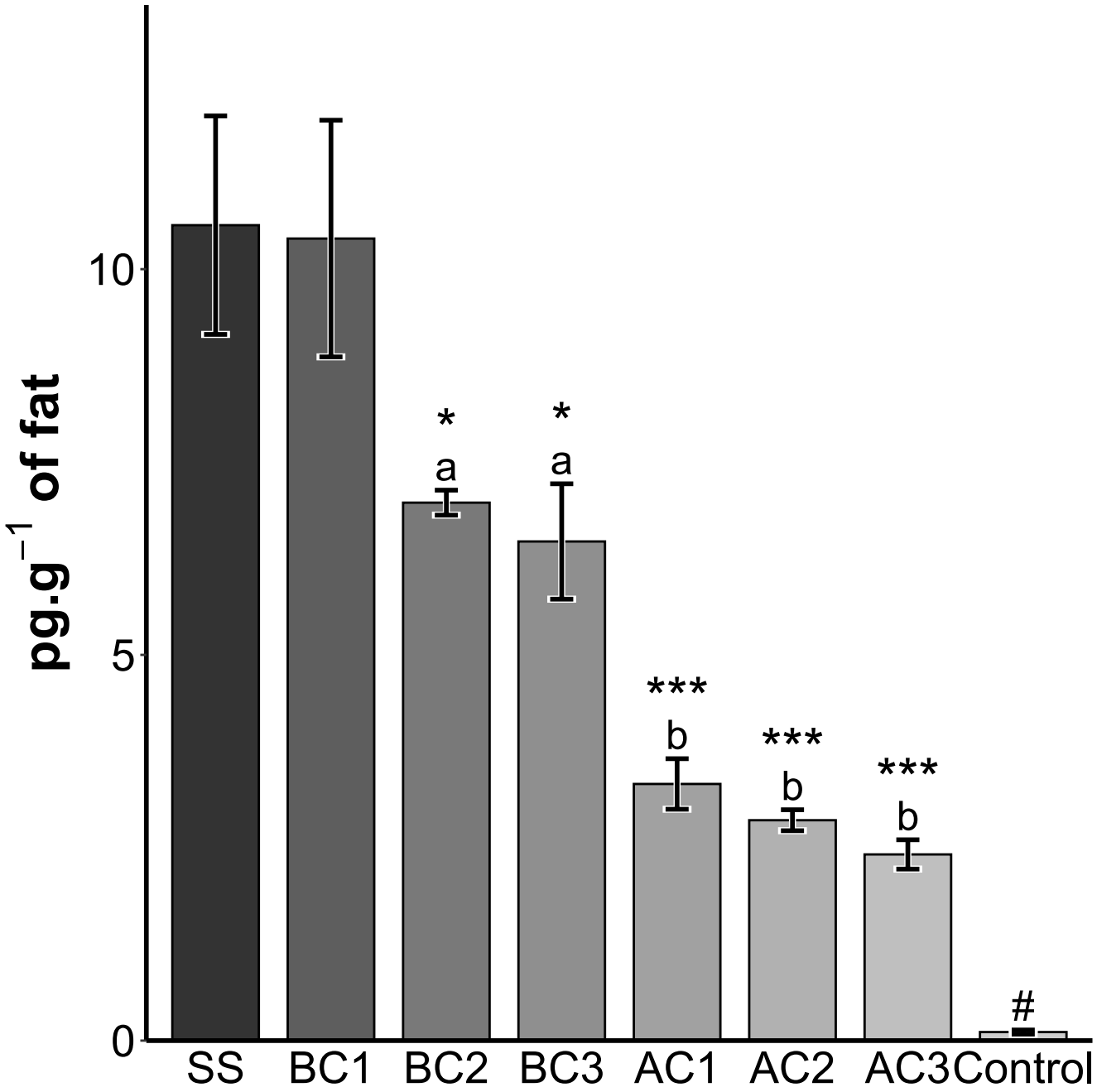

Figure S9: Concentrations of 2,3,7,8-TCDF in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 2,3,7,8-TCDF in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

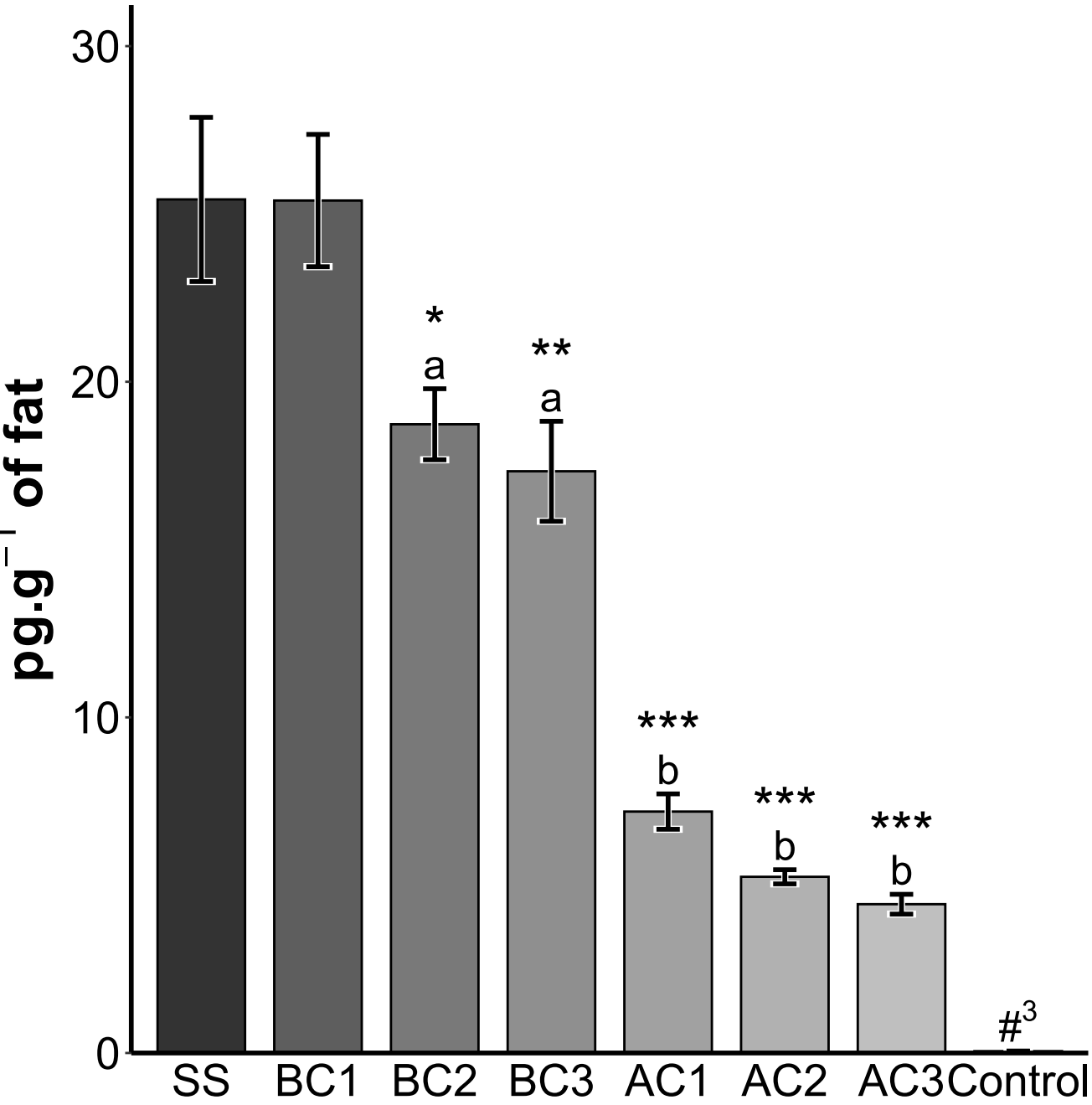

Figure S10: Concentrations of 1,2,3,7,8-PeCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,7,8-PeCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

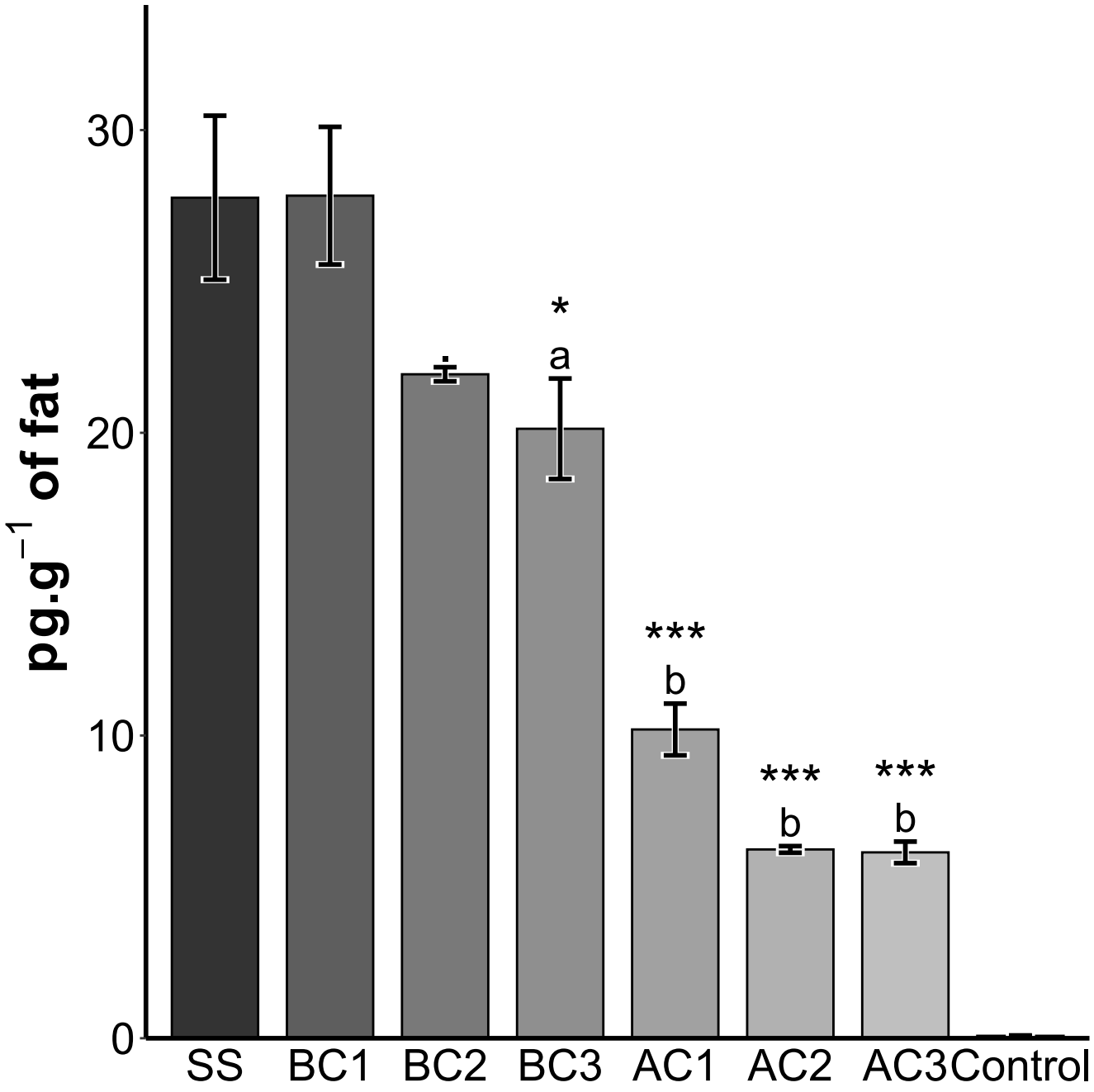

Figure S11: Concentrations of 2,3,4,7,8-PeCDF in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 2,3,4,7,8-PeCDF in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values

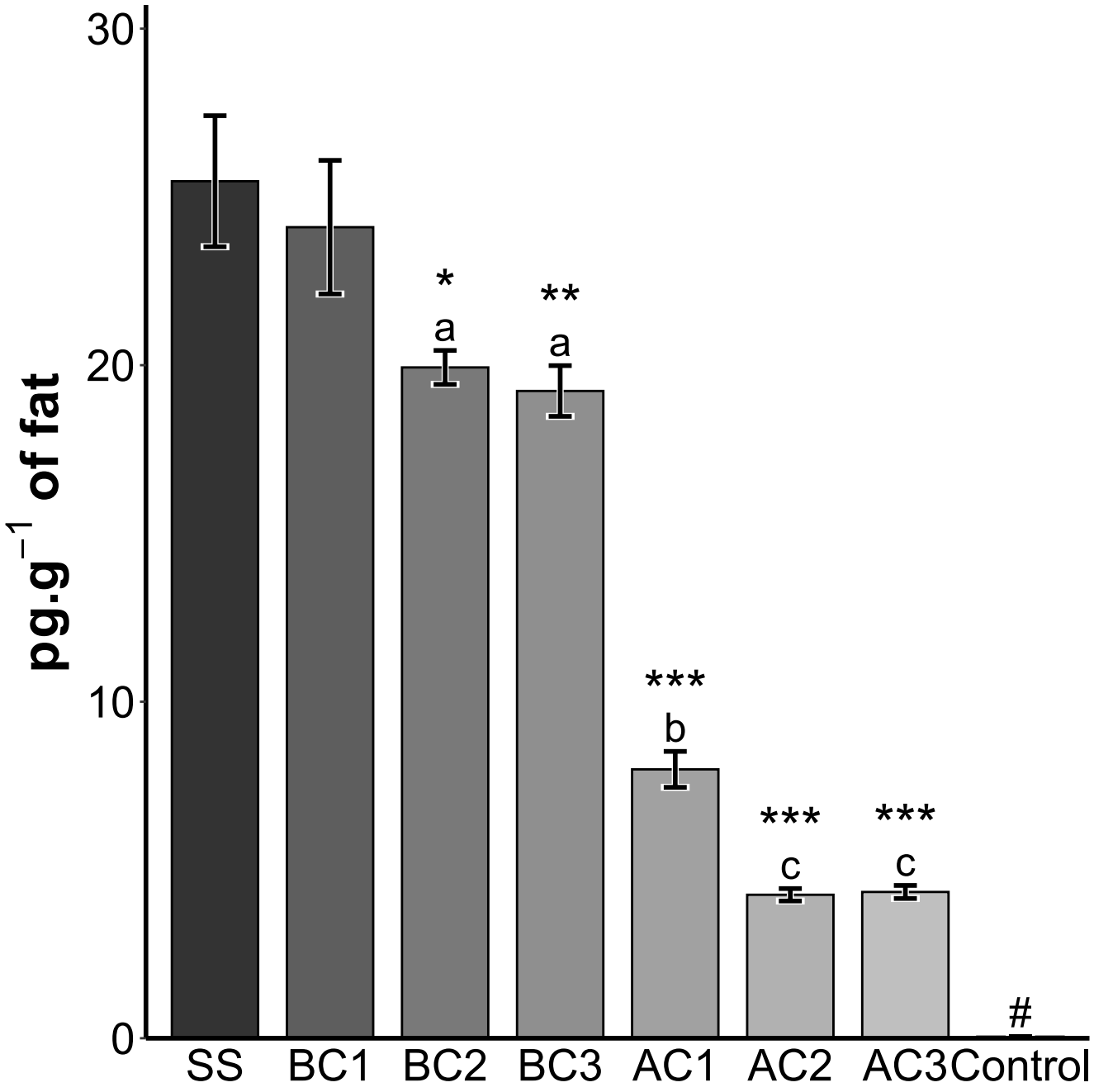

Figure S12: Concentrations of 1,2,3,4,7,8-HxCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,4,7,8-HxCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
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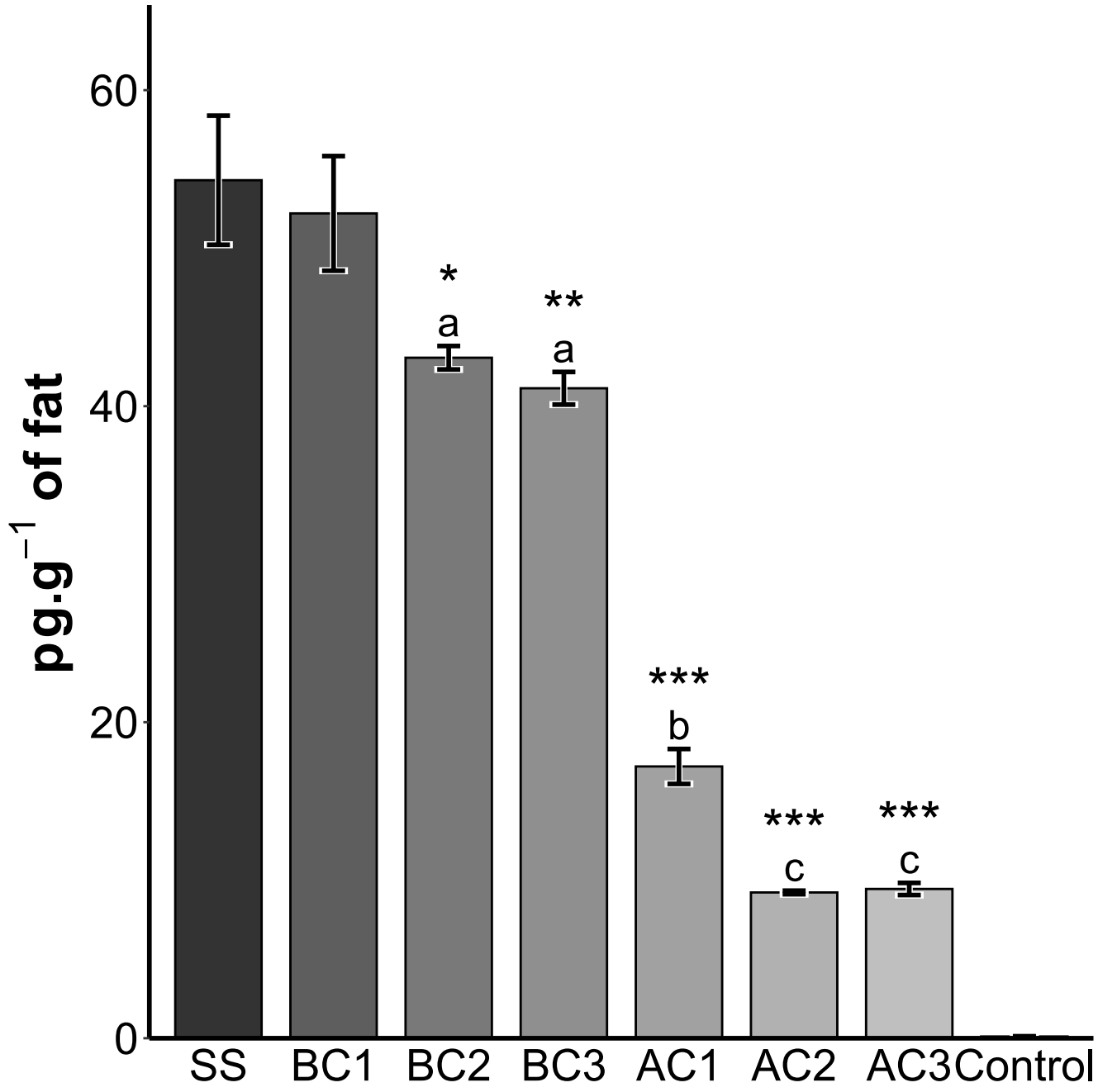

Figure S13: Concentrations of 1,2,3,6,7,8-HxCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,6,7,8-HxCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
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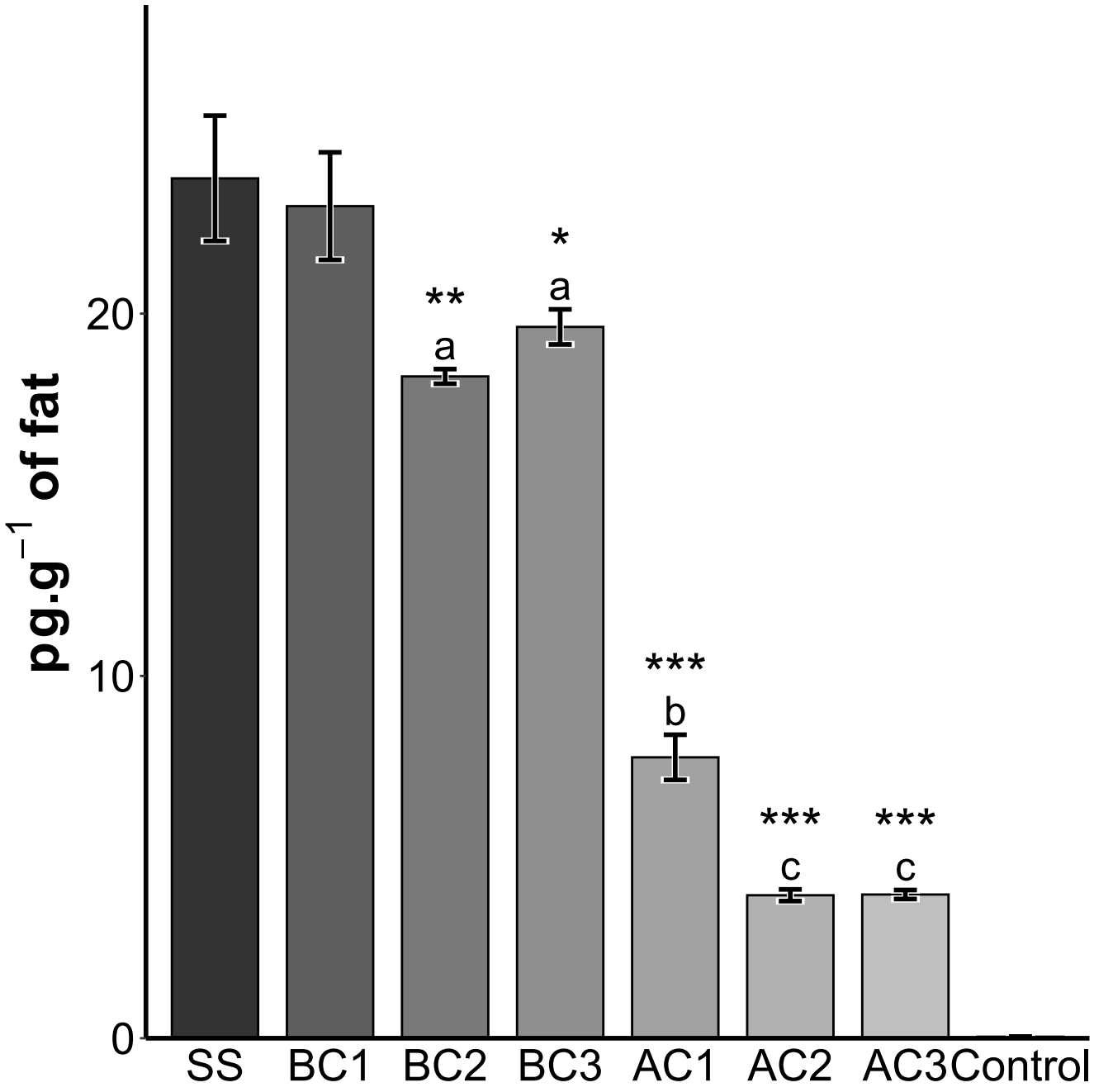

Figure S14: Concentrations of 1,2,3,6,7,8-HxCDF in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,6,7,8-HxCDF in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
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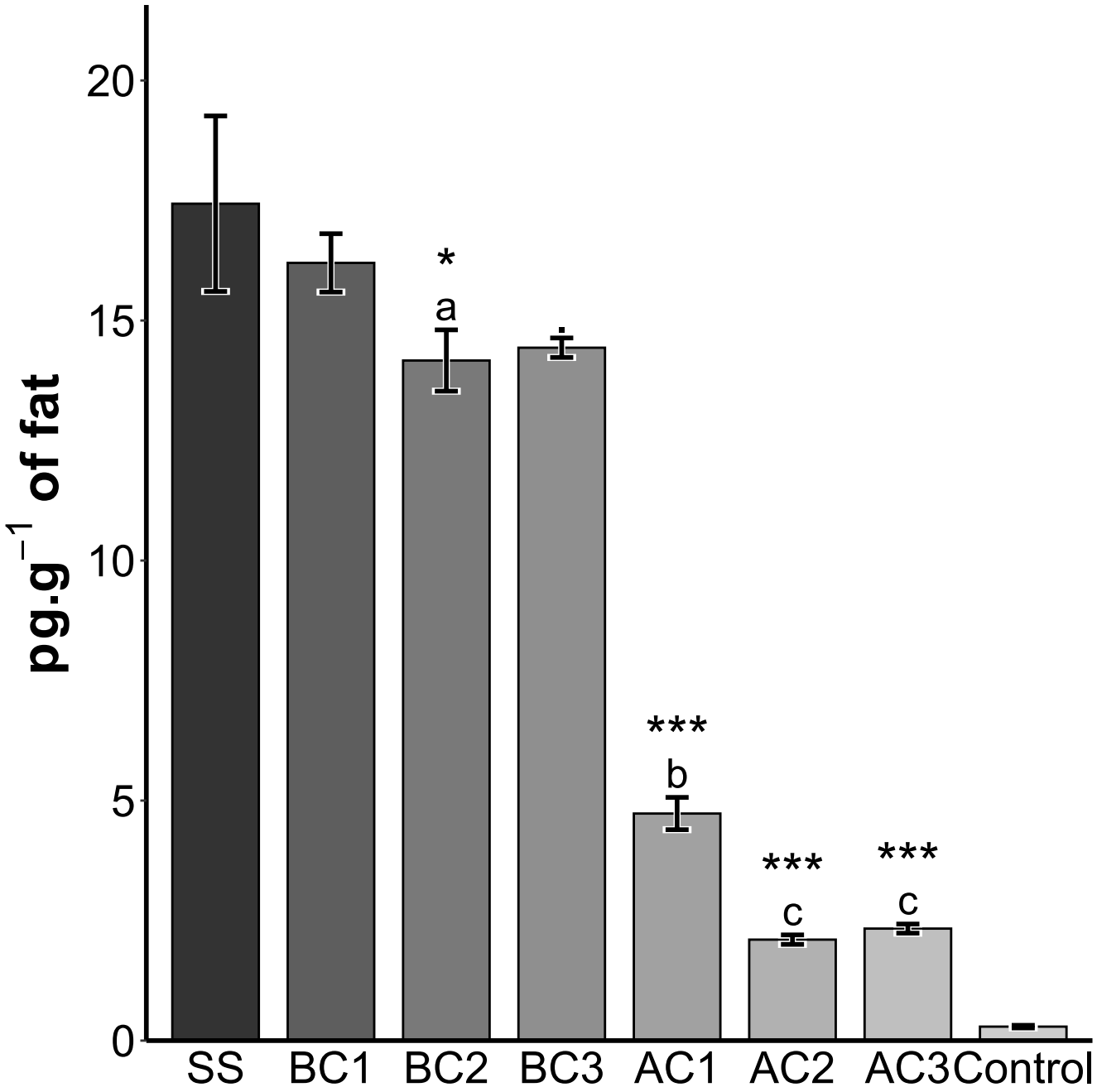

Figure S15: Concentrations of 1,2,3,4,6,7,8-HpCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,4,6,7,8-HpCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
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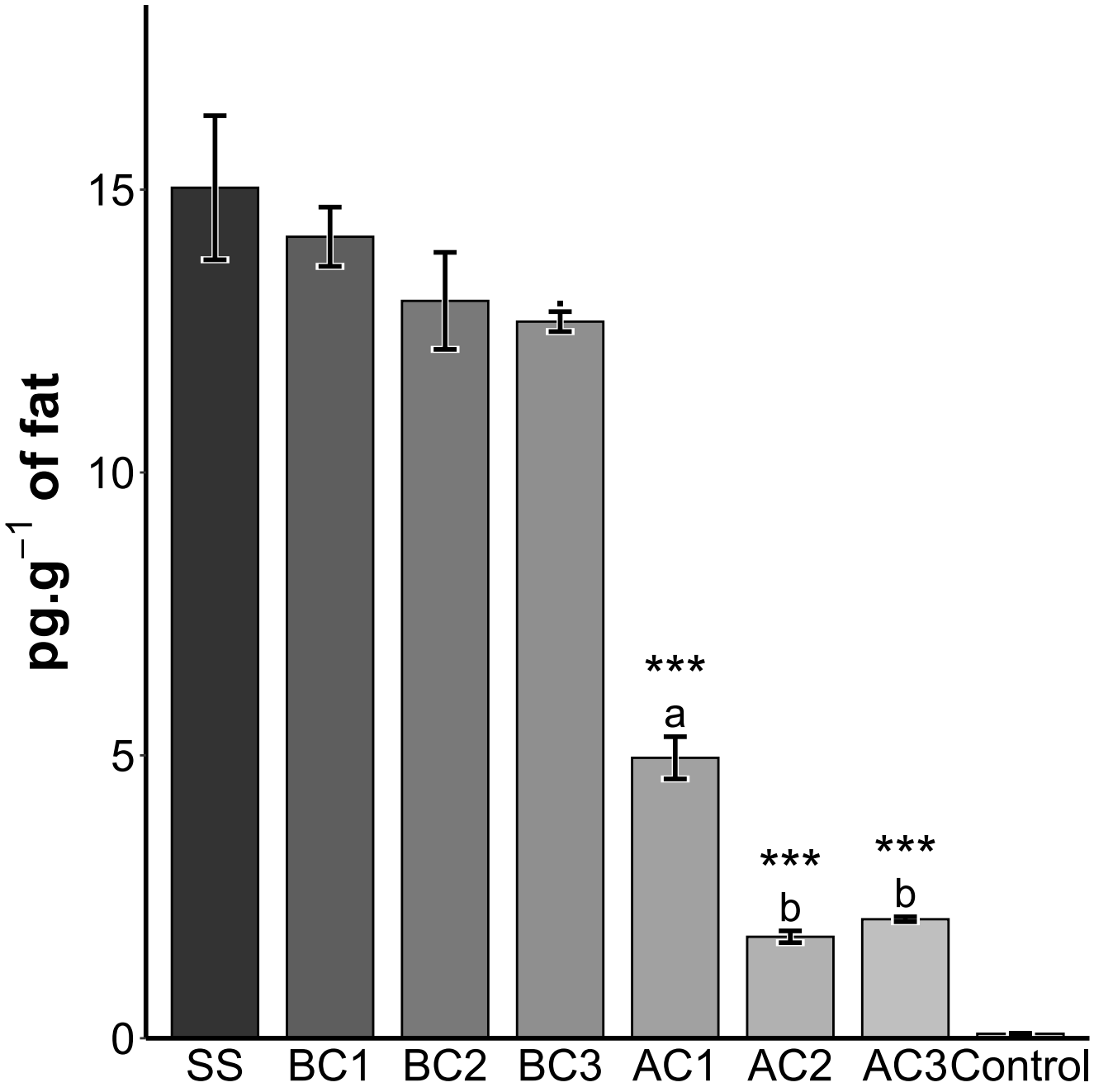

Figure S16: Concentrations of 1,2,3,4,6,7,8-HpCDF in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,4,6,7,8-HpCDF in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found significant groups using dunnetts.
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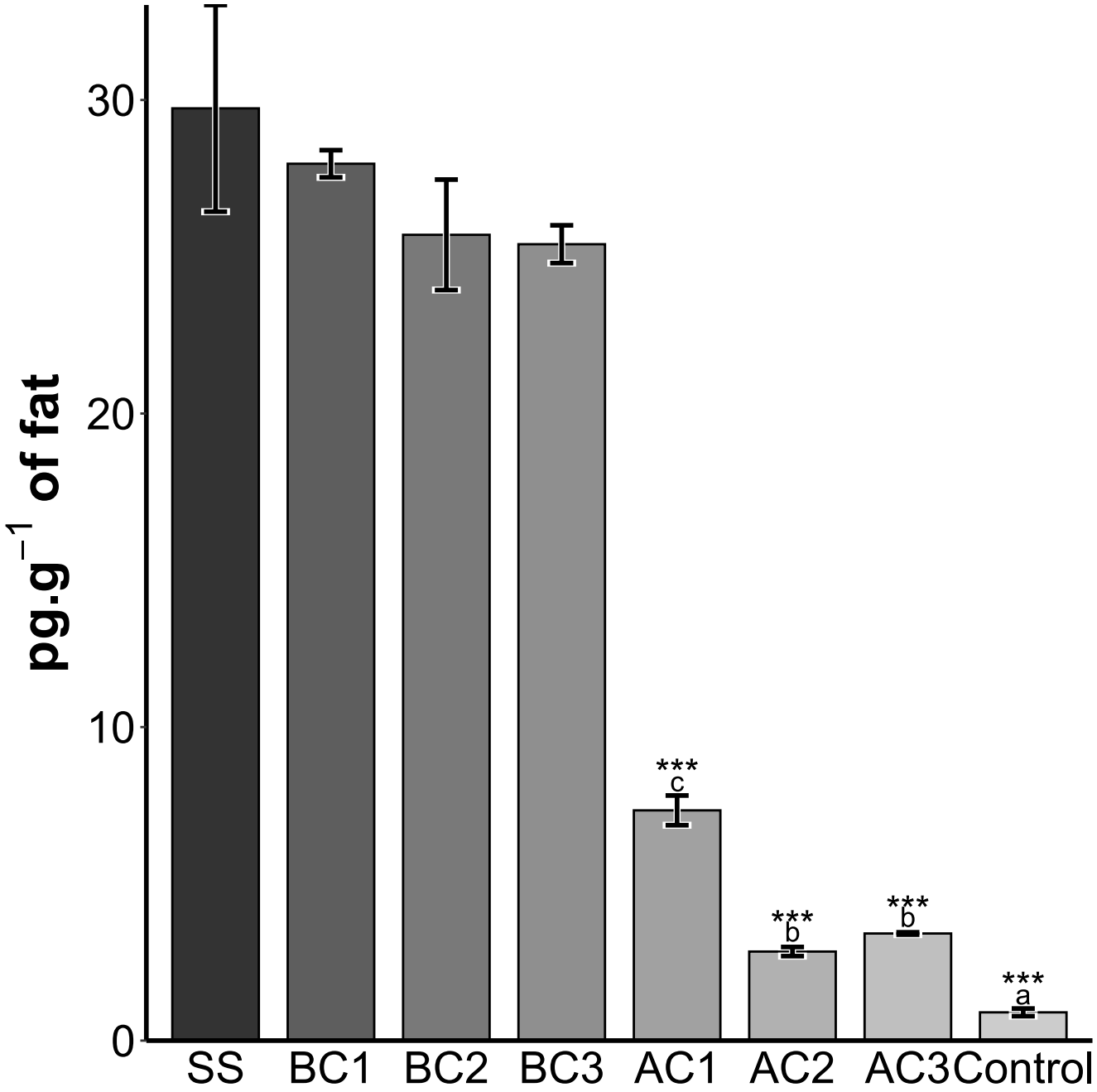

Figure S17: Concentrations of 1,2,3,4,6,7,8,9-OCDD in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,4,6,7,8,9-OCDD in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found signif icant groups using dunnetts.
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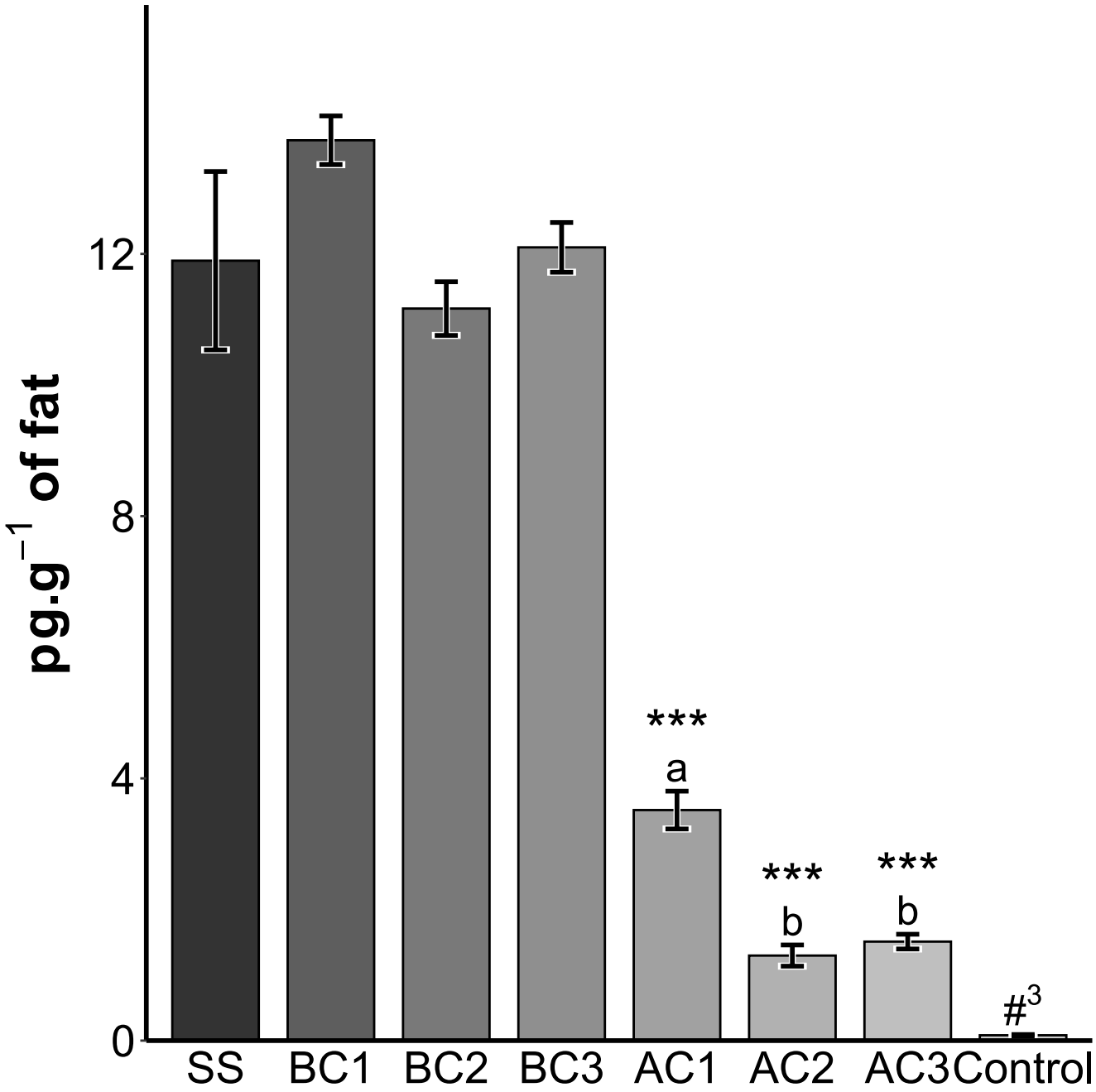
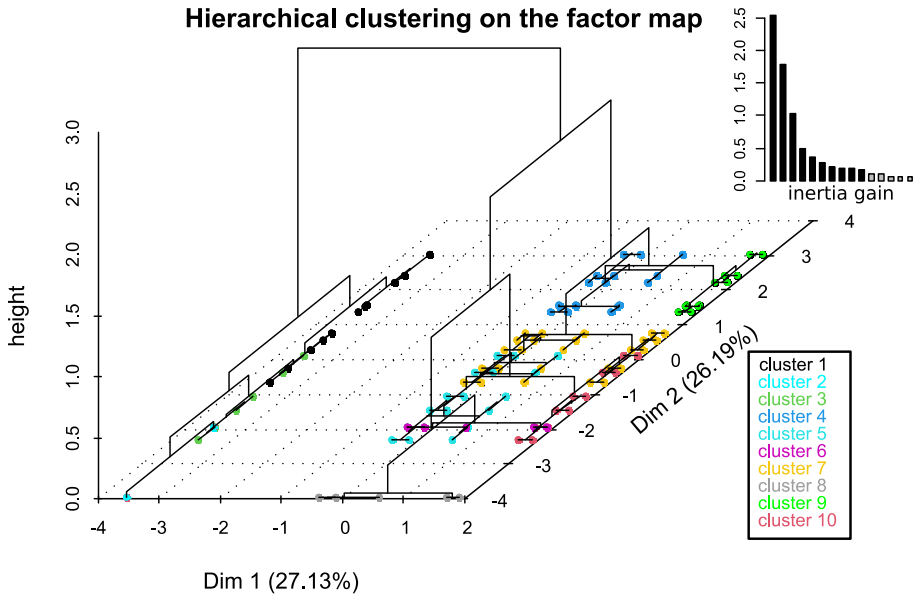


Figure S18: Concentrations of 1,2,3,4,6,7,8,9-OCDF in egg yolks.

Values correspond to the mean ± SE (n=3) concentrations of 1,2,3,4,6,7,8,9-OCDF in egg yolks. Group mean values with superscript asterisks are statistically different from non-amended soil (SS) (0.1: . > 0.05 : * >0.01 : ** > 0.001 : ***) using a variance analysis and a Dunnett post-hoc test. Controls were not included in the tested dataset. Groups mean concentrations with different superscript letter (a, b, c) are statistically different (P<0.05) from each other using a complementary variance analysis followed by a Tukey post-hoc test on previously found signif icant groups using dunnetts.
#: Values below LOQs were replaced by the corresponding LOQ, superscript numbers indicate the number of replaced values



cluster #	BR (%) :	TEF :	BCs / ACs :	Compounds :
1	93±24% (3)	0,25	BC1	L+H-DF
3	69±57% (3)	0,0001	BC1	PCB 28 ; PCB 77
2	94±8,1% (6)	-	BC1	CLD ; PCB-NDL
4	66±21% (9)	0,037	BC2, BC3, CA1	H-DF
5	94±4,2% (18)	-	BC2, BC3, CA1	CLD ; PCB-NDL
6	56±11% (15)	0,0001	BC2, BC3, CA1, CA2, CA3	PCB77
7	40±13% (15)	0,5	BC2, BC3, CA1, CA2, CA3	L-DF
8	47±11% (15)	-	BC2, BC3, CA1, CA2, CA3	PCB 28
9	14±3,0% (6)	0,037	CA2, CA3	H-DF
10	81±8,9% (12)	-	CA2, CA3	CLD ; PCB-NDL

Figure S19: First attempt of unsupervised hierarchical classification of characterization data and cluster composition
For each cluster are presented:
Relative bioavailability factor (%) (mean ± p90), in parenthesis the number of individuals associated with this value.
Average TEF: the average TEF of the cluster
BCs /ACs : matrices present in the cluster
Compounds: compounds present in the cluster:
L-DF (low weight dioxins: 2378TCDF, 2378TCDD, 23478PCDF, 12378PCDD, 123678HxCDF); H-DF (high weight dioxins: 123678HxCDD, 123478HxCDD, 1234678HpCDF, 1234678HpCDD, 12346789OCDF, 12346789OCDD); PCB-NDL (Non dioxin like PCB (without PCB 28): PCB 52, PCB 101, PCB 138, PCB 153, PCB 180); PCB 77; PCB 28
- : value not computable.