Chemosphere 270 (2021) 129698

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Average transfer factors are not enough: The influence of growing cattle physiology on the transfer rate of polychlorinated biphenyls from feed to adipose

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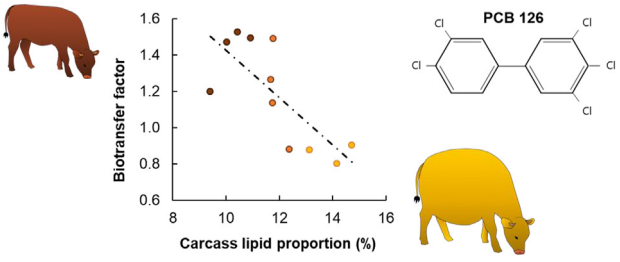
HIGHLIGHTS

- Growing cattle fed with diets at PCB background levels were studied.
- Higher chlorinated PCBs have higher transfer factors than lower chlorinated PCBs.
- Growth rate had no significant effect on PCB transfer factors.
- Higher carcass lipid proportion decreased PCB bioconcentration & -transfer factors.
- Transfer factor equations in function of carcass lipid proportion were established.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 December 2020
Received in revised form
15 January 2021
Accepted 17 January 2021
Available online 23 January 2021

Handling Editor: J. de Boer

Keywords:
Persistent organic pollutant
Bioconcentration factor
Biotransfer factor
Cattle growth rate

ABSTRACT

Food of animal origin accounts for >90% of the overall human exposure to polychlorinated biphenyls (PCBs). Food regulatory maximum levels help to control this exposure, but bovine meat has been found to be prone to exceed those occasionally. In order to ensure the chemical safety of bovine meat, the aim was to explore the dependency of the bioconcentration (BCF) and biotransfer (BTF) factor, and assimilation efficiency (AE) of PCBs on carcass lipid proportion and growth rate of beef cattle. Eleven bulls were fattened for 293 days with three different diets (7.0, 7.4, 7.5 MJ net energy for growth kg^{-1} dry matter) at PCB background levels, until slaughter at 530 or 600 kg body weight. Feed and perirenal adipose tissue were sampled for PCB analyses via GC/HRMS and carcass lipid proportion was estimated by the 11^{th} rib dissection technique. For all tested PCBs, BCF (ranging from 0.7 to 18.4) and BTF (ranging from 0.1 to 2.7) decreased at least 1.5 up to 10.6-fold when the carcass lipid proportion increased by 4%, resulting from a typical dilution process. For a faster growth rate of 0.18 kg d^{-1} however, only a non-significant increasing trend in transfer factors (1.1 to 2.1-fold) was seen. Besides, the transfer factors increased with PCB chlorination degree, non-ortho substitution and lipophilicity. These results underpin the complex

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https://doi.org/10.1016/j.chemosphere.2021.129698

Carcass lipid proportion
Chemical risk assessment

interaction between animal physiology and PCB physicochemical properties, making it challenging to interpret average transfer factors to support chemical risk assessment and management.

1. Introduction

Persistent organic pollutants (POPs) are carbon-based organic molecules that are highly toxic to humans and ecosystems. They easily disperse and exhibit very long half-lives (decades or even centuries) in the environment, and further bioaccumulate into the food chain due to their poor degradability and high lipophilicity (Guo et al., 2019; Weber et al., 2019). Among others, polychlorinated biphenyls (PCBs) were listed as POPs under the Stockholm Convention in 2001 (Ryan et al., 2013). Polychlorinated biphenyls are anthropogenic chemicals that had a widespread use in various applications due to their non-flammability, chemical stability and low heat conductivity (EFSA, 2018). However, PCBs have several adverse health effects, such as endocrine disruption, carcinogenicity or neurotoxicity (EFSA, 2018; Guo et al., 2019). Although PCBs were banned in the 1970s in the USA and Europe, around 80% of the total PCB amount ever produced is estimated to be still present and remains to be eliminated (UNEP, 2015), so that the concern for human health endures due to the risk of chronic exposure (EFSA, 2018; Guo et al., 2019).

In humans, consumption of food of animal origin (meat, dairy, eggs and fish) embodies the major exposure pathway to PCBs. In Western countries, this route accounts for more than 90% of the overall human exposure, of which ruminant products are the largest contributor (EFSA, 2018; Ryan et al., 2013; U.S.EPA, 1994). Due to this dietary exposure, between 1 and 53% of the European population exceeds the tolerable weekly intake (TWI) of 14 pg toxic equivalent (TEQ) kg^{-1} body weight (BW) (EFSA, 2012), an issue that may become worse since the European Food Safety Authority (EFSA) recommended in 2018 to reduce the TWI by a factor of seven (EFSA, 2018). In order to control and reduce human exposure, several national and international agencies have set up maximum regulatory levels in food items of animal origin (e.g. Regulation (EU) No. 1259/2011) (EFSA, 2018). Over the past decades, occasional exceedance of maximum regulatory levels in food products of animal origin mostly originated from contaminated feed incidents (e.g. the Belgian PCB and dioxin crisis in 1999) (Weber et al., 2018). Among ruminant products, bovine meat, which accounts for around 15% of the total human exposure to PCBs (BAG, 2013), has been found to be more sensitive and prone to exceed these regulatory levels in recent years, even without obvious identifiable contamination sources (Weber et al., 2018). These findings are based among others on two monitoring studies, where more than 50% of the bovine meat samples from extensive farming systems exceeded either the action (product sale permitted, contamination source should be identified and eliminated) or maximum level (product sale prohibited, confiscation and incineration) (BAG, 2012; BVL, 2013; EFSA, 2018). Also U.S. surveys from 2002 to 2019 demonstrate that over time bovine meat median dioxin/furan and PCB levels stagnate or even increase, whereas the median levels in pork, chicken and turkey meat are 2 to 7-fold lower than for beef and decrease continuously over time (Lupton et al, 2015, 2019). Therefore, in order to ensure the chemical safety of bovine meat and further the long-term sustainability of beef cattle farming systems, it is mandatory to mechanistically understand and quantify the transfer of POPs in beef cattle.

In livestock, the fate of POPs is primarily investigated via feeding experiments (Mclachlan, 1993a), from which transfer factors from feed to body tissues [adipose tissues (AT), muscles or offal], including the biotransfer factor (BTF), the bioconcentration factor (BCF) or the assimilation efficiency (AE), can be derived. In such kind of feeding experiments, the animal system is often seen as a 'black box', where the POP transfer rate parameters from input (oral exposure concentration) to output (body tissue/organ concentration or burden) is given as a single average value without taking into consideration the animal physiological status. When one aims to go deeper into a comprehensive and mechanistic approach, it is important to better understand and quantify how much the transfer rate from feed to the target tissues can be affected by the animal physiological status. Indeed, some studies with broilers, pigs or sheep highlight the dependency of POP toxicokinetics on growth rate and body lipid dynamics (Jondreville et al., 2017; Lerch et al., 2016, 2020b; Rey-Cadilhac et al., 2020). In the case of beef cattle, feeding studies regarding PCB transfer from feed to meat are rather scarce and focus only on few congeners (Vemmer et al., 1992, 1993). Besides, the extent and the mechanism by which physiological traits of growing cattle, especially growth rate and body lipid proportion, can affect such transfer parameters were not addressed at all so far.

The aims of this study were therefore i) to determine the transfer rates from feed to the blood and adipose tissue of growing cattle for a large set of PCB congeners with diets at background contamination levels and ii) to explore further the dependency of the transfer factors on the animal physiology (i.e. growth rate and carcass lipid proportion).

2. Materials and methods

2.1. Animals and diets

All procedures performed on animals were approved (approval number: 2013_48_FR) by the committee on animal experimentation of canton Fribourg (Switzerland). The experiment was conducted at the experimental farm of Agroscope (Posieux, Switzerland). Thirteen mostly crossbreed bulls, aged 43 ± 13 d and weighing 70 ± 6 kg BW, were selected from a larger experiment of 89 individuals purchased from dairy farms (detailed experimental design in Lerch et al. (2020a)). During the pre-experimental period, the bulls were fed with an average of 3.6 kg d^{-1} milk replacer (130 g dry matter (DM) kg^{-1} milk replacer) for seven weeks, completed with hay, maize silage and concentrate *ad libitum* until reaching 154 ± 5 kg BW at the age of 126 ± 17 d. Two bulls (age 153 ± 6 d, 154 ± 5 kg BW) were slaughtered at day 0 of the experimental period to determine the initial body composition and POP carcass burden. The remaining eleven bulls were allocated into three groups based on BW and growth rate during the pre-experimental period, and subsequently received three different total mixed feed rations for the duration of the experimental period. Rations were composed of maize and grass silages, as well as three different concentrates (76:24 forage/concentrate ratio) to reach three distinct energy levels (maize/grass silage ratio on DM basis of 70:30, 80:20 and 85:15, resulting in 7.0, 7.4 and 7.5 MJ of net energy for growth kg^{-1} DM). Rations were prepared and distributed once per day at 09:00 h. In order to record the individual daily feed

intake, the bulls had controlled access to electronic feed bunks on balances. Animals were kept in an indoor straw-bedded free-stall barn with access to an outdoor walking area and had free access to fresh water. Details regarding animal breed and performance are provided in Table S1, whereas details regarding chemical composition and nutritive value of the total mixed rations are provided in Table S2. When reaching a BW of either 530 ($n = 4$) or 600 kg ($n = 7$) at an age of 415 ± 36 d, the bulls were slaughtered at the commercial slaughterhouse "Marmy Viande en gros SA" (Estavayer-le-Lac, Switzerland). Slaughter took place according to the legally defined procedure, being stunning via cranial perforation followed by exsanguination.

2.2. Measurements, sampling and POP analyses

Body weight was recorded every 4 weeks at 08:00 h before feed distribution. The DM content of each ration offered was determined by desiccation (103 °C, 24 h) three times a week. The daily DM intake composited for each bull was based on the individual fresh matter intake and the mixed ration DM content data. A pool sample per mixed feed ration and for the three concentrates was composited from subsamples collected weekly along the experiment. These fresh samples were oven dried at 60 °C for 24 h before being grinded through a 1 mm grid. At slaughter, 30–60 mL full blood and 50–100 g perirenal AT were sampled and stored at −20 °C before POP analyses. After cooling the carcass for 24 h at 4 °C, the total carcass was weighed and the 11th rib of a half carcass was collected and dissected to determine its relative proportions of muscle, AT and bone.

A slightly modified previously reported analytical method for POPs was followed (Bogdal et al., 2017) with the detailed procedure and quality assurance in the Supporting Information (sections 1.1., 1.2. and 1.3.). Sample extraction was performed with Soxhlet for feed pool and perirenal AT and liquid-liquid extraction for blood. The extracts were spiked with $^{13}C_{12}$-labeled indicator PCB (iPCB) and dioxin-like PCB (dlPCB) internal standards. Purification was based on silica, alumina and carbon column chromatography clean-up. For the detection of the PCBs, gas chromatography/high resolution mass spectrometry (GC/HRMS) was used.

2.3. Calculations and statistical analyses

The following transfer parameters were calculated (Jondreville et al., 2017; Takaki et al., 2015):

$$BCF\ (unit\ less) = PCB\ concentration\ in\ AT\ (ng\ kg^{-1}\ lipid) / PCB\ concentration\ in\ feed\ (ng\ kg^{-1}\ DM) \quad (1)$$

$$BTF\ (d\ kg^{-1}) = PCB\ concentration\ in\ AT\ (ng\ kg^{-1}\ lipid) / daily\ PCB\ intake\ (ng\ d^{-1}) \quad (2)$$

$$AE\ (\%) = PCB\ carcass\ burden\ (ng) / total\ PCB\ intake\ (ng)\ x\ 100 \quad (3)$$

The daily PCB intake equals the PCB concentration in feed DM multiplied by the average DM intake over the experimental period. Total PCB intake was obtained by multiplying the daily PCB intake by the number of days of the experimental period. Polychlorinated biphenyl carcass burden was calculated by multiplying the perirenal AT PCB lipid-normalized concentration by the carcass lipid mass, assuming equal PCB lipid-normalized concentrations in both carcass and perirenal AT (Richter and McLachlan, 2001). The carcass lipid mass was estimated from the proportion of AT of the 11th rib and the cold carcass weight applying the predictive equation of Robelin et al. (1975):

$$Carcass\ lipid\ mass\ (kg) = (0.568 \times AT\ proportion\ of\ 11^{th}\ rib\ (\%) + 3.52) \times carcass\ weight\ (kg) / 100 \quad (4)$$

To test whether the animal physiology has an effect on the transfer parameters, the growing bulls were classified into low, medium or high carcass lipid proportion [LL > 0.5 × standard deviation (SD) below, ML ± 0.5 SD from and HL > 0.5 × SD above the carcass lipid proportion mean, respectively] and into slow, medium or fast growth rate (SG > 0.5 × SD below, MG ± 0.5 SD from and FG > 0.5 × SD above the growth rate mean, respectively).

Statistical analyses were performed with the Statistical Analysis Systems software (SAS, version 9.3., SAS Institute Inc., NC, USA). A mixed model in total randomization was used for testing the effect of feeding treatments on physiological parameters and PCB concentrations in perirenal AT and blood, with feeding treatment as a fixed and animal as a random effect. Further, a mixed model was used for testing the effect of carcass lipid proportion and growth rate on BTF, BCF and AE, including feeding treatment, carcass lipid proportion class (LL, ML, HL), growth rate class (SG, MG, FG) and their interaction as fixed effects and animal as a random effect. Feeding treatment and interactions were never significant ($P > 0.10$) and were removed from the final model. A logarithmic transformation was applied when needed, to comply with the assumptions of normality and homoscedasticity of residuals. When transformation was needed, least square means and standard errors were estimated from untransformed data, whereas P-values reflect statistical analyses of transformed data. Additionally, to assess the individual effect of carcass lipid proportion and growth rate on BTF, BCF and AE, linear regression analyses were performed using the GLM procedure. Significance was declared at $P \leq 0.05$. Trends towards significance were assumed at $0.05 < P \leq 0.10$.

3. Results

3.1. Feed intake, growth rate and carcass traits

Body weight, feed intake, growth rate, carcass weight and lipid proportion are presented in Table 1. The BW of the two bulls slaughtered at the initiation of the experimental period was representative of the initial BW of the eleven bulls studied (154 and 154 kg, respectively), and their carcass weight of 74 kg was estimated to contain 7.6% lipids.

Along the experimental period, the average daily DM intake ($P < 0.01$) and the growth rate ($P = 0.11$) were higher for the high energy compared to the low energy treatment, whereas no treatment effect was recorded for carcass weight and lipid proportion ($P > 0.43$). Differences in DM intake across treatments were mostly observed during the second half of the experimental period (Fig. S1).

3.2. Feed PCB concentrations

The PCB levels of the three mixed rations are presented in Table 2. The low, medium and high energy rations had total PCB concentrations of 2.5, 2.3 and 1.7 μg iPCBs kg^{-1} DM and 600, 500 and 340 ng dlPCBs kg^{-1} DM (0.08, 0.06 and 0.05 ng TEQ_{05} kg^{-1} DM, respectively). Accordingly, the bulls had a daily exposure of 16 μg iPCBs d^{-1} and 3.9 μg dlPCBs d^{-1} (0.52 ng TEQ_{05} d^{-1}) for low, 16 μg iPCBs d^{-1} and 3.6 μg dlPCBs d^{-1} (0.42 ng TEQ_{05} d^{-1}) for medium, and 13 μg iPCBs d^{-1} and 2.6 μg dlPCBs d^{-1} (0.35 ng TEQ_{05} d^{-1}) for high energy rations (Table S4). The pattern of the iPCBs in feed was dominated by the congeners 52 and 101 (34% and 31% of the total iPCBs, respectively). For dlPCB raw concentrations PCB 118 and 105 (69% and 20%, respectively) were the most abundant, whereas for the TEQ_{05} transformed concentration PCB 126 dominated, followed

Table 1
Growing bulls' age, body weight, intake, growth rate, carcass weight and lipid proportion depending on feeding treatment.

		Pre-experiment[1]	Ration			
			Low[2]	Medium[2]	High[2]	*P-value*
Age (d)	Initial (day 0)	153 ± 6	122 ± 7	126 ± 9	117 ± 7	0.74
	Slaughter		427 ± 14	411 ± 20	405 ± 24	0.70
Body weight (kg)	Initial (day 0)	154 ± 4	149[b] ± 1	156[a,b] ± 5	158[a] ± 1	0.05
	Slaughter		569 ± 21	574 ± 23	588 ± 17	0.78
Intake (kg DM d^{-1})			6.5[c] ± 0.1	7.1[a,b] ± 0.1	7.7[a] ± 0.3	<0.01
Growth rate (kg d^{-1})			1.38 ± 0.01	1.46 ± 0.02	1.51 ± 0.06	0.11
Carcass weight (kg)		74 ± 6	322 ± 13	322 ± 18	330 ± 7	0.88
Carcass lipid prop. (%)		7.6 ± 1.2	11.0 ± 1.1	12.7 ± 1.0	12.0 ± 0.5	0.43

[a,b,c] Values in the same row (excluding pre-experiment) with different superscripts differ ($P < 0.05$).
[1] Pre-experiment data correspond to the two initially slaughtered bulls at the initiation of the experiment, for which initial (day 0) and slaughter day are the same.
[2] Observations per category: low energy level has $n = 4$, medium $n = 3$ and high $n = 4$.
Data represented are the least square means ± standard error and *P*-values of the non-transformed data mixed model.
Abbreviations: DM: dry matter, carcass lipid prop.: carcass lipid proportion

Table 2
Concentrations of polychlorinated biphenyls (PCBs) in feed, perirenal adipose tissue and blood at slaughter depending on feeding treatment.

	Feed ration[2]			Perirenal adipose tissue[2]					Blood[2]				
	Low[1]	Med.[1]	High[1]	Pre-experiment[1]	Low[1]	Medium[1]	High[1]	*P-value*	Pre-experiment[1]	Low[1]	Medium[1]	High[1]	*P-value*
iPCB	µg kg^{-1} DM			ng g^{-1} lipid					ng g^{-1} lipid				
28	0.23	0.22	0.17	1.3 ± 0.20	0.38 ± 0.17	0.37 ± 0.29	0.34 ± 0.23	0.90	5.5 ± 0.79	13 ± 4.0	12 ± 3.9	6.9 ± 2.6	0.51
52	0.85	0.79	0.56	0.81 ± 0.09	0.30 ± 0.09	0.45 ± 0.18	0.30 ± 0.15	0.72	3.1 ± 1.1	3.8 ± 0.69	5.3 ± 0.62	2.6 ± 0.40	0.08
101	0.78	0.72	0.52	1.9 ± 0.27	0.61 ± 0.26	0.73 ± 0.52	0.64 ± 0.45	0.93	3.3 ± 1.9	3.9 ± 0.82	3.9 ± 1.4	2.2 ± 0.58	0.24
138	0.31	0.23	0.20	3.4 ± 0.53	1.9 ± 0.31	2.2 ± 0.33	1.7 ± 0.57	0.55	3.5 ± 0.95	4.5 ± 0.96	2.9 ± 0.70	2.4 ± 0.89	0.07
153	0.28	0.23	0.19	3.5 ± 0.67	1.9 ± 0.24	2.1 ± 0.30	1.7 ± 0.51	0.55	4.4 ± 1.2	4.7 ± 0.63	3.5 ± 0.82	2.5 ± 0.75	0.13
180	0.06	0.05	0.04	1.3 ± 0.25	0.67 ± 0.15	0.78 ± 0.27	0.68 ± 0.30	0.86	1.6 ± 0.40	1.8 ± 0.48	1.1 ± 0.21	1.2 ± 0.56	0.51
Sum	2.5	2.3	1.7	12 ± 2.0	5.8 ± 1.2	6.7 ± 1.9	5.4 ± 2.2	0.70	21 ± 6.3	32[a] ± 7.6	28[a,b] ± 7.7	18[b] ± 5.8	0.05
dlPCB	ng kg^{-1} DM			pg g^{-1} lipid					pg g^{-1} lipid				
77	11	9.7	6.5	110 ± 13	35 ± 20	39 ± 33	37 ± 30	0.95	450 ± 150	500 ± 190	440 ± 240	290 ± 63	0.60
81	0.54	0.49	0.32	5.4 ± 0.65	1.9 ± 1.0	1.9 ± 33	1.6 ± 31	0.91	50 ± 33	32 ± 5.8	21 ± 250	16 ± 81	0.14
105	120	98	66	560 ± 39	250 ± 49	200 ± 46	190 ± 65	0.61	940 ± 380	3200 ± 2400	590 ± 29	460 ± 120	0.24
114	9.1	7.5	4.6	40 ± 0.65	25 ± 5.2	25 ± 3.2	17 ± 4.0	0.35	78 ± 17	120[a] ± 37	66[a,b] ± 12	37[b] ± 8.8	0.03
118	410	350	230	2400 ± 240	1400 ± 250	1200 ± 120	870 ± 170	0.25	4300 ± 1000	4600[a] ± 660	3100[a,b] ± 10	2100[b] ± 460	0.03
123	4.9	4.7	2.5	29 ± 0.50	18 ± 2.9	14 ± 1.8	12 ± 2.6	0.30	64 ± 12	84[a] ± 19	40[a,b] ± 15	26[b] ± 7.9	0.03
126	0.59	0.42	0.34	5.7 ± 0.30	4.8[a] ± 0.57	3.7[a,b] ± 0.47	2.9[b] ± 0.27	0.04	23 ± 1.9	18 ± 3.6	11 ± 2.0	12 ± 3.3	0.41
156	24	19	15	270 ± 20	160 ± 40	150 ± 17	120 ± 37	0.52	390 ± 87	750 ± 300	230 ± 32	263 ± 130	0.24
157	4.7	3.3	2.5	51 ± 2.7	30 ± 6.4	30 ± 2.1	22 ± 6.2	0.45	110 ± 34	180 ± 88	52 ± 11	54 ± 22	0.24
167	12	9.2	6.7	120 ± 7.8	72 ± 16	66 ± 7.9	53 ± 16	0.54	270 ± 55	290 ± 69	180 ± 16	160 ± 51	0.18
169	0.05	0.03	0.03	0.65 ± 0.05	0.50 ± 0.07	0.43 ± 0.09	0.35 ± 0.05	0.33	n.d.	n.d.	n.d.	n.d.	
189	1.4	1.1	0.78	25 ± 0.0	14 ± 4.7	14 ± 5.3	12 ± 5.2	0.93	150 ± 26	190 ± 30	110 ± 4.0	92 ± 20	0.10
Sum	600	500	340	3600 ± 300	2000 ± 400	1700 ± 270	1300 ± 360	0.37	6800 ± 1800	9960 ± 3800	4900 ± 620	3500 ± 970	0.06
TEQ$_{05}$ 3[3]									0.14 ± 0.03	0.15[a] ± 0.02	0.10[a,b] ± 0.001	0.07[b] ± 0.02	0.03
TEQ$_{05}$ min	0.08	0.06	0.05	0.71 ± 0.02	0.56 ± 0.07	0.43 ± 0.05	0.34 ± 0.04	0.06	2.5 ± 0.17	2.2 ± 0.44	1.3 ± 0.18	1.1 ± 0.40	0.27
TEQ$_{05}$ max	0.08	0.06	0.05	0.71 ± 0.02	0.56 ± 0.07	0.43 ± 0.05	0.34 ± 0.04	0.06	2.8 ± 0.11	2.4 ± 0.40	1.5 ± 0.16	1.7 ± 0.30	0.33

[a,b] Values in the same row (excluding day 0) with different superscripts differ ($P < 0.05$).
[1] Pre-experiment data correspond to the two initially slaughtered bulls at the initiation of the experiment, low energy level has $n = 4$ observations, medium $n = 3$ and high $n = 4$.
[2] The gray numbers are concentrations close to the blank (< 2.5-fold higher in concentration; blank concentrations are given in **Table S3**).
[3] TEQ$_{05}$ 3: represents the TEQ$_{05}$ sum of the 3 congeners (PCB 118, 157, 167) in blood, which exceeded the analytical blank by more than 2.5-fold for more than 75% of the 11 samples.
Data represented are the least square means ± standard error of the non-transformed data and *P*-values of the log-transformed data overall mixed model.
Abbreviations: iPCB: indicator polychlorinated biphenyl, dlPCB: dioxin-like polychlorinated biphenyl, TEQ: toxic equivalent, DM: dry matter, n.d.: non-detected congener

by 118 and 105 (73%, 16% and 5%, respectively, Table 2).

3.3. Adipose tissue and blood PCB concentrations and carcass PCB burdens

Table 2 presents the PCB levels in perirenal AT and blood at slaughter. The PCB concentration in AT at day 0 was 2.1-fold higher compared to the average PCB concentration in the AT at the end of the experiment. Further, there was a 1.5-fold decreasing trend in dlPCB AT concentrations from low to high energy fed bulls, being significant for PCB 126 ($P = 0.04$). The iPCBs, contributing the most to the concentration in AT, were PCBs 138 and 153 (33% and 32%, respectively). For dlPCBs, PCB 118, 105 and 156 dominated the sum (68%, 13% and 9%, respectively), whereas for TEQ$_{05}$ transformed concentrations PCB 126 and 118 dominated (85% and 8%, respectively).

For blood, a more pronounced decreasing trend from low to high energy ration as for AT was seen, being significant for PCB 118 ($P = 0.03$). Partition coefficients between AT and blood (PCB concentration in AT divided by the concentration in blood, both on lipid weight basis) were 0.36, 0.38 and 0.32 for PCB 118, 157 and 167, respectively. The focus relied on these three congeners, as they are the only ones where more than 75% of the blood measurements were 2.5-fold above the analytical blank.

The PCB carcass burden was 68 μg iPCBs and 20 μg dlPCBs (3.9 ng TEQ_{05}) at the initiation of the experiment (day 0, two initially slaughtered bulls). The estimated burden of the bulls at the end of the experimental period was 2.5 to 4-fold higher, resulting in 200 μg iPCBs and 67 μg dlPCBs (19 ng TEQ_{05}) for the low, 270 μg iPCBs and 69 μg dlPCBs (18 ng TEQ_{05}) for the medium and 200 μg iPCBs and 51 μg dlPCBs (13 ng TEQ_{05}) for the high energy rations (Table S4).

3.4. Bioconcentration and biotransfer factors, and assimilation efficiencies of PCB

3.4.1. Effect of PCB physicochemical properties

The BCFs ranged from 0.4 to 16.5 (Table S5), the BTFs from 0.1 to 2.3 (Table S6) and the AE from 1 to 31% (Table S7), with none diet effect ($P > 0.14$). The overall trend was that BCF, BTF and AE increase with increasing chlorination degree (Fig. S2). The only congeners that showed higher transfer factors as their equal chlorination degree counterparts were PCBs 77, 81, 126 and 169. This was in accordance with the fact that transfer factors increased with increasing lipophilicity, i.e. with the logarithm of the octanol-water partition coefficient (log K_{ow}, Fig. 1).

3.4.2. Effect of growth rate

The growth rates of the bulls were divided into three classes: SG 1.36 kg d^{-1}, MG 1.44 kg d^{-1} and FG 1.54 kg d^{-1}. Fig. 2A and Table S6 represent the BTFs, Fig. S3 and Table S5 the BCFs and Fig. 2C and Table S7 the AEs for each PCB congener based on growth rate classes. No significant effect of growth rate class was noticed for neither BCF nor BTF ($P > 0.16$, Fig. S3, Fig. 2A, Tables S5 and S6). Although for the hexa- and hepta-chlorinated PCBs, a FG rate appeared to be coupled to a somewhat higher BCF and BTF compared to the SG rate. For the AEs, the connection between a faster growth and higher AE seemed to be more pronounced and was already visible for the tetra-chlorinated PCBs 77 and 81 as well as for the penta-chlorinated congeners, especially for PCB 126 ($P < 0.10$, Fig. 2C and Table S7). For the hexa-chlorinated PCB 169 a significant difference between SG and FG ($P < 0.01$) and between SG and MG class ($P = 0.05$) was seen. This last result has to be interpreted with caution, as the concentration of PCB 169 in perirenal AT was around the analytical blank.

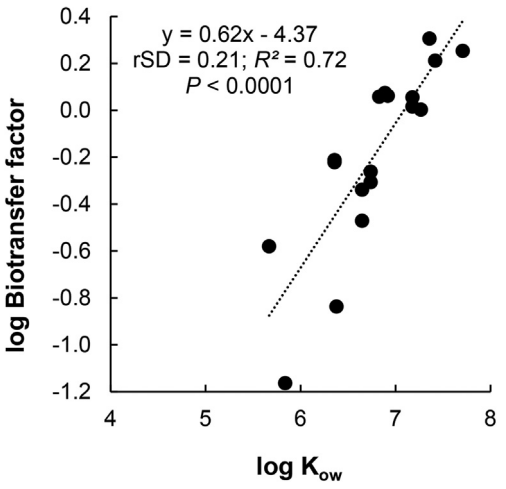

Fig. 1. Relationship between the biotransfer factor for polychlorinated biphenyls (PCBs) in growing bulls ($n = 11$) and the corresponding PCB octanol-water partition coefficient (K_{ow}) (IARC, 2016).

3.4.3. Effect of carcass lipid proportion

The carcass lipid proportions of the bulls were divided into three classes: LL 10%, ML 12% and HL 14%. Fig. 2B and Table S6 represent the BTF, Fig. S3 and Table S5 the BCF and Fig. 2D and Table S7 the AE for each tested PCB congener based on the carcass lipid proportion classes. Conversely to the growth rate, BTF and BCF decreased when the carcass lipid proportion increased. This effect was significant for BTF in the case of PCB 105, 114, 118, 123, 126 and 156 ($P < 0.05$), whereas a trend was observed for PCBs 157 and 167 ($P < 0.10$, Fig. 2B and Table S6). For BCF, the carcass lipid proportion class effect was significant for PCBs 105, 123 and 126 ($P < 0.05$), whereas for PCBs 118 and 156 a trend was observed ($P < 0.10$, Fig. S3 and Table S5). For the AE, no significant effect of carcass lipid proportion was detected ($P > 0.14$); even if numerically, the AE is also lower for the high carcass lipid proportion class, compared to the low one (Fig. 2D and Table S7).

3.5. Predictive transfer factor equations depending on physiological status

By performing linear regressions, a decreasing effect on BCFs and BTFs of an increasing carcass lipid proportion was confirmed on an individual basis, as demonstrated for the BTF of PCB 126 in Fig. 3. The BTF for this congener decreased 1.65-fold when the carcass lipid proportion increased by 4% (Table 3).

Similar linear equations were set-up for other PCB congeners, as listed in Table 3. For AE only one regression was significant ($P = 0.04$), namely the effect of growth rate on the AE of PCB 169:

$$AE \text{ for PCB 169 (\%)} = 47.28 \times \text{growth rate} - 47.57 \pm 5.60 \text{ (rSD)}; R^2 = 0.40 \quad (5)$$

4. Discussion

The studies focusing so far on PCB transfer from feed to bovine meat are rather scarce and did not assess the influencing effect of physiological traits of growing cattle on transfer parameters. Novel aspects of the present feeding experiment therefore include the determination of PCB transfer rates from feed to growing cattle for the 18 regulated iPCB and dlPCB congeners and an assessment of the effects of growth rate and carcass lipid proportion on PCB transfer factors. The effects of the dietary treatment on animal performances of the eleven bulls were comparable to the ones recorded on the whole set of 89 individuals, which were previously discussed in detail elsewhere (Lerch et al., 2020a).

4.1. Levels of PCBs in feed and growing bulls

Since the transfer factors are based on PCB concentrations, the PCB levels in the three total mixed rations, adipose tissue and blood were quantified. The PCB concentrations between the three rations differed presumably due to the grass silage content. The more grass silage was included within the total mixed ration, the higher the PCB concentration was, which may come from a higher soil inclusion in grass silage, as soil is a major environmental sink for PCBs

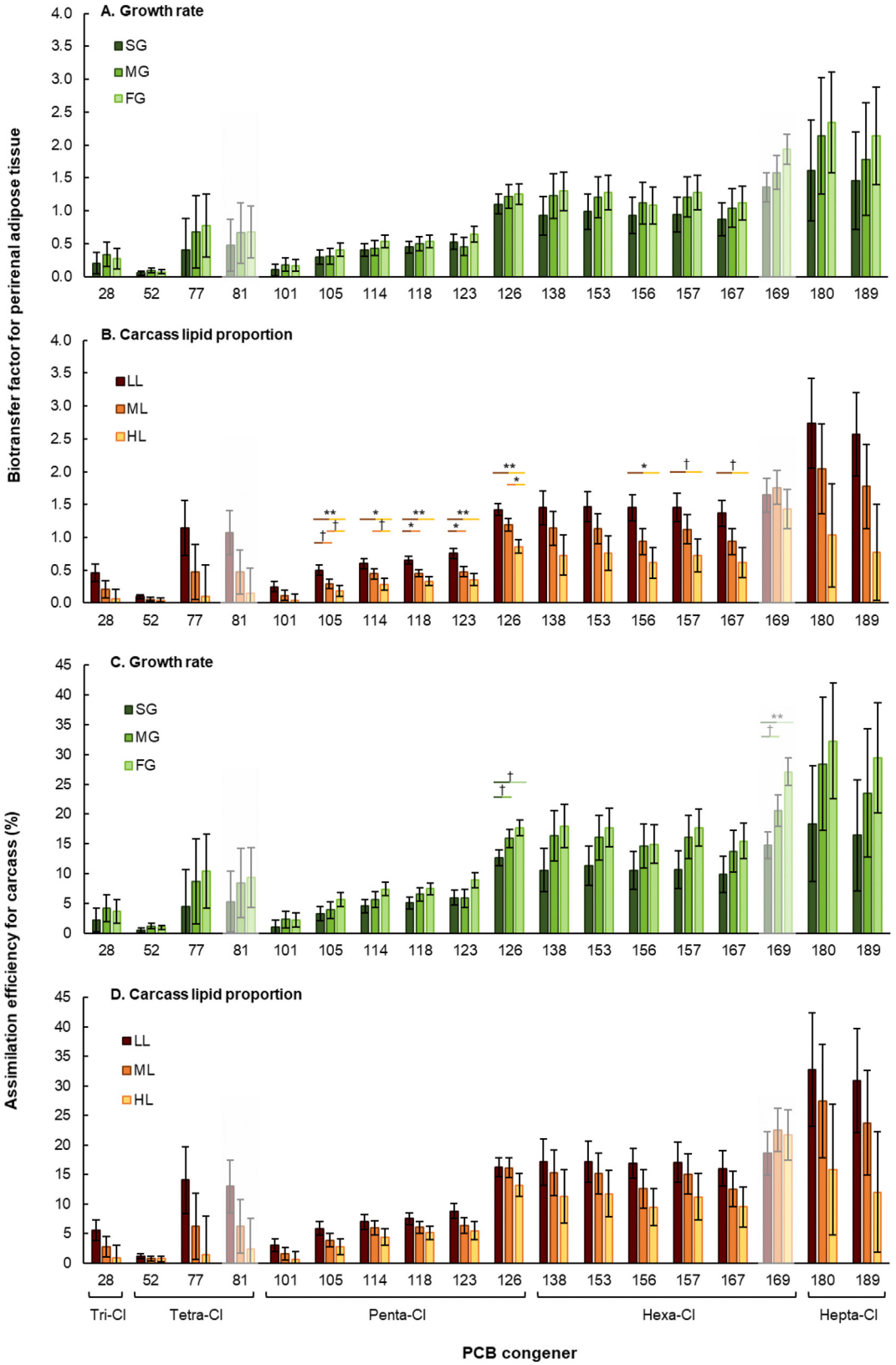

Fig. 2. Biotransfer factor in perirenal adipose tissue lipids and assimilation efficiency in carcass for polychlorinated biphenyls (PCBs) depending on growth rate (A and C, respectively): slow (SG), medium (MG) and fast (FG) growth class, or carcass lipid proportion (B and D, respectively): low (LL), medium (ML) and high (HL) lipid class. The shaded PCBs 81 and 169 concentrations are close to the analytical blank (< 2.5-fold higher in concentration). Least square means ± standard error are displayed. Trend and significant differences are indicated as follows: †$P < 0.10$, *$P < 0.05$, **$P < 0.01$.

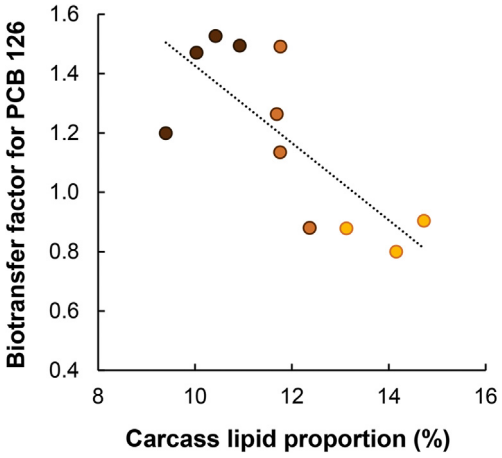

Fig. 3. Relationship between the biotransfer factor for polychlorinated biphenyl (PCB) 126 and the carcass lipid proportion. The color-coding corresponds to the different carcass lipid proportion classes: ● low, ● medium and ● high lipid proportion. Linear regression parameters are reported in Table 3. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

(Rychen et al., 2014). Nevertheless, all PCB levels in feed remained 4 to 8-fold below the EU maximum regulatory level for iPCBs [11 μg kg^{-1} DM (Regulation (EU) No. 277/2012)] or the action level for dlPCBs [0.39 ng TEQ_{05} kg^{-1} DM (Regulation (EU) No. 277/2012)]. This environmental background exposure could also be confirmed based on the feed iPCB congener profile, since the pattern within this study did not fit any industrial mixture, paint or joint sealant pattern (Weber et al., 2015). Further, the mean dlPCB background exposure of 430 pg TEQ_{05} d^{-1} was in accordance with the dioxin/furan and dlPCB chronic dietary exposure of beef cattle fed a maize silage-based diet reported by EFSA with a mean upper bound of 420 pg TEQ_{05} d^{-1} (EFSA, 2018). This chronic dietary exposure resulted in a 3-fold increase in carcass PCB burden from day 0 until the end of the experiment, as the PCBs bioaccumulate over the lifespan of the bulls.

The differences in PCB concentration between the three rations resulted subsequently in differences in daily PCB intake by the bulls. The bulls fed the high energy ration (i.e. the poorest in grass silage) had a 1.5-fold lower daily PCB intake compared to the low energy ration (i.e. the richest in grass silage), which was remarkably close to the 1.5-fold decreasing trend in average adipose tissue dlPCB concentration from low to high energy ration fed bulls. However, also the PCB levels in adipose tissue remained 3 to 7-fold below either the EU maximum regulatory level for iPCBs [40 ng g^{-1} fat (Regulation (EU) No. 1259/2011)] or the action level for dlPCBs [1.75 pg TEQ_{05} g^{-1} fat (Recommendation, 2013/711/EU)]. The iPCBs, which contributed the most to the adipose tissue concentration, were the higher chlorinated, more persistent PCBs 138 and 153 (33% and 32%, respectively). The lower chlorinated PCB 28, 52 and 101, on the other hand, accumulated in lesser extent, which can be explained by their higher metabolic clearance rate (Thomas et al., 1999; Vemmer et al., 1993).

In contrast to the adipose tissue, where all regulated PCB congeners were detected, only PCBs 118, 157 and 167 were detected within blood due to the 8-fold higher limit of detection in blood than in adipose tissue, resulting mainly from the very low blood lipid content. The detected PCB congeners in blood were 2 to 6-fold higher in concentration compared to the adipose tissue on lipid basis, presumably because PCBs not only diffuse into blood lipids, but may also bind to plasma proteins (Patterson et al., 1989).

4.2. Bioconcentration and biotransfer factors and assimilation efficiencies of PCBs

Transfer factors represent a way to describe the bioaccumulation potential of contaminants into specific tissues (Amutova et al., 2020). The BCFs and BTFs from feed to adipose tissue, reported in the present study, are in broad accordance with previous reports in growing beef cattle (Vemmer et al., 1992, 1993), but also in dairy cows, even if in dairy cows the BCF for PCBs 105, 114, 118 and 167 were 3 to 5-fold higher (Thomas et al., 1999). These differences between the calculated BCFs here and the ones determined in lactating cows may result from varying lipid dynamics between growing and lactating bovines: body lipids accretion for growing cattle *vs.* dynamic body lipid mobilization and subsequent accretion combined with lipid secretion through milk for lactating cattle.

4.2.1. Effect of PCB physicochemical properties

As suggested by Travis and Arms (1988) and Amutova et al. (2020), the physicochemical properties of contaminants widely affect transfer factors. In the present study, BCF, BTF and AE increased with increasing chlorination degree of the PCB congener, which is generally linked to an increasing lipophilicity (i.e. log K_{ow}) (IARC, 2016). The more chlorines are bound, the more lipophilic the

Table 3
Polychlorinated biphenyl (PCB) bioconcentration and biotransfer factor predictive equations depending on carcass lipid proportion of growing bulls.[a]

	Carcass lipid proportion (X)											
	Bioconcentration factor (Y)[2]						Biotransfer factor (Y)[2]					
PCB	Slope (a)			Intercept (b)			rSD	R^2	Slope (a)			Intercept (b)			rSD	R^2
105									-6.65	±	3.11	1.13	±	0.37	0.16	0.34
114									-6.01	±	3.25	1.17	±	0.39	0.17	0.28
118	-35.55	±	18.41	7.68	±	2.20	0.97	0.29	-6.47	±	2.62	1.26	±	0.31	0.14	0.40
123	-47.64	±	25.90	9.5	±	3.10	1.36	0.27	-8.31	±	3.55	1.53	±	0.42	0.19	0.38
126	-68.46	±	24.36	16.46	±	2.91	1.28	0.47	-13.01	±	3.70	2.73	±	0.44	0.19	0.58

[1] Only linear regression models with $P < 0.10$ for the effect of carcass lipid proportion on the slope are reported.
[2] Linear regression equations where *Y (BCF or BTF) = a × X (carcass lipid proportion) + b ± residual standard deviation (rSD).*
Slope and intercept are given as least square means ± standard error.
All intercepts are significantly different from 0 ($P > 0.05$).

compound, the more persistent and bioaccumulative it is. Several authors, however, question this relation, when the log K_{ow} exceeds 6.5, as the absorption rate in the ruminant digestive tract sharply decreases for such highly hydrophobic molecules due to their inability to easily cross the water layer surrounding the intestinal wall by passive diffusion (Fries et al., 1999; Kelly et al., 2004; Mclachlan, 1993b). Indeed, Vemmer et al. (1992, 1993), who studied iPCBs in growing cattle, demonstrated that the BCF and BTF from PCB 138 to the higher chlorinated and more lipophilic PCB 180 slightly decreased (18.73 to 14.98 and 2.65 to 2.12, respectively). The same slight decreasing trend was also seen in growing lambs (Hoogenboom et al., 2015). However, such a decline was not obvious in the present study, although the highest log K_{ow} of the tested congeners in the present study was 7.71 for PCB 189 (IARC, 2016). The PCBs 77, 81, 126 and 169 showed higher transfer factors as their equal chlorinated counterparts. This discrepancy can be attributed to their distinct chlorine substitution pattern, since these are the only PCBs tested with a non-ortho substitution (Fig. S4). Similarly, higher BCFs for the PCB non-ortho substituted congeners were seen in broiler adipose tissue, although such factors were 1.3 to 4.3-fold lower in poultry than in ruminant (Hoogenboom et al., 2004).

4.2.2. Effect of growing cattle physiology

The physiological status has previously been shown to also affect transfer factors (Jondreville et al., 2017; Lerch et al., 2016, 2020b; Rey-Cadilhac et al., 2020). One of the aims of the present study was to explore the effect of growth rate on transfer factors, but only moderate differences between growth rate classes were achieved, resulting in only a slight increasing trend of the PCB transfer factors from slow to fast growth rate. A similar increasing effect of the growth rate on the AE of α-hexabromocyclododecane (α-HBCDD, another lipophilic POP) from feed to abdominal adipose tissue was reported in broilers (Jondreville et al., 2017). Along the fattening period, an increased growth rate is accompanied by a faster body lipid deposition, which results in a lower POP concentration in adipose tissue and subsequently in blood. The presumptive lower blood PCB concentration for the fast growing compared to the slow growing animals could generate a higher POP concentration gradient between the digestive tract and the blood, resulting in a presumably increased absorption rate and further a higher AE. Indeed, the PCB blood concentrations for the bulls decreased from slow to fast growth (PCB 118 decreased in blood from 4200 to 2300 pg g^{-1} lipid, PCB 157 from 180 to 59 pg g^{-1} lipid, PCB 167 from 270 to 180 pg g^{-1} lipid, respectively). For the BCF, however, broiler results were in contrast to the ones reported here for bulls, since the BCF of adipose tissue was higher in slow growing than in fast growing broilers (Jondreville et al., 2017).

The increase in carcass lipid proportion had a decreasing effect on the PCB BCFs and BTFs. This suggests that the higher the carcass lipid proportion, the more dilution space is available for lipophilic contaminants and the lower the transfer factors will be (Fernandes et al., 2011). Alternatively, the decrease in BCF and BTF may also be due to a higher rate of PCB metabolism in cattle having a higher carcass lipid proportion, a hypothesis that deserves further research focusing not only on PCB parent compounds, but also on their metabolites. Since BCFs and BTFs are usually used in risk assessment and management, it is important to consider this carcass lipid proportion as influencing factor. To do so, linear equations that link BCF and BTF to carcass lipid proportion are proposed in Table 3 and may help to further refine existing regressions for beef cattle chemical risk assessment purposes (Dowdy et al., 1996; Hendriks et al., 2007). Indeed, it was shown that an increase in carcass lipid proportion of 4% resulted in a 1.65-fold decrease in the BTF of PCB 126.

For AE from feed to carcass, no effect of carcass lipid proportion was expected, as the carcass burden calculation already accounts for its lipid content, suggesting that equal amounts of PCBs are transferred to the carcass for a given PCB intake independently of carcass lipid proportion. This is an insensitivity of the AE on the lipid accretion rate in growing cattle, which is comparable to the insensitivity of the carry over rate (fraction of the daily PCB intake that is eliminated through the milk) regarding milk fat yield, body lipid dynamic and feed intake in lactating cows (McLachlan and Richter, 1998).

5. Conclusion

The results of this study showed that PCB physicochemical properties, such as the chlorination degree, substitution pattern and lipophilicity, together with the animal physiological status, mainly the carcass lipid proportion, largely affect transfer factors from feed to growing cattle. It was demonstrated that BCF, BTF and AE increased the higher the chlorination degree and that transfer factors for non-ortho substituted congeners were higher compared to their equal chlorinated counterparts. Besides, carcass lipid proportion is negatively correlated with BCF and BTF, which at least in part was attributed to an increase in the dilution space for lipophilic contaminants. The highest correlation between carcass lipid proportion and transfer factors was seen for PCB 126, the most important TEQ contributing congener, where the BTF decreased 1.65-fold with a carcass lipid proportion increase of 4%. Conversely, the effect of growth rate on transfer factors was less obvious and deserves further research in beef cattle, showing larger differences in growth rate classes compared to this study. Such results underpin the complexity and limitations of using average transfer factors for chemical risk assessment and management in livestock farming systems. In order to deal with this complex interaction between POP physicochemical properties and animal physiology, equations were proposed that could help to decrease the uncertainties in transfer factor calculations in case reliable estimates of carcass lipid content can be determined. Another way, to deal with such a complex interaction is to generate more integrative tools, such as physiologically based toxicokinetik models, for which this unique PCB dataset in beef cattle can serve for calibration or validation purposes. For such an aim, also the transfer to other tissues, such as liver and muscle, and its influence by physiology would be mandatory, since toxicokinetic models usually consist of several body compartments.

CRediT author statement

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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.

Acknowledgement

The authors would like to thank Aeby Y. (Agroscope, Switzerland) and his team for diligent feeding, management and weighing of the growing cattle; Oberson JL. (Agroscope) for his technical support along the study; Dougoud B. and the team of the Agroscope Animal Biology unit (Posieux) for the blood and perirenal adipose tissue sampling and 11th rib sampling and dissection. They also want to acknowledge Bongard L., Cavaliere D., Kälin O. and Perrone D. of Empa (Dübendorf, Switzerland) for the POP analyses support and the team of the Agroscope Feed Chemistry unit (Posieux) for feed chemical and nutritive value analyses. C. Driesen acknowledges financial support for a PhD scholarship provided by the Federal Office for Agriculture and Federal Food Safety and Veterinary Office (Switzerland) under the project "AgroPOP" (project number: 4.17.b).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2021.129698.

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