Supplemental Material to

Estimation of Human Body Concentrations of DDT from Indoor Residual Spraying for Malaria Control

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1 Input parameters

1.1 Body weight and body lipid for South African women

In Fig. S1 the temporal development of a South African woman's body weight and body lipid weight is shown. The corresponding lipid fraction is listed in Table S1. The growth-related increase of body weight of South African females was modeled by using mean body weight data of female children (Cameron, 2003), teenagers (Department of Health, 2007), and adults (Puoane et al., 2002). For the pharmacokinetic (PK) model, we linearly interpolated the data to obtain a growth curve for an average South African female. We used the age-dependent lipid fractions for girls from Veldhuis et al. (2005) for the age of 0–17 years; the final lipid fraction was assumed to be 30% for the age of 20 and above (Levitt et al., 2005) for a nulliparous female. With every birth, the mother loses more water than fat (ICRP, 1975). Consequently, her lipid fraction increases accordingly and was set constant for the rest of her life. Two studies report lipid fraction in South African children (Amusa et al., 2011; Monyeki et al., 2005). Amusa et al. (2011) reported fairly constant body lipid fraction of 25% for girls from grade 1 to 7, and Monyeki et al. (2005) reported 15.6–18.6% for girls aged 7–14 years. Because of this high range found in these two studies, we used the values presented by Veldhuis et al. (2005), which are between the extremes. Pregnancy weight gain was assumed to be 0.3 kg/week (IOM, 1996). After childbirth, the woman reaches the pre-pregnancy weight after almost four months. Pregnancy duration was assumed to be 270 days (~ 39 weeks) (Kruger, 2005). For subsequent children, the same weight gain and weight loss pattern was assumed.

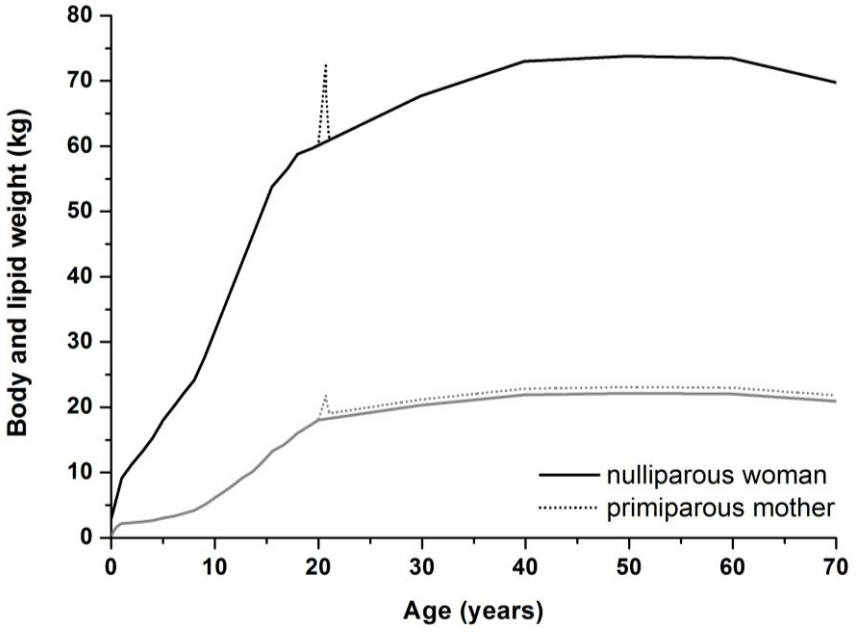

Fig. S1: Development of the body (black) and lipid weight (grey) of a nulliparous (solid line) and a primiparous South African woman (dotted).

Table S1: Body weight and lipid fraction at different ages of a nulliparous South African woman.

age	body weight (kg)	lipid fraction (–)
birth	3.0	0.15
1 month	3.5	0.16
2 months	4.0	0.20
3 months	4.5	0.24
4 months	5.0	0.25
5 months	5.5	0.26
6 months	6.0	0.26
7 months	6.5	0.26
8 months	7.0	0.26
9 months	7.5	0.25
10 months	8.1	0.25
11 months	8.6	0.24
1 year	9.1	0.24
2 years	11.3	0.21
5 years	18.0	0.17
10 years	31.6	0.20
15 years	51.7	0.24
20 years	60.1	0.30
40 years	73.0	0.30
60 years	73.5	0.30
70 years	69.8	0.30

1.2 Uptake via inhalation

Uptake via inhalation of ΣDDT (=*p,p'*-DDT and *o,p'*-DDT) and ΣDDE (=*p,p'*-DDE and *o,p'*-DDE) was estimated according to Eq. S1:

$$U_{i,\mathrm{inh}}(t_{\mathrm{age}}) = E_{\mathrm{inh}} \times f_{\mathrm{indoor}} \times r_{\mathrm{inh}}(t_{\mathrm{age}}) \times c_{i,\mathrm{air}} \qquad \text{(Eq. S1)}$$

where $U_{i,\mathrm{inh}}$ (t_{age}) is the age-dependent uptake of substance *i* (*i* = ΣDDT or ΣDDE) (ng_i/d) via inhalation, E_{inh} is the uptake efficiency (dimensionless-), f_{indoor} is the time spent indoors (h/d), $r_{\mathrm{inh}}(t_{\mathrm{age}})$ are the default values for an age-dependent inhalation rate for women (m^3/d), $c_{i,\mathrm{air}}$ is the ΣDDT or ΣDDE concentrations in indoor air (ng/m^3) used for our base case scenario. The corresponding values are shown in Table S2 and Table S3.

Table S2: Inhalation rates (r_{inh}) for women (U.S. EPA, 1997)

age group (years)	inhalation rate (r_{inh}) in m^3/d
0–1	4.5
1–2	6.8
2–5	8.3
5–8	10
8–11	13
11–19	12
>19	11.3

Table S3: Values for ΣDDT and ΣDDE concentrations in indoor air (Bouwman et al., 2009; Singh et al., 1992; Ritter et al., 2011; van Dyk et al., 2010), uptake efficiency (E_{inh}) and time spent indoors (f_{indoor}) (Bouwman et al., 2009)

parameter	symbol	unit	value
ΣDDT concentration	$c_{\Sigma DDT,air}$	ng/m^3	5 000
ΣDDE concentration	$c_{\Sigma DDE,air}$	ng/m^3	185
uptake efficiency via inhalation	E_{inh}	–	1
time fraction spent indoors	f_{indoor}	h/d	8

1.3 Uptake via diet

The daily uptake of ΣDDT and ΣDDE was calculated for each food item individually, namely chicken muscle, chicken fat, chicken egg, leafy vegetables, and fish according to Eq. S2 and S3:

$$U_{i,\mathrm{diet}}(t_{\mathrm{age}}) = E_{\mathrm{diet}} \times \sum_{\mathrm{j}} (r_{i,j}(t_{\mathrm{age}}) \times c_{i,j}) \qquad \text{(Eq. S2)}$$

$$U_{i,\mathrm{fish}}(t_{\mathrm{age}}) = E_{\mathrm{diet}} \times (r_{i,fish}(t_{\mathrm{age}}) \times f_{lip,fish} \times c_{i,fish}) \qquad \text{(Eq. S3)}$$

where $U_{i,\mathrm{diet}}(t_{\mathrm{age}})$ is the overall daily uptake of substance i (i = ΣDDT or ΣDDE) (ng/d) from food items j (j = chicken muscle, chicken fat, chicken egg, leafy vegetables), E_{diet} is the dietary uptake efficiency (dimensionless; here assumed to be equal to 1) for all food items, $r_{i,j}(t_{\mathrm{age}})$ is the daily average consumption rate per capita (g/d) of the food item j, and $c_{i,j}$ is the ΣDDT or ΣDDE concentration (ng/g) in each food item j on wet-weight basis (van Dyk et al., 2010). Because Barnhoorn et al. (2009) reported lipid-normalized concentrations for the fish *O. mossambicus*, we used Eq. S3 for the uptake of chemical with this particular fish; in Eq. S3, $U_{i,\mathrm{fish}}(t_{\mathrm{age}})$ is the daily uptake of i from fish (ng/d), $r_{i,\mathrm{fish}}(t_{\mathrm{age}})$ is the consumption rate of fish (g/d), $f_{\mathrm{lip,fish}}$ is the lipid fraction of *O. mossambicus* (dimensionless), and $c_{i,\mathrm{fish}}$ is the lipid-normalized concentration (ng/g_{lip}).

The daily consumption rates for chicken meat, fish, chicken eggs, and leafy vegetables were derived from Nel and Steyn (2002), see Table S4. Nel and Steyn (2002) reported daily consumption rates per capita for the age categories 1–5 years, 6–9 years, and >10 years. Because we used differently defined age categories in our model calculations (0.5–3 years, 3–6 years, 6–10 years, 10–50 years, and >50 years), we attributed the values from Nel and Steyn (2002) to the age categories 3–6 years, 6–10 years, and 10–50 years. In order to obtain consumption rates for the additional age categories

of 0.5–3 years and >50 years, we applied proportionality factors according to the recommended calorific intake for an adult female by Rose et al. (2002), see Table S5. The amount of chicken meat presented by Nel and Steyn (2002) was assumed to consist of roughly 90% muscle and 10% fat. The overall fat content of native South African chicken was calculated to be about 10% based on chicken carcass composition reported by van Marle-Köster and Webb (2000) and on the lipid content in chicken skin, white meat, and dark meat (50% white and 50% dark meat, rough assumption) presented by van Heerden et al. (2002) for South African chicken.

For the concentration of ΣDDT or ΣDDE in chicken fat, we combined the data reported by van Dyk et al. (2010) and by Barnhoorn et al. (2009). Both studies investigated the contaminant concentrations in chicken fat. However, based on the arithmetic mean, the values reported by both studies differ by a factor of 6 for DDT and by 24 for DDE. Because both studies cover chickens which lived in villages where IRS is performed regularly and seem to be equally valid, we gave the same weight to the data from these two studies. In doing so, we (i) used the median values (as presented by van Dyk et al., 2010, and re-calculated from the data reported by Barnhoorn et al., 2009) instead of mean values since the median is more robust and outliers have less weight; (ii) accounted for the high variability by choosing the median values of van Dyk et al. (2010) as the upper bound and the median values of Barnhoorn et al. (2009) as the lower bound; and (iii) used the median of the medians of both studies as the intermediate concentrations and set them as our default concentrations for ΣDDT and ΣDDE, see Table S6.

Barnhoorn et al. (2009) measured the ΣDDT and ΣDDE concentrations in fish fat of Mozambique tilapia (*Oreochromis mossambicus*). The fat content of this fish was assumed to be 3.6% (Naeem et al., 2011; Abou et al., 2011). The median ΣDDT and ΣDDE concentrations on wet-weight basis in chicken eggs were provided by Riana Bornman (unpublished data).

Table S4: Food item consumption rates from Table 26 in Nel and Steyn (2002)

age group (years)	chicken meat (g/d)	chicken eggs (g/d)	fish (g/d)	leafy vegetables (g/d)
0.5–3	6.3	4.8	3.6	18.1
3–6	8.7	6.7	5.0	25.2
6–10	10.3	5.2	3.9	21.9
10–50	16.9	13.9	5.4	41.6
>50	14.5	12.0	4.6	35.8

Table S5: Proportionality factors used to obtain age-adjusted food consumption rates for 0.5-3 years and >50 years (Rose et al., 2002)

age group (years)	proportionality factor (–)
0.5–3	0.59
10–50	1
>50	0.86

Table S6: Median ΣDDT and ΣDDE concentrations in different food items (Barnhoorn et al., 2009; van Dyk et al., 2010). Concentration in chicken eggs was provided by Riana Bornman (unpublished).

food item	unit	ΣDDT	ΣDDE
chicken muscle	ng/g	134	263
chicken fat (default[1])	ng/g	24 440	47 086
chicken fat (upper bound[2])	ng/g	45 380	87 072
chicken fat (lower bound[3])	ng/g	3 500	7 100
chicken eggs	ng/g	4 037	6 879
leafy vegetables	ng/g	70	10
fish	ng/g_{lip}	3 721	3 697

[1] median of median concentrations presented by van Dyk (2010) and Barnhoorn et al. (2009)
[2] sum of median concentrations of *p,p'*- and *o,p'*-isomer of DDT and DDE (van Dyk et al., 2010)
[3] median concentrations of *p,p'*-isomers of DDT and DDE (Barnhoorn et al. 2009)

1.4 Uptake via breast milk

We assumed that ΣDDT and ΣDDE lost by the mother via breastfeeding is directly ingested by the infant. The ΣDDT and ΣDDE uptake via breast milk, $U_{i,\mathrm{milk}}(t_{\mathrm{age}}^{\mathrm{infant}})$, in ng/d, was calculated as

$$U_{i,\mathrm{milk}}(t_{\mathrm{age}}^{\mathrm{infant}}) = E_{\mathrm{milk}} \times k_{\mathrm{bf}}(t_{\mathrm{age}}^{\mathrm{mother}}, t_{\mathrm{bf}}) \times m_i(t_{\mathrm{age}}^{\mathrm{mother}}) \qquad \text{(Eq. S4)}$$

where $k_{\mathrm{bf}}(t_{\mathrm{age}}^{\mathrm{mother}}, t_{\mathrm{bf}})$ is the rate constant (1/d) for breastfeeding from Eq. 5 of the main text, E_{milk} is the uptake efficiency from breast milk (dimensionless), and $m_i(t_{\mathrm{age}}^{\mathrm{mother}})$ is the mass (ng) of substance *i* in lipids as a function of the mother's age. Table S7 shows the values used for the calculation of the uptake via breast milk. The infants were always exclusively breastfed and therefore, $U_{i,\mathrm{diet}}$ was replaced by $U_{i,\mathrm{milk}}$ for the whole duration of breastfeeding. The corresponding mass balance (Eq. S5) for the infant is described as

$$\frac{dm_i(t_{\mathrm{age}}^{\mathrm{infant}})}{dt} = U_{i,\mathrm{milk}}(t_{\mathrm{age}}^{\mathrm{infant}}) + U_{i,\mathrm{inh}}(t_{\mathrm{age}}^{\mathrm{infant}}) - \left(k_{i,\mathrm{met}}(t_{\mathrm{age}}^{\mathrm{infant}}) + k_{ex}(t_{\mathrm{age}}^{\mathrm{infant}})\right) \times m_i(t_{\mathrm{age}}^{\mathrm{infant}}) \qquad \text{(Eq. S5)}$$

Table S7: Values used for breast milk uptake

Parameter	Symbol	Unit	Value	Reference
lipid content in breast milk	$f_{\mathrm{lip,milk}}$			
0–4 months		%	3.27	Bouwman (1990)
5–8 months		%	3.82	Bouwman (1990)
9–12 months		%	4.24	Bouwman (1990)
13–24 months		%	4.99	Bouwman (1990)
breast milk consumption rate during 1. year	r_{milk}	g/d	800	Bouwman et al. (2006)
breast milk consumption rate during 2. year	r_{milk}	g/d	600	da Costa et al. (2010)
uptake efficiency via breast milk	E_{milk}	–	1	

1.5 Intrinsic elimination

The overall intrinsic elimination of ΣDDT and ΣDDE is the sum of metabolic degradation and non-metabolic excretion (Kreuzer et al., 1997). Because excretion is represented directly in the mass-balance equation of the model, the uptake efficiency of the chemical is implicitly calculated by the model itself. Therefore, we assumed an initial uptake efficiency of 100% from all exposure routes (E_{diet}, E_{inh}, E_{milk}). The daily lipid excretion via feces was linearly interpolated between birth and the age of 18 (= adult) (ICRP, 1975). For female adults over 18 years, we assumed constant fecal lipid excretion. All parameter values are shown in Table S8.

Table S8: Parameter values for elimination processes

parameter	symbol	unit	value	reference
liver weight as fraction of body weight	f_{liver}	%	2.4	ICRP (1975)
reference liver weight	$m_{\text{liv,ref}}$	kg	1.8	this work
reference lipid weight	$m_{\text{lip,ref}}$	kg	21.9	this work
density of liver	p_{liv}	kg/L	1.0	Brown et al. (1997)
density of lipids	p_{lip}	kg/L	0.9	Brown et al. (1997)
reference metabolic rate constant for ΣDDT	$k^{\text{ref}}_{\Sigma\text{DDT,met}}$	1/d	6.6E-4	this work
reference metabolic rate constant for ΣDDE	$k^{\text{ref}}_{\Sigma\text{DDE,met}}$	1/d	1.0E-4	this work
female fecal lipid excretion	$r_{\text{lip,feces}}$			
birth		g_{lip}/d	3.0	ICRP (1975)
18 years		g_{lip}/d	4.5	ICRP (1975)
>18 years		g_{lip}/d	4.5	ICRP (1975)

2 Sensitivity analysis

We performed a sensitivity analysis by increasing and decreasing 15 different model parameters by factors of 1.5 and 0.67, respectively, to identify which parameters most strongly influence the modeled concentration curve shown in Fig. 1 in the main text. We recorded the change in concentration levels by adjusting one parameter at a time; Table S9 presents the 15 parameters considered plus the lipid fraction in breast milk for which we test a constant and a time-variant value. For the last five parameters listed in Table S9, the model sensitivity is very low and, therefore, their effects are not displayed graphically.

Fig.S2 shows the effects of changes in body weight and body lipid fraction on the value of the predicted concentration. These two parameters have the strongest influence on the model results, because they determine the conversion of mass into concentration (Eq. 2 in the main text) and also affect the all the rate constants (metabolic elimination, non-metabolic elimination, and the ΣDDT and ΣDDE elimination during breastfeeding), see Eq. 3–5 in the main text. The effects of these two variables are identical because they occur in the model always in the product of $bw \cdot f_{\text{lip}}$.

Varying the reference values of liver and lipid volume of a 40-year-old South African woman has a similar effect on the predicted concentration as shown in Fig.S2, see Fig. S3. These two parameters influence the first-order rate constant for metabolic elimination $\left(k_{i,\text{met}}, k^{\text{ref}}_{i,\text{met}}\right)$. The reference liver volume and lipid volume are calculated by dividing the reference masses of liver ($m_{\text{liv,ref}}$) and lipid ($m_{\text{lip,ref}}$) by their densities (p_{liv}, p_{lip}). Therefore, we varied the reference mass of liver and lipid by a factor of 1.5 and 0.67. The predicted concentrations differ by a factor of approximately 1.5 from the

base case scenario of a nulliparous woman when either of these parameters is changed a by a factor of 1.5 or 0.67.

Fig. S4 shows the effects of five parameters related to the consumption of chicken on the value of the predicted concentration. The five parameters have similar influences on the model results; the concentrations calculated with the model increase by a factor of approximately 1.2 when any of these parameters is increased by a factor of 1.5. The parameter for which there is considerable model sensitivity as well as a wide range of actual variability is the concentration of total DDT in chicken fat. This is why the ranges shown in Fig. 1 of the main text are based on the variability of this parameter.

Fig. S5 shows the effect of a constant vs. time-variant lipid fraction of the breast milk over the course of breastfeeding as well as the effect of variation of the consumed amount of breast milk by factors of 1.5 and 0.67. The change in predicted concentrations is less than a factor of 1.3 for both parameters.

Fig. S6 shows the age-dependent intrinsic elimination half-lives of ΣDDT and ΣDDE. As described in Kreuzer et al. (1997), infants and children have a much faster elimination rates, which finally level off at the values reported in Ritter et al. (2009).

Table S9: Model parameters for which the sensitivity of the modeled concentration of total DDT (=ΣDDT and ΣDDE) is determined.

parameter	symbol	unit	shown in
female body weight	bw	kg	Fig.S2
female body lipid fraction	f_{lip}	–	Fig.S2
reference liver weight	$m_{liv,ref}$	kg	Fig. S3
reference lipid weight	$m_{lip,ref}$	kg	Fig. S3
mass of chicken consumed daily	$m_{chicken}$	g/d	Fig. S4
concentration in chicken fat	$c_{chicken\ fat}$	ng/g	Fig. S4
concentration in chicken egg	c_{egg}	ng/g	Fig. S4
fraction of chicken fat to overall mass of chicken	$f_{chicken\ fat}$	–	Fig. S4
mass of chicken egg consumed	m_{egg}	g/d	Fig. S4
lipid fraction of breast milk (constant or variable)	$f_{lip,milk}$	–	Fig. S5
mass of breast milk consumed	m_{milk}	g/d	Fig. S5
fraction of time spent indoors	f_{indoor}	–	not shown
concentration in indoor air of ΣDDT and ΣDDE	c_{air}	ng/m^3	not shown
inhalation rate	r_{inh}	m^3/d	not shown
mass of fish	m_{fish}	g/d	not shown
mass of leafy vegetables	m_{veg}	g/d	not shown

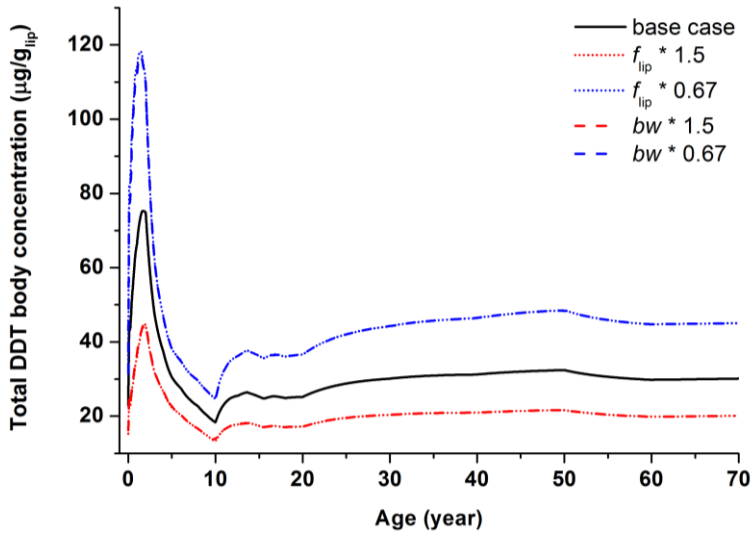

Fig.S2: Effect of body weight (*bw*) and body lipid fraction (f_{lip}) on the calculated concentrations (changes of the base case values by factors of 1.5 and 0.67).

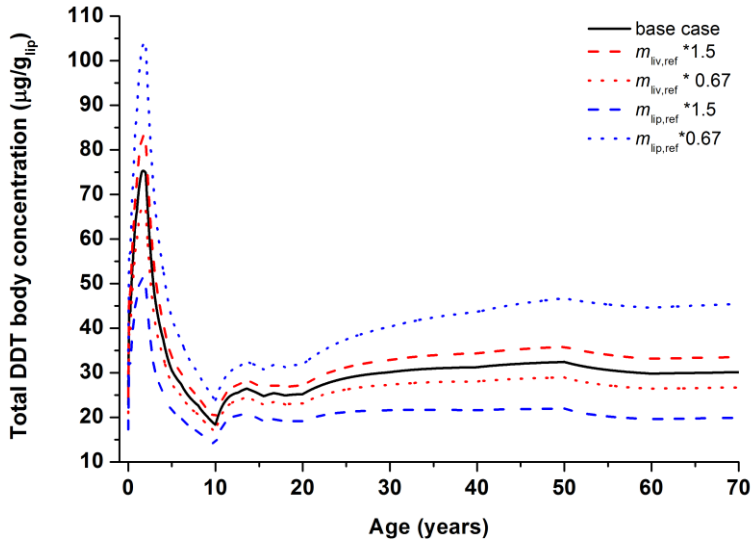

Fig. S3: Effect of reference liver weight ($m_{liv,ref}$) and reference lipid weight ($m_{lip,ref}$) on the calculated concentrations (changes of the base case values by factors of 1.5 and 0.67).

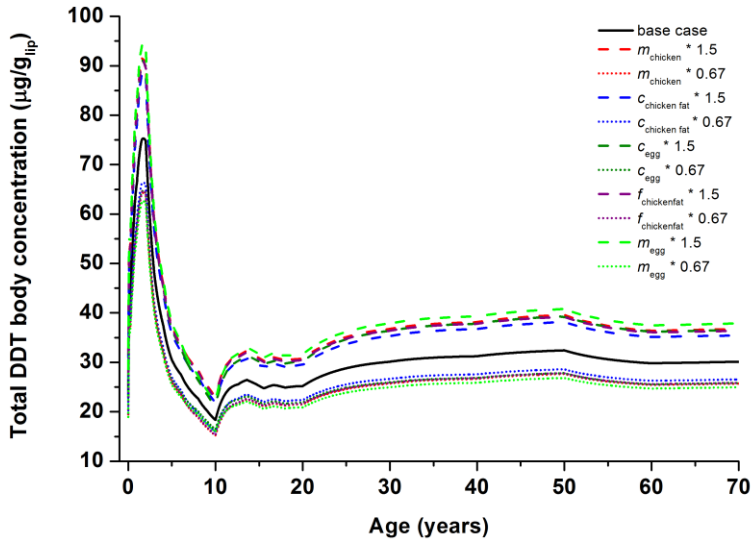

Fig. S4: Effect of the five model parameters related to consumption of chicken on the calculated concentrations (changes of the base case values by factors of 1.5 and 0.67). For abbreviations used in the legend, see Table S9.

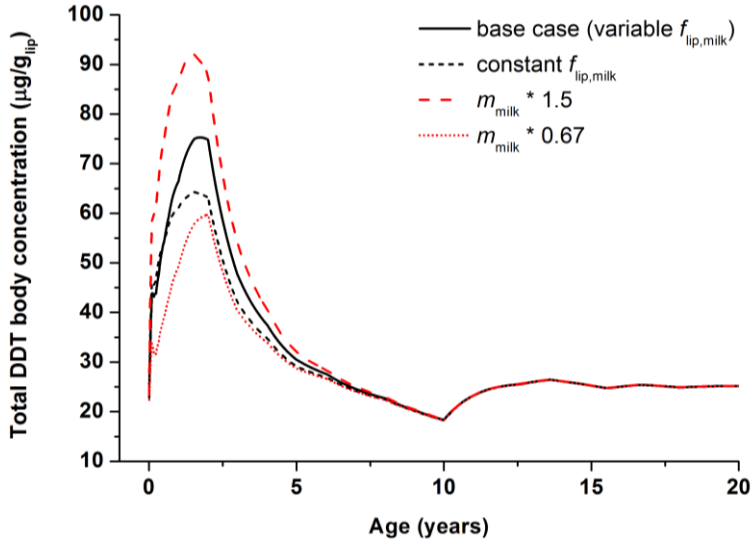

Fig. S5: Effect of the daily consumption rate of breast milk (m_{milk}) on the calculated concentrations (changes of the base case values by factors of 1.5 and 0.67). Further, the effect of constant and time-variant lipid fraction of breast milk is shown ($f_{lip,milk}$).

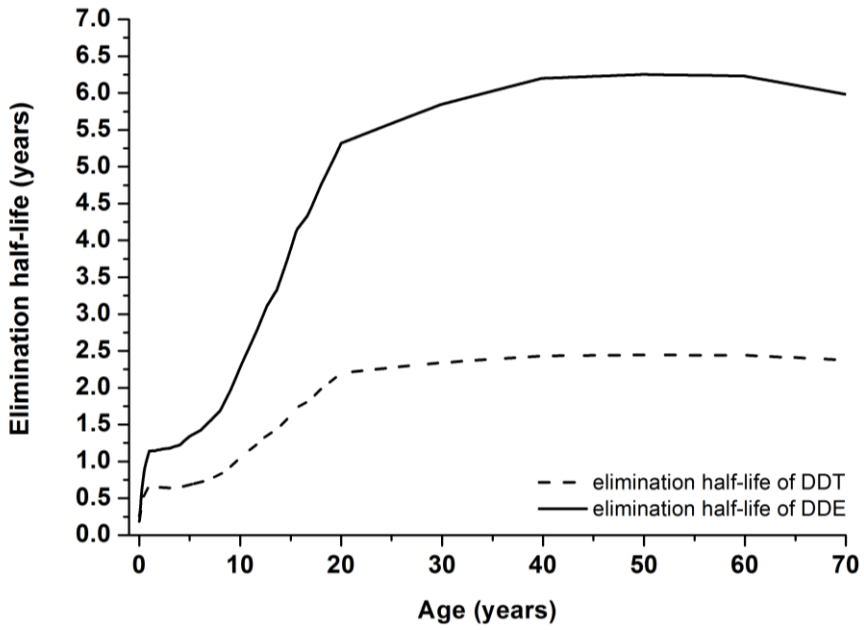

Fig. S6: Age-dependent intrinsic elimination half-lives of ΣDDT and ΣDDE calculated by the model.

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