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Assessment and improvement of biotransfer models to cow's milk and beef used in exposure assessment tools for organic pollutants

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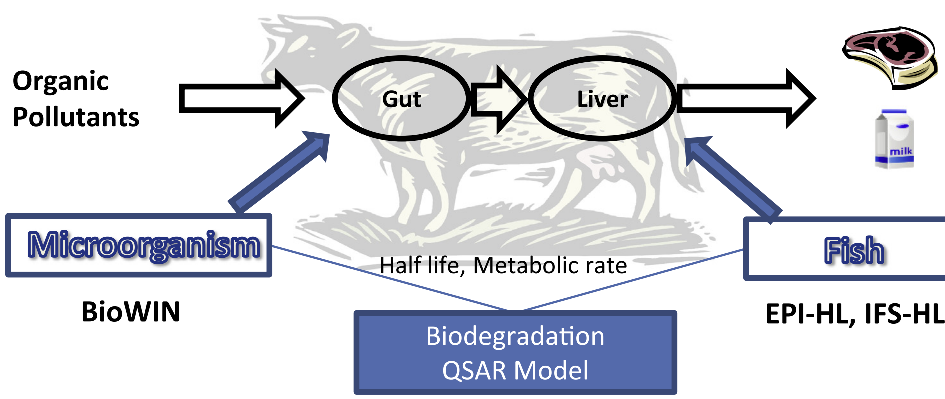
HIGHLIGHTS

- A simple method for estimating cattle biotransfer of organic compounds is proposed.
- The biodegradation of microorganisms mimics the cattle metabolism in gut.
- The biodegradation of fish mimics the cattle metabolism after the absorption.
- The two component metabolic rate significantly improves the model performance.

GRAPHICAL ABSTRACT



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ABSTRACT

The aim of this study was to assess and improve the accuracy of biotransfer models for the organic pollutants (PCBs, PCDD/Fs, PBDEs, PFCAs, and pesticides) into cow's milk and beef used in human exposure assessment. Metabolic rate in cattle is known as a key parameter for this biotransfer, however few experimental data and no simulation methods are currently available. In this research, metabolic rate was estimated using existing QSAR biodegradation models of microorganisms (BioWIN) and fish (EPI-HL and IFS-HL). This simulated metabolic rate was then incorporated into the mechanistic cattle biotransfer models (RAIDAR, ACC-HUMAN, OMEGA, and CKow). The goodness of fit tests showed that RAIDAR, ACC-HUMAN, OMEGA model performances were significantly improved using either of the QSARs when comparing the new model outputs to observed data. The CKow model is the only one that separates the processes in the gut and liver. This model showed the lowest residual error of all the models tested when the BioWIN model was used to represent the ruminant metabolic process in the gut and the two fish QSARs were used to represent the metabolic process in the liver. Our testing included EUSES and CalTOX which are K_{OW}-regression models that are widely used in regulatory assessment. New regressions based on the simulated rate of the two metabolic processes are also proposed as an alternative to K_{OW}-regression models for a screening risk assessment. The modified CKow model is more physiologically realistic, but has equivalent usability to existing K_{OW}-regression models for estimating cattle biotransfer of organic pollutants.

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1. Introduction

Biotransfer of organic pollutants to cattle is an important process in quantifying the exposure of humans to toxic chemicals. Surveys in Germany and Canada have demonstrated that over 50% of dioxin and furan exposure to humans was through ingestion of cattle products (Birmingham et al., 1989; Fürst et al., 1990). In 1988, Travis and Arms (1988) proposed simple regressions between the octanol–water partition coefficient (K_{OW}) and a biotransfer factor (BTF) for milk and beef from experimental data, and this model has been incorporated by international regulatory authorities (e.g. European Chemical Agency, California Environmental Protection Agency) into their chemical exposure assessment tools for human health (Mckone, 1993; European Chemicals Bureau, 2010). These tools are routinely used by regulatory authorities to determine the risk to human health from organic polluted soils; the accuracy of the cattle biotransfer model is therefore critical for robust health risk assessment.

Despite the extensive adoption of the Travis and Arms (1988) model, its validity has been questioned by many authors (McKone and Ryan, 1989; McLachlan, 1993; Staples et al., 1997; Birak et al., 2001). Their criticisms were based on: a limited amount of data, which all relate to persistent chemicals with a narrow K_{OW} range ($3 < \log K_{OW} < 7$), a high residual error in the derived regression equations and, when K_{OW} exceeds $10^{6.5}$, the model has an increase in the BTF but observations show that BTF decreases as K_{OW} increases (McKone and Ryan, 1989; McLachlan, 1993; Staples et al., 1997; Birak et al., 2001). An alternative approach was to generate new K_{OW}-regression models using a larger amount of experimental data, as proposed by MacLachlan and Bhula (2008) and the Research Triangle Institute (RTI) (2005). Hendriks et al. (2007) reported a very weak correlation between K_{OW} and BTF when labile and persistent chemical data were analysed separately (r^2 = 0.35 for labile, 0.02 for persistent). The BTF was more significantly affected by the metabolism of individual chemicals in cattle rather than their hydrophobicity (Staples et al., 1997; Hendriks et al., 2007). Therefore, the widely used K_{OW}-regressions would appear to have a limited theoretical basis. A regression model using the molecular connectivity index (MCI) to characterise the chemical behaviour and metabolism in cattle instead of K_{OW} was proposed by Dowdy et al. (1996), although the USEPA reported that there was no significant difference in performance between this approach and the Travis and Arms (1988) model using their data set (US EPA, 2005).

Mechanistic cattle biotransfer models have also been constructed, for example, ACC-HUMAN (Czub and McLachlan, 2004) based on Mclachlan (1994) model, RAIDAR (Arnot and Mackay, 2008) and OMEGA (Hendriks et al., 2007). All these models are based on mass balance of pollutants between the input, e.g. ingestion of pollutants, and the output, e.g., excretion with milk, faeces, and urine, and metabolism. McLachlan (1994) noted that the metabolic rate and absorption efficiency were the key parameters. However, the specific metabolic rate in cattle for each pollutant needs to be known in all the three models and there are few actual data. To date no simple cattle model has been developed for the metabolism of chemicals based on their chemical properties and this has resulted in the limited applicability of mechanistic models to a broad range of pollutants.

More recently, Rosenbaum et al. (2009) introduced a linear regression of the metabolic rate and K_{OW} into their newly developed model, CKow. When using this approach the model fit to observed BTFs was better than the K_{OW}-regressions of Travis and Arms (1988) and RTI (2005). However, the model accuracy might be limited for a wide range of organic chemicals because of the considerable deviation of the measured from the estimated metabolic rate in their model set (up to two orders of magnitude). Therefore, alternative approaches for deducing the metabolic rate in cattle for various pollutants need to be considered.

In regulatory risk assessment, Quantitative Structure–Activity Relationships (QSARs) are often preferred practically for filling data gaps to reduce costs and prevent animal studies which may have ethical barriers (Cefic & VCI, 2009). Another case for filling the data gaps is using parameter values obtained from other species, called species read-across in this study. For example, Arnot et al. (2010) used the measured metabolic rate in fish as a substitute for avian and mammalian species with the biological explanation of each metabolism. These substitution techniques should also be useful for estimating the cattle metabolism of a wide scope of organic pollutants targeted by the regulatory authorities.

The aim of this study was to assess and improve the accuracy of biotransfer models of organic pollutants to cow's milk and meat for use in human exposure assessment, focusing on the metabolism and the absorption of these contaminants in cattle. This was achieved through QSARs and the species read-across approach, specifically the metabolic rate in cattle was estimated by QSAR biodegradation models of microorganisms (the Biodegradation Probability Program for Windows, BioWIN) and fish (EPI-HL and IFS-HL). The performance of cattle biotransfer estimation using the estimated metabolic rate was then assessed with experimental data and predictions of other existing models.

2. Methods

The iterative process for improving performance of cattle biotransfer models was:

(1) check the performance of existing models, based on an assessment of the residual error between the simulated and observed BTFs, against a broad range of experimental data;
(2) introduce the QSAR and the species read-across approach to these models to deduce the metabolic rate;
(3) check the improvement of the model performance following the optimisation of parameters like the absorption efficiency;
(4) re-build the model regression using the simulated metabolic rate as a predictor.

The biotransfer of organic pollutants to milk and meat can be expressed in three ways: bio-concentration factor (BCF), biotransfer factor (BTF), and carry-over rate (COR) (Thomas et al., 1999):

$$BCF = \frac{\text{concentration in meat or milk } (\text{mg} \cdot \text{kg}^{-1})}{\text{concentration in feed } (\text{mg} \cdot \text{kg}^{-1})} \quad (1)$$

$$BTF = \frac{\text{concentration in meat or milk } (\text{mg} \cdot \text{kg}^{-1})}{\text{daily intake of chemicals } (\text{mg} \cdot \text{kg}^{-1})} \quad (2)$$

$$COR = \frac{\text{chemical flux in the milk } (\text{mg} \cdot \text{day}^{-1})}{\text{daily intake of chemicals } (\text{mg} \cdot \text{day}^{-1})} \quad (3)$$

In addition, BTFs to whole milk (BTF_{milk}) and meat (BTF_{meat}) were adopted in this study and other criteria such as BTFs to milk lipid were converted to BTF using values of daily intake of feed (16 kg d^{-1} for lactating cow, 8 kg d^{-1} for non-lactating cattle), milk mass flow (23 kg d^{-1}), and lipid fraction in milk (0.04) and meat (0.25) in manner of previous models (Dowdy et al., 1996; Hendriks et al., 2007; Rosenbaum et al., 2009).