ABSORPTION AND EXCRETION OF ^{14}C- PERFLUOROOCTANOIC ACID (PFOA) AND PERFLUOROOCTANE SULFONATE (PFOS) IN BEEF CATTLE

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Introduction

Perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) are industrially produced chemicals. Due to the amphiphilic nature of PFOA and PFOS, they can be used as surfactants and coatings in many industrial, commercial and consumer products. However, the extensive use of these chemicals has led to a widespread occurrence in humans[1-2], wildlife[3-5] and the environment[6-7]. As a result of the persistence of PFOS in humans and the environment, PFOS was listed as a persistent organic pollutant under the Stockholm Convention in 2009[8]. PFOS and PFOA are slowly excreted by humans and have been observed in biosolids[6-7], suggesting exposure of agricultural animals to these chemicals may occur through application of biosolids to cattle pastures and animal food crops. Possible accumulation of PFOS and PFOA in edible tissues of agricultural animals would be a cause for concern for the United States food supply and human health. As such, the United States Department of Agriculture (USDA) has undertaken a study to determine the absorption, distribution, metabolism, and excretion of PFOA and PFOS in beef cattle following an oral dose. This study will help to determine the extent to which these chemicals are likely to be observed in edible tissues of beef cattle. The data will also be used to assess if consumption of beef plays a significant role in human exposure and accumulation of PFOA and PFOS.

Materials and methods

Four Lowline Angus steers (281-366 kg) were given single oral bolus doses containing [1-^{14}C]-PFOA (1 mg/kg, 0.6 mCi per steer) and three steers were provided simultaneous single oral bolus doses of unlabeled PFOS (10 mg/kg). Steer 181 was not dosed with PFOS and served as a PFOS control. Plasma, urine and feces were collected prior to and after dosing at various time intervals from each steer for 28 d. After 28 d steers were euthanized, deboned, and tissues collected. The animal protocol was approved by the local Institutional Animal Care and Use Committee and by the USDA Radiation Safety Committee.

^{14}C-PFOA derived radioactivity in plasma and urine was determined by liquid scintillation counting (LSC) and in the feces and tissues by combustion analysis followed by LSC. An ion pairing liquid-liquid extraction (IP-LLE) was utilized for PFOA and PFOS extractions[7] prior to quantification by liquid chromatography-quadrupole time of flight mass spectrometry (LC-QToF).

Results and discussion

Figure 1 shows the relationship of plasma ^{14}C-PFOA concentration and time post-dose. Peak concentrations of ^{14}C-PFOA in plasma occurred between 24 and 36 h post-dose, and the first-order elimination half-life in plasma for ^{14}C-PFOA was 19.2 ± 3.3 h using a two compartment analysis model. For comparison the PFOA serum half-lives in male rodents and non-human primates were on the order of 10-30 d, while in female rats PFOA is eliminated in serum even faster with a half-life of 3-5 h[9-10]. The ^{14}C-PFOA half-life in steer plasma is on the order of the female rats' serum half-life. Conversely, plasma PFOS concentrations (Figure 2) in the cattle did not decline appreciably during the 28 d study and an elimination half-life could not be determined. PFOS plasma concentrations increased over the first 48 h post-dose and remained elevated for the rest of the study (Figure 2). An estimated serum half-life in monkeys was 200 days in a long term study[11] indicating that PFOS is retained for longer periods than PFOA and

possible monitoring of this compound may be necessary. PFOS serum half-life in humans is estimated to be on the order of years[12] indicating possible accumulation and continual exposure.

Differences in elimination patterns were observed between PFOA and PFOS. The majority of the ^{14}C-PFOA derived radioactivity was quantitatively recovered in urine (100.8 ± 3.3%) with over 90% eliminated within 8 d of dosing having an elimination curve quantitatively similar to that observed in the plasma. Appreciable renal excretion of PFOS did not occur, with only 0.054 ± 0.0071% of the PFOS dose recovered in urine during the 28 d study. Although little PFOS was excreted in urine on a percentage basis, the urinary concentration of PFOS remained relatively consistent (0.127 ± 0.014 µg/mL) throughout the study reaching a plateau approximately 3 d after dosing. Total fecal excretion of ^{14}C-PFOA was 4.6 ± 2.8% with the majority eliminated within the first 9 d after dosing. Within the first 11 d post-dose, 5.8 ± 1.4% of the PFOS dose was excreted in feces with levels still elevated (0.997 ± 0.26 µg/g) on day 11. PFOS in feces collected from 12 to 28 d post-dosing are still being analyzed.

Considering that the majority of ^{14}C-PFOA was eliminated in the urine during the first week of the study, accumulation in edible tissues is unlikely. Radioactive residues in the muscle, liver, kidney, lung, spleen and carcass remainder were not detectable at the time of slaughter (28 d). Also no likely metabolites were observed in the tissue or plasma by LC-MS. PFOS concentrations remained elevated in the plasma over the course of the 28 d study and PFOS was quantified in muscle, liver, kidney, lung, spleen and the remainder of the carcass; accumulation of PFOS in edible tissues is likely and warrants further study (Figure 3).

The percentage of the dosed PFOS still circulating in the plasma fraction after 28 d was relatively high, approximated to be 30-40% based on total blood volumes of steers calculated from live body weights[13]. Average percentages of the dosed PFOS recovered in muscle, liver, kidney, lung, spleen and carcass remainder at time of kill were 4%, 2%, 0.1%, 0.2%, 0.05% and 6%, respectively. These data indicate that while distribution and accumulation of PFOS occurs in body tissues, specifically the muscle and liver, the blood plasma appears to be the major PFOS reservoir in the scope of this study. Remaining tissues are still being analyzed for PFOS residues.

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Figure 1. ^{14}C-PFOA elimination curve in steer plasma with concentrations (µg/mL) through day 15. Concentrations were at background by day 10.

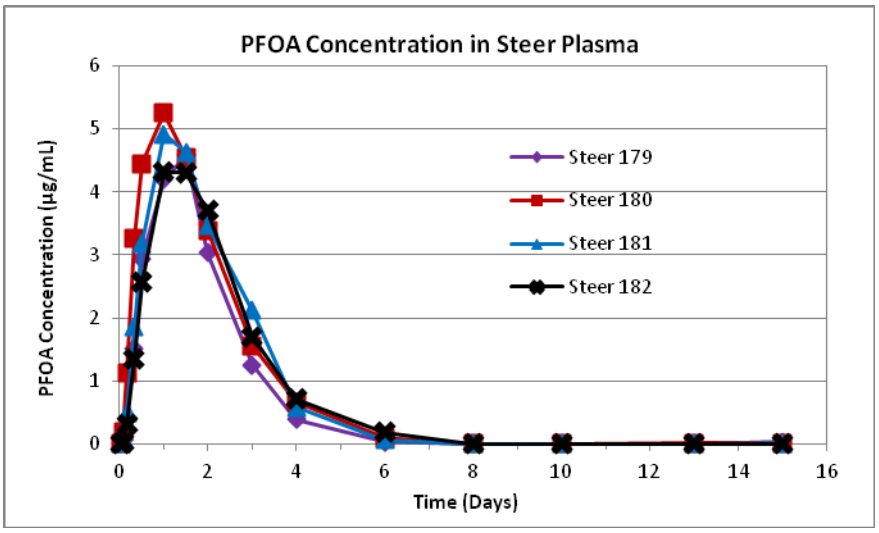

Figure 2. PFOS concentrations (µg/mL) in steer plasma through slaughter (day 28).

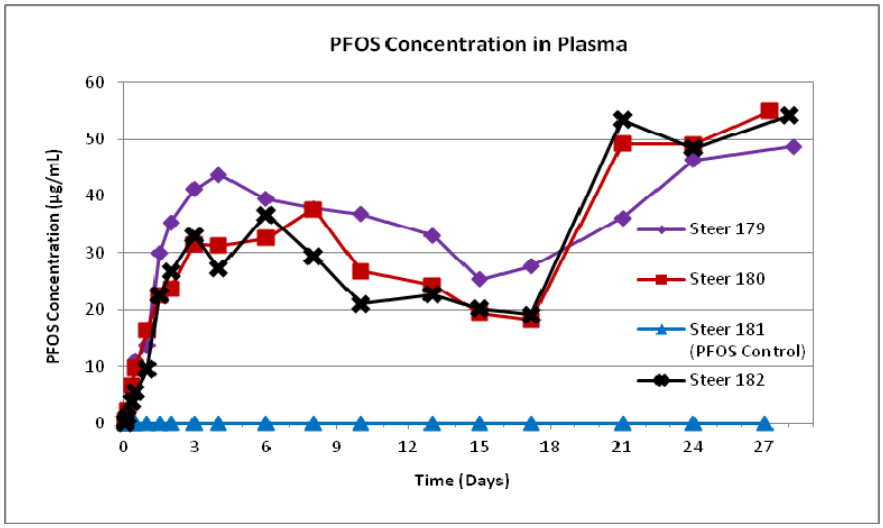

Figure 3. PFOS concentrations in steer tissues and excreta at 28 days post dosing. Plasma and urine reported as µg/mL. Feces and other tissues are µg/g wet weight.