Fluxes of the $^{238}$U series within the Dicalcium Phosphate industrial production and the biokinetical analysis of $^{210}$Pb and $^{210}$Po in broilers due to its ingestion

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Dicalcium Phosphate

- Dicalcium Phosphate is a calcium feed supplement for domestic animals (cattle, poultry, etc.).

- High calcium availability (93%).

- Produced through the rock acid digestion with either HCl or H$_2$SO$_4$ (Gäfvert et al., 2001).

- Replacement of calcium by uranium in the apatite structure

$^{238}$U > 10$^3$ Bq·kg$^{-1}$
(Burnett and Veeh., 1992)
Dicalcium Phosphate

- Previous studies shown that depending on the acid used, different radionuclides are accumulated in the final product (Casacuberta et al., 2009).

Specific concentrations $^{210}\text{Pb} \sim 2000 \text{ Bq} \cdot \text{kg}^{-1}$ and $^{210}\text{Po} \sim 800 \text{ Bq} \cdot \text{kg}^{-1}$
Radionuclide incorporation in animals

- $^{210}\text{Pb}$ and $^{210}\text{Po}$ are of special interest since its accumulation in food might pose a potential radiological dose by ingestion. $h(g) \, ^{210}\text{Pb} = 6.9 \cdot 10^{-7} \text{ Sv} \cdot \text{Bq}^{-1}$; $h(g) \, ^{210}\text{Po} = 1.2 \cdot 10^{-6} \text{ Sv} \cdot \text{Bq}^{-1}$

- Accumulation of radionuclides in animals and humans depends on:
  - the rate of intake,
  - gastrointestinal absorption, and
  - turnover in tissues.
Aims of the work

• Elucidate the fluxes of the isotopes of the $^{238}\text{U}$ decay series in the production process of DCP;

• Examine the accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in chicken tissues during its growth as a function of the type and amount of DCP in chicken diets as well as its contents of radionuclides; and

• Build a suitable kinetic model to understand the distribution of $^{210}\text{Pb}$ and $^{210}\text{Po}$ within chicken tissues after ingestion.
Materials and Methods

- Fluxes of radionuclides within the DCP production process;
  - Sampling
  - Radionuclide analysis

- Accumulation of $^{210}$Pb and $^{210}$Po in chickens;
  - Experimental set-up
  - Biokinetic model for $^{210}$Pb and $^{210}$Po in chickens.
Industrial samples

x 3 sampling dates:
- May 2007
- November 2007
- April 2008
Radionuclides analysis

- $^{238}$U, $^{234}$U, $^{230}$Th: radiochemical purification (Horwitz et al., 1992), electrodeposition and alpha spectrometry (EG&Ortec Mod. SSB 450 R).

- $^{226}$Ra: gamma spectrometry (GMX,EG&G Ortec): $^{214}$Pb (295, 351 keV) and $^{214}$Bi (609 keV).

- $^{210}$Pb, $^{210}$Po: acid digestion, deposition of $^{210}$Po in silver disks and alpha spectrometry. Ingrowth decay corrections of $^{210}$Pb and $^{210}$Po at sampling date (Masqué et al., 2002).
Accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in chickens

Diet A: blank diet (~2 Bq·kg$^{-1}$ $^{210}\text{Pb}$ and $^{210}\text{Po}$)

Diet B: 2.5% DCP* (~60 Bq·kg$^{-1}$ $^{210}\text{Pb}$ and $^{210}\text{Po}$)

Diet C: 5% DCP* (~100 Bq·kg$^{-1}$ $^{210}\text{Pb}$ and $^{210}\text{Po}$)

* DCP: 1700 Bq·kg$^{-1}$ $^{210}\text{Pb}$ and $^{210}\text{Po}$
Results and discussion

- Fluxes of radionuclides within the DCP production process.
- Accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in chickens.
- Biokinetic model for $^{210}\text{Pb}$ and $^{210}\text{Po}$ in chickens.
Fluxes of radionuclides within the DCP production process
RESULTS

Fluxes in the sludges line (kBq·h⁻¹)

![Diagram showing the sludges line process]

**Sludges line**

- **Phosphate rock** → **digestor** → **decanter 1** → **Redigestion** → **decanter 2** → **decanter 3** → **Sludge filter** → **Sludges**

**Fluxes in the sludges line (kBq·h⁻¹):**

- **S1**
  - S1_1st
  - S1_2nd
  - S1_3rd

- **S3**
  - S3_1st
  - S3_2nd
  - S3_3rd

- **S9**
  - S9_1st
  - S9_2nd
  - S9_3rd

**Isotopes:**

- $^{238}$U
- $^{230}$Th
- $^{226}$Ra
- $^{210}$Pb
- $^{210}$Po
Fluxes in DCP production line (kBq·h\(^{-1}\))

RESULTS
RESULTS

Fluxes waters and recirculation line (kBq·h⁻¹)

![Graph showing fluxes waters and recirculation line with bars for different samples S8_1st, S8_2nd, S8_3rd.]

- **238U**, **230Th**, **226Ra**, **210Pb**, and **210Po** markers are used to indicate different isotopes or elements.

**Notes:**
- S7
- Fluxes waters and recirculation line (kBq·h⁻¹)

![Diagram showing residual waters line and decanter 4 connection.]

**Diagram Details:**
- Residual waters line
- Decanter 4
- Residual water

**Legend:**
- Yellow: 238U
- Orange: 230Th
- Red: 226Ra
- Green: 210Pb
- Purple: 210Po
Radionuclide outputs

- U-238: 54%
- Th-230: 95%
- Ra-226: 61%
- Pb-210: 86%
- Po-210: 4%

Legend:
- Sludges
- DCP
- Water effluent
RESULTS

Specific concentrations of radionuclides

PHOSPHATE ROCK

SLUDGES

DCP

WATER DISCHARGES

- $^{238}$U
- $^{230}$Th
- $^{226}$Ra
- $^{210}$Pb
- $^{210}$Po

[Graphs showing specific concentrations of radionuclides in different matrices: Phosphate Rock, Sludges, DCP, and Water Discharges.]
Exemption and clearance criteria

New European Basic Safety Standards:

The exempt activity concentration values (Bq·g⁻¹) for the materials involved in the practice for Naturally Occurring Radionuclides is:

- Natural radionuclides from the U-238 series: 1 kBq·kg⁻¹
- Natural radionuclides from the Th-232 series: 1 kBq·kg⁻¹
- K-40: 10 kBq·kg⁻¹

Some elements in the decay chain, e.g. ²¹⁰Pb and ²¹⁰Po may warrant the use of values by up to two orders of magnitude.
Exemption and clearance criteria

PHOSPHATE ROCK

SLUDGES

DCP

WATER DISCHARGES

- \(^{238}\)U
- \(^{230}\)Th
- \(^{226}\)Ra
- \(^{210}\)Pb
- \(^{210}\)Po

\[ \text{Bq·kg}^{-1} \]

\[ \text{Bq·m}^{-3} \]
Accumulation of $^{210}$Pb and $^{210}$Po in chickens due to the ingestion of DCP
Accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in broilers

**RESULTS**

Accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in broilers over 21 and 42 days of feeding.
Accumulation of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in feces
Biokinetic model: first order approach

Single-compartment model: **STEADY STATE CONDITIONS**

\[
\frac{d}{dt} x(t) = -\lambda x(t) - Kx(t) + b(t)
\]

- \( x(t) \) specific activity of \(^{210}\text{Pb} \) and \(^{210}\text{Po} \) into the chicken body (whole animal);
- \( b(t) \) input of \(^{210}\text{Pb} \) and \(^{210}\text{Po} \) to the chicken;
- \( Kx(t) \) output rate of \(^{210}\text{Pb} \) and \(^{210}\text{Po} \);
- \( \lambda x(t) \) radioactive decay (also includes \(^{210}\text{Po} \) ingrowth from \(^{210}\text{Pb} \) decay)
First order model results: whole animal

Diet B, $^{210}\text{Pb}$

Diet C, $^{210}\text{Pb}$

Diet B, $^{210}\text{Po}$

Diet C, $^{210}\text{Po}$
Biokinetic model: non-linear approach

Single-compartment model: NON STATIONARY CONDITIONS

\[
\begin{aligned}
\frac{d}{dt} [x(t)p(t)] &= -\lambda x(t)p(t) - k(t)x(t)p(t) + b(t) + x(t) \frac{d}{dt} p(t) \\
K(t) &= k_x \frac{F(t)}{p(t)} \overline{p} = k_x N(t)
\end{aligned}
\]

- \(F(t)\) food weight
- \(p(t)\) animal weight
- \(k_x\) transfer rate at stationary state
- \(F\) food weight at stationary state
- \(\overline{p}\) animal weight at stationary state
RESULTS

Diet B, $^{210}$Pb and $^{210}$Po

$K (^{210}\text{Pb}) = 3.20 \pm 0.41$

$K (^{210}\text{Po}) = 1.26 \pm 0.06$

$K = [\text{d}^{-1}]$

Diet C, $^{210}$Pb and $^{210}$Po

$K (^{210}\text{Pb}) = 4.65 \pm 0.61$

$K (^{210}\text{Po}) = 1.84 \pm 0.25$
Conclusions: fluxes of radionuclides in DCP industrial process

- About $30 \cdot 10^3$ kBq·h$^{-1}$ of $^{238}$U, $^{230}$Th, $^{226}$Ra, $^{210}$Pb and $^{210}$Po enter the production system.
  - $^{238}$U out-fluxes are divided between sludges and DCP.
  - $^{230}$Th and $^{210}$Po are discharged in sludges.
  - $^{226}$Ra is mainly eluted through water effluents.

- Limits of radionuclides established in the new BSS are 1 kBq·kg$^{-1}$. DCP industries are not exempted.
  - > $10^3$ Bq·kg$^{-1}$ of $^{238}$U
  - > $10^4$ Bq·kg$^{-1}$ of $^{230}$Th and $^{210}$Po
  - > $2 \cdot 10^3$ Bq·kg$^{-1}$ of $^{210}$Pb
  - $^{226}$Ra?
Conclusions: $^{210}\text{Pb}$ and $^{210}\text{Po}$ in chickens

- $^{210}\text{Pb}$ and $^{210}\text{Po}$ are accumulated in chicken tissues proportional to the initial contents in diets.
  - $^{210}\text{Pb}$ accumulates in bones
  - $^{210}\text{Po}$ accumulates in liver and kidneys
  - Accumulation is small compared to the amounts excreted.

- First order kinetic approach model would not fit the experimental data due to the fact that the model does not take into account the growing conditions of the organism.

- A model based on a non-stationary based function is capable to model the experimental results when growing conditions occur. Allows calculation of transfer rates $k$ useful for first-order models if extrapolating $k(t)$ when $t$ tends to a steady state.
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