

## <sup>99</sup>Tc transfer from water to monogastric

D. Berthol<sup>1,2</sup>, C. Feidt<sup>1</sup> and J. Brun Bellut<sup>1</sup>

<sup>1</sup> Laboratoire de Sciences Animales, INRA-INPL-UHP, ENSAIA,  
BP. 172, 54505 Vandœuvre-lès-Nancy, France

<sup>2</sup> EDF-CNPE de Cattenom, BP. 41, 57570 Cattenom, France

**Abstract.** Technetium-99 ( $T_{1/2}$ : 212 000 years) is derived from the fission of uranium-235 and produced more particularly by nuclear fuel reprocessing plants. We studied, during a chronic contamination by water, excretion, distribution and retention of <sup>99</sup>Tc in rats as monogastric model.

More than 85% of ingested <sup>99</sup>Tc is excreted by urine and feces, urinary way being dominant from the first week, to reach 72% of total excretion at the end of the experiment (14 weeks).

The thyroid and hair content increased until the end of the treatment. They represented respectively 0.1 and 16% of the given dose, after 98 days of intake. The liver and the kidneys content reached a plateau between the 40<sup>th</sup> and the 56<sup>th</sup> days of the exposure period. There was no <sup>99</sup>Tc in the muscle and the intestine.

## 1 INTRODUCTION

<sup>99</sup>Tc is a radioelement which contributes in a important way to the radiotoxicity with long-lived waste. It is a beta emitter ( $E_{max}$  = 295 keV) with a half-life of 212 000 years. Formed during the fission of <sup>235</sup>U, its presence in the environment results mainly from anthropological activities: nuclear shots aeriels, medical use of the <sup>99m</sup>Tc, cycle of the fuel. Information on <sup>99</sup>Tc transfer during long-term exposure at low dose is scanty. This work was, therefore, performed to study retention and excretion of <sup>99</sup>Tc in rats given orally low amounts of <sup>99</sup>Tc for a long period, and during the post-exposure period.

## 2 MATERIALS AND METHODS

This experiment was performed with 35 male Wistar-strain rats (~100 g). They were all kept in individual cages with food and water *ad libitum*.

Rats were separated into 2 groups:

- Animals of the first group (18 rats) were used to study chronic exposure over 98 days. So, until day 98, they were given 5 Bq.g<sup>-1</sup> of ammonium pertechnetate obtained from Dupont<sup>1</sup>, by an intra-esophageal canula. They were weighed every week and the dose was corrected again.
- Animals of the second group (13 rats) were used to study post-exposure period. Consequently, they received NH<sub>4</sub>TcO<sub>4</sub> (5 Bq.g<sup>-1</sup>) during only 56 days, and the intakes were stopped afterwards.

For each group, the urine and the feces of 4 animals kept in metabolism cages were collected daily from D<sub>0</sub> to D<sub>98</sub>.

<sup>1</sup> Dupont NEN Life Science Products France S.A., Paris, France.

Rats were anesthetized with chloroform and then sacrificed by cervical dislocation at different times:

- at D<sub>0</sub>, D<sub>28</sub> and D<sub>56</sub>, 3 rats from the first group
- at D<sub>63</sub>, D<sub>70</sub> and D<sub>84</sub>, 2\*3 rats from each group
- at D<sub>98</sub>, 2\*4 rats in the metabolism cages, from each group

Liver, kidneys, intestine, muscle, thyroid and hair were sampled for measurement of <sup>99</sup>Tc content by liquid scintillation, in a Packard model Tri-Carb 1500 beta counter. At D<sub>98</sub>, only 3 rats from each group were used for tissues analysis.

Results were compared by regression and variance analysis using STAT-ITCF software.

### 3 RESULTS

#### 3.1. Excretion of <sup>99</sup>Tc

##### 3.1.1. Exposure period

Over the whole experimental period, we observed an increase of <sup>99</sup>Tc content with time, in the urine and the feces. This increase may be explained by this of the intake by animals, until day 56. But after, the dose was constant, as animals didn't gain longer weight. Excretion in the urine was greater than in the feces. The maximal values were 14.2 kBq at D<sub>98</sub> for the urine, and 2.8 kBq for the feces.

The distribution of cumulative excretion in urine and feces combined, showed that during the first week, rats excreted about 44.5% of the weekly dose in the urine and 30.2% in the feces (figure 1). But, during the experimental period, we observed an increase of the relative part excreted in the urine and consequently, a decrease of relative part excreted in the feces. After 14 weeks, the feces represented only 16% of the relative excretion, and the urine, 81%.

At the end of experiment, the total activity excreted represent 72% of the given dose in the urine, and 15% in the feces.

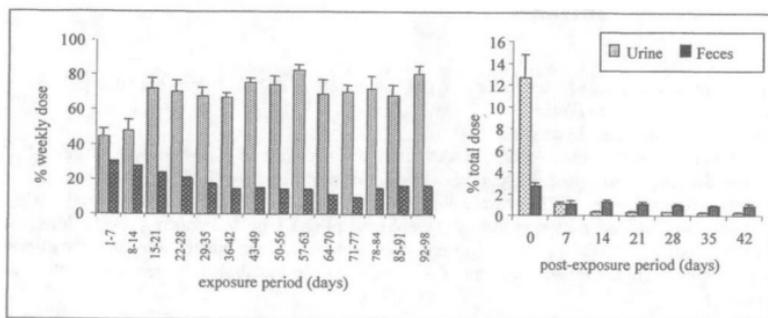


Figure 1: Excretion of <sup>99</sup>Tc in the urine and feces

### 3.1.2. Post-exposure period

We observed a decrease of the urinary concentrations of more than 91 %, from the first week following the stopping of the intakes. After 3 weeks, the urine activities were very low (0.2 kBq) and represented only 0.2% of the total given dose (figure 1).

The fecal activities, decreased slowly (less than 65% in one week) and reached an average level of 0.7 kBq maintained until the end of experiment which represents almost 0.9% of the dose ingested over the 56 days.

## 3.2. Distribution of $^{99}\text{Tc}$ in tissues

### 3.2.1. Continuous intakes

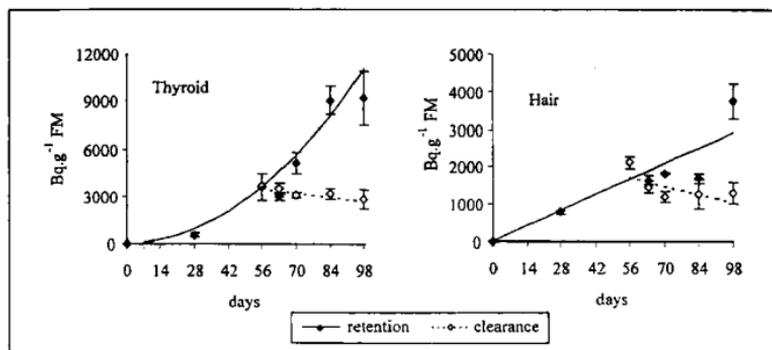


Figure 2: Kinetics of  $^{99}\text{Tc}$  in the thyroid and the hair

In both thyroid and hair, the concentrations increased until the 98<sup>th</sup> day (figure 2) to maximal values of 9184 Bq.g<sup>-1</sup> FM for the thyroid and 3730 Bq.g<sup>-1</sup> FM for the hair. At the end of the experimental period, the quantities of  $^{99}\text{Tc}$  in the thyroid and the hair represented respectively 0.1% and 15.6% of the total given dose.

The kidneys and the liver displayed lower  $^{99}\text{Tc}$  concentrations. In these tissues, it seems that, at 6-7 weeks,  $^{99}\text{Tc}$  content stabilized around a maximal value between 150 and 200 Bq.g<sup>-1</sup> FM for the kidneys and between 5 and 15 Bq.g<sup>-1</sup> FM for the liver (figure 3). At D<sub>98</sub>, the percentage of the total administered dose was 0.3% in rats' kidneys and 0.1% in the liver.

$^{99}\text{Tc}$  was found neither in the intestine nor in the muscle.

### 3.2.2. Post-exposure period

There was no significant loss of  $^{99}\text{Tc}$  from the rats' thyroid or hair over the 6 weeks following the interruption of the intakes (figure 2).

The concentrations of  $^{99}\text{Tc}$  in the kidneys and the liver decreased by approximately 52 and 42%, respectively, between the first and 7<sup>th</sup> days after the end of the intake (figure 3). By 28 days, the  $^{99}\text{Tc}$  content in the liver had fallen near the detection limit. The clearance was significantly slower for the kidneys.

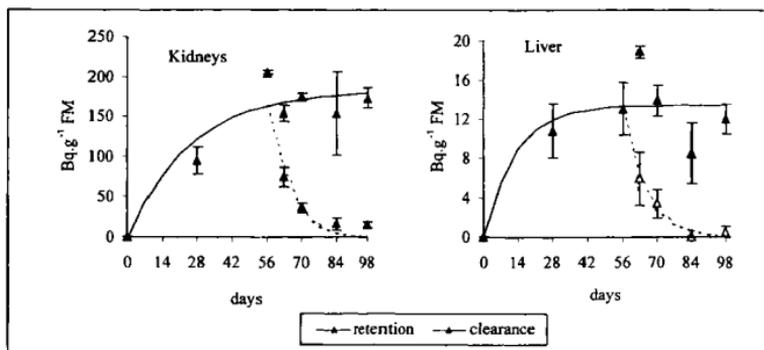


Figure 3: Kinetics of  $^{99}\text{Tc}$  in the kidneys and liver

#### 4 DISCUSSION

In the present study, the cumulative exposure showed on one hand, a decrease of the fecal relative excretion and on the other hand, an increase of both urinary relative excretion and uptake by the tissues. This better apparent digestibility may be due to the adaptation of the gastrointestinal tract to  $^{99}\text{Tc}$  during the first weeks of intake.

We also showed the presence of  $^{99}\text{Tc}$  in the feces during the post-exposure period. This endogenous  $^{99}\text{Tc}$  could be explained by the recycling of  $^{99}\text{Tc}$  in the digestive tract (by the saliva, the bile or the hair ingested).

We observed different retention and elimination kinetics for  $^{99}\text{Tc}$  according to tissues.

- These results seem to indicate that the muscles and intestine don't retain the  $^{99}\text{Tc}$ . However, Jones [1] showed Tc's presence in the muscle and Archimbaud *et al.* [2] also put in evidence Tc's uptake by the gastrointestinal tract. It would be possible that these tissues may not retain Tc at low doses, even during chronic exposure. We can also think that the activity of these tissues is too low to be measured.
- $^{99}\text{Tc}$ 's presence in the kidneys and the liver is doubtless due to their purifier role. They do not seem to fix Tc in a specific way. We could rather think that they play the role of filter as for others xenobiotics or organic poisons (cadmium, arsenic, lead, ...).
- Cataldo *et al.* [3] had put in evidence Tc's incorporation in sulfurated amino acids into vegetables. The fixation of Tc in hairs could be due to the same phenomenon. In the thyroid, Tc seems also bound in a specific way because the stopping of the supplies does not induce a significant decrease of the concentrations. These results agree with those of Garten *et al.* [4] and Van Bruwaene *et al.* [5] who not observed elimination of Tc in the thyroid, in rats and sheep.

The transfer factor is defined as the ratio between Tc content in the tissues at equilibrium and the daily activity intake. However, some authors calculate it, with more or less similar definitions, during experiment realized in single exposure, even if they can't reach equilibrium conditions. We obtained transfer factors of 0.05 for kidneys, and 0.07 for the liver. This result is close to those to Gerber *et al.* [6] and Jones [1] from which we calculated respective transfer factors of 0.07 and 0.05 for the kidneys, and 0.08 and 0.03 for the liver. On the other hand, if we are interested in tissues where the balance is not reached, we can give as indication a "temporary" transfer factor of 4.27 for the thyroid and 1.38 for the

hair at day 98. The single exposure underestimates the real uptake of Tc by these tissues. Indeed, Gerber *et al.* [6] obtains transfer factors of only 1.4 and 0.1 for the thyroid and the hair, respectively.

The differences between the tissues may be linked with the form of  $^{99}\text{Tc}$ . Additional studies on speciation of  $^{99}\text{Tc}$  (free or linked, reduced or oxidized) within these tissues may allow answering this question. In addition, more than 85% of the  $^{99}\text{Tc}$  administered is excreted, and the animal remains likely to reject additional quantities 42 days after stopping the intake. It seems therefore interesting to determine its form in the urine and the feces, possible paths for the return of  $^{99}\text{Tc}$  into the environment.

### Acknowledgments

This work was supported by a grant from EDF.

### References

- [1] Jones B., *Health Phys.* **57** (2) (1989) 331-336.
- [2] Archimbaud Y., Poncy J.L., Masse R., Cogneau M., *Radioprotection.* **27** (1) (1992) 35-41.
- [3] Cataldo D.A., Garland T.R., Wildung R.E., Fellows R.J., *Health Phys.* **57** (2) (1989) 281-287.
- [4] Garten C.T. Jr, Myttenaere C., Vandecasteele C.M., Kirchmann R., Van Bruwaene R., Technetium in the environment (Elsevier Applied Science Publishers, 1986) pp.319-332.
- [5] Van Bruwaene R., Hegela M., Gerber G.B., Kirchmann R., Maisin J.R., Technetium in the environment (Elsevier Applied Science Publishers, 1986) pp.391-395
- [6] Gerber G.B., Van Hess M., Garten C.T. Jr., Vandecasteele C.M., Vankerlom J., Van Bruwaene R., Kirchmann R., Colard J., Cogneau M., *Health Phys.* **57** (2) (1989) 315-319.