TECHNETIUM-99M: FROM NUCLEAR MEDICINE APPLICATIONS TO FINE SEDIMENT TRANSPORT STUDIES

Jefferson V. Bandeira¹ & Lécio H. Salim¹

Nuclear Technology Development Center (CNEN/CDTN) – Belo Horizonte, Brazil
Objective

To rescue the history of the development of the application of $^{99m}$Tc, widely used in Nuclear Medicine, to its use as a tracer for studying the transport of fine sediment in suspension, in water environment.
Background

IAEA TC Project URU/8/009 – 1997

Study of the environmental quality of Montevideo Bay - Sedimentological studies with the application of nuclear techniques.
Background

IAEA TC Project URU/8/009 – 1997

• It was scheduled to label mud with $^{198}\text{Au}$ imported from Brazil

• $^{198}\text{Au}$ did not arrive and was substituted by $^{99m}\text{Tc}$ ($T_{1/2} = 6.02h$), eluted from Mo/Tc generator as pertechnetate ($\text{TcO}_4^-$), obtained from the Center of Nuclear Medicine - University of the Republic of Uruguay (UdelaR).

• Quick labeling tests for the reduction of $\text{TcO}_4^-$ with the reductants:
  
  stannous chloride ($\text{SnCl}_2\cdot\text{2H}_2\text{O}$)

  sodium thiosulfate ($\text{Na}_2\text{S}_2\text{O}_3\cdot\text{5H}_2\text{O}$)
Background

Montevideo Harbour

Sub-superficial injection of mud labelled with $^{99m}$Tc
After this first use, and considering the favourable characteristics of $^{99m}$Tc:

- Convenient half-life = 6.02 h for studies of transport of fine sediment in suspension;

- Low gamma ray energy (140 keV) $\rightarrow$ moderate shielding and ease of manipulation in field conditions;

- Availability even in places where there are no nuclear facilities $\rightarrow$ widespread application in Nuclear Medicine;

- Convenient half-life of the $^{99}$Mo parent nuclide (66 h) $\rightarrow$ possible to obtain $^{99m}$Tc for one week or more in field studies.

The feasibility of its use for labeling mud was studied: IAEA Research Contract BRA-10891.
Lab labeling results – CDTN/Brazil

Effect of reducing agent nature for the sorption of $^{99m}$Tc in fine sediments.

Variation of sorption of $^{99m}$Tc in 100 mg of sediment with mass of SnCl$_2$.2H$_2$O

Labeling yield as function of different factors
Lab labeling results – CDTN/Brazil

Effect of pH in the sorption of $^{99m}$Tc

Labeling yield as function of different factors

Influence of the concentration in the sorption of $^{99m}$Tc by fine sediment
Lab labeling results – CDTN/Brazil

Influence of contact time in the sorption of $^{99m}$Tc by fine sediment

Labeling yield as function of different factors

Desorption of $^{99m}$Tc in river water for different sediment concentrations
Lab labeling results – CDTN/Brazil

Hydrodynamic behavior of labeled and not labeled sediment.
Sedimentation tests – Stokes law.

Andreasen pipette (1) & test tube (2)

Natural sediment in distilled water

Labelled (but not floculated) sediment using minimum quantities of SnCl2 and HCl to reduce the Tc (VII)
Pampulha reservoir dredging – Belo Horizonte, Minas Gerais State, Brazil (1999-2004)
Pampulha studies: Injection points (PI) & detection points (D)
Pampulha tracer studies: injection of mud with $^{99m}$Tc

Onça Creek

Double labelling: sediment with $^{99m}$Tc & water with Rhodamine WT

Injection Point: PI-2

Detection station: D3

ISCO Sampler

Upstream view

Downstream view
Pampulha tracer studies: Eulerian Detection

Onça Creek

Station D4
Pampulha studies: Some results

Detection & mathematical model curves

Onça creek (Jul./03/2001)

Rhodamine WT stretch (D3-D5)
\( v = 0.282 \text{m/s}; D = 9.4 \text{m}^2/\text{s} \)

99mTc stretch (D3-D5)
\( v = 0.290 \text{m/s}; D = 11.4 \text{m}^2/\text{s} \)
Pampulha studies: Some results

Mathematical model used – (Sing & Beck, 2003)

\[
\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2} - v \frac{\partial c}{\partial x}
\]

One-dimensional dispersion equation (1)

\[
c(x,t) = \frac{M}{A\sqrt{4Dt\pi}} \exp\left(-\frac{(x-vt)^2}{4Dt}\right)
\]

A solution of (1) for point and instantaneous injection → Taylor equation (2)

\[
\frac{c}{c_0} = \frac{1}{2} \text{erfc}\left(\frac{x-vt}{2\sqrt{Dt}}\right)
\]

Sing & Beck equation (3)

Equation (3) corresponds to equation (2) for large values of \(v x / D\) and a point and instantaneous injection (SINGH & Beck, 2003), as is the case of the experiments performed in the studied creeks and rivers, in the region downstream of Pampulha reservoir.
<table>
<thead>
<tr>
<th>Location</th>
<th>Date</th>
<th>Trecho</th>
<th>Trecho</th>
<th>Trecho</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ribeirões Pampulha e Onça</td>
<td>27/09/2000</td>
<td>Trecho D1-D2</td>
<td>Trecho D1-D3</td>
<td>Trecho D2-D3</td>
</tr>
<tr>
<td>Água (Rodamina WT)</td>
<td>$v = 0,441m/s$</td>
<td>$v = 0,741m/s$</td>
<td>$v = 0,936m/s$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 9,2m^2/s$</td>
<td>$D = 15,1m^2/s$</td>
<td>$D = 5,5m^2/s$</td>
<td></td>
</tr>
<tr>
<td>Sedimento fino ($^{99m}$Tc)</td>
<td>$v = 0,714 m/s$</td>
<td>$v = 0,741 m/s$</td>
<td>$v = 0,936 m/s$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 8,8m^2/s$</td>
<td>$D = 15,1m^2/s$</td>
<td>$D = 5,5m^2/s$</td>
<td></td>
</tr>
<tr>
<td>Ribeirão da Onça</td>
<td>03/10/2000</td>
<td>Trecho D3-D4</td>
<td>Trecho D4-D6</td>
<td></td>
</tr>
<tr>
<td>Água (Rodamina WT)</td>
<td>$v = 0,379m/s$</td>
<td>$v = 0,344m/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 11,2m^2/s$</td>
<td>$D = 10,2m^2/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sedimento fino ($^{99m}$Tc)</td>
<td>$v = 0,386m/s$</td>
<td>$v = 0,326m/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 11,9m^2/s$</td>
<td>$D = 54,0m^2/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rio das Velhas</td>
<td>26/06/2001</td>
<td>Trecho D7-D8</td>
<td>Trecho D8-D9</td>
<td></td>
</tr>
<tr>
<td>Água (Rodamina WT)</td>
<td>$v = 0,550m/s$</td>
<td>$v = 0,480m/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 16,2m^2/s$</td>
<td>$D = 14,6 m^2/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sedimento fino ($^{99m}$Tc)</td>
<td>$v = 0,541m/s$</td>
<td>$v = 0,525m/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>$D = 13,9m^2/s$</td>
<td>$D = 24,8m^2/s$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ribeirão da Onça</td>
<td>03/07/2001</td>
<td>Trecho D3-D4</td>
<td>Trecho D3-D5</td>
<td>Trecho D4-D5</td>
</tr>
<tr>
<td>Água (Rodamina WT)</td>
<td>$v = 0,282m/s$</td>
<td>$v = 0,264m/s$</td>
<td>$v = 0,290m/s$</td>
<td>$v = 0,300m/s$</td>
</tr>
<tr>
<td></td>
<td>$D = 9,4m^2/s$</td>
<td>$D = 7,3m^2/s$</td>
<td>$D = 11,4m^2/s$</td>
<td>$D = 21,5m^2/s$</td>
</tr>
<tr>
<td>Sedimento fino ($^{99m}$Tc)</td>
<td>$v = 0,314m/s$</td>
<td>$v = 0,290m/s$</td>
<td>$v = 0,264m/s$</td>
<td>$v = 0,300m/s$</td>
</tr>
<tr>
<td></td>
<td>$D = 12,8m^2/s$</td>
<td>$D = 11,4m^2/s$</td>
<td>$D = 7,3m^2/s$</td>
<td>$D = 21,5m^2/s$</td>
</tr>
</tbody>
</table>
Pampulha studies: Some results

Temporal variation of sediment concentration, downstream Pampulha dam, as a function of dredging dumping at D1, with 9 hours duration, obtained with a convolution model calibrated with the results of experiments of fine sediment labelled with $^{99m}$Tc
Pampulha studies

The above studies made it possible to calculate physical environmental impacts:

- increase of sediment concentration in the watercourses due to the dumping of dredged sediment downstream the Pampulha reservoir dam;

- the possibility of deposition of the dumped sediment.

The measurement of physical-chemical parameters of the water (DO; pH; Eh; conductivity & temperature) allowed to evaluate the possibility of desorption of the metals adsorbed in the sediment to be dredged, and dumped downstream.

As for the impacts studied, it was concluded that there was no impediment to the dumping of fine material in the water courses downstream from Pampulha reservoir (Bandeira, 2004).
Labelling & injection boat

Detection boat

Orinoco River - Labelling & injection of mud with $^{99m}$Tc

Lead shielding (3 mm)

Injection tube
Detection orientation floats together with injection
2.5 Orinoco River – Venezuela (2005-2006) - VEN/8/019

1st injection: sediment in suspension with $^{99m}\text{Tc}$

Detection lines

2\textsuperscript{st} injection: sediment in suspension with $^{99m}\text{Tc}$

Detection lines

Synthesis

Results of suspended sediment studies

<table>
<thead>
<tr>
<th>Injection</th>
<th>Probe</th>
<th>$S_R$ (g/ton/s)</th>
<th>$D_L$ (m²/s)</th>
<th>$D_T$ (m²/s)</th>
<th>$D$ (m/s)</th>
<th>$U$ (m/s)</th>
<th>$T_{half}$ (min)</th>
<th>$L_{half}$ (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1 (1.5 m)</td>
<td>520</td>
<td>1.373</td>
<td>0.495</td>
<td>500</td>
<td>0.723</td>
<td>22.2</td>
<td>963</td>
</tr>
<tr>
<td>1</td>
<td>2 (0.5 m)</td>
<td>532</td>
<td>1.081</td>
<td>0.450</td>
<td></td>
<td>0.720</td>
<td>21.7</td>
<td>937</td>
</tr>
<tr>
<td>2</td>
<td>1 (1.5 m)</td>
<td>372</td>
<td>-</td>
<td>0.278</td>
<td>30</td>
<td>0.609</td>
<td>31.1</td>
<td>1136</td>
</tr>
<tr>
<td>2</td>
<td>2 (0.5 m)</td>
<td>421</td>
<td>-</td>
<td>0.265</td>
<td></td>
<td>0.609</td>
<td>27.4</td>
<td>1001</td>
</tr>
</tbody>
</table>

$S_R$ - Sedimentation rate

$D_L$ - longitudinal dispersion coefficients for time $T_{half}$.

$D_T$ - transversal dispersion coefficients for time $T_{half}$.

$D$ - Dilution

$U$ - Advection velocity

$T_{half}$ - the necessary time for half of the suspended sediment dumped to settle

$L_{half}$ - distance travelled from the dumping site
Orinoco River studies

The results obtained for the behavior of the natural sediment in suspension, in the end of the low water season of the Orinoco River (April), could be used for preliminary designs of outfalls for industrial effluents which discharge particulate material with the density of fine sediment or for pollutant material that could be adsorbed by the fine sediment.
Environmental impact due to fine sediment originated from bottom discharge of Small Hydroelectric Power Plant (SHP)

Paciência SHP – Paraibuna River, MG - Brazil

**Study goals**

**A**- Evaluating, using tracers and related techniques, the physical environmental impact: advection, dispersion and sedimentation rate of the dumping in downstream waterways, of the material removed from the reservoir through bottom discharge and / or dredging;

**B**- Assess the environmental impact on its biological aspects, using as bioindicator the zoobenthos aquatic community existing in the Paraibuna River, downstream the reservoir;

**C**- Perform the physical and chemical characterization of sediments deposited in the reservoir, which is periodically removed through bottom discharge;

**D**- Establish a basic methodology considering hydrosedimentological, chemical and biological aspects together, to be applied, adapted to each case, for other SHP.
Key Map
Area under study: EMF (physical) and EMB (biological) monitoring stations.
Site characterization

Upstream view from the dam during bottom discharge

Paciencia SHP dam
Hydro sedimentological work

- **Sampling of bottom sediments from the reservoir and downstream region, just before and just after bottom discharge**, to evaluate the influence of bottom discharge in the sediment distribution.

- **Two bathymetric surveys were performed in the backwater region of the dam, just before and just after bottom discharge**, to quantify: the accreted volume and the sediment transferred downstream due to bottom discharge.
Hydro sedimentological work

• Experiments with tracers to assess the parameters of transport of fine sediment in suspension: advection, dispersion and sedimentation rate, arising from dredging / bottom discharge;

• Mathematical modeling, with the above parameters measured, to simulate the bottom discharge and assess physical environmental impacts arising from it.
Tracer studies

Equipment for tracer injection

Scintillation detectors operating

Tracers injection

Analog and digital meters
Tracer studies

Examples of measured and modelled results (Field experiments – August 2012)
Tracer studies

Paciência SHP and the study region downstream, with monitoring stations.
Tracer studies

Experiments of August 2012

<table>
<thead>
<tr>
<th>Stretch/Day</th>
<th>08/21/2012</th>
<th>08/22/2012</th>
<th>08/23/2012</th>
</tr>
</thead>
<tbody>
<tr>
<td>EMB2 – EMB4</td>
<td>$v = 0.39 \text{ m/s}$</td>
<td>$v = 0.38 \text{ m/s}$</td>
<td>$v = 0.38 \text{ m/s}$</td>
</tr>
<tr>
<td></td>
<td>$D = 9.40 \text{ m}^2/\text{s}$</td>
<td>$D = 9.85 \text{ m}^2/\text{s}$</td>
<td>$D = 9.00 \text{ m}^2/\text{s}$</td>
</tr>
</tbody>
</table>

Experiments of October 2012

<table>
<thead>
<tr>
<th>Stretch/Day</th>
<th>24/10/2012</th>
<th>25/10/2012</th>
</tr>
</thead>
<tbody>
<tr>
<td>Power house – EMB1</td>
<td>$v = 0.21 \text{ m/s}$</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>$D = 5.7 \text{ m}^2/\text{s}$</td>
<td>--</td>
</tr>
<tr>
<td>EMB1 – EMB4</td>
<td>$v = 0.36 \text{ m/s}$</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>$D = 13.3 \text{ m}^2/\text{s}$</td>
<td>--</td>
</tr>
<tr>
<td>Power house – EMB2</td>
<td>--</td>
<td>$v = 0.33 \text{ m/s}$</td>
</tr>
<tr>
<td></td>
<td>--</td>
<td>$D = 5.7 \text{ m}^2/\text{s}$</td>
</tr>
<tr>
<td>EMB2 – EMB4</td>
<td>--</td>
<td>$v = 0.33 \text{ m/s}$</td>
</tr>
<tr>
<td></td>
<td>--</td>
<td>$D = 13.0 \text{ m}^2/\text{s}$</td>
</tr>
</tbody>
</table>

Values of advection velocity ($v$) and dispersion coefficient ($D$) obtained with the adjustment of Singh & Beck model (2003) to field data.
Temporal representation of the concentration of sediment in suspension due to bottom discharge lasting for 4.5 days, considering a unit concentration in the release point. It is the simulation, obtained by convolution, of the continuous bottom discharge, using average values of $v$ and $D$ from the experiments.
Main results - Paciência SHP

The attenuation of the concentration of fine sediment in suspension, arising from a bottom discharge, in the Paciência SHP, was calculated up to the mouth of Preto river (18% of the initial concentration), about 34 km downstream. A mathematical model calibrated with the results of tracer studies was used for this calculation.

This information can be used by CEMIG (Electric Utility Company) as a tool in the management of bottom discharge, associated with the currently employed controls, based on the level of DO <3.
Main conclusions

• The above mentioned case studies confirm that labeling of fine sediment with $^{99m}$Tc can be successfully accomplished and that the labelled sediment has the same hydrodynamic behavior as the natural one.

• The application of this radioactive tracer, widely used in Nuclear Medicine, became available in the field of Sedimentology for which fine sediments are instrumental in the transportation of heavy metals and other pollutants in the water environment.

• The work performed in the laboratory research and in some field applications used second week generators obtained, at no cost, from Nuclear Medicine laboratories. The reason is that the $^{99m}$Tc detector for environmental applications is placed into the water, in $4\pi$ geometry and for medical applications it is situated externally to the patient. So, the necessary activity concentrations for environmental use (Bq/mL of water) are much lower ($10^{-7}$) than in Nuclear Medicine utilization (Bq/mL of blood).
Acknowledgements

• IAEA (Research Contract Nr. 10891) and CDTN/CNEN-Brazil;
• Edmundo Garcia Agudo (IAEA-Retired), Technical Officer of TC Project URU/8/009;
• The researchers and collaborators from Uruguay, who participated in the IAEA TC Project URU/8/009, outstandingly the Pharmaceutical Chemist Beatriz Souto Ameigenda, for her original idea of trying to replace the scheduled labelling of fine sediment with $^{198}\text{Au}$, by the labeling with $^{99m}\text{Tc}$;
• Authors would like to thank their colleagues of CDTN involved in the development of the technique and in the field works performed;
• ECOGRAF and ECOAR Nuclear Medicine laboratories (Dr. Ivana Sena Nascimento), from Belo Horizonte, Brazil, for supplying the $^{99m}\text{Tc}$ used in the laboratory research and for part of the field work performed in Pampulha studies (second week generators);
• Special thanks to Dr. Patrick Brisset (IAEA), for his ever-present role in disseminating the use of tracers and nuclear techniques in the study of sediment transport and dispersion of pollutants in water environment.
Thanks for your attention