HelmholtzZentrum münchen
German Research Center for Environmental Health

Transaqua AP 3.1:
Untersuchungen zur Biokinetik inkorporierter Radionuklide aus aquatischen Ökosystemen zur verbesserten Dosisabschätzung

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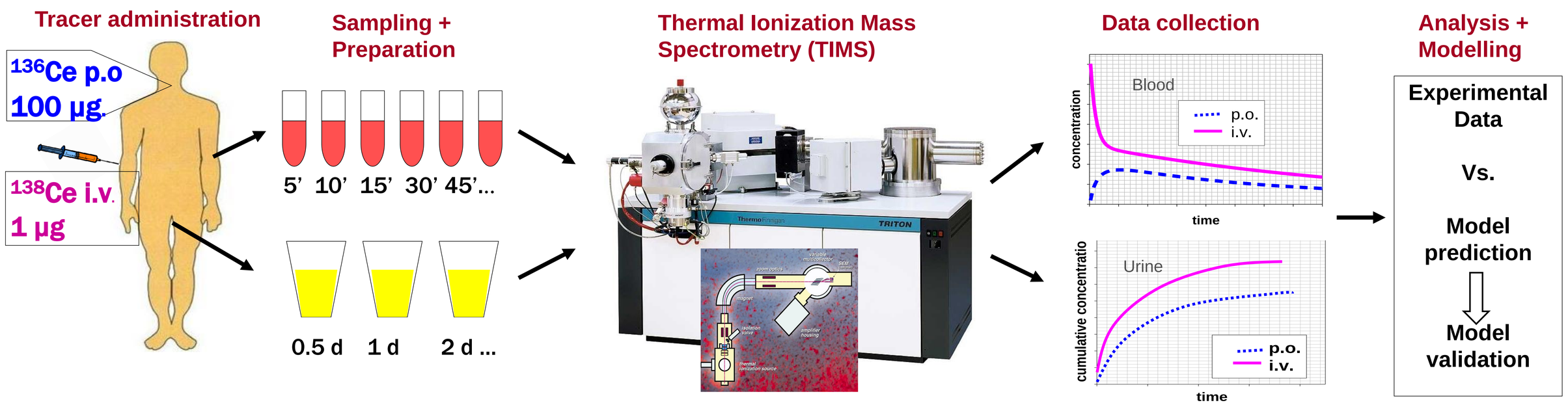
Background

Biokinetic models are used to assess internal radiation doses. In order to create such models, the **biokinetic behaviour of radionuclides** in the human metabolism must be known. This can be achieved by estimation from animal experiments or ideally from direct measurements on humans.
Based on the assumption that isotopes behave chemically similar, it is possible to use stable isotopes as tracers in order to collect data from human volunteers, without exposing them to ionizing radiation. Tracer solutions are **administered orally and intravenously to volunteers** (see figure below). The measured isotopic distribution in these samples provides information about the absorbed fraction from the gut, clearance from plasma, and urinary excretion from the body. These are the most important characteristic data needed **to validate or modify the existing basic ICRP biokinetic model of cerium.**

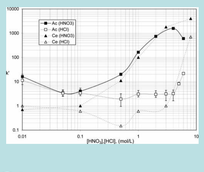


Challenges for measurement technique

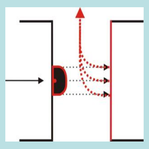
1. Measurements in (sub)nanogram range 2. Separation of Ce-isotopes from organics and other metals 3. Controllable sources of interference (Ba)

Sample preparation

a) Addition of internal standard (Ce-142) to biological sample
b) Microwave-powered acid digestion
c) Separation of cerium isotopes by extraction chromatography
e.g. RE resin, Pb resin, Sr resin, MnO_2 resin, **DGA resin**



EVA IONI

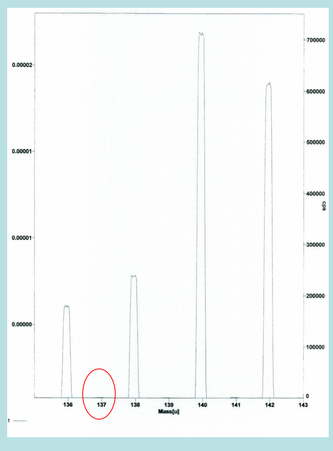


d) Transfer to tantalum double filaments (EVA, IONI)
f) TIMS measurement with single electron multipliers on masses 136, 138, 140, 142 (all cerium), and 137 (for barium isobaric correction)

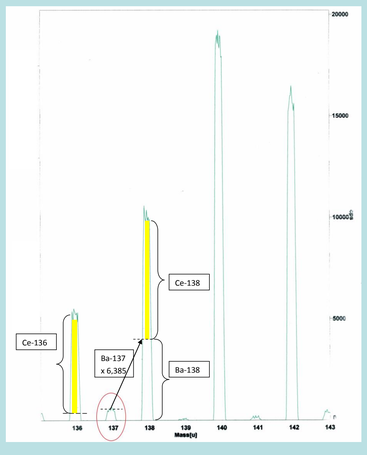
Results: Measurement signals during mass scans

Test solutions of barium (50 ng) and cerium (10 µg -10 ng) using different resins:

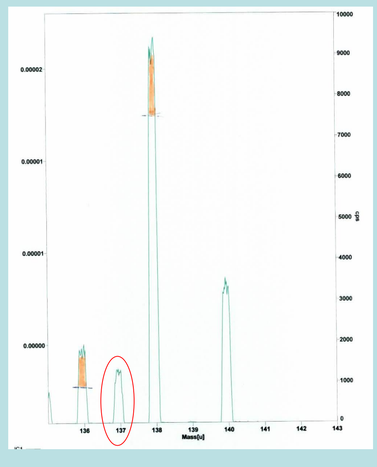
a) **DGA** resin
10 µg Ce-isotopes



b) **DGA** resin
10 ng Ce-isotopes



c) **Pb** resin
10 ng Ce-isotopes



Isotopic composition of used Ba- and Ce-solutions, in (atom%)

	136	137	138	139	140	141	142
Ba	7.854	11.232	71.698				
Ce	0.19		0.25		88.45		11.11
Ce-136	30.6		0.7		64.2		4.5
Ce-138	0.04		41.6		55.81		2.55
Ce-142	0.02		0.1		6		93.88

Barium correction

$$\left(\frac{^{138}Ce}{^{140}Ce}\right)_{corr} = \left(\frac{138}{140}\right)_m - \left(\frac{137}{140}\right)_m * 6.385\,;$$

$$\left(\frac{^{136}Ce}{^{140}Ce}\right)_{corr} = \left(\frac{136}{140}\right)_m - \left(\frac{137}{140}\right)_m * 0.6994\,;$$

„stepwise” TIMS measurement

- Pre-heating (evaporation) at 800 mA current for 16 h to release barium from sample
- Measurement at constant ionization at 4800 mA current and stepwise increase of evaporation to 1600 mA and to 2400 mA
- Monitoring of barium signal till its decrease
- Actual start of sample measurement on masses 136, 137, 138, 140, 142 to determine Ce isotope ratios

Results: Recovery experiments

Test Urine (10ml)	recovery Ce-136 (ng)	Ce-136 spiked (ng)	recovery Ce-138 (ng)	Ce-138 spiked (ng)
1	0.491	0.5	0.807	0.2
2	0.489	0.5	2.374	0.2
3	0.480	0.5	-0.190	0.2
4	0.463	0.5	-1.355	0.2

Results: Human study

Human study p.o. Ce-136	Ce-136 (ng)	Ce-138 (ng)
Urine 1 (10ml)	0.022	-0.014
Urine 2	0.470	6.862
Urine 3	0.173	1.000
Urine 4	0.175	1.949
Urine 5	0.586	4.456

Interference reductions on masses 136 und 138 were insufficient. Used method is not sensitive enough to measure Ce-isotopes in nanogram range without interferences due to barium.
Reliable, reproducible determinations of Ce-isotopes in these biological samples were not possible.

TransAqua
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