



LUND
UNIVERSITY

Sensitivity analysis of the ICRP biokinetic model predicting the activity of Gd in lungs and 24-hour excretion samples.

BELIKSE RAMLJAK, ALEXANDR MALUSEK, CHRISTOPHER RÄÄF, KRISTINA STENSTRÖM



Summary of the project

Help predict, monitor, and follow-up the internal doses from accidentally release particles from tungsten target area of the ESS to workers and to members of the public.



Strål
säkerhets
myndigheten

Swedish Radiation Safety Authority

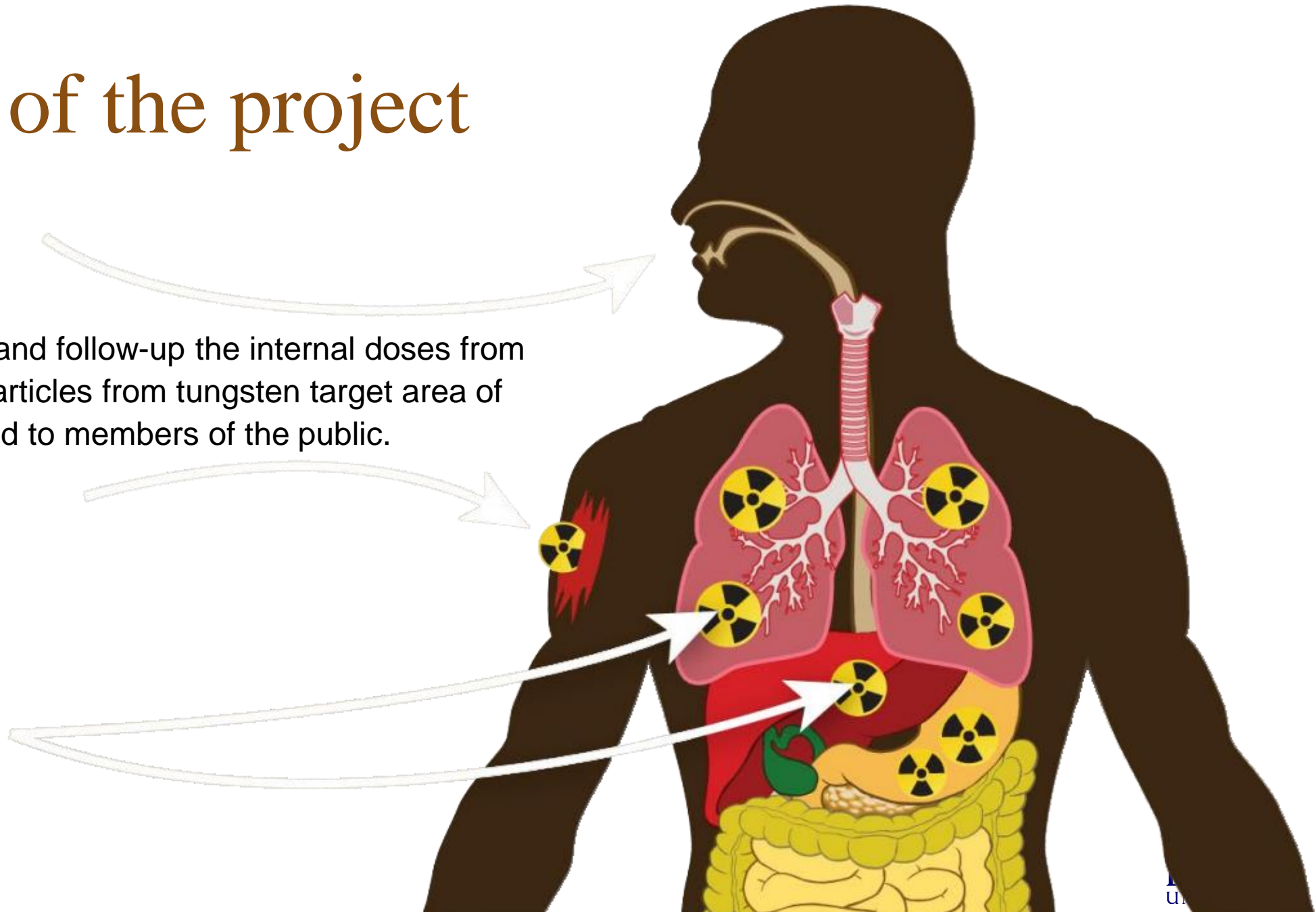
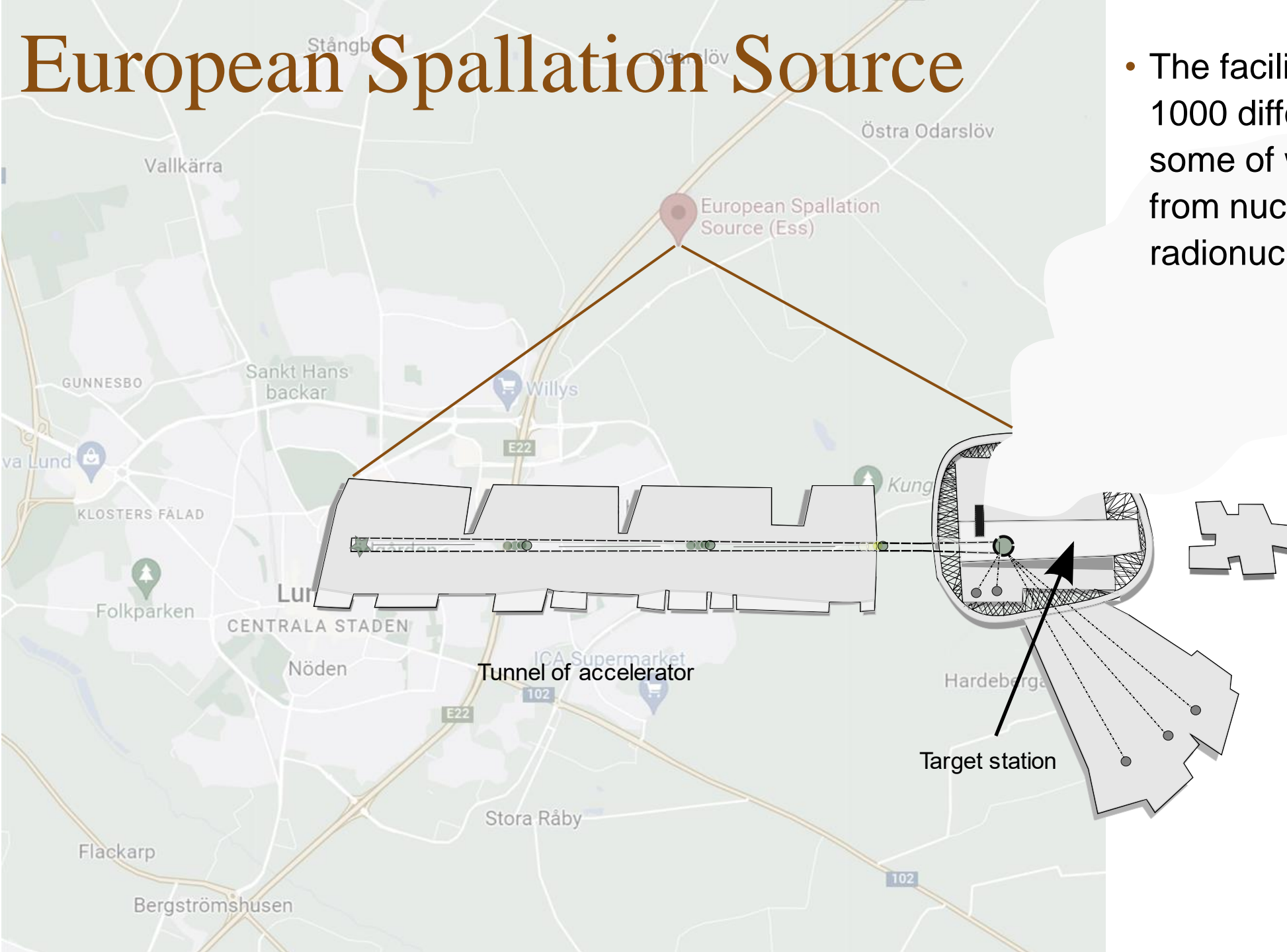


Image taken from CDC

European Spallation Source

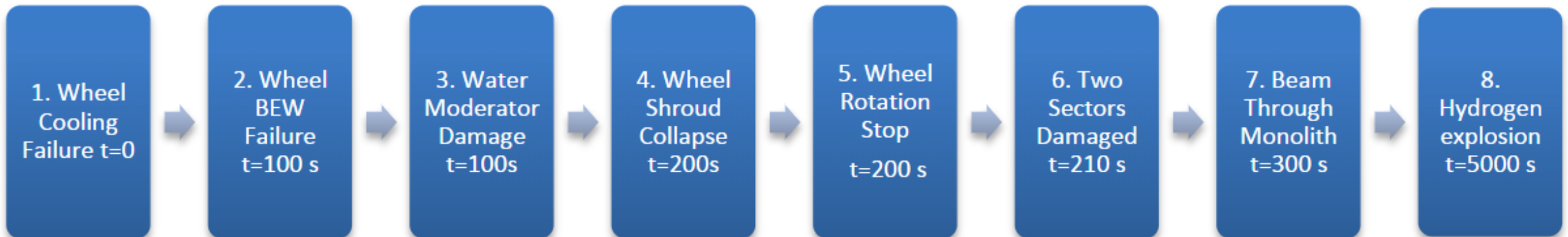
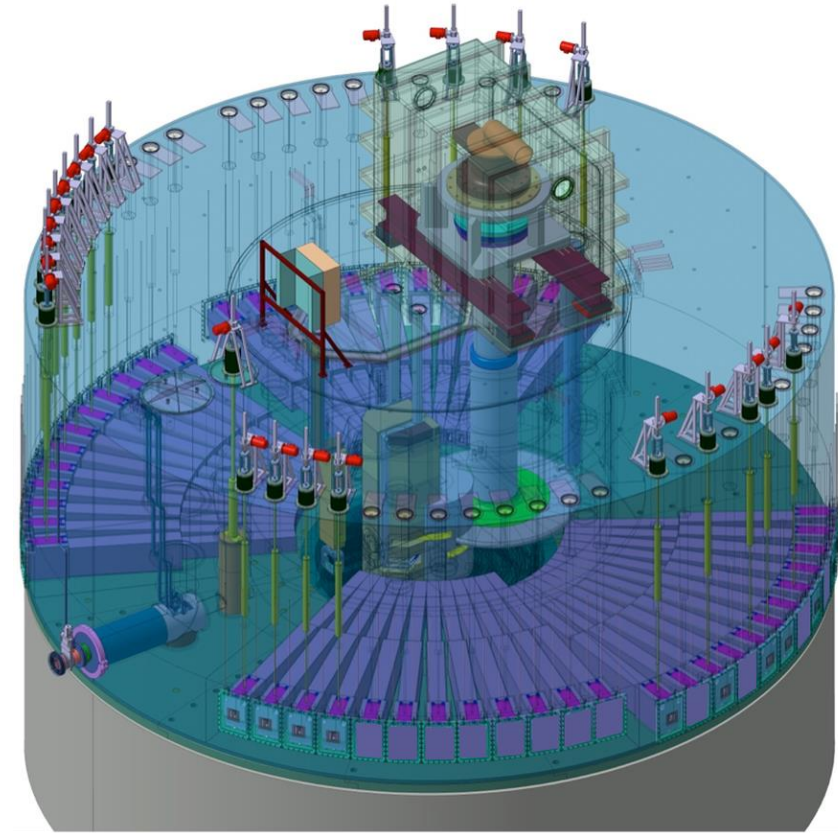
- The facility will produce over 1000 different radionuclides, some of which will be different from nuclear power plant radionuclides.



Worst case scenario

Loss of cooling in target wheel during beam time.

^{148}Gd , ^{187}W , ^{172}Hf , ^{182}Ta $^{178\text{m}}\text{Hf}$



Worst case scenario

Radionuclide	Half life	Dose coefficient (Sv/Bq)
148Gd	71.1 y	5.6E-8
187W	23.9 h	6.3E-10
172Hf	1.87 y	1.0E-9
182Ta	115.0 d	1.5E-9
178mHf	31 y	4.7E-9

50% of the total dose

ICRP, 2012. Compendium of Dose Coefficients based on ICRP Publication 60. ICRP Publication 119. Ann. ICRP 41(Suppl.), Spanier Leif, AA03a_LP-104B Source term to SSM, Internal document

Internal dosimetry



LUND
UNIVERSITY

Examination of excretion samples



Image taken from 3AW Melbourne

Whole body counting



Image taken from SciencePhotoLibrary

Collaboration with Linköping University

- Their previous work:
 - Bioassay calculations for retention of radionuclides in lungs
 - Simulation of gamma spectra
 - Method for determination of minimum detectable dose in lungs and urine

Internal dosimetry of radionuclides that can be released during an accident at the European Spallation Source (ESS)

2022:10

Author: Håkan Pettersson, Alexandr Malusek, Marie Karlsson
Linköpings universitet, Linköping
Report number: 2022:10
ISSN: 2000-0456
Available at: www.ssm.se

MDA vs lungs and daily urine excretion activity

Criteria for workers

CED=20 mSv

Detectability time window

Nuclide	γ -energy [keV]	Day1		Day2		Day3		Day4		Day10	
		urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]
¹⁸⁷ W	686	991900	37.3	52080	11.3	2278	5.2	356	3.5	2.1	1.3
¹⁷² Hf	126	28.9	2.0	69.8	55.8	71.4	17.2	66.6	10.9	42.7	5.0
¹⁸² Ta	1189	3.87	32.2	8.9	12.9	8.5	6.5	7.3	4.4	3.3	1.5
^{176m} Hf	326	1.8	44.2	4.3	10.4	4.4	2.7	4.1	1.4	2.6	0.6
^{178m} Hf	426	1.8	49.9	4.3	11.7	4.4	2.8	4.1	1.4	2.6	0.4
¹⁸³ Ta	108	5.1	233	10.2	56.7	8.5	17.7	6.5	11.3	1.3	5.4
¹⁸³ Ta	246	5.1	124	10.2	29.3	8.5	7.5	6.5	4.0	1.3	1.8
¹⁷³ Lu	172	400	154	321	37.0	138	10.9	62.3	6.9	17.0	3.9
¹⁶⁹ Yb	198	486	76.1	384	18.1	163	5.0	73.1	3.1	18.5	1.7
¹⁷¹ Lu	740	511	17.4	378	6.5	149	3.6	62.1	2.4	10.3	0.9
¹⁷² Lu	1094	280	11.4	203	4.5	78.7	2.2	32.1	1.5	4.8	0.5
¹⁴⁸ Gd	115	33.6	58.7	27.0	14.2	11.8	4.4	5.6	2.8	1.7	1.3
¹⁴⁸ Gd	116	33.6	58.7	27.0	14.2	11.8	4.4	5.6	2.8	1.7	1.3
¹⁴⁸ Eu	747	4.7	8.2	7.0	3.2	4.3	1.8	2.5	1.2	1.3	0.4
¹⁵³ Gd	103	28.8	125	23.4	30.4	10.3	9.5	4.9	6.1	1.6	2.9
¹⁴⁸ Gd	alpha	1.23		1.00		0.44		0.21		0.07	

Criteria for public

CED=1 mSv

Nuclide	γ -energy [keV]	Day1		Day2		Day3		Day4		Day10	
		urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]	urine [Bq]	MDA [Bq]
¹⁸⁷ W	686	49595	8.4	2604	2.6	114	1.2	17.8	0.80	0.11	0.30
¹⁷² Hf	126	1.4	51.5	3.5	12.5	3.6	3.9	3.3	2.5	2.1	1.1
¹⁸² Ta	1189	0.19	7.3	0.44	2.9	0.42	1.5	0.37	1.0	0.16	0.39
^{178m} Hf	326	0.09	9.9	0.21	2.3	0.22	0.60	0.20	0.32	0.13	0.13
^{178m} Hf	426	0.09	11.2	0.21	2.6	0.22	0.63	0.20	0.31	0.13	0.10
¹⁸³ Ta	108	0.25	52.2	0.51	12.7	0.43	4.0	0.32	2.5	0.07	1.22
¹⁸³ Ta	246	0.25	27.8	0.51	6.6	0.43	1.7	0.32	0.91	0.07	0.41
¹⁷³ Lu	172	20.0	34.5	16.1	8.3	6.9	2.5	3.1	1.6	0.85	0.88
¹⁶⁹ Yb	198	24.3	17.0	19.2	4.1	8.1	1.1	3.7	0.70	0.93	0.38
¹⁷¹ Lu	740	25.5	3.9	18.9	1.5	7.5	0.81	3.1	0.56	0.52	0.21
¹⁷² Lu	1094	14.0	2.6	10.2	1.0	3.9	0.51	1.6	0.35	0.24	0.13
¹⁴⁸ Gd	115	1.7	13.1	1.35	3.2	0.59	0.99	0.28	0.63	0.08	0.30
¹⁴⁸ Gd	116	1.7	13.1	1.35	3.2	0.59	0.99	0.28	0.63	0.08	0.30
¹⁴⁸ Eu	747	0.25	1.9	0.35	0.72	0.21	0.40	0.15	0.26	0.07	0.10
¹⁵³ Gd	103	1.4	28.0	1.2	6.8	0.52	2.1	0.25	1.4	0.08	0.66
¹⁴⁸ Gd	alpha	0.061		0.050		0.022		0.011		0.0035	

green colour - favourable conditions for activity quantification,
 yellow colour - possibly favourable conditions for quantification with larger HPGe-detectors and/or evaporation of urine sample of about a factor of 10.
CED – committed effective dose

What can affect detectability?

- ✓ Chemical form of radionuclide
- ✓ Aerosol size of the radionuclide
- ✓ Biokinetics of specific radionuclide
- ✓ Individual size, age, gender and lifestyle habits
- ✓ Detector specifications



Sensitivity analysis needed!

Sensitivity analysis

- Investigate feasibility of internal dose assays of alpha, beta, and gamma emitters using excretion samples:
 - Find biokinetic parameters with biggest influence on activity in 24h lungs, urine and faeces for most radiotoxic radionuclides;
 - Perform the minimum detectable activity analysis for different spectra scenarios;
 - Compare minimum detectable activity with detector at LiU and LU;

Collaboration with



LINKÖPING
UNIVERSITY

SENSITIVITY ANALYSIS OF THE ICRP BIOKINETIC MODEL PREDICTING THE ACTIVITY OF Gd IN LUNGS AND 24-HOUR EXCRETION SAMPLES

Belkise RAMLJAK¹, Alexandr MALUSEK², Kristina ERIKSSON STENSTRÖM³, Christopher RÅÅF³
¹Division of Particle and Nuclear Physics, Lund University; ²Division of Diagnostics and Specialist Medicine, Linköping University; ³Medical Radiation Physics, Lund University
belkise.ramljak@hep.lu.se; alexandr.malusek@liu.se; kristina.stenstrom@hep.lu.se; christopher.raaf@med.lu.se

Abstract: ¹⁴⁶Gd may contribute to around 50% of the dose from inhalation in case of an accident at the European Spallation Source. Methods for its detection have been developed. They rely on biokinetic models predicting activities of the gamma emitters ¹⁴⁶Gd ($T_{1/2} = 48.3$ d) and ¹⁵²Gd ($T_{1/2} = 240.4$ d) as tracers in the lungs, urine, and faeces. The optimal choice of model parameters and associated uncertainties, which propagate to the estimates of the minimum detectable activity of ¹⁴⁶Gd ($T_{1/2} = 84.4$ y), have yet to be systematically investigated. This work presents a step in this direction. The authors have implemented the ICRP biokinetic model for Gd and performed a sensitivity analysis of the model to identify the most influential parameters. They will use this knowledge in subsequent uncertainty analysis and determination of the optimal measurement time window.

Keywords: ESS, radioactive aerosols, internal dosimetry

1. Introduction

The European Spallation Source (ESS) is a neutron production facility being built in the vicinity of Lund in southern Sweden. Its main components will be a pulsed (14 MeV) linear accelerator emitting 2 GeV protons, a tungsten target, and scientific stations dedicated to experiments using neutron spallation reactions [9]. Radiation protection systems comprise, e.g., shielding of the accelerator via 5 m of soil, steel and concrete around the target and filters in the ventilation exhaust [9]. These systems may not shield entirely the environment and the public from radiation exposure in case of an accidental release of radionuclides. The worst-case scenario accident is caused by an initial loss of the helium cooling of the target while the target is still irradiated by the 5 MW proton beam. A series of events (including melting and oxidizing of the tungsten target and hydrogen deflagration) will lead to the release of helium, filter particles and aerosols formed from the target material [4]. Radionuclide exposure to the workers and public will be mainly through inhalation, ingestion,

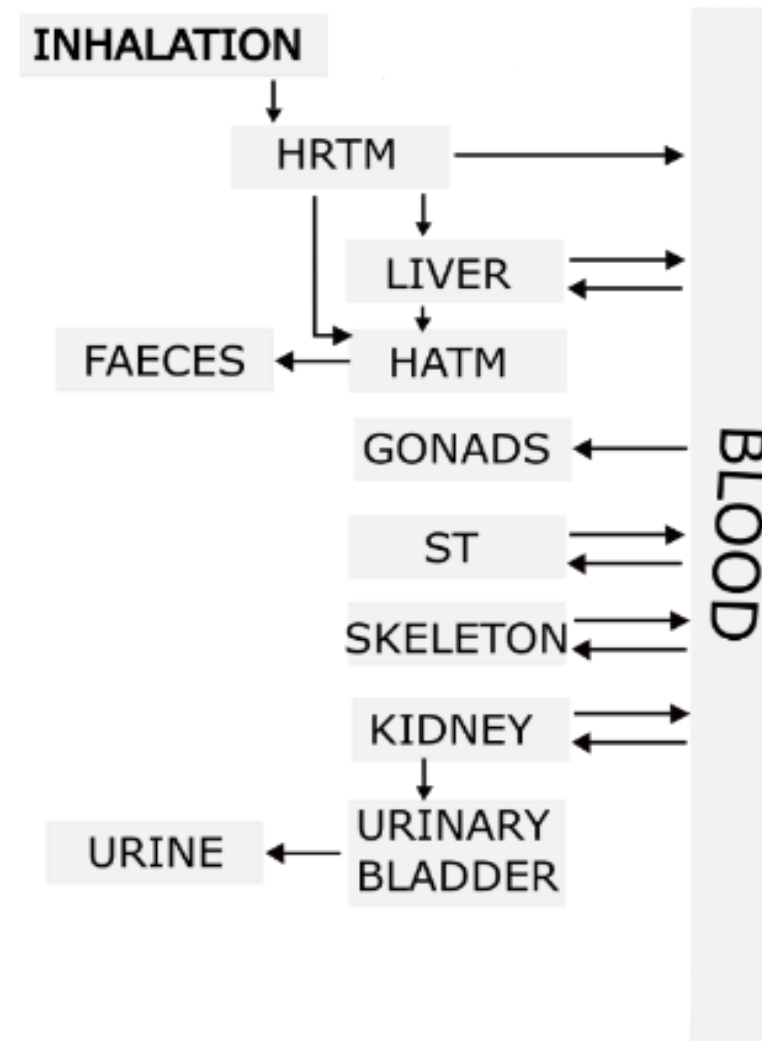
groundshine and cloudshine. In this article, we focus on the inhalation route only. Current estimates of the released radionuclide mix predict that the most radiotoxic radionuclides would be ¹⁴⁶Gd ($T_{1/2} = 84.4$ y [15]), ¹⁰⁷W ($T_{1/2} = 23.7$ h), ¹³²Te ($T_{1/2} = 1.87$ y), ⁹⁰Ta ($T_{1/2} = 114.4$ d), and ¹²⁵I ($T_{1/2} = 59.49$ d), with ¹⁴⁶Gd contributing to around 50% of the dose [8]. Since ¹⁴⁶Gd contributes most of the dose and is an alpha emitter, entailing a higher radiobiological effect, its detection is the main focus of this article. ¹⁴⁶Gd is one of the ESS-specific radionuclides which is not relevant in waste from nuclear power plants. Hence, standard methods for assessing ¹⁴⁶Gd in the environment or humans have not yet been developed.

Internal contamination assessment is usually done through whole body counting, urinal, faecal and blood sample examination. Biokinetic models predicting the 24-hour excretion in urine and faecal as a function of time and their relation to the organ and whole-body retention of the radionuclides can be used to assay internal dose from excretion samples. The latest ICRP biokinetic models described in publications 130, 134, 137, and 141 are implemented in the commercially available Taurus [19] and IDEApuls codes. These codes allow the user to specify values of model parameters. These parameters are in many cases known with limited accuracy, especially for the exposure pathway through the respiratory tract, and may vary widely depending on the physical size and chemical form of particles containing the radionuclides. The large number of model parameters complicates the corresponding uncertainty analysis. A commonly used approach is to perform a sensitivity analysis of the model and focus on the parameters that notably affect the resulting uncertainty. Sensitivity analysis is a technique used to determine how different values of an independent variable impact a particular dependent variable under a given set of assumptions. For instance, local sensitivity analysis methods are used in metrology, where the interest is mainly in the true value and uncertainty [1]. Global methods are of interest in disciplines where the input quantities vary notably. Regression methods do not fully



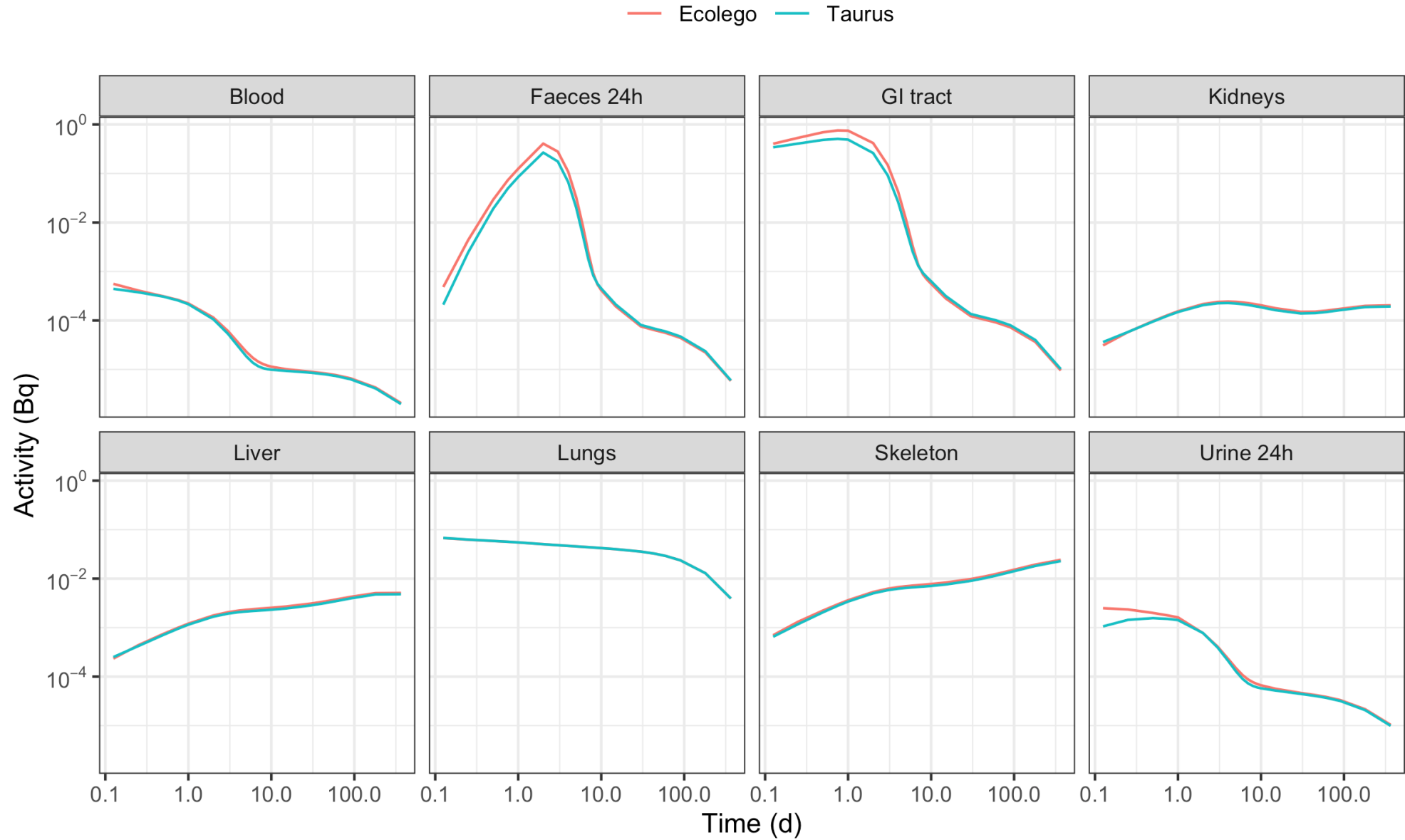
^{148}Gd Biokinetic model

- The model is developed in Ecolego by following recommendations in ICRP 100, ICRP 141 and ICRP 130.
- The model is made for
 - male reference worker
 - $5\mu\text{m}$ aerodynamic median activity diameter (AMAD)
 - Oxide form



Model validation

- Ecolego model outputs were compared to outputs from the same conditions from Taurus.

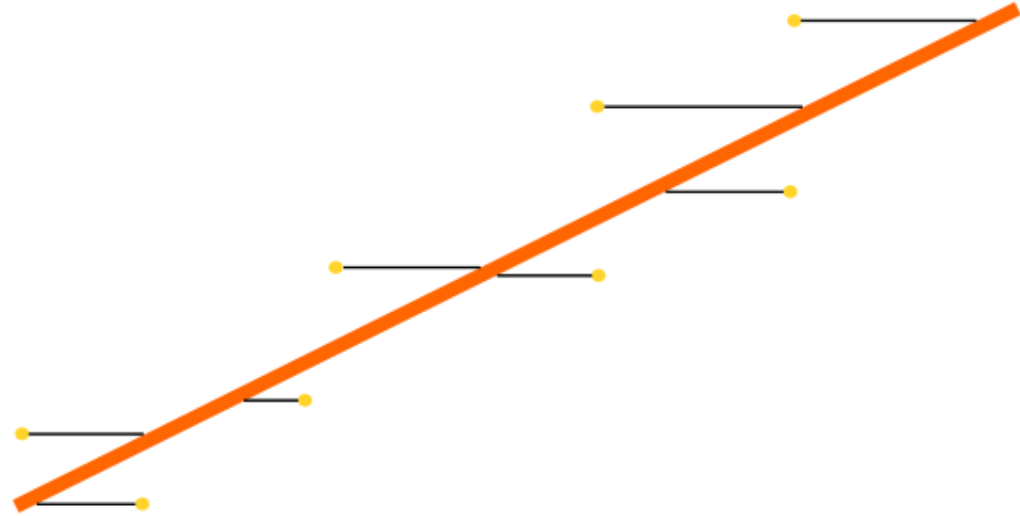


Sensitivity analysis

- Analysis is done by calculating standard regression coefficients (SRC). They indicate how many standard deviations the dependent variable (lung activity and excretion activity) is likely to change based on changes in independent variables (biokinetic transfer parameters).
- a sensitivity coefficient is defined as:

$$\left| \frac{\partial f}{\partial x_i} \right|$$

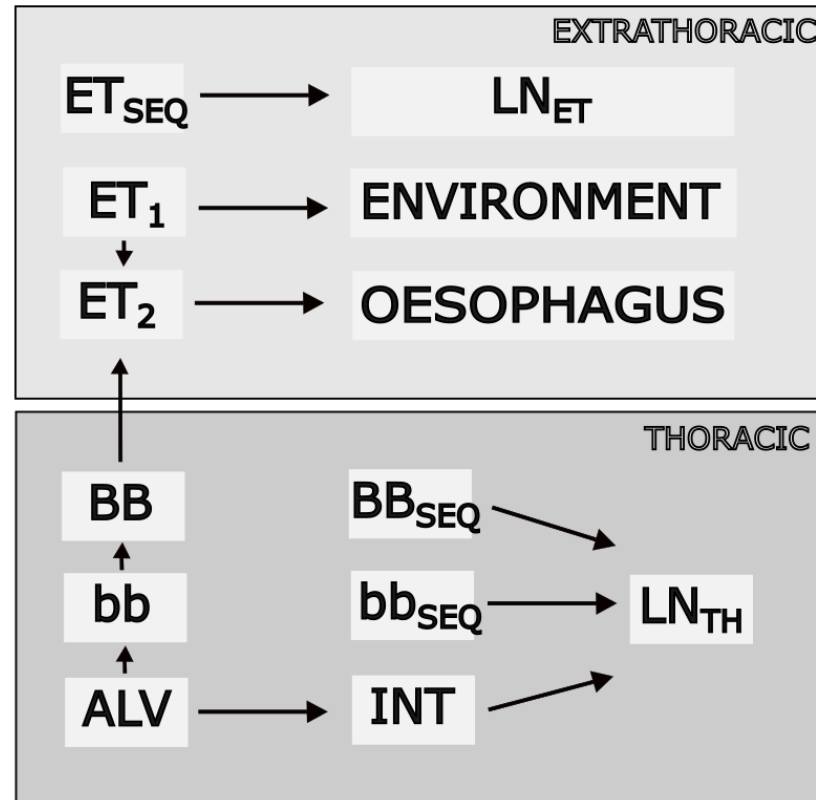
- *we assume that all other inputs are not changed.*



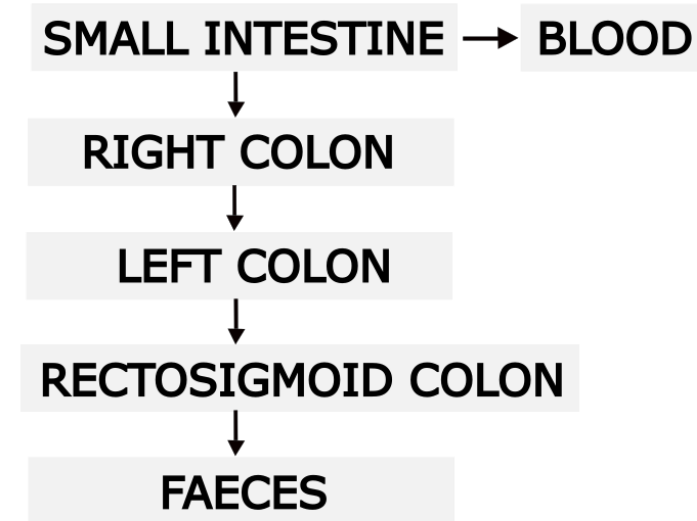
Sensitivity analysis

- Sensitivity analysis was done by using Ecolego toolbox.
- All transfer coefficients were selected as independent variables, with assumed normal distribution.

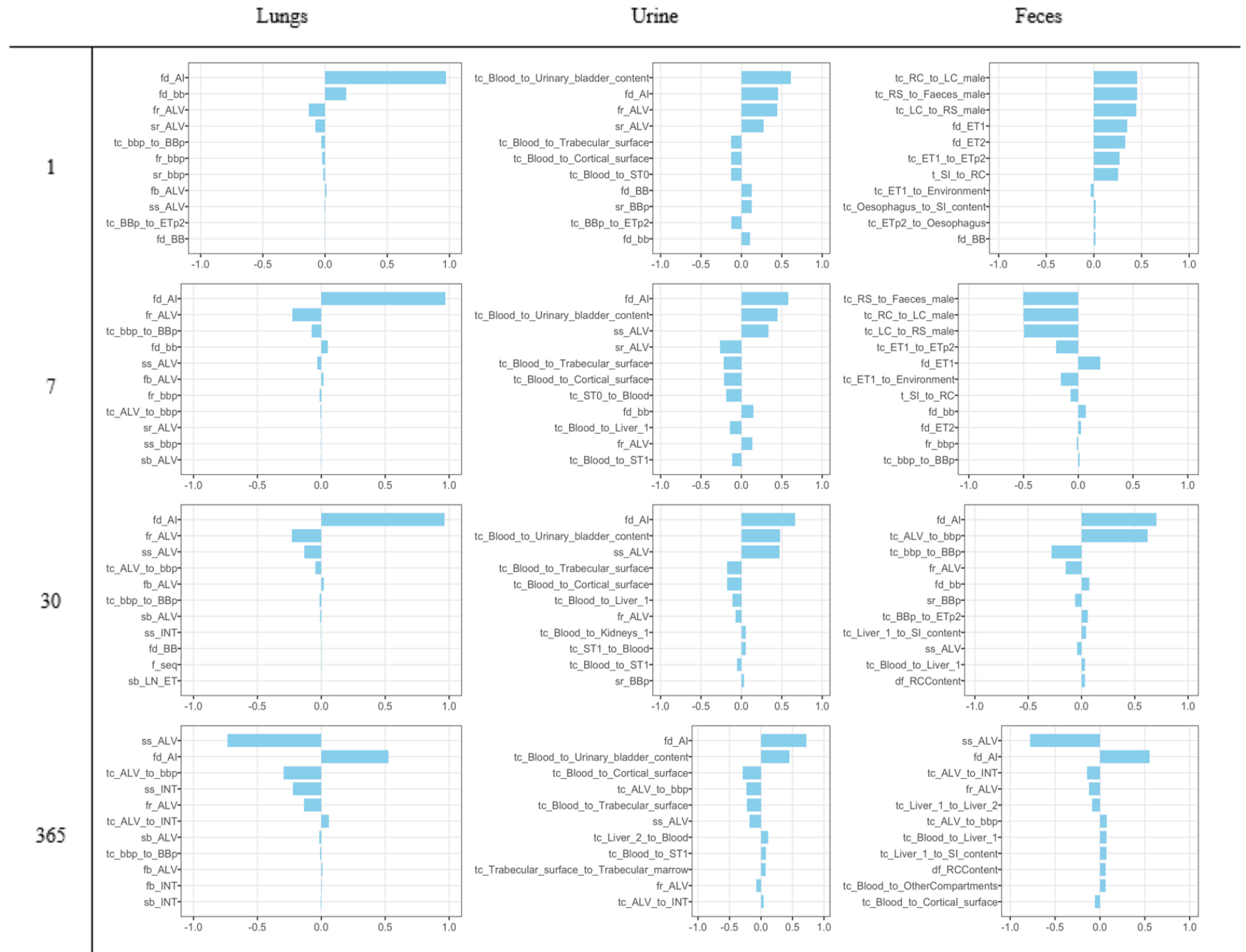
Lung model



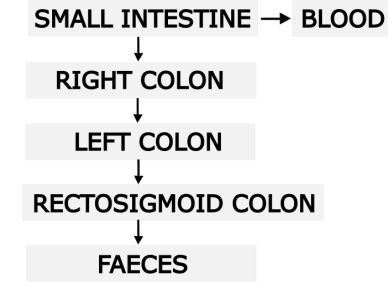
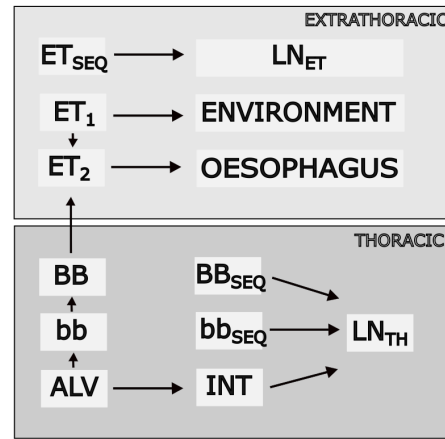
GI tract



Sensitivity analysis results



Sensitivity analysis results



Comp.	Order	1	7	30	365
Lung	1	$f_{d,AI}$ +0.97	$f_{d,AI}$ +0.97	$f_{d,AI}$ +0.96	$S_{s,ALV}$ -0.74
	2	$f_{d,bb}$ +0.17	$f_{r,ALV}$ -0.23	$f_{r,ALV}$ -0.23	$f_{d,AI}$ +0.53
	3	$f_{r,ALV}$ -0.13	$\lambda_{bb' \rightarrow BB'}$ -0.08	$S_{s,ALV}$ -0.13	$\lambda_{ALV \rightarrow bb'}$ -0.29
	4	$S_{r,ALV}$ -0.08	$f_{d,bb}$ +0.05	$\lambda_{ALV \rightarrow bb'}$ -0.05	$S_{s,INT}$ -0.22
Urine	1	$\lambda_{blood \rightarrow UBC}$ +0.61	$f_{d,AI}$ +0.58	$f_{d,AI}$ +0.66	$f_{d,AI}$ +0.72
	2	$f_{d,AI}$ +0.45	$\lambda_{blood \rightarrow UBC}$ +0.45	$\lambda_{blood \rightarrow UBC}$ +0.48	$\lambda_{blood \rightarrow UBC}$ +0.45
	3	$f_{r,ALV}$ +0.44	$S_{s,ALV}$ -0.26	$S_{s,ALV}$ +0.47	$\lambda_{blood \rightarrow CS}$ -0.29
	4	$S_{r,ALV}$ +0.28	$S_{r,ALV}$ +0.34	$\lambda_{blood \rightarrow TS}$ -0.18	$\lambda_{ALV \rightarrow bb'}$ -0.23
Faeces	1	$\lambda_{RC \rightarrow LC}$ +0.46	$\lambda_{RS \rightarrow Faeces}$ -0.50	$f_{d,AI}$ +0.70	$S_{s,ALV}$ -0.78
	2	$\lambda_{RS \rightarrow Faeces}$ +0.46	$\lambda_{RC \rightarrow LC}$ -0.50	$\lambda_{ALV \rightarrow bb'}$ +0.62	$f_{d,AI}$ +0.56
	3	$\lambda_{LC \rightarrow RS}$ +0.45	$\lambda_{LC \rightarrow RS}$ -0.49	$\lambda_{bb' \rightarrow BB'}$ -0.28	$\lambda_{ALV \rightarrow INT}$ -0.15
	4	f_{d,ET_1} +0.35	$\lambda_{ET_1 \rightarrow ET_2}$ -0.20	$f_{r,ALV}$ -0.15	$f_{r,ALV}$ -0.12

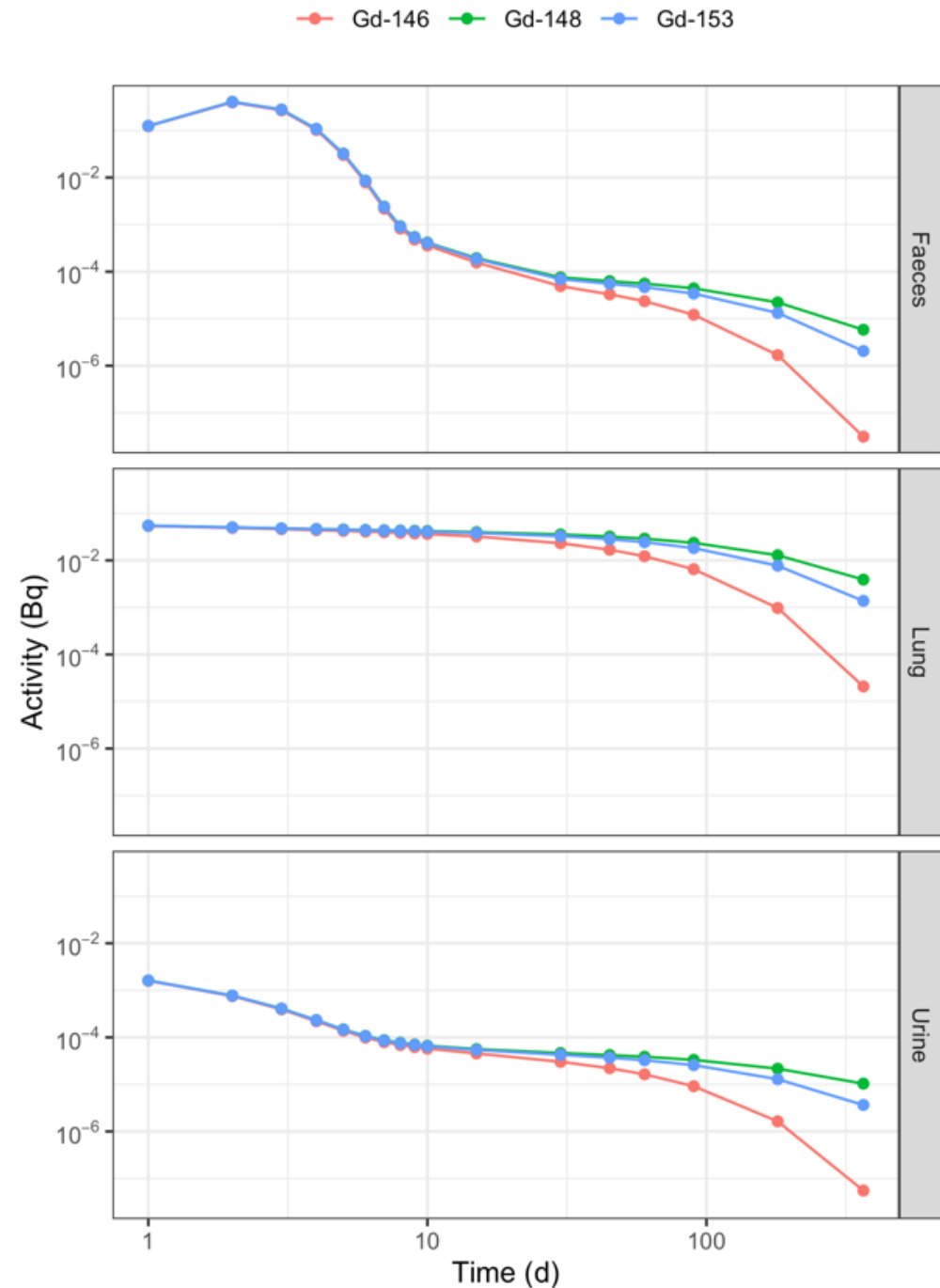
24h lung activity and 24h urine and faecal excretion for ^{148}Gd , ^{146}Gd and ^{153}Gd

^{148}Gd	Alpha emitter	86.9 y
^{146}Gd	Gamma emitter	48.27 d
^{153}Gd	Gamma emitter	240.6 d

- Isotopic ratios $^{146}\text{Gd}/^{148}\text{Gd}$ and $^{153}\text{Gd}/^{148}\text{Gd}$ can be used to estimate whole body burden of ^{148}Gd .

Consult:

- Rääf C., Barkauskas V., Eriksson Stenström K, Berhardsson C, Pettersson HBL, Internal dose assessment of ^{148}Gd using isotope ratios of gamma emitting ^{146}Gd or ^{153}Gd in accidentally released spallation target particles, *Sci. Rep.* 2020 Dec 14;10(1):21887



Future plans

- Uncertainty analysis of sample activities and associated MDAs.
- Other ESS relevant radionuclides will be added to the model.



LUND
UNIVERSITY