DE LA RECHERCHE À L'INDUSTRIE

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CODE DE CALCUL TRIPOLI-4® - APPLICATION EN DOSIMÉTRIE POUR LES DEUX VICTIMES DE L'ACCIDENT DE CRITICITÉ DE TOKAI-MURA (1999)

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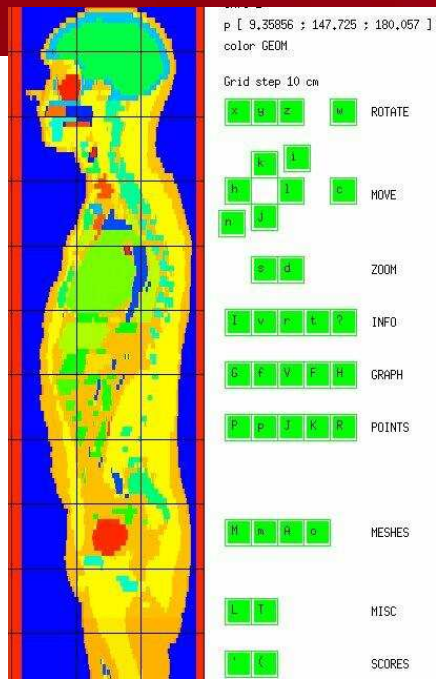
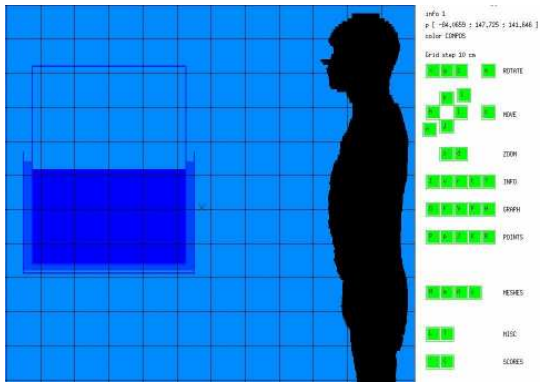
SFRP JOURNÉES TECHNIQUES, FONTENAY-AUX-ROSES, 10/03 2023

OUTLINE



- Introduction
- Tokai-Mura criticality accident (1999)
- TRIPOLI-4 Monte Carlo Code
- ICRP Voxel phantoms modeling I & II
  - T4G Display Tool & Organ dose calculations
- Conclusions



INTRODUCTION

TRIPOLI-4 is a general-purpose Monte Carlo radiation transport code developed by the Service d'Études des Réacteurs et de Mathématiques Appliquées (SERMA) at CEA-Saclay, France.

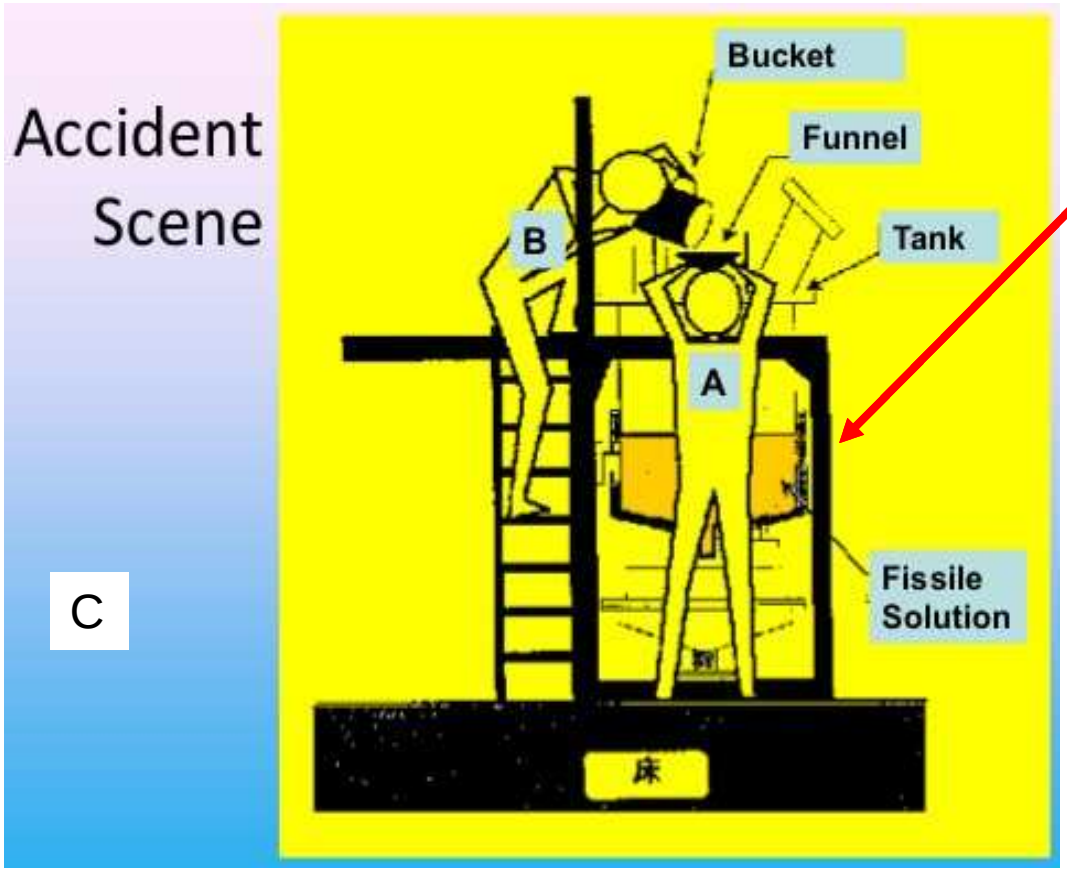
It uses continuous-energy nuclear data to simulate neutron, photon, electron and positron transport in fields like radiation protection and shielding, fission reactor physics, and nuclear criticality safety.

To study radiation dosimetry in human organs, computational phantoms were recently modeled using the TRIPOLI-4 geometry.

To easily use the ICRP voxel-based phantoms in different exposure scenarios, a newly developed PHANTOM option is helpful to model one or more phantoms in diverse irradiation environments.

In this study, this new phantom option was applied to radiation dose-reconstruction for the victims of 1999 Tokai-Mura criticality accident.

TOKAI-MURA CRITICALITY ACCIDENT 1999



- JCO fuel processing plant
- Uranyl (U-235 18.8%) nitrate in tank ➔ Joyo fuel
- Solution reached criticality ➔ about 16.6 kg U / 45 L
- A: standing beside the tank and holding a funnel
  ➔ 16-20 Gy ➔ 82 days
- B: bending over the tank and pouring the uranyl nitrate solution
  ➔ 6-10 Gy ➔ 210 days
- C: team leader 4 m away
  ➔ 1.2 – 5.5 Gy

Ref: R. A. Knief, SAND2013-6983P
K. Miyamoto et al. Health Physics, 83 (2002) 19

➔ High dose estimation: ^{24}Na in blood, Biodosimetry

TOKAI-MURA CRITICALITY ACCIDENT 1999

Table 5

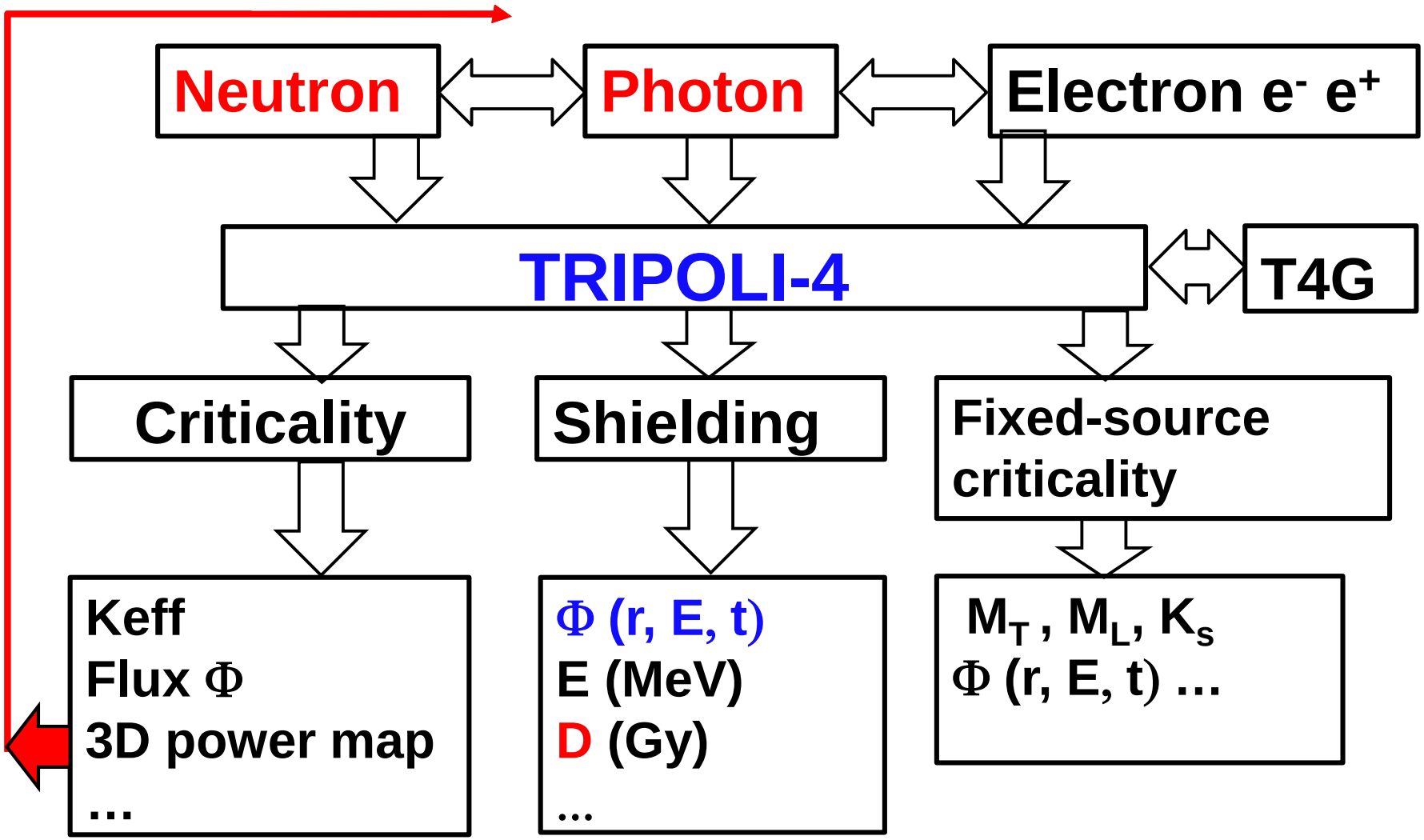
Estimated doses (Murata and Akashi, 2002) (γ-ray equivalent dose: GyEq).

Method	Worker		
	A	B	C
(1) Prodromal symptoms	Over 8	4–6 or over 6	Less than 4
(2) Lymphocyte counts	16–23	6–8	1–5
(3) Chromosome analysis	16 to over 30	6.9–10	2.8–3.2
(4) ^{24}Na in the body			
1) Specific activity of ^{24}Na in blood (Neutron, γ-ray: Gy)	(5.4, 9.9)	(2.9, 4.1)	(0.81, 1.5)
Total dose (assuming RBE = 1.7)	19	9.0	2.9
2) Whole-body counting (Neutron, γ-ray: Gy)	–	–	(0.62, 1.1)
Estimated doses based on the methods (1)–(4)	16–25	6–9	2–3

Ref: A. Endo et al. Radiation Measurements, 45 (2010) 1484

➔ **High dose estimation: Biodosimetry**

TRIPOLI-4 RADIATION TRANSPORT CODE
T4G – 2D & 3D I/O VIEWER



Ref.: J.-P. Both, Y.-K. Lee et al., Journées scientifiques, SFRP, Oct., 2003, **Sochaux**

RADIATION PROTECTION DOSIMETRY CALCULATIONS
OPERATIONAL & PROTECTION QUANTITIES

TRIPOLI-4 Radiation transport code

Neutron & photon flux

Flux-to-dose conversion coefficients & exposure time

Ambient dose equivalent
H*(10)
Personal dose equivalent
Hp(10)

Energy deposition
- **Neutron**
- **Photon** ➔ **Kerma K**
- **Electron**
- **Positron**

Organ mass, W_R, W_T

Organ absorbed dose D_T
Organ equivalent dose H_T
Effective dose E

Ref.: Y.-K. Lee, J. of Nucl. Eng. and Rad. Sci., Oct. 2020 / 041105-3

TRIPOLI-4 INPUT DATA

-. GEOMETRY …… Lattice & Voxel Phantoms

-. COMPOSITION ….. Uranyl nitrate & ICRP 110

-. GEOMCOMP ……. Organ ID

-. CALCULATION ….. Volume & Mass U, U-235, H

-. GRID_LIST ….. 4 groups of energy

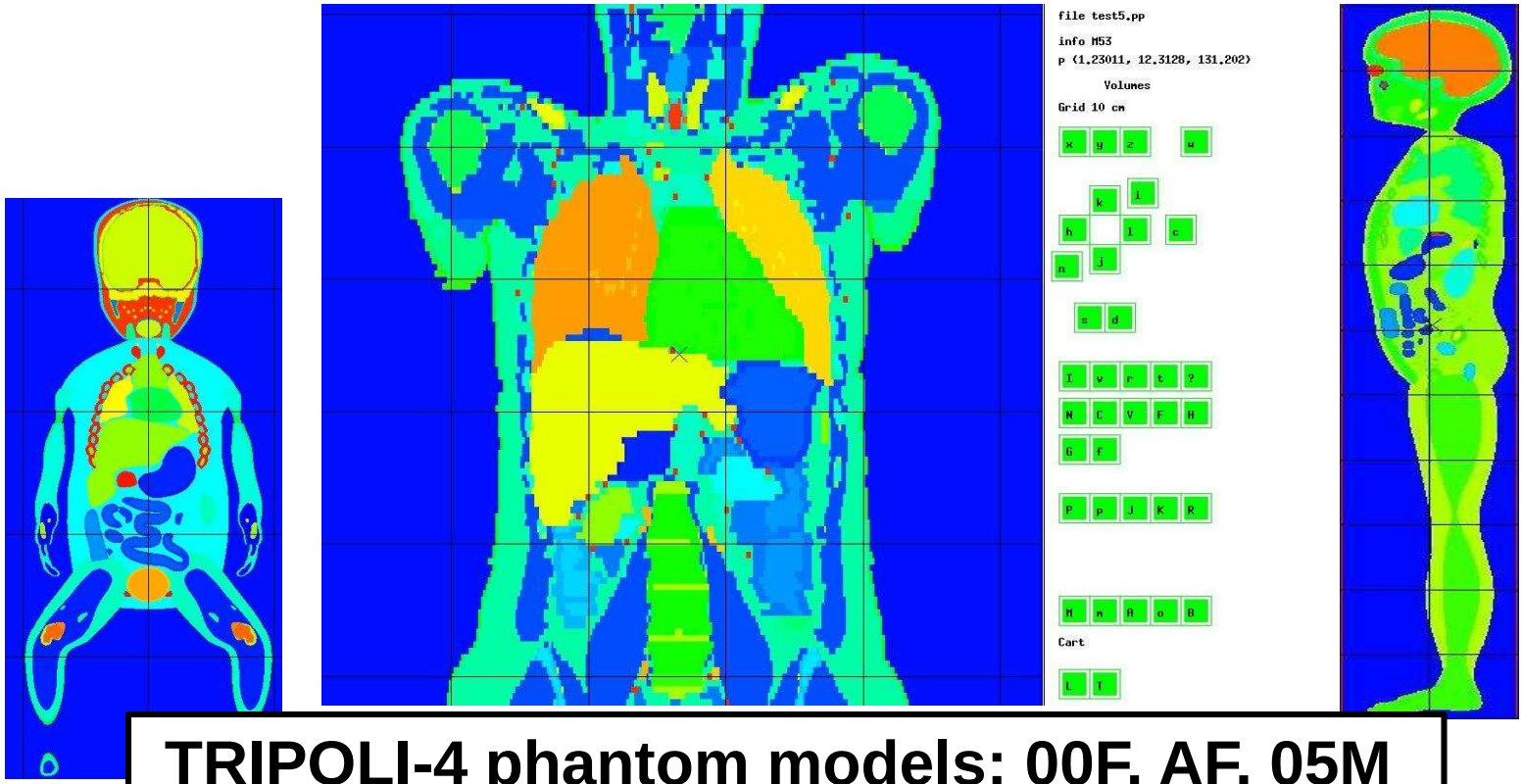
-. RESPONSES …… Energy deposition, neutron & γ

-. SCORE …… Flux & Organ dose

-. SOURCES_LIST … Intensity & distribution

-. SIMULATION …… Keff & shielding, neutron & γ

ADVANCES IN COMPUTATIONAL PHANTOMS



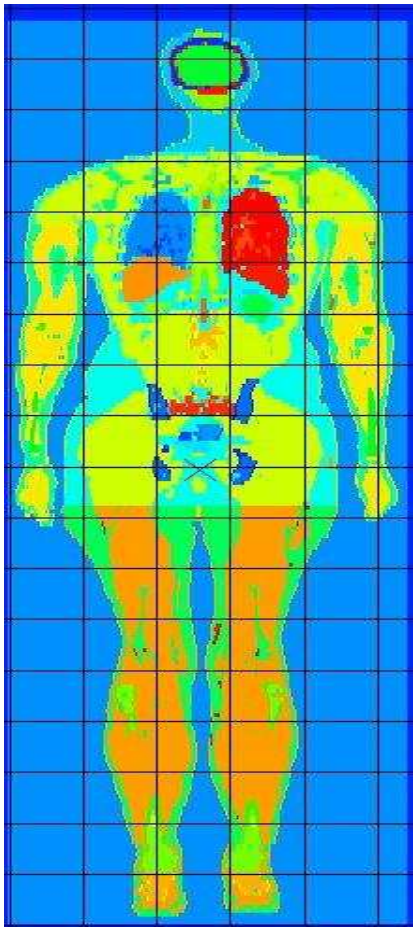
TRIPOLI-4 phantom models: 00F, AF, 05M

2009 ICRP 110 Voxel-type Reference phantoms (60 MB)

2020 ICRP 143 Pediatric Reference phantoms (>200 MB)

TRIPOLI-4 INPUT DATA – SINGLE VOXEL PHANTOM

```
VOLU 502 RESC VOLU 501  254 127 222  BASIS 1 1 1
          EXCEPT
          1950255
```



```
151 79 155
152 79 155
153 79 155
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159 80 155
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162 81 155
144 82 155
145 82 155
163 82 155
143 83 155
164 83 155
142 84 155
165 84 155
142 85 155
```

/. ICRP 110
Male Phantom (AM)

➔ **140 organs & 52 media in 7.2 M voxels (29 MB)**

➔ **1.76 m & 73 Kg**

➔ **1.95 M tissue voxels**

➔ **5.21 M void voxels**

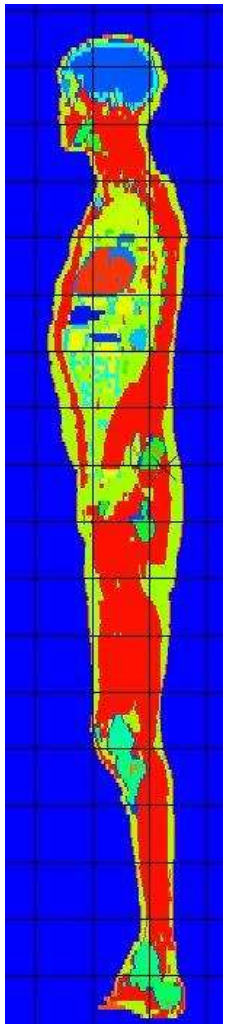
➔ **In 254 x 127 x 222 array**

➔ **Voxel dimensions**
2.137 x 2.137 x 8.0 mm^3

Ref.: Y.-K. Lee, J. of Nucl. Eng. and Rad. Sci., Oct. 2020 / 041105-3

TRIPOLI-4 INPUT DATA – SINGLE VOXEL PHANTOM INPUT OPTIMIZATION

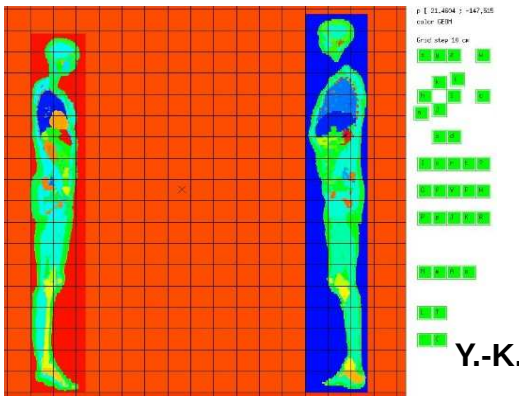
```
PHANTOM 10000
AM_organs.dat  AM.dat          //  ICRP 110 data files

DIMENSION
254       127       222        //  Voxel 3D array size
0.2137    0.2137    0.8        //  Voxel dimensions (cm)

PLUS
1000      2000  Mat            //  Increment for organs' names

FRAME
27.14     13.57     88.8       // (x, y, z) in cm & center of 3D array
1.        0.        0.         // X-axis of phantoms AM
0.        1.        0.         // Unit vector of Y-axis
0.        0.        1.         // Single AM standing phantom
ENDP
```



Ref.: Y.-K. Lee, F.-X. Hugot, Y. Jin, « New Route in TRIPOLI-4 for Radiation Dosimetry Calculations Using ICRP 110 Voxel Phantoms," ANS M&C2021, Oct. 2021.

TRIPOLI-4 INPUT DATA – TWO VOXEL PHANTOMS INPUT OPTIMIZATION

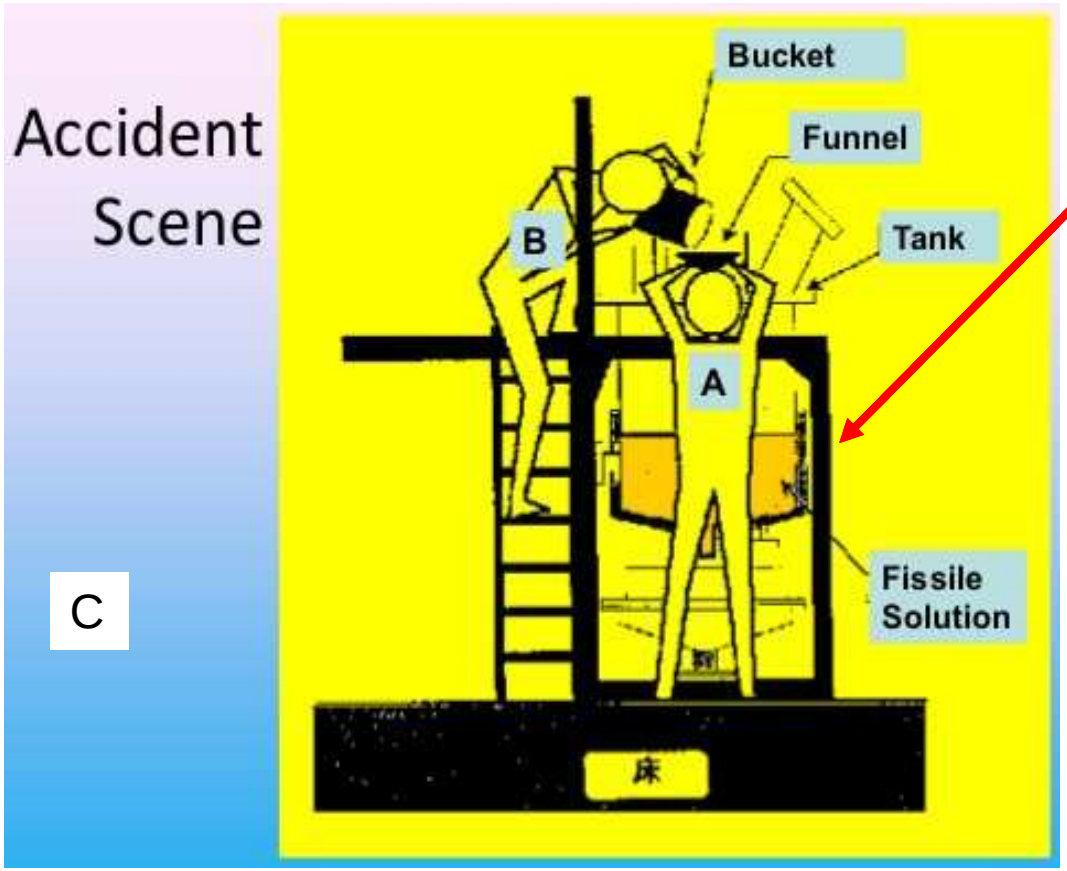
```
PHANTOM 10000                          PHANTOM 20000          // Two voxel phantoms
AM_organs.dat  AM.dat                  AF_organs.dat  AF.dat  //  ICRP 110 data files

DIMENSION                              DIMENSION
254      127      222                  299      137      348  //  Voxel 3D array size
0.2137   0.2137   0.8                  0.1775   0.1775   0.484 //  Voxel dimensions (cm)

PLUS                                   PLUS
1000     2000  Mat                     1000     2000     Mat  //  Increment for organs' names
                                                               //  Ex: Liver 11095 (AM) & 21095 (AF)
FRAME                                  FRAME
27.14    13.57    88.8                 26.536   -108.    84.216 // (x, y, z) in cm & center of 3D array
1.       0.       0.                   -1.      0.       0.     // X-axis of phantoms AM & AF
0.       1.       0.                    0.      -1.      0.     // Unit vector of Y-axis
0.       0.       1.                    0.      0.       1.     // Two face-to-face standing phantoms
                                                               // With 1 m distance at chest level
ENDP
```



Y.-K. Lee

Ref.: Y.-K. Lee, F.-X. Hugot, Y. Jin, M&C 2021.

TOKAI-MURA CRITICALITY ACCIDENT 1999



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Ref: R. A. Knief, SAND2013-6983P
K. Miyamoto et al. Health Physics, 83 (2002) 19
➔ High dose estimation: ^{24}Na in blood, Biodosimetry

CRITICALITY ACCIDENT IN TOKAI-MIRA 1999

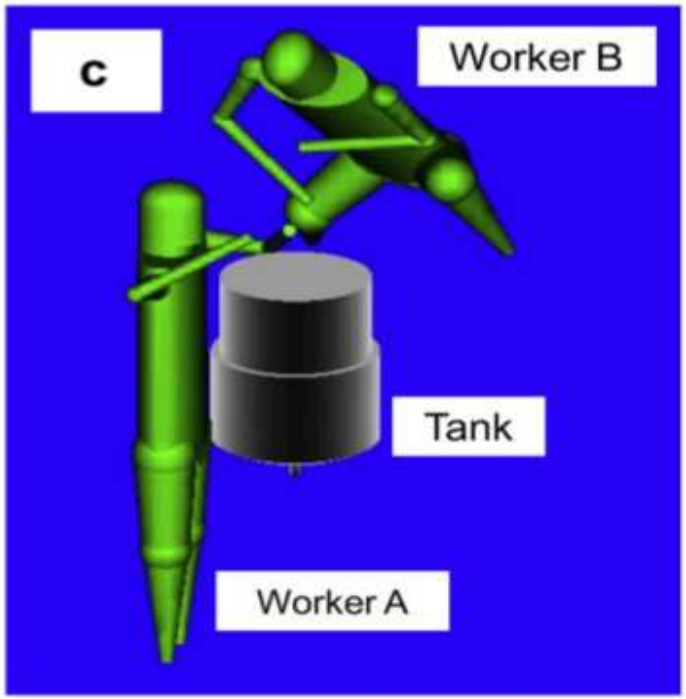


Fig. 4. Procedures for establishing simulation model.

- ➔ **MIRD phantom in simulation**
- ➔ **Organ dose of worker A: SI**
- ➔ **N: 5-12 Gy, G: 15-22 Gy**

Ref: A. Endo et al. Radiation Measurements, 45 (2010) 1484

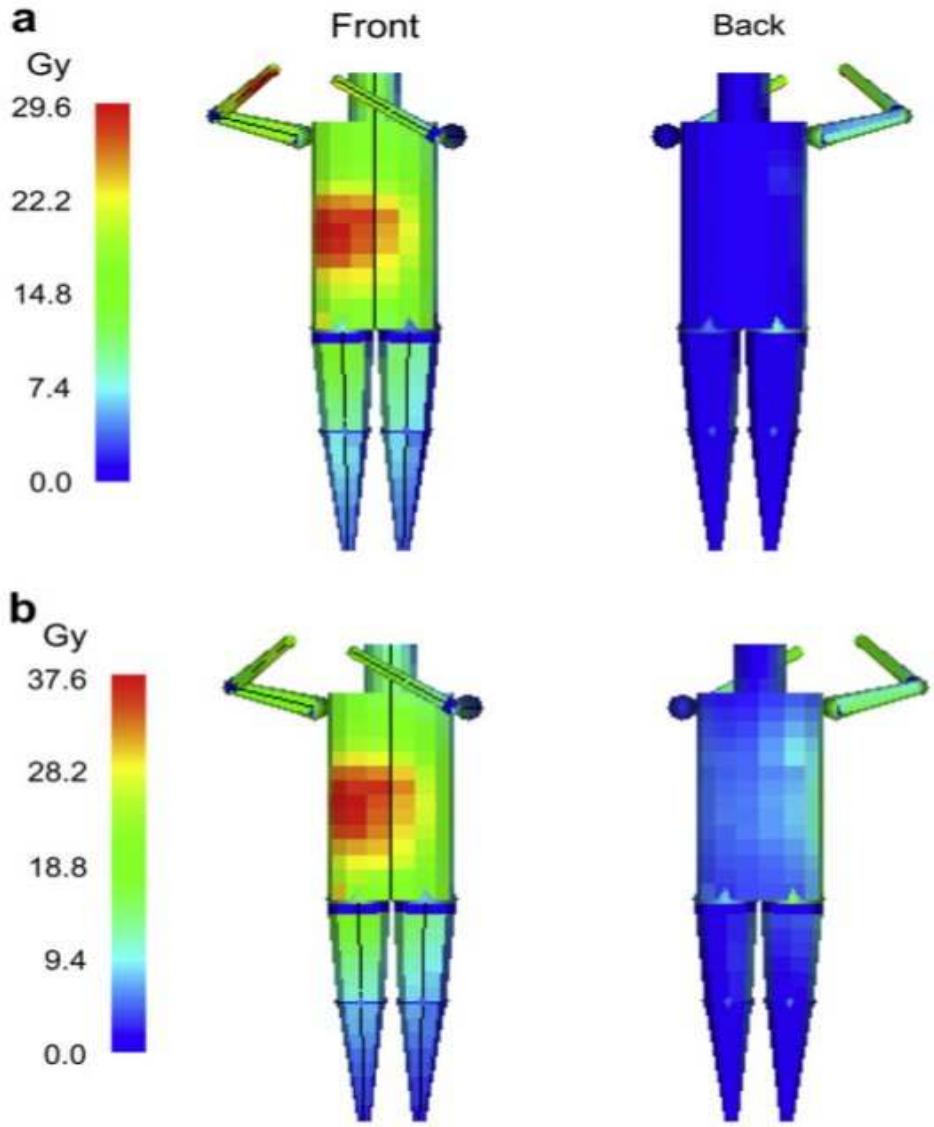


Fig. 5. Absorbed dose distribution in the skin of Worker A. (a) Neutrons, (b) γ-rays.

CRITICALITY ACCIDENT IN TOKAI-MIRA 1999

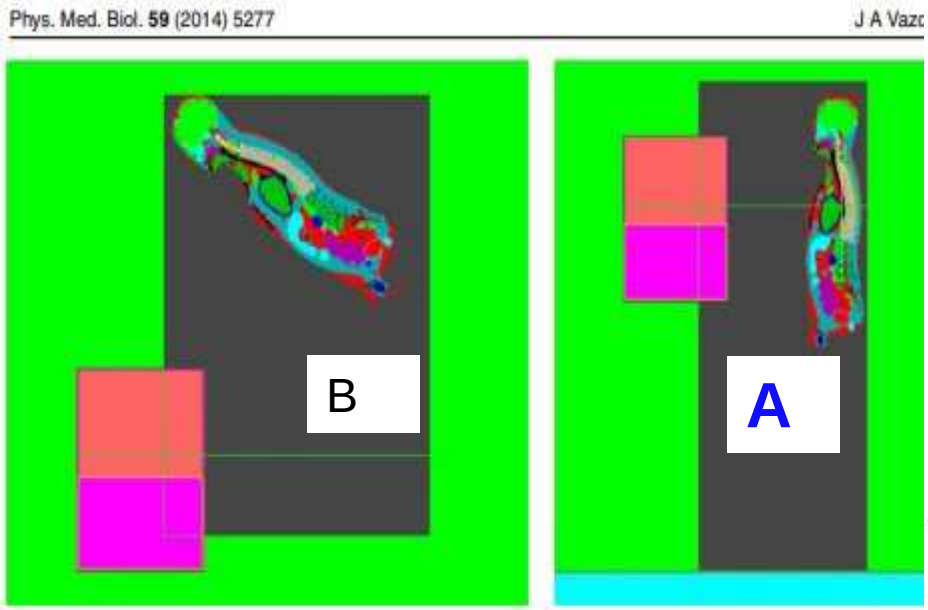
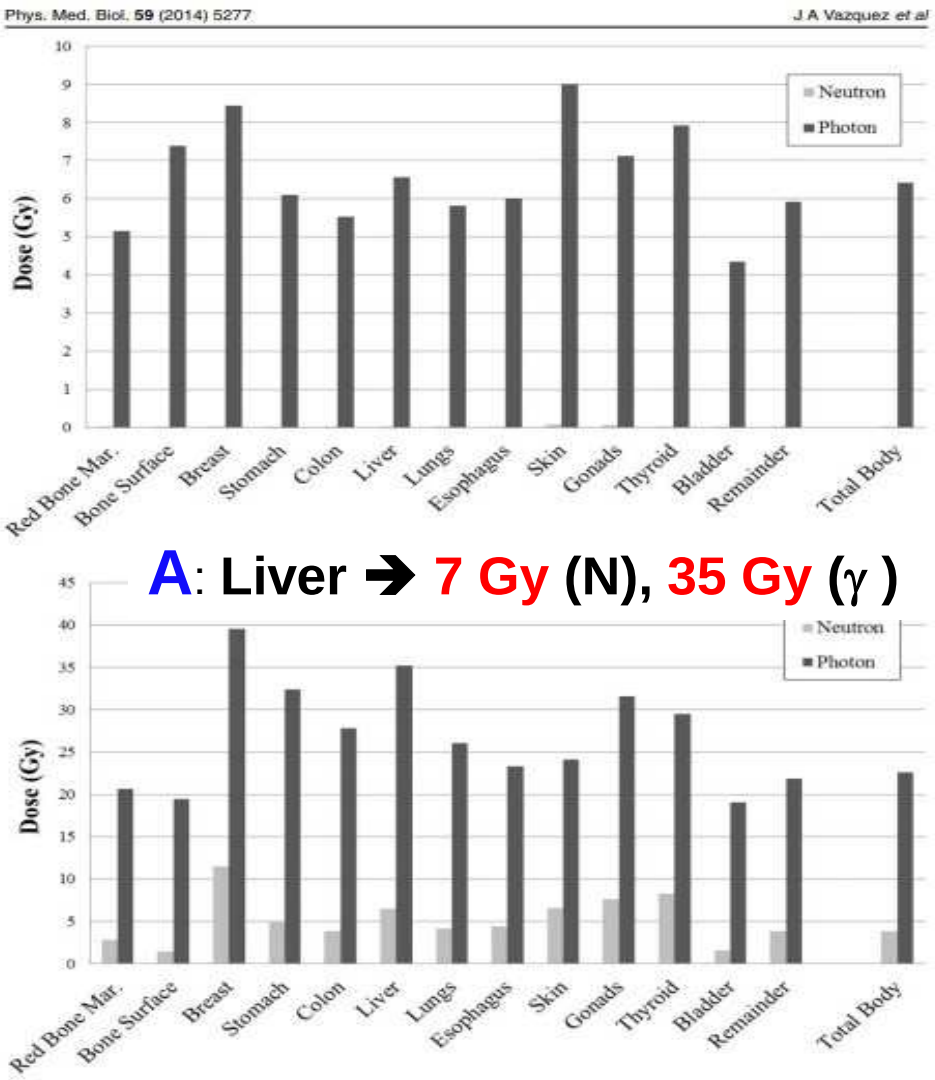


Figure 2. Cross section view of the crouching (left) and standing (right) phanto
in the MCNPX simulation. Different colors represent different organs.



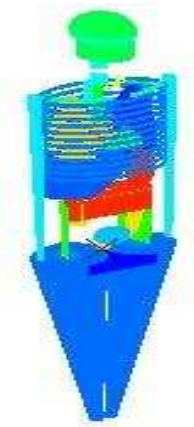
A: Liver ➔ 7 Gy (N), 35 Gy (γ)

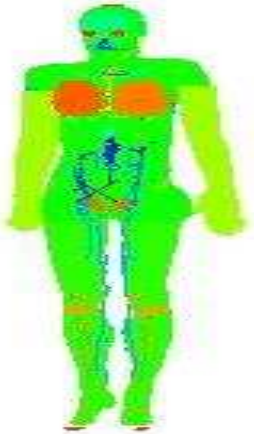
Figure 4. Calculated organ doses for the crouching-posture worker (top) and the standing-posture worker (bottom) for selected radiation-sensitive tissues.

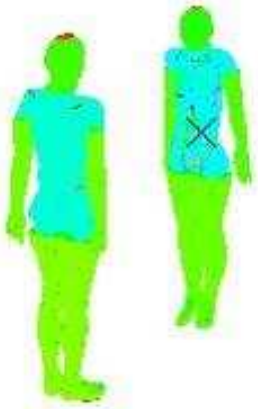
- ➔ Only one phantom in simulation
- ➔ No water layer around tank
- ➔ Organ dose: neutron < gamma
- ➔ Small neutron dose for B

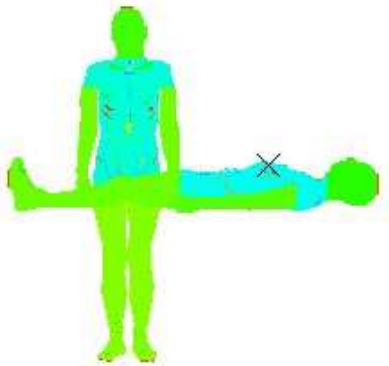
Ref: J. A. Vazquez et al.
Phys. Med. Biol., 59 (2014) 5277

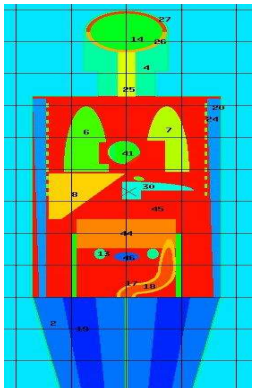
TRIPOLI-4 – T4G - MIRD AND VOXEL PHANTOMS

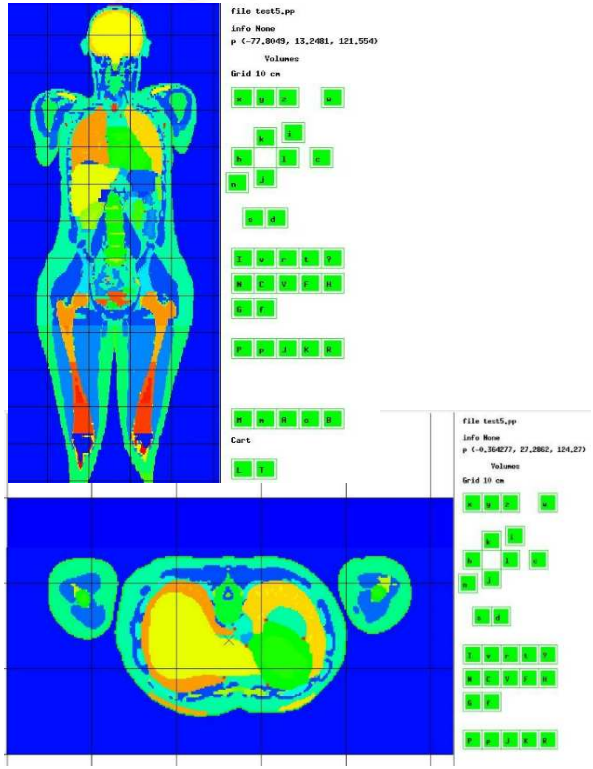


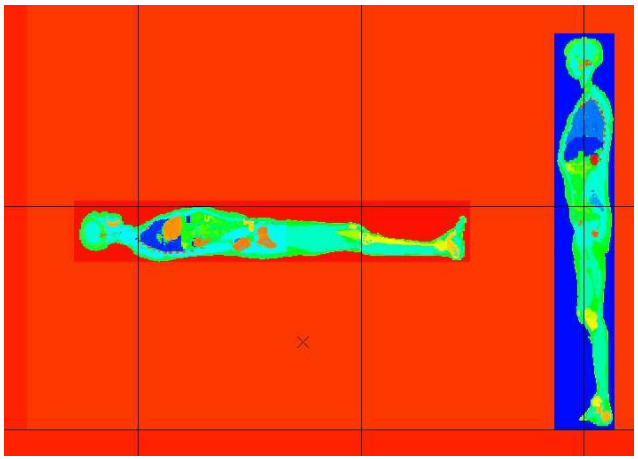


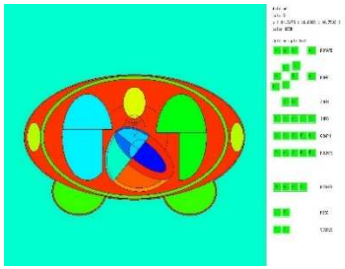










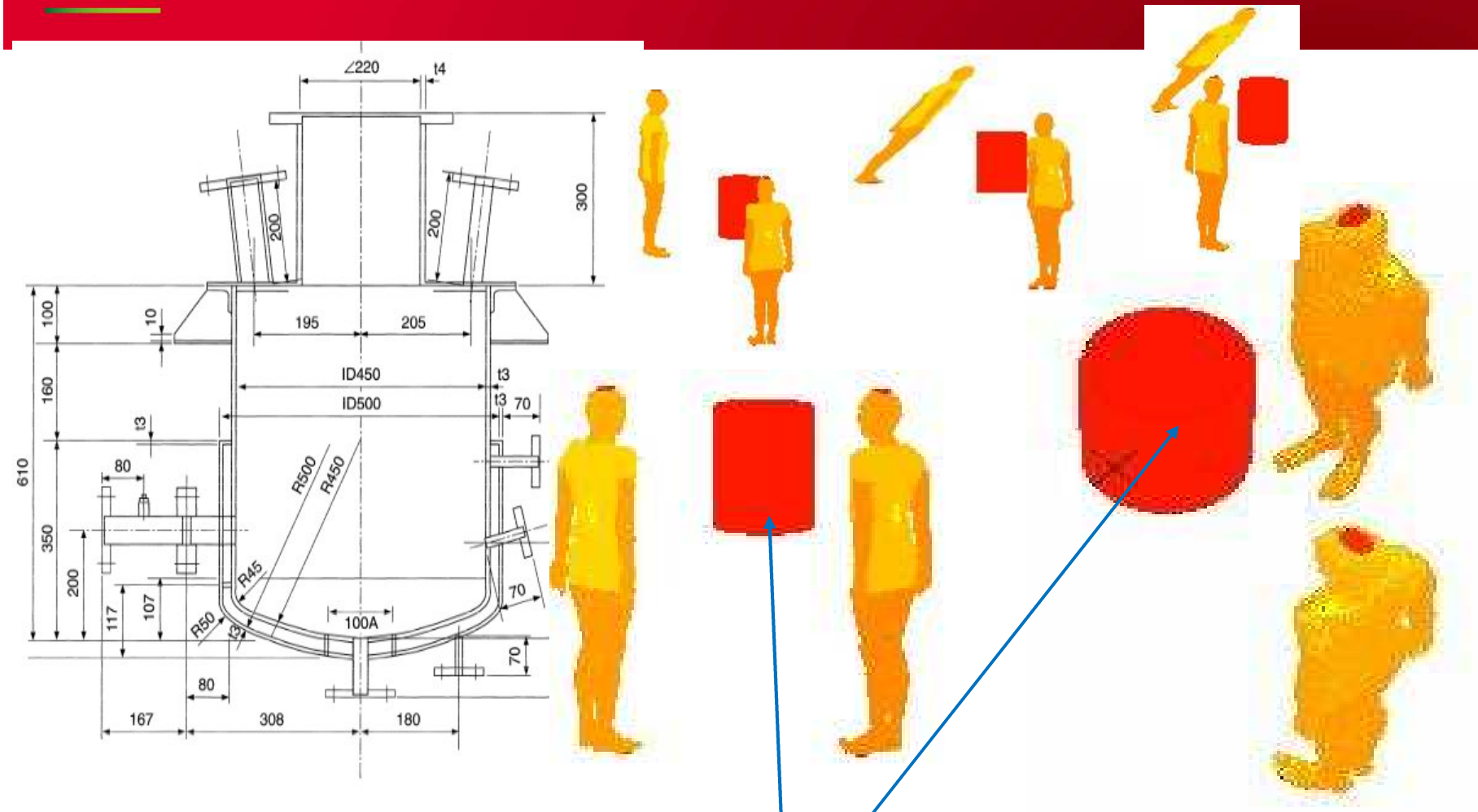


T4G tool ➔ 2D/3D display

TRIPOLI-4 ➔ PHANTOM option

Criticality accident victims ?

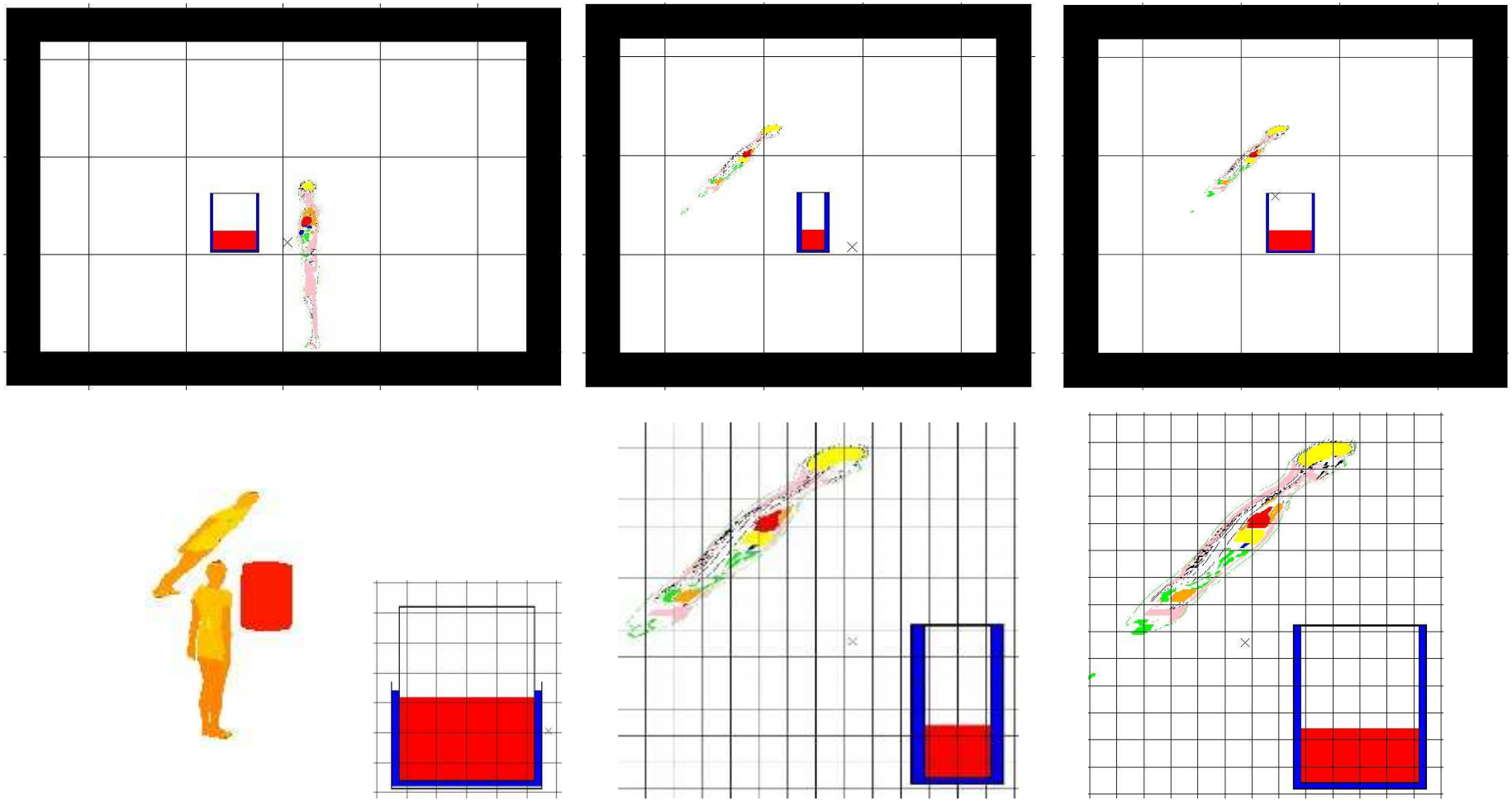
TOKAI-MURA CRITICALITY ACCIDENT VICTIMS – T4G



TRIPOLI-4 modeling of uranyl nitrate in tank + operators A and B

Ref: IAEA mission report, 1999. & LA-13638

TOKAI-MURA CRITICALITY ACCIDENT – T4G



TRIPOLI-4 modeling of fissile solution in tank & workers A and B

Ref: IAEA mission report, 1999. & LA-13638

CRITICALITY ACCIDENT - MATERIALS

$$U_3O_8 + 8HNO_3 \rightarrow 3UO_2(NO_3)_2 + 4H_2O + 2NO_2$$

Uranyl nitrate is a **water**-soluble yellow uranium salt with the formula $UO_2(NO_3)_2 \cdot n\,H_2O$ **n=3, 4, 6**

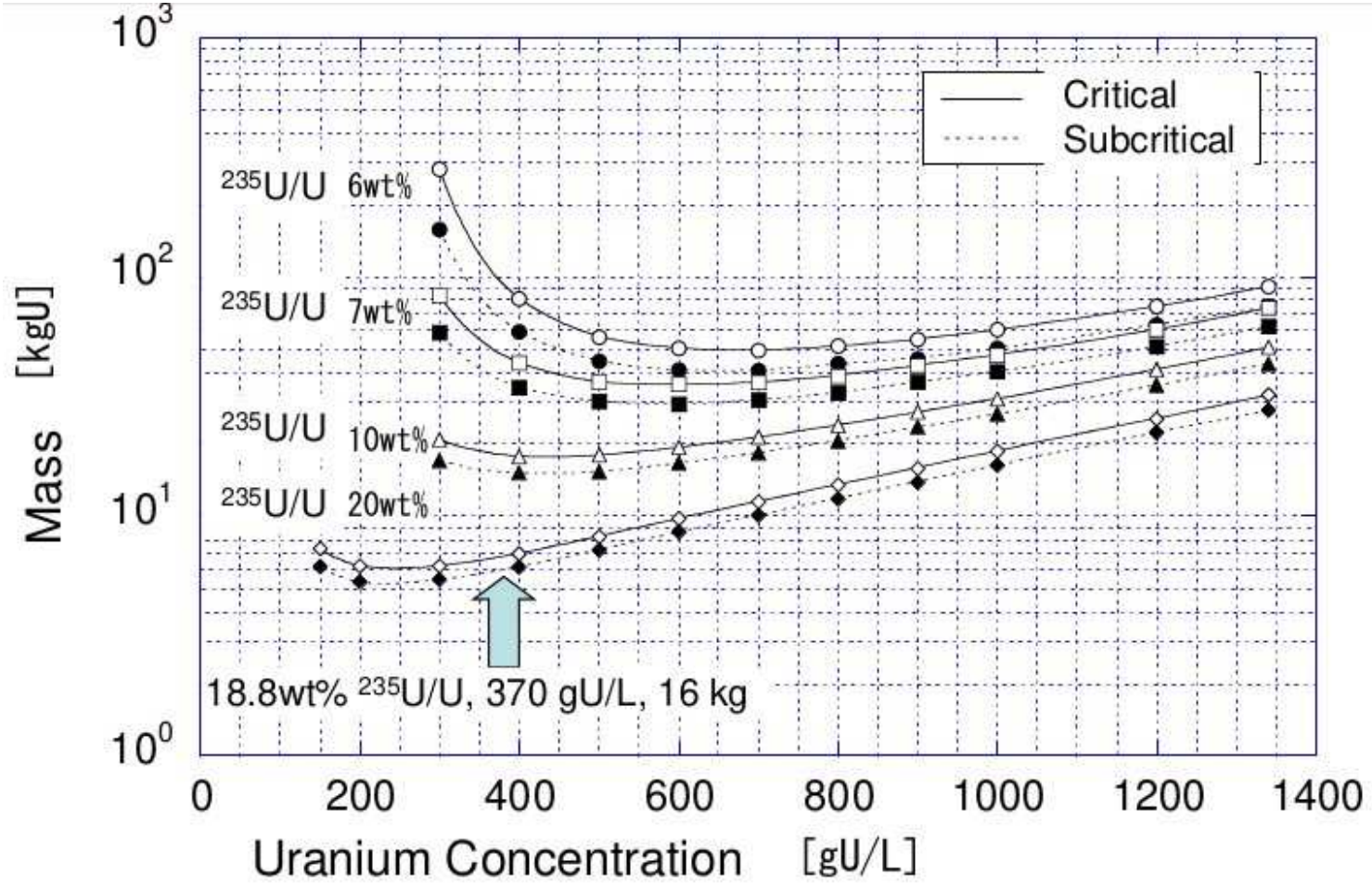
➔**Keff (H/U) ➔➔ H/U : 12 ➔ ??? (1M, 2M, …)**

➔**Tokai-Mura JCO solution ➔ 370 g U / L**

TRIPOLI-4 Volume and Mass verification

Ref: H. C. Paxton, LA-13638, 2000.
A. Endo, et al., J. Nucl. Sci. Tech. 40 (2003) 628

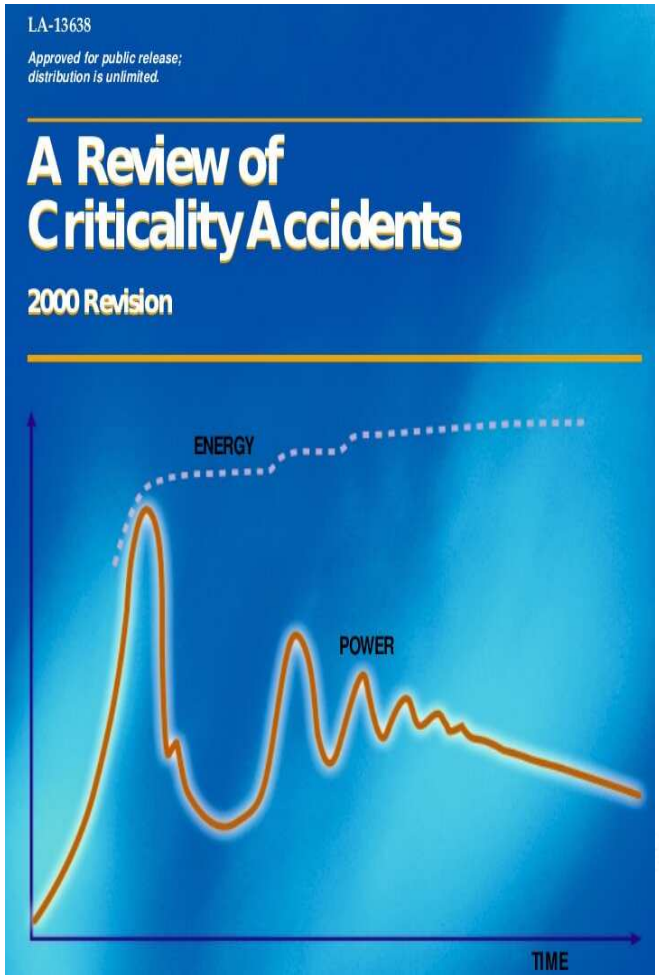
CRITICALITY ACCIDENT - MATERIALS



Criticality mass curves for uranium nitrate solution

Ref: H. Okuno, IAEA workshop, Chiba Japan,1-4 Oct. 2013

CRITICALITY ACCIDENT - CRITICALITY EXCURSION



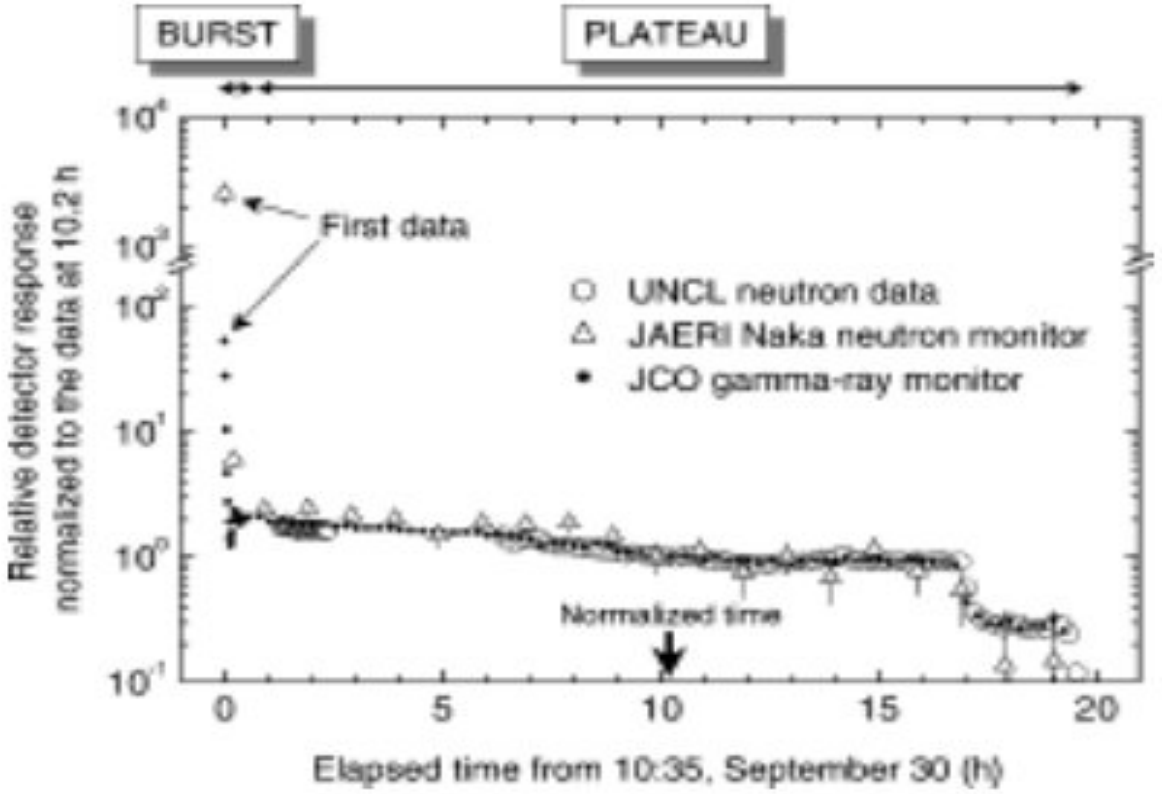
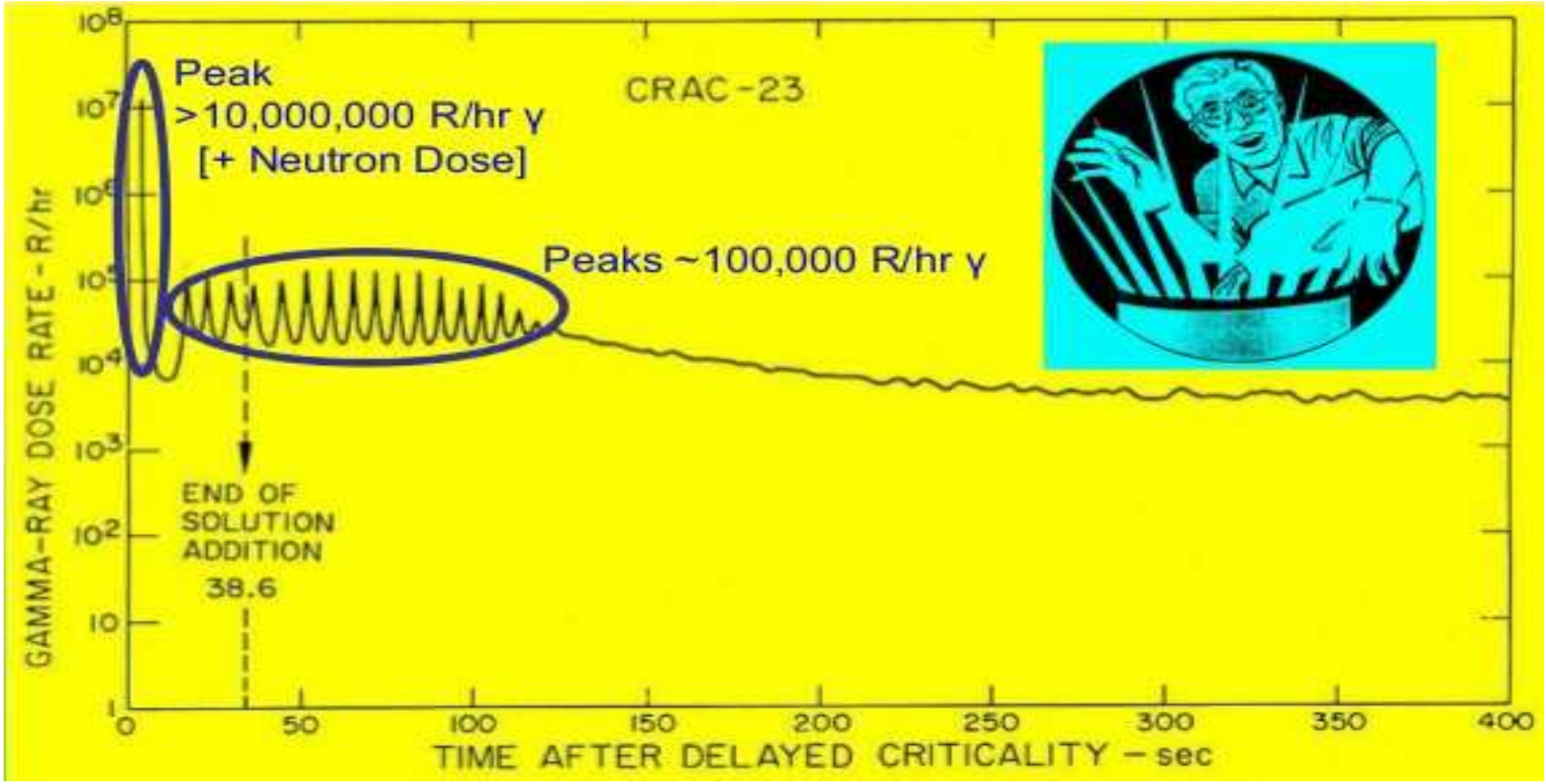


Fig. 3 Comparison of the time evolution of the counting rates measured with UNCL and dose rates measured with the neutron monitor and gamma-ray monitor

Fission power (t)
Ref: LA-13638

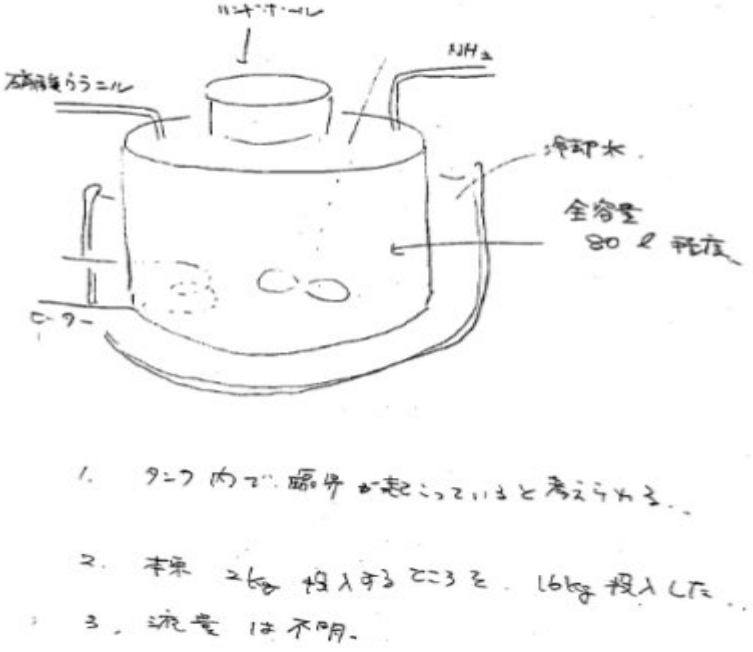
Neutron monitor count rate (t) ➔ 20 h
A. Endo, et al., J. Nucl. Sci. Tech. 40 (2003) 628

CRITICALITY ACCIDENT - CRITICALITY EXCURSION



CRiticality ACcidents in solution (CRAC and SILENE, CEA-Valduc)
Gamma dose rate (t) & Temperature feedback (heating – cooling)
Ref: F. Barbry, et al., Nucl. Sci. Eng. 161 (2009) 160

TOKAI MURA ACCIDENT - STOP CRITICALITY



- **Outline drawing of the precipitation tank and planning of countermeasures to stop criticality**
  1. **To draw cooling water outside of tank**
  2. **To inject neutron absorber into the tank**
  3. **Is it possible to inject from NH_3 line?**

- **Keff = Production / (Absorption + Leakage)**

Production ➔ Safety Mass & Volume control
Absorption ➔ Neutron poisons
Leakage ➔ Geometry, Reflection, Separation
P, A, L ➔ Enrichment (LEU, HEU, Pu)
➔ Moderation (H/U)

Ref: H. Okuno, IAEA workshop, Chiba Japan,1-4 Oct. 2013

- **Initial burst (blue flash reported) ➔**

1.2 E17 fissions	(J. A. Vazquez, Phys. Med. Biol. 59 (2014) 5277)
2.- 4.E17 fissions	(H. Okuno, JAEA, 2013, 10:35-11:00)
0.4 - 8.1E17 fissions	(R. A. Knief, Sandia Nat. Lab. 2013)
2.75 E17 fission ??	(A. Endo, JAEA, 2003 & 2010)

TRIPOLI-4 mode ➔ Criticality mode + Shielding mode

- **S (r, E, Ω, t) = C S(r) S(E) S(Ω) S(t)**

  Neutron source ➔ 1.2E17 * 2.43 = 2.92E17 n

  Gamma source ➔ 1.2E17 * 8.31 = 9.76E17 γ

  S(r) = S(R) * S(Z)

  S(E) : U-235 thermal fission for neutron & prompt gamma-rays

TRIPOLI-4 CALCULATION – H*(10) PROMPT GAMMA

TRIPOLI-4 Point flux FLUXPT position (cm)	Distance to the center of fissile solution (-50, 150, 118) (cm)	TRIPOLI-4 ICRP 74 (Sv)	MCNPX PMB (2014) Organ dose (Gy)
(x ,y, z)		Sigma < 2%	**35 (Liver)**
Radial direction – X (Outside of solution tank)			
(-80, 150, 90)	30	73.3	
(-100, 150, 90)	**50**	**34.2**	**> 30 (Gonads)**
(-120, 150, 90)	**70**	**20.0**	
Radial direction + X (Outside of solution tank & Phantom A present)			
(**0**, 150, 90)	**50**	**35.3**	**> 30 (Gonads)**
(10, 150, 90)	60	**20.8**	
(20, 150, 90)	**70**	**9.9**	

TRIPOLI-4 CALCULATION – ORGAN DOSE OF VICTIM A CENTER OF PHANTOM (25, 150, 88.8)

Target Organ	ICRP110 AM		TRIPOLI-4 (Gy)	MCNPX (Gy)		Ratio TRIPOLI-4 / MCNPX
	Organ ID	Mass (kg)		PMB (2014)	RM (2010)	
Prompt Fission Gamma rays						
Liver	95	1.8	14.5	35		**0.41**
SI wall	74	0.65	17.3		15 - 22	**1.15 - 0.79**
Prompt Fission Neutron						
Liver	95	1.8	9.6	7		**1.37**
SI wall	74	0.65	10.9		5 - 12	**2.18 – 0.91**
Secondary Gamma rays						
Liver	95	1.8	**25.5**	?		
SI wall	74	0.65	**25.6**		?	.

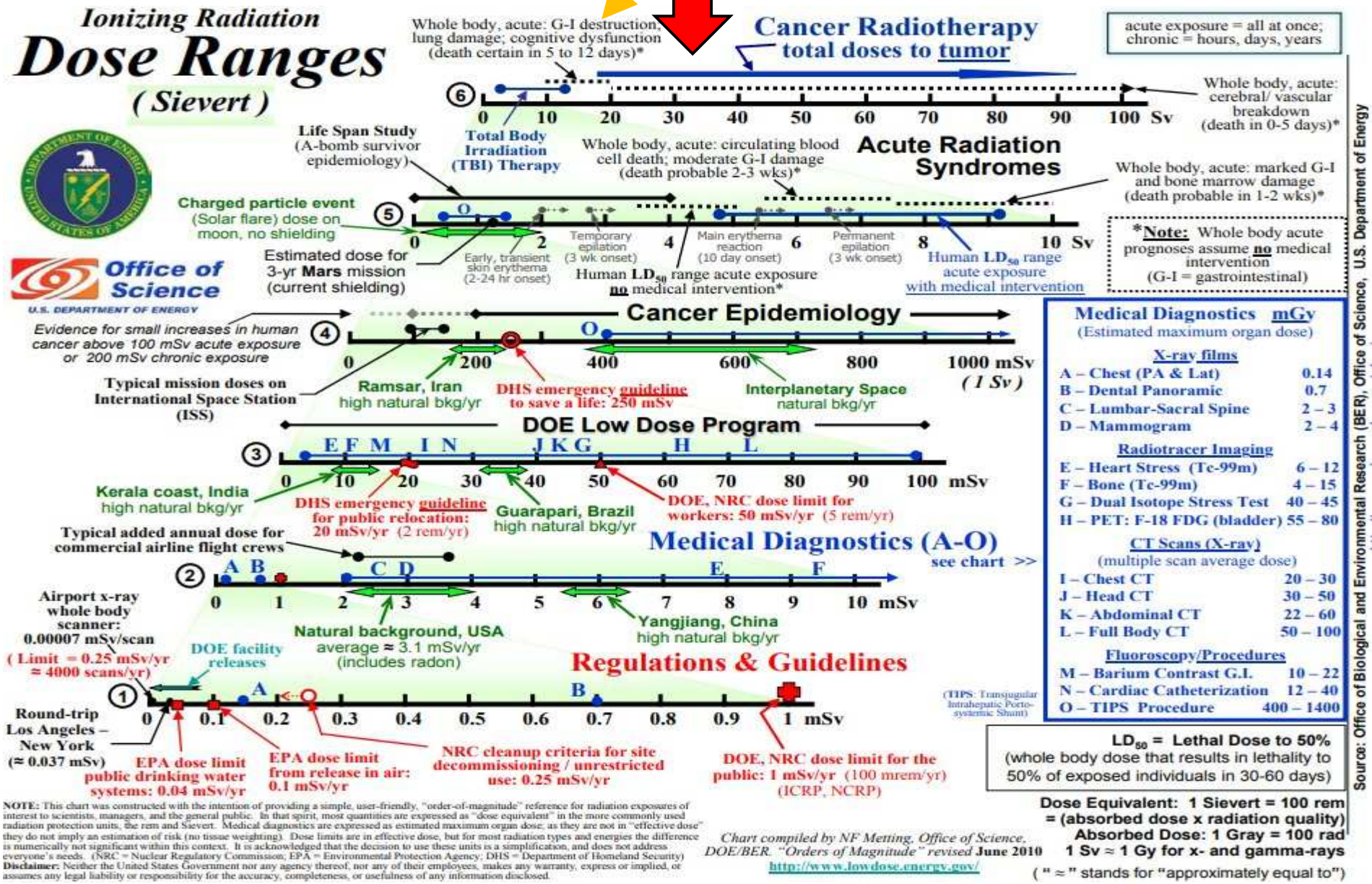
TRIPOLI-4 CALCULATION – ORGAN DOSE OF VICTIM A CENTER OF PHANTOM (5, 150, 88.8)

Target Organ	ICRP110 AM		TRIPOLI-4 (Gy)	MCNPX (Gy)		Ratio TRIPOLI-4 / MCNPX
	ID	Mass (kg)		PMB (2014)	RM (2010)	
Prompt Fission Gamma rays						
Liver	95	1.8	27.7	35		0.79
SI wall	74	0.65	29.3		15 - 22	1.95 - 1.33.
Prompt Fission Neutron						
Liver	95	1.8	17.2	7		2.46
SI wall	74	0.65	19.3		5 - 12	3,86 - 1.61
Secondary Gamma rays						
Liver	95	1.8	**49.3**	?		
SI wall	74	0.65	**46.6**		?	.

CONCLUSIONS

- The TRIPOLI-4 radiation dose study was successfully performed using two ICRP 110 voxel phantoms for the victim workers of 1999 Tokai-Mura criticality accident.
- The T4G display tool was useful to debug and to navigate the TRIPOLI-4 models of voxel phantoms including millions voxels.
- To improve the organ dose calculation efficiency, an optimization of TRIPOLI-4 input has been performed and a new route in TRIPOLI-4 has been developed.
- TRIPOLI-4 calculation results of prompt fission neutron are generally close to the published neutron organ doses.
- For the next steps both the refinement of modeling uncertainties and the sensitivity study of different parameters are necessary.

TOKAI-MURA CRITICALITY ACCIDENT 1999



CRITICALITY ACCIDENT IN TOKAI-MIRA 1999

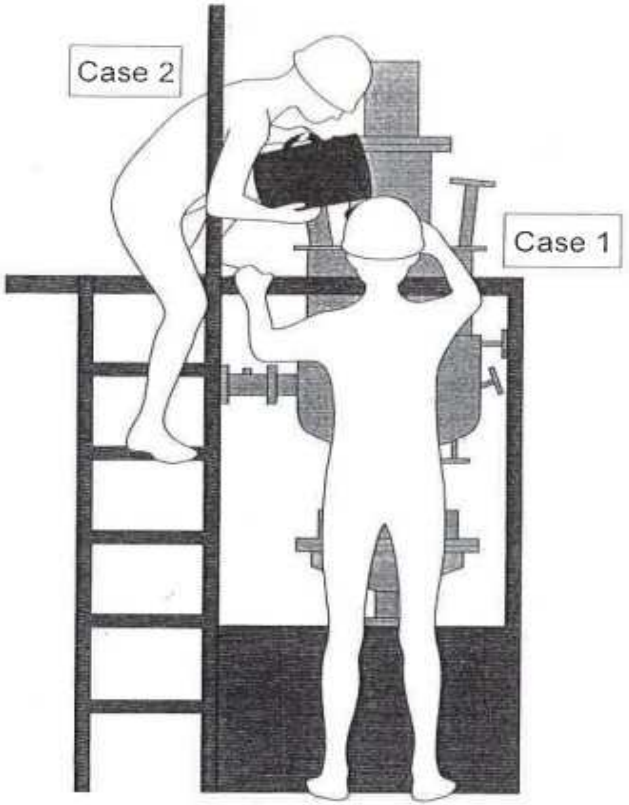


Fig. 1. Schematic diagram of the precipitation tank and likely positions of two victims.

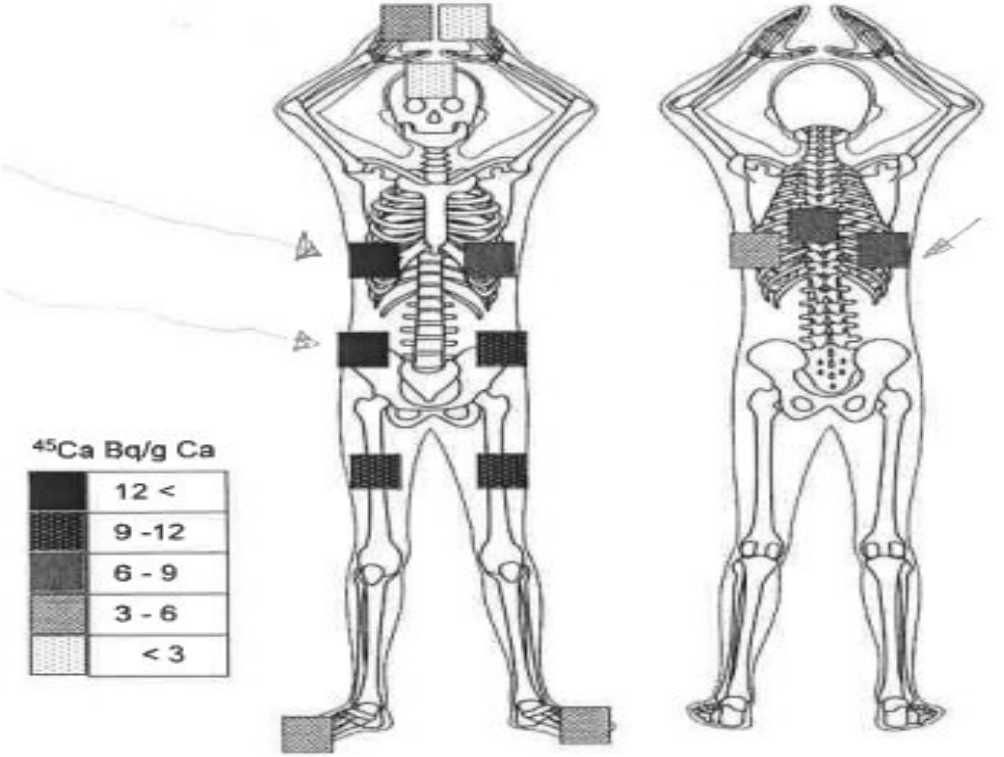


Fig. 4. Illustration of Case 1's orientation assumed at the moment of the burst. Cubes show ^{45}Ca activity level of the spots from which bones were taken.

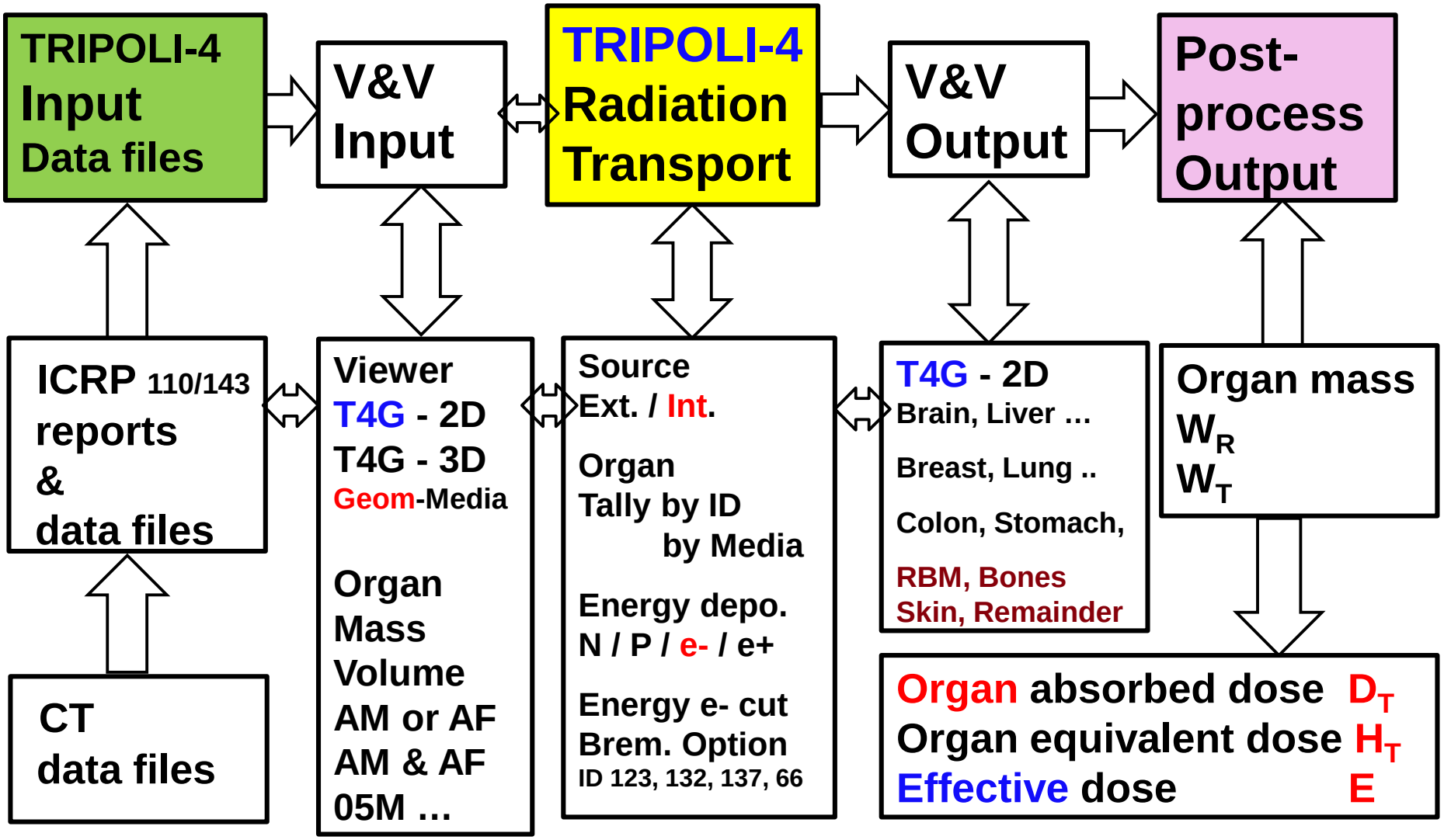
Neutron activation of ^{44}Ca in bone samples

➔ ^{45}Ca Bq/g Ca ➔ (Right iliac bone / Left iliac bone) = 1.7

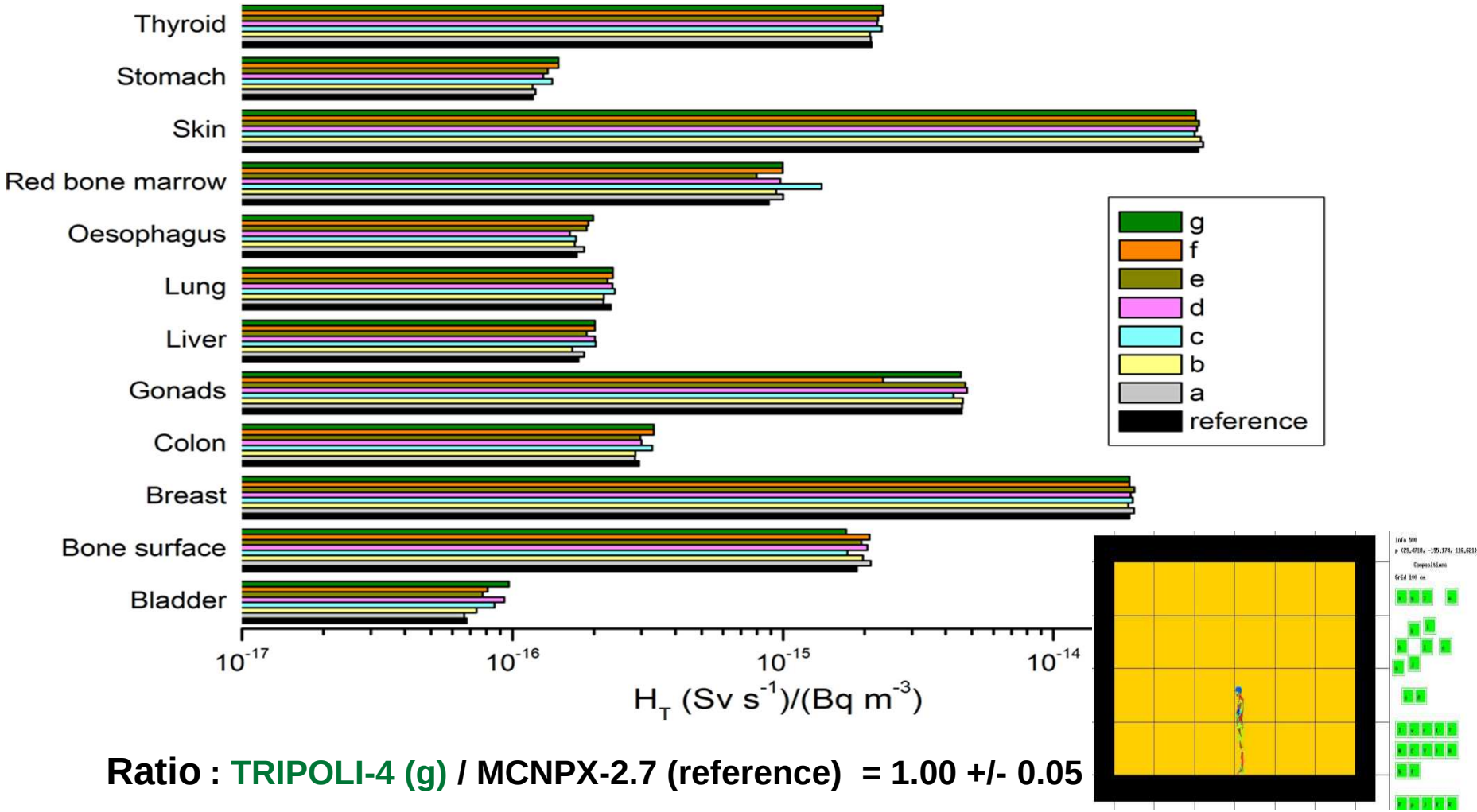
➔ Self-shielding of human body ➔ anterior & posterior ribs

Ref: K. Miyamoto et al. Health Physics, 83 (2002) 19.

TRIPOLI-4 ORGAN DOSE CALCULATION USING VOXEL PHANTOMS



VALIDATION OF TRIPOLI-4 CALCULATIONS (WG6-TASK 4)
VOXEL PHANTOM IMMERSED IN N-16 CONTAMINATED AIR



Ratio : TRIPOLI-4 (g) / MCNPX-2.7 (reference) = 1.00 +/- 0.05

Ref.: J.M. Gomez, … Y.-K. Lee, Radiation Measurements, 145 (2021) 106612.