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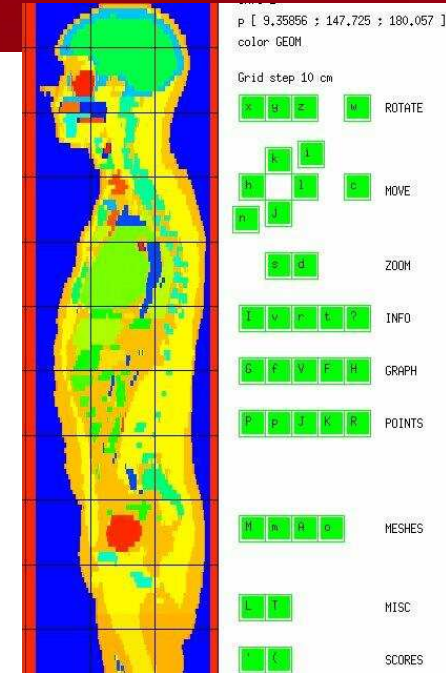
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CODE DE CALCUL TRIPOLI-4[®] - APPLICATION EN DOSIMÉTRIE POUR LES DEUX VICTIMES DE L'ACCIDENT DE CRITICITÉ DE TOKAI-MURA (1999)

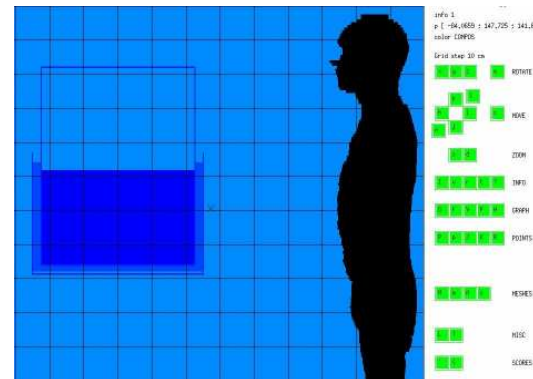
Yi-Kang Lee

**CEA-Saclay, SERMA
France**

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



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



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.

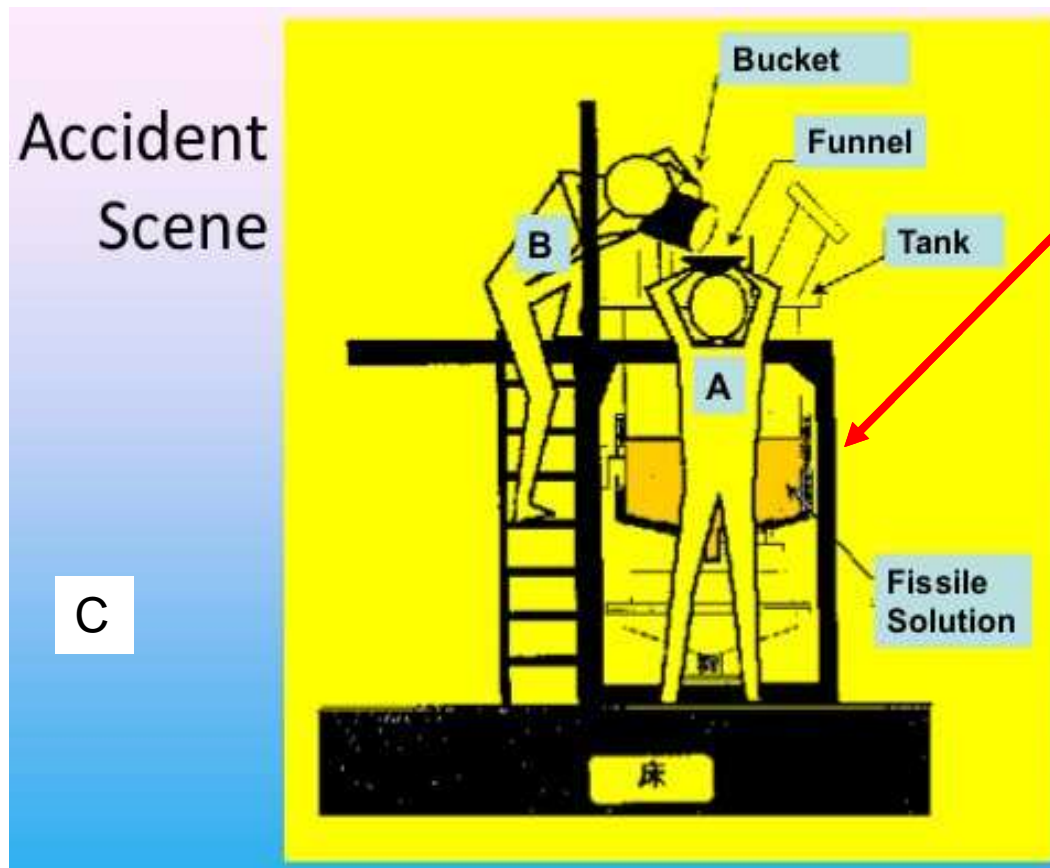
It uses continuous-energy nuclear data to simulate neutron, photon, electron and positron transport in fields like **radiation protection and shielding**, fission reactor physics, and **nuclear criticality safety**.

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**



Table 5

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

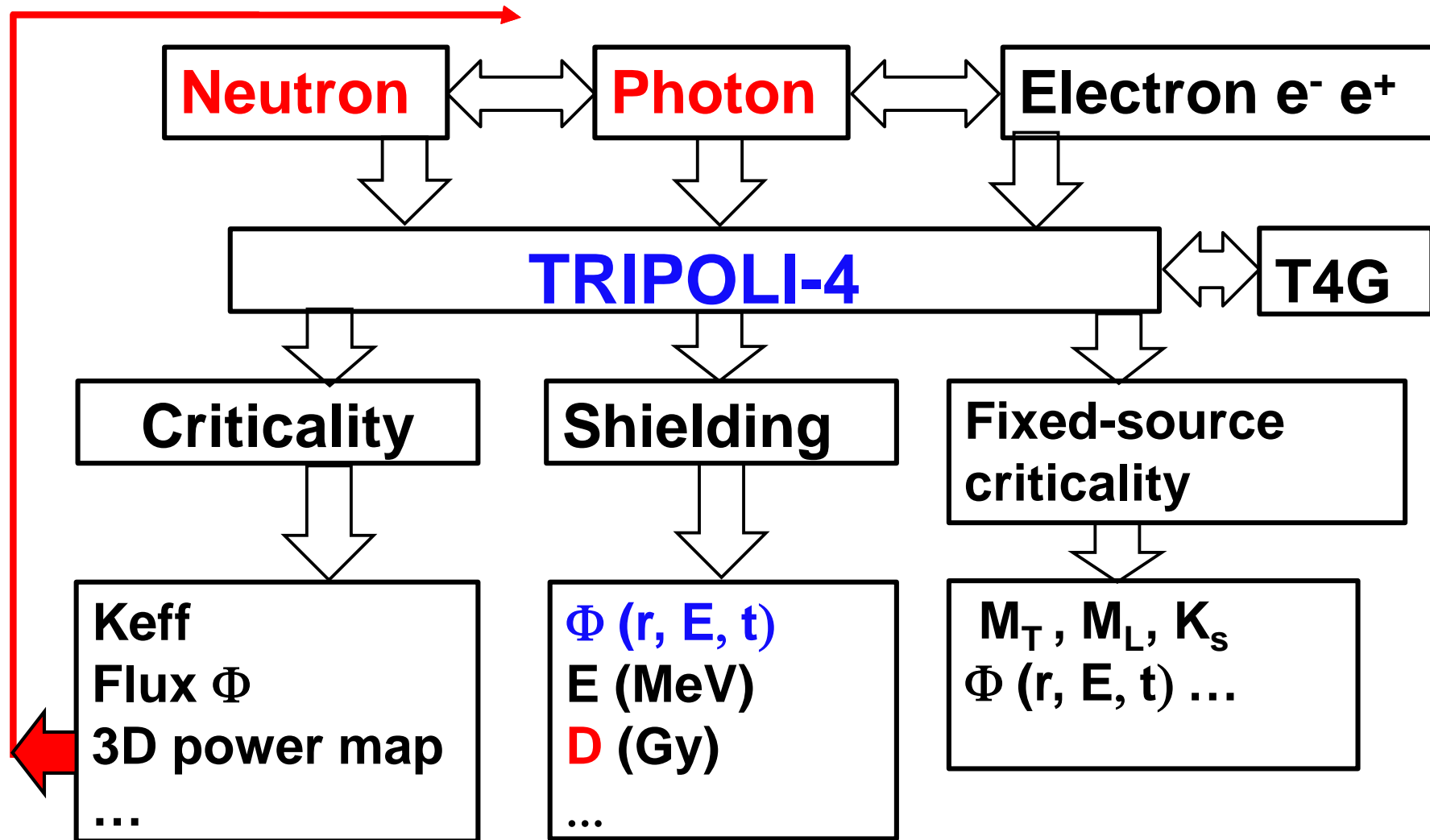
Method	Worker		
	A	B	C
(1) Prodromal symptoms	Over 8	4–6 or over 6	Less than 4
(2) Lymphocyte counts	16–23	6–8	1–5
(3) Chromosome analysis	16 to over 30	6.9–10	2.8–3.2
(4) ^{24}Na in the body			
1) Specific activity of ^{24}Na in blood (Neutron, γ -ray: Gy)	(5.4, 9.9)	(2.9, 4.1)	(0.81, 1.5)
Total dose (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

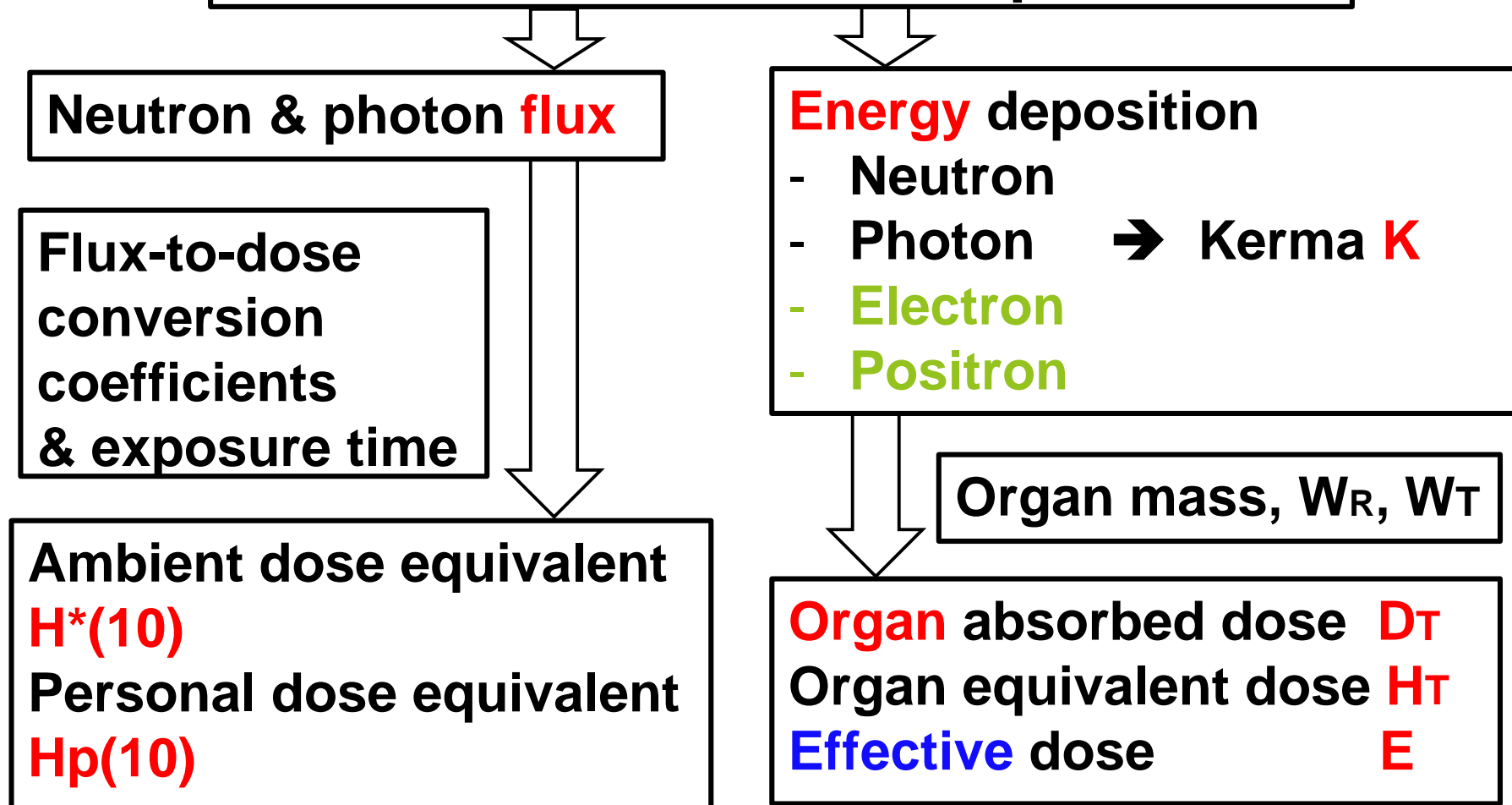
→ High dose estimation: Biodosimetry

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

TRIPOLI-4 Radiation transport code

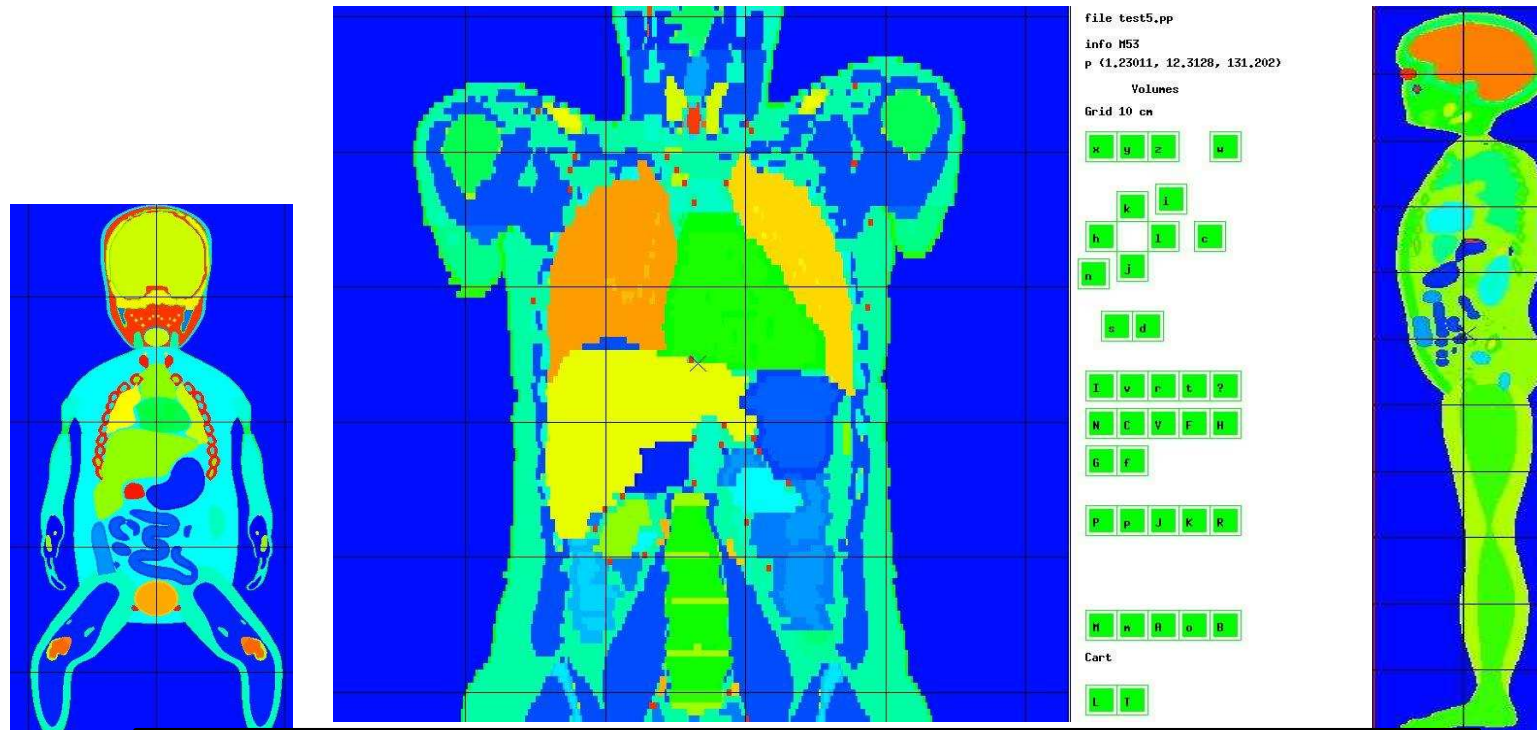


Ref.: Y.-K. Lee, J. of Nucl. Eng. and Rad. Sci., Oct. 2020 / 041105-3

- **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**

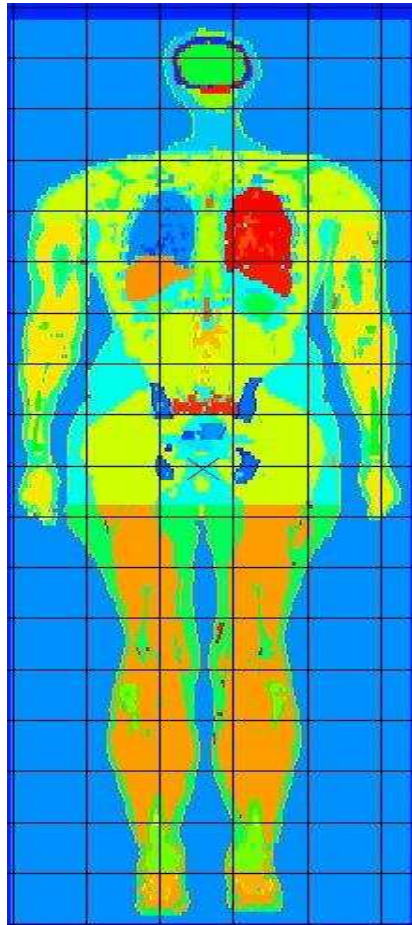


TRIPOLI-4 phantom models: 00F, AF, 05M

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

2020 ICRP 143 **Pediatric Reference phantoms (>200 MB)**

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



```
151 79 155
152 79 155
153 79 155
154 79 155
155 79 155
156 79 155
157 79 155
158 79 155
148 80 155
149 80 155
150 80 155
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161 80 155
146 81 155
147 81 155
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
165 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 mm³

PHANTOM 10000

`AM_organ.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

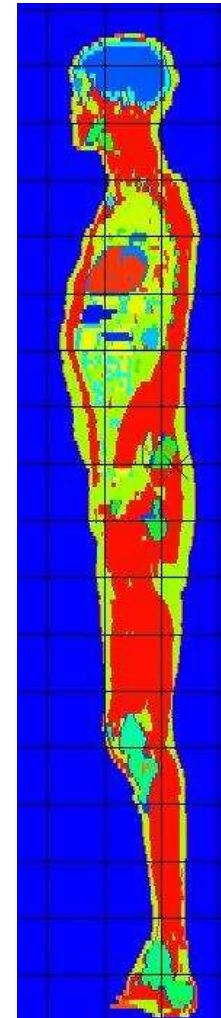
1000 2000 Mat // Increment for organs' names

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

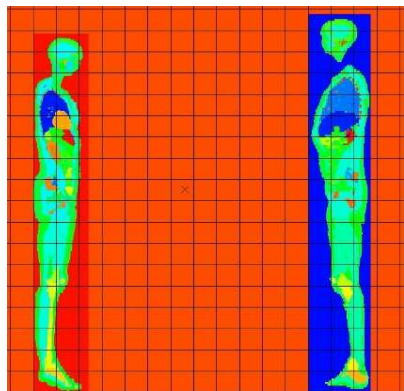
PHANTOM 10000
AM_organs.dat AM.dat

DIMENSION
254 127 222
0.2137 0.2137 0.8

PLUS
1000 2000 Mat

FRAME
27.14 13.57 88.8
1. 0. 0.
0. 1. 0.
0. 0. 1.

ENDP

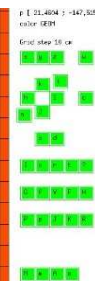
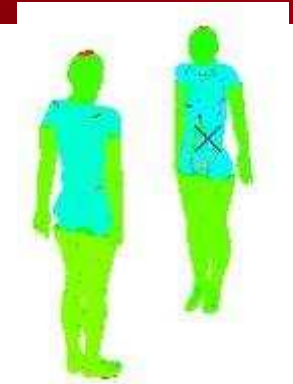


PHANTOM 20000 // Two voxel phantoms
AF_organs.dat AF.dat // ICRP 110 data files

DIMENSION
299 137 348 // Voxel 3D array size
0.1775 0.1775 0.484 // Voxel dimensions (cm)

PLUS
1000 2000 Mat // Increment for organs' names
// Ex: Liver 11095 (AM) & 21095 (AF)

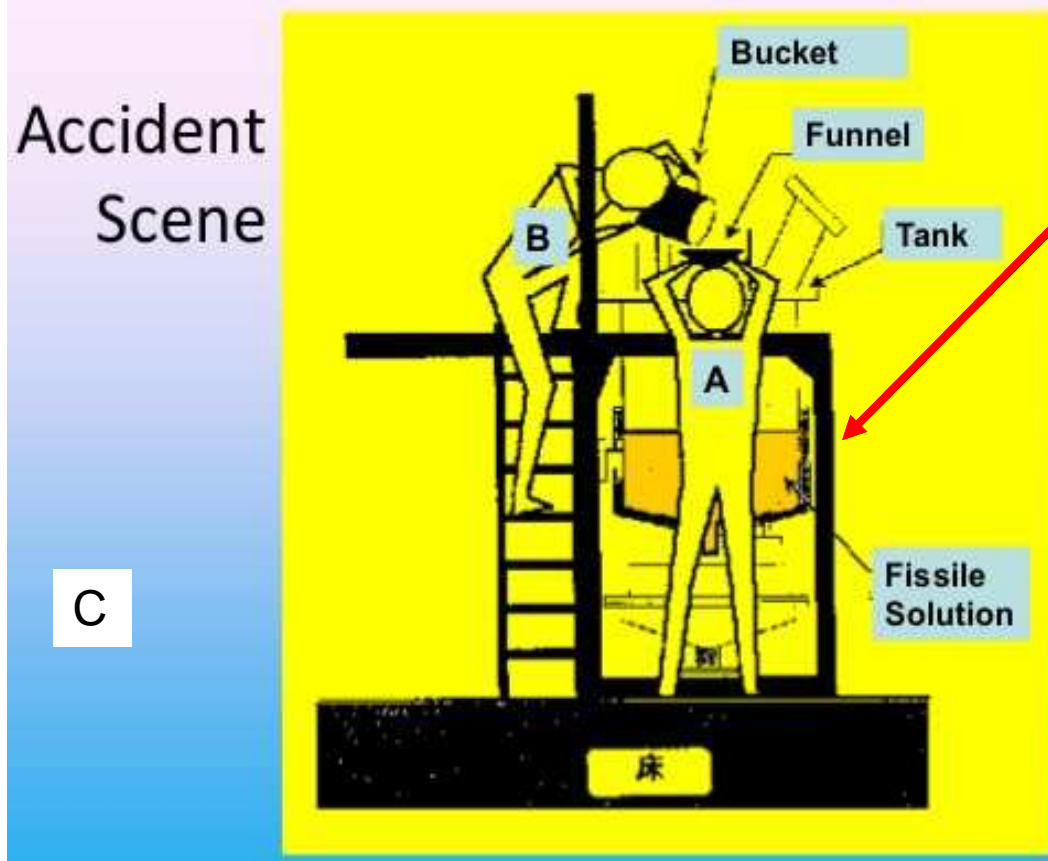
FRAME
26.536 -108. 84.216 // (x, y, z) in cm & center of 3D array
-1. 0. 0. // X-axis of phantoms AM & AF
0. -1. 0. // Unit vector of Y-axis
0. 0. 1. // Two **face-to-face standing phantoms**
// With 1 m distance at chest level



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: ²⁴Na in blood, Biodosimetry

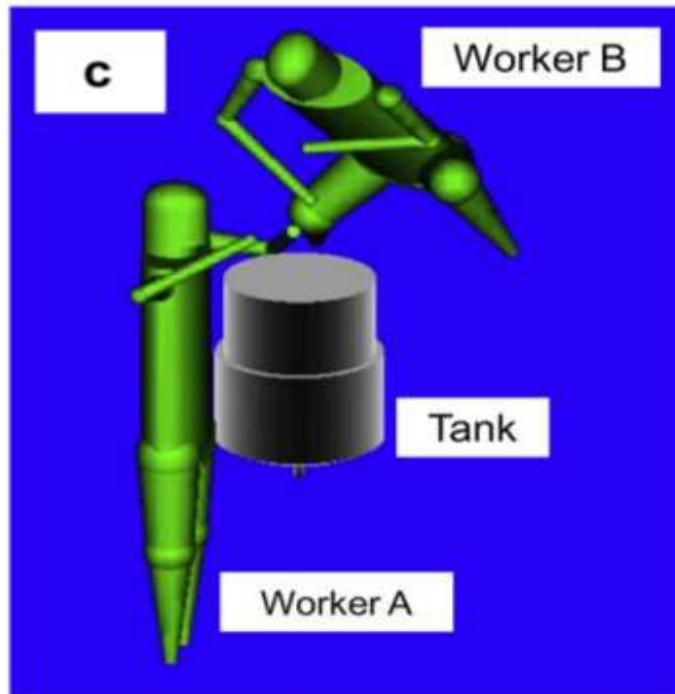


Fig. 4. Procedures for establishing simulation model.

- 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

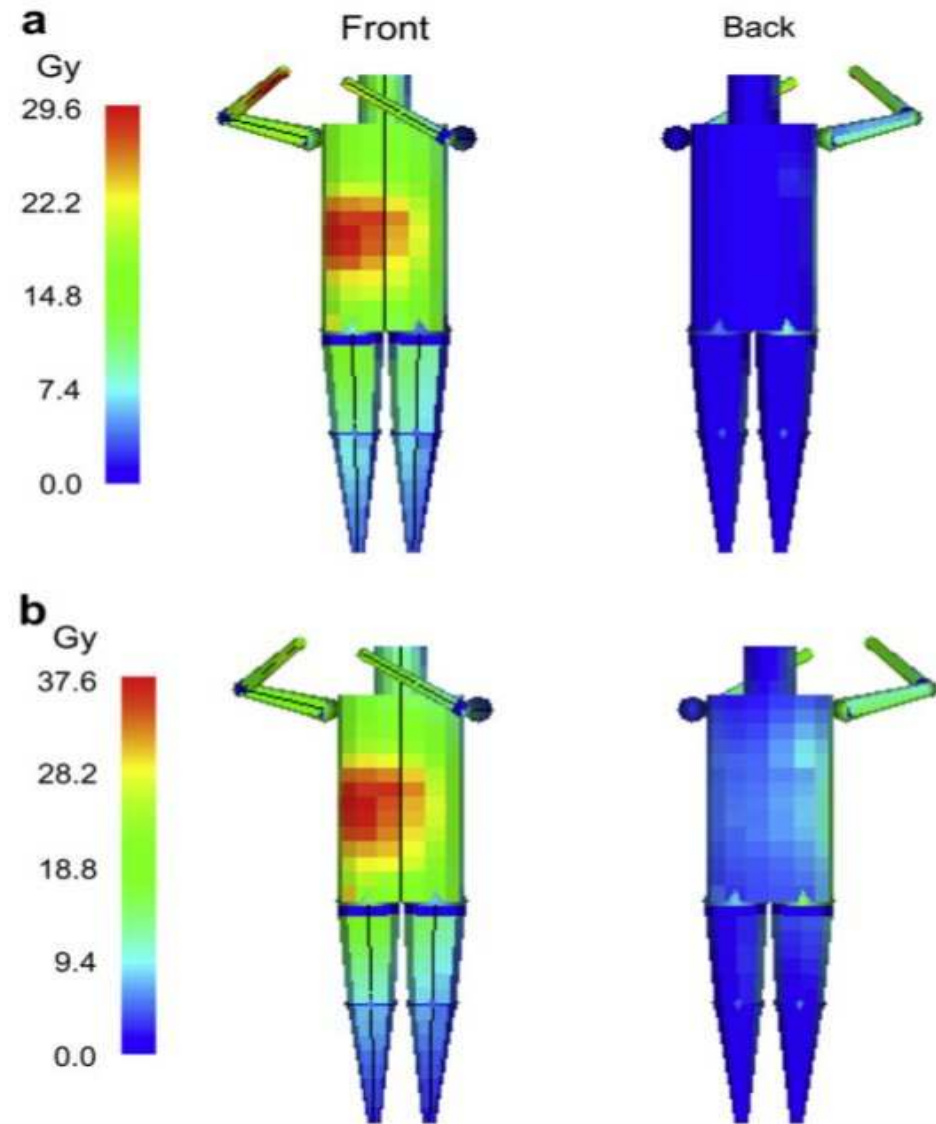


Fig. 5. Absorbed dose distribution in the skin of Worker A. (a) Neutrons, (b) γ -rays.

Phys. Med. Biol. 59 (2014) 5277

J A Vazq

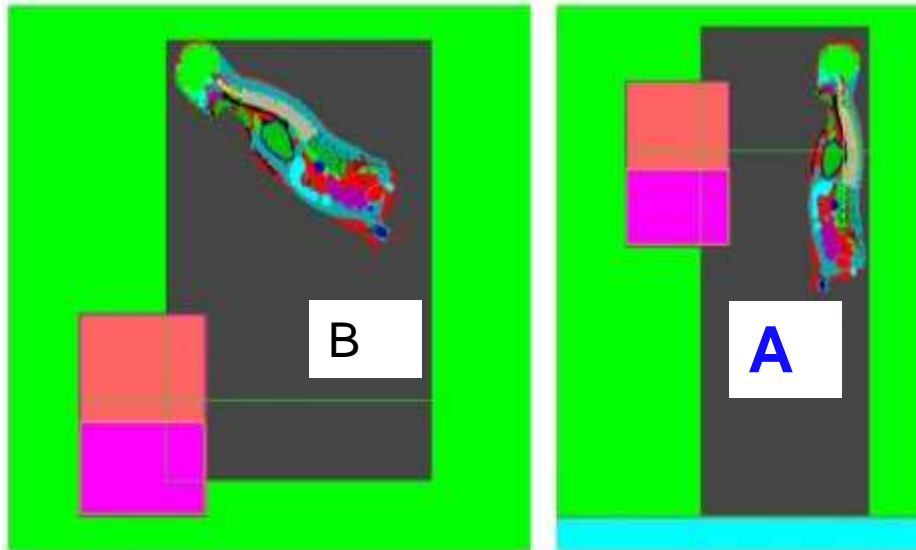


Figure 2. Cross section view of the crouching (left) and standing (right) phantom in the MCNPX simulation. Different colors represent different organs.

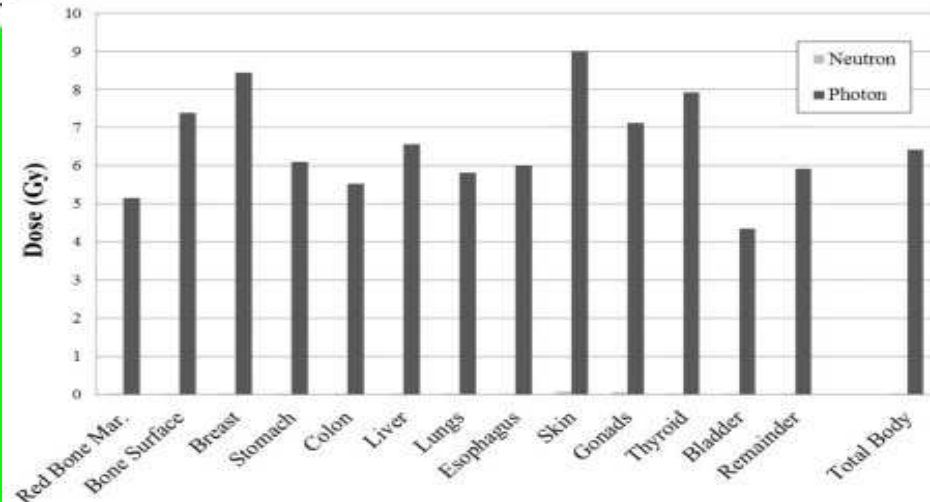
- Only one phantom in simulation
- No water layer around tank
- Organ dose: neutron < gamma
- Small neutron dose for B

Ref: J. A. Vazquez et al.

Phys. Med. Biol., 59 (2014) 5277

Phys. Med. Biol. 59 (2014) 5277

J A Vazquez et al



A: Liver → 7 Gy (N), 35 Gy (γ)

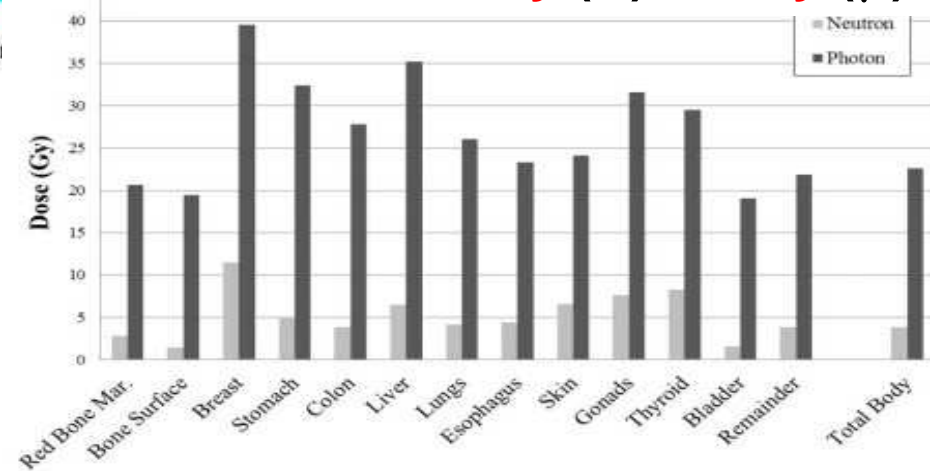
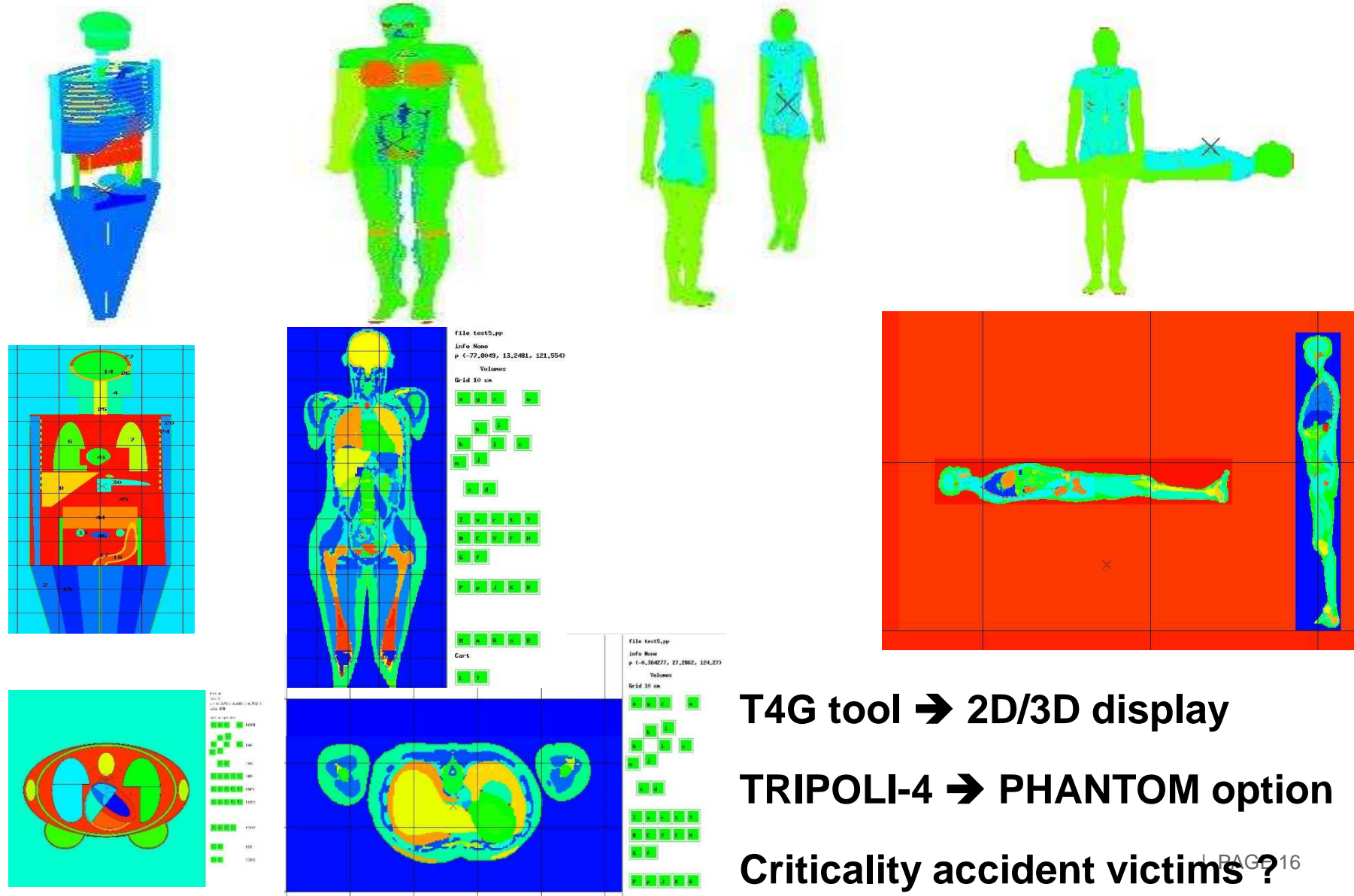


Figure 4. Calculated organ doses for the crouching-posture worker (top) and the standing-posture worker (bottom) for selected radiation-sensitive tissues.



TRIPOLI-4 – T4G - MIRD AND VOXEL PHANTOMS

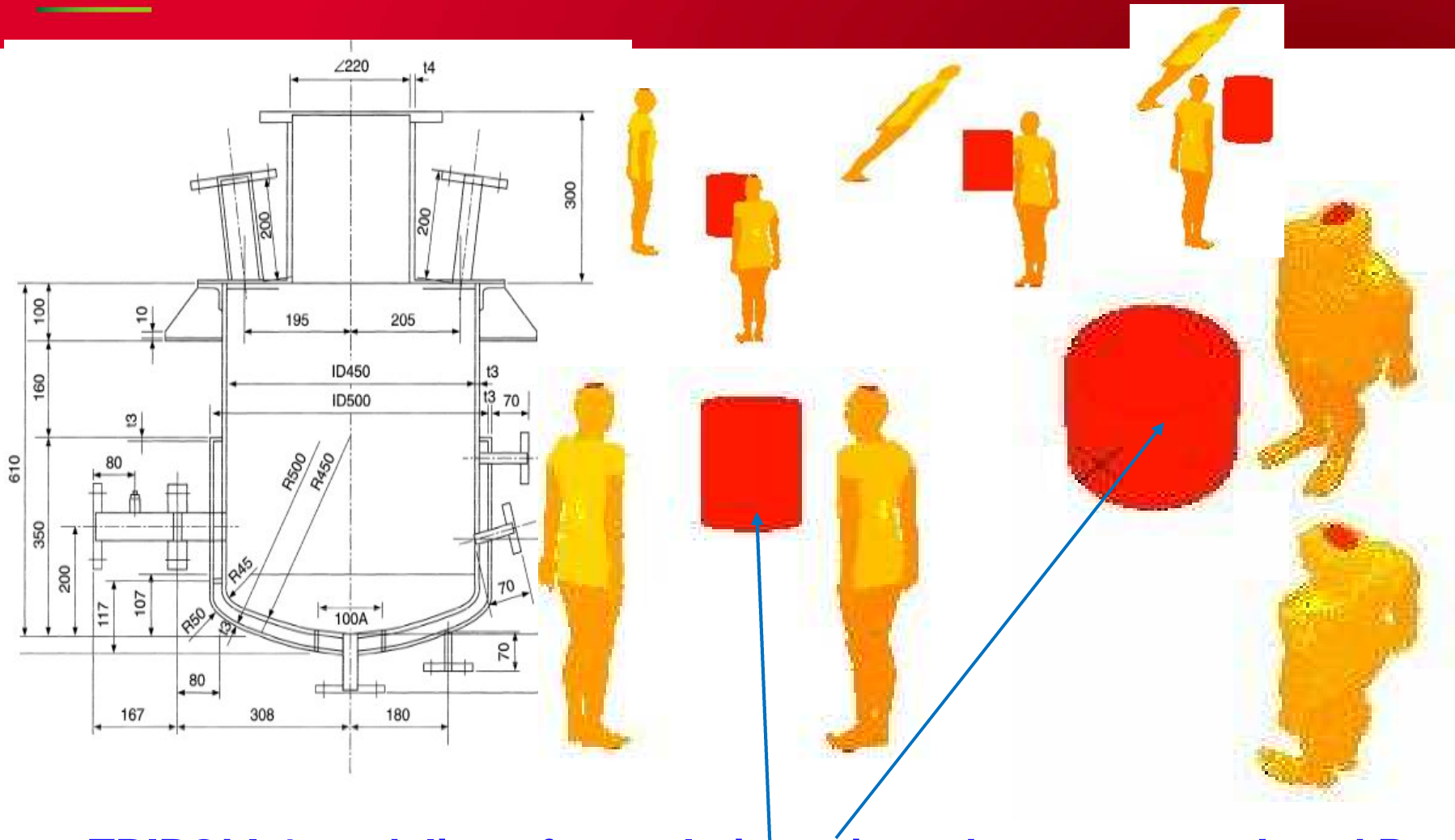


T4G tool → 2D/3D display

TRIPOLI-4 → PHANTOM option

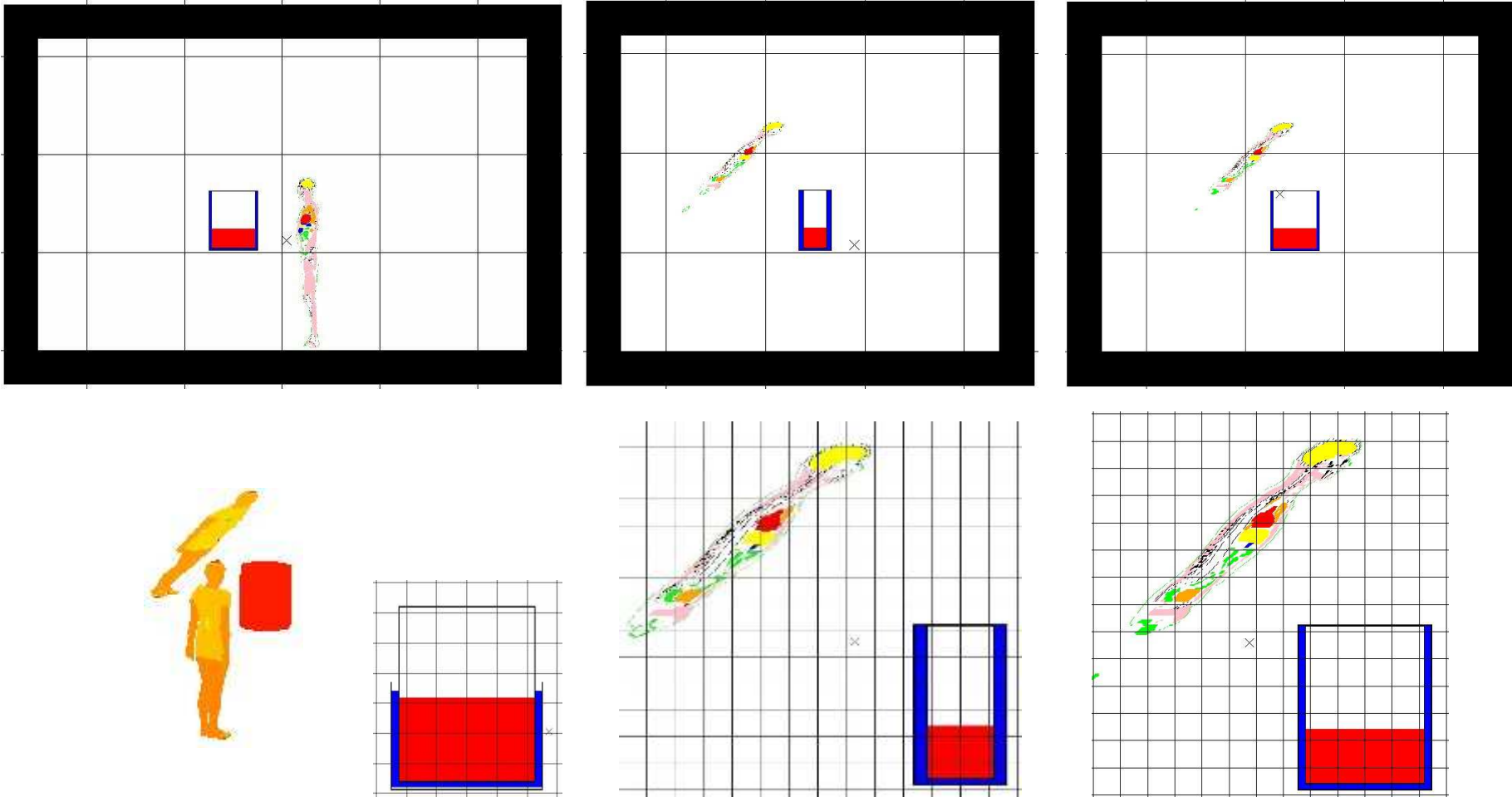
Criticality accident victims ?

TOKAI-MURA CRITICALITY ACCIDENT VICTIMS – T4G



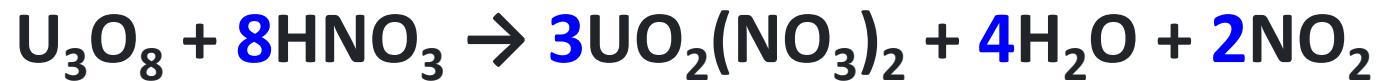
TRIPOLI-4 modeling of uranyl nitrate in tank + operators A and B

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



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

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



Uranyl nitrate is a **water**-soluble yellow uranium salt with the formula $\text{UO}_2(\text{NO}_3)_2 \cdot n \text{H}_2\text{O}$ $n=3, 4, 6$

→ $K_{\text{eff}}(\text{H}/\text{U}) \leftrightarrow \text{H}/\text{U} : 12 \rightarrow ??? (1\text{M}, 2\text{M}, \dots)$

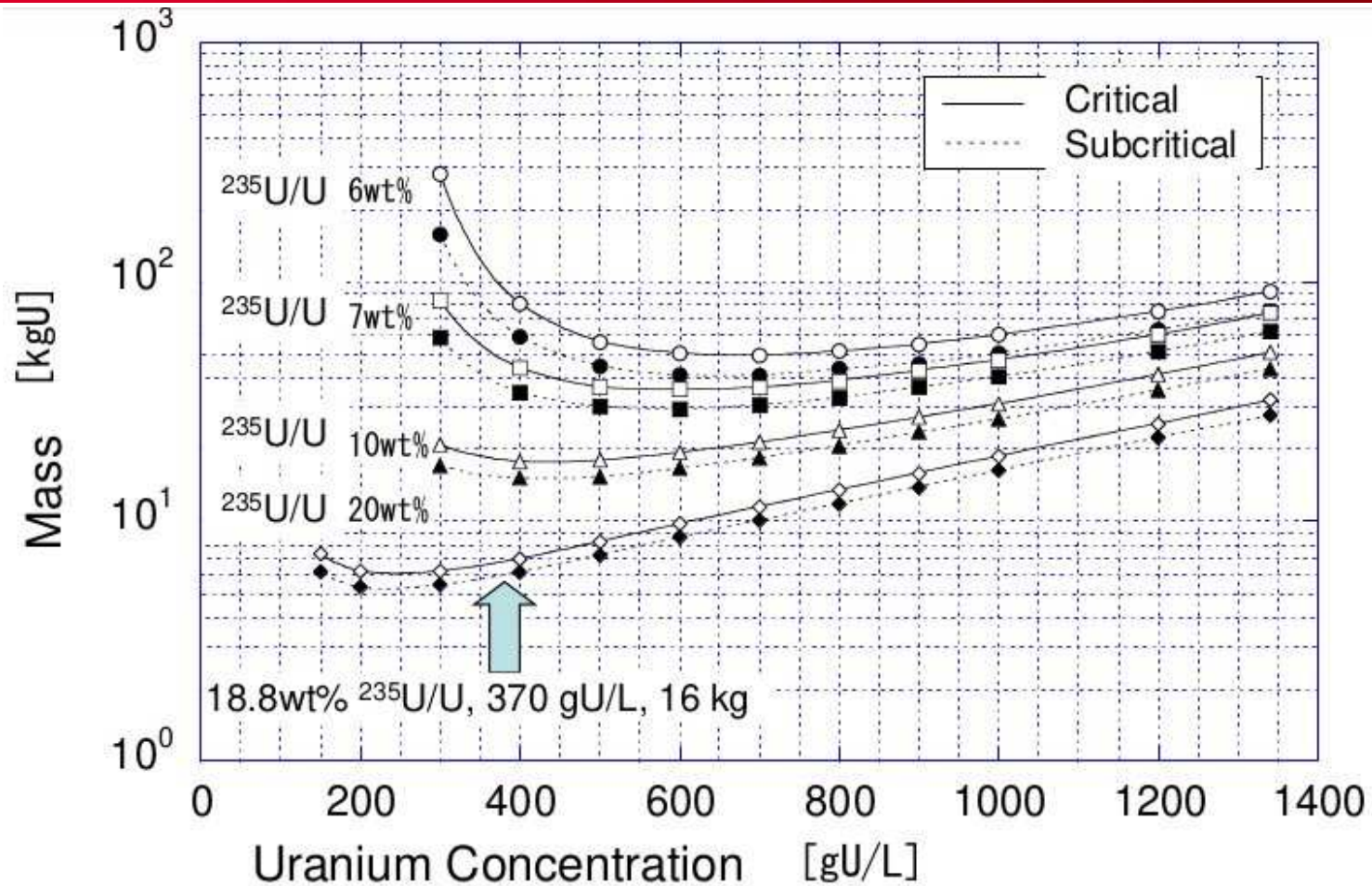
→ Tokai-Mura JCO solution → **370 g U / L**

TRIPOLI-4 **Volume** and **Mass** verification

Ref: H. C. Paxton, LA-13638, 2000.

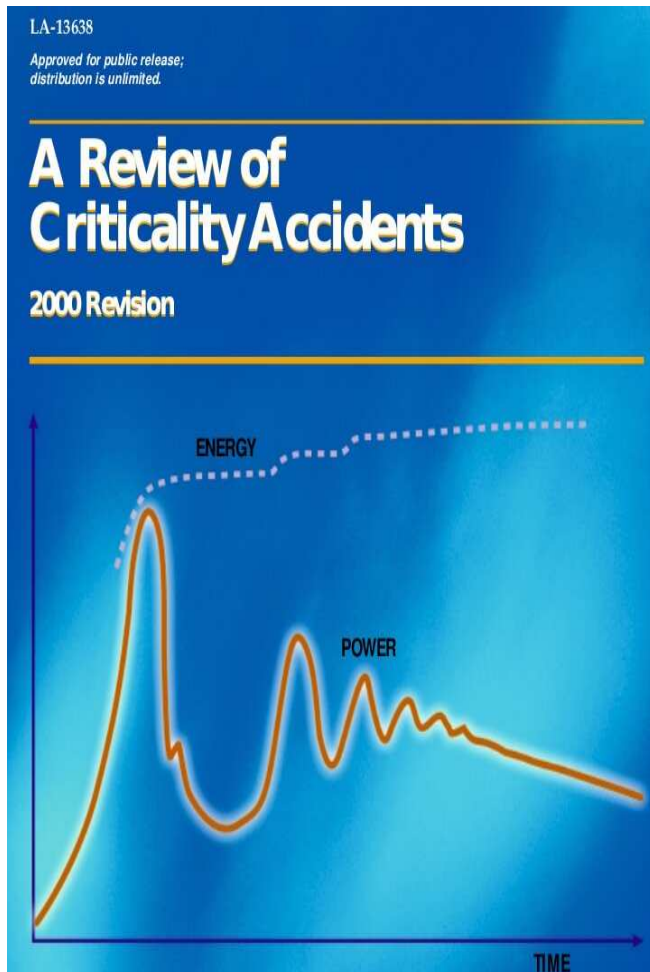
A. Endo, et al., J. Nucl. Sci. Tech. 40 (2003) 628

CRITICALITY ACCIDENT - MATERIALS



Criticality mass curves for uranium nitrate solution

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



Fission power (t)

Ref: LA-13638

Y.-K. Lee

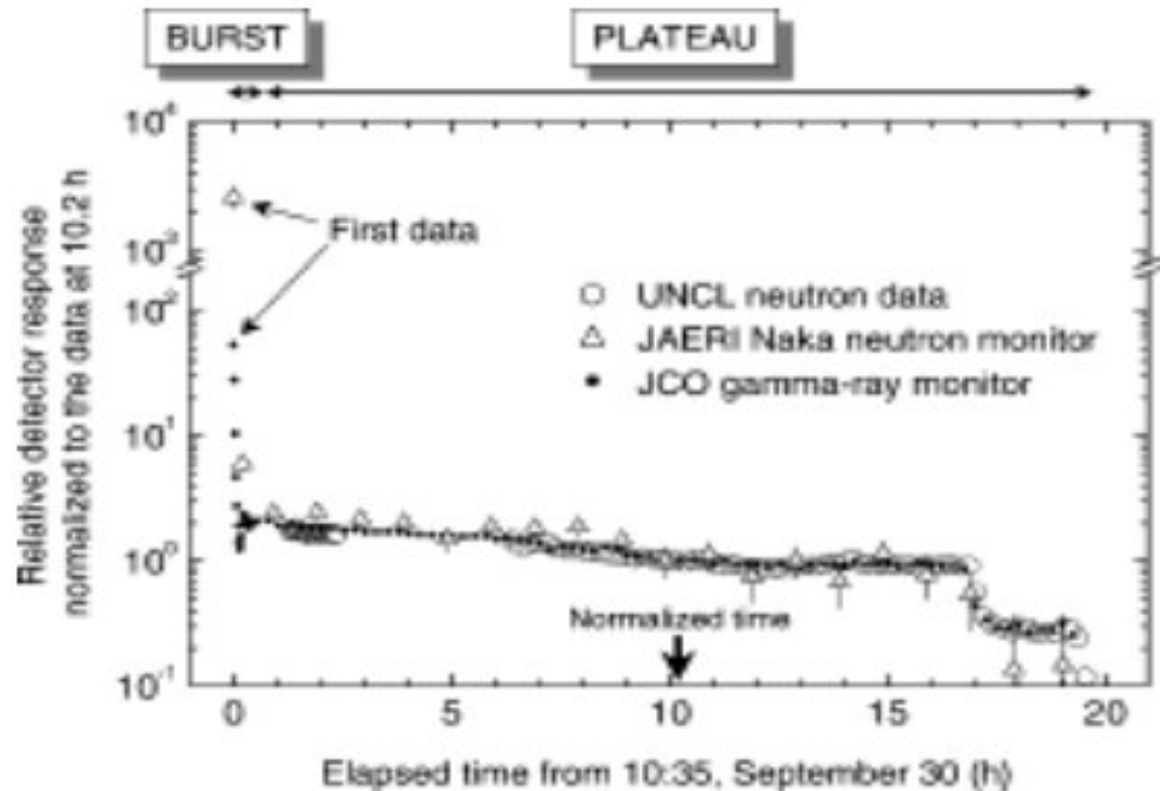
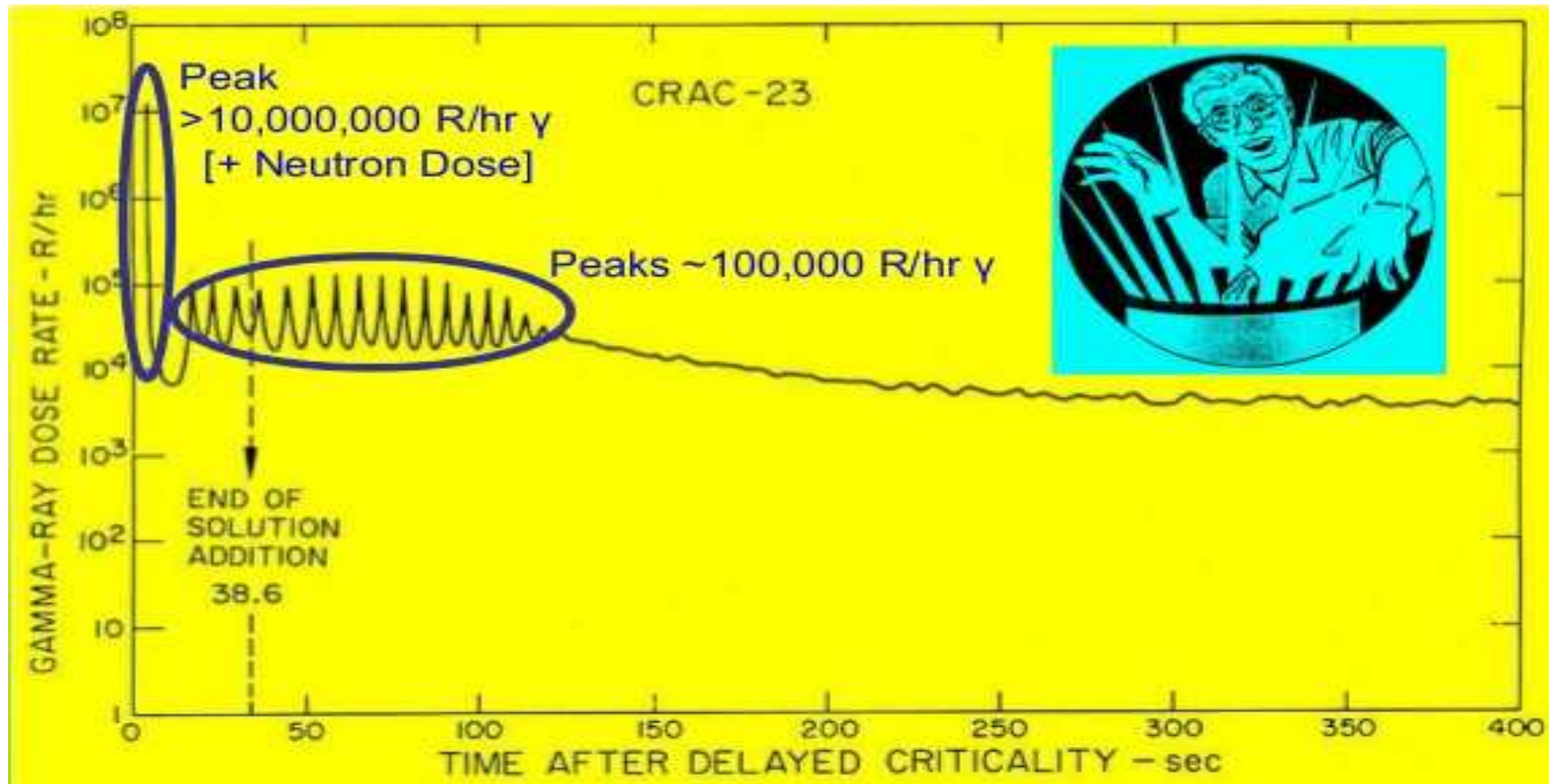


Fig. 3 Comparison of the time evolution of the counting rates measured with UNCL and dose rates measured with the neutron monitor and gamma-ray monitor

Neutron monitor count rate (t) → 20 h

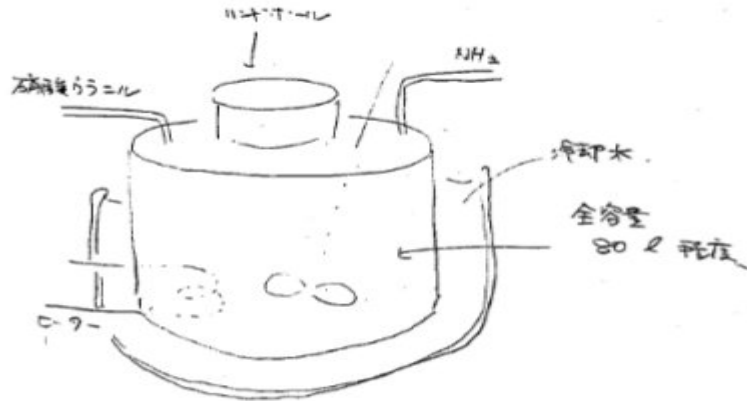
A. Endo, et al., J. Nucl. Sci. Tech. 40 (2003) 628

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



1. 9-7 内での漏れが起るとは考えられず...
2. 本来 2kg 投入するところ、16kg 投入した...
3. 流量は不明。

- **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 NH₃ line?

■ **$K_{eff} = \text{Production} / (\text{Absorption} + \text{Leakage})$**

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

■ $S(r, E, \Omega, t) = C S(r) S(E) S(\Omega) S(t)$

Neutron source → $1.2E17 * 2.43 = 2.92E17 n$

Gamma source → $1.2E17 * 8.31 = 9.76E17 \gamma$

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

$S(E)$: 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		?	.

- ❑ 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

Ionizing Radiation Dose Ranges (Sievert)



Evidence for small increases in human cancer above 100 mSv acute exposure or 200 mSv chronic exposure

Typical mission doses on International Space Station (ISS)

Ramsar, Iran high natural bkg/yr

DOE Low Dose Program

Kerala coast, India high natural bkg/yr

Typical added annual dose for commercial airline flight crews

Airport x-ray whole body scanner: 0.00007 mSv/scan (Limit = 0.25 mSv/yr = 4000 scans/yr)

Round-trip Los Angeles - New York (≈ 0.037 mSv)

EPA dose limit public drinking water systems: 0.04 mSv/yr

EPA dose limit from release in air: 0.1 mSv/yr

NRC cleanup criteria for site decommissioning / unrestricted use: 0.25 mSv/yr

DOE, NRC dose limit for the public: 1 mSv/yr (100 mrem/yr) (ICRP, NCRP)

LD₅₀ = Lethal Dose to 50% (whole body dose that results in lethality to 50% of exposed individuals in 30-60 days)

Dose Equivalent: 1 Sievert = 100 rem = (absorbed dose x radiation quality) Absorbed Dose: 1 Gray = 100 rad 1 Sv ≈ 1 Gy for x- and gamma-rays ("≈" stands for "approximately equal to")

Whole body, acute: G-I destruction, lung damage; cognitive dysfunction (death certain in 5 to 12 days)*

Cancer Radiotherapy total doses to tumor

acute exposure = all at once; chronic = hours, days, years

Whole body, acute: circulating blood cell death; moderate G-I damage (death probable 2-3 wks)*

Whole body, acute: cerebral/vascular breakdown (death in 0-5 days)*

Whole body, acute: marked G-I and bone marrow damage (death probable in 1-2 wks)*

Life Span Study (A-bomb survivor epidemiology)

Total Body Irradiation (TBI) Therapy

Charged particle event (Solar flare) dose on moon, no shielding

Estimated dose for 3-yr Mars mission (current shielding)

Temporary epilation (3 wk onset)

Main erythema reaction (10 day onset)

Permanent epilation (3 wk onset)

Human LD₅₀ range acute exposure with medical intervention

Human LD₅₀ range acute exposure no medical intervention*

Human LD₅₀ range acute exposure with medical intervention

*Note: Whole body acute prognoses assume no medical intervention (G-I = gastrointestinal)

Cancer Epidemiology

Interplanetary Space natural bkg/yr

DHS emergency guideline to save a life: 250 mSv

DOE, NRC dose limit for workers: 50 mSv/yr (5 rem/yr)

Medical Diagnostics (A-O) see chart >>

Guarapari, Brazil high natural bkg/yr

Yangjiang, China high natural bkg/yr

Regulations & Guidelines

(TIPS: Transjugular Intrahepatic Porto-systemic Shunt)

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Chart compiled by NF Metting, Office of Science, DOE/BER. "Orders of Magnitude" revised June 2010 <http://www.lowdose.energy.gov/>

NOTE: This chart was constructed with the intention of providing a simple, user-friendly, "order-of-magnitude" reference for radiation exposures of interest to scientists, managers, and the general public. In that spirit, most quantities are expressed as "dose equivalent" in the more commonly used radiation protection units, the rem and Sievert. Medical diagnostics are expressed as estimated maximum organ dose; as they are not in "effective dose" they do not imply an estimation of risk (no tissue weighting). Dose limits are in effective dose, but for most radiation types and energies the difference is numerically not significant within this context. It is acknowledged that the decision to use these units is a simplification, and does not address everyone's needs. (NRC = Nuclear Regulatory Commission; EPA = Environmental Protection Agency; DHS = Department of Homeland Security) Disclaimer: Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information disclosed.

Source: Office of Biological and Environmental Research (BER), Office of Science, U.S. Department of Energy

CRITICALITY ACCIDENT IN TOKAI-MIRA 1999

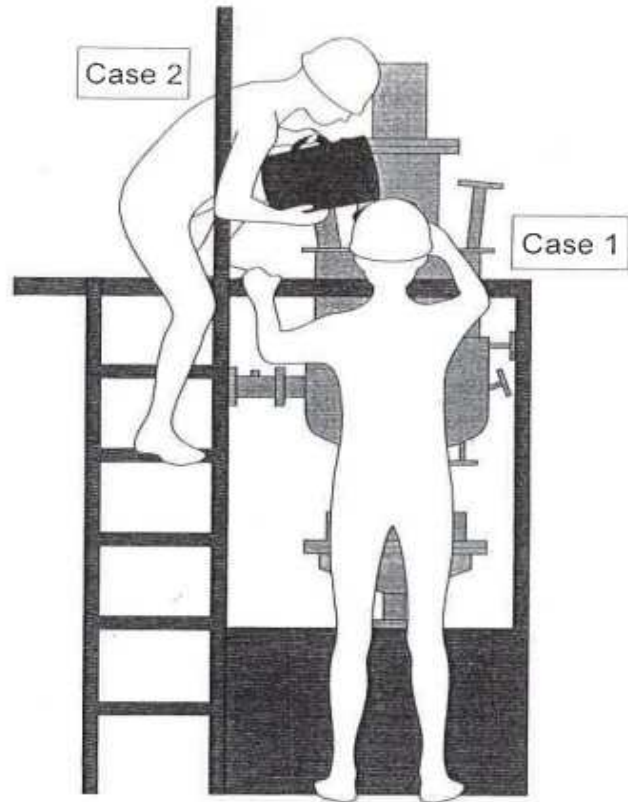


Fig. 1. Schematic diagram of the precipitation tank and likely positions of two victims.

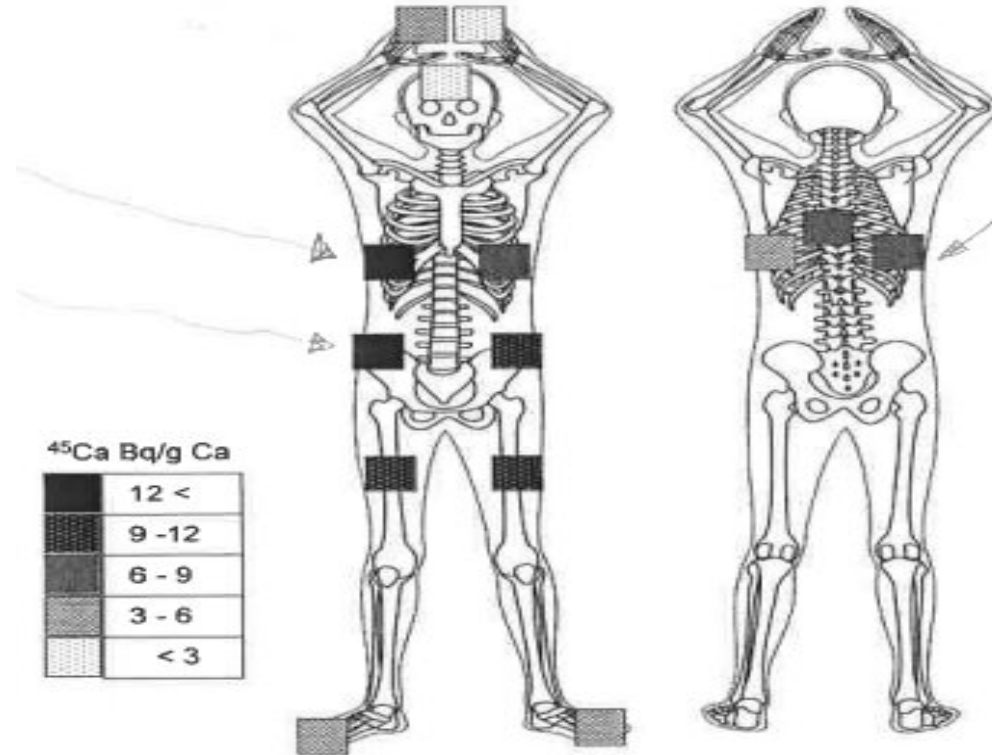


Fig. 4. Illustration of Case 1's orientation assumed at the moment of the burst. Cubes show ⁴⁵Ca activity level of the spots from which bones were taken.

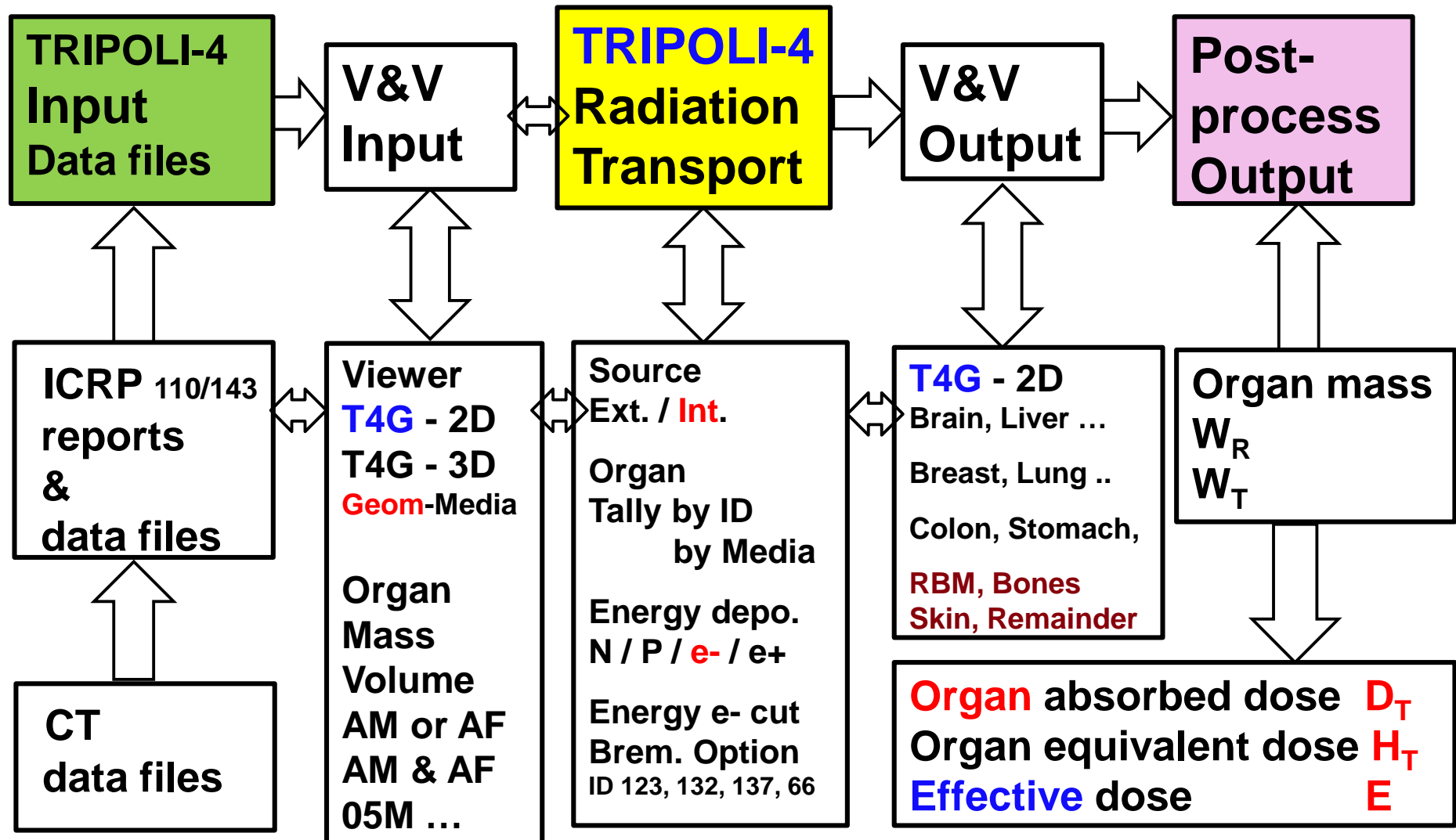
Neutron activation of ⁴⁴Ca in bone samples

→ ⁴⁵Ca Bq/g Ca → (Right iliac bone / Left iliac bone) = 1.7

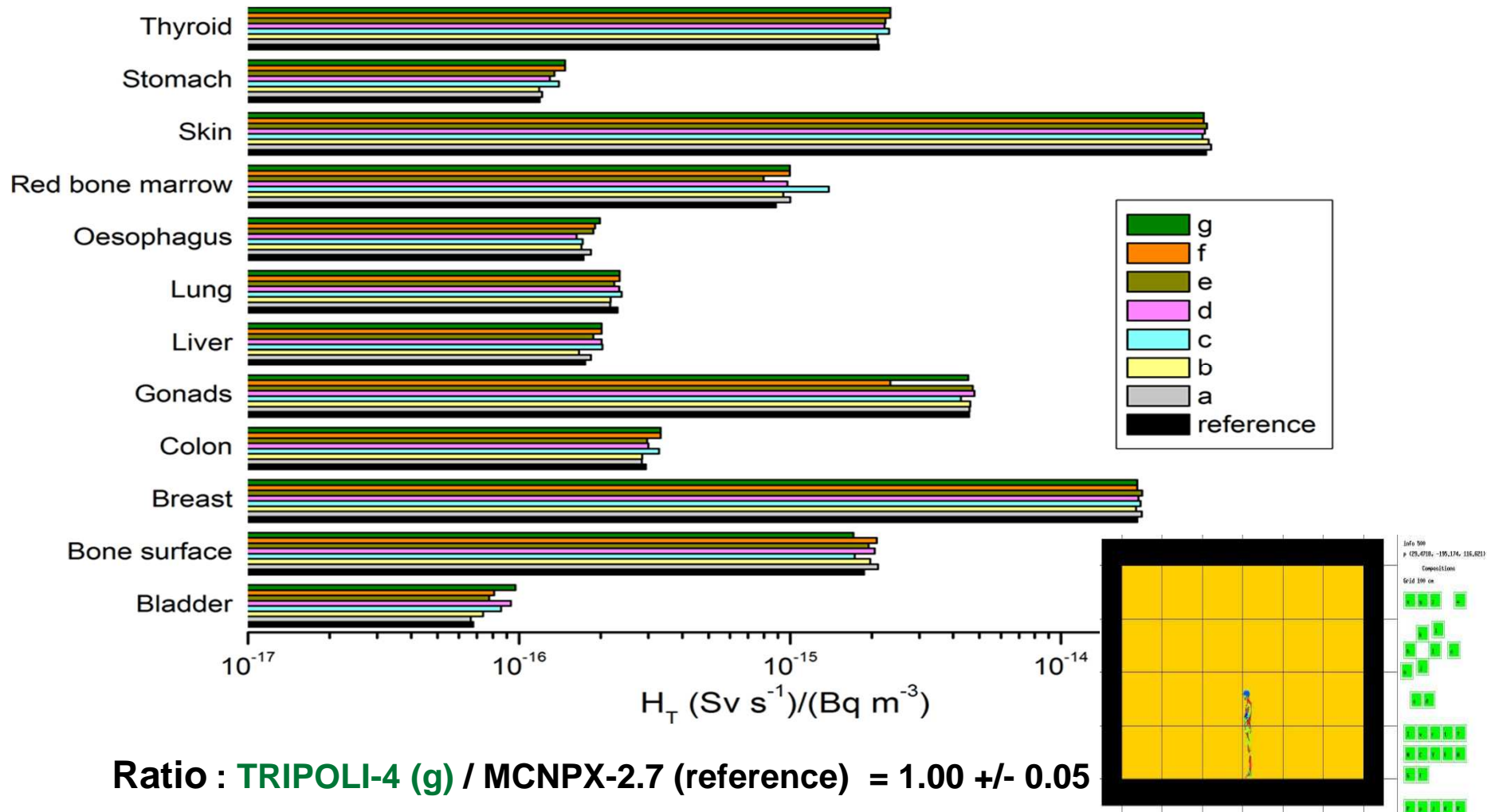
→ Self-shielding of human body → anterior & posterior ribs

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



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