H2020-MSCA-RISE-2014-GA641540, SKPLUS

Radioactive Contaminations in precision WČ Physics ; the SuperK-Gd case

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Outline

1. main sources of RI contaminations; main types of radiation produced

2. main physics measurements affected by radiation from RI in WC
3. relevant RI contaminants in [pure water] and doped WČ detectors; techniques for measuring RI contamination of materials for/in WČ

4. the case of Super-Kamiokande Gd (SuperK-Gd)
   - the regular Gd salt in the market
   - main physics affected and corresponding RI purity goals
   - measuring the very low contaminations in high purity Gd salts
   - the R & D for a high purity Gd salt

Main sources of radioactive contaminations

- long lived (> earth age) primordial isotopes
  - ✓ high Z are grouped into the 3 (4) decay chains: **^{238}U**, ^{235}U, **^{232}Th**
  - ✓ middle or low Z come alone, normally less important in *WČ* phys.

- cosmogenic production of long-lived radioactive isotopes in materials due to the exposure to cosmic rays on Earth
  - ✓ Cosmic-ray-muon spallation-induced in water RI with β decays: ^{12}B, ^{12}N, ^{16}N, ^{11}Be, ^{9}Li, ^{8}He, ^{9}C, **^{8}Li**, ^{8}B, and ^{15}C

- Anthropogenic radionuclides from nuclear weapons, nuclear power plants and uranium mining. The most recent strong contributions are from Chernobyl (1986) and Fukushima (2011) accidents. Most important RI are ^{90}Sr and ^{137}Cs.

- *Radon, mainly **^{226}Rn** in **^{238}U** decay chain, being a gas it is a special but very important source of RI*

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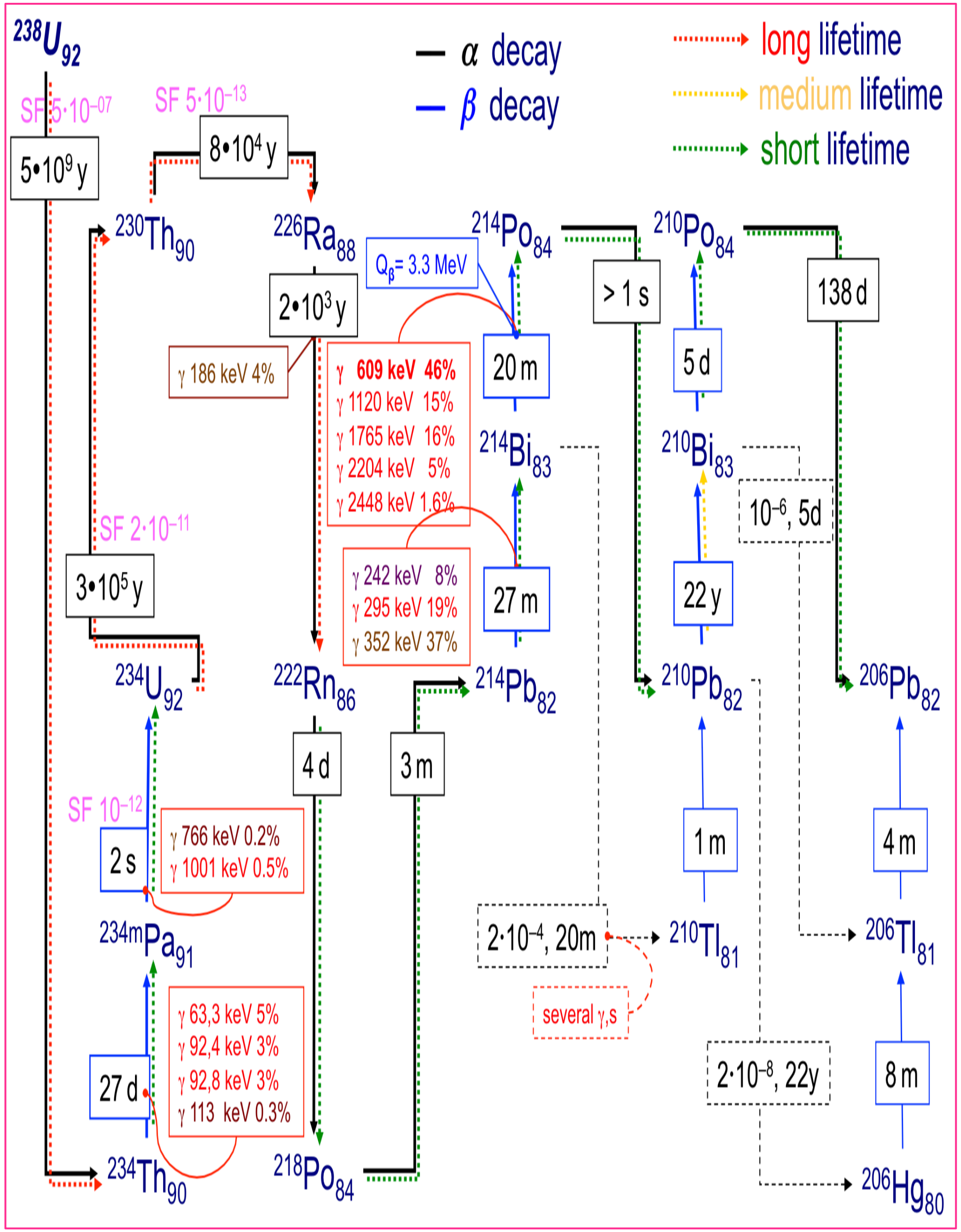
Main radiation produced by radioactive isotopes from contaminations

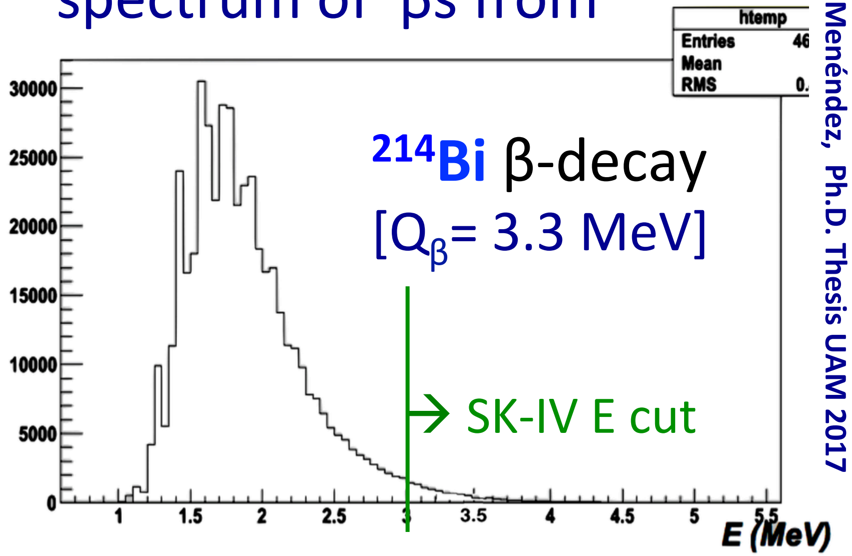
- spontaneous fission →
  - ✓ neutrons, gammas (can be of significant energy)
- α decays → α ($^4He_2$) particles
  - ✓ neutrons from (α, n) reactions maybe more dangerous
- β decays → β particles (electrons)
- nucleus stabilization after decay processes → gammas
- special care when gammas, neutrons, β are produced in coincidence

Main radiation produced by RI: the case of ^{238}U chain and ^{222}Rn



Radon is a gas →
diffusion along its surroundings
τ (^{222}Rn) ≈ 4 d
→ **spread of RIs**

For illustration, in SK:
reconstructed Energy
spectrum of βs from



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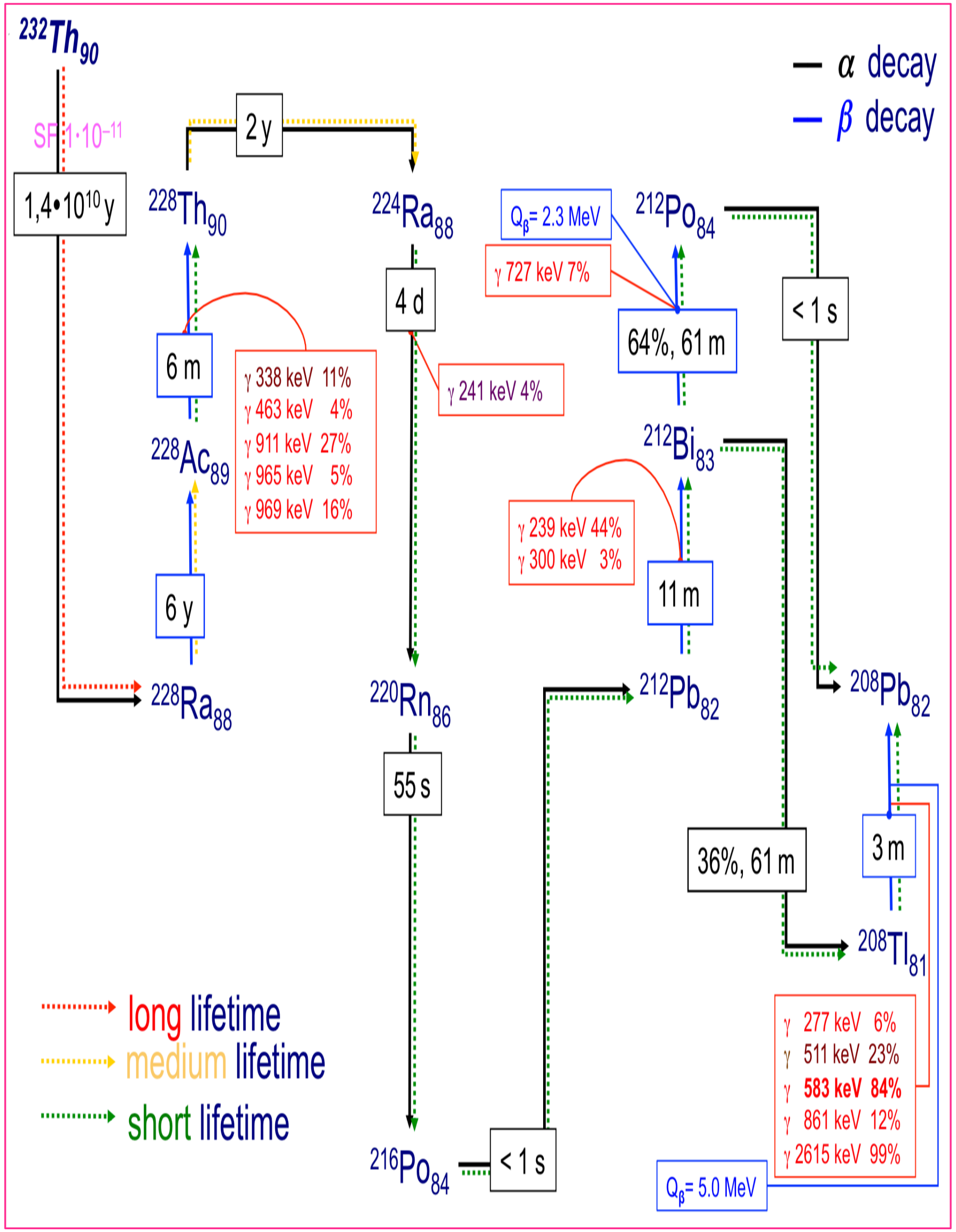
Main radiation produced by RI: the case of ^{232}Th chain (and ^{224}Ra)



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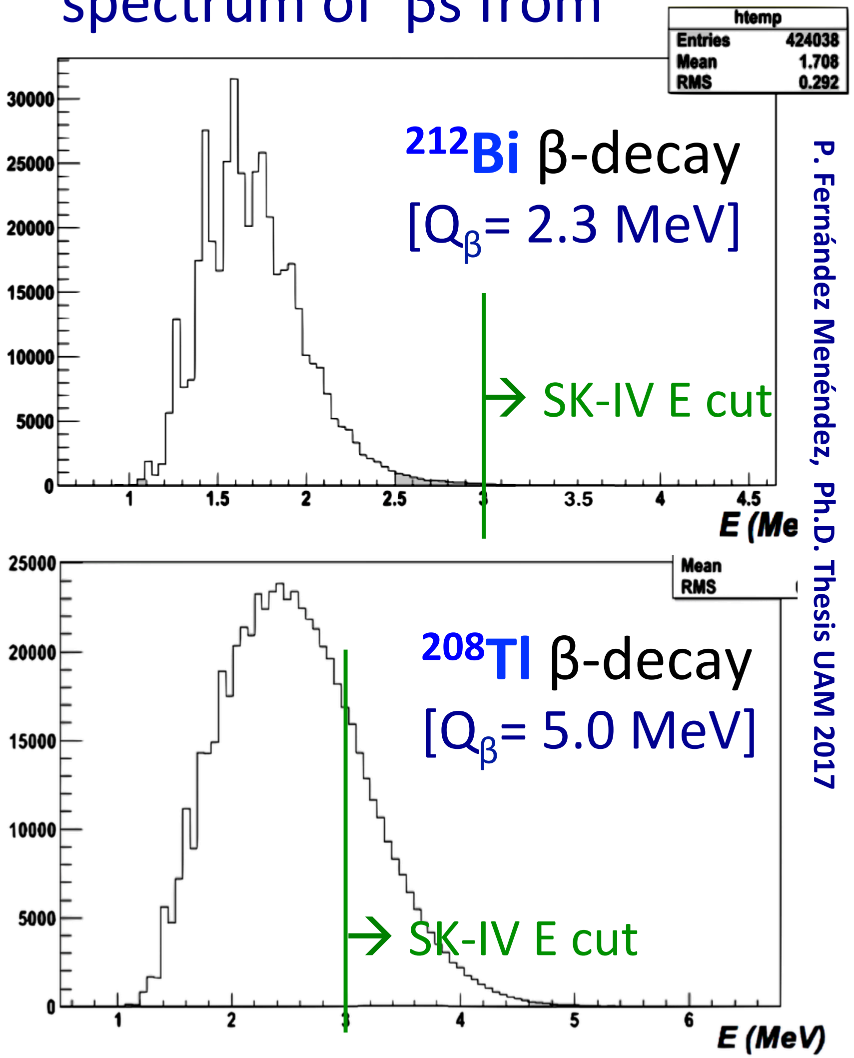

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neutrons

- there are many, naturally produced
- relevant are neutrons produced from ***α*** decays in the naturally present radioactive chains
- for instance that of ^{238}U
- ***α***s interact with the water :

$$^{18}O + ^{4}\alpha(\sim 6MeV) \rightarrow ^{22}Ne^{*}[\sim 15MeV] \rightarrow$$

$$^{22}Ne + \gamma \qquad \qquad ^{21}Ne + n + \gamma$$

$$\rightarrow ^{21}Ne^{*}[\sim 12.5MeV] + n(\sim 2.5MeV) \rightarrow ^{20}Ne + 2 \cdot n(\sim 2.3MeV)$$

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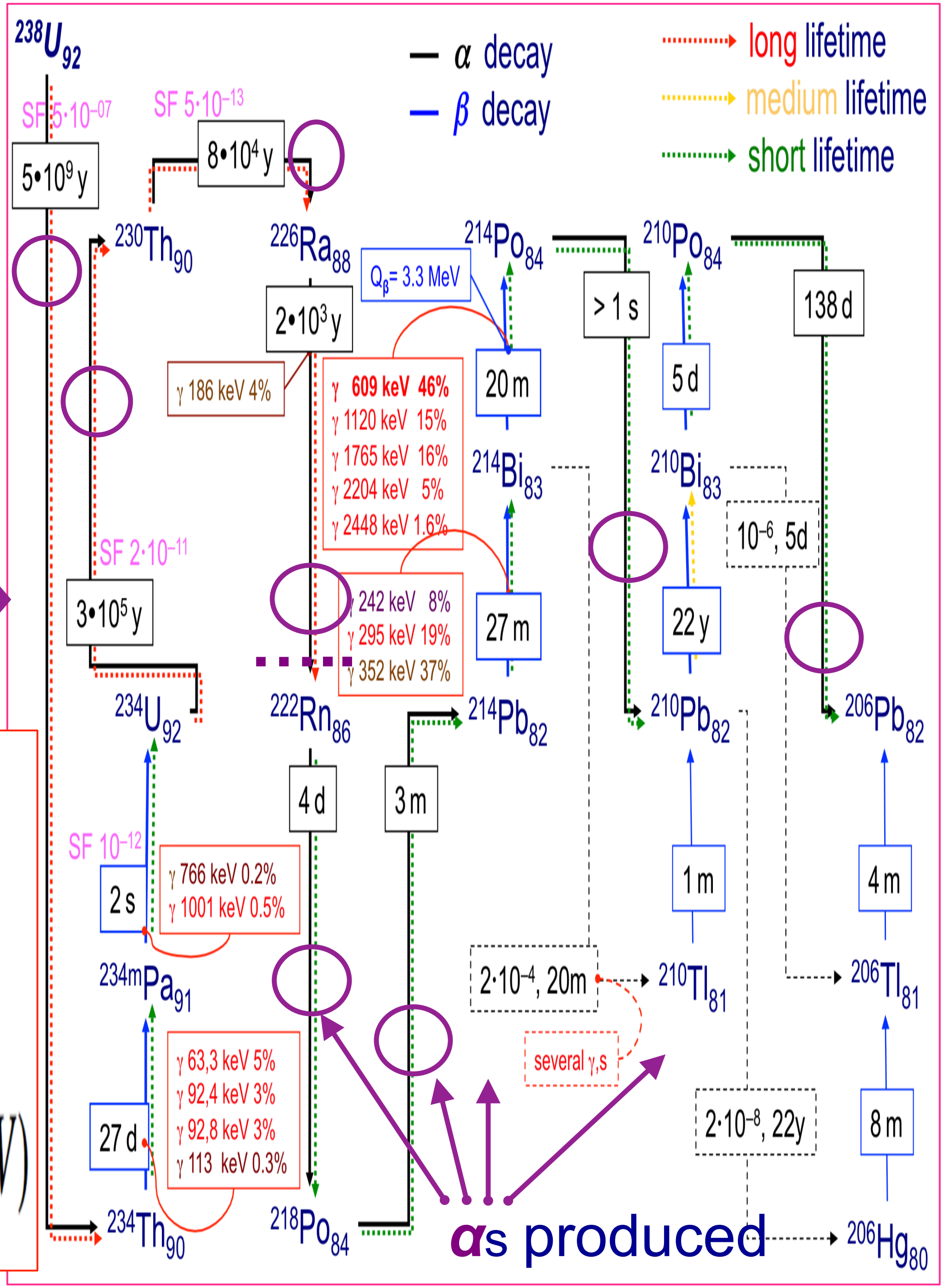

Main radiation produced by RI: neutrons

example from ^{232}Th, ^{235}U, ^{238}U chains in SuperK-Gd for "market standard" $Gd_2(SO_4)_3$

- relatively high RI levels (see later)
- computed with SOURCES4C
- dominated by (α,n) reactions on oxygen ^{18}O
- multiplicity of two in them
- for the radioactivity levels taken $5.1 \cdot 10^{-13}$ neutrons/s/cm^3

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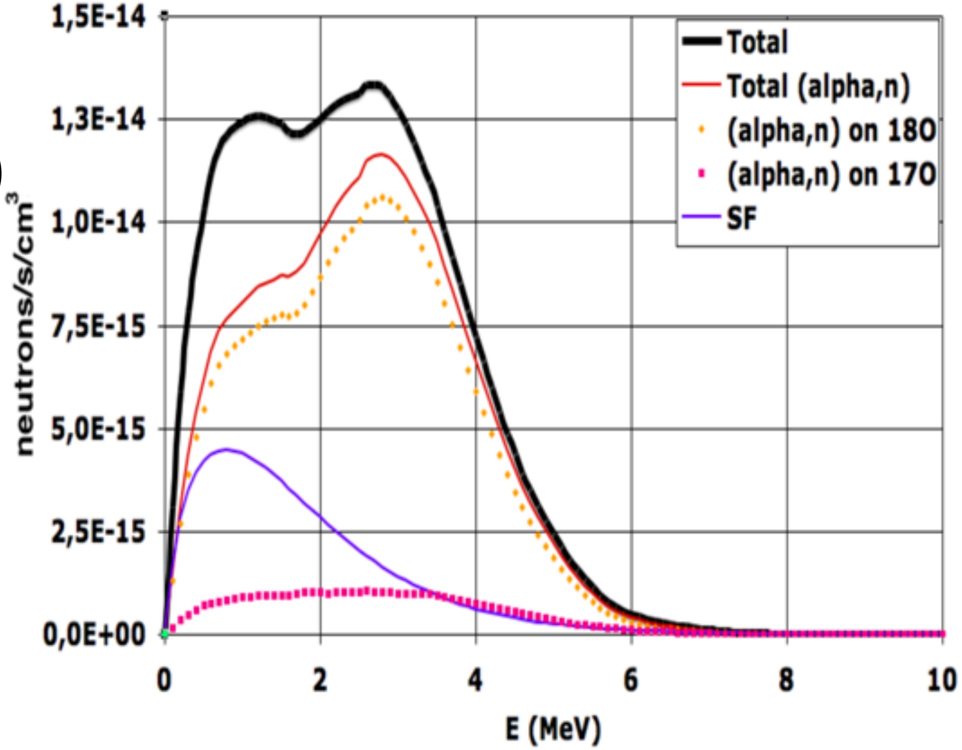


$$N_{rad}^{neutrons} = 316.3 \frac{single\ neutrons}{day \cdot SKFV}$$

Main physics measurements affected by radiation from RI in WC
→ low energy neutrino physics

- general: worse reconstruction because of increased "dark noise"

- neutron "blind" detector (SK-I, II, III …)
  - solar elastic
  - SRN

- neutron sensitive detector (SNO, SK-IV, SK-Gd …)
  - solar elastic
  - solar NC (**can be very severe**, see next slide)
  - SRN
  - reactor
  - pre-supernova

neutrons at Neutral Current scattering measurement by SNO

$$\nu_x + d \rightarrow p + n + \nu_x$$

sensitive to **all three** ν_e , ν_μ , ν_τ

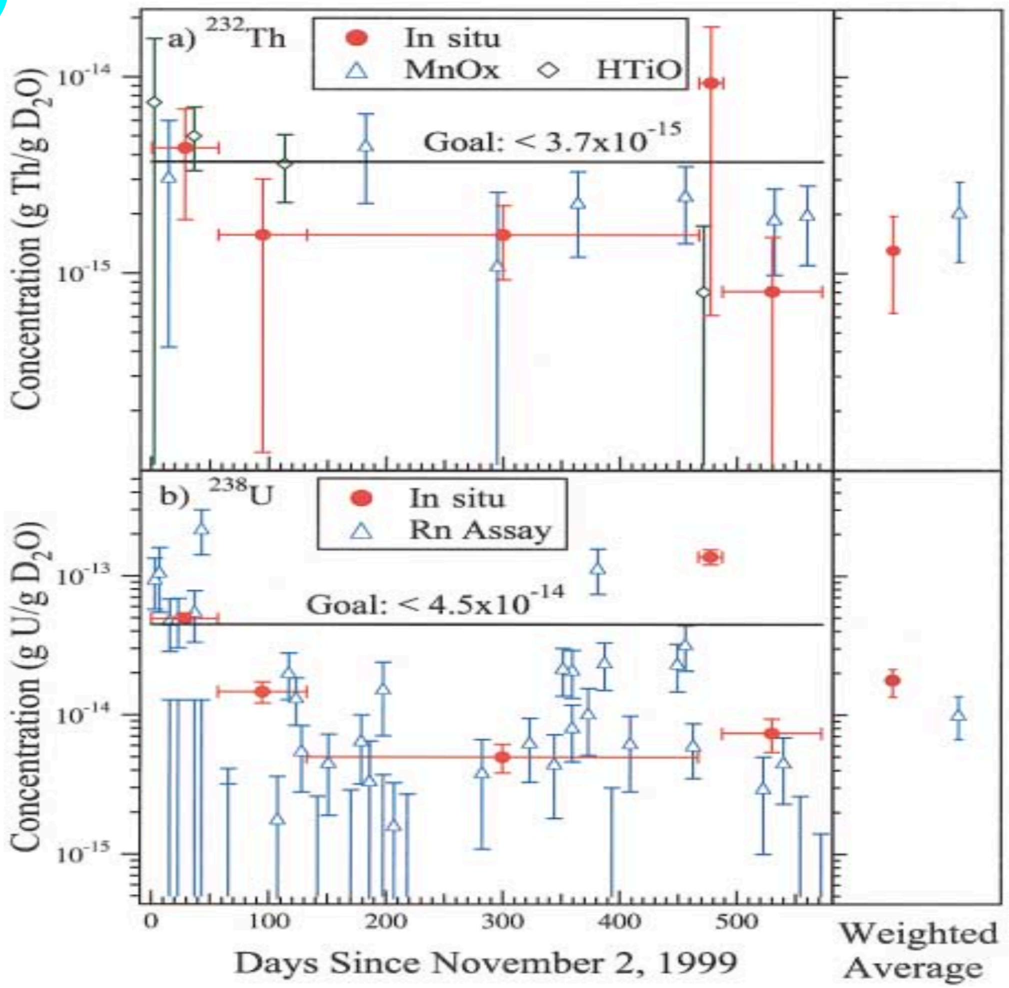
with E[ν_x] > 2.2 MeV (binding E.)

neutron capture:

$$^2H + n \rightarrow {^3H} + \gamma \text{ [6.2 MeV]}$$

Cerenkov ring; **just event counting**

SNO; Phys. Rev. Lett. 89, 011301 (2002)



n are **irreducible background**

a) minimize to the maximum
- purest D_2O
- acrylic vessel to isolate D_2O from external contamination

b) quantify to the highest precision:
- permanent monitoring by
- 2 ex-Situ radioactivity cont. meas. systems
- 1 in-situ technique

relevant RI contaminants in regular WČ detectors (pure water)

- materials close to ID:
  - PMT glass
  - in SK FRP cover
  - other

→ increase "dark noise"
→ reduce FV

Y. Nakano, previous talk

- other sources of radon diffusion:
  - from water after purification cycle
  - gaskets in water inlets ! H. Sekiya @LRT 2015

→ radon into FV

relevant RI contaminant [-ions] in doped WČ detectors

- those as in pure water
- contaminations in the solute
  → these might be much more dangerous since they RIs spread out along the whole fiducial volume

techniques for measuring the RI contamination of materials for/in WČ

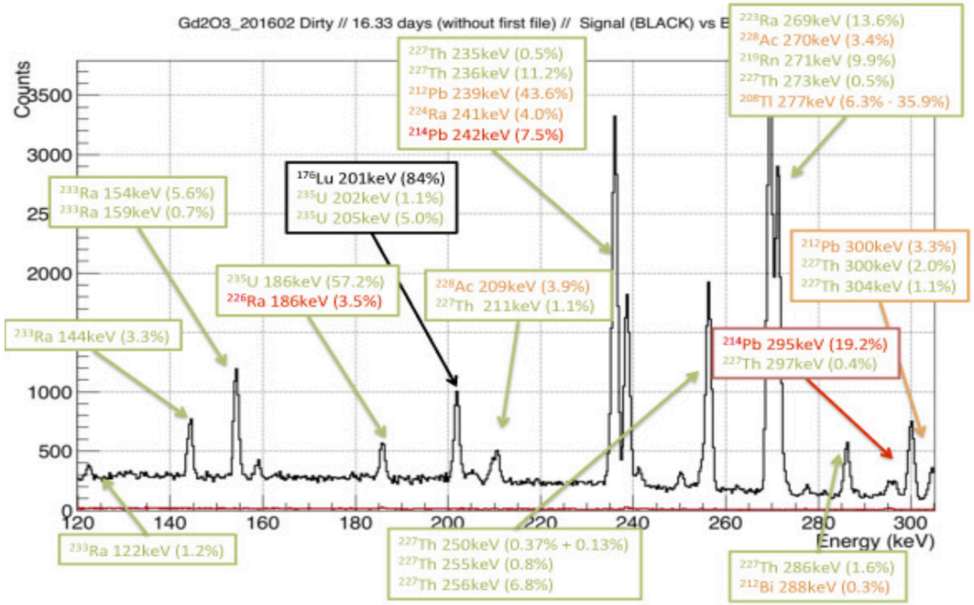
- High purity Ge detectors
  - ✓ measuring of gammas from radioactive decays: allows to investigate basically all the contaminants
  - ✓ for extremely high RI purity physics it might be not enough

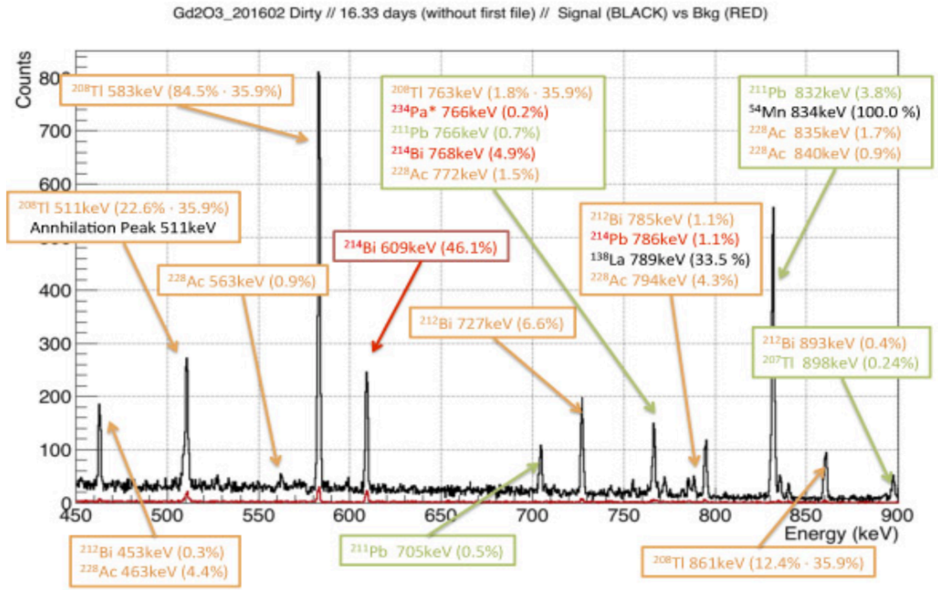
- ICPMS
  - ✓ can detect extremely low amounts of RIs
  - ✓ sensitive only to very long lived RIs ($\gtrsim 10^8$ years)

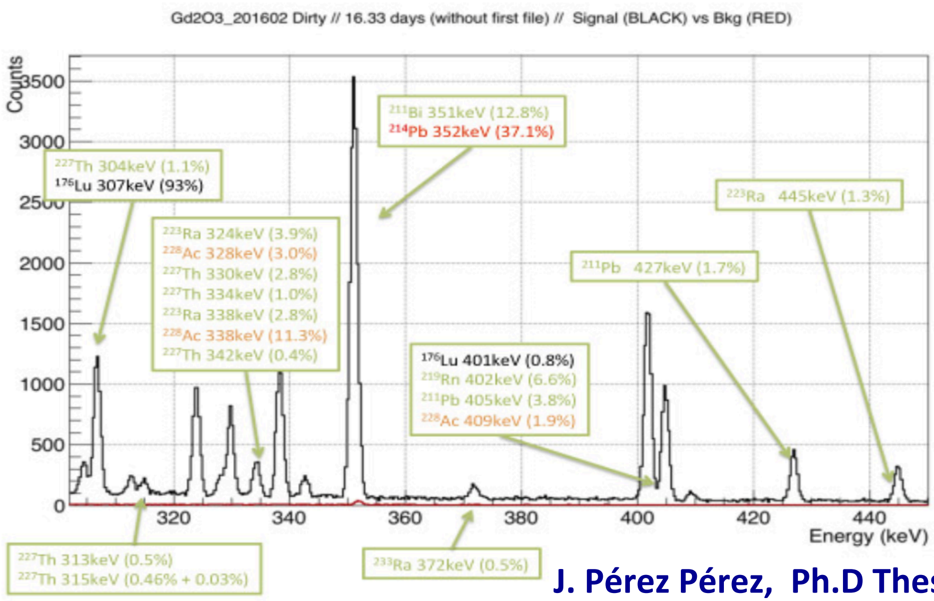
- radon detection
  - ✓ possibility of sampling in-situ, even online measuring
  - ✓ very high sensitivities can be achieved
  - ✓ very difficult technique

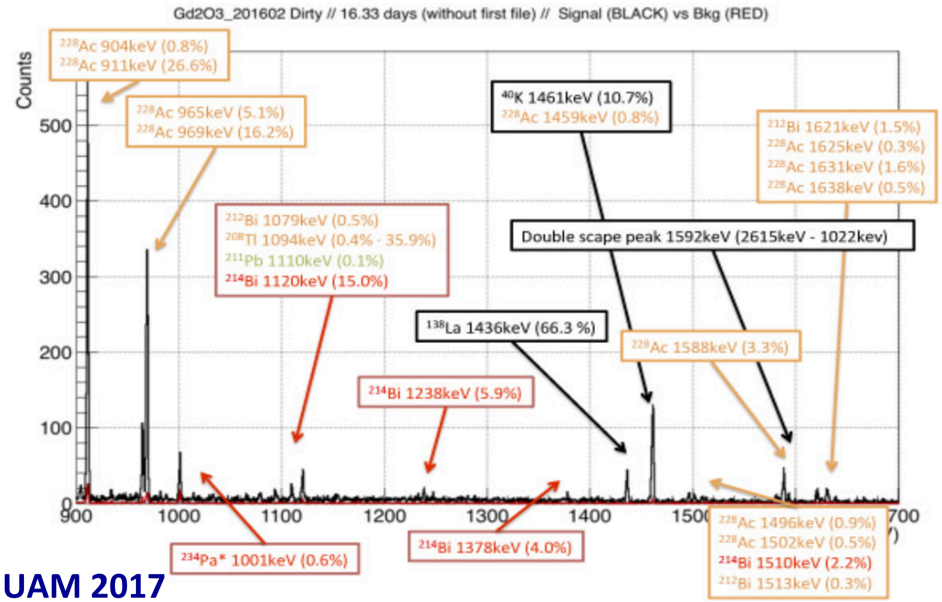
- ….

example of HP-Ge measurement:
measured gamma spectra of a highly RI contaminated sample of Gd salt









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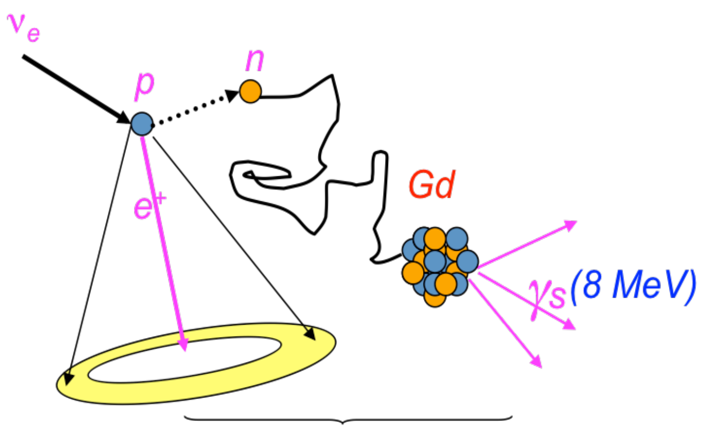
the case of SuperK-Gd



H.W:Sobel et al. PRC 7-4 1973

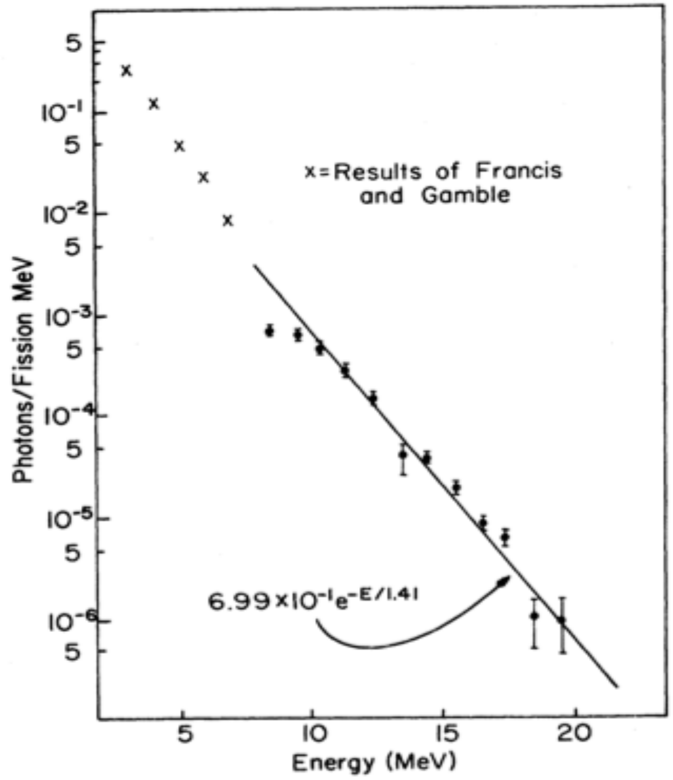


FIG. 10. Photons/fission MeV versus photon energy.

Background for Supernova Relic Neutrino:

^{238}U Spontaneous Fission:

- coincidence 1 neutron – 1 γ (Eγ>10 MeV)
- irreducible background !
- fortunately not a serious issue

Solar ***ν*** in accidental coincidence with neutrons or βs from radioactivity

spatial (2m sphere) and time coincidence (60μs after prompt) between

- prompt "signal": solar final E>10 MeV (~1460 events/year/FV)
- delayed "signal": β from RI decays / capture of radioactivity neutrons

the case of SuperK-Gd

Background for lowest Energy solar ν

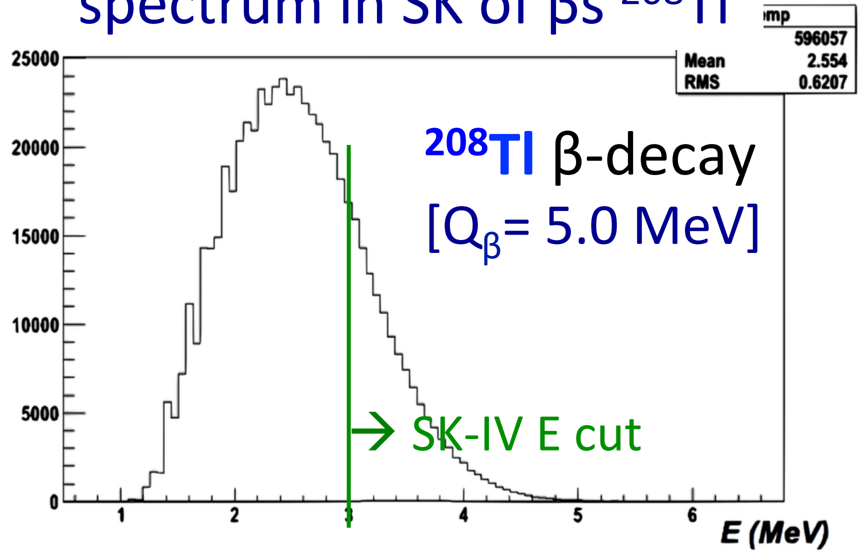
βs from ^{232}Th, ^{238}U chains

- very severe background source

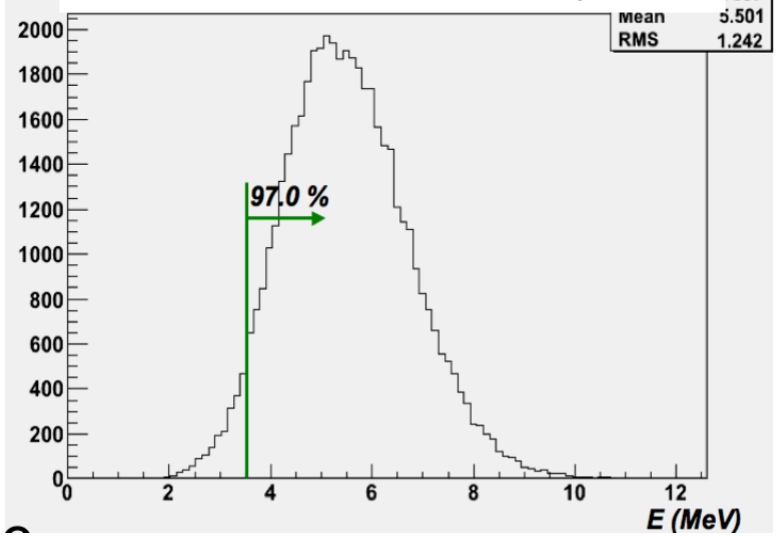
Gd captures of neutrons produced by the radioactive contamination

- sizable but less severe

reconstructed Energy spectrum in SK of βs ^{208}Tl



approximate E spectrum of gamma cascade from Gd neutron capture



$Gd_2(SO_4)_3$ "regular market" survey: radioactivity contaminations

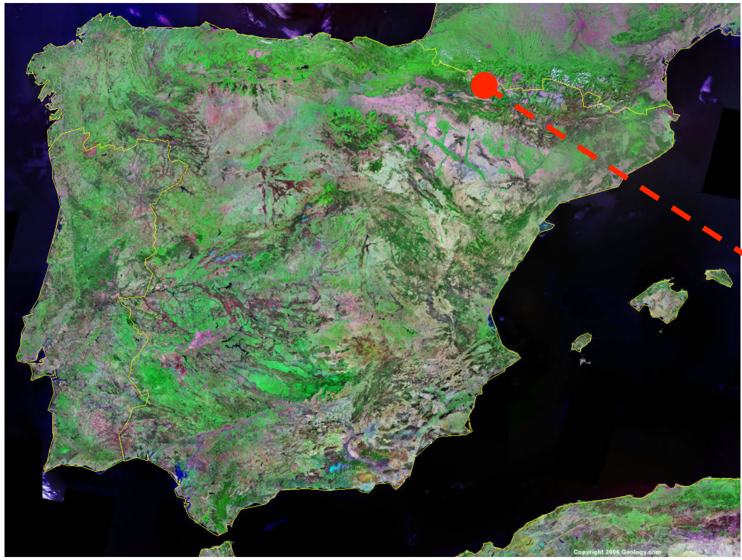
		Measured radioactivity in mBq/kg for the $Gd_2(SO_4)_3$ batches purchased to date **June 2015**								
Chain	**Sub-chain**	**Standford Materials** 09/04	**Standford Materials** 10/08	**Beijing Jinghonganxin** 12/08	**Changshu Huanyu** 13/02	**Beijing Jinghonganxin** 13/03	**Standford Materials** 13/08	**HK Tai Kun** 13/07a	**HK Tai Kun** 13/07b	**Standford Materials** 14/12
^{238}U	^{238}U	51 ± 21	< 33	292 ± 6	74 ± 28	242 ± 6	71 ± 20	47 ± 26	73 ± 27	< 76
	^{226}Ra	8 ± 1	2.8 ± 0.6	74 ± 2	13 ± 1	13 ± 2	8 ± 1	5 ± 1	6 ± 1	< 1.4
^{232}Th	^{228}Ra	11 ± 2	270 ± 16	1099 ± 12	205 ± 6	21 ± 3	6 ± 1	14 ± 2	3 ± 1	2 ± 1
	^{228}Th	28 ± 3	86 ± 5	504 ± 6	127 ± 3	374 ± 6	159 ± 3	13 ± 1	411 ± 5	29 ± 2
^{235}U	^{235}U	< 32	< 32	< 112	< 25	< 25	< 32	< 12	< 30	< 1.8
	^{227}Ac	214 ± 10	1700 ± 20	2956 ± 30	1423 ± 21	175 ± 42	295 ± 10	< 6	< 18	190 ± 6
Others	^{40}K	29 ± 5	12 ± 3	101 ± 10	60 ± 7	18 ± 8	3 ± 2	3 ± 2	8 ± 4	< 5
	^{138}La	8 ± 1	$<$	683 ± 15	3 ± 1	42 ± 3	5 ± 1	< 1	< 2	23 ± 1
	^{176}Lu	80 ± 8	21 ± 2	566 ± 6	12 ± 1	8 ± 2	30 ± 1	1.6 ± 0.3	< 2	2.5 ± 0.6

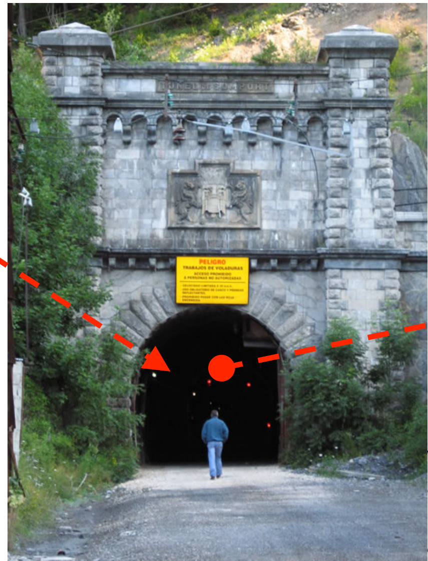
work done mostly at the *Canfranc Underground Laboratory*

- salts from different providers have in general similar contaminations
- some improvement along time seen
- in any case, Superk-Gd can not afford those amounts of RIs

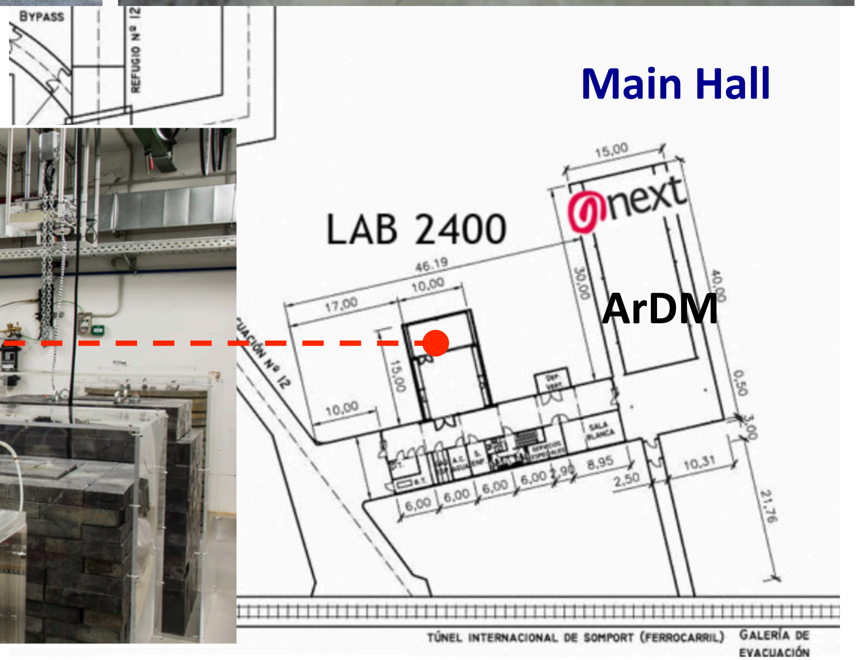
A "propaganda" slide

[***LSC***, *Laboratorio Subterráneo de Canfranc*]





HPGe detector farm



impact on physics of RI contaminations in "regular market" $Gd_2(SO_4)_3$

Typical activities of salts in the market:
(from over 10 samples from 5 providers)

June 2015

Radioactive chain	Part of the chain	mBq/kg
^{238}U	^{238}U	50
	^{226}Ra	5
^{232}Th	^{228}Ra	10
	^{228}Th	100
^{235}U	^{235}U	32
	^{227}Ac / ^{227}Th	300

For DSNB

Expected signal ~5 events/year/FV

- ^{238}U Spontaneous Fission:

  ~ 5.5 [γ($E\gamma$>10.5 MeV) + 1n] / year / FV

an approx. **x10 reduction desirable**

For solar neutrino

Current BG ~200 events/day/FV

- U (n) ~320events/day/ FV

an approx. **x10 reduction desirable**

- Th/Ra (β,γ)~3 x 10^5 events/day/ FV

x10^3 reduction needed !

Physics based requirements for RI contamination at $Gd_2(SO_4)_3$

Typical activities of salts in the market:
(from over 10 samples from 5 providers)

Physics based requirements

Radioactive chain	Part of the chain	mBq/kg	SRN (mBq/kg)	Solar ν (mBq/kg)
^{238}U	^{238}U	50	< 5	-
	^{226}Ra	5	-	< 0.5
^{232}Th	^{228}Ra	10	-	< 0.05
	^{228}Th	100	-	< 0.05
^{235}U	^{235}U	32	-	< 3
	^{227}Ac / ^{227}Th	300	-	< 3

- Superk-Gd can not afford those amounts of RI, **approaches to reduce** them
  - ✓ by ourselves from received batches [a lot of work being done in Kamioka, not discussed here]
  - ✓ Cooperative development of pure salts with chemical Companies
    Shin-Etsu Chemical Co. Ltd., Kanto Chemical Co. Inc., Wako Pure Chemical Ind. Ltd., Nippon Yttrium Co. Ltd.

M. Ikeda, H. Sekiya et al.

R&D of “ultra” pure Gd powder

^{238}U: γ and neutrons from S.F.
^{226}Ra: β from ^{214}Bi （Q=3.27MeV）
^{232}Th: γ from ^{208}Tl （=2.6MeV）
^{235}U: neutrons from decay chain

← SRN BG < 0.5 events/year

< solar ν BG level.

Unit: [mBq/kg $(Gd_2SO_4)_3$ $8H_2O$]

* Goal for 0.2% Gd-sulfate loading

Chain	Isotope	Typical	Goal*	Company A		Company B		Company C	
				Ge	ICPMS	Ge	ICPMS	Ge	ICPMS
^{238}U	^{238}U	50	< 5	-	~ 0.04	< 11	< 0.04	< 10	< 0.04
	^{226}Ra	5	< 0.5	-	—	<0.2	—	< 0.2	—
^{232}Th	^{232}Th	100	< 0.05	-	~ 0.09		0.02	—	0.06
	^{228}Ra	10	< 0.05	-	—	< 0.3	—	< 0.2	—
	^{228}Th	100	< 0.05	-	—	< 0.3	—	< 0.3	—
^{235}U	^{235}U	30	< 3	-	—	< 0.4	—	< 0.3	—
	$^{227}Ac/Th$	300	< 3	-	—	< 1.7	—	< 1.2	—

Ge detector: Sensitive to almost 0.1 mBq/kg (Canfranc, Boulby and Kamioka)

ICPMS: For isotopes w/ long life (Kamioka)

Company B achieved goals for U, 226Ra and 232Th

Summary / Conclusions / Outlook

- Radioactive contaminations can jeopardize the physics outcome of **WČ** detectors; mostly for low energy reactions
- particular care has to be put if a solute is dissolved in the water: RI might be spread along the whole fiducial volume

- In SuperK-Gd the measurement most severely affected by radioactivity contamination is low energy solar neutrinos
- SuperK-Gd has carried out a very hard but successful campaign / R&D program in order to external companies achieving the needed high purity $Gd_2(SO_4)_3$ in a regular production mode

- SuperK-Gd will dissolve 100 ton of $Gd_2(SO_4)_3$; its quality, particularly its radio-purity has to be scrutinized for every production batch (~0.5 tons) → large international effort involving Boulby, Canfranc, Kamioka (+ others ?) laboratories additional

First measurement of radioactive isotope production through cosmic-ray muon spallation in Super-Kamiokande IV;
Super-Kamiokande, PRD 93, 012004 (2016)

TABLE I. Possible radioactive isotopes induced by cosmic-ray muon spallation at SK [13,22,23]. The fourth column lists the end point kinetic energy ($E_{\mathrm{kin.}}$). The fifth column lists the primary generation process of the radioactive isotopes.

Radioactive isotope	τ (s)	Decay mode	$E_{\mathrm{kin.}}$ (MeV)	Primary process
^{11}Be	19.9	β^-	11.51	$^{16}O(n, \alpha + 2p)^{11}Be$
		$\beta^-\gamma$	$9.41 + 2.1(\gamma)$	
^{16}N	10.3	β^-	10.44	$^{16}O(n, p)^{16}N$
		$\beta^-\gamma$	$4.27 + 6.13(\gamma)$	
^{15}C	3.53	β^-	9.77	$^{16}O(n, 2p)^{15}C$
		$\beta^-\gamma$	$4.51 + 5.30(\gamma)$	
^{8}Li	1.21	β^-	~13.0	$^{16}O(\pi^-, \alpha + {}^2H + p + n)^8Li$
^{8}B	1.11	β^+	~13.9	$^{16}O(\pi^+, \alpha + 2p + 2n)^8B$
^{16}C	1.08	$\beta^- + n$	~4	$^{18}O(\pi^-, n + p)^{16}C$
^{9}Li	0.26	β^-	13.6	$^{16}O(\pi^-, \alpha + 2p + n)^9Li$
		$\beta^- + n$	~10	
^{9}C	0.18	$\beta^+ + p$	3–15	$^{16}O(n, \alpha + 4n)^9C$
^{8}He	0.17	$\beta^-\gamma$	$9.67 + 0.98(\gamma)$	$^{16}O(\pi^-, {}^3H + 4p + n)^8He$
		$\beta^- + n$		
^{12}Be	0.034	β^-	11.71	$^{18}O(\pi^-, \alpha + p + n)^{12}Be$
^{12}B	0.029	β^-	13.37	$^{16}O(n, \alpha + p)^{12}B$
^{13}B	0.025	β^-	13.44	$^{16}O(\pi^-, 2p + n)^{13}B$
^{14}B	0.02	$\beta^-\gamma$	$14.55 + 6.09(\gamma)$	$^{16}O(n, 3p)^{14}B$
^{12}N	0.016	β^+	16.38	$^{16}O(\pi^+, 2p + 2n)^{12}N$
^{13}O	0.013	$\beta^+ + p$	8–14	$^{16}O(\mu^-, \mu^- + p + 2n + \pi^-)^{13}O$
^{11}Li	0.012	β^-	20.62	$^{16}O(\pi^+, 5p + \pi^0 + \pi^+)^{11}Li$
		$\beta^- + n$	~16	

Radioactive Contamination in Neutrino Experimental Physics: the Cases of NEXT and Super-Kamiokande Experiments;
J. Pérez Pérez, Ph.D Thesis UAM 2017

The 32 Primordial radionuclides are: ^{40}K, ^{48}Ca, ^{50}V, ^{76}Ge, ^{82}Se, ^{87}Rb, ^{96}Zr, ^{100}Mo, ^{113}Cd, ^{115}In, ^{116}Cd, ^{128}Te, ^{130}Te, ^{130}Ba, ^{136}Xe, ^{138}La, ^{144}Nd, ^{147}Sm, ^{148}Sm, ^{150}Nd, ^{151}Eu, ^{152}Gd, ^{174}Hf, ^{176}Lu, ^{180}W, ^{186}Os, ^{187}Re, ^{190}Pt, ^{209}Bi, ^{232}Th, ^{235}U and ^{238}U.

The most used cosmogenic radionuclides are ^{3}H, ^{10}Be, ^{14}C, ^{21}Ne, ^{26}Al, and ^{36}Cl; usually for dating geologic materials or rocks.

In Xenon, has been observed these isotopes: ^{7}Be, ^{85}Sr, ^{88}Zr, $^{91*}Nb$, ^{99}Rh, ^{101}Rh, $^{110*}Ag$, ^{113}Sn, ^{125}Sb, $^{121*}Te$, $^{123*}Te$, ^{126}I, ^{131}I, ^{127}Xe, $^{129*}Xe$, $^{131*}Xe$, ^{133}Xe, and ^{132}Cs. In copper, has been observed: ^{46}Sc, ^{48}V, ^{54}Mn, ^{59}Fe, ^{56}Co, ^{57}Co, ^{58}Co and ^{60}Co.