短半減期核種の内部被ばく
Dose reconstruction issues regarding internal exposure due to short half-lives radionuclides

Ichiro Yamaguchi, NIPH

Alex Malins, JAEA

☑ The authors have no conflict of interest to disclose with respect to this presentation.

Contents
20 minutes talk

- Motivation / Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion
- The other issues

Contents

- Motivation / Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion

UNSCEAR 2017 White paper

- 27. Within the next two to three years (i.e. by 2020), these ongoing studies are expected to provide:
- (d) Better estimates of the doses received by members of the public, in particular doses from internal exposure to short-lived radionuclides in the early phase of the accident, doses from external exposure and how they varied according to behaviour and location, and doses from the ingestion of foodstuffs from the second year after the accident onwards;

http://www.unscear.org/unscear/en/publications/Fukushima_WP2017.html

放射性ヨウ素の同位体の寄与

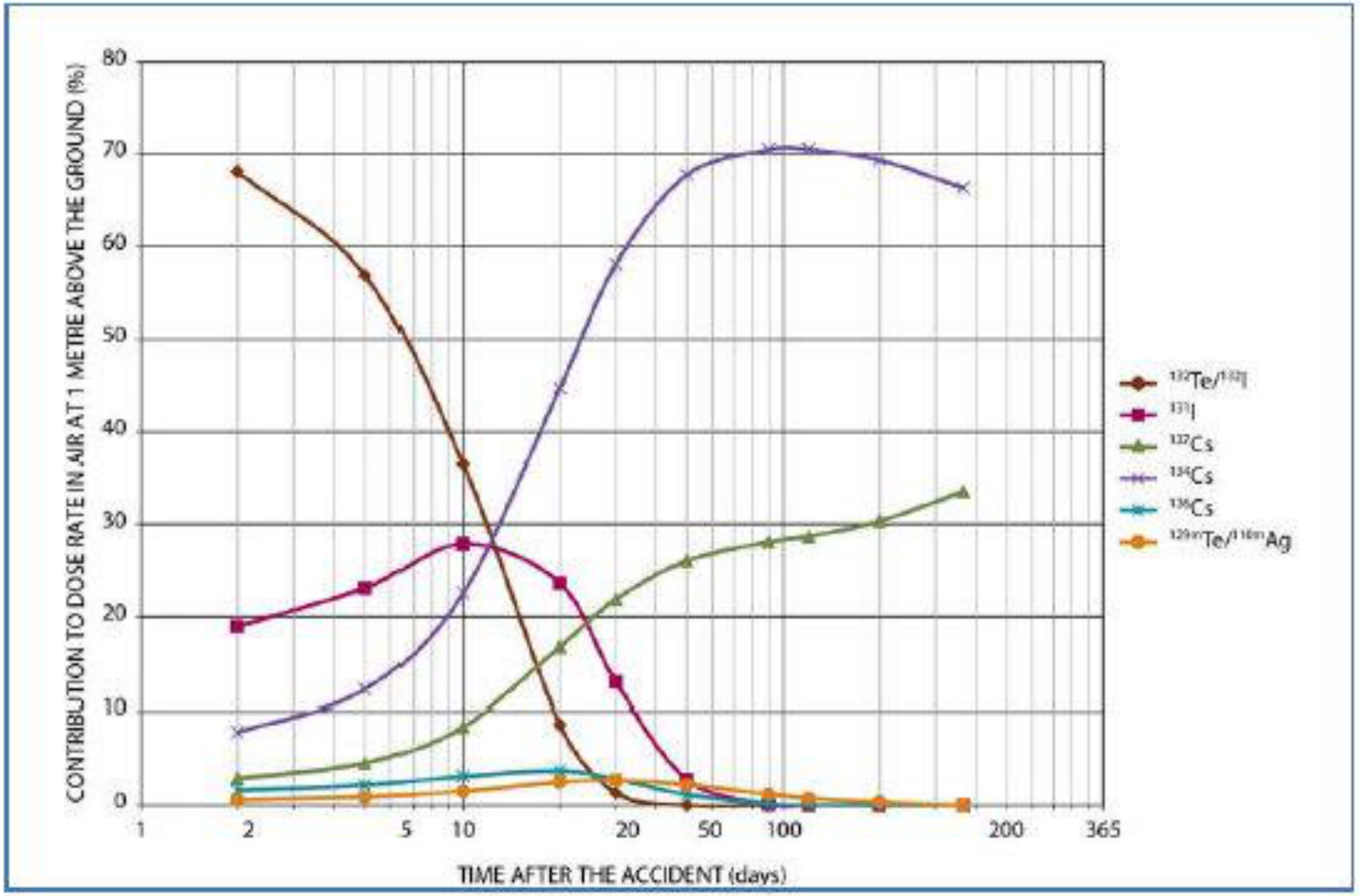
表３－１　事故以前の運転によって蓄積された各号機炉内のヨウ素放射能
（事故直後の放射性ヨウ素の放射能）

核種	半減期	平成 23 年 3 月 11 日時点での放射能(GBq/core)		
		1 号機	2 号機	3 号機
I-128	24.99m	8.1×10^{6}	1.2×10^{7}	1.2×10^{7}
I-129	1.57×10^{7}y	4.8×10^{1}	5.9×10^{1}	5.6×10^{1}
I-130	12.36h	1.9×10^{7}	2.8×10^{7}	2.6×10^{7}
I-131	8.0252d	1.3×10^{9}	2.3×10^{9}	2.3×10^{9}
Te-132/I-132	3.204d/2.295h	1.9×10^{9}	3.4×10^{9}	3.4×10^{9}
I-133	20.8h	2.8×10^{9}	4.8×10^{9}	4.9×10^{9}
I-134	52.5m	3.1×10^{9}	5.5×10^{9}	5.5×10^{9}
I-135	6.58h	2.6×10^{9}	4.6×10^{9}	4.6×10^{9}

https://www.nsr.go.jp/data/000085734.pdf

放射性ヨウ素以外の核種

Figure C-VIII. Percentage contribution of different radionuclides to the dose rate in air at 1 m above the ground in the first months after the accident



http://www.unscear.org/docs/reports/2013/13-85418_Report_2013_Annex_A.pdf

Questions from a citizen and atmospheric environmental researchers

測定試料採取点*		採取日時	放射能濃度(Bq/m^3)					
			^{131}I	^{134}Cs	^{137}Cs	^{132}I	^{132}Te	その
【3-1】(33km北西)1回目	相馬郡飯舘村長泥	3月25日 12:28～12:52	38	1.6	1.9	110	1.6	不検出
【3-1】(33km北西)2回目		3月25日 13:28～13:50	440	8.1	10	680	14	^{99m}Tc：5.2
【3-1】(33km北西)3回目		3月25日 14:28～14:50	330	6.2	7.7	410	5.4	^{99m}Tc：3.7
【3-1】(33km北西)4回目		3月25日 15:28～15:49	290	9.6	14	380	6.3	^{99m}Tc：3.7
Aコース		3月19日 13:16～13:36	25	6.1	7.0	29	11	^{99m}Tc：1.6
Dコース		3月19日 13:41～13:58	1.2	不検出	不検出	14	不検出	不検出
Cコース		3月19日 13:54～14:14	1.7	不検出	不検出	15	不検出	不検出
福島大学		3月18日 12:50～13:00	不検出	不検出	不検出	11,000	不検出	不検出
福島飯坂IC		3月18日 13:51～14:01	不検出	不検出	不検出	11,000	不検出	不検出
福島西IC		3月18日 14:30～14:40	不検出	不検出	不検出	9,100	不検出	不検出
杉妻会館		3月18日 14:56～15:06	不検出	不検出	不検出	11,000	不検出	不検出
Bコース 国道399号-114号交差地点		3月18日 14:11～14:31	3.3	1.3	不検出	4,000	2.0	^{58}Co：1.8
Dコース 常葉小学校入り口(田村市)		3月18日 15:00～15:20	不検出	不検出	不検出	4,600	不検出	不検出
二本松市役所		3月18日 10:55～11:05	2.2	不検出	不検出	16,000	不検出	不検出
道の駅　ふくしま東和		3月18日 11:37～11:47	2.1	不検出	不検出	17,000	不検出	不検出
道の駅　安達		3月18日 12:20～12:30	不検出	不検出	不検出	15,000	不検出	不検出

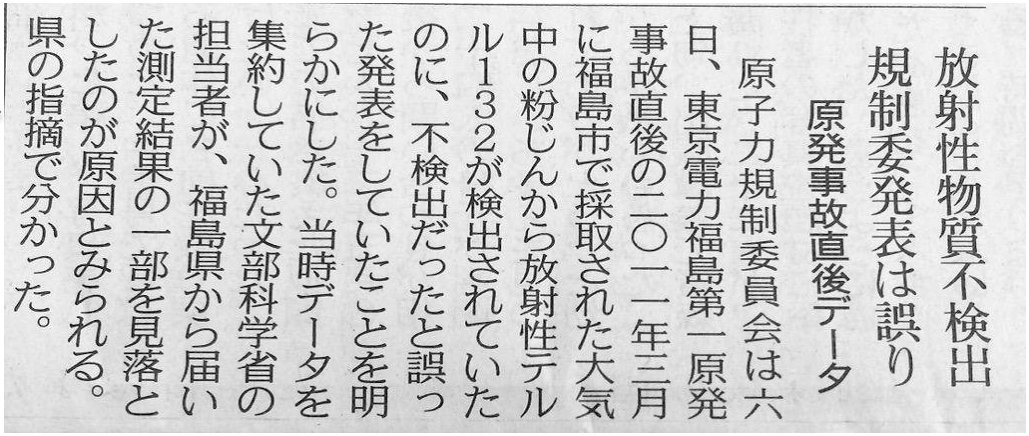
^{99m}Tcの「m」は、原子の持つ軌道電子がエネルギーの高い軌道に遷移してエネルギーの高い励起(準安定)状態であることを表す。
備考欄の番号は、モニタリングカーによる測定箇所を示す。
＊空欄の場所の詳細については確認中。
なお、ここに掲載された測定は、日本原子力研究開発機構により実施されています。

Is this data correct?
I-132 >> Te-132

http://radioactivity.nsr.go.jp/ja/contents/4000/3698/24/1210_201103.pdf

ダストサンプリングの測定結果（平成23年5月31日まで）中の平成23年3月の福島市方木田における測定結果に誤りがありました。

県の指摘で
分かった
県に指摘したのは
どなたか？



放射性物質不検出
規制委発表は誤り
原発事故直後データ

原子力規制委員会は六日、東京電力福島第一原発事故直後の二〇一一年三月に福島市で採取された大気中の粉じんから放射性テルル132が検出されていたのに、不検出だったと誤った発表をしていたことを明らかにした。当時データを集約していた文部科学省の担当者が、福島県から届いた測定結果の一部を見落としたのが原因とみられる。県の指摘で分かった。

東京新聞. 朝刊 (July 7 2015) 3面

ダストサンプリングの測定結果（平成23年5月31日まで）（抜粋）

試料採取日時	放射能濃度（Bq/m^3）					
	I-131	Cs-134	Cs-137	I-132	Te-132	その他検出された核種
2011/3/20 18:30-18:50	203	26.0	32.2	50.0	~~不検出~~ 30.3	~~不検出~~ ^{136}Cs:3.40
2011/3/21 18:30-18:50	2.50	不検出	不検出	不検出	不検出	不検出
2011/3/22 18:30-18:50	3.06	不検出	不検出	9.19	不検出	不検出
2011/3/23 19:38-19:58	3.69	1.71	1.20	4.95	~~不検出~~ 0.966	不検出
2011/3/24 18:30-18:55	不検出	不検出	不検出	不検出	不検出	不検出
2011/3/25 19:10-19:20	24.0	16.1	14.2	不検出	~~不検出~~ 8.54	~~不検出~~ ^{136}Cs:1.53

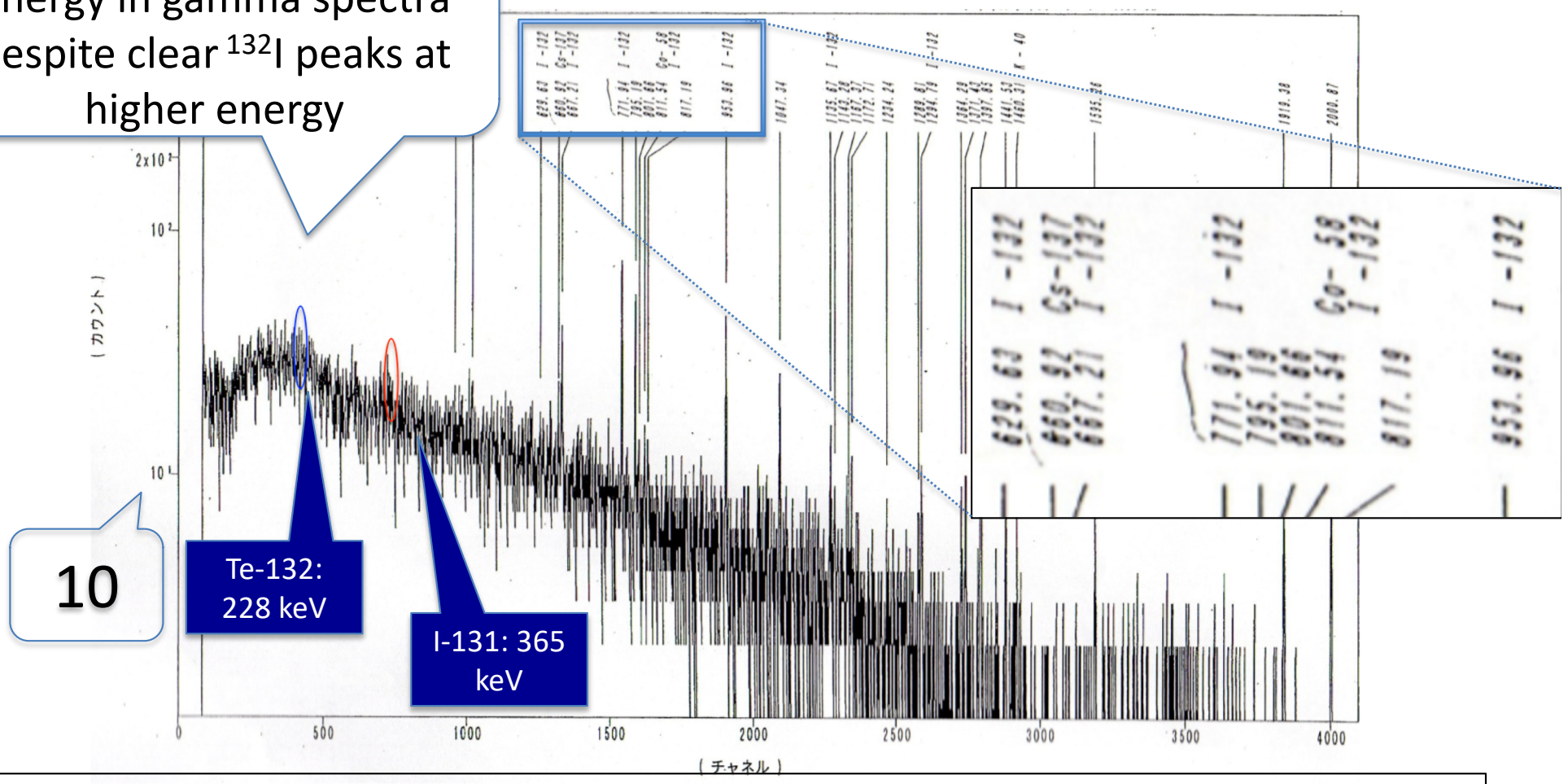
http://radioactivity.nsr.go.jp/ja/list/222/list-1.html

日本保健物理学会シンポジウム Ⅰ「福島事故後の内部被ばくの課題の解決に向けて －不溶性粒子と短半減期核種－」

The other issues?

Observed spectrum

Missing ^{132}Te peaks at low energy in gamma spectra despite clear ^{132}I peaks at higher energy



Location: Fukushima city, air sampling time: 18 Mar, 2011 12:50-13:00, sampled air volume: 1 m^3, filter: charcoal, measurement time: 19 Mar, 2011 15:51-16:08 (1,000 s), detector: HPGe semiconductor

Unusual shape of spectrum

- The low energy peaks may be missing as the spectrum could be showing radiation from the outside environment penetrating through the shield surrounding the germanium detector
- Note: there was no background subtraction
- This would explain the missing ^{132}Te & ^{131}I peaks, i.e. the low energy peaks have been attenuated by the shield
- We will test this theory using simulations

Contents

- Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion

Ultimate goal

- To reconstruct the concentration of radioactive iodine-132 in the air

Contents

- Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion

Methods

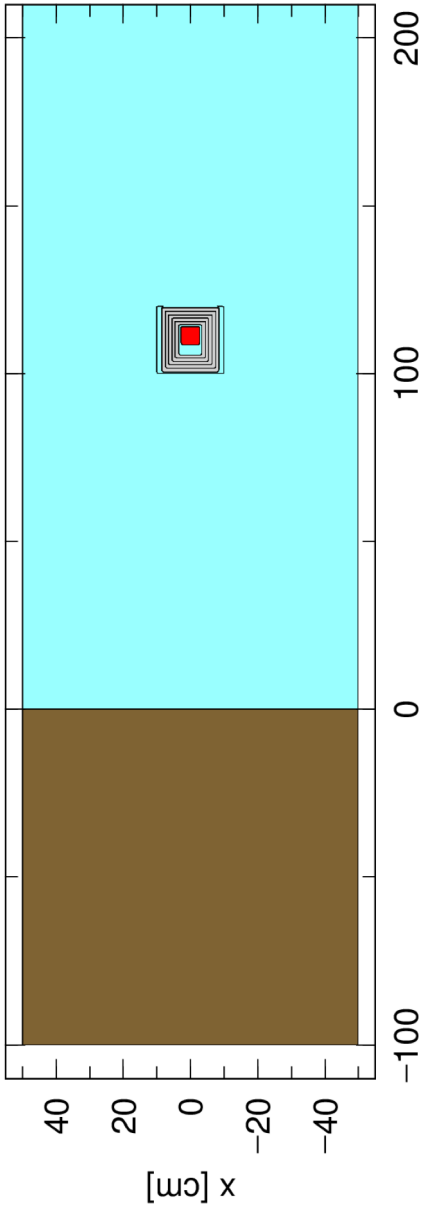
Huge source in the outside environment, small detector, & thick lead shield makes simulations challenging. We adopt a multi-step simulation approach using PHITS.

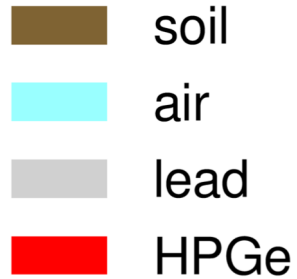
1. Transport photons from radionuclides within environment to edge of shield
   - Using periodic boundary conditions in x,y directions
2. Transport photons to inside of the shield
   - Generate weight windows automatically [T-WWG]
3. Re-transport photons through shield using weight windows
   - Much improved statistics on inside surface of shield
4. Turn on electron transport, calculate response of HPGE detector
   - Required modification of PHITS [T-Deposit] tally

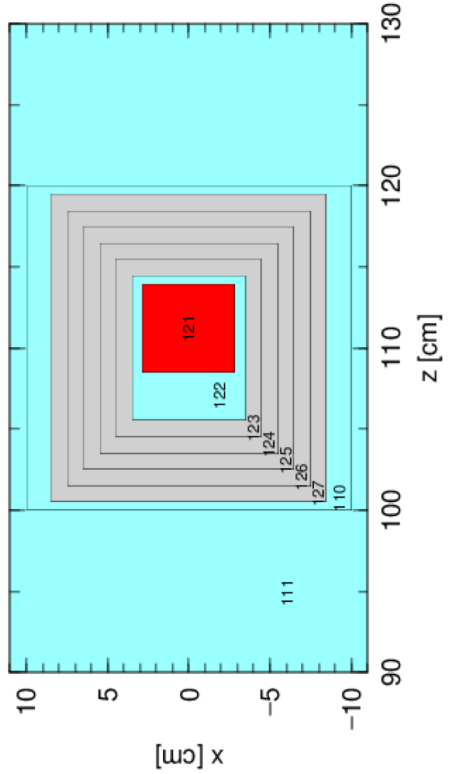
For step 4:
cut-off energy for electron: 100keV
cut-off energy for photon: 3keV

PHITS has been improved to be able to calculate uncertainty correctly for weighted transportation by Dr. Sato

Geometry







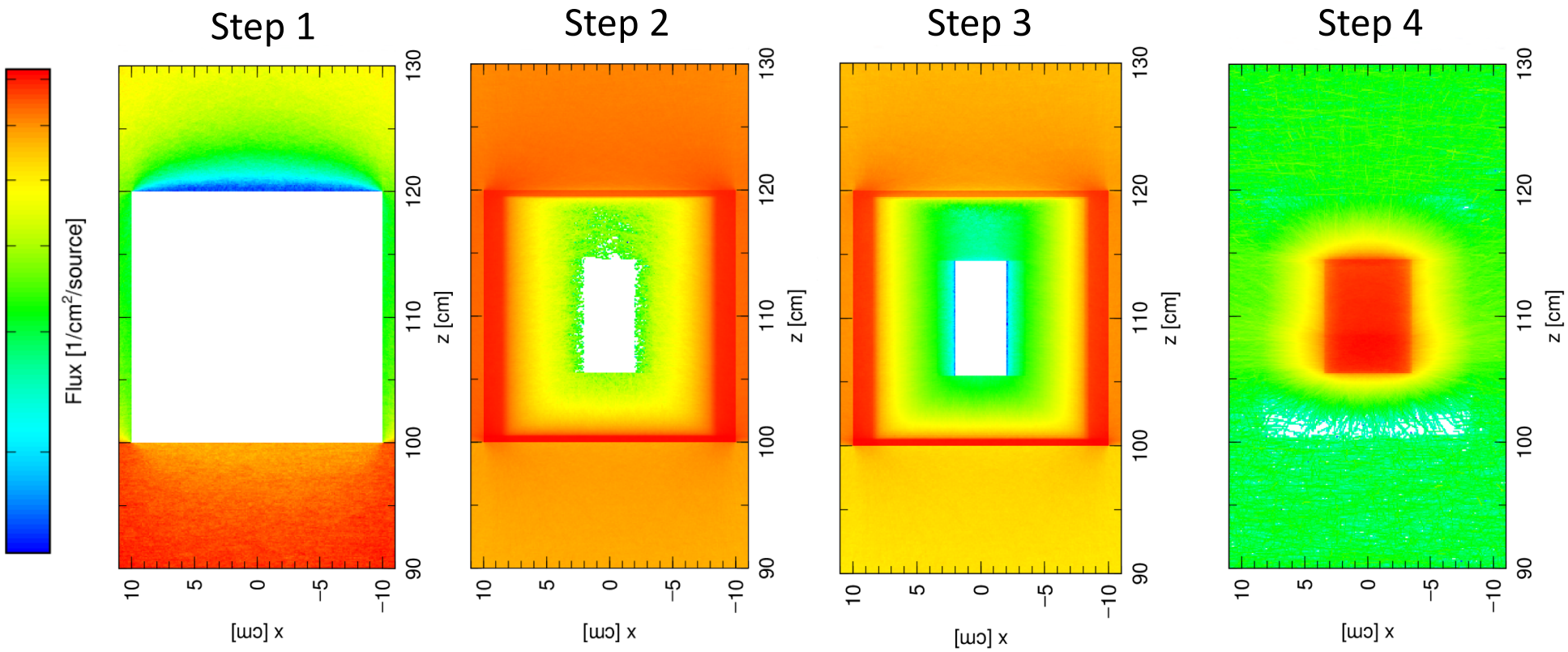
- Variable thickness lead shield (0, 3, 5 cm)
- 5.42 cm length, 5.72 cm diameter HPGe crystal
- Radioactive sources (^{131}I, ^{132}Te, ^{134}Cs & ^{137}Cs) distributed exponentially within soil, β = 0.1 g/cm^2

RI sources
- ground sources -

- Distributed exponentially within the soil, β = 0.1 g/cm^2
  - ^{132}Te considering ingrowth of ^{132}I
    - 0.57 MBq/m^2
  - ^{131}I considering ingrowth of ^{131m}Xe
    - 7.0 MBq/m^2
  - ^{134}Cs
    - 1.0 MBq/m^2
  - ^{137}Cs considering ingrowth of ^{137m}Ba
    - 1.0 MBq/m^2

Activities applicable on 19th March 2011, based on simple decay correction of UNSCEAR 2013 atmospheric release figures

Multi-step simulation



Transport photons to outer edge of shield

Generate weight windows for shield (unbiased)

Transport photons through shield (biased)

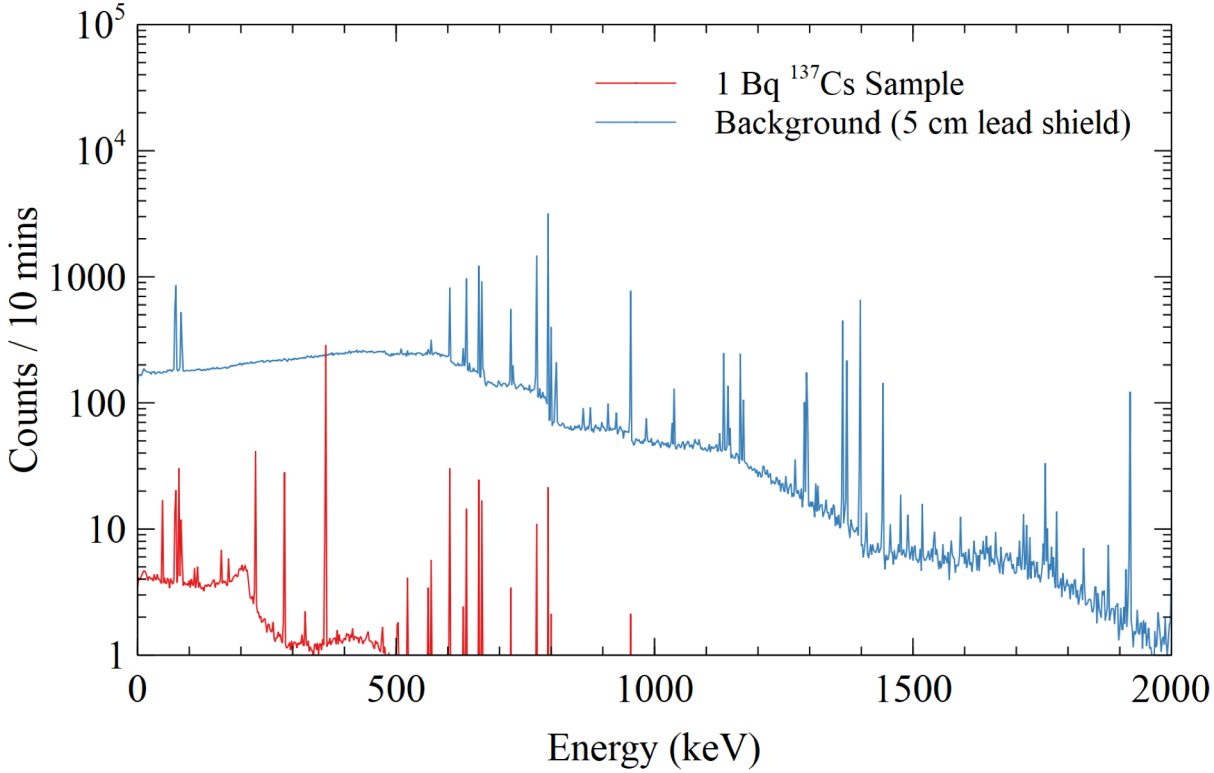
Turn on electron transport, calculate spectrum in detector

Note colours are not directly comparable between figures, but show sampling performance within each step

Contents

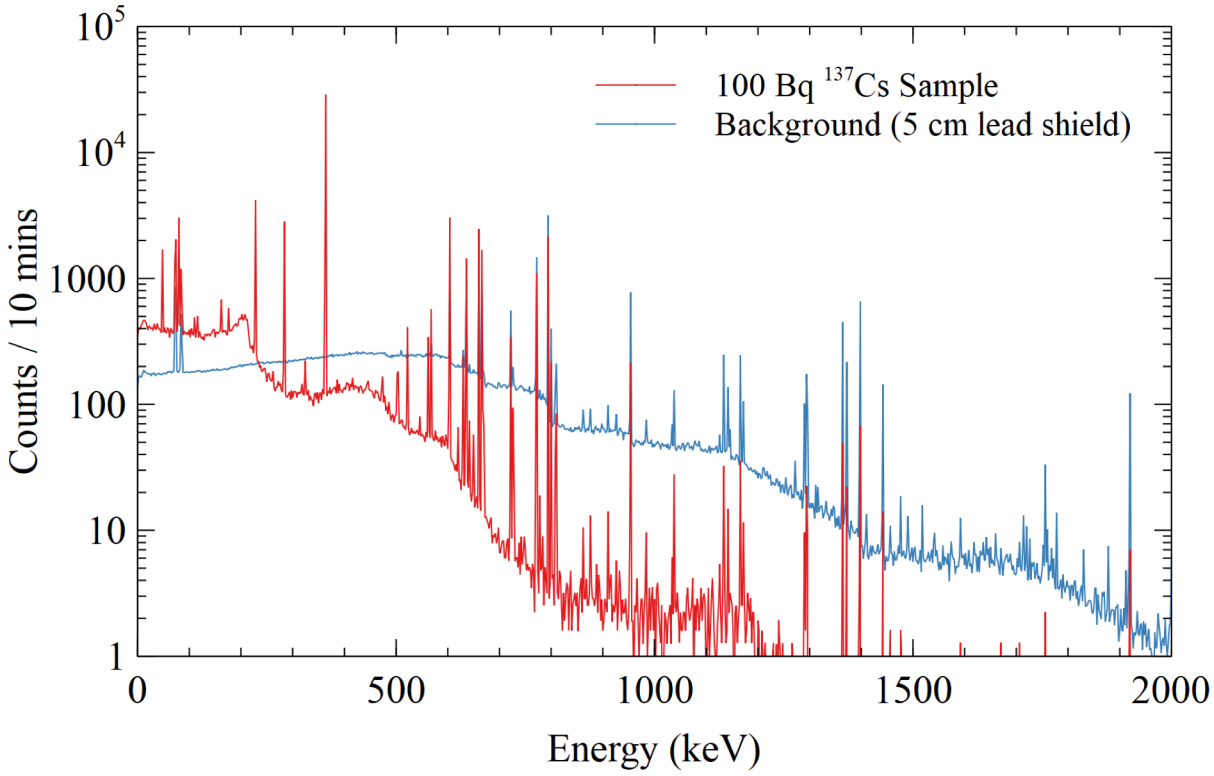
- Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion

Simulated spectra from outside radiation and from 1 Bq ^{137}Cs sample



[Assumptions]
Counting time: 10 min
Detector: HPGe
Outside source: 1 MBq/m^2 ^{137}Cs, activity of ^{131}I, ^{132}Te & ^{134}Cs in UNSCEAR decay-corrected proportions
Sample: 1 Bq ^{137}Cs, and ^{131}I, ^{132}Te & ^{134}Cs with corresponding activity proportions

100 Bq ^{137}Cs on sample



- Increasing the sample activity, the sample peaks at low energy now show up above the spectra from the outside environment (5 cm lead shield)
- However the high energy peaks are still mainly from the radiations coming from the outside environment

Contents

- Background
- Ultimate goal
- Methods
- Results
- Discussion
- Conclusion

Discussion

- Results suggest the shape of the spectrum depends on the relative radioactivity of the sample vs the radioactivity within the outer environment
- Missing peaks at low energy are consistent with the sample activity being very low, and the spectra being due to radiations coming from the outer environment
- If low energy peaks are present, these are most likely to be from the sample. Correspondence with high energy peak count rates could be checked to determine if high energy peaks are from sample or from the outside environment

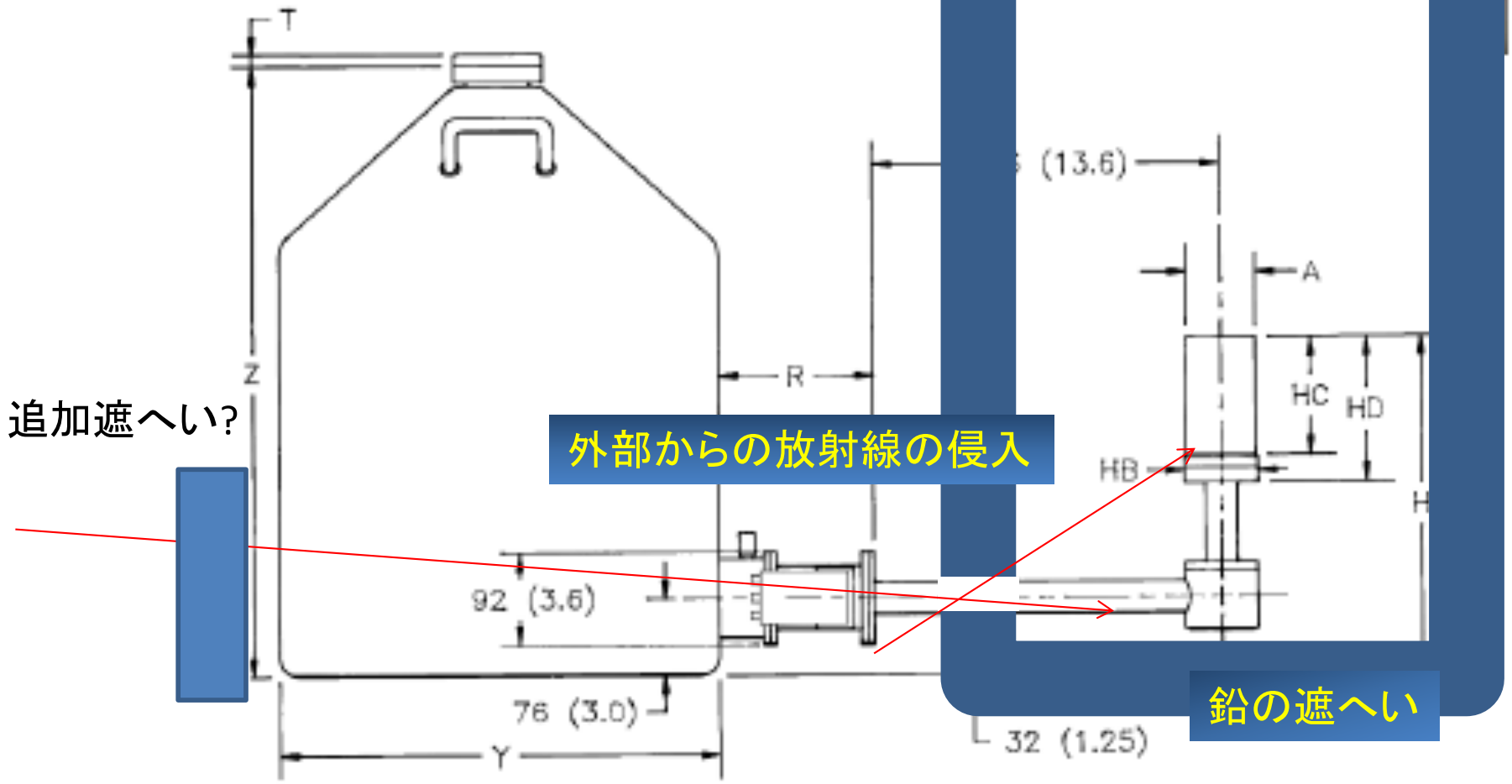
Limitations

- Other issues to be considered
  - Distribution of radionuclides in the environment
    - Non-Uniform source distribution which was fallout deposited on the ground
    - Contamination of equipment and a monitoring car
    - Chemical characteristics differences between a parent and a daughter nuclides

Future plans

- Need to check these effects suggested by the simulations are correct
- Proposed an experiment by taking the remote monitoring vehicle to a high radiation environment (e.g. Ottozawa, Okuma, next to Fukushima Daiichi)
- Check for penetration of ^{137}Cs 667 keV gammas from the outside environment through the shield (i.e. is peak visible with no sample inside the lead shield)

モニタリング・カー



Additional Future plan

- More detailed analysis of measured spectrum
  - Subtraction of back ground data
  - Comparison of each peak considering efficiency of the detector
  - Confirmation on handling regarding HE-40T filters and CHC cartilages

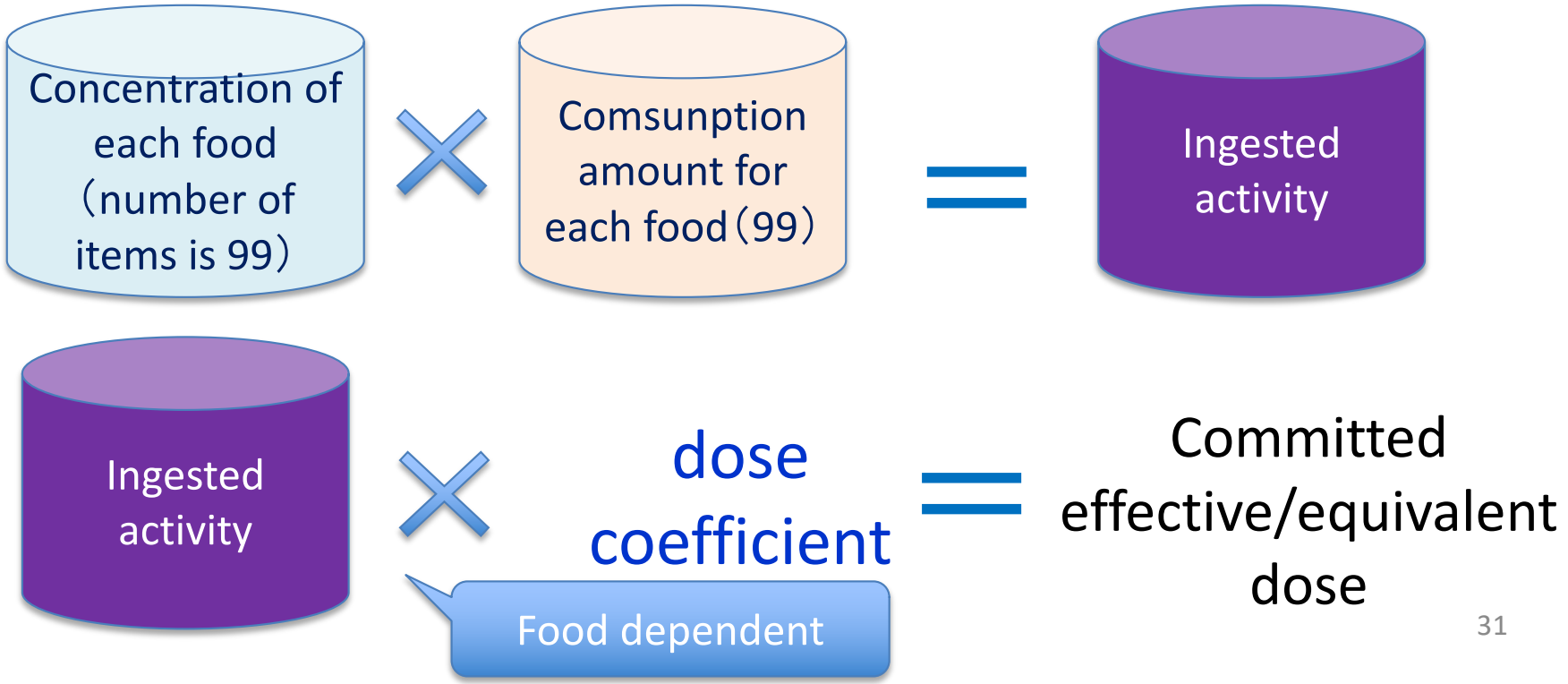
Conclusion of this part

- Re-analysis of the spectrum would be able to improve the quality of dose reconstruction for the early stages of the nuclear accident
  - It has been prepared to carry out simulation for plausible conditions by improving PHITS
  - It has been established to carry out collaborative approaches with relevant researchers including the first responders for environmental monitoring

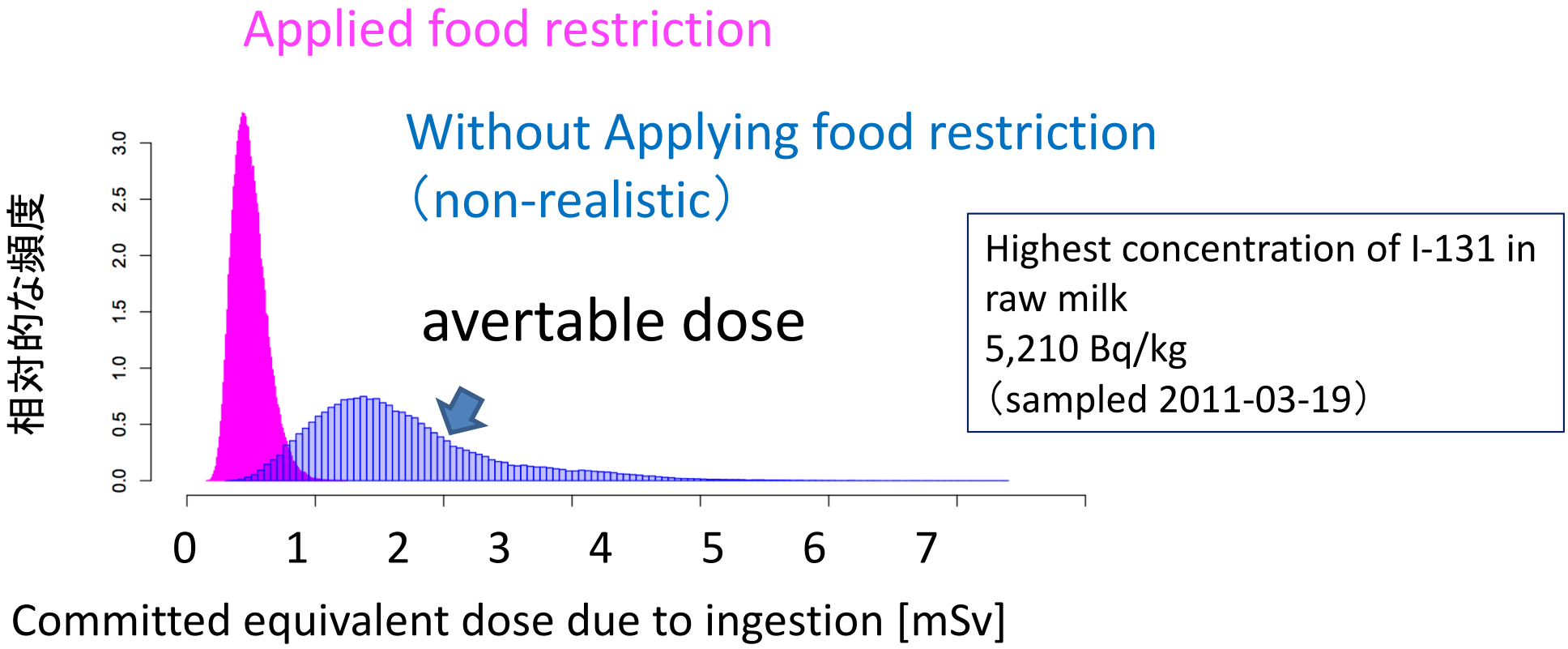
Ingestion dose?

Method

- Ingested activity was calculated by combining concentration (monitoring data) and food consumption and then converted to dose（0.1 million times）



Committed equivalent dose to thyroid due to ingestion, 2011



Highest concentration of I-131 in raw milk
5,210 Bq/kg
（sampled 2011-03-19）

Acknowledgements

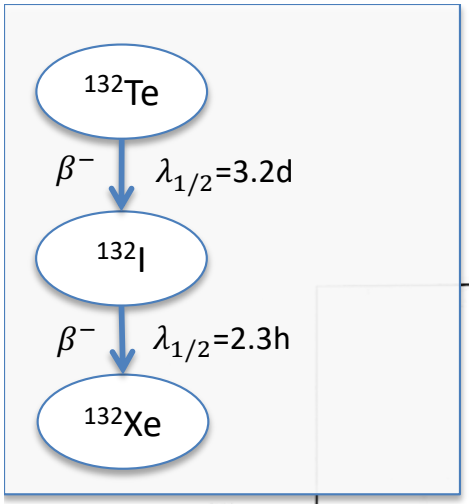
- Dr. Kazumasa Shimada; for his discussions.
- Dr. Tatsuhiko Sato; for his help with the PHITS simulations
- Dr. Takuya Furuta, Dr. Jun Saegusa and Dr. Minoru Takeishi ; for their technical advices.
- @iPatrioticmom; for her many questions.
- This work was supported by Japan Health Physics Society and 環境省．戦略的研究開発領域（戦略研究プロジェクト専門部会）（5-1501）

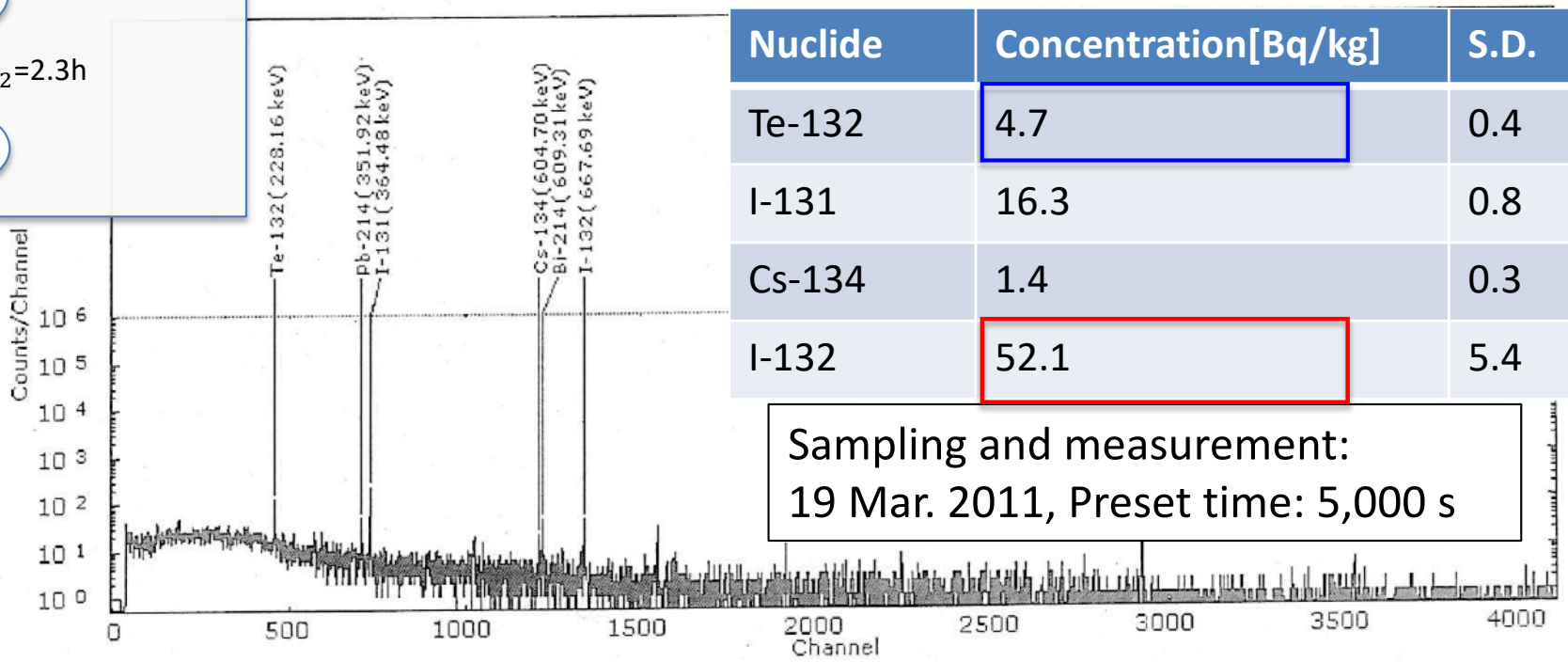
References

- Furuta, Takuya; Takahashi, Fumiaki. Analyses of radiation shielding and dose reduction in buildings for gamma-rays emitted from radioactive cesium in environment discharged by a nuclear accident. JAEA-Research 2014-003,2014
- Takuya Furuta and Fumiaki Takahashi. A computational approach using reflection boundaries for dose calculation in infinitely expanded radiation field. Radiation Protection Dosimetry. 167(4), 392-398,2015.

Supplemental material

The other example of decay adjustment without considering “ingrowth”(Tomato)





Nuclide	Concentration[Bq/kg]	S.D.
Te-132	4.7	0.4
I-131	16.3	0.8
Cs-134	1.4	0.3
I-132	52.1	5.4

Sampling and measurement:
19 Mar. 2011, Preset time: 5,000 s

Software decay corrected back from measurement time of the sample in the spectrometer to the time the tomato was picked from the field.
To be conservative, it performed the correction without considering ingrowth, which gives a higher I-132 activity, i.e.:

$$A_{\mathrm{Te}}(t) = A_{\mathrm{Te}}(0)e^{-\lambda_{\mathrm{Te}}t}$$
$$A_{\mathrm{I}}(t) = A_{\mathrm{I}}(0)e^{-\lambda_{\mathrm{I}}t}$$

However the correct solution (considering ingrowth) is actually:

$$A_{\mathrm{Te}}(t) = A_{\mathrm{Te}}(0)e^{-\lambda_{\mathrm{Te}}t}$$
$$A_{\mathrm{I}}(t) = A_{\mathrm{I}}(0)e^{-\lambda_{\mathrm{I}}t} + \frac{\lambda_{\mathrm{I}}}{\lambda_{\mathrm{I}} - \lambda_{\mathrm{Te}}} A_{\mathrm{Te}}(0)\left[e^{-\lambda_{\mathrm{Te}}t} - e^{-\lambda_{\mathrm{I}}t}\right]$$

Ingrowth term