

2021-FNCA

Surveillance of radioactivity in terrestrial environment and quality control of data

Sang-Han LEE (s.lee@kriss.re.kr)

Korea Research Institute of Standards and Science











Current status of radioactivity monitoring

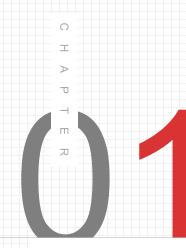






Better Standards, Better Life





Introduction



Better Standards, Better Life 📄

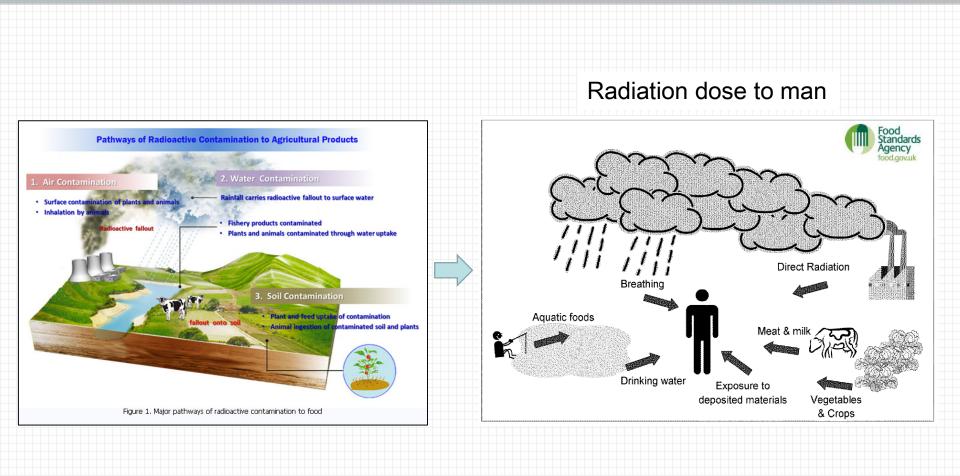
Radioactivity outflow in the environment

- ✓ Following the Fukushima NPP disaster, Nuclear power plant operation and accidental release from the specific source, the concerns regarding radioactive contamination of terrestrial products have continued to spread to the general public
- ✓Thus leading to increased demand for intensive monitoring and survey of radioactivity in the environment.



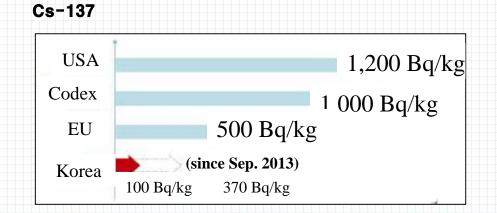
KRISS

Major pathways of radioactive contamination to terrestrial **KRISS**



Guidance levels (Bq/kg) for I-131 and Cs isotopes in Korea KRISS

radionuclide	Fo	od	Guidance level (Bq/kg, L)
	Infant	foods	100
131 I	M	ilk	100
	Foods other	ood It foods Iilk I than infant al foods Drinks Milk and milk products General food	300
	Genera	ıl foods	100
		Drinks	10
$^{134}Cs + ^{137}Cs$	Food imported from Japan		50
		General food	100



- The Brief Information of NORM Regulation KRISS

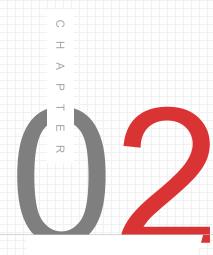
• Definition and Registration level

- **Reference**: IAEA RS-G-1.7, IAEA SRS-49
- U-238 series, U-235 series, Th-232 series, and K-40
- The handler (operator) **should be registered** if they deal with raw

materials and residues over registration levels

Cort	Definition		Registration level	
Sort	Bq/g	kBq	Bq/g	kBq
Raw materials	0.1 (U, Th) 1 (K-40)	100	1 (U, Th) 10 (K-40)	1,000 10,000
Residues	0.5 (U, Th) 5 (K-40)		1 (U, Th) 10 (K-40)	1,000 10,000





Current status of radioactivity monitoring in South Korea



Better Standards, Better Life 📄

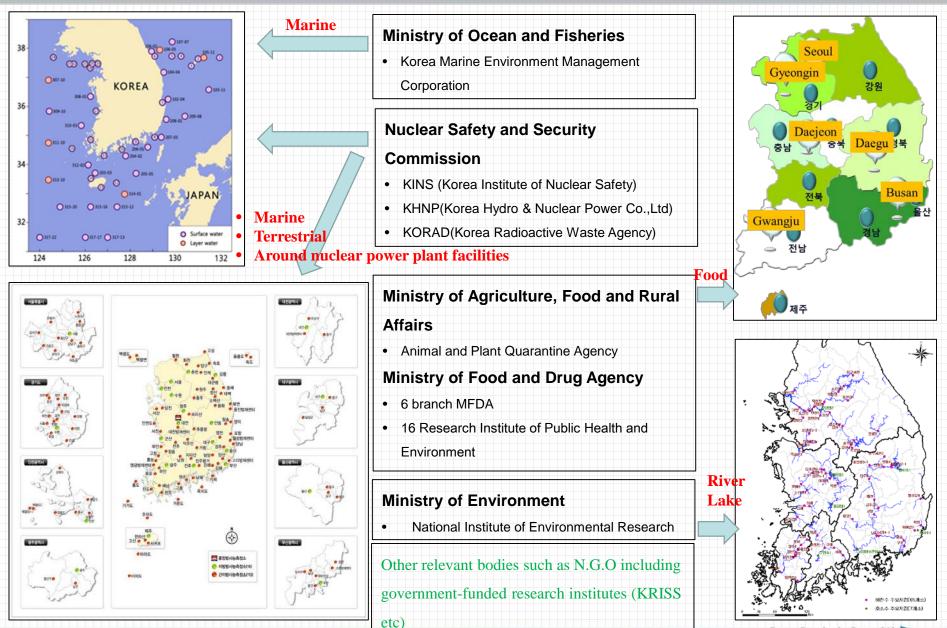
National terrestrial monitoring programme

Why do we need monitoring on environment?

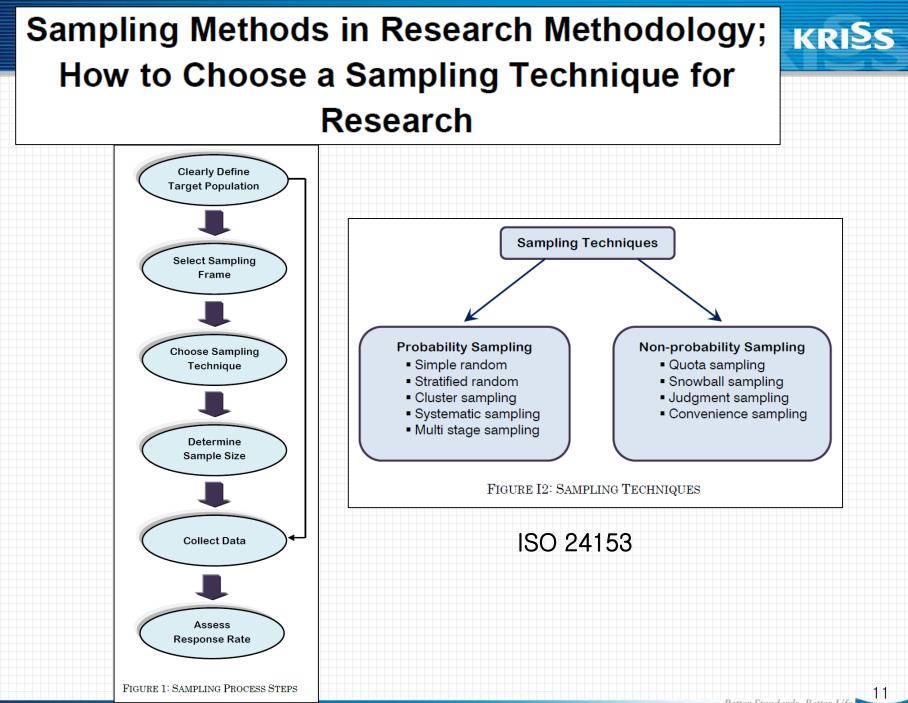
- ✓ Ensuring terrestrial environment is safe
- ✓ Public reassurance
- \checkmark Check against statutory limits and target
- ✓ Carry out dose assessments
- ✓ Long-term trends of radioactivity in environment
- ✓ Identify early signs of change

KRISS

Network of national radioactivity monitoring system in S. Korea KRISS



Better Standards, Better Life



 \mathcal{O}



1. Non destructive methods

Gamma spectrometry (Cs-137, I-131) Nuclear Activation Analysis, XRF etc.

2. Destructive methods

Alpha spectrometry (U-238, Pu isotopes) Beta spectrometry (Sr-90) AAS ICPMS etc. (Isotope mass)

Soil sampling and analysis





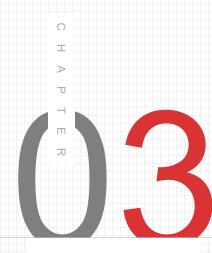
Better Standards, Better Life

Aerosols, rainwater, undergroundwater



KRISS





Case study (soil, foods and atmospheric materials)





Year	Number of samples tested	0.1-<100 (Bq/kg)	¹³⁷ Cs (Bq/kg)
2011	23,144	73	0.1-97.9
2012	26,608	167	0.33-24.7
2013	27,453	66	0.86-9.8
2014	27,567	15	0.92-26
2015	31,502	8	1.0-3.0
2016	32,946	7	1.0-2.0
2017	35,230	4	1.0
2018.8	24,850	3	1.0-28

KRISS

Mushroom sample (edible)



























Better Standards, Better Life

The activity concentration of ¹³⁷Cs in the mushrooms

 \bigcirc



Scientific name	Figure	Number of analyzes	Number of detections (>1 Bq/kg)	¹³⁷ Cs (Bq/kg, fresh)
Lentinula edodes		256 (6	9 from Jeju Isla	1.4 ~ 23 ind)
Phellinus linteus		26	1	1.3
<i>Umbilicaria esculenta</i> lichen	S Lat	3	3	5.9 ~ 10
Tricholoma Matsutake	Ste	14	5	1.3 ~ 2.2
Ramaria botrytis (Pers.) Ricken		3	3	4.0 ~ 20
Sarcodon aspratus	R.	4	4	<mark>9.5</mark> ~ 77
Inonotus Obliquus		6	6	9.1 ~ 36

¹³⁷Cs Contribution from Fukushima NPP



✓ (Sampling site : Jeju Island, Korea)

May 2017	Cs-134 (Bq/kg, dry wt.)	Cs-137 (Bq/kg, dry wt.)	Ν
<i>Lentinula edodes</i> (Shiitake)	0.17-0.29	27-37	3
March 2011 (Decay corrected)	Cs-134 (Bq/kg, dry wt.)	Cs-137 (Bq/kg, dry wt.)	Ν

✓ The activity ratio of ¹³⁷Cs/¹³⁴Cs from Fukushima : *ca.* 1

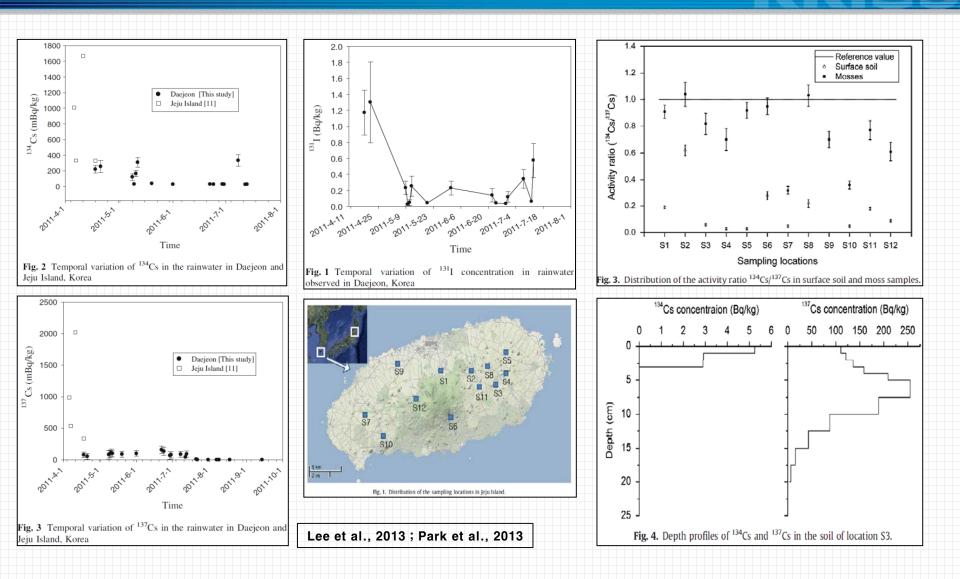
(Leon et al., 2012; Kim et al., 2012; Lujanien et al., 2012)

Therefore, the ¹³⁷Cs contribution from Fukushima NPP accident could

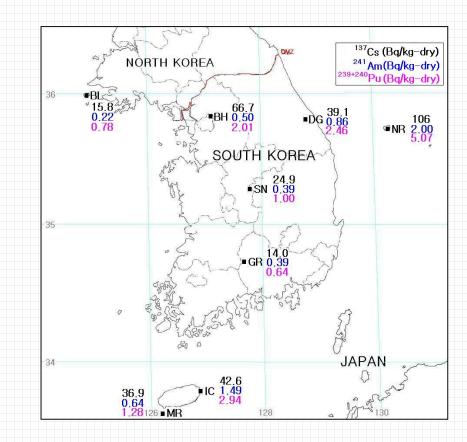
estimate to be ca. 3% of total ¹³⁷Cs content in the Lentinula edodes in Jeju

Island, Korea

- Radioactive impact to Korea from Fukushima NPP KRISS



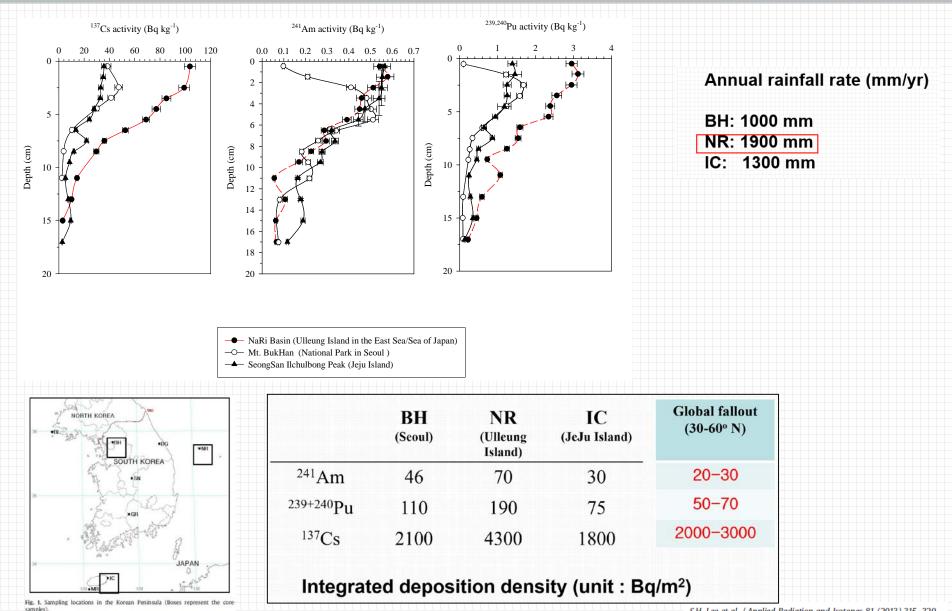
Distribution of ¹³⁷Cs, ²⁴¹Am and ^{239,240}Pu isotopes in surface soils



KRISS

Comparison of vertical profile of radionuclide concentration in the soils

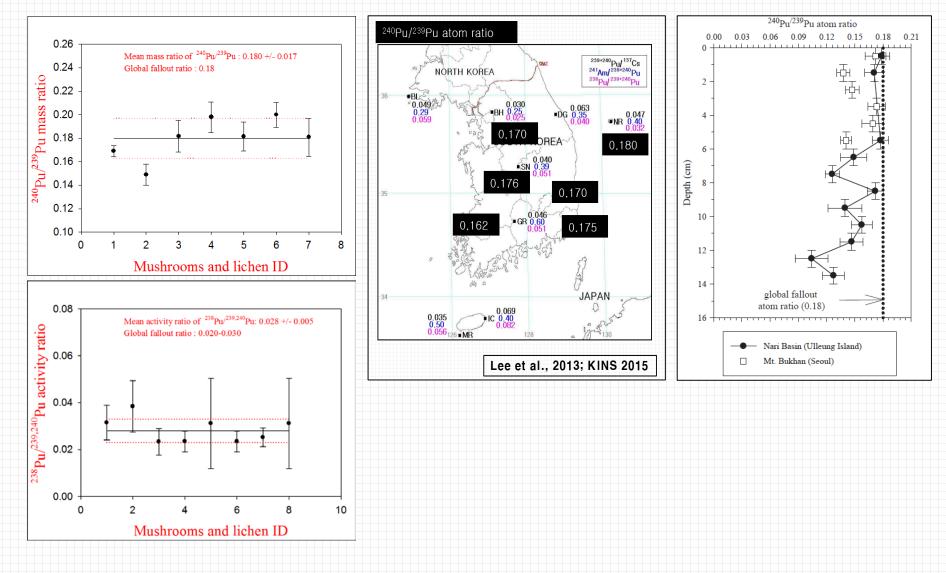




S.H. Lee et al. / Applied Radiation and Isotopes 81 (2013) 315-320

Source of artificial radionuclide in food in Korea KRISS

²³⁸Pu/^{239,240}Pu activity ratio and ²⁴⁰Pu/²³⁹Pu mass ratio



Assessment of dose

KRISS



Annual effective dose for a man in Korea experienced as a result of ingestion of ¹³⁷Cs from mushrooms and fish.

Species	¹³⁷ Cs	Effective dose	Annual	Annual committed
	(Bq/kg of	coefficient ^b	intake ^c	effective dose
	fresh wt.)	(Sv/Bq)	(kg/yr)	(mSv/yr)
Mushroom Fish	8.40^{a} 0.12^{a}	$1.30 imes 10^{-8} \ 1.30 imes 10^{-8}$	2 24	$2.00 imes 10^{-4} \ 3.85 imes 10^{-5}$

^a Mean activity concentration of ¹³⁷Cs measured in the mushrooms and fishes.

^b ICRP 103(2007).

^c National Food & Nutrition Statistics: based on Korea National Health and Nutrition Examination Survey.

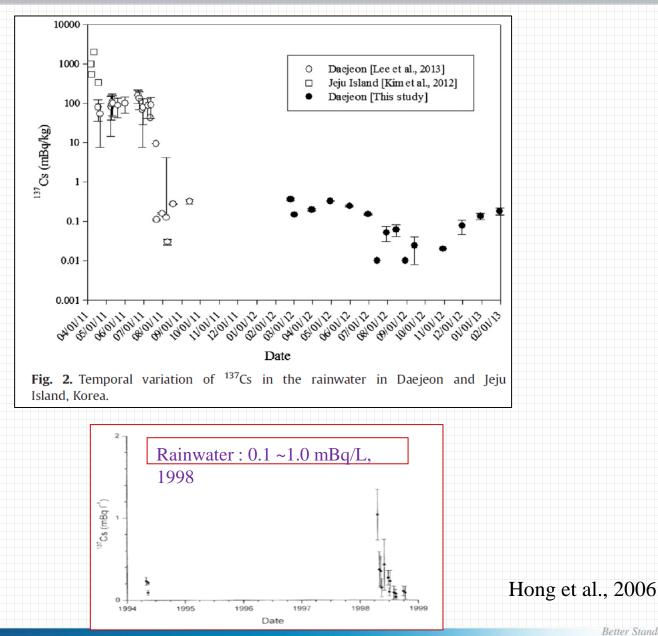
Measurement of I-131 in rainwater after Fukushima NPP accident

Figure 1 Temporal variation of ¹³¹I concentration in rainwater in Daejeon, Korea

Date	¹³¹ (mBq/kg)	Measured Time(s)	
2011-04-18	1,170 ± 24%	86,400	
2011-04-22	1,300 ± 39%	86,400	
2011-05-09	230 ± 36%	71,600	
2011-05-10	24.1 ± 59%	85,000	3.0
2011-05-11	47.0± 76%	86,400	2.5 The theoretical decayed concentration of ¹³¹ I for the rainout event
2011-05-12	252 ± 50%	86,400	$\begin{array}{c} 30\\ 30\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\ 3\\$
2011-05-20	<42.0	86,400	(28 March 2011) (28 March 2011) (28 March 2011) (28 March 2011)
2011-06-01	229 ± 37%	86,400	
2011-06-22	138 ± 60%	86,400	
2011-06-24	<40.3	74,600	$0.0 \qquad $
2011-06-29	<33.4	86,400	20 ¹ , 20 ¹ , Time
2011-06-30	116 ± 58%	86,400	
2011-07-08	343 ± 34%	86,400	
2011-07-12	< 60.4	86,400	
2011-07-13	574 ± 34%	86,400	Better Standard

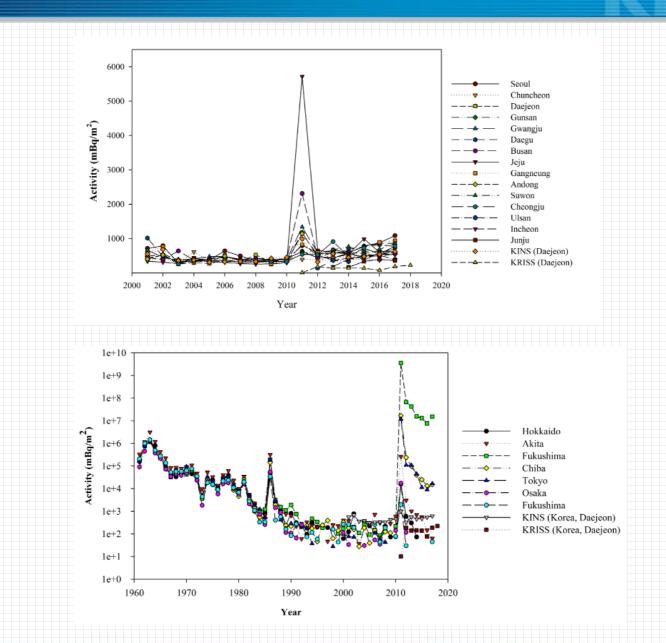
Better Standards, Better Life

Measurement of Cs-137 in rainwater after Fukushima KRISS



Better Standards, Better Life

Annual variation of Cs-137 in the fallout



Better Standards, Better Life

KRISS

Þ	K	R	S	5

	Rainwater		^{239,240} Pu		
Date of Sampling	sample weight (kg)		Uncertainty (<i>k=1</i>)		
May 2012	31.34	7.7	2.6	1.77	
Jun. 2012	53.30	2.6	1.3	1.73	
Jul. 2012 a	61.06	<mda< td=""><td></td><td>2.52</td></mda<>		2.52	
Jul. 2012 b	65.71	<mda< td=""><td></td><td>3.48</td></mda<>		3.48	
Aug. 2012 a	83.16	<mda< td=""><td></td><td>2.37</td></mda<>		2.37	
Aug. 2012 b	73.38	<mda< td=""><td></td><td>3.67</td></mda<>		3.67	
Sep. 2012 a	84.26	<mda< td=""><td></td><td>2.21</td></mda<>		2.21	
Sep. 2012 b	97.14	<mda< td=""><td></td><td>2.27</td></mda<>		2.27	

Concentration of Pu isotopes in rainwater

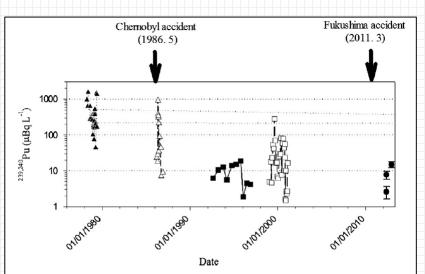
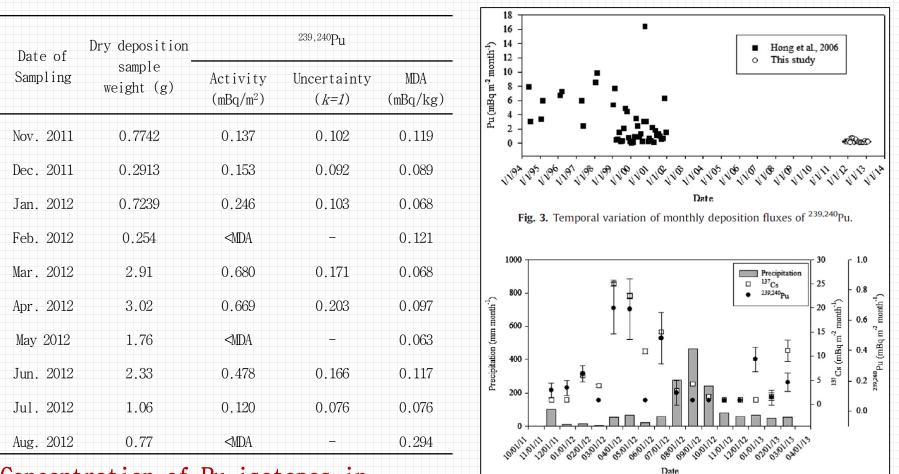


Fig. 1. Temporal variation of ^{239,240}Pu concentrations in rainwater (▲: Thein et al., 1980; ∆: Ballestra et al., 1987; ■: Rubio-Montero and Martin Sanchez, 2001; □: Lee et al., 2002; •: this study).

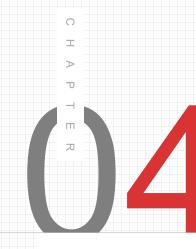




Concentration of Pu isotopes in dry deposition

Fig. 4. Temporal variation of ^{239,240}Pu and ¹³⁷Cs in rainwater with precipitation in Daejeon, Korea.





Regulation of (TE)NORM in COMMODITIES

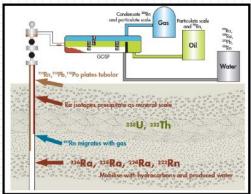


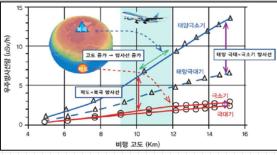
Natural Radiation Source

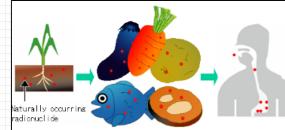
KRISS

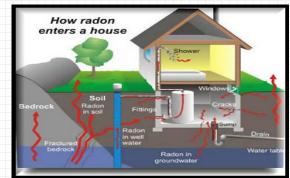
Cosmic radiation

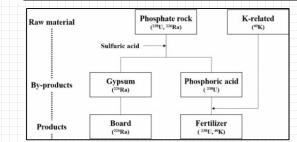
- Cosmic rays : high-energy particles that originate in outer space
- Cosmogenic radionuclides : ³H, ⁷Be, ¹⁴C, ²²Na
- Terrestrial Radiation
- External exposures : soils (²³⁸U series, ²³²Th series, ⁴⁰K), construction materials
- Internal exposures : inhalation (air dust) & ingestion (food, water)
- Radon : gaseous radioactive products (decay of the radium isotopes)
- Enhanced exposures from industrial activities
- Phosphate processing, Metal ore processing, Uranium mining, Zircon sands, Titanium pigment production, Fossil fuels, Oil and gas extraction, Building materials, Thorium compounds, Scrap metal industry, Emissions.











Increased social interest due to radioactivity event (Radioactive asphalt)

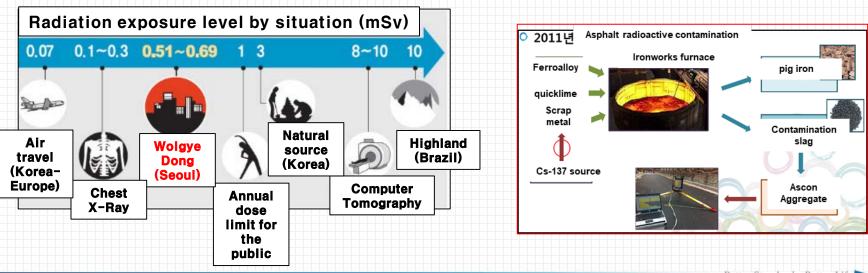




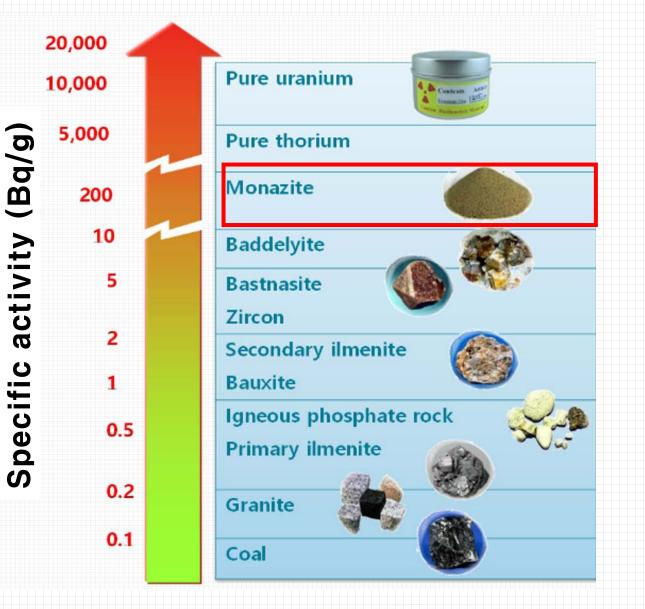
The annual radiation dose that can be received by people living in Wolgye 2-dong residential areas is measured as 0.51-0.69 mSv.

(Annual radiation dose assumes that an adult male stays for 1 hour each day for 1 year).

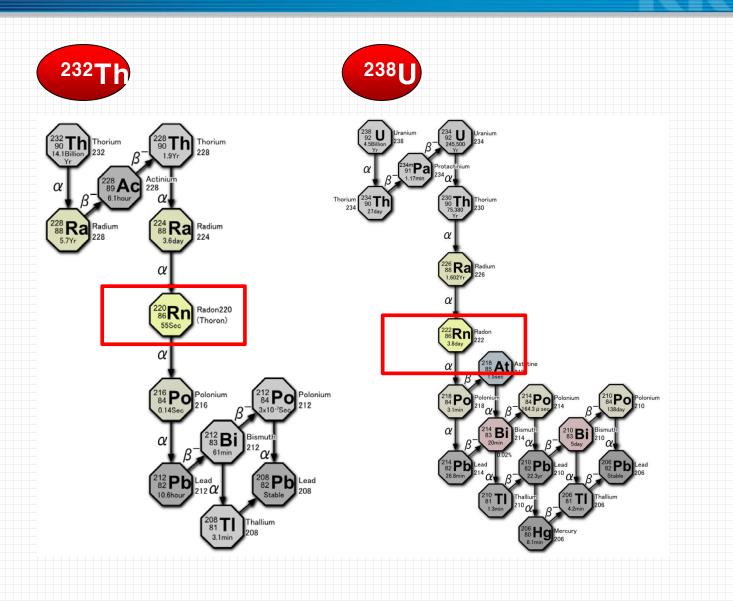
- It is about a sixth to a quarter of the average annual radiation dose in Korea of 3 mSv, which is received from nature such as land, terrestrial, air, and space
- ✓ It is less than 1 mSv, the annual dose limit set by the Nuclear Safety Act.



Radiation levels of major industrial raw materials kriss



Natural radionuclide decay series



KRISS

Increased social interest due to radioactivity event (household products (or commodity) around daily life : anion effect KRISS







국민일보

2009년 10월 09일 금요일 002 종합

건강팔찌·음이온매트 '방사능 범벅'

방사능 방출 분석 결과

시간당 0.19~0.5 # SV

시간당 0.12~0.4 µ SV

7.45

시중에 유통 중인 건강 용품과 건	건축자재 생활	용품 방사능	농도 검출 1
자재에서 기준치를 초과하는 밤사		팔찌(구슬)	- 왕형판(태.5
물질이 다량 검출된 것으로 확인	至最(Th232)	3.9	50
다.	2)唇(Ra226)	0.6	1.3
교육과학기술부가 8일 한나리당 해결 의원에게 제출한 '저준위 등 의 자연 및 인공 발사성 핵종을 함 한 물질에 대한 안전 규제 당한 수	*?** 도표 국제왕	토륨'등	기준 초:
이 관한 연구'에 따르면 팔찌와 지	특히 건강 4	용품으로 인가	

A제품

B제품

되는 라돈 가스는 폐암을 유발하는 트 제료) 지압용 제품 음이온 타월 1급 발암물질로 분류돼 있다. 콘그리 81 0.7 트와 시멘트에서는 라듐이 기준치 이 하로 검출됐지만 견축 환경에 따라 적 은 양으로도 실내 방사능 농도를 높일 과- 건축자재서도 검출 수 있다는 지정이다 방사능 물질이 검출 시메트에는 N라하이 패셨고(이사 지안요 제문에서는 도를 높도가 치

YTN



건강 돌침대서 어울치 이상 방



방사능 된서리 돌침대 "억울해요" [mbn TV 2007-02-12 17:33]



The Brief Information of NORM Regulation KRISs

• Purpose

- To prescribe those matters concerning safety management of radiation encountered in environments
- To protect the public health and the environment and improving quality of life while contributing to public safety

• Structure and other information

- 1 Act, 1 Enforcement Decree, 1 Enforcement Regulation, and 2 Notifications
- Enacted in 2011, and implemented in 2012 by Nuclear Safety and Security Commission(NSSC)

(Due to the issues of radiation exposure from the NORM products

and Fukushima accident)

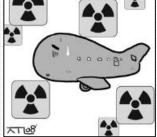
37

The Brief Information of NORM Regulation KRISS

Target of radiation

- NORM : Raw materials, Residues, The Product
- Cosmic radiation (flight exposure to air crew)
- Terrestrial radiation (radon excluded because radon
 - is already regulated by another act)
- Radioactive materials in the scrap metal

Cosmic ray



By-product

Recycled scrap metal

Raw materials

Terrestrial radiation









- The Brief Information of NORM Regulation KRISS

- Definition and Registration level
 - Reference: IAEA RS-G-1.7, IAEA SRS-49
 - U-238 series, U-235 series, Th-232 series, and K-40
 - The handler (operator) **should be registered** if they deal with raw

materials and residues over registration levels

Sort	Definition		Registration level	
	Bq/g	kBq	Bq/g	kBq
Raw materials	0.1 (U, Th) 1 (K-40)	100	1 (U, Th) 10 (K-40)	1,000 10,000
Residues	0.5 (U, Th) 5 (K-40)		1 (U, Th) 10 (K-40)	1,000 10,000

Assessment of dose-Raw material



- The survey on raw materials and by-products was conducted for a total of 15 companies in four areas: workplaces handling raw materials, workplaces generating significant substances, facilities generating by-products, and workplaces suspected of significant substances.
- ✓ The maximum radiation dose rate in areas where there is a risk of external exposure was 0.62 µSv/h, and the maximum radiation dose was evaluated as low as 0.21 mSv/y for the total internal and external exposure.

(KINS/GR-547, 2019)

Assessment of dose-Drinking water



 \checkmark In the production of drinking water, ²¹⁰Pb, a daughter nuclide of radon

discharged with groundwater, can be concentrated in the filter.

- However, as a result of radioactivity analysis, it did not meet the criteria for defining by-products.
- ✓ In addition, the radiation dose to workers was 0.0039 mSv/y.
 (KINS/GR-547, 2019)

Assessment of dose-processed products



- \checkmark The survey on processed products is conducted to analyze the concentration
 - of radioactivity for a total of 513 products and evaluate the annual exposure dose.
- The radon and toron concentrations of all 513 products were measured, and internal exposure doses for 411 products were evaluated. (KINS/GR-547, 2019)
- Of the 411 products, 68 products exceeded the exposure dose of 1 mSv. (17 mattresses, 14 electric mats, 8 blankets, 6 pads, 6 thermal mats, 5 pillows, 4 suit-type underwear, 4 blankets, 2 latex mattresses, 1 hot water mat, 1 sofa, etc.) (KINS/GR-547, 2019)

Assessment of dose-air transportation business kriss



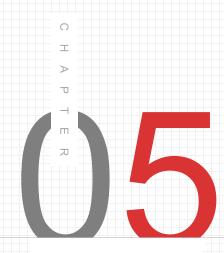
 $\checkmark\,$ According to the survey on air transportation business operators in 2019,

there are 9 business operators in Korea.

- ✓ The number of crew members was 22,484
- ✓ The business operator is using CARI-6/6M and NAIRAS as a program for evaluating the exposure dose by cosmic radiation.
- ✓ It was found that the annual average dose of crew members received 2.09 mSv.
- According to the safety instructions, the flight attendant is recommended that does not exceed 6 mSv
- \checkmark Therefore, it is investigated that no one exceeds the value.

(KINS/GR-547, 2019)





QA/QC for radionuclide data



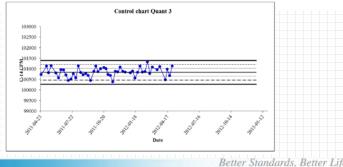
ISO-17025



Quality Assurance & Quality Control

QA : The process used to create the deliverables, and can be performed by a manager, client, or even a thirdparty reviewer (ex. checklist, audits, methodology and standards development)

QC : Quality related activities associated with the creation of project deliverables. Quality control is used to verify that deliverables are of acceptable quality and that they are complete and correct (ex. calibration, control charts, proficiency tests)



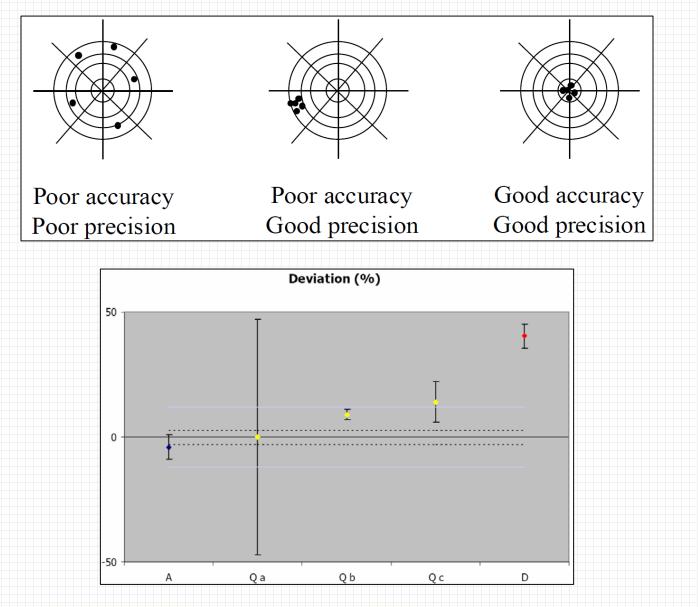
Accuracy and Precision

- Accuracy is a measure of how close the measured value is to the 'true' result (Certified value or reference value)
- The accuracy of any technique is assessed through the measurement of CRMs (certified reference materials) and through participation in national and international proficiency tests.
- Where no standards or proficiency tests are available for the particular analyte of matrix, the accuracy of the measurement may be assessed by spiking a matrix-matched blank (containing none of the analyte) with a known quantity of a standard solution of the analyte and using this as a standard
- Precision is a measure of the scatter of results for repeated analyses.
- The precision of a technique will be dependent on the uncertainties associated with the technique and will deteriorate as the analyte concentration decreases.
- The precision of a technique is assessed by multiple analysis of a homogenous material containing the analyte of interest and determining the standard deviation on the mean of the measurement results.

KRI

Accuracy and Precision





- For any analytical measurement, the reported result will be accompanied by a measurement uncertainty quoted with a given confidence level (usually 95% or 2 s.d.). The quoted uncertainty provides an indication of the precision of the measurement. The true value for the sample will lie (with the degree of confidence quoted) somewhere within the range specified by the uncertainty.
- For example a reported results of 10 ± 2 Bq/kg (2 s.d.) would indicate that there is a 95% probability that the true value is between 8 and 12 Bq/kg. However, there is a 5% chance that the true activity concentration is either lower than 8 or greater than 12 Bq/kg.

Counting statistical uncertainties

Arising from the random nature of radioactive decay, these uncertainties are associated with the radiometric measurement of the purified source and instrument background. The uncertainties will decrease relative to the measured value with increasing counts detected (not count rate). Hence the uncertainties can be reduced by increasing the activity measured (e.g. by increasing the sample size taken) and increasing the count time.

Other method uncertainties

Other uncertainties arising from the methodology and instrument calibration including

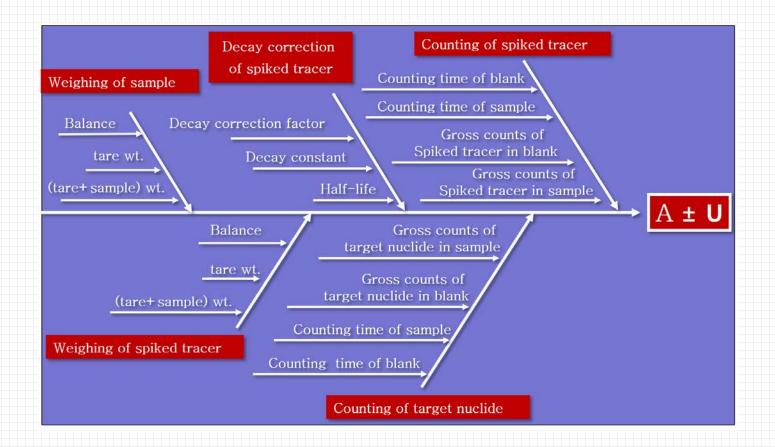
- Sub-sampling
- Weighing / pipetting operations
- Calibration of pipettes, balances or radiometric equipment
- Yield monitor concentration / activity
- Spectral deconvolution / peak fitting
- Decay correction half-life

Uncertainties from all sources are combined to give the total expanded uncertainty. Typically, total expanded uncertainties are around the 5 - 10% of the measured value.

* Fishbone

Measurement uncertainty

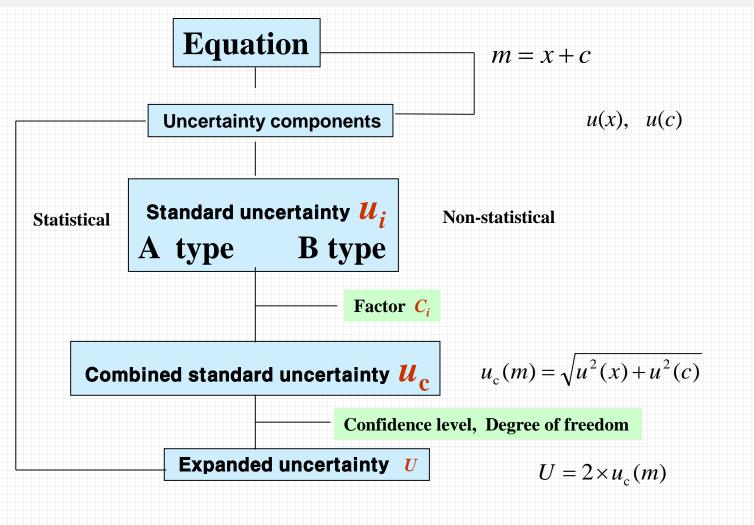
• Expression of uncertainty in measurement results (fishbone)



Better Standards, Better Life



• 5 steps to calculate uncertainty



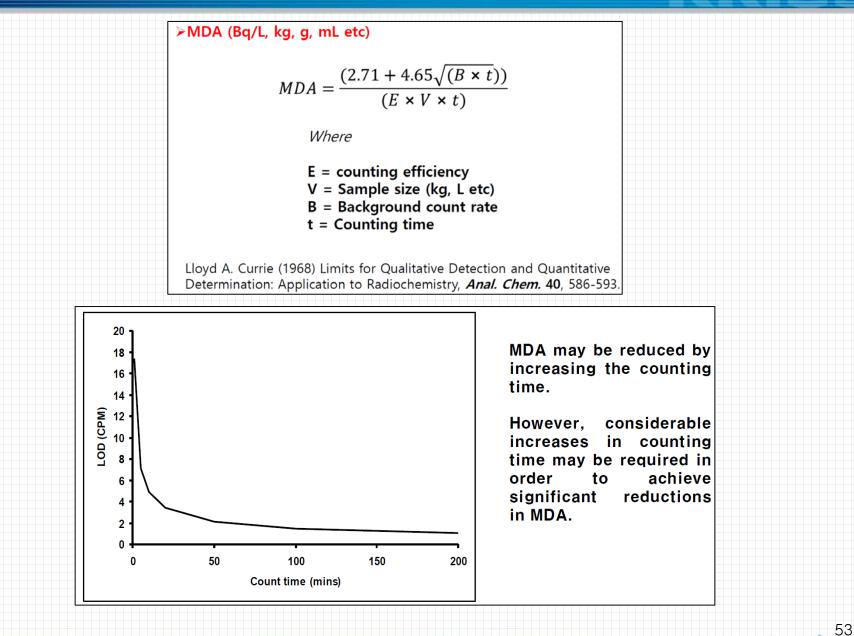
LOD and MDA

- The limit of detection (LOD) is defined for a given measurement process as the smallest true net count rate that is 'certain' to be detected with a specified degree of confidence.
- The minimum detectable activity (MDA) is the detection limit corrected to units of Bq per unit volume or mass of sample.
- > A number of factors affect the limit of detection
- ✓ Method used for calculation !
- ✓ Instrument background
- ✓ Counting efficiency
- ✓ Amount of sample analysed
- ✓ Length of count time

When planning an analytical programme it is important to determine the limits of detection required. In general, lower limits of detection result in higher analytical costs so in many instances project costs can be reduced by realistically setting limits of detection required rather than requesting 'the lowest possible'.

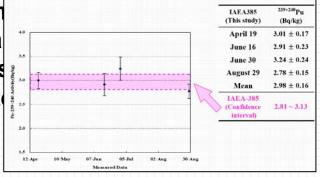
— MDA





The laboratory shall have a procedure for monitoring the validity of results. The resulting data shall be recorded in such a way that trends are detectable and, where practicable, statistical techniques shall be applied to review the results. This monitoring shall be planned and reviewed and shall include, where appropriate, but not be limited to:.

- a) use of reference materials or quality control materials;
- b) use of alternative instrumentation that has been calibrated to provide traceable results;
- c) functional check(s) of measuring and testing equipment;
- d) use of check or working standards with control charts, where applicable;
- e) intermediate checks on measuring equipn
- f) replicate tests or calibrations using the sa
- g) retesting or recalibration of retained items
- h) correlation of results for different charact
- i) review of reported results;
- j) intralaboratory comparisons;
- k) testing of blind sample(s).



The laboratory shall monitor its performance by comparison with results of other laboratories, where available and appropriate. This monitoring shall be planned and reviewed and shall include, but not be limited to, either or both of the following:

a) participation in proficiency testing;

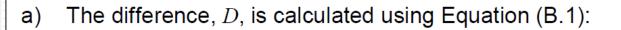
NOTE ISO/IEC 17043 contains additional information on proficiency tests and proficiency testing providers.

Proficiency testing providers that meet the requirements of ISO/IEC 17043 are considered to be competent.

b) participation in interlaboratory comparisons other than proficiency

testing.

KRIS



$$D = (x - X)$$

where

- *x* is the participant's result;
- *X* is the assigned value.
- b) The percent difference, $D_{\%}$, is calculated using Equation (B.2):

$$D_{\%} = \frac{(x-X)}{X} \times 100$$

c)

The *z* scores are calculated using Equation (B.3):

$$z = \frac{x - X}{\hat{\sigma}}$$

where $\hat{\sigma}$ is the standard deviation for proficiency assessment.

As described in ISO 13528, $\hat{\sigma}$ can be calculated from the following:

- a fitness for purpose goal for performance, as determined by expert judgement or regulatory mandate (prescribed value);
- an estimate from previous rounds of proficiency testing or expectations based on experience (by perception);
- an estimate from a statistical model (general model);
- the results of a precision experiment; or
- participant results, i.e. a traditional or robust standard deviation based on participant results.

KRISS

(B.3)

ISO/IEC 17043-performance for quantitative results

e) E_n numbers are calculated using Equation (B.5):

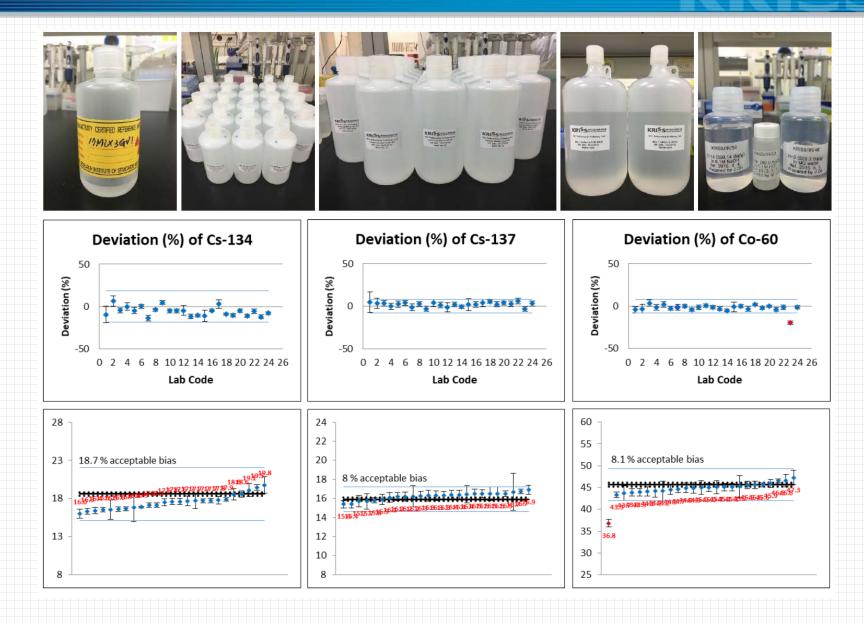
$$E_n = \frac{x - X}{\sqrt{U_{\mathsf{lab}}^2 + U_{\mathsf{ref}}^2}}$$

where

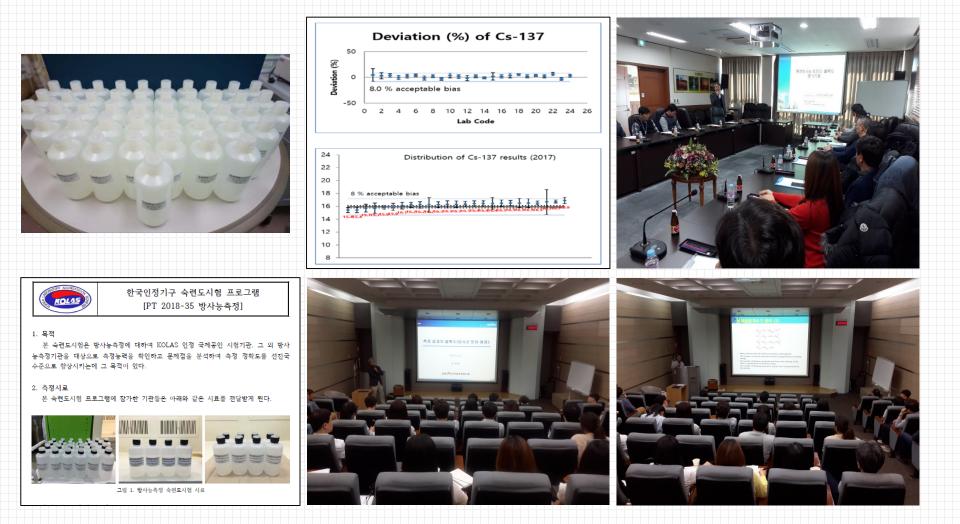
 U_{lab} is the expanded uncertainty of a participant's result;

U_{ref} is the expanded uncertainty of the reference laboratory's assigned value.

Proficiency Test



– Proficiency Test-seminar, symposium



- Crucial activities for production for CRM кы§s ISO Guide 30~35

- All or some of the following activities can be crucial in RM
- production and their quality assessment can be crucial to the quality of the final RM:
- ✓ Sourcing of materials including synthesis
- Processing of materials including purification, grinding,
 - particle size separation
- Homogeneity and stability testing
- ✓ Development and validation of measurement procedures,

including consideration of the traceability

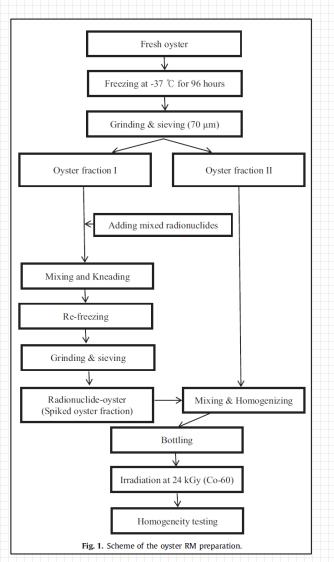
Measurement of property values and evaluation of

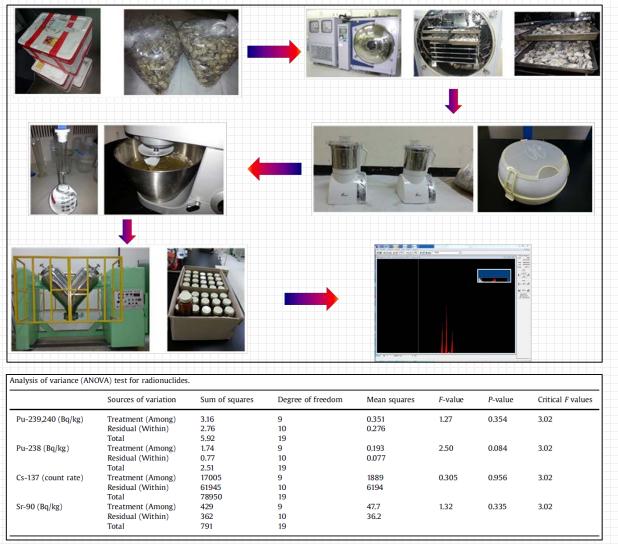
measurement uncertainty

Certification and sign off of the RM

Korea Laboratory

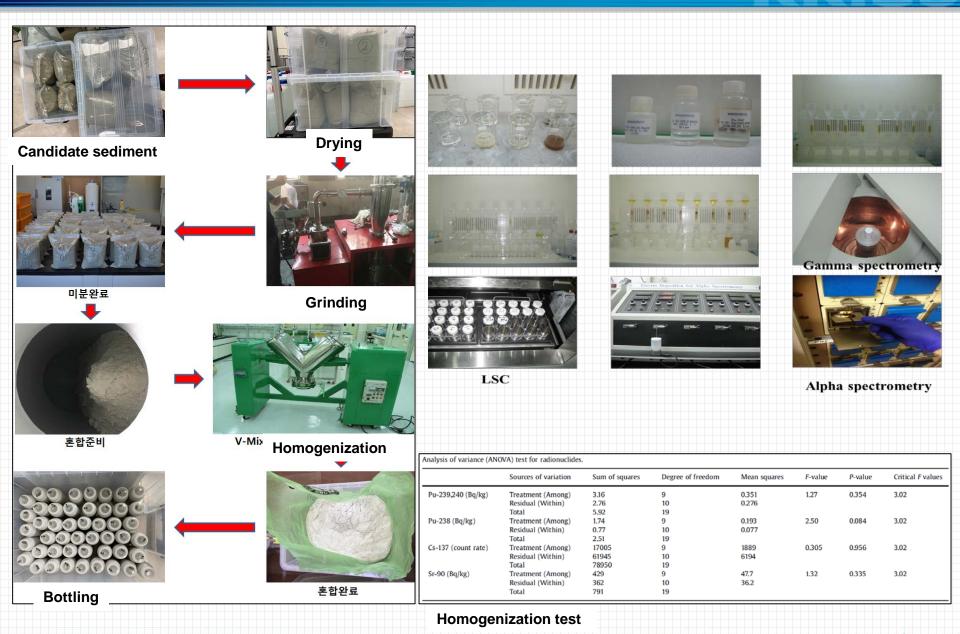
-Development of Oyster CRM (ISO Guide 35) KRISS





S.-H. Lee et al. / Applied Radiation and Isotopes 109 (2016) 109–113

Development of Sediment CRM (ISO Guide 35)



CRMs and international comparison exercise

- International comparison exercise materials for radioactivity measurement of (mushroom) material developed by KRISS (BIPM CCRI(II)-S15)
- International comparison exercise materials for radioactivity measurement of (rice) material developed by KRISS (BIPM CCRI(II)-S9)



ご清聴ありがとうございました

표준이 올라가면 생활이 즐거워 집니다!

Thank you for your attention!

감사합니다 KRISS 한국표준과학연구원