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Note

A Comparative Study of the Outdoor Absorbed Dose Rate in Air by *In-situ* and Soil-sampling-based Measurement Methods

Chutima Kranrod[1, 2]*, Supitcha Chanyotha[2], Phongphaeth Pengvanich[2], Rawiwan Kritsananuwat[2], Masahiro Hosoda[1, 3] and Shinji Tokonami[1]

[1]*Institute of Radiation Emergency Medicine, Hirosaki University, Hirosaki, Aomori 036-8564, Japan.*
[2]*Natural Radiation Survey and Analysis Research Unit, Department of Nuclear Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand*
[3]*Hirosaki University Graduate School of Health Sciences, Hirosaki, Aomori 036-8564, Japan*

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Outdoor absorbed dose rates in air were evaluated in the environs of Eastern, Western, and Southern Thailand using the gamma-ray pulse height distribution obtained by *in-situ* NaI(Tl) scintillation spectrometer, and by analyses of soil samples for ^{226}Ra, ^{232}Th, and ^{40}K activity concentration using an HPGe gamma spectrometry. The geometric mean values of the outdoor gamma dose rates from the direct measurements and the soil analyses were 45 ± 8 nGy/h and 69 ± 3 nGy/h respectively. The ratio of the average absorbed dose rate in air inferred from the activity concentrations of radionuclides in soil to the average absorbed dose rate in air from the *in-situ* measurement in this study is 1.5.

Key words: car-borne survey, *in-situ*, gamma spectrometry

1. Introduction

The daily radiation exposure to human normally comes from various natural background sources such as food, air, elements in the body, building materials, ground and outer space[1]. Gamma radiation emitted from primordial radionuclides and their progenies are one of the main external sources of radiation exposure to human. The most important naturally occurring radionuclides in rocks and soils are the radioactive isotope of potassium (^{40}K) and the radionuclides from the thorium (^{232}Th) and uranium (^{238}U) decay series[2]. The contributions on the terrestrial absorbed dose rate in air as gamma radiation from these primordial radionuclides are approximately 35% from ^{40}K, 25% from ^{238}U series and 40% from ^{232}Th series[3]. The global mean value of this dose according to the distribution of the population ranges from 50 to 59 nGy/h[4].

Collection of soil samplings for laboratory radioactivity analysis using gamma-ray spectroscopy has been employed by many researchers for environmental radioactivity monitoring[5-7]. The reliability and simplicity of this method has been well established by using the same geometry for the reference standard material and the sample from the point of measurement. In-situ gamma spectrometry on the other hand identifies the average contributions of individual radionuclides over a large area around the point of measurement, resulting in less statistical precision of the obtained data than that

*Chutima Kranrod : Institute of Radiation Emergency Medicine, Hirosaki University, Hirosaki, Aomori 036-8564, Japan.
E-mail: kranrodc@hirosaki-u.ac.jp

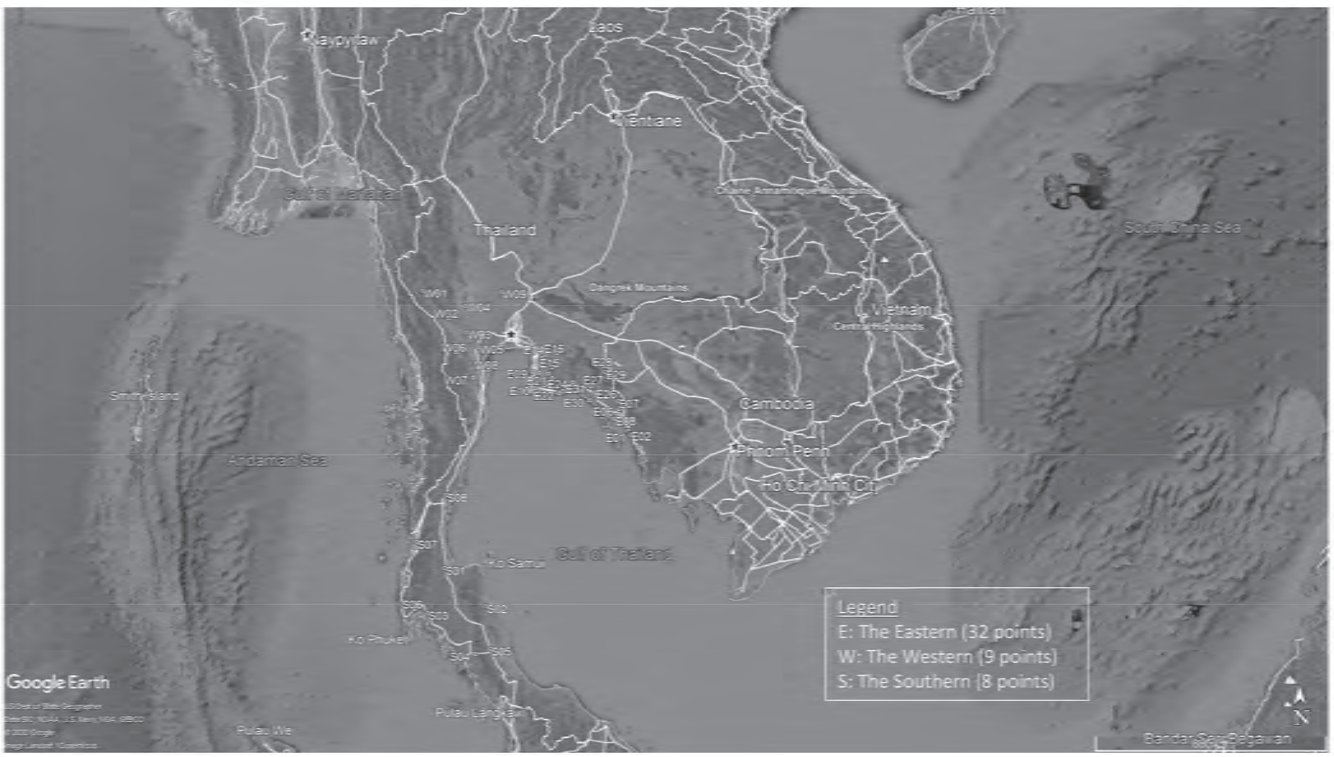

Fig. 1. The *in situ* measurement and soil sampling points[26]

obtained by soil-sampling-based evaluations[8-10].

The objective of the present study is to make comparison between two outdoor gamma dose rate measurement methods, in-situ and soil-sampling-based. For this purpose, *in-situ* measurement utilizing the gamma-ray pulse height distribution obtained by a 3-in × 3-in NaI(Tl) scintillation spectrometer was carried out in three regions of Thailand. The data obtained in the frame of this study was used to compare against the results from the recently published soil-sampling-base measurement[11] in the same areas.

2. Material and Methods

2.1 Survey Area

The absorbed dose rates in air (nGy/h) from the natural radionuclides including ^{40}K, ^{238}U series and ^{232}Th series were measured during March–May, 2017, June, 2017 and February, 2018 in the eastern, western, and southern Thailand, respectively. The study areas are shown in Figure 1.

2.2 In-situ measurements by the system for car-borne survey

In-situ measurements were carried out using the system for car-borne survey with a 3-in × 3-in NaI (Tl) scintillation spectrometer (EMF-211, EMF Japan Co., Osaka, Japan). Measurement of gamma-ray pulse height distributions were performed at 1 m above the ground surface outside the car for 15 min at 49 points (Fig. 1). About 70% of measurement points were taken in the agricultural fields along several main roads near city centers and residential area and 30% were measured at reclamation land. These observations were carried out on private land after obtaining specific permissions from the land owners and it was also confirmed that the field studies did not involve endangered or protected species. The gamma-ray pulse height distributions were unfolded using a 22 × 22 response matrix for the estimation of absorbed dose rate in air[12]. In this study, the energy bins were set to 1.39–1.54 for ^{40}K, 1.69–1.84 MeV and 2.10–2.31 MeV for ^{214}Bi (^{238}U series) and 2.51–2.72 MeV for ^{208}Tl (^{232}Th series) to unfold the gamma-ray pulse height distribution. The energy intervals for the bins were given by Minato[13].

2.3 Soil sample collection and measurement of radionuclide activity concentrations

A total of 49 surface soil samples (from 0 to 5 cm soil profile) were collected from those locations where in-situ measurement was performed (Fig. 1). The soil samples were oven dried at 110℃ until they reached constant weight. The dried samples were pulverized into a fine powder and sieved through a 250 μm mesh

Table 1. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq/kg) and the absorbed dose rate in air by *in-situ* measurement.

Sampling region (no. of sample point)	Concentration (Bq/kg)						The outdoor absorbed gamma dose rates (nGy/h)	
	^{238}U		^{232}Th		^{40}K			
	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)
The eastern (32)	9-77	34 (3)	4-62	29 (4)	23-3043	353 (29)	7-101	42 (7)
The western (9)	13-92	44 (2)	23-118	39 (3)	30-486	281 (16)	24-96	57 (14)
The southern (8)	15-75	41 (2)	15-79	35 (1)	36-304	170 (7)	15-90	49 (6)
Average	9-92	36 (1)	4-118	31 (2)	15-3043	305 (11)	7-101	45 (8)

Table 2. The activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq/kg-dry) and the outdoor absorbed gamma dose rate in air estimated for the soil samples.

Sampling region (no. of sample)	Concentration in soil (Bq/kg-dry)						The outdoor absorbed gamma dose rates (nGy/h)	
	^{226}Ra(^{238}U)		^{228}Ra(^{232}Th)		^{40}K			
	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)	Range	Average (S.D.)
The eastern (32)	5-132	41 (4)	2-165	55 (1)	10-1270	380 (15)	6-191	68 (3)
The western (9)	13-92	31 (3)	23-118	51 (1)	47-1414	465 (20)	23-170	65 (5)
The southern (8)	11-77	40 (4)	20-126	63 (1)	30-965	448 (20)	18-124	75 (6)
Average	5-132	37 (2)	1-165	56 (1)	10-1414	431 (11)	6-191	69 (3)
Worldwide median	16-110	35	11-64	30	140-850	400		51

size sediment sieve. All homogenized samples were analyzed for activity concentration of ^{226}Ra, ^{228}Ra and ^{40}K using gamma spectroscopy. The samples were packed into airtight plastic containers and sealed to prevent the escape of radon (^{222}Rn) and thoron (^{220}Rn) gases. Prior to measurement, the sample was stored for at least four weeks in order to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their radioactive progenies. Activity concentrations in the samples were measured using a high-purity germanium (HPGe) detector (GEM25P4-76, ORTEC, USA.) with a relative efficiency of 30% and 2.1 keV energy resolution (FWHM) at 1.332 MeV of ^{60}Co. The detector is connected to the multi-channel analyzers (MCA; DSPEC jr 2.0, ORTEC, USA.) and gamma-ray energy peak analysis using the MAESTRO Mca32 program. The detector was shielded with 10 cm lead to reduce the gamma radiation from the environment into interference with the radiation metering system. Energy and efficiency calibrations of the detector were carried out using three different IAEA standard reference materials including IAEA-RGU-1, IAEA-RGTh-1 and IAEA-RGK-1. The calibrations used known activity standards (IAEA) presented in the same geometry as the soil samples by determining the absolute efficiency as a function of energy and then used emission probabilities from the literature to calculate concentrations in soil. Counting time for each sample was set at 24 h. By assuming secular equilibrium with their progenies in the ^{238}U and ^{232}Th decay chains, the activity concentration of ^{226}Ra was calculated using gamma-rays associated with the decay products of ^{214}Bi (609.3 keV) and ^{214}Pb (351.9 keV). In the case of ^{232}Th activity concentration, the gamma-ray lines of 911.2 keV from ^{228}Ac and 583.2 keV from ^{208}Tl were used. The activity concentration of ^{40}K was derived directly from the measured intensity of its photon peak at 1,460.8 keV[14-15].

2.4 The absorbed dose rate in air from soil sample

In secular equilibrium conditions, activity concentrations of ^{238}U and ^{226}Ra in uranium series and ^{228}Ra and ^{232}Th in thorium series remain the same. Thus, the absorbed dose rate in air can be evaluated from the activity concentrations of the ^{226}Ra, ^{228}Ra and ^{40}K measured in soil samples. If natural gamma sources uniformly distributed in the ground, the absorbed dose rate in air at 1 m above the ground can be calculated by using the following equation (1) based on the data provided by Saito and Jacob[16]

$$D_{air} = 0.462 \cdot A_U + 0.604 \cdot A_{Th} + 0.0417 \cdot A_K \quad (1)$$

where A_U, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K respectively in Bq/kg. The coefficients of 0.462, 0.604 and 0.0417 are conversion factors (absorbed dose rate in air per unit activity per unit of soil mass, in units of nGy/h per Bq/kg) evaluated for ^{238}U-series, ^{232}Th-series and ^{40}K, respectively[17].

Table 3. *In-situ* and laboratory measurements of average activity concentrations of ^{238}U, ^{232}Th, and ^{40}K and their respective ratios for comparison.

In-situ measurements (Bq/kg) (S.D.)			Laboratory measurements (Bq/kg) (S.D.)			Ratio (In situ/Laboratory)		
^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K
36 (1)	31 (2)	305 (11)	37 (2)	56 (1)	431 (11)	0.97	0.55	0.71

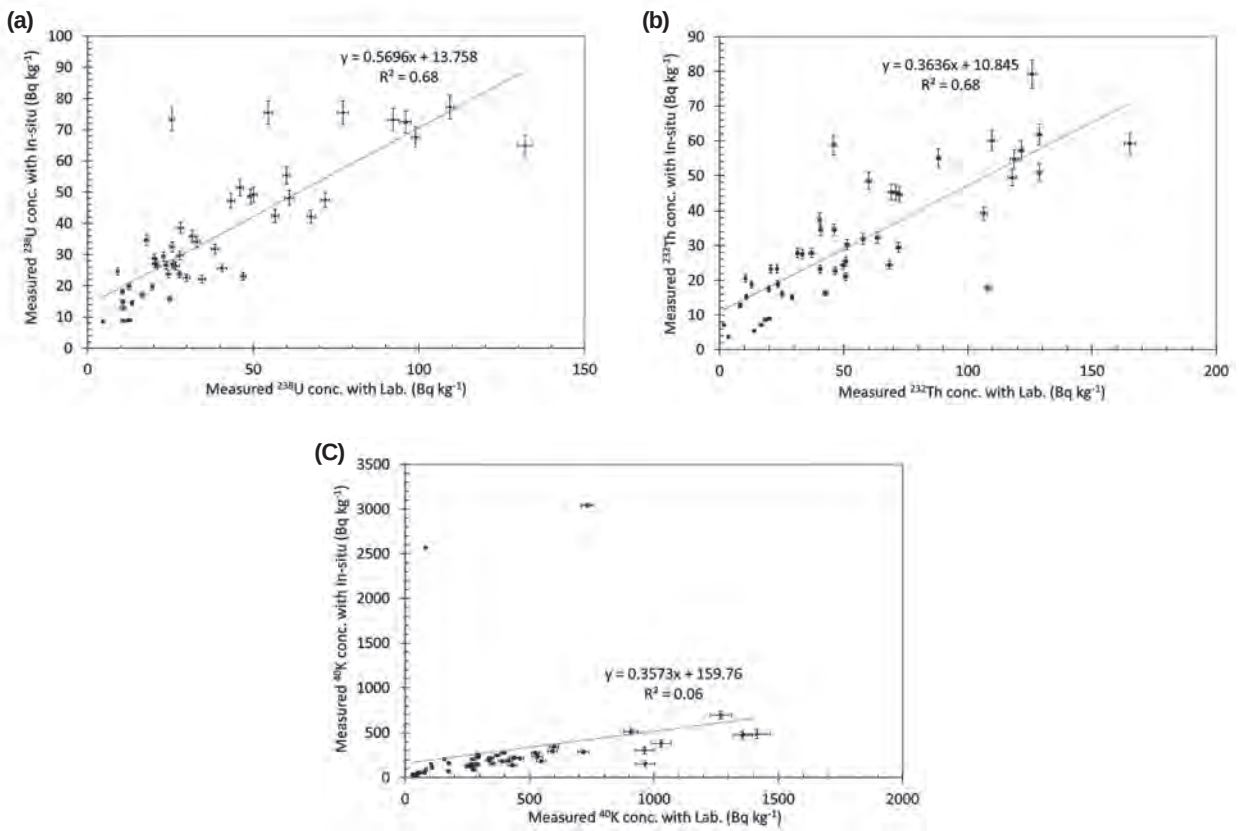

Fig. 2. The relation of measured activity concentration of (a) ^{238}U, (b) ^{232}Th and (c) ^{40}K using *in-situ* and laboratory gamma spectroscopy techniques

3. Result and discussion

3.1 Evaluation of specific activity concentration of natural radionuclides

Activity concentrations of natural radionuclides of ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K in Eastern, Western, and Southern Thailand were measured using the *in-situ* measurement with car borne survey system and the laboratory soil-sampling-based gamma spectrometry technique as shown in Table 1 and Table 2, respectively. A comparison between the measured values of ^{238}U, ^{232}Th and ^{40}K with the two techniques of in-situ and laboratory gamma ray spectrometers are presented in Figure 2. The mean activity concentration of ^{238}U, ^{232}Th and ^{40}K were 36 ± 1 Bq/kg, 31 ± 2 Bq/kg and 305 ± 11 Bq/kg respectively for the *in-situ* measurement. The mean activity concentration of ^{226}Ra (^{238}U), ^{228}Ra (^{232}Th) and ^{40}K were 37 ± 2 Bq/kg, 56 ± 1 Bq/kg and 413 ± 11 Bq/kg respectively for the laboratory soil-sampling-based measurement. The median values of ^{238}U-series, ^{232}Th-series and ^{40}K in the earth' s crust are 35, 30 and 400 Bq/kg respectively[17]. Thus, all the average values of the radionuclides from the 3 regions of Thailand measured in the soil samples from this study are slightly higher than the worldwide median values. The high value of the activity concentration may correlate with the presence of the geological lineaments in the area (Granite belts)[18] and also this variation of ^{40}K may be due to the excess use of agricultural fertilizers.

3.2 Comparison between in-situ and laboratory activity concentration of ^{40}K, ^{238}U and ^{232}Th

Comparative analyses were done between *in-situ* and laboratory measurements. The ratio between the two measurements were 0.97, 0.55 and 0.71 for ^{238}U, ^{232}Th and

^{40}K respectively as shown in Table 3. The laboratory results obtained in this work were greater than the *in-situ* results for ^{232}Th and ^{40}K. The average % difference of -3%, -81% and -41% was obtained for ^{238}U, ^{232}Th and ^{40}K respectively. Moreover, a moderate correlation between these techniques can be observed in Figure 2 with linear regression lines of 68% for ^{228}U and ^{232}Th whereas a very-weak correlation for ^{40}K, the correlation was 6%. Large deviation recorded between the *in-situ* and laboratory measurements in this work maybe due to the fact that the samples collected for the laboratory analysis only accounted for a very small volume of surface soil (0-5 cm.) as against the *in-situ* measurements with gamma contributions from the top 30 cm layer soil[19] and surface area up to hundreds of m^3 [20]. Moreover, the water content, and soil density affect the attenuation of gamma within the soil. In regards to the water content of the soil, attenuation coefficients show a difference of less than 3% for soil with water contents of 0% and 25%[21]. Therefore, high moisture in soil would underestimate the specific activity of radionuclides in the soil by *in-situ* measurement. On the other hand, Corbacho and Baeza[22] reported that there was an evidence of a slight increase of natural radionuclides in the top soil layer (0-5 cm) at the areas nearby the ash pond due to the deposition of fly-ash whereas a depth soil layer was a homogeneous distribution of natural radionuclides. Likewise, the use of fertilizers on agricultural lands may lead to an increase of natural radioactivity in farm soil[23-25].

3.3 Comparison between in situ and laboratory outdoor absorbed dose rate in air

The average absorbed dose rates in air estimated for the soil samples varied from 65 to 75 nGy/h with a mean of 69 ± 3 nGy/h. The average absorbed dose rates in air from direct measurement using *in-situ* technique varied from 42 to 57 nGy/h with a mean of 45 ± 8 nGy/h. The average percent difference was 53%. Additionally, the uncertainty in the absorbed dose rate in air using the laboratory gamma spectrometry was about 4% while as the uncertainty using the *in-situ* gamma ray was about 18%. The *in-situ* measurement uncertainty was 4.5 times higher than laboratory gamma spectrometry. This may cause by the non-uniform distribution of radioactivity in soil, the largest source of *in-situ* uncertainty was likely to be because a uniform reference calibration does not accurately represent the true efficiency. Moreover, the ratio of the average absorbed dose rate in air inferred from activity concentrations of radionuclides in soil to the average absorbed dose rate in air from *in-situ* measurement using car borne survey system in this study is 1.5. A factor of 1.5 is likely due to shielding of the soil moisture and the climate condition during the measurement time. The difference between the two methods could be further explained that the *in-situ* gamma spectrometry gives representative source concentration in the horizontal plane. In practice, an infinite half space can be taken as a large area of open ground with a radius up to 70 m where there are few obstructions[19] whereas the gamma spectrometry in laboratory measures radioactivity in soil samples collected from the top 5 cm of a 1 m^2 area providing a dry mass of approximately 200 g. It is important to acknowledge this measurement error when employing the two methods.

4. Conclusion

A comparative study of outdoor gamma dose rate evaluation by *in-situ* and soil-sampling-based radioactivity measurement methods was performed in the eastern, western, and southern Thailand. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K from soil were determined with *in-situ* Na(Tl) and conventional laboratory HPGe detectors. Largest difference between the *in-situ* and laboratory measurements were observed for the measurement of activity concentration of ^{232}Th at the ground level of soil. The absorbed dose rate in air by the laboratory measurement are 1.5 times of the *in-situ* measurement. On the other hand, the mean value of the *in-situ* measurement (45 ± 8 nGy/h) is in agreement with the value reported in a previous publication (41 ± 8 nGy/h)[11] using car-borne survey.

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Conflict of interest disclosure

The authors declare that they have no conflict of interest.

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Supplementary information

The latitude, longitude and altitude of the In-situ measurement and soil sampling points

Location	Site code	Latitude (°N)	Longitude (°E)	Altitude(m a.s.l.)
The Eastern	E01	11.811458	102.853475	16
	E02	11.824867	102.842693	6
	E03	11.838502	102.832968	7
	E04	11.886980	102.801908	7
	E05	11.895453	102.786613	4
	E06	12.315335	102.625288	38
	E07	12.468680	102.618123	35
	E08	12.132743	102.547307	3
	E09	13.046533	100.927088	6
	E10	12.730072	100.984170	36
	E11	13.027143	101.072865	120
	E12	13.302883	101.114518	74
	E13	13.368263	101.195657	29
	E14	13.514833	101.267810	56
	E15	13.127977	101.317985	100
	E16	13.402930	101.407420	66
	E17	12.790897	101.155347	66
	E18	13.009872	101.247840	97
	E19	12.784258	101.250015	35
	E20	12.921992	101.316473	35
	E21	12.632005	101.456797	22
	E22	12.684377	101.833462	8
	E23	12.836345	101.727978	25
	E24	12.653128	101.453040	16
	E25	12.766120	101.563613	51
	E26	12.640042	102.170993	17
	E27	12.920037	102.434685	153
	E28	13.135523	102.355592	142
	E29	13.027268	102.361165	182
	E30	12.491285	102.049473	4
	E31	12.760395	102.083108	40
	E32	12.760788	101.965262	24
The Western	W01	14.618232	98.7280730	86
	W02	14.095860	99.2533470	94
	W03	13.817620	99.6264430	51
	W04	14.325465	99.5981350	58
	W05	13.533613	99.8699320	7
	W06	13.436737	99.4085300	143
	W07	12.940515	99.7479520	62
	W08	13.094630	100.062573	6
	W09	16.886085	99.1197080	122
The Southern	S01	9.2526250	99.1459080	18
	S02	8.5089050	99.9583320	5
	S03	8.3614230	98.7840370	85
	S04	7.5607120	99.7125020	30
	S05	7.6649120	100.023640	10
	S06	8.4435930	98.5204530	18
	S07	9.7560450	98.5921170	33
	S08	10.674867	99.2023000	46