Evaluation of 2019 Ar-41 Measurements and Analysis

Several non-conformities (NCs) between the 2019 Ar-41 measurement and analysis and those from previous years have been identified. These are described below with corresponding Corrective Actions (CAs). Based on new analysis of 2019, 2014, 2009 and 2008 Ar-41 data and review of the Ar-41 release reports dating back to 2005, several Opportunities for Improvement (OFIs) have also been identified that could improve the consistency and reliability of the measurement and analysis procedures going forward.

NC1: Non-conformity in sample collection procedures

Summary: Vent fans #1, #2, and #3 were turned on during the 2019 measurement. However, an air sample was only collected from vent fan #3 and only the fan flow rate for vent fan #3 was used in the calculation of the outside Ar-41 concentration. Therefore, the 2019 measurements would have resulted in non-conservative Ar-41 concentration values. It is not possible to exactly quantify the difference as new analyses (presented below) of the 2008, 2009, 2014 and 2019 measurements reveal that the measured Ar-41 concentrations were consistently below detection limits.

- CA 1.1. SOP 654 shall be revised to specify the status of each fan (on or off) used in the procedure.
- OFI 1.1. When airflow paths from the pool surface above the core to the vent fans are equal, the highest capacity vent fan will result in the shortest Ar-41 residence time in the bay and therefore the highest outside concentration. Thus, use of fans #1 or #2 during Ar-41 measurement is expected to be most conservative for the purposes of estimating gaseous effluent release. Meanwhile, measurements with only fan #3 operating are expected to result in the most conservative values for the purposes of estimating occupational doses. That said, it is not clear if airflow paths to fans #1, #2, and #3 are exactly comparable as their position relative to the core, bay floor grating, and various obstructions in the bay differ. In revising the measurement procedures, it would be prudent to individually measure the concentrations at all three vent fans using a consistent methodology and then determine which combination of measurement and dose limit (occupational or dose to general public) imposes the most restrictive administrative limit on energy released. A suitable criterion for accepting the results could be that the measured Ar-41 concentrations exceed the quantification limit.
- OFI 1.2. In the 2019 and 2014 data, four additional photopeaks at 511, 610, 1460, and 2613 keV were observed in the background and sample spectra. These peaks are attributed to the naturally occurring radionuclides K-40, Tl-208, and Bi-214 which are commonly encountered in gamma ray spectra in NaI detectors [1,2]. While K-40 can be partially attributed to potassium impurities in the NaI crystal, the Tl-208 and Bi-214 peaks are members of the thorium and uranium decay series, respectively, and are likely present in the masonry and building materials in the counting lab. Although a detector shield was used, the door of the shield was partially open in order to accommodate the large Marinelli flask. Additional lead sheet was used to further reduce background contamination in the spectra. Based on the aforementioned photopeaks, however, it is likely that the amount of shielding was insufficient. Although background contamination was mostly removed through background subtraction, lower detection limits could have been achieved with additional shielding. In future measurements, the detector shield should be further extended using lead brick.
- OFI 1.3. SOP 654 specifies a 1800 second count time. Detection limits can be further improved by increasing the count time.

- OFI 1.4. The Na-22 Marinelli source had decayed to ~35 Bq by 2019. This resulted in a weak signal and a corresponding 10% uncertainty in the detector efficiency. A new Na-22 source should be procured and used in future tests. Sources older than 7 half-lives should not be used in the Ar-41 measurement.

NC2: Non-conformity in spectral analysis

Summary: Upon inspection of the raw spectra files, it became apparent that two different energy calibrations were used in the analysis. One calibration was applied to both the Na-22 standard and background spectrum while the other was applied to the four air samples. The calibration of the Na-22 spectrum was deemed to be correct as two photopeaks, centered at 1274 keV and 511 keV, coincide with the Na-22 characteristic gamma and annihilation photon, respectively. The energy calibration applied to the four air sample spectra were also determined to be correct through comparison of spectra collected during the 2014 Ar-41 analysis. When the same calibration used with the sample spectra was applied to the background spectrum, the locations of the four aforementioned background peaks and continuum shape overlapped (see Fig. 1). However, as mentioned above, the energy calibration used for the Na-22 spectrum was applied to the background spectrum. The situation is represented in Fig. 2.

A shaded rectangle indicates the region of interest (ROI) used to quantify the Ar-41 activity. No photopeak can be visually identified in any of the spectra. Therefore, the ROI gross counts in each of the sample spectra primarily reflect the continuum background. If a photopeak was present, the chosen ROI would be slightly narrower than the FWHM of the photopeak, which would result in underestimation of photopeak area. Use of 50 channels is called for in the counting procedure. However, in the years when that procedure was developed, the detector and multi-channel analyzer (MCA) was configured to have an approximately 10 keV/ch bin width. In 2014 and 2019, 2 keV/ch were used.

In the quantification procedure, the gross background counts were subtracted from the gross sample counts. Due to the use of the incorrect energy calibration in the background spectrum, the gross background counts, having been measured at a higher energy (around 4000 keV), were significantly lower than the continuum background of the sample spectra in the Ar-41 region. Thus, the quantity reported as the net Ar-41 counts was essentially a difference in gross counts within two different energy ranges of the same background, a physically meaningless quantity. This explains why the reported Ar-41 concentrations at each sampling location were all similar in value (3.11×10^{-6} - 3.27×10^{-6} μCi/mL) and an order of magnitude larger than expected based on the SAR and results from previous years.

As the Na-22 spectrum had a different energy calibration, it is possible that the high voltage (HV) or gain settings were changed for that acquisition. Indeed, the continuum background in the Na-22 spectrum was significantly lower than the background spectrum (Fig. 3). It is therefore doubtful that a simple method of gross background count subtraction would result in an accurate efficiency determination.

- CA 2.1. SOP 654 shall be revised to establish an ROI selection procedure based on detector type, HV, gain and MCA settings.
- CA 2.2. SOP 654 shall be revised to establish detailed procedures that ensure a common and correct energy calibration is applied to the spectra. It will also provide additional guidelines for selecting HV and gain settings.

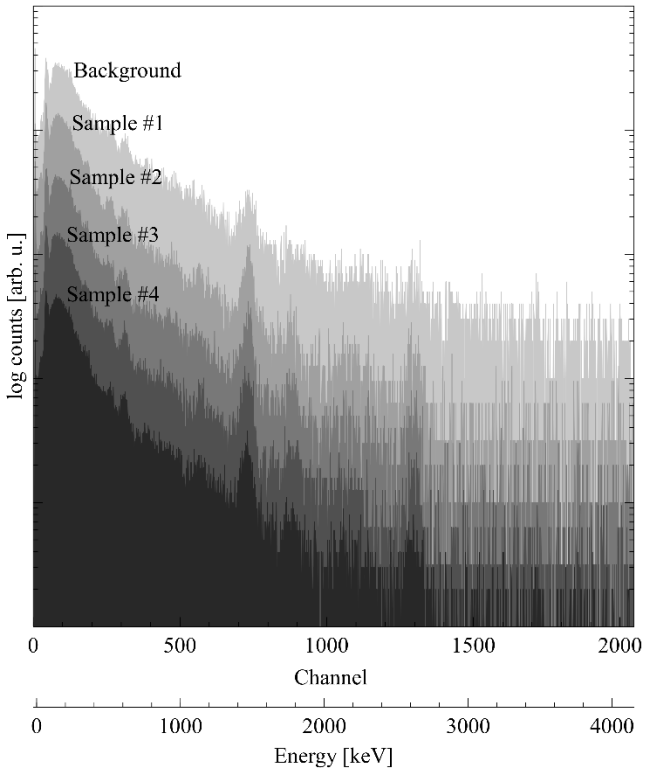

Fig. 1. Background and sample spectra from 2019 measurement with correct calibration. Spectra are shifted vertically.

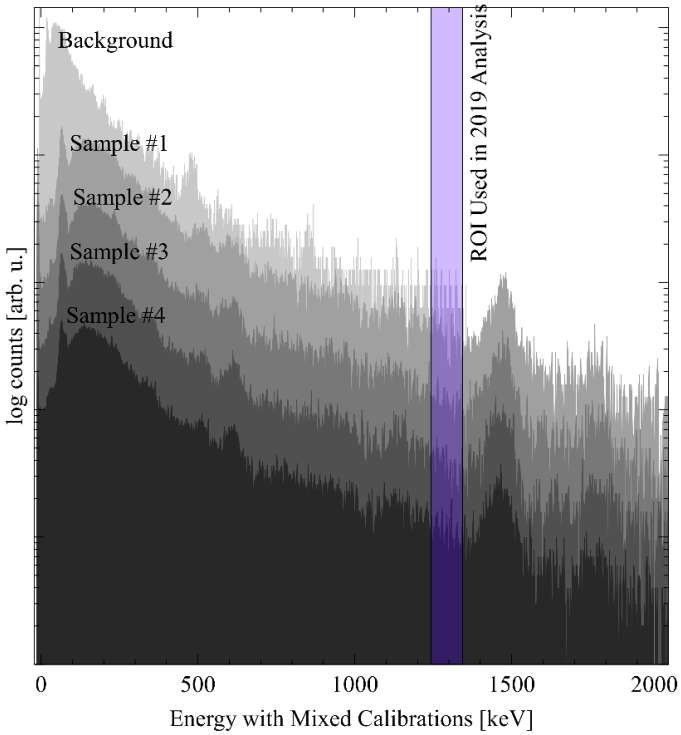

Fig. 2. Background and sample spectra as used in 2019 analysis. Spectra are shifted vertically.

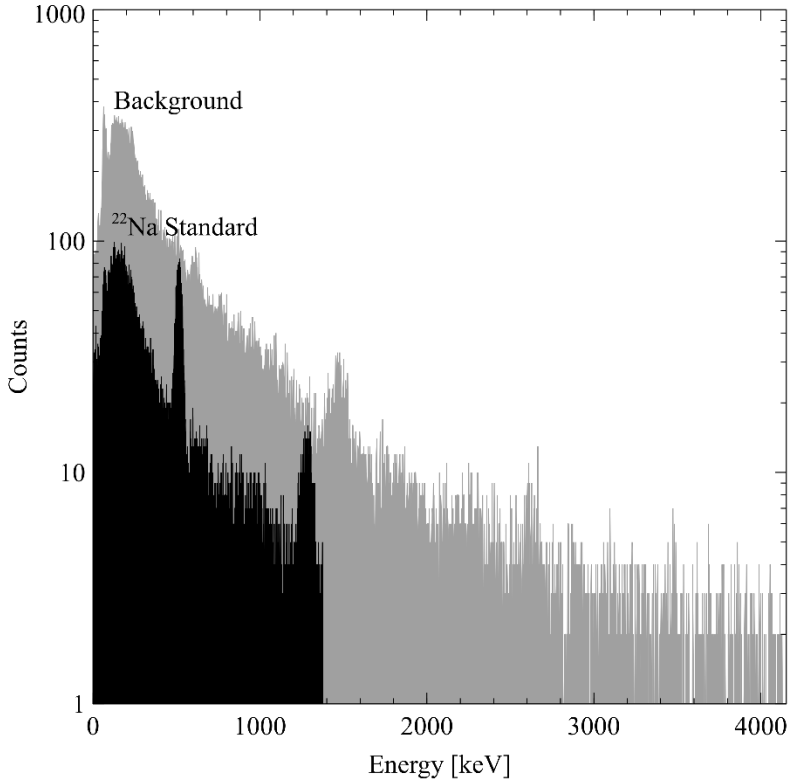

Fig. 3. Comparison of spectra from Na-22 standard and background. Note that the counting times were the same (1800 s).

NC3: Non-conformity in calculation of downwind Ar-41 concentration

Summary: An incorrect vent flow rate correction factor was used in calculating the downwind Ar-41 average annual concentration. The flow rate of vent fan #3 (5000 cfm) was assumed instead of the combined flow rates of vent fans #1, #2, and #3. Thus, the average concentration calculated was inaccurate in the less conservative direction. It was further noted that in previous years, the flow rate of vent fan #3 was used even though measurements were taken at either fan #1 or #2. In calculating the Ar-41 concentration in the Marinelli flask, an incorrect flask volume was also used. In data going back to at least 2005, a 400 mL flask volume was assumed. The actual flask volume is 4000 mL. This resulted in a 10 times higher concentration in all years.

- CA 3.1. Revisions in the analysis procedures shall be made such that the correction factor(s) used in the determination of downwind concentration accurately reflect the fans used in a year's operations.

Reanalysis of 2019, 2018, 2009, and 2008 Measurements

The 2019, 2018, 2009 and 2008 spectral data were reanalyzed with the following modifications:

1. The correct calibration was used for the background spectrum. This only affected the 2019 results as the correct calibration was used in previous years.

2. The linear continuum estimation method was used to determine net counts from photopeak ROI.

3. Detection limits and quantification limits were determined from the background spectrum using the Currie method with 95% confidence intervals for Type I and Type II errors [3].

4. Selection of ROIs was based on the FWHM of the data and interference of the K-40 peak at 1460 keV.

5. Use of the correct Marinelli flask volume (4000 mL instead of 400 mL).

6. Use of correct fan flow rates.

A comparison of the results from the original analysis and the new analysis is shown in Table I. There is greater consistency in the results under the new analysis though it should be noted that in many cases the Ar-41 concentration was found to be below detection limits. After applying corrections for Marinelli flask volume and fan flow rates, the concentrations (or detection limits) before 2008 were reasonably consistent with the 2014 and 2019 values though detailed spectral reanalysis was not performed for the years 2005, 2006, and 2007. Annual Ar-41 doses were recalculated and are shown in table II. In all cases, the doses are well below the 10 mrem limit of 10 CFR 20.1102d.

Table I

	Original Analysis				New Analysis			
Location	2019	2014	2009	2008	2019	2014	2009	2008
6” above pool near core	3.2×10^{-6}	3.9×10^{-6}	4.7×10^{-6}	5.0×10^{-7}	$*6.3\times10^{-8}$	3.8×10^{-7}	5.6×10^{-7}	$*1.0\times10^{-7}$
6” above fuel storage pit	3.2×10^{-6}	2.2×10^{-6}	$*2.3\times10^{-7}$	4.9×10^{-7}	$*6.3\times10^{-8}$	1.1×10^{-7}	$*1.2\times10^{-7}$	$*1.0\times10^{-7}$
Bay area**	3.3×10^{-6}	2.0×10^{-6}	$*2.9\times10^{-7}$	$*1.6\times10^{-7}$	$*6.3\times10^{-8}$	1.3×10^{-7}	$*1.3\times10^{-7}$	$*1.0\times10^{-7}$
Roof by Fan	3.1×10^{-6}	1.4×10^{-6}	1.7×10^{-6}	$*2.7\times10^{-7}$	$*6.3\times10^{-8}$	$*5.5\times10^{-8}$	$*1.2\times10^{-7}$	$*1.1\times10^{-7}$
Detector efficiency	0.64%	0.64%	1.53%	1.53%	1.32%	1.32%	0.79%	0.98%

*Denotes detection limit which is used as an upper bound in cases where counts were below the detection limit.

**Sampling location in reactor bay varies from year to year

Table II

Reporting Year	Energy Generated [MWhr]	Dose [mrem]
2005	40.8	1.27
2006	18.7	0.50
2007	14.1	0.38
2008	2.7	0.11
2009	17.9	0.90
2010	11.7	0.26
2011	25.5	0.56
2012	23.0	0.50
2013	35.5	0.78
2014	12.7	0.65
2015	9.7	0.57
2016	8.8	0.52
2017	16.9	0.99
2018	17.5	1.02
2019	6.5	0.38
2020	6.7	0.39

Summary and Additional Corrective Actions

While reevaluation of Ar-41 release data from 2019 and previous years has shown that effluent release from MSTR has been well below the 10 mrem ALARA threshold specified in 10 CFR 20.1102d, an unacceptably large number of errors were uncovered in sample collection, spectral analysis, and concentration calculations. The Corrective Actions mentioned above specify changes to methods and procedures intended to prevent those errors from reoccurring. However, a key underlying cause of those errors is a lack of quality assurance and knowledge atrophy. Historically, a single health physicist performed most of the analysis tasks using methods and procedures handed down from previous health physics personnel, without questioning those methods or verifying their origin or bases. This led to the duplication of the same error over multiple years. Additional Corrective Actions will be implemented at an administrative level to address these underlying issues.

1. Future Ar-41 data analysis and effluent calculations shall be checked by a member of the faculty, unaffiliated with the reactor or campus health physics and who has practical knowledge of radiation measurement and spectroscopy. That faculty member will be encouraged to suggest areas for improvement in the methodology.

2. Ar-41 measurements will be taken every three years instead of every five years to reduce knowledge atrophy.

References

1. G. R. Gilmore, “Appendix D: Gamma-Ray Energies in the Detector Background and Environment” in “Practical Gamma-Ray Spectrometry,” John Wiley and Sons, Warrington, UK, 2008.

2. P. Adhikari et al., “Understanding internal backgrounds in NaI(Tl) crystals toward a 200 kg array for the KIMS-NaI experiment,” *The European Physical Journal C*, **76**, 185 (2016).

3. L. A. Currie, “Limits for qualitative detection and quantitative determination. Application to radiochemistry,” *Analytical Chemistry*, **40**, 586 (1968).