

Methods for Radiological Assessment of C-14 in Air Effluents – 15419

James Westmoreland *, Robert Wills *

* GEL Laboratories, LLC

ABSTRACT

The purpose of this paper is to report on methods used for the assessment of “dose significant” C-14 being discharged into the air from nuclear facilities. GEL laboratories, LLC and GEL Engineering LLC, (GEL) undertook the task of developing new and improved methods to better identify the release of C-14 in the various chemical forms released from nuclear reactors and fuel reprocessing facilities.

To completely understand the environmental releases and impact to the public a two step approach was selected. The first step was to develop a sampling method that would enable an accurate assessment of the ratio of C-14 discharged as carbon dioxide compared to organic gases like methane. This is very important to dose modeling calculations to members of the public. Once this ratio was understood, the second step was to develop a method to track inorganic forms of C-14 with standard environmental air monitoring equipment and achieve a detection limit that was in line with NRC 10CFR Appendix I dose modeling requirements.

GEL development efforts focused on using a solid sample cartridge that would fit into standard air sampling equipment used at nuclear plants. The cartridge and sorbent needed to be capable of running 7 days at 24hrs per day and not saturate with stable carbon dioxide. The process developed for analyzing the sorbent involves chemically removing the carbon dioxide and ultimately counting by liquid scintillation counting. The method yields a detection limit for C-14 of 0.02 Bq/m³ which provides a dose calculation of approximately 0.1 mSv/yr when applied to the child model. This paper will describe the methods and results used to determine field environmental monitoring for C-14 using GEL cartridges also known as “GELsorb C14”.

INTRODUCTION

C-14 has become a principle radionuclide in the air effluent releases of active nuclear power plants. This shift is largely the result of continuous improvement and reduction of other radionuclides emitted over time. In the early stages of nuclear power production C-14 was correctly considered as a minor dose contributor compared to other fission and activation products discharged to the environment. The calculated dose contribution of C-14 was less than 0.1% of the total exposure pathway in the earlier plant designs. With improvements in ALARA programs and reactor fuel integrity C-14 has shifted to the largest dose contributor to the general public via air borne environmental pathways accounting for almost 90% of the total dose to members of the public. The NRC requires in Title 10 CFR 50 Appendix I that all power reactors perform assessment for the following.

1. Provide Data on Measurable Levels of Radioactive Material Released in Liquid and Gaseous Effluents
2. Provide Data on Measurable Levels of Radioactive Material Released in the Environment

3. Identify Changes in Principal Exposure Pathways

Prior to 2010 C-14 was not a reportable isotope in reactor effluents or radiological environmental monitoring programs. After this date the NRC required utilities to report C-14 releases as part of the effluents regulatory guide 1.21 “MEASURING, EVALUATING, AND REPORTING RADIOACTIVE MATERIAL IN LIQUID AND GASEOUS EFFLUENTS AND SOLID WASTE”.

Since the utilities were not equipped with an analytical method to determine the amount of C-14 the plant was discharging they opted for a calculation approach based on effective full power reactor hours. This method was used to provide the frame work to calculate the discharge of C-14. This approach is good as an estimate but does not fully account for some field operating conditions that can arise.

1. The calculation does not account for non stack discharges due to reactor depressurization during refueling and maintenance outages.
2. The calculation does not take in account different plant equipment for off gas treatment such as “gas recombiners” which convert organic to inorganic carbon.
3. Calculated doses tend to yield higher results (overestimated) when compared to measured results

DISCUSSION

Determination of the proper solid sorbent, amount of material and optimal mesh sizes were tested in the laboratory. Solid media was preferred for ease of shipping and handling than a bubbler design. The challenge in developing the analytical method was in stripping the carbon without losses between sorbent and final liquid scintillation counting. Additionally handling approximately seven grams of stable carbon dioxide accumulated over a week of sampling was a challenge that the method needed to address. The C-14 method employs the wet oxidation of carbon compounds to volatile carbon dioxide CO₂ which is sparged through a dilute acid solution for trapping tritiated water present in the sample. After sparging through dilute acid, the CO₂ is trapped in a sorbing solution which is added to liquid scintillation cocktail and finally counted in a liquid scintillation counter. Figure 1 shows the method design configured for optimal separation of carbon from the solid sorbent.

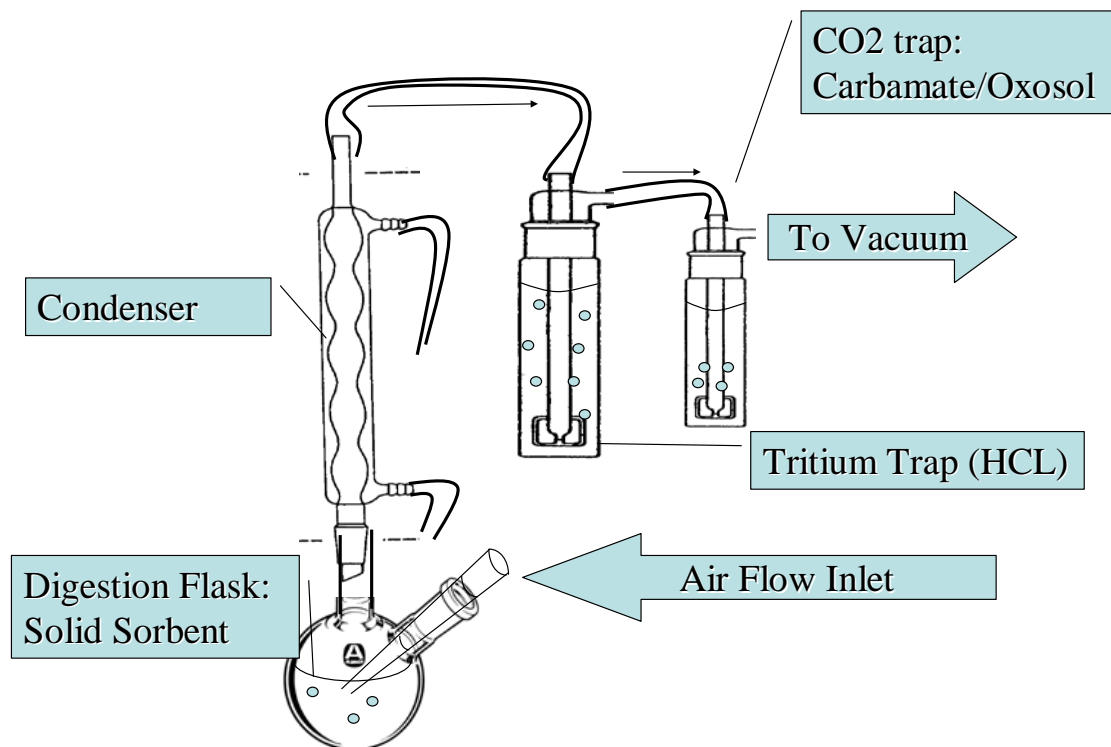


Fig. 1. C-14 Laboratory Apparatus

Validation of the solid sorbent was done per lot number. First a representative cartridge was prepared with material and subjected to a testing apparatus capable of monitoring breakthrough of carbon dioxide (CO_2) by mass on a secondary sorbent tube. The observed breakthrough on the secondary material was compared to the theoretical total amount of carbon dioxide relative to natural levels in air. With a flow Rate of 1.0 liters per minute and total flow 10080 liters of air the method showed greater than 99.9% retention for the one week interval.

A second capacity validation test was done with three cartridges subjected to pure CO_2 . A gas cylinder was connected in line with each cartridge and the CO_2 was passed through the cartridge at 1 liter per minute for 4 minutes. The theoretical mass of CO_2 retained on the cartridge was 7.2 grams which is greater than the mass expected to be captured under normal use. The cartridge effluent was bubbled through a sodium hydroxide trap with calcium chloride to precipitate any CO_2 not captured as calcium carbonate. The precipitate was collected on a filter for mass determination.

Table I represents cartridges tested for carbon dioxide capacity at a flow rate of 1.0 liters per minute and a total flow of 4 liters.

TABLE I. Capacity Testing for C-14 Cartridges

Sample ID	Pump Flow (Lpm)	Total Liters of CO₂ Sampled (L)	Theoretical CO₂ (g)	Observed CO₂ (g) Breakthrough	Percent Efficiency (%)	PASS/FAIL
Sample 1	1.0	4	7.2	2.27e-4	99.99	PASS
Sample 2	1.0	4	7.2	None measured	100	PASS
Sample 3	1.0	4	7.2	None measured	100	PASS

Once the method was shown to be capable of good capture of carbon dioxide in the field, GEL deployed cartridges at three nuclear power plants from 2012 to 2014. Results of this study are shown in Figure 2. No weekly measurement was greater than the minimum detectable concentration (MDC) in this study. The anomaly on the 20th of April was caused by a pump that did not run for the entire week causing an increase in the MDC observed. This weekly confirmation of data that was less than the MDC allows the plant to monitor for compliance and adjust should any anomaly be observed. The MDC values fluctuate based on air flow in the field which at the start of the study were set at lower flow rates. Resulting MDC values ranged from 0.02 Bq/m³ (0.5pCi/m³) to just over 0.04 Bq/m³ (1.0 pCi/m³).

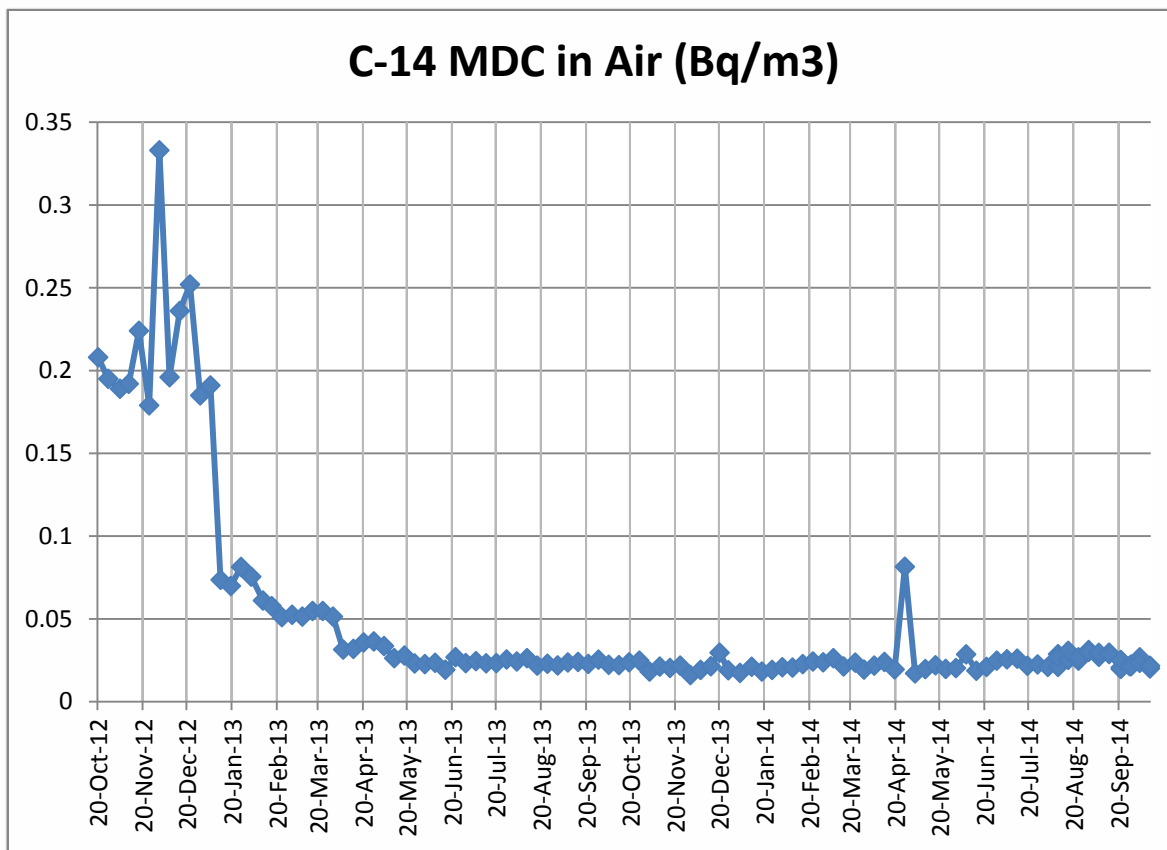


Fig. 2. Summary of C-14 MDC results in air

CONCLUSIONS

Actual testing for effluents is recommended for nuclear utilities to avoid over reporting of releases and to provide weekly monitoring for C-14 at perimeter locations. The cartridge approach proved effective and met design expectations in high heat and high humidity environments. Laboratory validations show the method capable of 99.9% retention of C-14 in air at collection rates of approximately 1.0 liter per minute. The cumulative collection capacity over a 1 week sampling interval was demonstrated accurate over a wide range of concentrations. The methodology has been shown free from interferences with detection limits of approximately 0.02 Bq/cubic meter.