TRANSPORTABLE AUTOMATED IODINE-131 AND XENON-133 SAMPLING SYSTEM

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Abstract

The fission products Iodine-131 and Xenon-133 from medical radioisotope production waste at Atomic Energy Canada Ltd's (AECL) Chalk River Laboratories (CRL) are monitored under AECL's Environmental Protection Plan. At site, most radioactive waste monitoring is done automatically, however a portable system is being developed to gather information about possible localized emissions and support continuous improvements in the emission monitoring program. Considering the operator exposure time to radiation and measurement limitations caused by manual operations, the sampling system has been fully automated. This paper outlines the design and development of the Transportable Automated Iodine and Xenon Sampling System. This system features a fully automated instrument with configuration flexibility, sampling repeatability, reduced operator exposure time to radiation and ease of transportability.

1. Introduction

The medical radioisotope Molybdenum-99 (^{99}Mo) is produced as a fission product (provides 40% of the world production of medical isotopes) in the National Research Universal (NRU) reactor at AECL's Chalk River Laboratories. The process of extracting ^{99}Mo from the irradiated target results in radioactive liquid waste. The liquid waste goes through a cementation process to be solidified, and this solidified waste is transported to and stored in below grade tile holes in the Waste Management Area (WMA) at CRL. Extremely small amounts of fission products ^{131}I and ^{133}Xe are released from the solidified waste. These two fission products are of special interest because, the ^{131}I isotopes are volatile and ^{133}Xe isotopes are gaseous found in airborne emissions and have longer half-lives compared to other emitted fission products. Currently emission of ^{131}I from these tile holes is sampled and measured under AECL's Environmental Protection Plan. The emission rates do not constitute a significant dose to personnel at site, and regular bioassay monitoring confirms that the current waste emplacement practice results in emission rates that are acceptable for the WMA workers and site staff.

Presently, I-131 samples are collected manually by operators which may lead to undesirable exposures. AECL is committed to improving worker safety and reducing emissions from the radioactive waste when it is feasible to do so. The methodology for sampling I-131 manually was developed and proven previously [1]. The previous system was operated manually and was only capable of gathering one iodine sample at a time. Also the system was only applied to filled (plugged) tile holes in the WMA. Unlike the previous manual system, the new automated sampling system is capable of sampling both I-131 and Xe-133 from higher radiation field environment such as transporting flasks enabling estimates of emanation rates during waste transportation and emplacement. Higher radiation exposure to the field operators is one of the main challenges. Application of this new automated system will reduce operator exposure time, gather 7 sets of iodine and xenon samples within short period of time, obtain accurate readings from the sensors in the tent, and run as many cycles as the operator pre-configures the software to collect acceptable amount of samples. Other challenges faced in the development of this system were: weather conditions and outdoor temperature variations especially cold temperatures; no electrical power line availability in the WMA; and measurement limitations from a manual operation. To overcome these challenges, the automated system has been designed to handle temperatures as low as -40°C, self powered with rechargeable batteries and programmed with power budgeting options.

The system consists of a rigid tent-like structure with a 1.3 m x1.3 m foot print. The central portion of the tent constitutes a trapped volume of air for accumulation of the isotopes as they are emitted from the tile hole or shipping flask. Carbon monoxide (CO) gas has a density close to air density and is used as a tracer to monitor the air leak rate from the tent. CO is injected in the tent for a few seconds and the concentration is tracked by a sensor, to trace the rate of air exchange between the inside and outside of the tent. Also the system monitors tent temperature, sampling air flow rate, battery power and tent humidity as this structure is utilized in the field under less-than-ideal condition. The top portion of the tent contains a slot to hold the sampling and control apparatus. This paper describes the development of the automated iodine and xenon sampling system and measurements techniques utilizing it. This automated sampling system will contribute to continuous improvements in AECL's emission monitoring program.

2. Radioactive waste storage configuration

The production process of ^{99}Molybdenum starts in the National Research Universal (NRU) reactor by irradiating specially designed targets containing highly enriched uranium. After irradiation, the targets are transported to the Molybdenum-99 Production Facility (MPF) where the extraction of Molybdenum-99 takes place. The liquid waste resulted from this process is solidified into small waste cans through a cementation process. The waste mixture is allowed to cure for 24 hours and any radioactive emissions during the curing process are monitored. After the curing process, the waste cans are transported in shipping flasks to engineered tile holes in WMA (Figure 1). Each tile hole in WMA can hold up to 9 waste cans. When the tile hole is full, the opening of the tile hole is capped with a concrete plug to prevent airborne emissions. Although the tile holes are plugged, trace quantities of gaseous emissions are still found in the air around the tile holes.

3. I-131 and Xe-133 concentration build up

As shown in Figure 2, the sampling system incorporates a tent-like enclosure for capturing tile hole emissions. Similarly, the tent can be placed on a shipping flask that contains waste cans, transported to WMA. The tent enclosure is placed on top of the tile hole or flask to accumulate emissions and allowing iodine and xenon concentrations inside it to rise after tent placement. After tent placement, the operator enables sampling system operation; the system can take up to 7 sets of iodine and xenon samples at preconfigured sampling times.

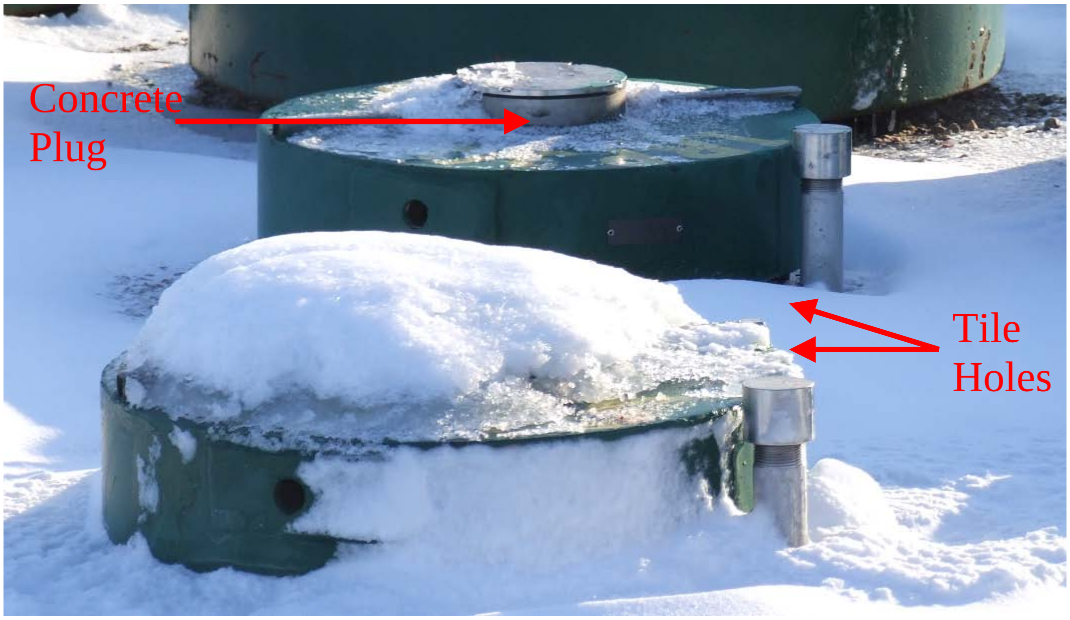

Figure 1 Tile holes in Waste Management Area (WMA)

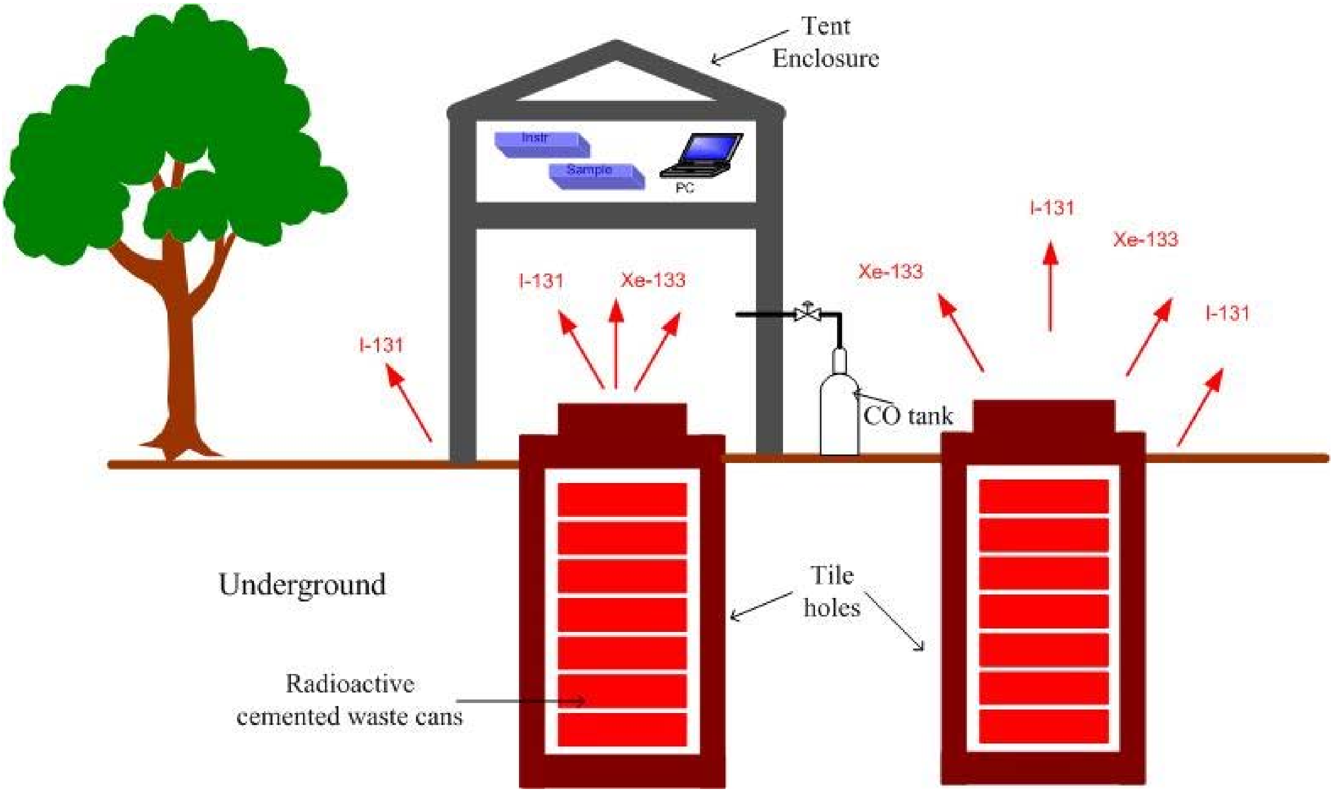

Figure 2 Schematic representation of I-131 and X-133 from tile holes, and the tent enclosure setup on a tile hole

For many reasons, the tent seal is not perfect; it is therefore necessary to determine the air leak rate over time during the sampling process. This leak rate may also vary due to weather conditions and further complicates the estimation of the loss rates for iodine and xenon from the tent. The system therefore incorporates the ability to inject a trace gas, carbon monoxide (CO) at the start of the sampling process; the concentration of CO is continuously recorded during the sampling cycle, providing a measure of the loss rates to the environment from the tent as well as the concentration of iodine and xenon in the tent. The change is concentration is defined as

$$\frac{dN}{dt} = E - \lambda_d N - \lambda_l N \qquad (1)$$

Where, *N* is the number of iodine or xenon atoms present in the tent at any moment, *E* is the emission rate of I-131 or Xe-133 from the tile hole (*number of atoms/s*), λ_d the decay constant given by $ln2/t_{0.5}(s^{-1})$ and $t_{0.5}$ is the half-life of I-131 (8.04 days) or X-133 (5.25days). λ_l is the air leak rate expressed as tent volume per unit time (s^{-1}).

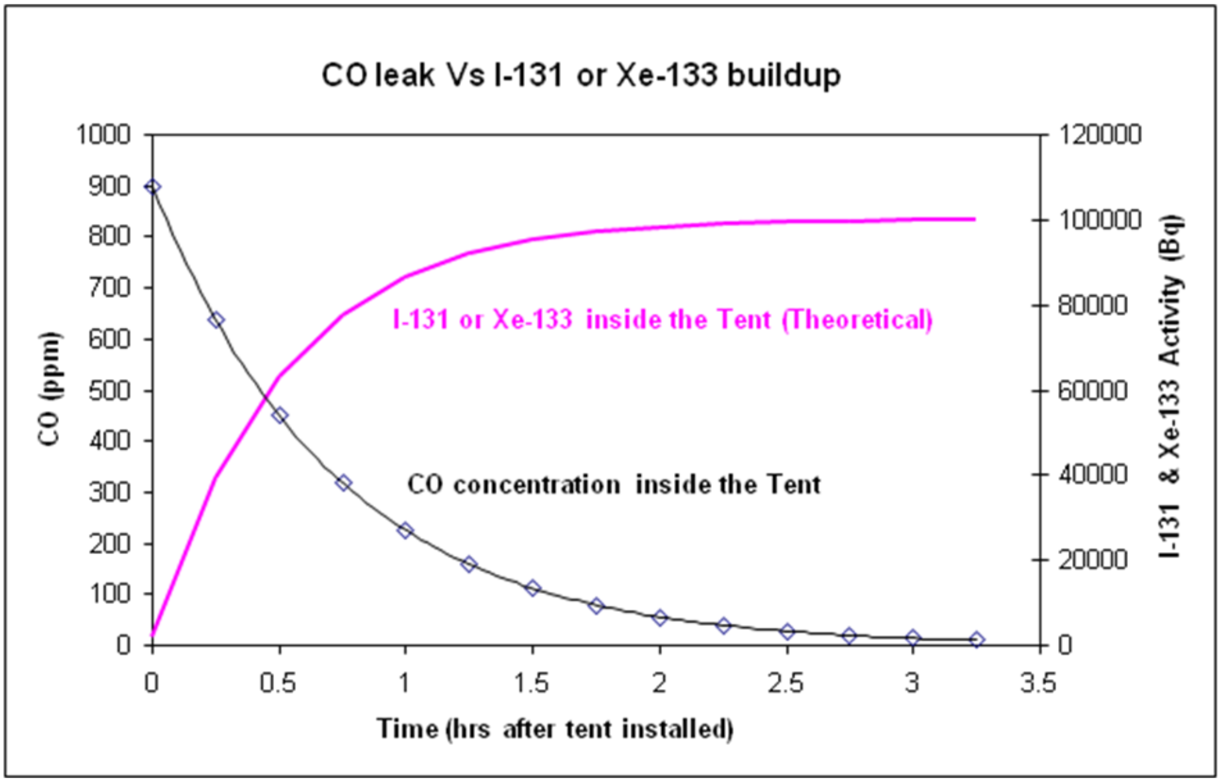

Figure 3 Theoretical diagram of CO leak Vs 1-131 or Xe-133 buildup

Figure 3 shows the theoretical concentration buildup of iodine and xenon, while CO leaks from the tent. After the injection of CO is complete, the concentration of CO will diminish and iodine and xenon level in the tent will continue to increase to an equilibrium level. Figure 4 shows the experimental results of CO concentration from the system under development for about 2 hours after the tent was injected with CO. As expected, the CO concentration decreased with time allowing iodine and xenon to build up in the tent.

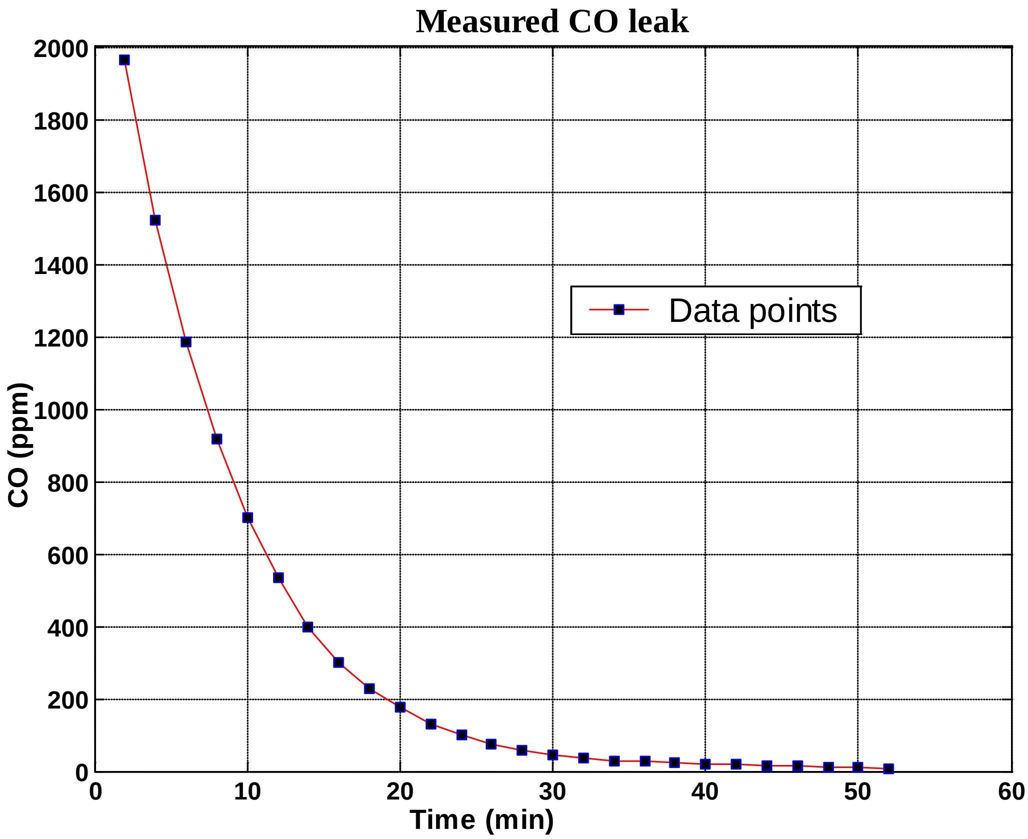

Figure 4 CO concentration in the tent measured by the system under development

4. Instrument Development

The automated iodine and xenon sampling system is a tent enclosure (Figure 5), which is placed over a tile hole or a flask and the sample collection process is started by the operator. The sequence is controlled by a LabVIEW program running on a portable PC controlling I/O hardware. The program executes the specified capture sequence and initiates, measures, controls, and directs the sample flow appropriately to fill the sample tubes. CO concentration, process flow, temperature and humidity are monitored throughout the sequence. Once the capture sequence is complete the removable sample vault containing the active gases is transported for gamma counting by a Germanium detector system.

4.1 Tent Enclosure

The tent enclosure consists of a cubic chamber, a compartment to hold the instrument package: sampling vault and a control case (PC and controller), and a roof to protect the system from the environment, all as shown in Figure 5. The tent weighs about 200lbs and it has 4 hoisting hooks on the top corners to be lifted by a crane, or there are 2 lifting handles on each side of the tent, two people can pick it up and move it in ideal conditions. The tent dimension is approximately 1.3 m x 1.3 m x 1.8 m. The interior surface is lined with plastic to minimize the surface absorption of iodine and xenon. The access panels on the sides can be opened and closed to have access to all devices in the instrumentation compartment.

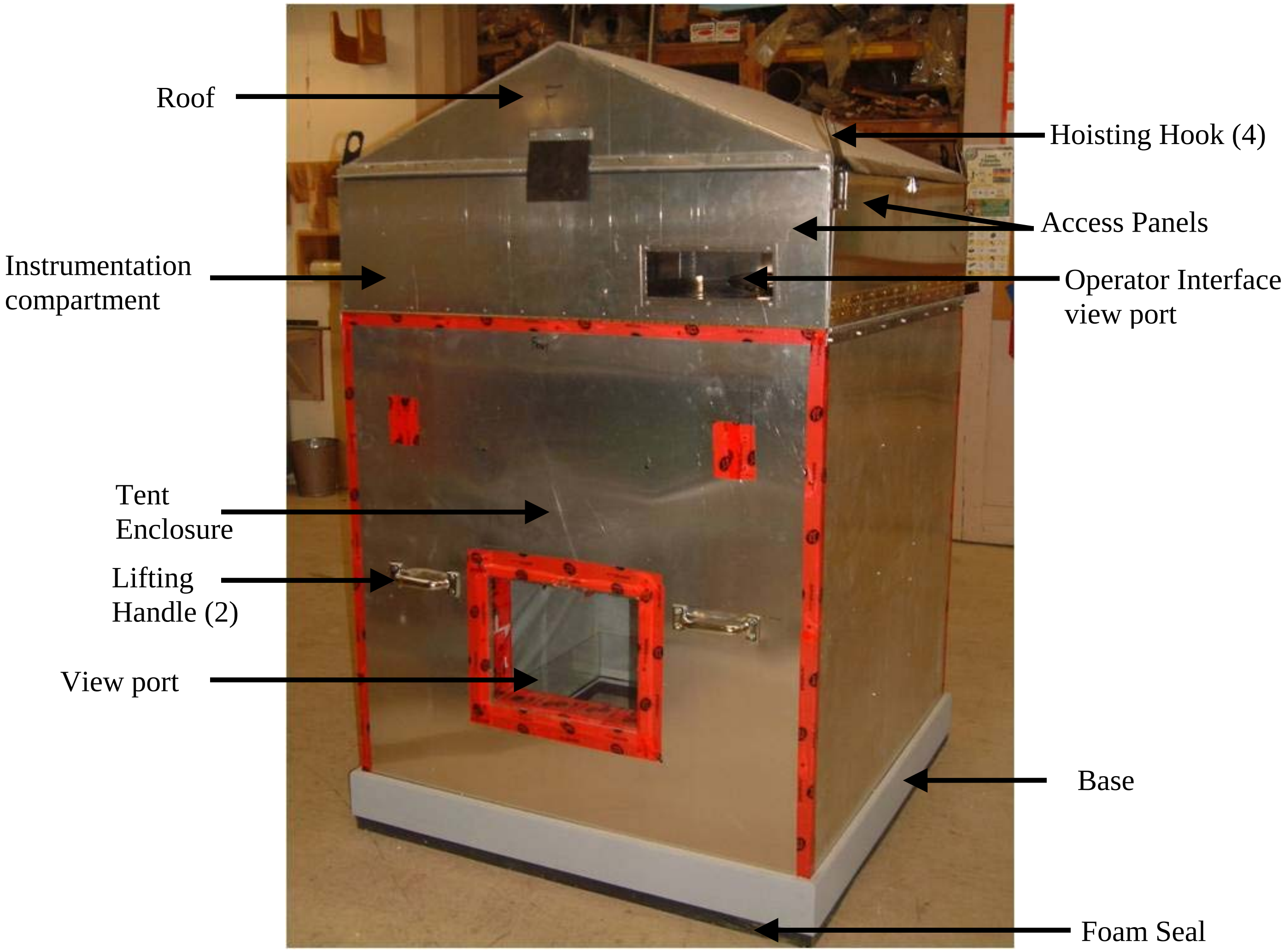

Figure 5 Tent Enclosure that placed on top of a tile hole or shipping flask to capture ^{131}I and ^{133}X

The carbon monoxide (CO) injection system consists of a CO bottle attached to the outside of the tent as shown in Figure 2. The tent contains a CO sensor, humidity sensor, and temperature sensor mounted inside the tent enclosure and wired to the Control case (Figure 9). The CO injection is controlled by a pressure regulator (< 1000 ppm) and a solenoid valve, monitored and recorded in the PC.

4.2 Sample Vault

The sample vault is a case that contains 7 iodine filters and 7 xenon tubes. As shown in Figure 6, there are a series of seven flow paths with inlet valves and outlet valves. All these components are plumbed together with lengths of tubing to create the required flow paths. Two sets of two wiring harnesses connect the valves electrically to a Bendix connector for actuation.

Experiments in the past have shown that, from many chemical forms of iodine, the majority of the emission from the tile holes are highly volatile organic forms of ^{131}I. The iodine filter is a Teflon tube with 3/8” diameter. To study the different forms of iodine emissions, the iodine filter was put together as three staged filter [1], which allowed the measurement of I-131 in three components: elemental iodine, low-volatility organic iodides and high-volatility organic iodides. The filter sections, in the order encountered by the incoming gas are: Front section, which contains CdI_2 (supported on Chromosorb P) and selectively absorbs elemental, highly reactive I_2. Middle section, which contains iodophenol and absorbs low volatility organic form of iodides. The last section, which contains tetraethylenediamine (TEDA) activated charcoal, absorbs high volatility organic form of iodides. The measurements indicated that the iodine present in the speciation tube is primarily in volatile organic form and no elemental iodine or less-volatile organic iodides were detected. Therefore, the speciation tube was modified to be completely filled with TEDA-activated charcoal (Figure 6). The presence of Xe-133 in the iodine speciation tubes are flushed by pumping air through the tube for a short period of time. Xenon tubes are Plexiglas tubes with 1 ¼" outer diameter and 1/8" wall thickness. Xe-133 in the air is the main radioactive gas present. The xenon tubes collect the air samples in the tent to later measure the Xe-133 in the air sample by gamma counting.

The automated sampling process runs until acceptable amount of samples are collected, which is preconfigured by the operator. Therefore it is not necessary for the operator to be present while the sampling process is in progress. Once the sampling process is completed, the sample vault that contains the speciation tubes are removed and taken for gamma counting.

Figure 6 Sample Vault that contains 7 sets of iodine filters and xenon tubes

4.3 Instrumentation case

The instrumentation case contains a flow sensor, pump and three solenoid valves to establish and measure the sampling air flow (Figure 7 and Figure 8). Normally the Pump Outlet valve V9 and Tent Vent valve V11 are open to re-circulate the pumped air back to the tent. The pump flow is set to a slow rate (< 0.5 litres/min) to allow iodine to react efficiently with the iodine filters. During a purge operation the Tent Vent valve V11 is closed and Atmospheric Vent valve V10 is opened to exhaust the purge air from the system to the atmosphere. All of these components are capable of functioning in low temperatures, as low as -40^{o}C. They are arranged on a floor plate inside the Instrumentation case with appropriate size transitions as needed to properly plumb the process lines.

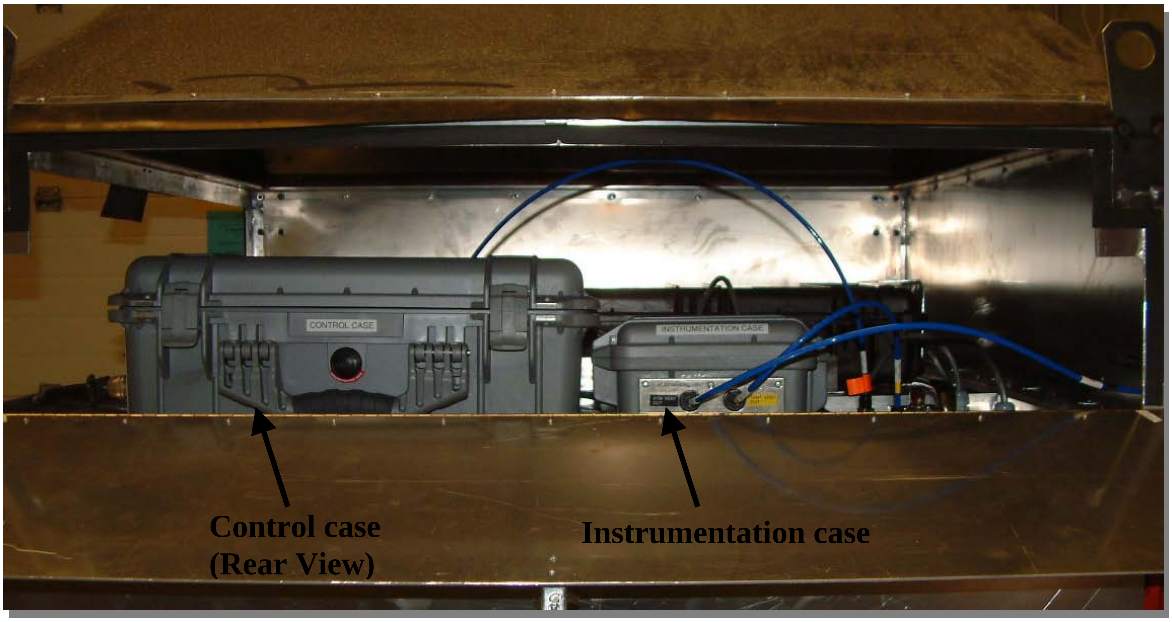

Figure 7 Instrumentation case and control case placed in instrumentation compartment

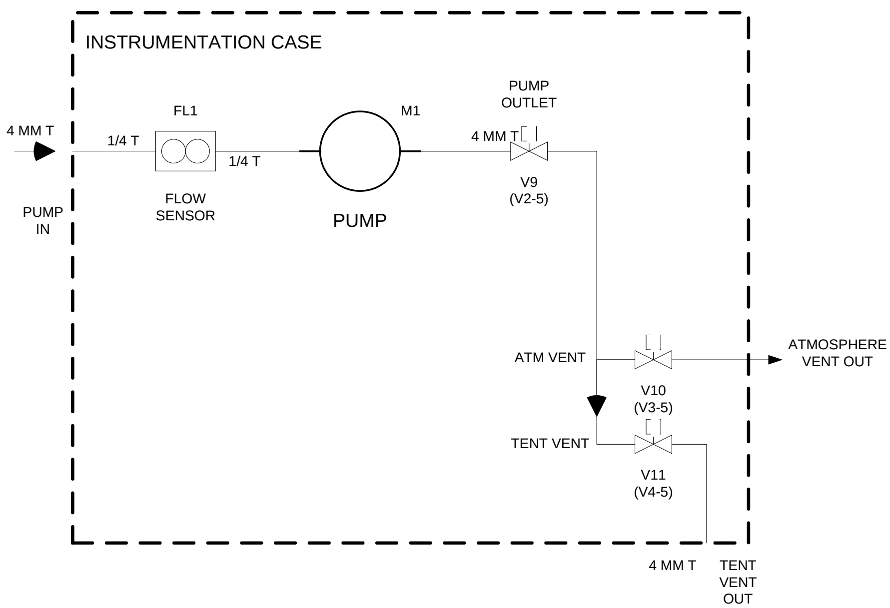

Figure 8 Instrumentation case process diagram

4.4 Control case

The Control case (Figure 9) houses the operator interface located on one side of the case where the operator can interact with the sampling process, process controller and Human Machine Interface (HMI). All sensor signals are wired to the Control case. The process controller directs the flow sequencing and records all sensor inputs for later analysis. The control of all the valves is performed through a matrix arrangement with a column and row selection to activate any one particular valve. There are five columns (inlet, xenon outlet, purge, outlet, and miscellaneous valves) and seven row selections for the seven sample sets.

(a) Control case – front view **(b) Control case – inside**

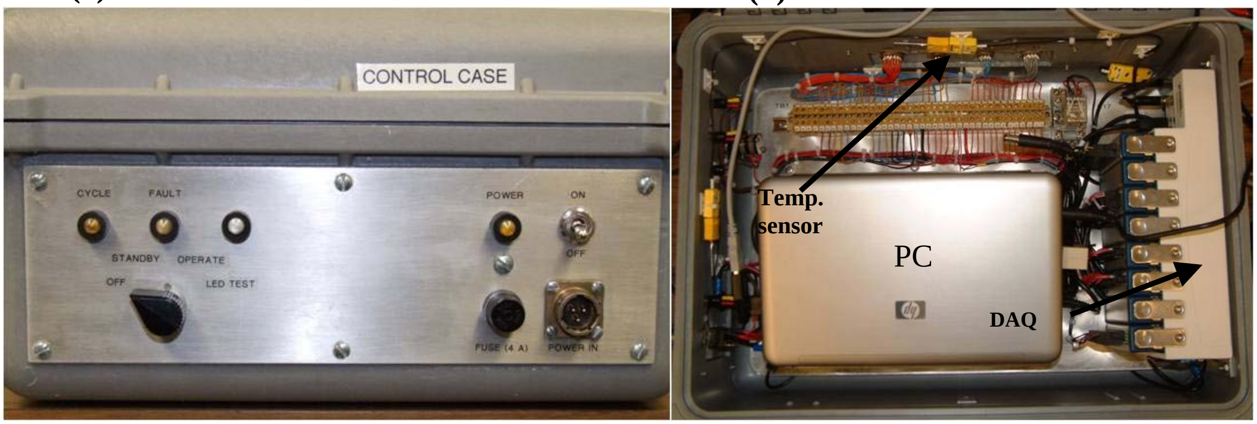

Figure 9 (a) Control case front view consists of operator console, accessed through the view port of the tent. (b) Control case consists of a PC, controller and 3 temperature sensors.

4.5 Software

The application software for the automated iodine and xenon sampling system was developed in LabVIEW. It consists of a software driver supporting the connectivity between the PC and the DAQ modules, and an integrated Human Machine Interface (HMI) and control program which incorporates a state machine that waits for a sequence of operations to be initiated. The sequence of operations is described in a textual 'language' which permits modification with a text editor. Once initiated the software sequences through each step and performs the required actions. As shown in Figure 10, the HMI, provides real time status indication of the equipment during the run; this is useful for debugging, and in future will be utilized for remote monitoring during sampling operations. The log file output generated by the software consists of sequential records containing time stamps, CO concentration, flow, tent temperature, battery voltage, tent humidity and three temperatures from inside the Control case. The log file is in text format, allowing it to be imported into many readily available analysis tools.

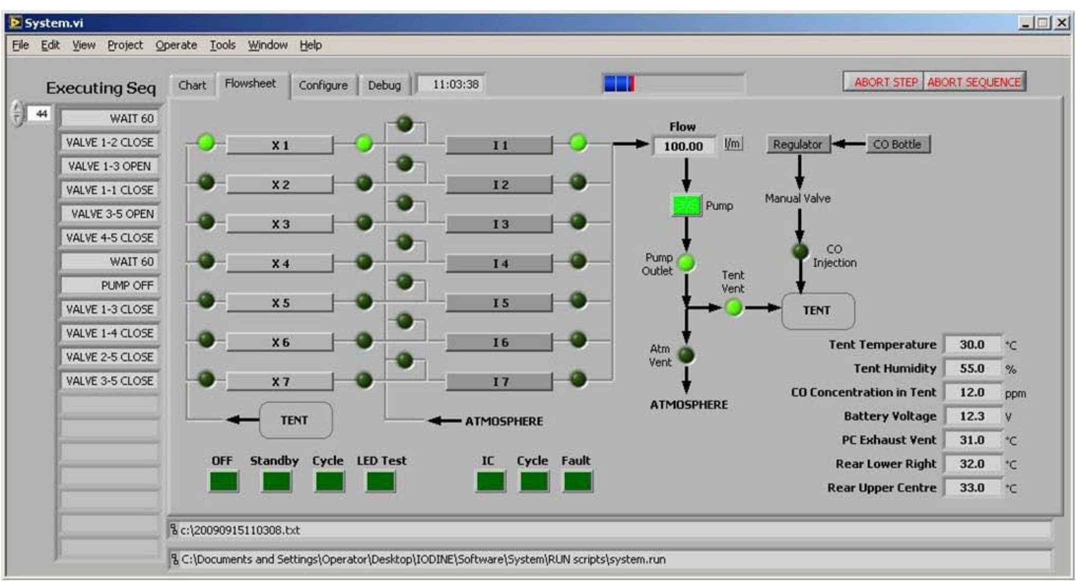

Figure 10 HMI (Human Machine Interface) screen shot showing the process controls and the sensor readings

5. Conclusions

In conclusion, a portable automated iodine and xenon sampling experimental system has been developed to sample ^{131}I and ^{133}Xe emanations from the tile holes and shipping flasks in WMA. This system was developed to achieve the main goals of automated sampling to reduce operator exposure time to radiation as well as to overcome the measurement limitations in manual operations in the past.

The automated sampling system is designed to handle summer and winter weather conditions. Also the instrument is self powered with rechargeable batteries to operate in WMA, where no electrical power line is available. The instrument captures 7 sets of iodine and xenon samples at a time, monitors tent temperature, air flow rate, CO concentration, battery power, tent humidity and records many other readings. The software is programmed with flexibility so that the operator can configure the sampling process according to the needs. Alarm conditions and wireless remote monitoring capabilities are being investigated to improve the system's capabilities.

Application of this transportable automated sampling system will contribute to many improvements in AECL's emission monitoring program in the future. The results obtained using the system will help decide whether or not mitigation is required, and if monitoring is needed on an occasional or on a regular basis.

6. References

[1] G.Jonkmans, D.Everall, N.Munir, T.Popistas, B.Sur, G.Tapp, P.Tonner, and S. Yue, "*Measurement of Controlled Iodine-131 Release in an Outdoor Environment*", Sixth American Nuclear Society International Topical Meeting on Nuclear Plant Instrumentation, Control and Human-Machine Interface Techno ˘ ˏ 2009.