Inspira- Journal of Modern Management & Entrepreneurship (JMME)
(A National Multidisciplinary bi-lingual Quarterly Double Blind Open Access Peer Reviewed Refereed Journal)
ISSN : 2231–167X (Print), Impact Factor: (Cosmos: 7.866 & I2OR: 5.675) Volume 16, No. 02, April-June, 2026, pp. 163-171

Revolutionizing Noble Gas Sampling: Method Development and Performance Analysis

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***Citation**: Jain, S. & Kamlesh, P. (2026). Revolutionizing Noble Gas Sampling: Method Development and Performance Analysis. Journal of Modern Management & Entrepreneurship, 16(02), 163–171. https://doi.org/10.62823/JMME/16.02.9058*

ABSTRACT

Radioactive xenon isotopes are members of the fission product noble gases (FPNG) family and help in the identification of production mechanisms and their extent in any nuclear installation. Therefore, it is necessary to assess the FPNG concentration in the air during the operation of any nuclear power plant. Presently, the available methodology of assessment of FPNG at almost all Nuclear Power plants worldwide (mostly light water-based) is through a computer-based gamma-ray spectrometry system. This method provides FPNG qualitative and quantitative identification through peak search within the acquired spectra of the collected air samples. For this purpose, the radioactive air sample from the active air of the plant is collected by the direct grab sampling method, which has inherent disadvantages as the sample size should be large enough to collect a sample representative of the entire stream. Usually, the collected samples are then analyzed in a high-purity germanium detector (HPGe) as a part of the gamma-ray spectrometer. This paper presents details about an alternative novel method for collecting and analyzing FPNG. This method is developed to overcome the disadvantages of the traditional method. The sampling here is based on cryogenic adsorption of noble gases on activated charcoal, enabling pre-concentration and improved detection sensitivity. Hence, compared to the traditional grab sampling technique, this method enhances the radioactive air collection capacity, ultimately reducing the minimum detectable activity and improving the sensitivity of environmental monitoring.

***Keywords**: Noble Gases, Gamma Spectrometry System, Grab Sampling, Adsorption Phenomena, Analytical Method.*

Introduction

In nuclear industries, there is a requirement to identify the sources of radiation and the specific radionuclides present in order to manage the radiation exposure rate effectively and to design appropriate shielding and safety systems. Gamma-ray spectrometry is widely used in nuclear facilities for qualitative and quantitative identification of radionuclides and for monitoring the state and performance of nuclear fuel during reactor operation **[1-3]**. Combining this method with the contaminant of interest concentration actions (e.g., water evaporation, sequestration cartridges, chemical extraction, etc.), the capacity of the spectroscopy system can be enhanced, and thus, minimum detectable activity concentration of the searched species in the sample can be reduced.

High-resolution gamma-ray measurements in this study were performed using a high-purity germanium (HPGe) detector system, which is commonly used for radionuclide identification due to its excellent energy resolution **[4,5]**.

Fission product noble gases (FPNGs) are produced by nuclear fission processes **[3-7]**. FPNG are mainly beta-gamma emitters **[1,2]** and contribute significantly to the early detection of eventually off-normal conditions in facilities managing irradiated nuclear fission fuels, e.g., nuclear power plants or fuel reprocessing facilities. Noble gases are chemically inert and exhibit weak interactions with most materials, allowing them to diffuse easily through many containment media. This property makes the representative collection of radioactive noble gases particularly challenging **[6,8]**.

Gases such as xenon and krypton can become trapped within cavities in a structure, similar to how water forms ice under high-pressure gas conditions. These trapped gas structures are known as clathrate compounds, or more specifically, in this context, noble gas hydrates. They have a formula approximating $6H_2O$:1 atom of gas. The noble gases of high atomic number can also be trapped in cavities of synthetic charcoal. Clathrates compound formation provides a scientific way of collection and concentration of radioactive isotopes of Kr and Xe **[2-12]**. The primary motivation behind developing this method is to improve environmental monitoring capabilities, particularly for detecting low concentrations of FPNGs that may escape beyond the containment boundaries of nuclear facilities. This newly developed sampling method enables more precise and accurate qualitative and quantitative analysis of FPNG gamma-emitting radioisotopes trapped within these structures.

Available Sampling Techniques

Any air sampling system consists of three basic parts:

- A collecting device that separates the contaminant from the air.
- A metering device for measuring the volume of air sampled.
- A source of suction (a vacuum pump) for drawing the air to be sampled

The airborne radioactivity concentration is determined by dividing the collected activity quantity by the collected air volume sampled. It is very much required that the air-sampling system, the flow meter, the collector, and the pump are calibrated as a whole system **[1,2]**. Vacuum pumps used in air sampling can vary in design, but for this study, a heavy-duty low-flow pump was used to ensure consistent air intake through the charcoal adsorption unit.

The requirements of any sampling device are determined by the type of air contamination targeted for collection. Filtration, thus sequestration of the contaminant to be measured on a specific cartridge, remains the most common method for collecting airborne particulate.

Each collection device has its own flow rate and physical, chemical, and filtration properties. Sampling of any contaminated air is done by two mechanisms, usually:

- Impaction **[1,2,11]** captures particles smaller than the pore size.
- Sieving **[1,2,11]**, which captures particles that are larger than the pore size,

In an impaction process **[2]**, airborne particles often keep a linear path because they are inertial. During the filtration process, the surrounding air travels through the twisting paths that are made of the filter's pores, but the airborne particles continue on their linear path. Eventually, the airborne particles collide with the structure of the filter, which is where impact occurs, and the airborne particles become trapped. Therefore, with a filter, it is beneficial for a large volume of air to move through the filter. Filters made of glass, fibre, or paper matrix perform filtration and trapping of particles in the medium, while membrane filters retain or trap particles just on the surface area of the filter. This difference is especially important when collecting samples for alpha or low energy beta particle analysis, as it will affect the corrections for self-absorption when measuring those radioactive particles. Due to this reason, and their high retention efficiency for respirable-sized particles, membrane filters are widely used and preferred for the sampling of radioactive aerosols.

Airborne contaminated radioactive gases are usually collected by several different techniques, relying on adsorption phenomena, some examples are given below. Adsorption is a process where a single layer of gas molecules adheres to the surface of specific granular materials known as adsorbents. Generally used adsorbents include activated charcoal, activated alumina (a very porous form of $Al_2O_3 \cdot 3H_2O$), silica gel (a hard, glassy form of SiO_2), and molecular sieves **[1,2,9]** (processed Na or Ca aluminosilicates of high porosity). An adsorbent's capacity for sample collection is determined by factors such as its specific surface area (m^2/g), the surrounding temperature, and the partial pressure of the gas involved. Their binding capacity is characterised by a curve called an adsorption isotherm **[5,9]** in which the equilibrium pressure at constant temperature is plotted against the amount of gas adsorbed per gram of adsorbent. Usually, the adsorbent is placed in an appropriate holder, and the air is pulled through the adsorbent.

For completeness, some commonly used airborne radionuclide sampling approaches for iodine vapours and radon gas are briefly mentioned here to highlight differences between their collection mechanisms and those required for radioactive noble gases. Radioiodine isotopes are known precursors of xenon isotopes in the nuclear fission decay chain, while radon is another radioactive noble gas commonly monitored in environmental radiation studies **[13,14]**. Radioiodine in airborne environments is typically collected using adsorption cartridges containing activated charcoal or chemically treated carbon media designed to trap reactive iodine species through strong chemical adsorption mechanisms **[15-17]**. However, such techniques are primarily suitable for reactive vapours and are not directly applicable to inert noble gases such as xenon and krypton. Radon is generally monitored using passive collectors based on adsorption or diffusion mechanisms **[18,19]**. Although radon monitoring methods are widely used in environmental radiation studies, their sampling principles differ from those required for fission product noble gases such as xenon.

When sequestration of the contaminant on the adsorbent media is particularly difficult, or cannot be applied, a simple grab sampling method can be used. In grab sampling, an evacuated glass washing bottle is opened in the radioactive atmosphere supposed to be sampled or water-filled glass bottle is emptied into the atmosphere, thus allowing the radioactive air to enter the glass bottle. The radioactivity level of this contaminated air was calculated by transferring the gas to an ionization chamber **[1,2**] and then measuring the ionization current due to the radioactive gases by gamma spectrometric analysis of the collected sample. The ion chamber provides quantitative analysis, and the semiconductor-based gamma spectrometer offers qualitative and quantitative analysis. The disadvantage of this method is the poor accuracy, representativeness and reliability of results on repeat sampling and analysis, but grab sampling is often used when the contaminant does not exhibit significant interaction with available adsorbent media or when rapid measurements are required. However, adsorption-based techniques using materials such as activated charcoal have also been investigated for the collection of radioactive noble gases including xenon **[20]**.

Development of Novel Sampling Methodology

Since FPNGs are inert gases, it is challenging to collect representative samples. A special sampling technique was developed for this, which is described in section 3.1.1.

In this study, the following methodology was adopted to describe the attachment of molecules and atoms to a surface, which can occur through two mechanisms **[10,12,13,17]**:

- Physisorption
- Chemisorption

Physisorption occurs by the interaction between the adsorbate and surface using weak van der Waals forces **[12,13,17]**. Van der Waals forces are long-range, but their strength is very low. The energy released during physisorption is similar to the enthalpy of condensation. It is of such a small quantity that the lattice vibrations of the substrate readily absorb it without acting to perform a work term - it is spontaneously dissipated as heat. Therefore, a freely travelling molecule over the surface will lose its kinetic energy until accommodation occurs and it becomes bonded to the surface.

In chemisorption (Chemical adsorption), the molecule sticks to the surface through a strong chemical bond, making the interaction much stronger and more specific than physisorption, which relies only on weak physical forces. The distance between the surface and the closest adsorbate atom is also typically shorter in Chemisorption than for physisorption.

Hence, according to the Chemical properties of Noble gases, they are trapped by following any of the above mechanisms in this newly developed method.

Main Components

- **Basis of Development of New Technique**

  A special sampling system was developed for this, for which the below items are required:

  - A Bubbler (gas washing bottle of 500 ml)
  - Semi/Partial Crushed Charcoal Filter

- Flow rate meter for sample volume calculation in cubic feet per minute (cfm)
- Low-flow heavy-duty pumps run on AC supply
- Liquid N_2 in an industry supply container can carry 3 Litre
- All the equipment can be mounted on a handheld tray to ease transportability

The sample setup diagram with the design is presented in Figure 1.

- **Sampling Method Steps**

The FPNG sample is collected using a newly developed method based on adsorption phenomena explained before. The steps listed hereafter are followed:

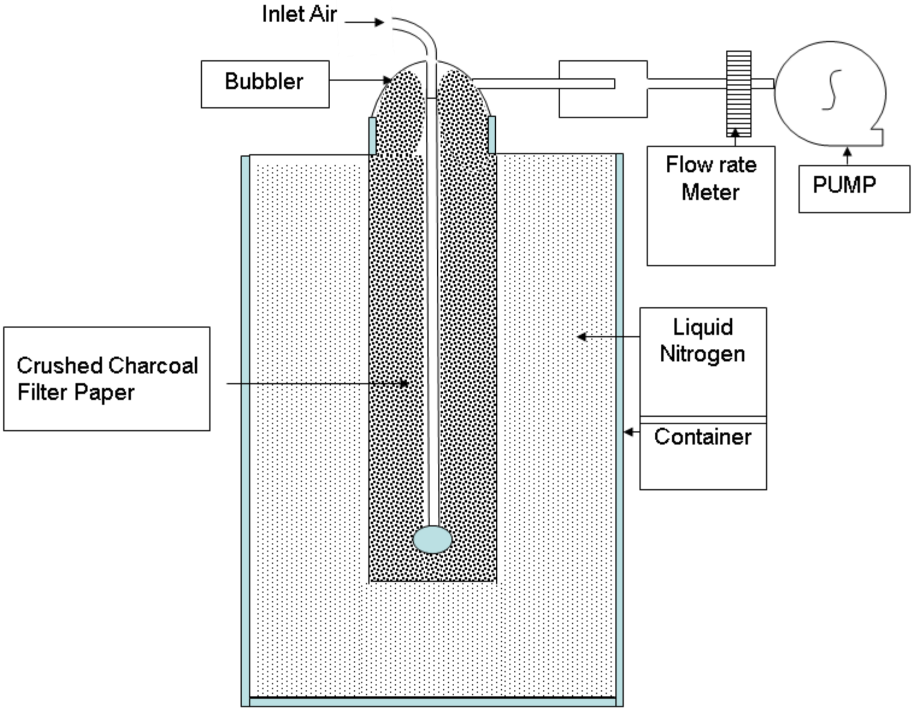

Figure 1: Novel sampling setup design

- An SS container is taken and filled with liquid N_2 up to the neck
- A fixed amount of semi crushed Charcoal filter is poured into a glass bubbler
- A heavy-duty electrically operated pump and flow rate meter are connected to a bubbler
- For the flushing of the sampling line, a PVC tube is used to discharge active air into a fresh water bottle.
- At the sampling line the top of the bubbler stem was connected and flow was adjusted to one cfm.
- The sample is collected at one cfm for a variable time, so total sampling time will be maintained in all experiments.
- After sample collection, bubbler is taken to the gamma spectrometry lab for analysis purposes.
- The gamma spectrometer was calibrated for energy and efficiency using standard sources with the same sample geometry prior to analysis. All xenon isotopes present in the collected sample are adsorbed simultaneously in the charcoal medium, and their identification and quantification are subsequently performed using gamma-ray spectrometry based on their characteristic gamma-ray energies.

- **Theoretical Basis of New Methodology**

This setup was designed based on the following property of noble gases by which we can collect a representative sample of FPNG.

Compound Formation of Noble Gases

During radioactive sample collection, noble gases can easily diffuse through different sampling media like plastic, rubber, glass, and some metals, making them difficult to handle in the laboratory. For the collection of representative samples of FPNGs, attention was paid to their special chemical properties, such as the formation of Clathrate **[12,17,21]** compound of Noble gases.

The gases FPNG may be trapped in cavities in the same way as water is frozen under the high pressure of the gas. These are Clathrate compounds, commonly termed as Noble gas hydrates. They have a simple formula approximating $6H_2O$:1 gas atom. The heavier noble gas may also be trapped in cavities of synthetic charcoal. Clathrates compound formation provides a convenient means of storing radioactive isotopes of Kr and Xe produced in nuclear reactors.

Basis of Compound Formation

The following literature survey basis is considered before the development of a novel method to ascertain the sampling methodology for future application:

- **Le Chatlier's Principle for Dissolution of Gases**

Dissolution **[12,17,21]** of gases in liquid (hydrate) is always exothermic. Therefore, the solubility of gases in liquids increases with a decrease in temperature.

- **Adsorption**

Under normal conditions, a surface exposed to a noble gas and continuously bombarded with its molecule forms a freshly prepared surface very quickly over the exposed surface. The number of gas molecules striking a unit area of surface per unit time can be estimated using the kinetic theory of gases, which is expressed in the collision flux:

$Z_w = p/(2\pi mkT)^{½}$

Where m= molar mass of gas (for air = 29 g/mol), p = pressure, T= temperature

At NTP, the collision flux for air is 3×10^{27} $m^{-2}s^{-1}$. Temperature is an important factor to maximize adsorption; adsorption is an exothermic process. Therefore, according to Le Chatelier's Principle, when temperature is decreased, this would increase adsorption. This is the reason for using liquid nitrogen (LN_2) to lower the adsorbent temperature from ambient conditions (298 K) to cryogenic temperatures, enabling efficient adsorption of noble gases such as xenon and krypton on adsorbents like charcoal. When the gas molecules are cooled to cryogenic temperatures, which means little to no thermal motion to effectively increase access for binding with the molecules on the adsorbent's surface, the binding action would be able to be done much more efficiently, maximizing the trapping capability of the cartdridge, thus enhancing the measurement sensitivity of the whole system, especially at environmental concentrations that are low. These principles form the basis for the use of cryogenic adsorption in the present sampling methodology.

- **Extent of Adsorption**

The extent of surface coverage is usually expressed as fractional coverage

μ = Number of adsorption sites occupied / Number of adsorption sites available

Result and Analysis

The results are obtained using the Canberra make (P-type coaxial detector) system and are discussed in Table 1.

- **Sample Collection Duration:** 10 minutes with a flow of 1 cfm (about 280 l of contaminated air per single measurement)

The reported activity concentrations (pCi/cc) are normalized with respect to the sampled air volume, where approximately 280 L of air is processed in the bubbler-based method and about 500 mL in the grab sampling method, ensuring that the values represent true activity concentration in air.

- **Spectrum acquiring time:** 600 seconds

Time for single sampling & analysis: 60 Minute

Minimum Detectable Activity (MDA) Concentration: 1pCi/cc for xenon isotopes per sampled air volume unit

Table 1: Estimation of Air Sample

Experiment	Xe-133 (pCi/cc)			Xe-135 (pCi/cc)			Residual Activity (pCi/cc)	
	New Method	Old Method	Ratio	New Method	Old Method	Ratio	Xe-133	Xe-135
I	266	6.5	40.9	126	3.5	36.0	1.6	1.0
II	300	7.9	38	160	3.8	42.1	1.7	1.1
III	316	7.2	43.9	299	7.1	42.1	1.8	1.6
IV	510	11.6	44	440	12	36.7	2.1	1.9
V	452	11.7	38.6	351	8.2	42.8	2.0	1.7
VI	1155	168	6.9	1249	71.6	17.4	6.1	7.3

Note: Residual activity represents the remaining activity measured in the second bubbler, used to assess trapping efficiency of the primary charcoal stage.

The residual activity values measured in the second bubbler stage are significantly lower, indicating that the primary charcoal unit captures the majority of xenon isotopes and confirming the high trapping efficiency of the proposed sampling system.

From the above tabulated experimental data, the following result was observed:

- Samples were collected from the same environment and during the same time period, but using different sampling volumes and collection mechanisms. Table 1 shows that samples collected with the bubbler method are approximately forty times higher than grab sampling data for both radionuclides, Xe-133 and Xe-135. The improved sensitivity of the proposed method arises from pre-concentration of xenon isotopes due to processing of a larger air volume. Since the same HPGe detection system with a minimum detectable activity (MDA) of approximately 1 pCi/cc is used for both methods, the increased collected activity in the bubbler-based technique enables detection of lower concentration levels that remain below the detection limit in the grab sampling method. Thus, the effective detectable concentration in air is significantly reduced using the proposed method. The experimental data indicate that activity values measured using the grab sampling method approach the detection limit for low concentration cases, whereas the proposed method provides significantly higher measurable activity for the same conditions. Figures 2 and 3 present the same data graphically to illustrate the consistent enhancement in measured activity obtained with the bubbler-based adsorption method across multiple experiments, highlighting the systematic difference between the two sampling techniques.

Moreover, during the experiment it was observed that the trapping efficiency of bubbler I charcoal is more that 99% (bubbler II charcoal was found practically clean), and we can say that first bubbler measurement is quite absolute. Hence it proves that a series of sampling bubblers is not required for the activity concentration range measured in the effective application carried out in this paper for our system (the second bubbler may be anyway useful in other applications to confirm that bubbler I makes an absolute measurement). Moreover, trapping the bubbler I the whole contaminant in the air, activity concentration values measured with the systems here developed can be assumed as true values of the contamination within the air sampled. From comparison of values with those coming from the grab sampling method, it can be seen that grab sampling method is underestimating the contamination in air, especially at lower activity concentration values.

- From the data presented in Table 1, it is evident that in all cases, grab sample results show that sample collection capacity by this way is low or the sample collected from grab sampling is not representative of actual radioactive air concentration.

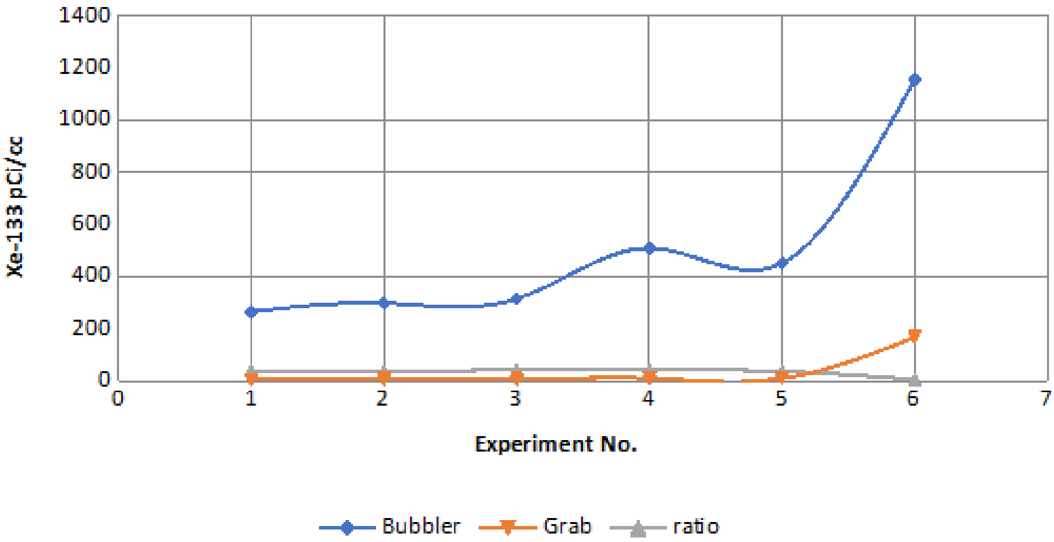

Figure 2: Comparison of Xe-133 activity levels measured using the new bubbler-based adsorption method and the conventional grab sampling method across multiple experiments. Each point represents a separate sampling event, with activity measured in pCi/cc

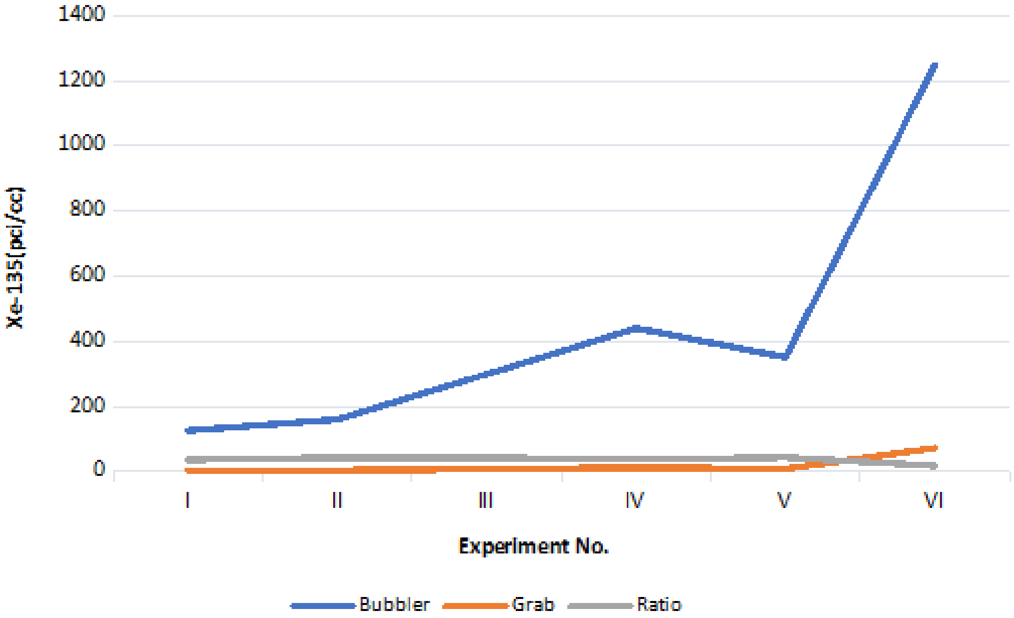

Figure 3: Comparison of Xe-135 activity levels obtained by the new bubbler-based technique versus the conventional grab sampling approach. The graph highlights the significantly improved detection capability of the new method, particularly at low concentrations

- It is also observed that in the case of a high concentration of air activity in grab samples, the results also favour new method sampling. This means the grab sampling method is not suitable for low and very low levels of radioactivity concentration sampling (Figures 2 and 3). However, it remains a simpler and faster option for intermediate and high levels of air concentration sampling and analysis.

- The experiment described above was performed to provide another opportunity to sample noble gases at low concentrations in a more reproducible way, which is supported by the obtained results.
- Figures 2 and 3 clearly indicate that the ratio of samples by both methods has a correlation in FPNG data. In both cases, it is observed that the concentration of xenon has a ratio, as we see that in the case of the 4 & 6 sampling results, both trends are increasing.

Regardless of the collection method, all samples were analyzed using the same calibrated gamma spectrometry system, minimizing inter-method bias. The consistently higher activity values (~40 times) observed with the new method across multiple trials indicate improved sensitivity and sampling efficiency. The observed enhancement arises from volumetric pre-concentration of xenon isotopes due to the significantly larger air volume processed in the bubbler-based method (~280 L) compared to grab sampling, leading to improved detection sensitivity. Since both samples were taken from the same ecosystem, time and place, and the same HPGe gamma spectrometry system was used to analyze both samples with the same parameters, the associated uncertainties, such as increases in varying air volume sampled (related to time sampled), the system efficiency, or the response of the detector, were all systematic, so they all ultimately represent the same uncertainty with respect to both techniques. Systematic uncertainties are expected to affect both methods similarly; therefore, the comparative evaluation remains valid. Accordingly, we felt it was unnecessary to provide the same level of detail with respect to the uncertainty calculations for the purpose of this study.

- **Gamma Spectrometry System Details**

Sample analysis was performed using a P-type coaxial HPGe detector system (150 cc volume) with a resolution of 0.8–1.2 keV/channel and a counting efficiency of 18% at 1 MeV. Energy and efficiency calibrations were conducted using certified multi-isotope gamma sources prior to each measurement series. The detector efficiency was determined for the same sample container geometry used in the experiments, thereby accounting for the geometrical efficiency of the detector–sample configuration during activity measurements. Identical borosilicate glass containers were used for both sampling methods, and all measurements were carried out with fixed geometry and consistent detector positioning.

Additionally, it should be noted that all activity measurements reported were derived using the HPGe detector system’s built-in analysis software, which outputs results at a 3σ confidence level above the lower limit of detection (LLD) per volume.

Summary

Based on the proposed methodology, multiple sampling experiments were conducted using both the conventional grab sampling method and the newly developed bubbler-based adsorption technique. The results demonstrated a consistent and significant enhancement in the detection of FPNGs using the new method here developed. The activity levels were measured by both approaches, with the bubbler method showing improved sensitivity and representativeness. From measured values, it should be noted that grab sampling is constantly underestimating the activity concentration of contaminants in the air, and the bias of the grab method decreases when contaminant activity concentration in air increases.

Key advantages of the newly developed method include:

- Sampling setup fabrication at low cost on its own without any manufacturer interventions & reusable.
- Reliable, consistent results in each sampling even at lower activity concentration values are also possible with this method.
- Direct sample analysis just after collection without any further process of the sample in the gamma spectroscopy (HPGe) system.
- The sample can be retained up to 72 hours after sample collection for further analysis in the same environmental conditions without any data loss.
- Sample collection location, duration, and time are very important for radioactive decay correction, and thus, the result will be very precise in analysis by this method.

- The volume of sample collected is measurable in this method which is not possible with the old grab sampling method.

The comparative results clearly highlight that the new method provides more consistent, repeatable, and accurate measurements, particularly in low-level activity scenarios with respect to grab sampling methods. Therefore, the bubbler-based adsorption technique presents a reliable and practical alternative for the environmental monitoring of radioactive noble gases and offers potential improvements for routine surveillance around nuclear facilities. The proposed method is especially suited for environmental surveillance applications where the detection of low-level FPNG concentrations outside nuclear installations is challenging using conventional methods.

Future work will focus on standardized calibration experiments using known activity sources to quantitatively benchmark the accuracy and efficiency of the new sampling technique against conventional methods.

- **Data Availability:** The data that support the findings of this study are available from the corresponding author upon reasonable request.

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