A simple method for sampling and measurement of radiocarbon by a passive diffusion sampling technique

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Passive diffusion sampling has been widely used to monitor organic and inorganic pollutants in the environment. We herein developed a new and simple passive diffusion sampling method to monitor $^{14}$C in the indoor working environment of radioisotope facilities. The badge-type Shikata sampler is an open-end type with no diffusion cap that fits a collection filter with a diameter of 25 mm. This sampler could efficiently collect indoor $^{14}$C with good reliability and a collection efficiency that was 10-fold higher than that of the conventional active pumped method. The Shikata sampler is small, simple, and easy to handle. It may be subsidiary used to monitor indoor $^{14}$C concentrations in radioisotope working rooms, in which radioactivity must be measured once a month as recommended by the Industrial Safety and Health Law.

Key words: indoor $^{14}$C sampling, passive diffusion sampler, Industrial Safety and Health Law, Shikata sampler, radioisotope facilities

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1. Introduction

Monthly measurements of radioactivity in the indoor air of all radiation facilities are required by the Industrial Safety and Health Law. Sampling of radiocarbon ($^{14}$C) and tritium ($^{3}$H) is generally conducted using the active pumped sampling method with special equipment. This equipment can collect large volumes of air and, thus, is expected to detect the lower concentrations of $^{14}$C and $^{3}$H in indoor environments. However, some limitations have been associated with the use of this equipment, including its heaviness, expense, and electrical power requirements. Extraction and concentration processes for $^{14}$C using a trapping reagent are also required before measurements can be made. $^{14}$C is not frequently used in most un-sealed radioisotope facilities in universities and/or research institutes, and its concentration level in the air is extremely low. Therefore, sampling of $^{14}$C cannot be always performed.

Passive diffusion samplers have been widely used to monitor organic and inorganic environmental pollutants, including NO$_x$, SO$_2$, O$_3$, NH$_3$, and formaldehyde. These samplers are based on the free flow of analyte molecules according to Fick’s first law of diffusion. Passive diffusion sampling methods have many advantages over active sampling methods: the equipment used is small, simple, inexpensive, easy to handle, and does not need electrical power or a pump. These methods are also suitable for simultaneous and multipoint samplings and measurements in indoor as well as outdoor environments. Furthermore, specialist training and maintenance are not required.

The aim of this study is to develop a passive diffusion sampling method to monitor $^{14}$C in the indoor working environment of radioisotope facilities. A newly designed badge-type sampler was used for this purpose. It enabled the adsorption and measurement of $^{14}$C with good reliability. This method may be subsidiary applied to regular working environment measurements together with the conventional active pumped sampling method in order to meet the requirements of the Industrial Safety and Health Law.

2. Materials and Methods

2.1 Passive sampler

A badge-type Ogawa sampler (Ogawa & Co., Ltd., Kobe, Japan) was tested in this study. This sampler consisted of a Teflon cylinder (2 cm in diameter and 3 cm in length) with two...
open and unconnected cylinders, each of which is equipped with a collection filter that was placed between two stainless steel screens and secured by a polyethylene diffuser end cap with 25×2 mm diameter holes, respectively (Fig. 1). The UMEx passive sampler (8.6 cm × 2.8 cm × 0.89 cm in size, SKC Inc., Eighty Four, PA, USA) equipped with a 112-hole diffusing plate was also used.

2.2 14C source

A 14C source was prepared as follows: 50–100 µl of [1, 2 - 14C] - sodium acetate (3.7 kBq/µl, Perkin Elmer, Inc., Boston, MA) was dropped onto cellulose filter paper for the wipe test (No. 63, Advantec Co., Ltd., Tokyo, Japan), which was pre-placed in a stainless steel sample dish (6 mm in depth and 25 mm in diameter, Chiyoda Technol Co., Ltd., Tokyo, Japan).

2.3 Comparison of 14C adsorption ability of filter papers

Three kinds of filter papers were evaluated for their 14C collection efficiencies. One was made of cellulose and glass fibers (HE-40T, Advantec Co., Ltd.), while the other two were made of glass fibers (GB-100R and GA-100, Advantec, Co., Ltd.). These filter papers have been used as pre-filter and/or collection filters in the conventional active pumped sampling method. A 14C radiation source was placed in a polycrylate chamber. The radiation source was removed 24 hours later and the air in the chamber was directly collected for 5 min (40 l/min) using the dust sampler (DSM-361, Hitachi Aloka Medical, Ltd., Mitaka, Tokyo, Japan) equipped with each of the filters, respectively.

The 14C radioactivity of each filter was measured using the liquid scintillation method.

2.4 Preparation of an improved passive sampler

Two types of passive diffusion samplers were produced in our laboratory. We designed a badge-type sampler with a single cylinder based on the Ogawa sampler. It consisted of three main parts: a lid of polyethylene vessel (Zinsser Minis 2001, Zinsser Analytic, Frankfurt, Germany) that served as a cylinder as well as a diffusion part, a collection filter placed between two stainless steel screens, and a diffuser end cap with holes that were 2 mm in diameter (PS sampler, Fig. 2). The diffuser end cap was made of polycrylate using a lathe and milling machine in the Workshop Center Division, Center for Engineering Innovation, Okayama University. Another badge-type sampler, which was based on the Willems badge, had two polyethylene cylinders (6250 sample vial lid, Thermo Scientific Inc., Rochester, NY, USA) and was an open-end type with no diffusion cap (Shikata sampler, Fig. 3). This sampler could fit a larger collection filter (up to 25 mm in diameter).

2.5 A chamber system

A polycrylate chamber box (with a volume of twenty-two liters) was prepared in order to evaluate the collection efficiency of 14C by the passive diffusion sampler. A 14C radiation source (185 kBq) was placed in the bottom of the chamber box. The
increase number of Ogawa samplers (up to 5 samplers with maximum 10 filters) were hung on the top (Fig. 4). After being exposed at room temperature for 5 days, the collection filters were removed and $^{14}$C radioactivity was measured.

2.6 Indoor $^{14}$C sampling

Indoor $^{14}$C sampling was conducted in a radioisotope working room. The $^{14}$C radiation source (370 kBq) on the desk (1 m in height) was placed on the bottom of the room (a volume of sixteen cubic meters). Several types of passive diffusion samplers, such as Ogawa sampler, PS sampler, UMEx passive sampler and Shikata sampler, fitting a total of 6 collection filters were hung on the ceiling approximately 1.5 m above the radiation source. Each of the samplers was tested separately. The door of the room was closed, and free, but weak airflow was allowed at room temperature (Fig. 5). After being exposed for 5 days, each collection filter was removed and $^{14}$C radioactivity was measured.

2.7 Radioactivity measurement

The removed collection filters were placed in a 4-ml liquid scintillator (Clear-sol II, Nacalai Tesque, Inc., Kyoto, Japan) containing assay tubes, and $^{14}$C radioactivity was measured using a liquid scintillation counter (LSC6100, Hitachi Aloka Medical Ltd.). The collection efficiency ($C_{ef}$) was calculated as:

$$C_{ef} = 100 \times \frac{A_t}{A_0 - A_s}$$

Where $A_t$ is $^{14}$C radioactivity adsorbed on the collection filter, $A_0$ is the $^{14}$C radioactivity of the radiation source, and $A_s$ is $^{14}$C radioactivity remaining in the radiation source after 5 days of release.

3. Results

3.1 Selection of collection filters

Three kinds of filters were evaluated for their $^{14}$C collection efficiencies by the active pumped method using the dust sampler. The glass fiber filters (GB-100R and GA-100) had the same capacities, with adsorption of more than 40%. The cellulose and glass fiber conjugated filter, HE-40T, was less effective for $^{14}$C adsorption (data not shown). By comparing the 2 glass fiber fil-
14C sampling by passive sampler

ters, we choose the low-cost GA-100 filter for application to the passive diffusion sampler.

3.2 Confirmation of 14C collection by passive diffusion sampling

We first analyzed the 14C collection efficiency of the passive diffusion sampler using the polyacrylate chamber system. The GA-100 filter, which was cut to a diameter of 14.5 mm, was fit to the Ogawa sampler and exposed to the 14C radiation source for 5 days. As shown in Fig. 6, a collection efficiency of up to 70% was observed with an increasing number of collection filters, thereby confirming the effectiveness of the sampler.

3.3 Effects of hole numbers in the diffuser on 14C collection efficiency of passive samplers

We analyzed the effects of hole numbers in the diffuser on 14C collection efficiency. In addition to the Ogawa sampler, we prepared a newly designed PS sampler with 7 and 17 holes in the diffuser. Each sampler with the same hole numbers was simultaneously exposed for 5 days to the 14C radiation source placed in the radioisotope working room. As shown in Fig. 6, a collection efficiency of up to 70% was observed with an increasing number of collection filters, thereby confirming the effectiveness of the sampler.

3.4 14C collection efficiency of the Shikata sampler in indoor 14C sampling

Since a larger number of holes in the diffuser had no effect on 14C collection as described above, we then examined the effects of the collection filter size. An open-end type of Shikata sampler, which fit the different sizes of collection filters, was developed and exposed to the 14C radiation source for 5 days in the radioisotope working room. As shown in Fig. 8, collection efficiency increased with an increase in the filter size. No significant differences were observed in the collection efficiencies of the Shikata sampler with a 14.5-mm filter and the Ogawa sampler with the same size filter. The Shikata sampler with a 25-mm filter had a collection efficiency of 1.3% and was 3.5-fold more efficient than that of the 14.5-mm filter. Indoor 14C sampling was also performed using the active pumped method with special equipment, which is the general method recommended by the Industrial Safety and Health Law. The 14C collection efficiency of passive diffusion sampling with the Shikata sampler was markedly higher than that of the general active pumped method, which showed a collection efficiency of up to 0.1% (data not shown).

4. Discussion

We herein developed a simple passive diffusion sampling method to monitor 14C in indoor working environments. By using the Shikata sampler equipped with a collection filter that

![Image](https://via.placeholder.com/150)

**Fig. 6.** Analysis of the 14C collection capacity of the Ogawa sampler using the polyacrylate chamber box system.

![Image](https://via.placeholder.com/150)

**Fig. 7.** Effects of hole numbers in the diffuser end cap on 14C collection efficiency.
was 25 mm in diameter, we demonstrated that indoor $^{14}$C could be efficiently collected with good reliability.

Under the Industrial Safety and Health Law, all facilities using unsealed radioisotopes must measure and monitor the concentration of indoor radioactivity in every working room once a month. $^3$H and $^{14}$C are generally sampled simultaneously using active pumped sampling with special equipment. Indoor air samples are continuously introduced into the combustion cylinder of the equipment, and $^{14}$C in the air is oxidized to the $^{14}$CO$_2$ form. The resulting $^{14}$CO$_2$ is trapped and dissolved into monoethanolamine. The concentration of $^{14}$C in the indoor environment is very low and requires a large volume of air to be sampled to detect above-background levels. This active pumped sampling method also involved several steps and is time consuming and costly.

Passive diffusion samplers have been widely used to monitor inorganic and organic pollutants in both indoor and outdoor environments. Although passive diffusion samplers are relatively sensitive to the effects of wind speed, they have several advantages, such as being small, simple, inexpensive, and easy to handle, and do not need electrical power or a pump\(^\text{10}\). There are passive diffusion radial-type samplers such as the Radiello sampler, tube-type such as the Palmes tube, and badge-types such as the Ogawa sampler and Willems badge\(^\text{11}\). Badge-type samplers are designed to have a large surface area for the collection filter in order to achieve useful sampling rates.

Taken together, passive diffusion sampling using the Shikata sampler was effective for indoor $^{14}$C monitoring in working environments. It may also be used to monitor personal exposure as well as outdoor environments. Further studies are needed to examine whether this sampler can be similarly used for a variety of $^{14}$C sources.

5. Conclusion

We developed a passive diffusion sampling method to monitor $^{14}$C in indoor working environments. The Shikata sampler is small, low cost and easy to handle. It can be subsidiary used to monitor indoor $^{14}$C concentrations in radioisotope working rooms, in which radioactivity must be measured once a month as recommended by the Industrial Safety and Health Law, together with the general active pumped sampling method. It may also be used as a personal dosimeter. The passive diffusion sampling method does not require specialist training or maintenance and can be easily applied in the field.

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