AIRBORNE IODINE MONITORING AT THE RADIO-ISOTOPE TEST PRODUCTION PLANT, JAERI

S. FUKUDA, M. NARITOMI, S. IZAWA and Y. IZUMI

Division of Health Physics and Safety, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken, Japan,

Abstract—During production of iodine-131 from an irradiated telluric acid (H₂TeO₄) by the distillation method, a considerable amount of airborne radioiodine is generated in the cell and its major portion is discharged to the atmosphere through a stack after passing through air cleaning systems, consisting of prefilters, charcoal filters and high efficiency particulate filter. Some minor portion of airborne iodine generated leaks directly out of the cell to the service area and operation area, although a negative pressure is maintained in the cell.

To evaluate the airborne iodine concentration in air and the amount discharged as accurately as possible, extensive radiation monitoring was performed using various kinds of monitoring devices. As one of these devices, an iodine sampler was developed which is composed of three sampling components, i.e. HV-70 filter paper, charcoal-impregnated filter paper and charcoal cartridges.

At several sampling positions of the air cleaning systems, the overall collection efficiency of the sampler was evaluated by taking into account activities collected in cold traps (containing granular charcoal) which are connected to the sampler in series. It was found that the overall collection efficiency ranges from 70% to 100%, highly dependent on whether the samples were taken at upper stream or downstream of the air cleaning systems.

Data obtained from the airborne iodine monitoring at the stack and in the working areas conducted during the iodine-131 test production are presented, and internal exposures of the operating personnel received by inhaling the airborne iodine are described in relation to the air contamination of the working areas.

INTRODUCTION

Many of the papers on methods or techniques of airborne radioiodine sampling are mainly concerned with the characterization and behaviour of airborne iodine under the accidental conditions of a reactor. There are few reports (1-7) concerning a routine sampling of airborne iodine released from a stack during normal operation of a radioiodine production facility or fuel processing plant, and also routine airborne iodine monitoring in working areas.

In general, it is much more difficult to sample efficiently and evaluate the concentration of radioiodine in air during routine operations than during accidental events. The more reactive components of airborne iodine have such properties as to be easily adsorbed and to be generally removed in passing through the filtering media of an air cleaning system. The

less reactive components constitute a major portion of the airborne iodine discharged. The behaviour of iodine leaking directly into the working areas from the processing cell, etc., would be rather similar to that in the accidental release.

In the radioisotope test production plant, which was constructed to gain experience for the construction of a large-scale radioisotope production plant, iodine-131, the chemical form of which is NaI in basic sodium sulfite, is produced from a sulfuric acid solution of irradiated telluric acid (H₂TeO₄) by the distillation method. During the processing, a considerable amount of airborne iodine is generated in the cells and its major portion is discharged into the atmosphere after passing through two existing air cleaning systems. Some minor portion of the airborne iodine leaks directly

into the service and operation areas from the cells.

In order to evaluate the iodine concentration in the air and the amounts released, extensive radiation monitoring was performed with various types of air monitoring devices. The present paper describes the airborne iodine monitoring carried out during the iodine-131 test production prior to the routine operation and also the performance of an iodine sampler employed in this monitoring program.

RADIATION MONITORING SYSTEM AND MONITORING DEVICES

Stack monitoring system

A schematic diagram of the air cleaning and monitoring systems for the iodine discharged into the atmosphere is shown in Fig. 1. The airborne iodine generated in the cells, mostly in a distillation cell, is filtered by two air cleaning systems before it is discharged through

the stack (17 m high). The first is a cell-ventilation air cleaning system, consisting of a glass wool filter, 4 charcoal filters (670 × 1000 × 57, Barnebey Cheney—M. granular charcoal used) and an absolute filter installed on the top of each cell. The second is an exhaust air cleaning system, consisting of a glass wool filter and a charcoal filter (Model 7 FE filter manufactured by Barnebey Cheney Co., U.S.A.).

In the normal operation, the concentration of airborne iodine discharged increases very rapidly 30 min after start of the distillation, reaches a maximum in about one hour, and then decreases at the end. However, the iodine still continues to be refleased at low concentration. The variation in the concentration of released airborne iodine amounts to about 2 orders of magnitude during and following the processing.

To cope with such problems as highly filtered exhaust air and the varying concentration,

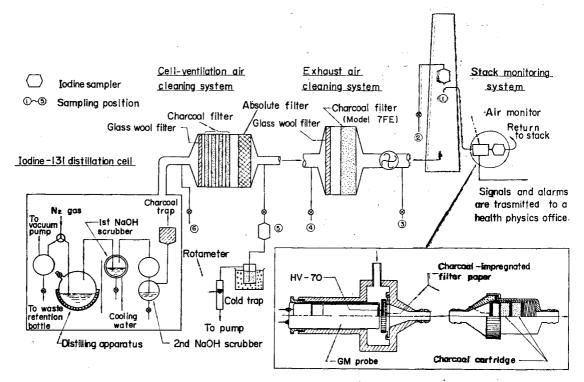


Fig. 1. Schematic diagram of airborne iodine monitoring and air cleaning systems at the radioisotope test production plant.

and to make it possible to evaluate the average concentration or the amount of airborne iodine released from the stack as accurately as possible, the iodine sampler, shown in Fig. 2 and in Table 1, was constructed in trial. The sampler was designed to ensure high overall collection efficiency under various sampling conditions, ease of handling and quick counting after the sampling, and low cost to be applicable to routine work.

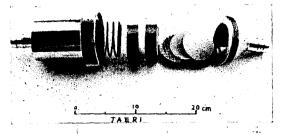


Fig. 2. Iodine sampler.

Table 1. Air Monitoring Devices

Monitoring devices	Characteristics	Sampling media
Iodine sampler	Air flow rate: 25~100 l./min External dimension: 70 mmØ, 215 mm or 230 mm long	HV-70 or HE-40 filter paper (50 mmØ, 9 mil thick) Charcoal filter paper* Charcoal cartridge† 2 or
Local air sampler‡	Air flow rate: 25 l./min	HV-70 or HE-40 filter paper Charcoal filter paper 2~
Stack air monitor	Air flow rate: 50~150 l./min Detector: end window type GM tube (window thickness 1~2 mg/cm²) Range: 6 ranges from 0.1 to 104 cps	HV-70 or HE-40 filter paper 1 Charcoal filter paper 1 2 (The iodine sampler is connected in series.)
Laboratory air monitor	Air flow rate: 25~50 1./min Detector: end window type GM tube or 1 3/4" × 2" NaI scintillator Range: 4 ranges from 150 to 1.5 × 10 ⁵ cpm	HV-70 or HE-40 filter paper Charcoal filter paper Charcoal cartridge
Iodine monitor§	Air flow rate: 50~100 l./min Detector: 1 3/4"Ø × 2" NaI scintillator Range: 6 ranges from 0.1 to 104 cps Single channel pulse height analyzer used	HV-70 or HE-40 filter paper 1 Charcoal filter paper 1 (The iodine sampler may be connected in series.)

^{*} Charcoal-impregnated filter paper, manufactured by Toyo Roshi (Toyo Filter Paper) Co. Ltd., Tokyo, Japan: 50 mm $\emptyset \times 2$ mm thick, average diameter of carbon particles $\sim 2\mu$.

[†] Charcoal cartridge, manufactured by Toyo Roshi Co. Ltd., Tokyo, Japan: 50 mmØ × 20 mm thick (effective thickness 18 mm), 30-mesh coconut shell charcoal used.

[‡] Installed to evaluate an air contamination in fixed points of working areas.

[§] A sample collected for a preselected interval, during which the sample is counted, is automatically moved and a succeeding filter paper is placed over a detector from a filter paper stocker.

The sampling positions in Fig. 1, except position No. 1, were used for measuring the collection efficiencies of the iodine sampler and the installed charcoal filters. These mositions were not used for routine monitoring. A continuous air monitor and the iodine sampler are installed at position No. 1 for routine stack monitoring. The primary praspose of installing the air monitor is to indicate the relative variations of iodine concentration with time by measuring continuously the activity accumulated in the charcoal filter parger and to actuate an alarm immediately if issuch an abrupt change of the concentration or san unusual release that exceeds a preset level should occur. It would be impossible to calificate the air monitor for iodine and determine the correct concentration because of the uncertainty of the collection efficiency of each sampling menedium for the various forms of airborne indime. For these reasons, the air monitor is employed

together with the iodine sampler to evaluate the correct average concentration or the amount released over a certain period. Characteristics of these monitoring devices are listed in Table 1.

Monitoring system for the working areas

The monitoring system for the working areas is shown in Fig. 3. The local air sampling system was designed for the purpose of examining the correlation between the iodine concentrations in the air and the actual thyroid burdens as determined by a whole body counter. Using these results necessary measures are taken to control the internal exposure by inhalation. The other purpose of this monitoring system is to find the origin of leaks of the airborne iodine and to improve the air-tightness of the cells and their equipment. In addition, a laboratory air monitor was operated to monitor continuously the general air in the service area and to actuate the alarm. The monitoring devices

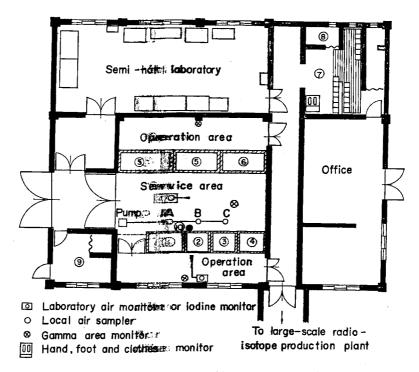


Fig. 3. Radiation monitoring systems: for working areas. 1-4. Iodine-131 processing cells.

1. Preparation. 2. Distillation. 3. The ispensation. 4. Storage. 5. Phosphor-32 processing cells.

6. Sulfur-35 processing cells. 7. Changing room. 8. Shower room. 9. Decontamination room.. 1991. Temporary liquid waste retention bottle.

employed are listed in Table 1 and shown in Fig. 4.

Counting of the samples

The counting method for the charcoal filter paper and charcoal cartridge is somewhat com-

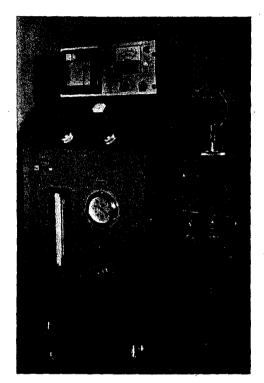


Fig. 4a. Laboratory air monitor.



Fig. 4b. Iodine monitor.

plicated. Two kinds of counting equipment were used: (1) a 400 channel pulse height analyzer with 5 in. \times 4 in. NaI(Tl) detector for the measurement of the collection efficiency of the iodine sampler; and (2) a scaler with $1\frac{3}{4}$ in. $\emptyset \times 2$ in. NaI(Tl) detector for the routine monitoring. The former was calibrated for the charcoal filter paper by counting the 0.364 MeV photopeak of standard iodine-131 sources. In this case, its overall counting efficiency is 17% and the minimum detectable amount is approximately $10^{-6}~\mu$ Ci for 4-min. counting.

The overall counting efficiency for the charcoal cartridge was determined to be 14%, assuming that the collected iodine was uniformly distributed throughout the cartridge. The efficiency selected was between 14% and 17%, according to the actual distribution of iodine in the cartridge.

The calibration of the scaler with $1\frac{3}{4}$ in. $\varnothing \times 2$ in. NaI(Tl) detector was also performed, using standard iodine-131 sources by suitably setting the discriminator. The minimum detectable amount is approximately $10^{-4} \, \mu \text{Ci}$ for 10-min counting.

MONITORING OF AIRBORNE IODINE AT THE VENTILATION DUCT AND STACK

In order to ensure the effectiveness of airborne iodine monitoring at the stack and in the working area, the field test for determining the collection efficiencies of the iodine sampler was carried out (using the airborne iodine produced during the processing) in parallel with the routine stack monitoring, because the results obtained by a laboratory test could not be directly applied to the field monitoring. The details of this test, conducted under the various sampling conditions, will be given in a later paper together with the counting methods for the sampling media. The present paper describes the measuring results relating to the stack monitoring.

The overall collection efficiency of the iodine sampler was determined, using one or two reference cold traps which were connected to the sampler in series and consisted of glass wool and activated granular charcoal (Barnebey Cheney-10, 130 g used) maintained at a temperature of less than -50° C with dry ice immersed in ethanol.

To examine the dependence of the collection efficiency of the iodine sampler on the degree of the filtration by the air cleaning systems, the samplings were carried out at several different positions in the air cleaning systems shown in Fig. 1. In passing through the air cleaning systems, the physical and/or chemical forms of iodine might be changed and the fraction of less reactive components in the discharged iodine would be considerably increased.

Sampling at the inlet of the cell-ventilation air cleaning system

Two types of samplings whose periods are different were adopted as follows: (1) short period sampling (less than 2 hr, in general 30 min), which was employed for the purpose of following the variations of concentration with

time and at the same time determining the collection efficiency in this sampling period; and (2) long period sampling (more than 8 hr) for examining the dependence of the collection efficiency on the concentration determined by the short period sampling, which is usually applied to the routine stack sampling. An example of results obtained is shown in Fig. 5.

In the short period sampling the overall collection efficiency was around 100%, while in the long period sampling it varied within the range of from 95% to 100%, depending on the concentrations and atmospheric conditions.

The distributions of the iodine collected in each component of the sampler during and following the processing are shown in Fig. 6 and Table 2 for the short period sampling and the long period sampling, respectively. As seen

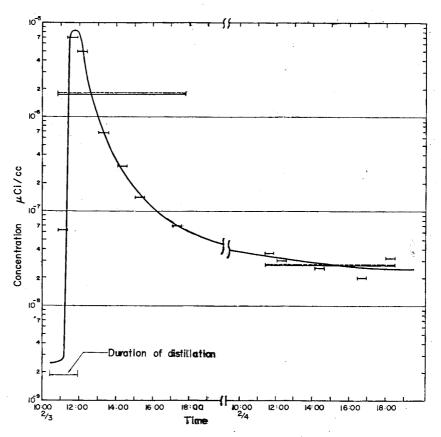


Fig. 5. Variation of concentration of airborne iodine at the inlet of the cell-ventilation air cleaning system during and following the processing.

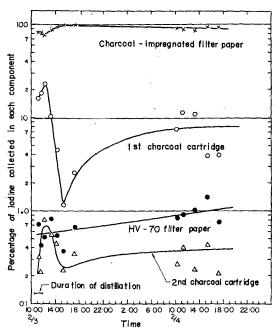


Fig. 6. Distribution of airborne iodine collected in each component of the sampler during a 30-min sampling at the inlet of cell-ventilation air cleaning system. Air flow rate is 45 l./min. Detectable quantity of iodine-131 is not collected in the cold trap.

in Fig. 6, more than 80% of the iodine is collected in a charcoal-impregnated filter paper. The percent of iodine collected in the charcoal filter paper increases up to approximately 100% and then gradually decreases with time after the end of the processing, although the released amount of iodine markedly falls off. The fraction of iodine in the first charcoal cartridge shows a remarkable fluctuation during the processing, in particular within about 3 hr of the start of the distillation. These results indicate that the chemical forms of airborne iodine, probably including I2, HI, HIO₃ and HIO₄, (8) may well be changed from the initial stage of generation to the end of distillation. In comparing Fig. 6 and Table 2, little difference is found between the distributions of iodine collected in each component of the sampler for the short period and for the long period, although in the long period sampling the fractions collected in the first and second

charcoal cartridge were of the same order of magnitude. The increase of the fraction in the second cartridge is caused by the fact that the iodine adsorbed in the activated charcoal during the abrupt change of concentration is desorbed in the course of the long period sampling under the low concentration.

Sampling at the exit of the cell-ventilation air cleaning system

In the sampling at the exit of the cell-ventilation air cleaning system, the distributions of the iodine collected in each component of the sampler is quite different from those collected at the inlet. As shown in Fig. 7 and Table 2, more than 60% of airborne iodine is collected in the first charcoal cartridge, whereas less than 20% of iodine is collected in the charcoal filter paper. This indicates that after passing through the cell-ventilation air cleaning system the fraction of the less reactive components in the residual airborne iodine increases relatively and the collection efficiency of the charcoal filter paper for the filtered iodine is only less than 20%. Despite such great differences between the iodine distributions among the components of the sampler obtained by the exit sampling and those obtained by the inlet sampling, overall collection efficiency of the sampler was still maintained at higher than 90% (94% on the average) in both the short period and the long period sampling.

Sampling at the exit of the exhaust air cleaning system and the stack

The exhaust air cleaning system originally consisted of a glass wool filter and an absolute filter and the discharged amount of the total processed iodine was rather high, ranging from 0.5% to 5%. In addition, the distributions of iodine among the components of the sampler and its overall collection efficiency at the stack or the exhaust air cleaning system, which ranged from 89% to 94% (91% on the average), were almost the same as those obtained at the exit of the cell-ventilation air cleaning system, indicating that the absolute filter was not efficient for removing the airborne iodine.

Based on these results, a charcoal filter (Model 7FE) was installed instead of the absolute filter in the exhaust air cleaning system, in order to

Table 2. Distribution of Iodine-131 Activity Collected in each Component of the Sompler during the Long Period Sampling at Various Positions of the Air Cleaning Systems.

Airflow Rate 25 I./mm.

					Activitie	s collected in ca	ch compo	Activities collected in each component of the sampler	pler					
Run No.	Sampling position*	Sampling	Total activity	HV-70 filter paper	Iter	Charcoal filter paper	filter	lst charcoal cartridge	lg 3	2nd charcoal cartridge	lag "	Cold trap	gi.	Overall collection
				io:	%	ğ	33.	Ō.	3 8	ğ	æ	₹	36	\$ 100 M
	7	The state of the	908	-01 × 02	10	60	-	2	2	## Cil	400	BE TH	-	32
	No. G	10:20 ~ 17:30	40 80 80 80	型 ×	ංක ාර			1.2 × 10-8	: O	1.2 × 10-1	° 0. ₽ 4 :	2.5 × 10	8.0	
		12/16	0.93	< 10-5	i	0.15	15.5	99.0	71	7.9×10^{-2}	8.5	4.6×10^{-2}	5.0	95
20	No. 5	$10:00 \sim 18:00$ 12/17 $10:00 \sim 18:00$	2.0×10^{-2}	< 10-5	I	2.6×10^{-2}	13	0.13	29	2.1×10^{-3}	Ξ	1.7×10^{-2}	9.0	91
		5/27	1.4 × 10 ⁻²	1 × 10-6	1	3.4×10^{-3}	24	6.8×10^{-3}	47	2.0×10^{-3}	14	2.2×10^{-3}	15	82
56	No. 3	$14:30 \sim 21:30$ 5/30 $10:45 \sim 17:45$	3.6×10^{-8}	< 10-5	I	2.5 × 10 ⁻⁴	2	1.8×10^{-8}	20	9.7×10^{-4}	27	5.8 × 10-4	16	25
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* Sampling positions of No. 6 and No. 5 are located at the inlet and exit of the cell-ventilation air cleaning system, respectively and a sampling position of No. 1 is located at the exit of 7 FE charcoal filter in the exhaust air cleaning system, as seen in Fig. 1.

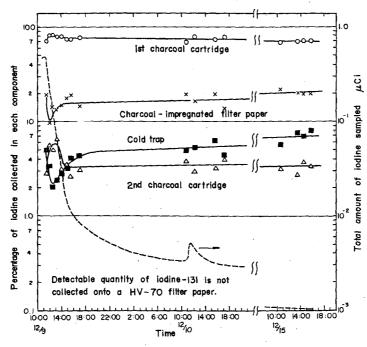


Fig. 7. Distribution of airborne iodine collected in each component of the sampler during a 30-min sampling and variation of concentration at the exit of cell-ventilation air cleaning system. Air flow rate is 45 l./min.

make the filtration much more efficient and to assure the environmental radiological safety. As a result of this change, the percent of the discharged iodine was reduced to less than 0.03%, as seen in Fig. 8. On the other hand, the overall collection efficiency of the sampler decreased down to a minimum of 70% in the long period sampling, showing that the relative amounts of less reactive components in the airborne iodine discharged increased considerably. However, there was not much difference between the distributions of the iodine among the components of the sampler in the samples taken at the exit of the cell-ventilation air cleaning system, and those taken at the exit of the exhaust air cleaning system.

The collection efficiencies of the sampler obtained in each stage of the filtration are summarized in Fig. 9. The overall collection efficiencies ranges from 70% to 100%, depending on the various sampling conditions; in particular, on whether the samples were taken upstream

or downstream of the air cleaning systems. Relative humidity and temperature during the period of this series of samplings were between 60% and 80% and about 20°C, respectively. Significant differences were not found between the overall collection efficiencies obtained at the exit of the exhaust air cleaning system and at the stack.

At every stage of the filtration, iodine was not collected appreciably onto the HV-70 filter paper, so that it is concluded that the airborne iodine generated during processing is in the vapour phase and little, if any, iodine is associated with particles.

AIR CONTAMINATION OF WORKING AREAS

Service area

In the early stage of the iodine test production, air contamination by the leaked iodine occurred in the service area, despite a negative pressure differential of about 40 mm H₂O in each cell. The variations of concentrations

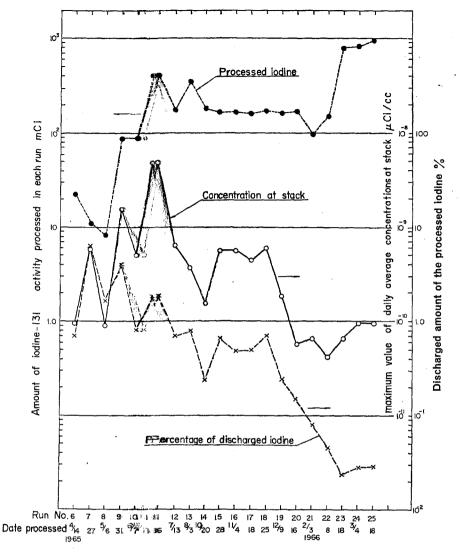
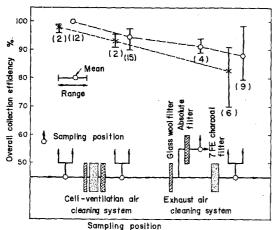


Fig. 8. Percentage of discharged indine and maximum value of daily average concentrations at stack in and at its concentration to iodine processed in each run.

obtained at three sampling points. Inchind the cells shown in Fig. 3 are illustrated in Fig. 10. The mode of variations is similar test that at the stack, but in Fig. 10 the first and income peak concentration appears about 10 him and 1 day, respectively, later than those at the estack and the magnitude of the concentration is different at each sampling point. These indicates that the airborne iodine leaks from the distillation weell nearly

proportionally to the amount generated, and gradually accumulates in the service area.

On the basis of such results, some attempts were made to prevent the leakage of the airborne iodine from the cells as follows: (1) the places where leakage was likely to occur, for example, joints of the cell air ventilation duct, openings of the cell attachment, etc., were sealed; and (2) the cell-ventilation air cleaning system



O Sampling of 30-min to 2-hr duration

Sampling of 7-hr to 8-hr duration

Air flow rate: 25t/min~50t/min

Fig. 9. Summary of collection efficiencies of the sampler at each stage of the filtration by air cleaning systems. Figures in brackets indicate number of measured samples.

was improved to maintain a negative pressure differential larger than 100 mm H₂O. In consequence, the air contamination of the service area decreased by about one order of magnitude or more, and at present the airborne iodine concentration is always kept below 5×10^{-11} μ Ci/cc during normal operation. An example of the air contamination after improving the air-tightness is shown in Fig. 10b. The mode of concentration variation in Fig. 10b is different from that in Fig. 10a and no correlation is present between the concentration variation at the stack and that in the service area. The air contamination in Fig. 10b might have been caused by the leakage of airborne iodine from a liquid waste bottle which was placed behind the cell access door to retain temporarily the irradiated telluric acid solution after the end of the processing.

Operation area

Airborne iodine monitoring is continuously performed in the operation area whenever operations are carried out. The results obtained in the operation area during the same run as in Fig. 10a are shown in Fig. 11. The concentration in the operation area reaches a maximum value

of approximately $10^{-10}~\mu\text{Ci/cc}$ about 2 days after start of the processing and is about one tenth of the concentration in the service area. This air contamination mainly occurs by the diffusion of airborne iodine from the cells into the service area. After various measures for preventing the leakage were taken, the air contamination decreased to the minimum detectable limit (8 \times $10^{-12}~\mu\text{Ci/cc}$) during normal operation, except that slightly higher concentrations than the minimum detectable limit were found under incidental conditions for a day or so, two or three days after the distillation.

INTERNAL EXPOSURE OF THE OPERATING PERSONNEL

In order to examine the effectiveness of the air monitoring system in evaluating the inhalation hazard, the thyroid burden of the operating personnel was measured each time after the end of the processing using a whole body counter. The results measured for the period April 1965 to March 1966 are summarized in Fig. 12. The total number of individuals measured over the period was 69, in 28 of which thyroid uptakes were detected. The maximum of the thyroid burdens summed for each individual over the period was 4.3 m μ Ci, which gives an integrated thyroid dose of 28 mrem.

The amount of actual uptake in the thyroids, as determined by the whole body counter, is at most about 200 times and on the average about 20 times as large as the maximum probable uptakes calculated using both the daily average concentrations in the working areas and the working time. No significant correlation was found between the actual uptake and the calculated uptake, as shown in Fig. 13. This could be caused by the fact that such marked variations of concentration might exist in both time and space that the exposure cannot definitely be assessed with the daily average concentrations determined by the samplers at fixed points. In addition, human exposure due to the inhalation could be highly influenced by personal factors such as working habits, respiratory rate, etc.

To permit more precise assessment of the air contamination to which individuals are exposed, personal air samplers (using a charcoal filter paper and a charcoal cartridge at a flow rate

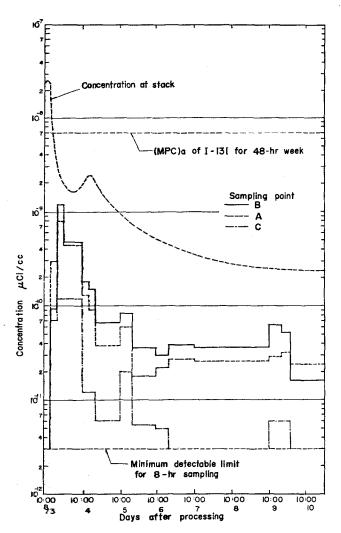


Fig. 10a. Variations of concentrations in the service area during and following the processing before improving an air-tightness of the cells. Air flow rate is 25 l./min.

of 10 l./min) are now employed. Preliminary results indicate a fairly good agreement between the activity on the charcoal paper, in which the collection efficiency is taken into consideration, and the actual thyroid burdens.

Further detailed investigations are planned to improve the existing air sampling system and to obtain reasonable correlations between the results of the samplers at fixed points, the results of the personal air sampler carried by operators, and the actual thyroid burdens.

CONCLUSIONS

The field test of the newly developed sampler conducted at the isotope test production plant shows that it can be satisfactorily employed for the purposes of routine monitoring within the error of $\pm 20\%$, and it is now widely applied to the airborne activity monitoring at JAERI.

Internal exposures of the operating personnel due to the inhalation of iodine-131 occurred during the test production. Although these exposures were not significant from the view-

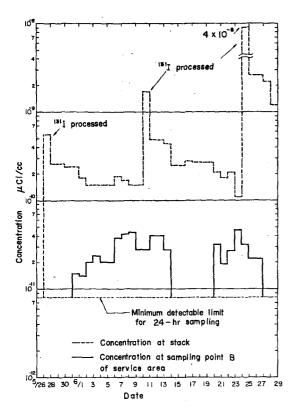


Fig. 10b. Variation of concentration in the service area after improving an air-tightness of the cells.

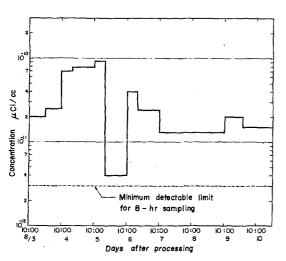


Fig. 11. Variation of concentration in the operation area in the same run as in Fig. 10a.

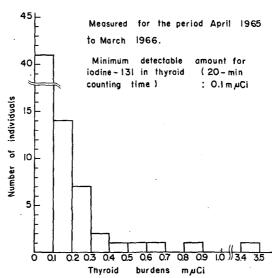
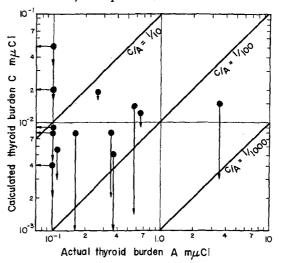


Fig. 12. Summary of thyroid burdens of operating personnel measured by a whole body counter. The total number of individuals measured over the period was 69, in 28 of which thyroid uptake was detected.



 Maximum probable thyroid burden obtained by calculation.

Fig. 13. Comparison of thyroid burdens measured by a whole body counter and those calculated using both the daily average concentration and the working time. Vertical arrow shows the extent of the uncertainty of the thyroid burden, which results from the concentrations of less than minimum detectable amounts in the working area. Horizontal arrow shows the thyroid burden of less than the minimum detectable amount of the whole body counter.

point of exposure accontrol, many efforts were made to reduce both: the airborne iodine generation and its leakagge, which resulted in the remarkable reduction of the air contamination in the working anexas. The amounts of iodine discharged through tithe stack were also reduced by about 2 orders for magnitude by installing a charcoal filter init; the exhaust air cleaning system.

The monitoring system for the radioisotope test production planest will be applied in the more improved form to these new large-scale radioisotope production planest where the production of iodine-131 is planeated at a cycle of 10 Ci/week.

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