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AIRBORNE RADIONUCLIDES OF CONCERN AND THEIR MEASUREMENT IN MONITORING A COMPREHENSIVE TEST BAN TREATY

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INTRODUCTION

The U. S. Department of Energy (DOE) is conducting radioanalytical developmental programs with the goal of providing near-real-time analysis technology for airborne signature radionuclides which are indicative of a nuclear weapons test in any of the earth's environments. If a test were conducted in the atmosphere or above the atmosphere, then the full spectrum of fission and activation products, together with residues from the device would be dispersed in the atmosphere. However, if a nuclear test were conducted underground or under water, the emission could range from a major to a very minor vent, and the material released would likely consist mainly of noble gas radionuclides and the radioiodines. Since many of the noble gases decay to form particulate radionuclides, these may serve as the more sensitive signatures. For example, Ba-140 is a daughter of Xe-140 (13.6 s), and Cs-137 is a daughter of Xe-137 (3.82 min). Both of these have been observed in large amounts relative to other fission products in dynamic venting of U.S. underground nuclear detonations.[1]

Large amounts of radionuclides are produced from even a comparatively small nuclear detonation. For example, a 10-KT fission device will produce approximately a megacurie of Ba-140 and of several other radionuclides with half-lives of days to weeks. If such a device were detonated in the atmosphere at mid-latitude, it would easily be observable at downwind monitoring sites during its first and subsequent circumnavigations of the earth.

Efficient and practical methods for the near-real-time analysis of both particulate and gaseous radionuclides are important to an effective monitoring and attribution program in support of a Comprehensive Test Ban Treaty (CTBT); methods for this purpose are being pursued.

DISCUSSION

During the period 1961 through 1989, concentrations of approximately 30 airborne radionuclides were monitored continuously at our sampling station in Richland, Washington[2]. Examples of the variations in concentrations of Cs-137 (30.17 yr) and Ba-140 (12.75 d) over this time period are shown in Figure 1. While Cs-137 shows gradual changes in its atmospheric concentrations, the relatively short-lived Ba-140 shows an abrupt increase following each atmospheric nuclear detonation. The key to high sensitivity detection of airborne radionuclides is the collection of debris from large volumes of air coupled with rapid analysis by high-sensitivity gamma-ray spectrometry.

Table 1 summarizes the nuclear detonations conducted by the Peoples Republic of China during the period October 16, 1964 through November 17, 1978. The observed airborne Ba-140 at Richland, Washington, from the atmospheric ~20 KT detonation of October 16, 1964; the >20 KT detonation of May 14, 1965; and the 300 KT detonation of December 18, 1966 are presented in Figures 2, 3, and 4, respectively. These data illustrate the ease with which debris from atmospheric nuclear detonations can be observed if large volume sampling is conducted, and if high-sensitivity measurements are performed.

Sampling of the airborne particulate radionuclides was conducted at approximately 12 m^3/min, and their measurements were made with either large NaI(Tl) detectors operating in coincidence with anticoincidence shielding[3] or with

high-resolution germanium detectors. These detection methods, together with improvements in sample collection, data processing, and more rapid analysis to avoid significant decay, provide a sound approach for monitoring of relatively short-lived airborne radionuclides indicative of a nuclear detonation.

In monitoring for possible violation of the Comprehensive Test Ban Treaty (CTBT), it is important to measure those radionuclides which are most uniquely indicative of a recent nuclear test. If monitoring stations were set up at points of interest over the earth's surface, then they should be capable of detecting and measuring those radionuclides which would enter the atmosphere from an atmospheric detonation or a vented underground or underwater detonation. The radionuclides which are most abundant at various times after a nuclear detonation are shown in Table 2 at 1 day, 3 days, 10 days, and 30 days post-detonation. For an atmospheric detonation, the easiest way to monitor for these radionuclides is large-volume air sampling and of particulate debris for gamma-ray spectrometric analysis. However, if the detonation occurred underground or underwater, it may be that only the noble gas radionuclides, and perhaps the radioiodines, would be vented.

As indicated in Table 2, there are several particulate radionuclides which could be observed just one day after a detonation and, in addition, the two xenon radionuclides, Xe-135 (9.10 h) and Xe-133 (5.243 d), would be among the 12 most abundant radionuclides. Even after three days, these two xenon radionuclides are still among the 12 most abundant radionuclides, and their ratios would be indicative of the time since the detonation occurred. For longer periods which would result in major decay of the 9.10-hour Xe-135, it would become increasingly difficult to use the xenon gas radionuclide pair for dating the time of the

detonation, and after five days the activity of the shorter-lived radionuclide Xe-135 would be so small that it would probably not be detectable.

In the 10- and 30-day time frame, as indicated in Table 2, the longer-lived radionuclides become the preferred signatures for detection. The use of several radionuclides as indicators of a nuclear detonation is important since there are many other possible sources of airborne radioactivity, including reprocessing of nuclear fuels, fixation of nuclear waste, possible emissions from nuclear reactors, and the use of radionuclides for medicinal and research purposes.

The DOE position in developing technology for monitoring of particulate and gaseous radionuclides has been that near-real-time analyses at the monitoring sites is preferred. The following discussion considers the measurement of both the particulate and particulate-associated radionuclides, as well as the measurement of noble gas radionuclides as indicators of nuclear detonations.

PARTICULATE RADIONUCLIDE MEASUREMENTS

As shown in Figures 2, 3, and 4, radioactive debris from an atmospheric nuclear detonation moves at a relatively high velocity from the point of detonation to downwind locations, presumably due to its transport by air masses in the upper troposphere. For example, the first Peoples Republic of China's nuclear test (see Table 2), which was conducted on October 16, 1964, carried debris to our monitoring site at Richland, Washington, which was approximately 10,000 km away, in just three days, with peak concentrations arriving approximately seven days after the detonation. The main air mass containing this activity apparently continued on a rapid trajectory and was again detected during second and third circumnavigations of the globe after consecutive two-week periods.

A second atmospheric nuclear detonation carried out by the Peoples Republic of China on May 14, 1965 (see Table 1), resulted in debris reaching our monitoring station in approximately five days, with maximum concentrations being observed there after seven days (see Figure 2).

A much larger detonation (approximately 300 KT) which was carried out on December 28, 1966, resulted in radioactive debris reaching our monitoring site approximately four days after the detonation, with maximum concentrations being present after five days. Thus, debris from atmospheric detonations does travel at a very high rate and can be expected to circumnavigate the earth in periods ranging from about 10 to 15 days.

The following factors influence the sensitivity with which airborne radionuclides can be measured:

- Large-volume air sampling
  A 100-fold increase in sample size can provide up to a 100-fold improvement in detection sensitivity;
- Low-background counting
  A 100-fold decrease in background may permit up to 10-fold improvement in detection sensitivity;
- High-efficiency counting
  A 10-fold increase in counting efficiency can provide up to a 3-fold improvement in detection sensitivity;
- Separation from interferences
  A 100-fold reduction in interfering radionuclides (radon and daughters in the case of Xe-133/Xe-135 measurements) could provide up to 10-fold improvement in detection sensitivity.

In addition to these considerations is the factor of timeliness in carrying out sample collection and analysis. As a general rule, sample collection time, as well as the measurement time, should not exceed the half-life of the shortest-lived radionuclide of interest. Thus, if it were important to measure a radio-

nuclide with a half-life of eight hours, very little would be gained by extending the collection time or the measurement time beyond eight-hour periods.

Maximizing the sensitivity for near-real-time measurements of the airborne particulate radionuclides can be accomplished by using a USDOE design with the following features:

- Air is filtered through a low-pressure drop electrostatic filter at a rate of 17 m^3 and a face velocity of about 1.2 m/sec for eight hours;
- The filter paper is folded and sealed in a thin plastic packet to provide a square of about 12 x 12 cm^2;
- The filter is immediately counted using a large-area intrinsic germanium detector for 8 hours, collecting data in a "time-stamped mode" to permit optimum sensitivity in discrimination against radon daughters;
- Complete spectral data and analytical results are immediately transmitted to the National Data Center (NDC) and on to the International Data Center (IDC).

The eight-hour collection plus an eight-hour measurement period are considered a minimum time period for normal monitoring. However, these time periods could be modified at any time to longer or shorter periods by use of a two-way hard-line or INMARSAT communication linkage. A schematic drawing of the system which is currently being tested for the near-real-time measurements is shown in Figure 5.

NOBLE GAS RADIONUCLIDE MONITORING

As indicated in Table 2, the Xe-133 (5.243 d) and Xe-135 (9.10 hr) are the most abundant noble gas radionuclides and appear to be by far the most useful indicators of vented material from a subsurface or underwater nuclear detonation. The very short half-life of Xe-135 requires on-site and near-real-time analysis if it is to be measured. One day after a detonation, the Xe-135 to Xe-133 ratio

is about 10:1. However, four days post-detonation this ratio is only about 1:20. Thus, the useful time period for measurement of xenon radioisotope ratios and determining detonation time is about four, or possibly five, days post-detonation.

The decay properties of Xe-133 and Xe-135 are summarized in Table 3. Both radionuclides emit low-energy gamma rays which can be readily measured by high-resolution gamma-ray spectrometry; however, the high internal conversion of the Xe-133 81-keV gamma-ray limits the sensitivity for its measurement. The concentration scheme for collection of xenon gas (and associated radioxenon) from the atmosphere is illustrated in Figure 6. Basically it involves processing filtered air through a molecular sieve aluminum oxide bed for removal of water, CO_2, and radon gas followed by cryogenically collecting the xenon on a cooled sorption bed. After a specified collection period of a few hours, the xenon is thermally desorbed and recaptured on a small sorption bed for gamma-ray spectrometric analysis of the Xe-133 and Xe-135.

This procedure, which is being developed and tested by the USDOE, allows the near-real-time measurement of Xe-133 and Xe-135 during consecutive six-hour intervals at concentrations as low as 1 mBq/m^3. We have observed concentrations of Xe-133 at these levels and higher in the northeast sector of the United States, and therefore this sensitivity seems adequate for measuring back- ground levels and any changes in these levels.

SUMMARY

The required technologies for the near-real-time high sensitivity detection and analysis of airborne radionuclides which could be indicative of atmospheric, underground, or underwater nuclear tests are being developed and demonstrated.

The key to high sensitivity detection and measurement is large-volume sampling combined with high-sensitivity and high-resolution gamma-ray spectrometric analysis.

Since many of the most characteristic radionuclides associated with nuclear weapons testing have relatively short half-lives, then a large sensitivity improvement is achieved by near-real-time analysis at the collection point. Technologies are being adapted for transmission of both the basic gamma-ray spectra and analysis of these data to the NDC and on to the IDC. The combined automatic and near-real-time separation, measurement, and transmission of airborne radionuclide data are currently being investigated and the selection of appropriate techniques for these activities is being made.

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2. Perkins, R.W., D.E. Robertson, C.W. Thomas, J.A. Young, 1989. "Measurement of Accident Versus Nuclear Test Debris." In Proceedings of IAEA Symposium on Environmental Contamination Following a Major Nuclear Accident, October 16-20, 1989. IAEA-SM-306/125, International Atomic Energy Agency, Vienna, Austria.

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TABLE I

PEOPLES REPUBLIC OF CHINA NUCLEAR DETONATIONS

Date of Detonation	Type	Yield	Location
Oct. 16, 1964	Tower	~20 KT	Lop Nor
May 14, 1965	Air Drop	>20 KT	
May 09, 1966	Air Drop	200 to 500 KT	
Oct. 27, 1966	Missile, not HH§*	<20 KT	
Dec. 28, 1966	Tower	300 KT	
June 17, 1967	Air Drop	3 MT	
Dec. 24, 1967	Air Drop	15 to 25 KT	
Dec. 27, 1968	Air Drop	3 MT	
Sept. 22, 1969	Underground	~25 KT	
Sept. 29, 1969	Air Drop	3 MT	
Oct. 14, 1970	Air Drop	3 MT	
Nov. 18, 1971	Tower	~20 KT	
Jan. 07, 1972	Atmospheric	<20 KT	
Mar. 18, 1972	Atmospheric	20 to 200 KT	
June 27, 1973	Missile?	1 to 3 MT ?	
June 17, 1974	Atmospheric	1 MT	
Oct. 27, 1975	Underground	20 KT	
Jan. 23, 1976	Atmospheric	<20 KT	
Sept. 25, 1976	Atmospheric	200 KT	
Oct. 27, 1976	Underground	200 KT	
Nov. 17, 1976	Atmospheric	4 MT	

* HH§ stands for launching by missile to high altitude.

TABLE 2

PRINCIPAL REMAINING FISSION PRODUCTS
AFTER 1-DAY, 3-DAY, 10-DAY AND 30-DAY DECAY PERIODS
(Gamma-ray Emitters)

1-Day Decay

Radionuclide	Half-Life
Xe-135	9.089 hr
I-133	20.8 hr
Zr-97	16.9 hr
Rh-105	1.473 d
Pd-109	13.46 hr
Ce-134	1.375 d
Mo-99	2.75 d
I-135	6.611 hr
Te-132	3.258 d
Sr-91	9.5 hr
Ru-105	4.439 hr
Xe-133	5.245 d

3-Day Decay

Radionuclide	Half-Life
Mo-99	2.75 d
Rh-105	1.473 d
Xe-133	5.245 d
Te-132	3.258 d
Ce-143	1.375 d
I-133	20.8 hr
Zr-97	16.9 hr
I-131	8.041 d
Ba-140	12.79 d
Xe-135	9.089 hr
Pd-109	13.46 hr
Ru-103	39.28 d
Ce-141	32.51 d

10-Day Decay

Radionuclide	Half-Life
Xe-133	5.245 d
Ba-140	12.79 d
I-131	8.041 d
Mo-99	2.75 d
Te-132	3.258 d
Ru-103	39.28 d
Ce-141	32.51 d
Nd-147	11.06 d
Zr-95	63.98 d
Rh-105	1.473 d
Sb-127	3.85 d
Ce-143	1.375 d

30-Day Decay

Radionuclide	Half-Life
Ru-103	39.28 d
Ce-141	32.51 d
Ba-140	12.79 d
Zr-95	63.98 d
I-131	8.041 d
Xe-133	5.245 d
Nd-147	11.06 d
Ce-144	284.3 d
Ru-106	1.008 yr
Te-129m	33.6 d
Te-132	3.258 d
Eu-156	15.19 d

TABLE 3

IMPORTANT DECAY PROPERTIES OF Xe-133 AND Xe-135

Xe-133

BETA ENERGIES, keV (ABUNDANCES, %)*	COINCIDENCE GAMMA OR (CS) X-RAY ENERGIES, keV (ABUNDANCES, %)*
346 (99.3) beta	81 (36.5) gamma
	30.62 (13.6) $K_{\alpha 2}$
	30.97 (25.3) $K_{\alpha 1}$
	35.0 (9.1) $K_{\beta 1}$
45 (53.3) CE[a]	

Xe-135

920 (96.1) beta	249.8 (90)

* Percent of disintegrations which involve emission of the beta particle, conversion electron or photon

a Conversion electron

FIGURE 1

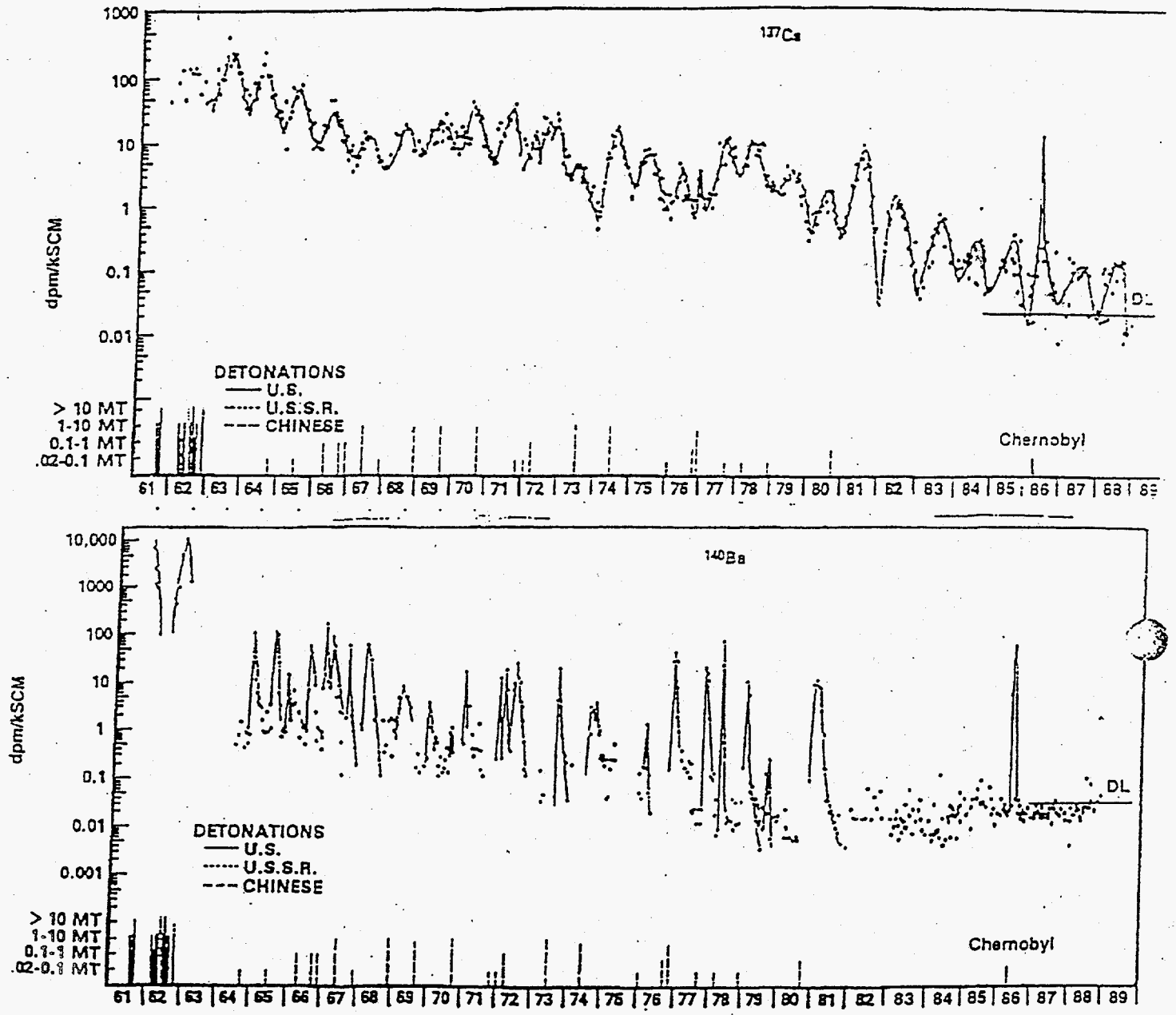
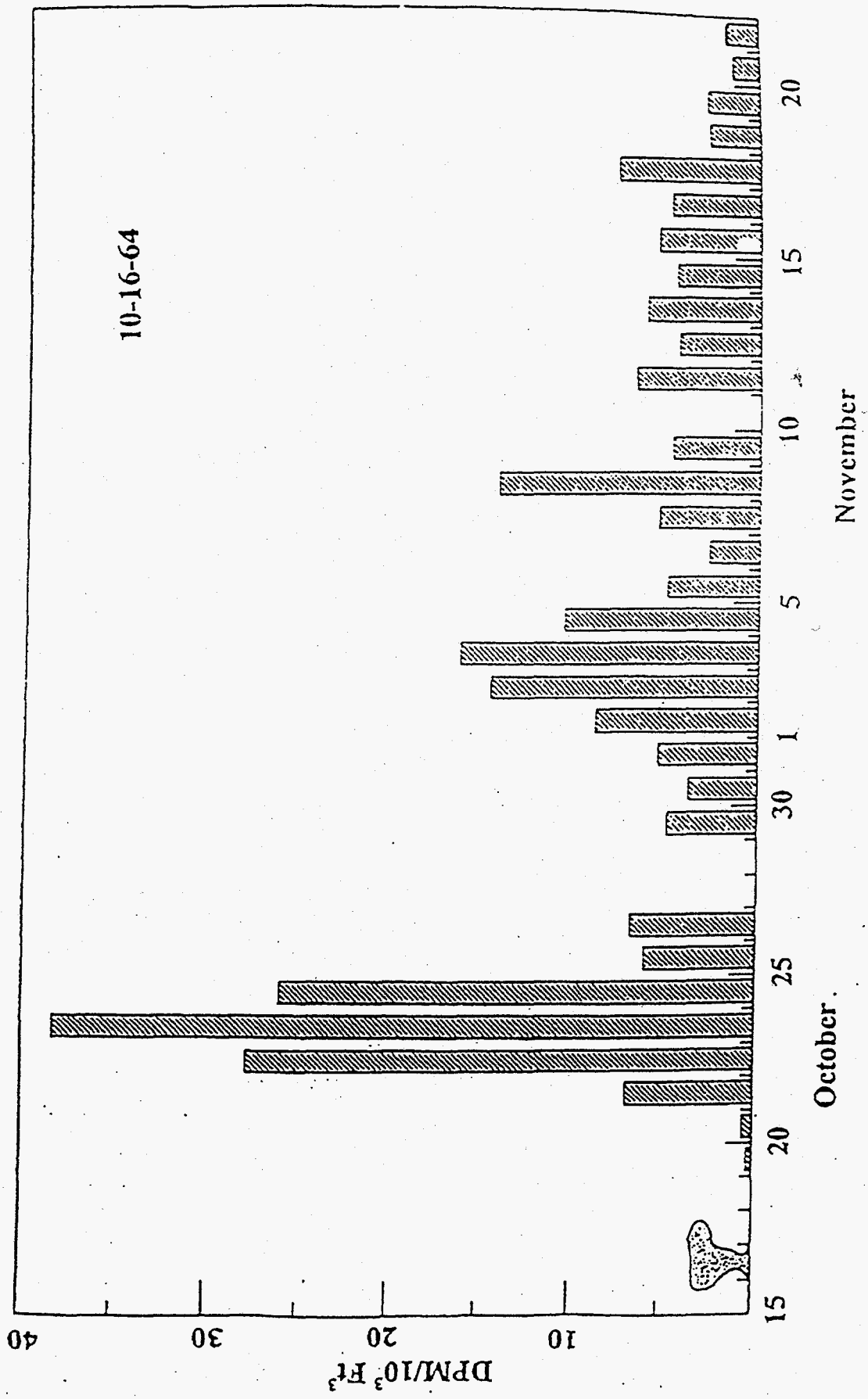

1000
100
10
1
0.1
0.01
dpm/kSCM
137Cs
DL
DETONATIONS
U.S.
U.S.S.R.
CHINESE
> 10 MT
1-10 MT
0.1-1 MT
.02-0.1 MT
Chernobyl
61 62 63 64 65 66 67 68 69 70 71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89
10,000
1000
100
10
1
0.1
0.01
0.001
dpm/kSCM
140Ba
DL
DETONATIONS
U.S.
U.S.S.R.
CHINESE
> 10 MT
1-10 MT
0.1-1 MT
.02-0.1 MT
Chernobyl
61 62 63 64 65 66 67 68 69 70 71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89

FIGURE 2
BARIUM-140 CONCENTRATION IN AIR SAMPLES
FROM CHINESE DETONATION



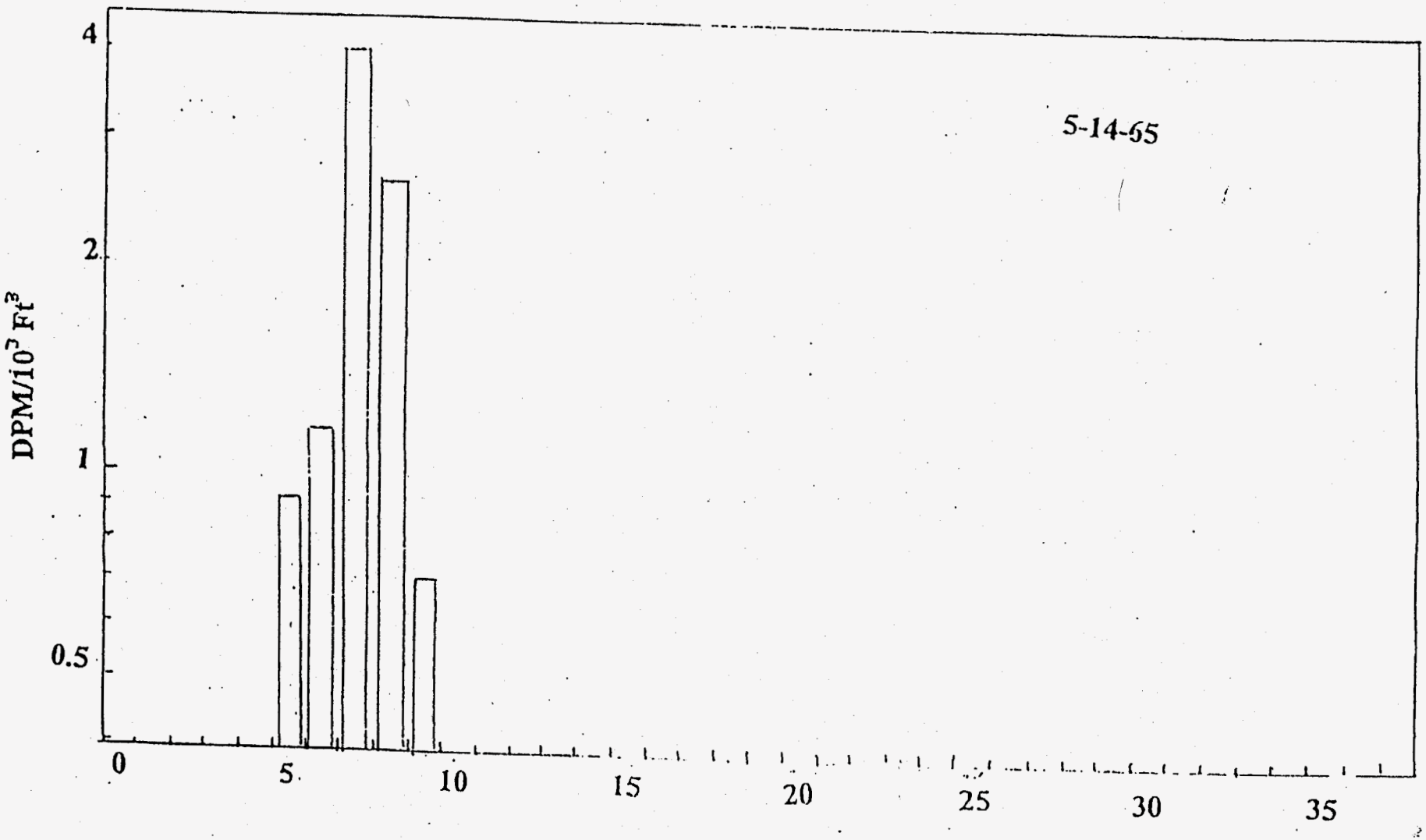


Figure 3. Ba^{140} Concentration in Air Samples from Chinese Detonation, Time After Detonation (days)

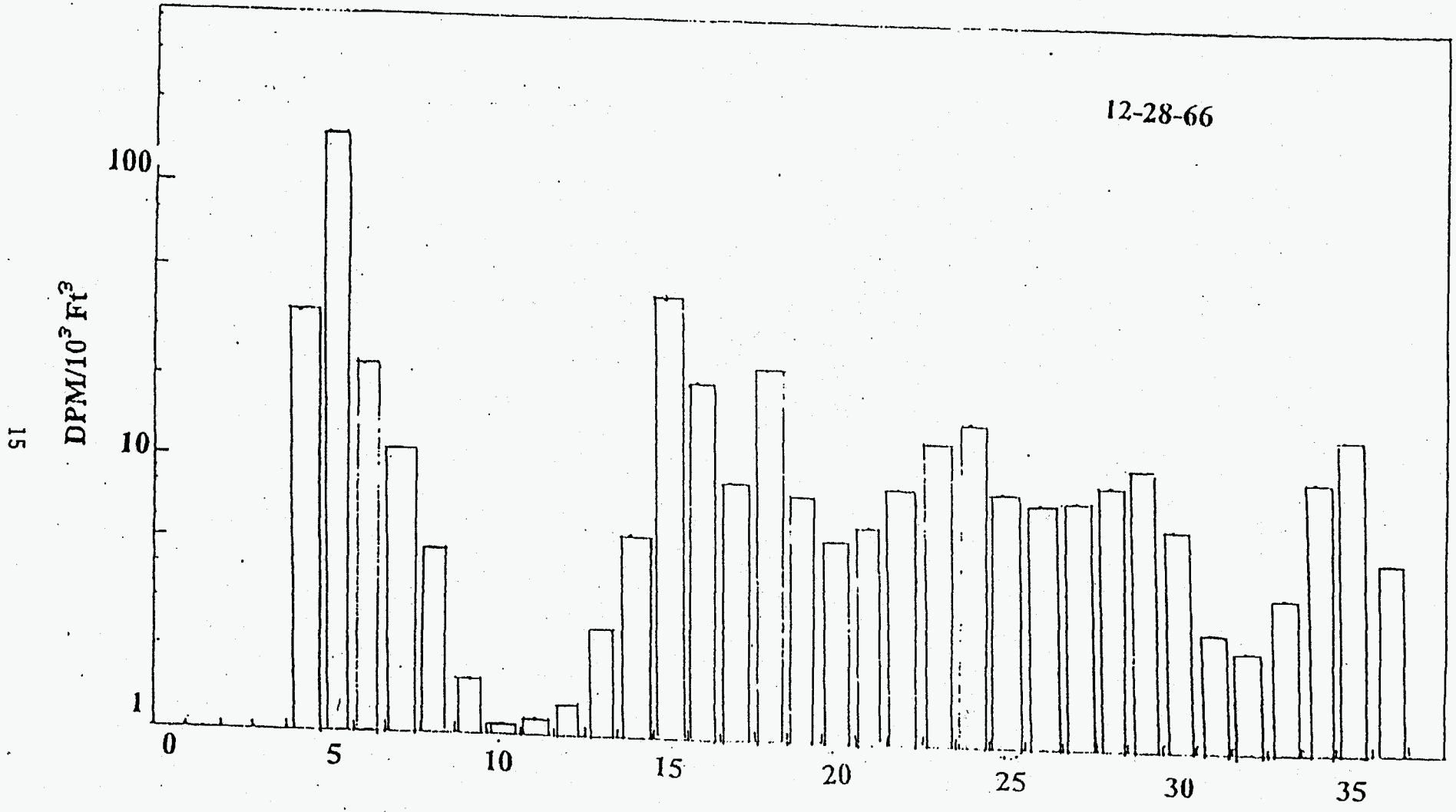


Figure 4. Ba^{140} Concentration in Air Samples from Chinese Detonation, Time After Detonation (days)

FIGURE 5

AIR FILTRATION GAMMA-RAY SPECTROMETRIC SYSTEM.
(Air flow 17 m^3/min; collection time 8 to 24 hours;
counting time 8 to 24 hours; data transmission immediate)

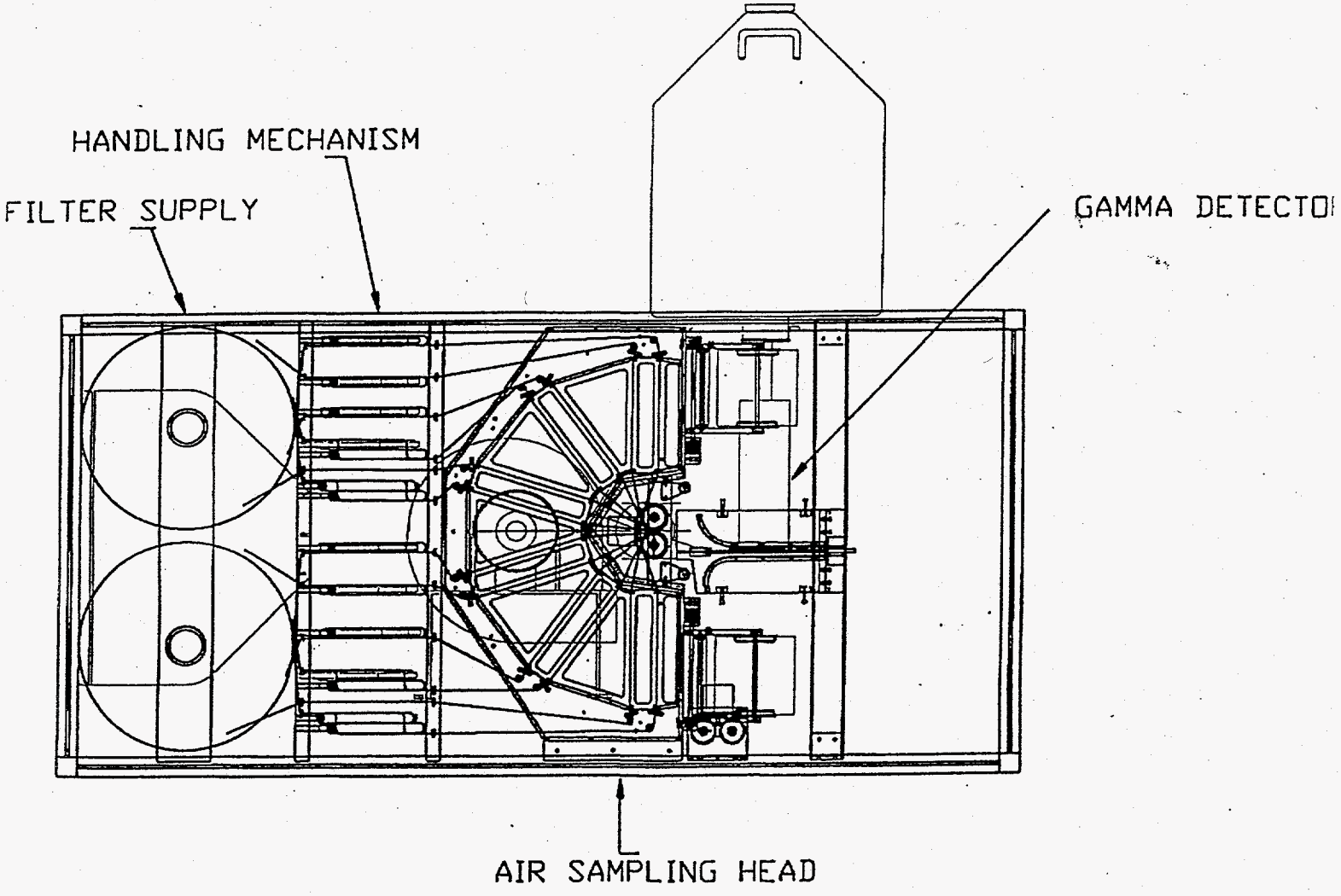


FIGURE 6
ATMOSPHERIC XENON COLLECTOR SYSTEM
(about 1.5 cc/hr)