# Radiation Physics Note No. 87
# Response of 1 1/2" by 1" NaI(Tl) with Respect to a Release Criterion

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## Introduction

We describe some calculations based on observations made with the 1.5 inch X 1 inch NaI(Tl) detector currently used at Fermilab to determine if items removed from accelerator enclosures are radioactive according to a criterion stated in the *Fermilab Radiation Guide*. That criterion is summarized as follows: Items are to be surveyed at contact with a Thyac™ or Bicron™ instrument which is based upon such a detector. If the highest reading exceeds 2000 cpm above background at contact on the X10 scale (10,000 cpm full scale), then the object is considered to be radioactive. This note explores the implications of this particular criterion.*

## Detector Sensitivity

Elwyn and Freeman[1] have studied the detection of low-levels of radioactivity using survey instruments equipped with these particular crystals. This work is a sequel to earlier studies by Cossairt[2] for a somewhat smaller crystal [1 inch X 1 inch NaI(Tl)] which had been used in the past. In both of these studies, a correlation was made between the count rates measured by the NaI(Tl) instruments and exposure rates measured with a properly calibrated Geiger-Müller tube-based survey meter in radiation fields due to both calibrated radioactive sources and accelerator-activated components. The sources chosen spanned the energy range of the $\gamma$-emitters expected in accelerator-activated materials. The results of the two studies were largely consistent; variations were mostly attributable to the expected higher detection efficiency of the larger crystal. For the larger crystal, Elwyn and Freeman found the following relationships:

| Source | $E\gamma$(Mev) | Exposure Rate (mR/h) @ 2000 cpm [NaI(Tl)] |
|---|---|---|
| $^{137}$Cs | 0.661 | 0.006 |
| $^{22}$Na | 0.511, 1.27 | 0.011 |
| $^{60}$Co | 1.17, 1.33 | 0.012 |
| | **Average** | **0.010 ± 0.003** |

* Natural background at Fermilab as measured with the Bicron and Thyac detectors is typically 1500-2500 cpm. Thus, the selected criterion is approximately equal to the background counting rate.

Typical $\gamma$-ray energies encountered with activated materials produced by particle accelerators are slightly less than 1 MeV.

The average response shown here can be checked for consistency by use of calculations of detector efficiency. Based on standard methods, we have calculated the absolute detection efficiency for a 1.5 inch X 1 inch crystal as a function of distance h from a point source emitting both 1 and 0.5 MeV photons. The results are shown in Figure 1. At 30 cm from such a source, the absolute efficiency at 1 MeV is 4 X $10^{-4}$. From Patterson and Thomas[3], 0.01 mR/h corresponds to a photon flux density of 5.61 $cm^{-2}s^{-1}$ for 1 MeV photons. Thus, the total emission from a point source which produces 0.01 mR/h at 30 cm distance is,

$$5.61 \times [4\pi(30)^2] \times 60 = 3.81 \times 10^6 \text{ photons/minute.}$$

If the detection efficiency is as given above, then the instrument should register the following count rate:

$$[3.81 \times 10^6] \times [4 \times 10^{-4}] = 1530 \text{ cpm.}$$

This result is within 24 % of the 2000 cpm empirical determination, and reasonably consistent with it in view of the uncertainties in all quantities.* In using this level as a release criterion, it is important to understand what sensitivity it actually corresponds to in practical circumstances, and this forms the basis for the remainder of the note.

## Benchmark Comparison with Some Regulatory Limits

DOE Order 5400.5 (as issued in February 8, 1990) states the public dose limits to residual radioactive materials to be 100 mrem/y above natural background. This limit explicitly applies to the dose equivalent which individuals could receive from materials released from DOE facilities. Since the 2000 cpm criterion corresponds to approximately 0.010 mrem/h, proximity of such materials to an individual could result in a maximum dose equivalent of 88 mrem in a year, in the unlikely event that the person was entirely surrounded by such material. Under any reasonable

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*The relative error due to counting statistics associated with the 2000 cpm counting rate based on a 1 second response time is

$$\frac{\sqrt{(S+2B)/60}}{(T-B)/60},$$

where S is the sample (alone) counting rate in cpm, B is the background rate, and T is the total rate. For S=B=2000 cpm, the relative error is ±30%.

circumstances, however, it is highly improbable that anyone would spend more than 10 per cent of the time in such a field. Therefore, the maximum dose equivalent to an individual exposed to such materials or equipment in a year is unlikely to exceed 9 mrem. Thus the criterion adequately protects against excessive exposure to the general public below dose level limits consistent with DOE Order 5400.5.

In the United States, possession of radioactive materials is, in general, regulated by the U.S. Nuclear Regulatory Commission. This commission has established quantities and specific activities exempt from its licensing procedures in the Code of Federal Regulations. Although U.S. DOE activities are not subject to these limits they provide a reasonable benchmark for comparison in the present discussion. For several radionuclides produced in reasonable abundance at particle accelerators including $^{7}$Be, $^{54}$Mn and $^{60}$Co, the exempt quantities and exempt concentrations in solid materials from 10 CFR § 30.71 Schedules B and A are listed in Table 1.

**Table 1**
**USNRC Exempt Quantities and Concentrations**

| Radionuclide | Exempt Quantity (μCi) | Exempt Concentration (μCi/g) |
|---|---|---|
| $^{7}$Be | 0.1 [a] | 2 X $10^{-2}$ |
| $^{24}$Na | 10 | 2 X $10^{-3}$ |
| $^{54}$Mn | 10 | 1 X $10^{-3}$ |
| $^{60}$Co | 1 | 5 X $10^{-4}$ |

[a]This nuclide does not appear in the exempt quantity table. The exempt quantity shown is that assigned to "unlisted by-product materials." Typical values for other nuclides of similar γ-energy and half-life are around 10 μCi.

The absence of specific radionuclides – in particular, the accelerator produced isotope $^{22}$Na – from the NRC tables is due to the fact that they are not produced in nuclear reactors and therefore are not "by-product materials" subject to regulation by the USNRC. Even so, to be within the spirit of these regulations, it is reasonable to show that the release criterion corresponds to the detection of less than approximately 100 nCi of activity for a point source or less than 100 pCi/g of specific activity in bulk materials.

## Applications of the Criterion

### (1) Point Source

Figure 1 shows that the absolute efficiency of the NaI(Tl)-based survey meter for 1 MeV photons at 1 cm (essentially at contact) is 7 X $10^{-2}$. Thus the 2000 cpm rate (33 $s^{-1}$) implies, with this efficiency, the total emission into $4\pi$ steradians of 471 $s^{-1}$. For a decay in which a single photon is emitted 100 % of the time this represents an activity of 12.7 nCi. If two photons are emitted (e.g., in $^{60}Co$ decay), the criterion corresponds to a limit of detection of 6.3 nCi. For the case of $^{7}Be$, for which a photon of 478 keV is emitted in only about 10% of the decays, the activity limit detectable according to this criterion would be, using a detector efficiency at 1 cm of 0.1 from Fig. 1, 90 nCi. Thus the criterion successfully detects point sources at, typically, far less than the exempt quantity limit for specific nuclides. This would apply to discrete radioactive sources as well as small parts (e.g., nuts and bolts) activated by the accelerator.

### (2) 55 Gallon Drum

Graden[4], in the course of developing a method for estimating the radioactivity content of 55 gallon (nominal volume) drums filled with miscellaneous solid materials of uniform specific activity by volume, has performed a calculation which relates the exposure rate at contact with the middle of the side of such a drum to the total activity of the contents. The result was that a uniform specific activity by volume of 1.55 nCi/cm$^3$ results in a contact exposure rate of 1 mR/h. Thus, a reading of 0.01 mR/h, equivalent to the release criterion, corresponds to a specific activity by volume of 15.5 pCi/cm$^3$. The average density of a mixture of aluminum, iron, concrete, and plastics expected to be found in such a drum would be about 3.5 g/cm$^3$. Hence, the minimum detectable specific activity by mass, $S_m$, is approximately 4.4 pCi/g. Thus, the detection criterion provides a reasonable screening level for such a bulk object of intermediate size and composition. This level is far below the typical USNRC exempt concentration value.

### (3) Slab of Bulk Material

The release criterion stated here can be related to specific activity in bulk materials. Morgan and Turner[5], among others, give a relationship between the photon flux density, $\phi$, at contact with the surface of a uniformly activated infinite slab of material and the specific photon emission rate (cm$^{-3}$s$^{-1}$) per unit volume of the source, $S_v$:

$$\phi = \frac{S_v}{2\mu},$$

where $\mu$ is the narrow beam linear absorption coefficient (cm$^{-1}$) of the photons in the material slab. This is the result of a simple integration over the volume of the infinite slab and neglects build-up from the distributed source. Inclusion of build-up increases the measured value of $\phi$, so that its neglect overestimates the value of $S_v$, and therefore underestimates the sensitivity of the stated criterion. Readings taken at contact with bulk material often simulate proximity to a slab of infinite extent. For comparison with the specific activities, the specific photon emission rate per unit mass, $S_m$ ($g^{-1}s^{-1}$), is needed.

Converting from $S_v$ ($cm^{-3}s^{-1}$) to $S_m$ ($g^{-1}s^{-1}$) leads to

$$S_m = \frac{2\phi\mu}{\rho},$$

where $\rho$ is the density ($g\text{-}cm^{-3}$), and $\mu = \rho\mu_m$ where $\mu_m$ is the mass absorption coefficient ($cm^2$ $g^{-1}$). Hence,

$$S_m = 2\phi\mu_m.$$

Since we have established that $\phi$ can be detected down to an exposure rate corresponding to 0.01 mR/h or, at 1 MeV photon energy, 5.61 photons $cm^{-2}s^{-1}$, and at 0.5 MeV,[3] 10.51 photons $cm^{-2}s^{-1}$, it is straightforward to calculate the approximate minimum detectable values of $S_m$ for typical bulk materials subject to activation by accelerators. The results are shown in Table 2. Inclusion of build-up reduces these values by factors between 1.3 and 2.5 for most bulk materials of interest. Thus, the numbers shown represent upper limits of minimum detectable activities. The values for activity in Table 2 assume each radioactive decay produces one photon. Nuclides which emit *n* photons in their decay, would be detected to a sensitivity of approximately *1/nth* of the specific activity shown here.

**Table 2**

**Minimum Detectable Specific Activities in an Infinite Slab for 0.5 and 1.0 MeV Photons**

| **Material** | **$\mu_m$ ($cm^2g^{-1}$)[6]** | | **$S_m$ ($g^{-1}s^{-1}$)** | | **pCi/g** | |
|---|---|---|---|---|---|---|
| | **1 MeV** | **0.5 MeV** | **1 MeV** | **0.5 MeV** | **1 MeV** | **0.5 MeV** |
| concrete | 0.0637 | 0.0877 | 0.71 | 1.84 | 19 | 50 |
| aluminum | 0.0613 | 0.0844 | 0.69 | 1.77 | 19 | 48 |
| iron | 0.0599 | 0.0840 | 0.67 | 1.76 | 18 | 48 |
| lead | 0.0708 | 0.161 | 0.79 | 3.38 | 21 | 91 |

It is obvious from this Table that the dependence of the detectable specific activity on atomic number of the material is generally small (except perhaps for lead at 0.5 MeV). The release criterion when used to check a large object for radioactivity is therefore sensitive to specific activities far smaller than the exempt concentrations. In fact, even with $^7Be$ for which a photon is emitted in only 10% of the decays the release criterion is sensitive to specific activity (about 500 pCi/g for most materials) much less than the exempt concentration limit (Table 1) (and, of course, the DOT limit of 2 nCi/g as well). This verifies that it is adequate for objects such as large pieces of iron or concrete used in quantity for shielding at particle accelerators.

The above discussion assumes that the bulk object has been irradiated uniformly so that the photon emission rate per unit volume $S_v$ is a meaningful concept. If however the irradiation is not uniform as, for example, may be the case for a magnet struck by a pencil-like particle beam, it is possible that the surface dose rate will indicate less activity than the activity at some depth within the material. For 1000 GeV protons VanGinneken and Awschalom[7] have shown that the peak of the hadronic cascade in either an iron or concrete cylinder occurs at a depth of 1.5-3 feet within the material and at a radial distance of 0-3 cm from the point of irradiation. As seen in Figs. VIII.8 and VIII.25, in Ref. 7, shown as Figs. 2 and 3 in the present report, the entrance activation level is only about a factor of two less than the value at this location. Therefore, even under these conditions, the criterion will generally detect radioactivity at levels less than the exempt quantity limit.

## The Incineration of Released Material

Subsequent to the release of bulk material based on the criterion discussed above, it may then be subjected to incineration. Such disposition of the item constitutes a much more efficient means than, e.g., machining operations, of releasing radioactivity which could potentially result in exposure to members of the public. On the assumption, based on Fermilab's experience with its magnet debonding oven, that the material, e.g., a large Fermilab magnet weighing 35000 lbs, is placed in an oven that can "burn" 50 metric tons a day (a rate of about 5000 lb per hour), the ground level concentration of radionuclides can be calculated as a function of the downwind distance from the stack. A Gaussian plume model is employed in the following form:[8]

$$\overline{X} = \frac{Q}{\pi \sigma_y \sigma_z \overline{u}} \exp\left[-\left(\frac{y^2}{2\sigma_y^2} + \frac{h^2}{2\sigma_z^2}\right)\right].$$

Here Q is the source term, $\sigma_y$, $\sigma_z$ are the lateral and vertical diffusion coefficients respectively, $\overline{u}$ is the mean wind speed, y is the lateral distance from the center of the plume, and h is the stack height at the furnace.

Table 2 shows minimum detectable activities that are somewhat less than 100 pCi/g. Nonetheless for this calculation we assume bulk material (a large Fe magnet, e.g.) with this concentration of radionuclides. The source term Q is, then,

$$100 \text{ X } 10^{-12} \frac{\text{Ci}}{\text{g}} \text{ X } 454 \frac{\text{g}}{\text{lb}} \text{ X } 5000 \frac{\text{lb}}{\text{hr}} \text{ X } \frac{\text{hr}}{3600 \text{ sec}} = 63 \frac{\text{nCi}}{\text{sec}} .$$

The average annual wind speed is taken as 4 m/sec, or 8.9 miles/h, typical for Fermilab. Diffusion coefficients associated with neutral wind conditions are used. They are shown in Table 3, taken from the figures on pages 102 and 103 in Ref. 8. The lateral distance y from the center of the plume is set equal to zero, the worst case. The results of the calculation, performed for stack heights of 10 m and 30 m, are shown in Figs. 4 and 5.

**Table 3**
**Diffusion Coefficients $\sigma_y, \sigma_z$ for Neutral Wind Conditions. From Ref. 8.**

| Downward Distance (meters) | $\sigma_y$ (meters) | $\sigma_z$ (meters) |
|---|---|---|
| 100 | 8.19 | 4.92 |
| 200 | 16.5 | 8.87 |
| 300 | 23.8 | 12.1 |
| 400 | 32.5 | 15.6 |
| 500 | 39 | 18.9 |
| 700 | 52.7 | 24.5 |
| 1000 | 72.4 | 32.2 |
| 2000 | 135.7 | 52.7 |
| 3000 | 194.5 | 64.8 |
| 4000 | 241.7 | 80 |
| 5000 | 300 | 90 |
| 7000 | 400 | 109 |
| 10000 | 547.7 | 136 |

Fig. 5 shows the concentration as a percentage of the Derived Concentration Guide (DCG), given by DOE Order 5400.5, for $^{60}Co$, the radionuclide with the most restrictive limit. The worst case–for the 10 m stack–shows values less than about 22% at all distances from the stack. This is, furthermore, a very conservative estimate. The DCG is based on a committed dose equivalent of 100 mrem taken into the body during <u>one</u> year, for an individual exposed to such a concentration during the whole time, while the radioactivity from the incineration of the 35000 lb magnet in this example would in fact be released in about seven hours. It is improbable that any individual or population would be exposed, in the worst case, to more than one such incineration in a given year. Inhalation at 22% of the DCG for only seven hours corresponds to a committed dose equivalent of $\frac{7}{8760}$ X 100 mrem X 0.22 = 0.02 mrem. Incineration of less massive items results in corresponding smaller values of the committed dose equivalent.

## ALARA Analysis

Radiation exposures are to be kept As Low As Reasonably Achievable (ALARA). This section reviews alternative screening methods from the ALARA perspective.

### (1) γ-ray Analysis

γ-ray spectroscopy is sensitive to specific activities as low as about 10 pCi/g. However, the technique is impractical for screening large scale quantities of material and equipment because of time and cost. A typical γ-ray analysis facility that costs about $50,000 can only process a few samples per day. On the other hand, Fermilab needs to be able to scan hundred of items every day. γ-ray sampling would not therefore provide as comprehensive coverage as the survey technique discussed above and truly representative sampling would be difficult to achieve. Thus, this alternative is not practical from either a cost or performance standpoint.

### (2) Survey Screening at a Lower Threshold

The present criterion is based on a screening survey at 2000 cpm above background. One might instead consider a lower threshold. However, significant fluctuations in the natural background in common situations could exceed the screening criterion and result in erroneous and inconsistent determinations. This alternative would not achieve a consistent significant reduction in the already small estimated worst case exposures from direct contact with material and from incineration.

### (3) Detector of Higher Sensitivity

Since NaI (Tl) scintillator crystals are in general the most efficient gamma ray detectors available, one might consider a larger sized crystal. However, the gain in sensitivity increases only modestly with detector size, and larger crystals are considerably more costly. Furthermore, finite source/detector size effects, which can complicate analyses, become more important as the size increases. Therefore, such an alternative will not necessarily lead to significantly improved performance.

## Conclusion

The screening criterion discussed in this report cannot detect the presence of $^3H$. However, the cross section for production of $^3H$ by high-energy physics processes is comparable in magnitude to that for other nuclides. From considerations of build-up and decay, the concentration of tritium in bulk material will always be comparable to that for other material to which the screening criterion is sensitive. Because of the much larger USNRC exempt quantity limit (1000 μCi) and exempt concentration limit (3 X $10^{-2}$ μCi/ml), and the DCG of 2 X $10^{-2}$ μCi/ml from DOE 5400.5, any $^3H$ component is not a significant exposure pathway.

It is concluded that the release criterion as discussed here provides reasonable means for protecting the public from receiving significant quantities of radioactivity in items released from Fermilab.

**REFERENCES:**

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3. H. W. Patterson and R. H. Thomas, *Accelerator Health Physics,* (Academic Press, 1973), p. 59.

4. K. Graden, "Activity Content Approximation in Radioactive Waste Drums," Radiation Physics Note. No. 78, Revised May, 1990.

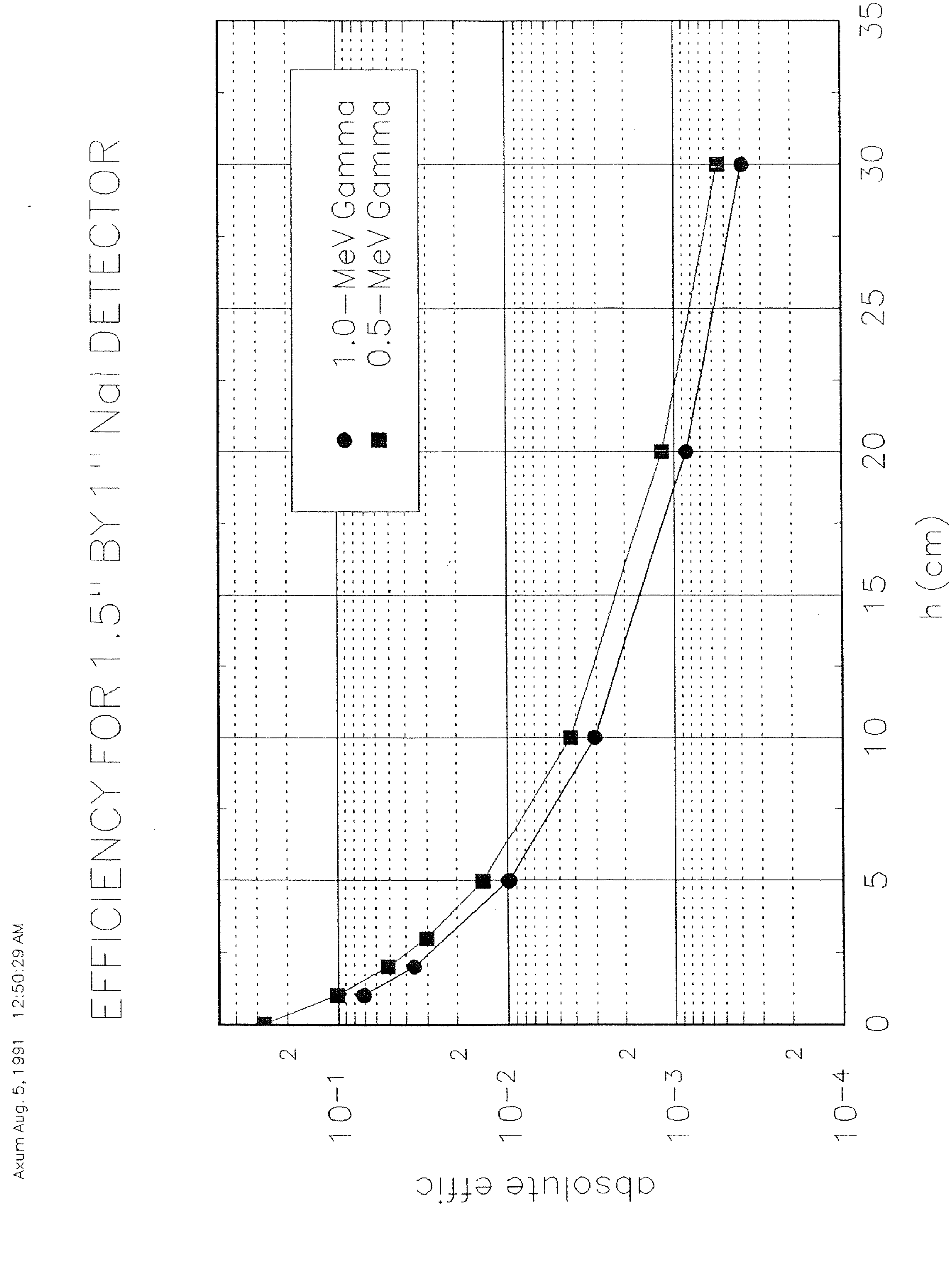
5. K. Z. Morgan and L. C. Emerson, "Doses from External Sources of Radiation," Chapter 9 in K. Z. Morgan and J. E. Turner, Editors, *Principles of Radiation Protection,* (Robert E. Krieger, Huntington, NY, 1973).

6. *The Health Physics and Radiological Health Handbook* (Nucleon Lectern Associates, Inc., 1984).

7. A. VanGinneken and M. Awschalom, "High Energy Particle Interactions in Large Targets, Vol. 1, p. 73" (Fermi National Accelerator Laboratory, Batavia, IL, 1975).

8. F.A. Gifford, "An Outline of Theories of Diffusion in the Lower Layers of the Atmosphere," Chapter 3 in D.H. Slade, Editor, Meteorology and Atomic Energy 1968, (USAEC Tech. Inf. Center, Oak Ridge, TN, July 1968), pp. 66-116.

**Figure 1**



Axum Aug. 5, 1991 12:50:29 AM
EFFICIENCY FOR 1.5" BY 1" NaI DETECTOR
1.0—MeV Gamma
0.5—MeV Gamma
absolute effic
10−1
10−2
10−3
10−4
2
0
5
10
15
20
25
30
35
h (cm)

# Figure 2

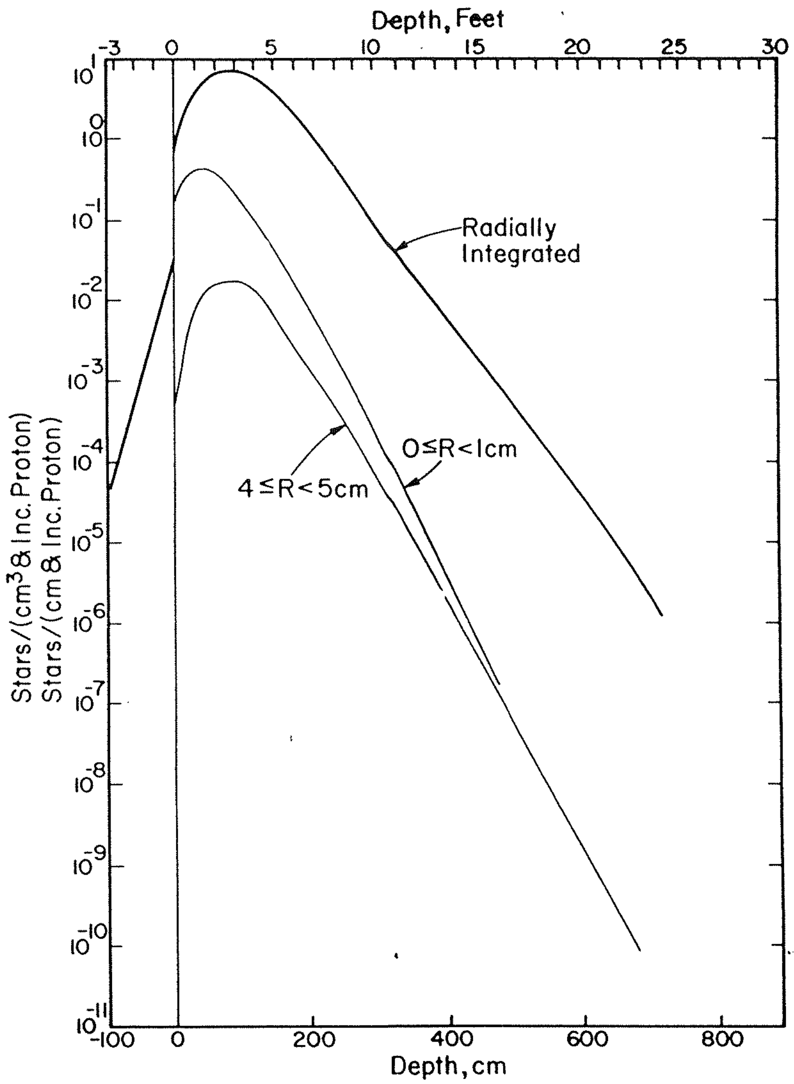



Fig. VIII.8. 1000 GeV/c protons incident on a solid iron cylinder. Depth dependence of the radially integrated star density (star/cm · inc. proton) and the star density at small radii (stars/cm$^3$ · inc. proton). The beam of $0.3 \times 0.3$ cm cross section is centered on the cylinder axis and starts to interact at zero depth. The star density includes only those due to hadrons above 0.3 GeV/c momentum. The star densities at small radii are not shown in the backward region due to statistical uncertainty.

# Figure 3

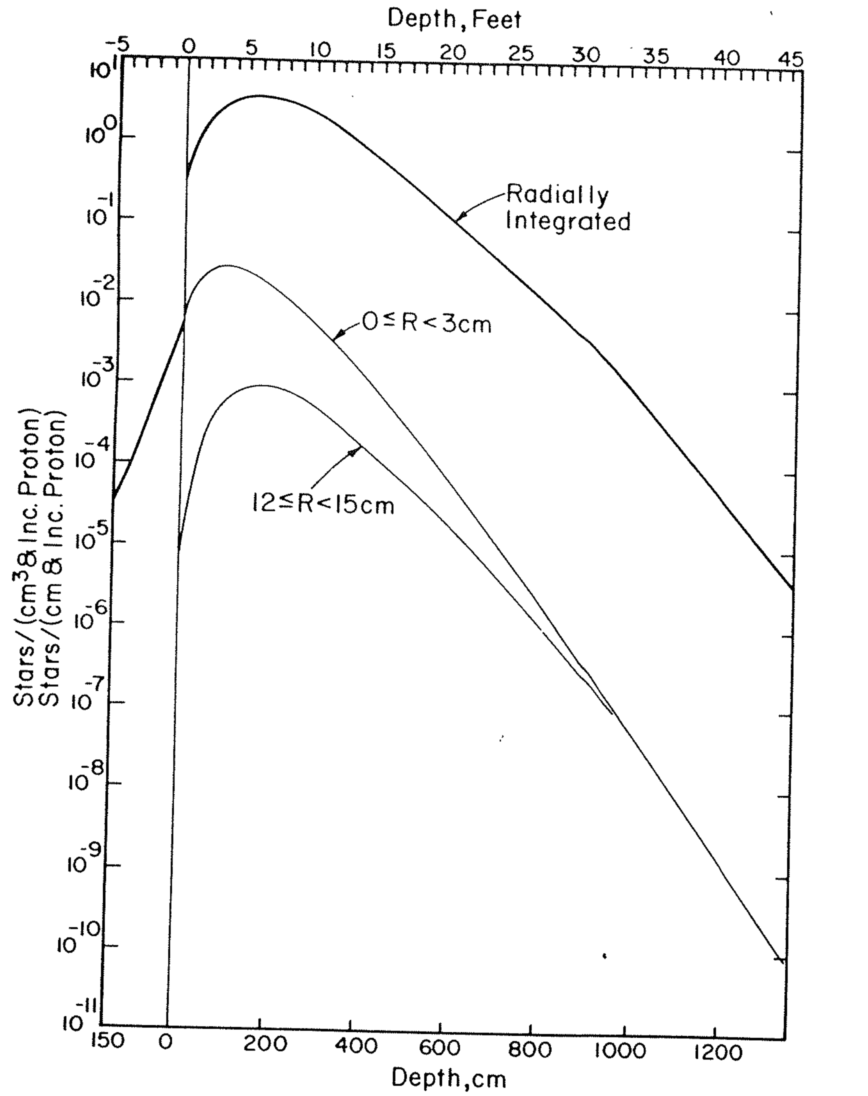



Fig. VIII.25. 1000 GeV/c protons incident on a solid concrete cylinder. Depth dependence of the radially integrated star density (stars/cm · inc. proton) and the star density at small radii (stars/$cm^3$ · inc. proton). The beam of 0.3 × 0.3 cm cross section is centered on the cylinder axis and starts to interact at zero depth. The star density includes only those due to hadrons above 0.3 GeV/c momentum. The star densities at small radii are not shown in the backward region due to statistical uncertainty.



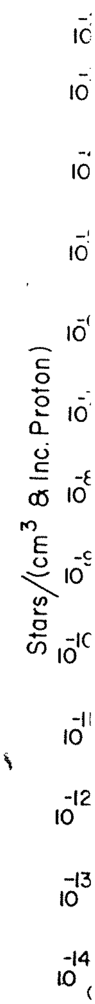
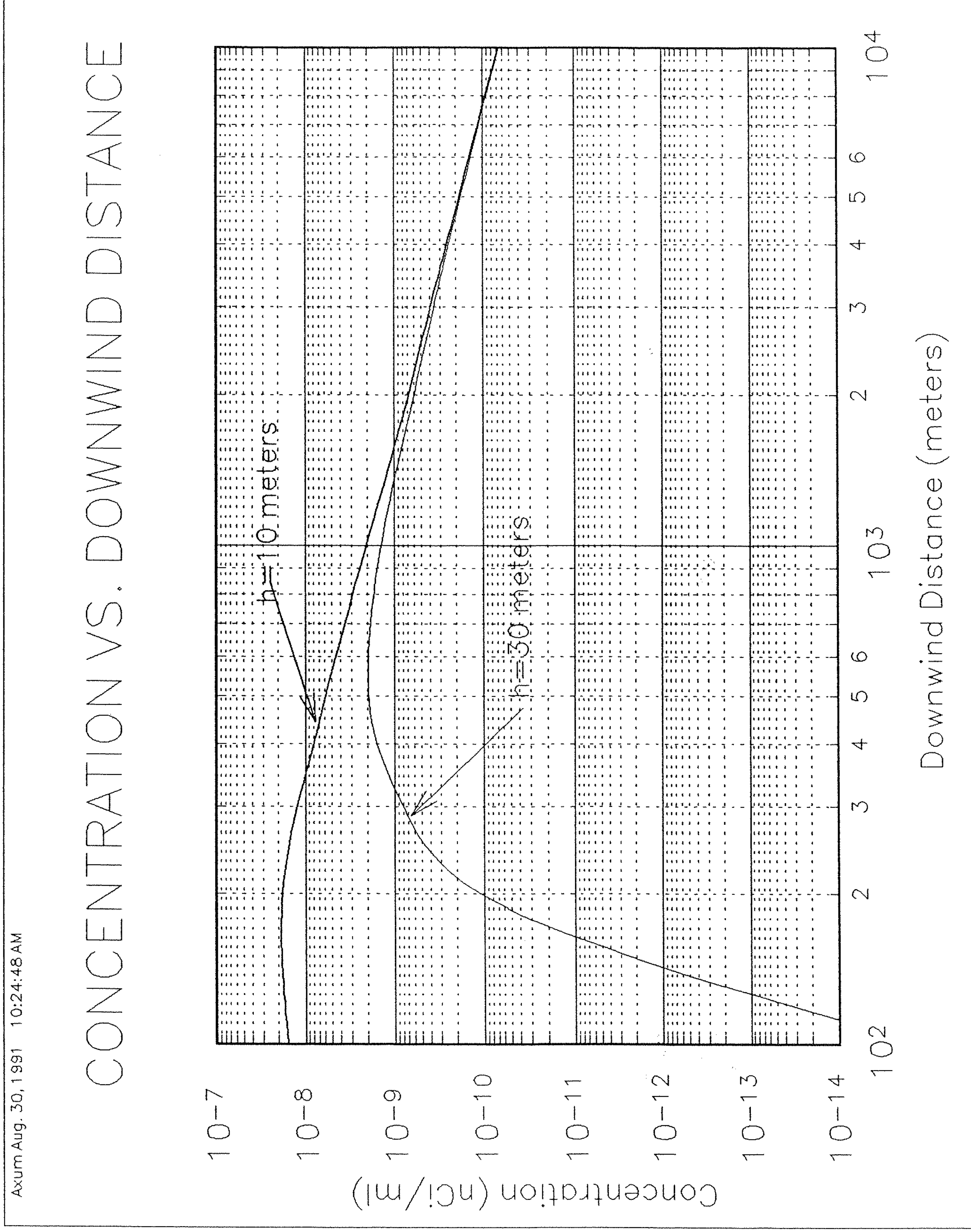


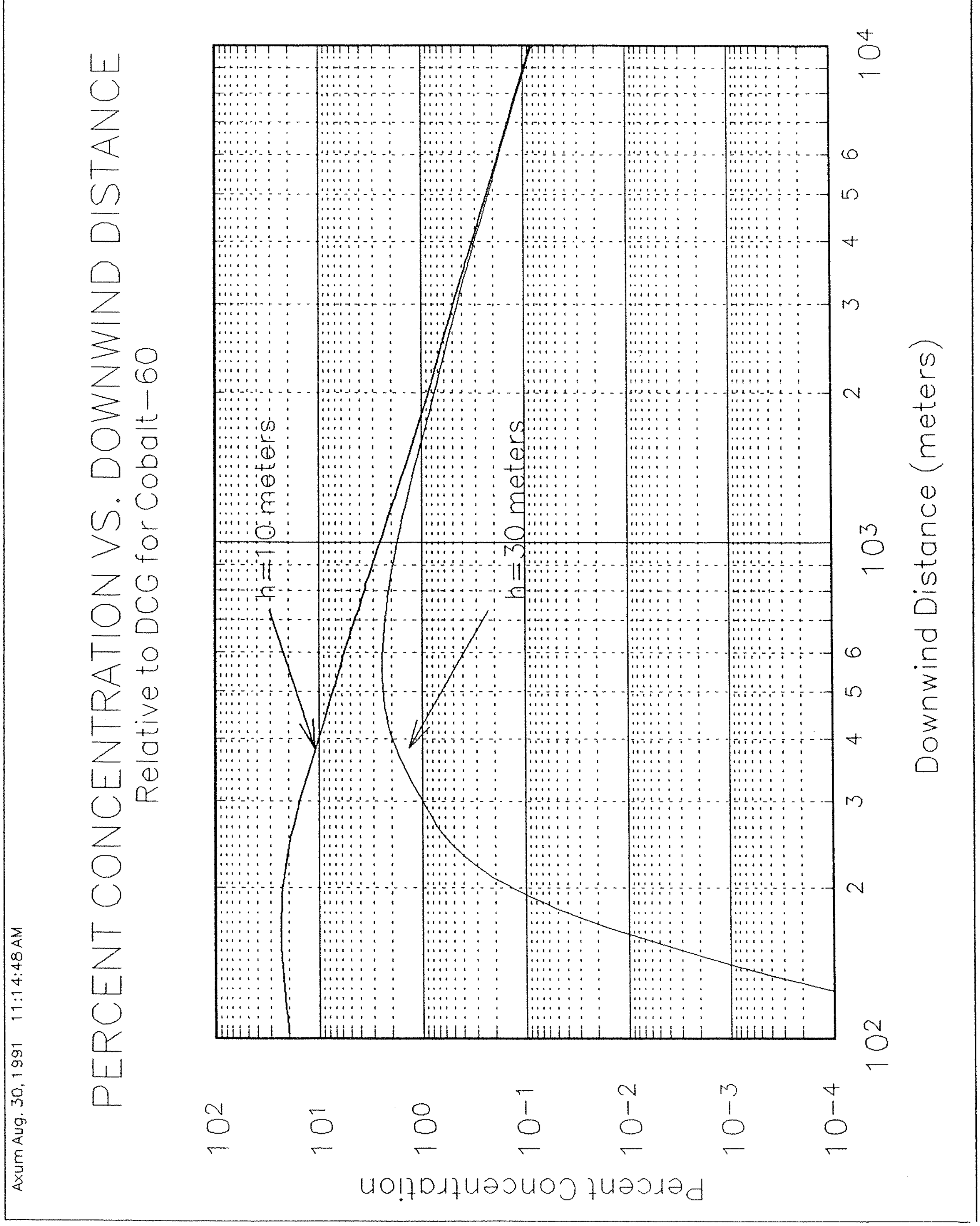

Fig. VIII.26.
star densit
section is c
above 0.3 G

**Figure 4**



Axum Aug. 30, 1991 10:24:48 AM
CONCENTRATION VS. DOWNWIND DISTANCE
h=10 meters
h=30 meters
Concentration (nCi/ml)
10-7
10-8
10-9
10-10
10-11
10-12
10-13
10-14
102
2
3
4
5
6
103
2
3
4
5
6
104
Downwind Distance (meters)

**Figure 5**



Axum Aug. 30, 1991 11:14:48 AM
PERCENT CONCENTRATION VS. DOWNWIND DISTANCE
Relative to DCG for Cobalt-60
h=10 meters
h=30 meters
Percent Concentration
10^2
10^1
10^0
10^-1
10^-2
10^-3
10^-4
10^2
2
3
4
5
6
10^3
2
3
4
5
6
10^4
Downwind Distance (meters)