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by

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ABSTRACT

Samples from the Islamic holy water Zamzam in Mecca, Saudi Arabia and the famous mineral water of Hot Springs, in Hot Springs, Arkansas were analyzed for trace elements content by thermal neutron activation analysis. Coupled with the respective standards, samples of each were subjected simultaneously to a thermal flux of $2x10^{12} \frac{n}{cm^2\text{-sec}}$ (except for the case of ^{82}Br and ^{76}As where the flux = $1.3x10^{13} \frac{n}{cm^2\text{-sec}}$ to produce the thermal neutron activation products ^{20}F, ^{37}S, ^{49}Ca, ^{24}Na, ^{27}Mg, ^{128}I, ^{38}Cl, ^{56}Mn, ^{31}Si, ^{42}K, ^{64}Cu, ^{76}As, and ^{82}Br. Interferences from undesirous nuclides for the procedure in question were minimized by applying an isotopic-ion-exchange method of separation. For Zamzam the concentration of ^{37}S, ^{49}Ca, ^{38}Cl, ^{31}Si, ^{42}K, ^{24}Na, and ^{82}Br were found, respectively, in parts per million, to be 3, 107, 11, 12, 4, 14, and 9; and that for Hot Springs Sample, replacing ^{82}Br with ^{27}Mg, are (in parts per million), 2, 44, 2, 10, 1, 4, and 5. The experimental limit of detection for pure standards of the nuclides ^{27}Mg, ^{128}I, ^{64}Cu, and ^{56}Mn were found to be (in μg) 8, $8x10^{-3}$, $6x10^{-2}$, and $2x10^{-4}$ respectively. These nuclides were not detected in Zamzam, therefore, it was concluded that in Zamzam the concentration levels of the nuclides ^{27}Mg, ^{128}I, ^{64}Cu, and ^{56}Mn were below that of the limit of detection of pure standards.

Aktivierungsanalyse mittels thermischer Neutronen des Zamzam-Wassers in Mekka, Saudi-Arabien, und des Wassers der 45 heißen Quellen in Hot Springs, Arkansas, USA

KURZFASSUNG

Proben des in der islamischen Religion heiligen Zamzam-Wassers in Mekka, Saudi-Arabien, sowie Proben des berühmten Mineralwassers der heißen Quellen in Hot Springs, Arkansas, wurden durch Aktivierungsanalyse mittels thermischer Neutronen auf ihren Gehalt an Spurenelementen untersucht. Zusammen mit den jeweiligen Standards wurden Proben beider Gewässer gleichzeitig einem thermischen Fluß von $2\mathrm{x}10^{12}\ \frac{n}{cm^2\text{-}sec}$ ausgesetzt (ausgenommen ^{82}Br und ^{76}As, bei denen der thermische Fluß $1{,}3\mathrm{x}10^{13}\ \frac{n}{cm^2\text{-}sec}$ betrug), um die Produkte der Aktivierung mit thermischen Neutronen herzustellen, d.h. ^{20}F, ^{37}S, ^{49}Ca, ^{24}Na, ^{27}Mg, ^{128}I, ^{38}Cl, ^{56}Mn, ^{31}Si, ^{42}K, ^{64}Cu, ^{76}As und ^{82}Br. Störende Einflüsse der für die durchgeführten Arbeiten unerwünschten Nuklide wurden durch Trennung mittels Isotopenaustausch auf ein Minimum begrenzt. Für Zamzam-Wasser wurde eine Konzentration in ppm gefunden, die für ^{37}S 3 beträgt, für ^{49}Ca 107, für ^{38}Cl 11, für ^{31}Si 12, für ^{42}K 4, für ^{24}Na 14 und für ^{82}Br 9. Die entsprechenden Konzentrationen in den Proben der heißen Quellen, wobei ^{82}Br durch ^{27}Mg zu ersetzen ist, betragen (in ppm) 2, 44, 2, 20, 1, 4 und 5. Die Nachweisgrenze ergab sich im Versuch für reine Standards der Nuklide ^{27}Mg, ^{128}I, ^{64}Cu und ^{56}Mn (in μg) zu $8{,}8\mathrm{x}10^{-3}$, bzw. $6\mathrm{x}10^{-2}$ und $2\mathrm{x}10^{-4}$. Die genannten Nuklide wurden im Zamzam-Wasser nicht nachgewiesen, was zu der Schlußfolgerung führte, daß die Konzentrationswerte der Nuklide ^{27}Mg, ^{128}I, ^{64}Cu und ^{56}Mn im Zamzam-Wasser unter der Nachweisgrenze für reine Standards liegen.

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1. INTRODUCTION

In Mecca, the center of Islamic World, the "Kabah" is the oldest building originally constructed by Abraham thousands of years before the birth of Islam, in A.D. 610, forty years after the birth of Prophet Mohamed(1).

Surviving within few yards on the North-East corner of this Holy Temple is the most famous well within Islamic World known as Zamzam. The Koraan, the official constitution of Islam, describes the well as being miraculously sprung when Abraham's wife, accompaning her husband in the "heavenly mission" of the first construction of Al-Kabah, was running up and down on the painfull sounds of her only child crying of hunger and thirst. Ever since then, residents of the area have protected the well from all sources of pollution and hazards.

The taste of Zamzam is completely different from other wells in the area or any ordinary drinking water brought into the city of Mecca from springs and wells found in lightly populated valleys within a radius of 30 miles.

In addition to its distinguished taste, Zamzam enjoys a unanimous popularity among 350 million Muslems all over the world. To a Muslem, Zamzam is the everlasting curing liquid that unquestionably contains medicinal properties for all illness. This has been "proved" repeatedly provided that faith dominates before and after dosages of Zamzam. The government of Saudi Arabia is in full charge of the well Zamzam; its purity; its security; its limited delivery to all over the Islamic World. Politically inclined propaganda from abroad has repeatedly rated Zamzam only as good as any drinking water found in the Middle East.

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[1] The Koraan informs that Abraham was the first constructor of the Kabah.

Thus, it is easily seen that investigation of its trace elements presents both an interesting and informative project. Due to the range of sensitivity such investigation is best when carried out by means of activation analysis. To an activation analyst the general routine of investigating trace elements in a sample is invested in the following three interdependent steps:

(1) to produce bombarding particles of suitable energy from suitable generator

(2) to bombard the target for a predetermined length of time

(3) to detect and identify the nuclear transmutation.

Bombarding particles can be neutral or charged particles. Targets can be prepared in small quantities. Length of irradiation, and consequently of delay and counting time can be predetermined by the characteristic parameter of the decaying particle, the half-life.

Knowledge of elemental components of Zamzam are not published largely simply to preserve the sacred flavor of the water at its best. Such lack of knowledge automatically characterizes the investigation as a non-routine analysis. This in turn introduces serious difficulties concerning the above three general interdependent steps in traces analysis by activation. The procedure to follow in circumstances as such is to compare Zamzam with another water in terms of the latter's known compositions. One such water of importance is the famous mineral water of Hot Springs, Arkansas. Furthermore, the investigation, then, will be termed as a semi-routine analysis which advantageously allows to select proper methods of irradiation and detection. Irradiation can take place by nuclear reactors, 14-Mev neutron generators, isotope neutron sources, cyclotron, and linear accelerators (high energy photons). Radiation can be detected by sodium iodide scintillators, solid state detectors, beta particle counting, and coincidence counting.

Selection of methods of irradiations and counting depends on nuclides under investigations. Many nuclides display high sensitivity for thermal neutrons. Bombardment by thermal neutrons results in nuclear reactions of the type (n,γ). The monoenergetic emitted gammas can be detected by scintillation and solid state detectors.

2. OBJECTIVE

Being born in Mecca, I am most interested in a thermal neutron analysis of Zamzam for its trace elements and a comparison of this analysis to one obtained for the mineral water of the famous springs of Hot Springs, Arkansas. Specifically, thermal activation is to be limited to the investigation of those elements reported to be present in the Hot Springs water(1). Among these elements only the following can be determined by thermal neutron bombardment:

Fluorine
Calcium
Sulfur
Potassium
Sodium
Magnesium
Chlorine
Silicon

Furthermore, investigation is to be extended to the detection of the following elements which are not reported to be found in Hot Springs water:

Iodine
Manganese
Copper
Bromine
Arsenic.

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[1] See notes in Appendix B

Therefore, in summary, the main objective is to compare quantitatively the components, as determined by thermal neutron activation analysis, of the two waters to illustrate their similarities and their differences.

3. THEORY

3.1 General Considerations

Activation analysis is the process of artificial production of radioactive nuclei and their qualitative and quantitative analysis. This process is generally represented by the following equation:

$$A + B \rightarrow C + D + (\text{Energy}) \qquad (1)$$

where,

	A	is the target nucleus
	B	the incoming projectile
	C	the artificially produced nucleus ($A=C$ or $A \neq C$)
and	D	the outcoming particle or photon.

Specifically equation (1) can be termed as:

1. Elastic scattering where total K.E. of $A + B$ is the same as that of $C + D$ ($C = A$).
2. Inelastic scattering where total K.E. of $C + D$ is less than that of $A + B$ ($C = A$); the difference being used to raise the nucleus A to an excited state.
3. Nuclear transmutation such that $C \neq A$ and D is one or more emitted particles or photons.

For a nuclear transmutation to occur, the incoming particle, except neutron or photon, must possess energy in excess to the height, V, of the potential barrier around a nucleus of charge Z_1^*:

$$V = 1.44 \frac{Z_1 Z_2}{R_1 + R_2} \quad \text{Mev}$$

where

Z_2 is the charge of the approaching particle with radius R_2;

R_1 is the radius of the nucleus; (R_1 & R_2 are expressed in fermis)

Examples of nuclear transmutations are represented by the following reactions:

1) $^{16}O(^{3}He,p)^{18}F$
2) $^{9}Be(\gamma,n)^{8}Be$
3) $^{19}F(n,2n)^{18}F$
4) $^{41}K(n,\gamma)^{42}K$

Reaction (1) is a specific example of charged particle activation analysis. In this type of nuclear analysis the bombarding particles, i.e. $^{3}He,p,\alpha,d,t$, or heavy charged particles, are usually produced and accelerated to the desired energy in Cockcroft-Walton voltage multipliers, Van de Graaff generators, multiple stage Van de Graaff generators, cyclotrons, or linear accelerators. Charged particle activation analysis has the following advantages:

A) suitability for activation of lighter elements
B) ability to avoid interferences by means of careful selection of projectile type and energy
C) controlability of penetration depth.

On the other hand, the disadvantages are due to the low depth penetration of the charged particles into matter and the possible consequent heat dissipation; this has the effect of limiting not only the type and form of sample that can be irradiated, but also the method of containment[3].

Reaction (2) is an example of photon activation analysis; because of the low neutron binding energy in Berillium. This reaction can be induced by the decay photons from an isotope such as Antimony-124.

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*Tunneling effects neglected

Usually the photo-neutron threshold exceeds 5 Mev. This form of activation analysis allows, among other things, determination of elements unsuitable to be determined by neutron induced transmutations, though large accelerators are required for most γ-photon activations.

Reactions (3) and (4) are neutron induced reactions. Mechanisms of neutron interactions are named direct interaction, potential or shape scattering, and compound nucleus formation. In direct interaction process a high energy neutron, usually not found in a thermal reactor, collides with a nucleus resulting in knocking out a nucleon, say proton (direct interaction (n,p) reaction), or emerging itself with reduced energy and leaving the struck nucleus in an excited state (direct interaction inelastic scattering); potential scattering is an elastic scattering which occurs with neutrons of any energy simply because of the presence of a nucleus. It is a function of the forces acting on a neutron as it moves in or near the nucleus. In compound nucleus formation an incident neutron with energy, in center of mass system, $E_c = 1/2\ n\ v_1^2$ + Binding Energy of neutron, where μ is the reduced mass and v_1 is the velocity of neutron in the lab-system, is usually shared among nucleons. This is known as an intermediate state. The compound nucleus may decay by one of the following modes:

1) compound elastic scattering: emission of a neutron with the nucleus returned to ground state
2) compound inelastic scattering: emission of a neutron with nucleus retained in excited state
3) radiative capture: emission of one or more gamma rays.

The probability that a neutron interaction would result in compound nucleus formation is a product of the probability, or cross section, for formation of compound nucleus, designated as $\sigma_{cn}(E_c)$, times the relative probability for the specified mode of decay[7].

Reaction (3) exemplifies nuclear transmutation induced by fast neutrons (14-Mev). Fast neutrons can be produced by number of reactions among which the D-T reaction is most energetic:

$$^{3}H + {}^{2}H \rightarrow {}^{4}He + n + 17.6 \text{ Mev.}$$

The neutron energy is approximately 14 Mev; such monoenergetic neutrons are sufficient to produce (n,2n), (n,p), and (n,α) reactions with nearly all elements. However, it is difficult with present technology to achieve a high flux of fast neutrons.

Finally reaction (4) is induced by thermal neutrons of high intensity produced in nuclear reactors. A thermal neutron, when designated to have the most probable velocity for a Maxwell-Boltzmann distribution at a specified temperature, is a neutron with kinetic energy and velocity of 8.61×10^{-5} T ev and 1.28×10^{4} $T^{1/2}$ cm/sec respectively; at 293 ^{0}K the thermal neutron energy is 0.025 ev and its velocity is 2.2×10^{5} cm/sec. Nuclear reactors allow the simultaneous irradiation of many targets subjected to high neutron flux of 10^{10} to 10^{14} $\frac{n}{cm^{2}\text{-sec}}$. Many elements, except O, C, H, and a few others, exhibit great sensitivity for the 0.025 ev. neutrons from thermal reactors: this increase in sensitivity, as compared to the 14-Mev induced transmutation, is accompanied by "interference".

3.2 Specific Development

Consider the artificial production of the nuclide ^{24}Na in a thermal reactor from a sample of single composition, Na, but unknown quatity. The transmutation reaction is symbolized by:

$$^{23}Na + n(\text{Thermal}) \rightarrow {}^{24}Na \rightarrow {}^{24}Mg + \beta + \gamma\left(E_{\gamma} = \begin{array}{l} 1.37 \text{ Mev} \\ 2.76 \text{ Mev} \end{array}\right)$$

with

% abundance (^{23}Na) = 100
cross section (^{23}Na) = 0.53 b

To produce the ^{24}Na nuclide, the single composition sample must be irradiated for a certain length of time, say 1 hour, at center of the core. The transmutation is then identified by the following two parameters:

E_γ = 1.37 Mev. (easily detectable); 2.76 Mev.
$t_{1/2}$ = 15 hr.

The procedure toward positive identification of the transformation is accomplished, in two steps, preferably with gamma spectrometer analysis. This analysis employs scintillation or solid state detectors. The qualitative analysis is achieved by locating two photopeaks - for 1.37 Mev. and 2.76 Mev. - and determining the characteristic parameter of the nuclide, its half life. The quantitative analysis starts with solving for w, weight in grams, in the activity equation:

$$\begin{array}{c}\text{activity in}\\ \text{disintegration/sec}\end{array} = \left(\frac{N_o W}{A}\right)(\sigma\phi)(1-e^{-\lambda t_a})\ (e^{-\lambda t_d})\ \frac{(1-e^{-\lambda t_c})}{\lambda}\ GE\Theta Br \qquad (2)$$

where,

N_o = Avogador's number = 6.02 x 10^{23} nuclei/mole
W = weight of element (^{24}Na) in grams
A = mass number of target nucleus = grams/mole = 22.98977
σ = cross section in cm^2 (1 b = 10^{-24} cm^2)
λ = decay constant (^{24}Na) = $\frac{0.693}{t_{1/2}}$ (sec^{-1})
G = geometry factor
E = efficiency factor
Θ = % abundance of ^{23}Na
Br = branching ratio

$t_{1/2}$ = half-life (^{24}Na) (sec)
t_a = irradiation time (sec)
t_d = time at start counting - time at end of irradiation = delay time (sec)
t_c = time at end of counting - time at beginning of counting (sec)
ϕ = neutron flux $\left(\frac{n}{cm^2\text{-sec}}\right)$

Now solving for W,

$$W(\text{grams}) = \frac{A\ (\text{activity in dist./sec})}{N_o\sigma\phi(1-e^{-\lambda t_a})\ (e^{-\lambda t_d})\ (1-e^{-\lambda t_c})\ GE\Theta Br}$$

Both geometry and efficiency factors can be experimentally determined.

Thermal neutron analysis of a mixture of elements in a sample requires the full inspection, for each element, of the parameters,

half-life (product)
gamma energies (product),
per cent abundance (reactant),
photopeak yield/gram of element (product),
limit of detection (product)$^{(1)}$

Accordingly, the proper length of irradiation, number of irradiations, and length of counting must be carefully planned. An approach to plan irradiation and consequent measurement of activity, in this case, is to divide the transmutations into short, intermediate, and long lived

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[1] Limit of detection and photopeak yield/(gram of element) for thermal neutron products are found in literature; one such source of information is a report entitled Nuclear Activation Analysis prepared by Wainerdi, et al., TAMU, 1966

artificial products. Then each division is to be detected by a single irradiation.

Now, to illustrate planning for irradiation consider Figure 1 where, as an example, activity in arbitrary units is plotted vs. time in hours for production of ^{56}Mn. The fact to observe, most pertinent to the estimate of a length of irradiation, is that when the target ^{56}Mn is bombarded for a length of time comparable to 3-4 times the half-life of the product ^{56}Mn ($t_{1/2}$ = 2.58 h), activity reaches virtually, saturation state. Therefore any target sample will not exhibit a significant increase in ^{56}Mn activity if irradiated for more than 3-4 half-lives. This conclusion is general for all nuclides.

For a long-lived reactor product, i.e. ^{182}Ta ($t_{1/2}$ = 115 d), to reach saturation activity irradiation must take place for many days: This is not economical or experimentally convenient. Nor can the neutron flux variation be correctly predicted over this period of time. Therefore, for a long-lived product irradiation should be planned only for an estimated length of time, providing a reasonable percentage of saturation activity, and a saturation correction must be applied; the term $(1-e^{-\lambda ta})$ in equation (2) is the saturation correction factor. On the other hand, a short-lived component can conveniently reach 50% of saturation activity by means of irradiating for an equivalence of one half-life.

Now the question arises whether one can detect each nuclide within each division or not? Obviously, consideration has been given so far only to the presence of photopeaks in γ-spectrometry. This is not the entire picture.

The three primary interactions of gamma rays with the detectors are named photoelectric, compton, and pair production. Each of these interactions is dependent on the radiaiton energy and the atomic number of the detector. In photoelectric interaction, the incident gamma ray collides with the electron cloud of the atom to eject one of the most strongly bound electrons with a kinetic energy equal to the difference between the incident photon energy and the binding energy of the electron. The cross section per atom for photoelectric interaction, τ, is related to the binding energy of the orbital electrons and the number of atoms per unit volume as $\tau \sim NZ^5(h\nu)^{-2.5}$ for the case of $h\nu << 0.51$ Mev[8]. Thus photoelectric effect is predominant at low gamma energies. Compton interaction is considered to take place between sufficiently high gamma ray energies and free electrons. As a result of not losing all of its energy, the photon will scatter with energy

$$h\nu' = \frac{h\nu}{1 + (1 - \cos\Theta)h\nu/mc^2}$$

where Θ is the angle between the scattered and the primary photon. The electron is recoiled with a kinetic energy E given by

$$E = h\nu - h\nu'$$

$$= h\nu \left[\frac{(1 - \cos\Theta)h\nu/mc^2}{1 + (1 - \cos\Theta)h\nu/mc^2} \right]$$

The cross section per atom for the Compton effect is expressed as

$$\sigma \sim \frac{NZ}{h\nu} (\ln \frac{2h\nu}{mc^2} + 1/2)$$

for the case of $h\nu >> 0.51$ Mev.[8] When the photon energy is greater than 1.02 Mev., the interaction termed pair production can occur in which the gamma ray disappears and a positron and an electron are produced with kinetic energy equals to $h\nu$ - 1.02 Mev. This process takes place in the presence of the nucleus so that conservation of momentum occurs. It is found that the probability for pair production increases with the square of the nuclear charge. And the partial absorption coefficient per atom, k, is written as

$$k \sim NZ^2 (h\nu - 2mc^2);\ \text{in the vicinity of 1 Mev.}^8$$

and

$$k \sim NZ^2 \ln h\nu;\ \text{at higher photon energies.}$$

In gamma spectrometry compton scattering presents the problem of shadowing nearby photopeaks. That is compton edges associated with higher photopeaks, especially of those with high sensitivity, fold or cover lower peaks. Unless the higher photopeak is short lived product, detection of lower peaks becomes difficult. Such difficulty is termed as interference. Elimination of predominant peaks can take place before or after counting. One can either plan to irradiate and count first, then unfold the complex spectrum by substracting out the interferring components; or one can plan to irradiate, carry out a chemical separation for undesired radioactive products, then measure desired activity.

The radiochemistry of a great many elements is summarized in a series of monographs issued by the U.S. National Academy of Science, Nuclear Science Series NAS-NS.[3]

A variety of techniques are employed among which most popular methods are precipitation, solvent extraction, differential migration, distillation, electro-analytical, and ion exchange. Selection of a method of separation depends upon the parameters of the nuclide (s) to be detected. Separation must be completed in a reasonable length of time and result in a high decontamination factor.

An ion exchange method for separation of ^{24}Na is reported by Chung-Wai Tang and Constantine J. Maletskos (Science, Vol. 167). In this method preheated (at 110 ^{0}C for 3 hrs.) sodium chloride (reagent grade granular) is introduced into a column; an aqueous solution of 1 ml of the activated sample combined with 10 ml of acetone and few drops of conc. HCl are to be poured through the column. The radioactive solution, as collected, is reported to be free of ^{24}Na activity. The authors have mentioned the following three important points about this simple method of separation:

1.) The method yields a decontamination factor greater than 10^6 (for 5 grams of NaCl in a column to separate ^{24}Na in microgram amounts).
2.) It was found, when tested, that the fractions of the following elements were also retained in the column: Sc, Mn, Sr, Ag, Ba, Ta.
3.) The method is applicable to other ions as well.

Now when two thermal neutron products must be identified by the same photopeak, i.e. the peak 0.84 Mev. for both $^{27}Mg(t_{1/2} = 9.5$ m) and $^{56}Mn(t_{1/2} = 2.58$ h), an alternate method of analysis, provided that the half-lives involved differ by a factor greater than 2, is to analyze a composite decay curve. To accomplish this the straight tail of the upward concave composite curve is extrapolated back to time zero. The extrapolated line and the straight tail of the composite curve combine to make up a straight line which is the decay curve of the long-lived nuclide. The decay curve of the short-lived nuclide is obtained by substracting the extrapolated line from the original curve.[6] Once the activity of each photopeak is obtained, it is only a matter of substitution in Eq. (2) to calculate individually the quantity of each nuclide. However, it should be noted that to apply Eq. (2) geometry and efficiency factors must be experimentally predetermined at the measurement of the individual activity. Also, each irradiation must be accompanied by a simultaneous irradiation of a flux monitor such as a gold foil, a solution of gold, or an aqueous solution of $KMnO_4$. The activity induced by the thermal neutrons is then measured by absolute counting of ^{198}Au or ^{56}Mn. The neutron flux is calculated from Eq. (2).

Now consider another method for quantitative analysis, with its own penalty, that eliminates the inconvenience of the experimental requirements for flux monitoring, efficiency determination, and geometry correction. This method is termed the comperator methods.

It essentially involves the similar treatment of unknowns and standards of similar composition. That is one subjects a known amount of an element to be detected and the unknown sample to the same neutron flux simultaneously; treat both (individually) to identical chemical separation (if any); count each individually with the same geometry. The activities obtained for the unknown and its standard are then to be compared to compared to compute the amount of the unknown element as follows:

$$\begin{array}{l}\text{activity (std)}\\ \text{(dist./sec.)}\end{array} = \left[\frac{N_o \sigma \phi}{A} \, W(\text{std})(1-e^{-\lambda t_a}) e^{-\lambda t_d(\text{std})} \left(\frac{1-e^{-\lambda t_c(\text{std})}}{\lambda}\right) \right] \times GE\Theta Br$$

$$\begin{array}{l}\text{activity (unk)}\\ \text{(dist./sec)}\end{array} = \left[\frac{N_o \sigma \phi}{A} \, W(\text{unk})\ (1-e^{-\lambda t_a}) e^{-\lambda t_d(\text{unk})} \left(\frac{1-e^{-\lambda t_c(\text{unk})}}{\lambda}\right) \right] \times GE\Theta Br$$

$$\frac{\text{activity(std)}}{\text{activity (unk)}} = \frac{(N_o \sigma \phi / A) W(\text{std})(1-e^{-\lambda t_a}) e^{-\lambda t_d(\text{std})} \left(\frac{1-e^{-\lambda t_c(\text{std})}}{\lambda}\right) \times GE\Theta Br}{(N_o \sigma \phi / A) W(\text{unk})\ (1-e^{-\lambda t_a}) e^{-\lambda t_d(\text{unk})} \left(\frac{1-e^{-\lambda t_c(\text{unk})}}{\lambda}\right) \times GE\Theta Br}$$

If $t_d(\text{std}) = t_d(\text{unk})$

$t_c(\text{std}) = t_c(\text{unk})$

then

$$\frac{\text{activity(std)}}{\text{activity (unk)}} = \frac{W(\text{std})}{W(\text{unk})}$$

Therefore,

$$W_{\text{unk}}(\text{grams}) = \frac{[\text{activity(unk)}](W_{\text{std}}(\text{grams})]}{[\text{activity (std)}]} \qquad (3)$$

Thus from equation (3) one observes that quantity of unknown is determined entirely by quantity and activity of standard and ativity of unknown. Therefore, to minimize effects due to variations in flux and self attentuation during irradiation, care must be taken such that the sample and standard are approximately the same weight, shape, thickness, and chemical composition.

4. EXPERIMENTAL

4.1 General Performance

A considerable amount of water, over one liter, was brought directly from the Well Zamzam at Mecca, Saudi Arabia, with special assistance from personnel at charge, and from all the fourty-five Hot Springs, stored collectively in a reservoir located directly behind the city museum, at Hot Springs, Arkansas. The waters were preserved in chemically purified bottles.

Standards were prepared from the standard room of the Activation Analysis Research Center at Texas A&M Univesity. Preparation was performed by repetitive mild heating, to evaporate moisture, before and after weighing until a constant weight value was obtained. Most standards used in this investigation are in compound forms of carbonate, ammonium, or oxide.

Except for the 14-hours irradiation, all samples were prepared in polyethelyne vials. The vials were pretreated with hydrochloric and sulfuric acid, and soaked in demineralized water for over a week. Vials were weighed before and after sample inclusion. Water samples were concentrated, by moderate heating, and then evaporated to dryness by infrared light. For simultaneous irradiation a larger container was used such that the sealed vials of the two water samples and a standard occupy tightly the bottom of the larger container.

After irradiation each product was transferred to a new vial. Water samples and standards were subjected to similar radiochemistry.

Before counting a suitable position from the detector was predetermined such that all counting of samples and standards takes place at the same distance from the detector with less than 10% dead time on the analyzer. Background radiation was minimized by removing nearby radioactive sources and shielding the detector by lead blocks. Alpha and beta radiations were absorbed by a polystyrene absorber of 2 inches thickness.

For gamma radiation measurement, two types of analyzers were used: a 400 multi-channel analyzer with sodium-iodine detector (3x3 inch) and a 1600 channel solid state analyzer with lithium-germanium detector (30 cc). Calibrations of NaI and Li(Ge) detectors, at 10 kev/ch. and 1 kev/ch. respectively, were performed using ^{137}Cs and ^{60}Co sources. The desired counting intervals were preset on the analyzer: all counting took place at clock time(1).

4.2 Separation Techniques

Depending on the half-life of a particular nuclide to be measured, two methods of separation were used: column or filtration.

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[1] Otherwise, consumption of extra time due to the inherent insensitivity of analyzers may not allow to obtain sufficient data points on decay curves for standards and unknowns. However, to apply equation (2) the term $\frac{1}{1-(\%\text{Dead Time})}$ must be added as dead time correction factor: position of samples from detectors were carefully selected to result in dead times less than 10%, in nearly every case, and consequently error resulting from such correction was assumed negligible.

Sodium separation by column technique was achieved by introducing 10 g. of sift (opening = 0.0117) sodium chloride, preheated for three hrs. at 110 ^{0}C, into a 12 inch long column moistened by acetone. One milliliter of water, ten milliliter of acetone, and a few drops (5-10) of conc. HCl were added to the irradiated unknown (or standard). After stirring, the solution was then poured through the column. Sodium free solution was collected in 17 minutes.

Filtration technique was performed by placing NaCl and the mixture solution in a beaker; stirring for 5 minutes; filtering and collecting sodium free solution in 6 minutes. Separation of chlorine was achieved by adding only one drop of HCl to avoid any chlorine hold back effect.

For the separation of potassium, calcium, sulfur, and silicon, the reagent sodium chloride was replaced with potassium chloride, calcium carbonate, zinc sulfide, and silicon respectively(1).

4.3 Specific Performance

The following is a tabulation of the specific performances for the determination of the indicated nuclides:

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[1] Solubility, in gramps per 100 cc (cold water)

NaCl	35.7
KCl	34.7
$CaCo_3$	$1.4x10^{-3}$
ZnS	$6.9x10^{-4}$
Si	Insoluble

Nuclide(s)	Length of Irradiation	Flux $\frac{n}{cm^2\text{-sec}}$	Component(s) to be Separated	Method of Separation	Counting Interval (unkn)	Detector Type
$^{20}F(E_\gamma$ = 1.63 Mev., $t_{1/2}$ = 11 S)	5 sec.	$2x10^{12}$	Na,K,Cl,Ca	Filtration	2 sec.	NaI
$^{37}S(E_\gamma$ = 3.13 Mev., $t_{1/2}$ = 5.1 M)	5 M	$2x10^{12}$	Na,Ca	Filtration	1 M	NaI
$^{49}Ca(E_\gamma$ = 3.1 Mev., $t_{1/2}$ = 8.8 M)	8 M	$2x10^{12}$	Na,S	Filtration	3 M	NaI
$^{27}Mg(E_\gamma$ = 0.84 Mev., $t_{1/2}$ = 9.5 M)	8 M	$2x10^{12}$	Na,K,Si,Mn	Filtration	7 M	Ge(Li)
$^{128}I(E_\gamma$ = 0.455 Mev., $t_{1/2}$= 25 M)	25 M	$2x10^{12}$	Na,Si	Column	25 M	Ge(Li)
$^{38}Cl(E_\gamma$ = 1.64 Mev., $t_{1/2}$= 37.3 M)	20 M	$2x10^{12}$	Na,K	Column	5 M	NaI
$^{56}Mn(E_\gamma$ = 0.845 Mev., $t_{1/2}$=2.58 h) $^{64}Cu(E_\gamma$ = 0.51 Mev., $t_{1/2}$= 12.8 h)	1 h	$2x10^{12}$	Na,K,Si	Column	10 M	Ge(Li)
$^{31}Si(E_\gamma$ = 1.26 Mev., $t_{1/2}$ = 2.62 h)	1 h	$2x10^{12}$	Na,K,Cl	Column	30 M	NaI
$^{42}K(E_\gamma$ = 1.53 Mev., $t_{1/2}$ 12.5 h)	1 h	$2x10^{12}$	Na,Cl	Column	10 M	NaI
$^{24}Na(E_\gamma$ = 1.37 Mev., $t_{1/2}$ = 15 h)	1 h	$2x10^{12}$			20 M	NaI
$^{76}As(E_\gamma$ = 0.555 Mev., $t_{1/2}$ = 1.10 d) $^{82}Br(E_\gamma$ = 0.77 Mev., $t_{1/2}$ = 1.5 d)	14 h	$1.3x10^{3}$	Na,K,Si	Column	6 hrs.	Ge(Li)

5. RESULTS

Experimental quantitative results are summarized in Tables (1-3); the respected spectra are presented in Appendix A. Figure (2) and Table (4) summarize results for experimental verification for the method of separation by Tang & Maletskos (in milligram regions). Figures (3) and (4) present an experimental evaluation of the limit of detection for Mn & Co and I and Mg, respectively.

6. DISCUSSION

Data points of photopeaks presented in Appendix A were synthesized, because of deviation due to the statistical nature of nuclear disintigration, by means of smoothing each data point by fitting a third order polynomial to five channels of data using the method of least squares. Such smoothing was accomplished by the following equation[11]:

$$D_i = \frac{1}{35} \left[-3C_{i-2} + 12C_{i-1} + 17C_{i+1} + 12C_{i+1} - 3C_{i+2} \right]$$

where

C_i = Observed number of counts in channel i
D_i = Smoothed number of counts in channel i

To set the experimental procedure as given on page 15 it was necessary to perform a prelliminary irradiation of short, intermediate, and long irradiation lengths; the qualitative interpretation of the results indicated that calcium and sodium dominated spectra of short and long irradiation lengths respectively.

The isotopic exchange method of Tang & Maletskos claimed to achieve a decontamination factor greater than 10^6. This was tested by preparing a mixture of KBr (1.1×10^{-2} g) and Na_2Co_3 (1.2×10^{-2} g) and irradiating this mixture for one hour with standards of KBr and Na_2Co_3 of the same amounts; the isotopic exchange reagents were NaCl and KCl.

One observes from Figure (2) that potassium peak is not shown before sodium separation, and from Table 4 that about 2% of Bromine(1) was retained or "trapped" in the column and only 84% and 81% of separation of sodium and potassium, respectively, were achieved. On the other hand, a reproducible 90% of separation was obtained for sodium and potassium when the mixture was made up of quantities in micrograms region. A greater percentage of separation was never accomplished. However, the simplicity of the method is attractive specially when the separated nulcide(s) is not to be collected and counted for its own analysis. Furthermore, the method allows one to separate more than one component simultaneously(2). Investigations were carried out first for those elements reported to be found in the Hot Spring sample; among these sodium, potassium, chlorine, and magnesium were determined in both unknowns (except magnesium in Zamzam) with relatively little difficulty. However, fluorine and sulfur determinations presented serious complications.

Though the gamma energies of calcium and sulfur coincide, it was difficult to detect these nuclides simultaneously; this difficulty arose from the fact that, first, calcium is found in Hot Springs sample in a larger amount than sulfur (45 ppm vs. 2.61 ppm). Second, the thermal neutron products of calcium and sulfur are nearly equally short-lived. The first fact results in calcium domination and the need to irradiate a larger amount of the unknowns to detect sulfur; the second fact imposes difficulty on separation length of time. It was found that to separate by means of a column the duration is around 17

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[1] It was concluded then that to reduce trapping additional acetone must be poured through the column.

[2] It should be noticed that group separation including chlorine necessitates the separation of chlorine first then the rest of the group collectively.

minutes. On the other hand, it takes only 6 minutes, but less effectively (80-82% separation), to separate by filtration. Therefore, calcium and sulfur were designed to be determined individually by separating sulfur and calcium respectively.

Large amounts (~25 ml) of the unknowns were, first, irradiated with a sulfur standard and then subjected to calcium and sodium separation by filtration. Analysis of the decay curves of the two unknowns showed calcium presence instead of sulfur. Therefore, it was concluded that larger amounts of unknowns (up to 50 ml) and calcium and sodium separating regents must be employed. To follow decay curve of sulfur standard, though detected, was not possible in this case due to time consumed in the process of unknown and standard separation and in acquiring data from the analyzer (~5 minutes). Therefore, it was concluded that simultaneous irradiation of unknowns and sulfur standard will not result in obtaining sufficient data points to prove sulfur presence in the unknowns and the standard. The experiment to determine sulfur was, then, redesigned, as to irradiate the unknowns and the standard individually within one hour period assuming the flux will remain constant(1) within this length of time. The resulting decay curve was a "composite" curve. It was difficult to resolve such composite curve into its short lived components and consequently only two data points were available to draw the decay curve for sulfur; this is actually statistically problematic.

To determine fluorine, steps similar to sulfur determination were approached. However, fluorine presents a deeper problem due to the fact that only 0.1 ppm of fluorine is reported to be found in Hot Springs water sample. The thermal reactor product of fluorine is F-20 with half-life 11 sec. and gamma energy of 1.63 Mev. To detect fluorine, then, it was necessary to irradiate large amounts of unknowns and

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[1] The validity of this assumption is based on experimental data acquired by the reactor personnel displaying a maximum variation of the flux within one hour period in the vicinity of 1%.

separate in less than one minute, Na,K, Cl, and Ca; such rapid separation is not within the capacity of the method by Tang & Maletskos.

Other methods were searched for and found for the separation of Na, K, Cl, and Ca. When tested on innert samples of unknowns (50 ml), separations were identified by the precipitated characteristic colorful compounds, but time duration was recorded to last over 20 minutes. The method of Tank & Maletskos, then, is simpler and quicker. It is applicable to detect even fluorine provided contaminations are present in microgram quantities; this was proved by subjecting the standard, which was irradiated with the unknowns, to separation by filtration of Na, K, Cl, and Ca contamination. The procedure was speeded up by allowing only 2 minutes to stir the solution and another two to collect contamination free solution. Fluorine was then detected; however, sufficient data was not subjected to separation but allowed for a delay time of only 5 sec. Sufficient data to construct a decay curve was not obtained either; because time interval greater than several fluorine half lives were consumed in acquiring data from the analyzer.

Therefore, to determine fluorine with such high contamination of Na, K, Cl, and Ca, the two unknowns must be treated chemically before irradiation such that newly introduced contamination will not be relevant to the detection of the gamma energy 1.63 Mev. In addition to this, data must be planned to be acquired from a fast output unit, such as magnetic tapes, to facilitate construction of decay curves. There was no access to such highly equipped analyzer near the pneumatic tube.

Investigation was extended to determine elements other than those reported to be found in Hot Springs water; specifically iodine, copper, manganese, arsenic, and bromine were designed to be detected by irradiating about 50 ml of unknowns for 25 minutes, 2 hours, and 14 hours (in glass containers), respectively. In the process of the investigation for Cu-64 and Mn-56, similar peaks were observed for the unknowns which when analyzed qualitatively were found to belong to chlorine and magnesium respectively; delay time for the unknowns were 10 hrs. for ^{64}Cu and 30 M for ^{56}Mn determination. When delay time for Mn-56 detection was increased to 7.5 hrs., the peak disappeared completely. The results of qualitative analysis for the presence of ^{128}I, ^{64}Cu, ^{56}Mn, ^{76}As, and ^{82}Br in the unknown samples were negative, except for ^{82}Br was observed in Zamzam. These results are affirmatively dependable, because the investigation was performed with very few complications as far as time is concerned; that is, the half-lives of nuclides involved are long enough to allow separation of unnecessary contamination by use of columns, to assure maximum efficiency, and acquire data from the slow output unit of the solid state detector, used for its higher resolution property, in "reasonable" time interval (~17 minutes). In addition, about 50 ml. of unknowns were irradiated and counted for a sufficient length of time for the detection of ^{128}I, ^{56}Mn & ^{64}Cu, and ^{76}As & ^{82}Br. However, it is not justifiable especially for Zamzam (for lack of any previous knowledge of contents) do declare that the holy water is free of iodine, copper, manganese, and magnesium. Therefore, the minimum amount detectable was experimentally determined for pure standards of the nuclides ^{128}I, ^{64}Cu, ^{56}Mn, ^{27}Mg.

Figures (3&4) represent an attempt to evaluate the sensitivity of the solid state analyzer employed to detect and identify magnesium, iodine, copper, and manganese: the solid straight lines were obtained by plotting pure standard quantities (in μg) vs. counts (dist./sec)(1). These solid lines were then extended, with dashed lines, to theoretical values for detection limits.(2) The least experimental amounts of standards, that can be detected and identified were, in μg, 8, 8×10^{-3}, 6×10^{-2}, and 2×10^{-4} for ^{27}Mg, ^{128}I, ^{64}Cu, and ^{56}Mn respectively(3).

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[1] The balance used in this investigation allowed to prepare standard only in milligram region; however, microgram amounts were obtained by dilution.

[2] See footnote on page 9.

[3] The corresponding levels of radiation were (in disintegration/sec) 876, 30, 55, and 69.

Basis for this decision is deduced from the fact that these amounts provide enough counts, after time consumption for necessary separation(s), to identify the peaks involved. On the other hand, peaks can be detected but not identified by amounts lesser than those stated above. The conclusion then can be stated as follows:

> For Zamzam and Hot Springs sample (except for ^{27}Mg), the oncentration levels of the nuclides ^{27}Mg, ^{128}I, ^{64}Cu, and ^{56}Mn were, respectively, below (in μg) 8, 8×10^{-3}, 6×10^{-2} and 2×10^{-4} and that these nuclides were not detected either because of their absences or for the shadowing effect caused by some other dominating nuclide(s) the experimenter was not aware of.

In the comparison of Zamzam with Hot Springs Water, the former is heavier and tastes with a unique distinction of "mineral flavor", while the latter, Hot Springs Water, is hot (Ave. 143 ^{0}F), with little distinctive taste at room temperature and contains natural radioactivity (0.81 mc$_{\mu}$c/L). Both contain detectable amounts of sulfur, calcium, chlorine, potassium, silicon, and sodium, while in addition, Zamzam and Hot Springs Water also contain detectable amounts of bromine and magnesium respectively.

The quantitative analysis of the Zamzam sample, as summarized in Table 3, is higher than that for the Hot Springs Water. Such higher concentration of minerals in Zamzam, then, must be accredited for its distinctive taste.

6.1 Error Analysis

Inherent error sources in Activation Analysis, among others, are
1) error in sample and standard weighing
2) error from flux variation
3) error arising from the process of sample container interchanging
4) fractional loss of sample in the process of separation technique
5) statistical error.

The comparator method employed in this investigation advantageously narrowed the error sources into two: Error in sample and standard weighing Statistical error. That is, errors from fractional losses in the process of container interchanging and separation technique and error from flux variation can be neglected due to the equal treatment of unknowns and standards. Error from statistical fluctuation were found to be negligible fraction of total error which can be attributed mainly to balance uncertainty in standards, unknown measurement, (±0.003) units, and moisture correction to standards weights, (±0.001) units. Finally, the concentrations of Hot Springs components obtained in this investigation are, in nearly every case, lower than the published ones: (in part per million) 2.61 vs. 2, 45 vs. 44, 4.9 vs. 5, 2 vs. 2, 21 vs. 20, 1.6 vs. 1, and 4vs. 4, for ^{37}S, ^{49}Ca, ^{27}Mg, ^{38}Cl, ^{31}Si, ^{42}K, and ^{24}Na respectively.

7. SUMMARY

Activation analysis provides no information concerning the chemical structural forms of elements found in Zamzam, however, this investigation has served its main purpose: the determination of trace elements of Zamzam in comparison to that of Hot Springs Water.

It is evident from the high concentration of the elements discovered so far that other trace elements exist possible in much lesser concentrations; therefore it will be necessary, for further investigation, to remove these highly concentrated elements preferably before irradiation (if possible).

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Sample	Wt. of Sample (mg)	Nuclear Reaction	Thermal absorp. Cross Section (Reactant) (barn)	Weight of Reactant Element (mg)	Half Life (Product)	Gamma Energy Readily Detectable (Mev)	Source Detector Distance (Inch)	% Dead Time	Delay Time	Counting Time	Peak Area	Decay correction factor during delay time	Decay correction factor during counting	Distingration per second
NH_4F	$6.6x10^{-1}$	$^{19}_{9}F(n,\gamma)^{20}_{9}F$	0.010	$3.3x10^{-1}$	11 sec	1.65	0	2	5 sec	2 sec	271	1.37	0.53	$2x10^{2}$
$(NH_4)_2SO_4$	4.8	$^{36}_{16}S(n,\gamma)^{37}_{16}S$	0.14	1.3	5.1 M	3.09	8	5	5 M	3 M	6663	1.97	0.41	$5.6x10^{3}$
$CaCo_3$	4.2	$^{48}_{20}Ca(n,\gamma)^{49}_{20}Ca$	1.10	1.8	8.8 M	3.10	10	4	40 M	3 M	4682	23.25	0.37	$4.2x10^{4}$
MgO	$2.2x10^{-1}$	$^{26}_{12}Mg(n,\gamma)^{27}_{12}Mg$	0.03	$1.1x10^{-1}$	9.5 M	0.842	4	4	7 M	7 M	16813	1.66	0.18	$5.3x10^{3}$
NH_4I	$3.4x10^{-1}$	$^{127}_{53}I(n,\gamma)^{128}_{53}I$	6.2	$2.8x10^{-1}$	25 M	0.441	6	4	219 M	25 M	2308	4.26	0.06	$5.7x10^{4}$
NH_4Cl	$3.9x10^{-1}$	$^{37}_{17}Cl(n,\gamma)^{38}_{17}$	0.43	$2.5x10^{-1}$	37.3 M	1.60	8	4	40 M	5 M	16854	2.10	1.00	$7.6x10^{3}$
$MnCo_3$	$4.5x10^{-1}$	$^{55}_{25}Mn(n,\gamma)^{56}_{25}Mn$	13.3	$1.9x10^{-1}$	2.58 h	0.847	10	8	11.33 hrs	30 M	2471	20.04	2.14	$1.2x10^{5}$
Silicon	1.1	$^{30}_{14}Si(m,\gamma)^{31}_{14}Si$	0.11	1.1	2.62 h	1.27	3	4	3 hrs	20 M	916	2.20	3.14	$6.6x10^{3}$
K_2Co_3	1.1	$^{41}_{19}K(n,\gamma)^{42}_{19}K$	1.1	$6.4x10^{-1}$	12.5 h	1.52	2	4	10 hrs	5 M	6412	1.73	12.06	$1.4x10^{5}$
$NaCo_3$	$2.7x10^{-1}$	$^{23}_{11}Na(n,\gamma)^{24}_{11}Na$	0.53	$1.1x10^{-1}$	15 h	1.368	11	15	90 M	5 M	32099	1.07	33.24	$1.3x10^{6}$
Cu_2O	2.1	$^{63}_{29}Cu(n,\gamma)^{64}_{29}Cu$	4.5	1.8	12.8 h	0.511	11	7	25.5 hrs	10 M	168428	3.97	6.22	$4.5x10^{6}$
As_2O_5	$4.1x10^{-2}$	$^{75}_{33}As(n,\gamma)^{76}_{33}As$	4.5	$2.6x10^{-1}$	1.1 d	0.559	10	3	3.2d	6 hrs	221657	7.46	4.47	$7.6x10^{6}$
NH_4Br	$1.5x10^{-1}$	$^{81}_{35}Br(n,\gamma)^{82}_{35}Br$	3.2	$1.1x10^{-1}$	1.5 d	0.554	10	4	3.49d	6 hrs	290766	4.98	4.24	$6.4x10^{6}$

TABLE 1: STANDARDS

Nuclear Reaction	Ther. Cross Section (barn)	Wt. of unknown (liquid) (g)	Half Life (Product)	Gamma Energy (Mev)	Source Detector Dist.	% Dead Time	Delay Time	Counting Time	Peak Area	Decay correct. factor during delay	Decay correct. factor during counting	Disintegration per sec.	Parts per Million
$^{36}_{16}S(n,\gamma)^{37}_{16}S$	0.010	44.125	5.1 M	3.09	8	4	5 M	1 M	199	1.97	1.07	4.4×10^{2}	2
$^{48}_{20}Ca(n,\gamma)^{49}_{20}Ca$	1.10	8.825	8.8 M	3.10	10	4	20 M	3 M	3699	4.80	0.37	6.9×10^{3}	44
$^{26}_{12}Mg(n,\gamma)^{27}_{12}Mg$	0.03	22.062	9.5 M	0.842	4	4	25 M	7 M	4158	6.17	0.18	4.9×10^{3}	5
$^{37}_{17}Cl(n,\gamma)^{38}_{17}Cl$	0.43	4.412	37.3 M	1.60	8	10	20 M	5 M	596	1.44	0.20	2.0×10^{2}	2
$^{30}_{14}Si(n,\gamma)^{31}_{14}Si$	0.11	26.475	2.6 h	1.27	3	4	40 M	30 M	1186	1.19	2.14	3.2×10^{3}	20
$^{23}_{11}Na(n,\gamma)^{24}_{11}Na$	0.53	4.412	15 h	1.368	11	8	19.5 M	20 M	59897	1.01	3.03	2.0×10^{5}	4
$^{41}_{19}K(n,\gamma)^{42}_{19}K$	1.1	22.062	12.5 h	1.52	2	5	8 hrs	10 M	574	1.55	6.02	5.7×10^{3}	1

TABLE 2: HOT SPRINGS SAMPLE

Nuclear Reaction	Thermal abs. Cross Sect. (barn)	Wt. of un-known (liquid) (g)	Half Life (Product)	Gamma Energy (Mev)	Source Det. Dist. (Inch)	% Dead Time	Delay Time	Counting Time	Peak Area	Decay Correct. factor during delay	Decay Correct. factor during counting	Dist. Per Second	Parts Per Million
${}^{36}_{16}S(n,\gamma){}^{37}_{16}S$	0.010	46.115	5.1 M	3.09	8	7	5 M	1 M	230	1.97	1.07	5.2×10^{2}	3
${}^{48}_{20}Ca(n,\gamma){}^{49}_{20}Ca$	1.10	9.922	8.8 M	3.10	10	5	28 M	3 M	6999	9.09	0.37	2.5×10^{4}	107
${}^{37}_{17}Cl(n,\gamma){}^{38}_{17}Cl$	0.43	4.611	37.3 M	1.60	8	12	29 M	5 M	3900	1.71	0.21	1.6×10^{3}	11
${}^{36}_{14}Si(n,\gamma){}^{37}_{14}Si$	0.11	6.616	2.62 h	1.27	3	7	80 M	30 M	594	1.42	2.14	1.9×10^{3}	12
${}^{41}_{19}K(n,\gamma){}^{42}_{19}K$	1.1	23.057	12.5 h	1.52	2	5	8.66hrs	10 M	2116	1.61	6.02	2.2×10^{4}	4
${}^{23}_{11}Na(n,\gamma){}^{24}_{11}Na$	0.53	4.611	15 h	1.368	11	28	41.5 M	20 M	180392	1.03	3.03	7.8×10^{5}	14
${}^{81}_{35}Br(n,\gamma){}^{82}_{35}Br$	3.2	46.115	1.5 d	0.554	10	4	2.5 d	6 hrs	1621542	3.16	4.24	2.7×10^{7}	9

TABLE 3: ZAMZAM SAMPLE

Table 4

Results of experimental testings of the method Tang & Maletskos of separation (in milligrams region):

Mixture: Na_2Co_3 + KBr
Separating Reagents: NaCl & KCl
Irradiation Time: 1 hr
Delay Time: 2 M
Separation Time: 17 M

Activity of ^{24}Na (1.37 Mev) in mixture before separation = 8001
Activity of ^{24}Na (1.37 Mev) standard of the same quant. = 8102
Activity of ^{24}Na (1.37 Mev) in mixture after separation = 1267
Activity of ^{24}Na (1.37 Mev) standard = 7853
% of ^{24}Na separation = 84
Activity of ^{42}K (1.53 Mev) in mixture before sep. = -
Activity of ^{42}K (1.53 Mev) standard (same quantity) = 880
Activity of ^{42}K (1.53 Mev) in mixture after sep. = 160
Activity of ^{42}K (1.53 Mev) standard = 842
% of ^{42}K separation = 81
Activity of ^{82}Br (0.55 Mev) before separation = 14480
Activity of ^{82}Br (0.55 Mev) standard = 14473
Activity of ^{82}Br (0.55 Mev) standard = 14197
% loss of ^{82}Br = 2

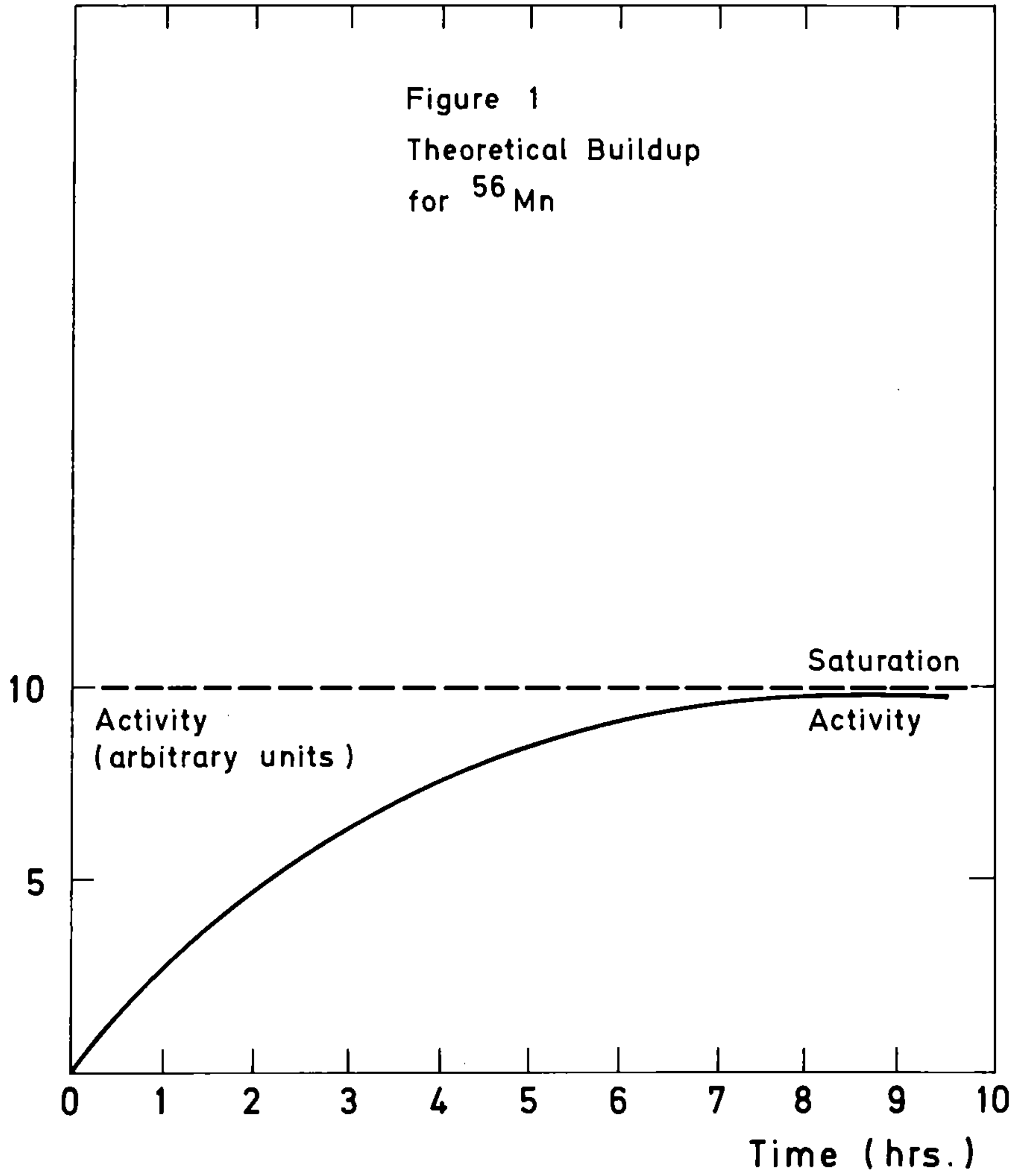

Figure 1
Theoretical Buildup
for ^{56}Mn
Saturation
Activity
10
5
Activity
(arbitrary units)
0
1
2
3
4
5
6
7
8
9
10
Time (hrs.)

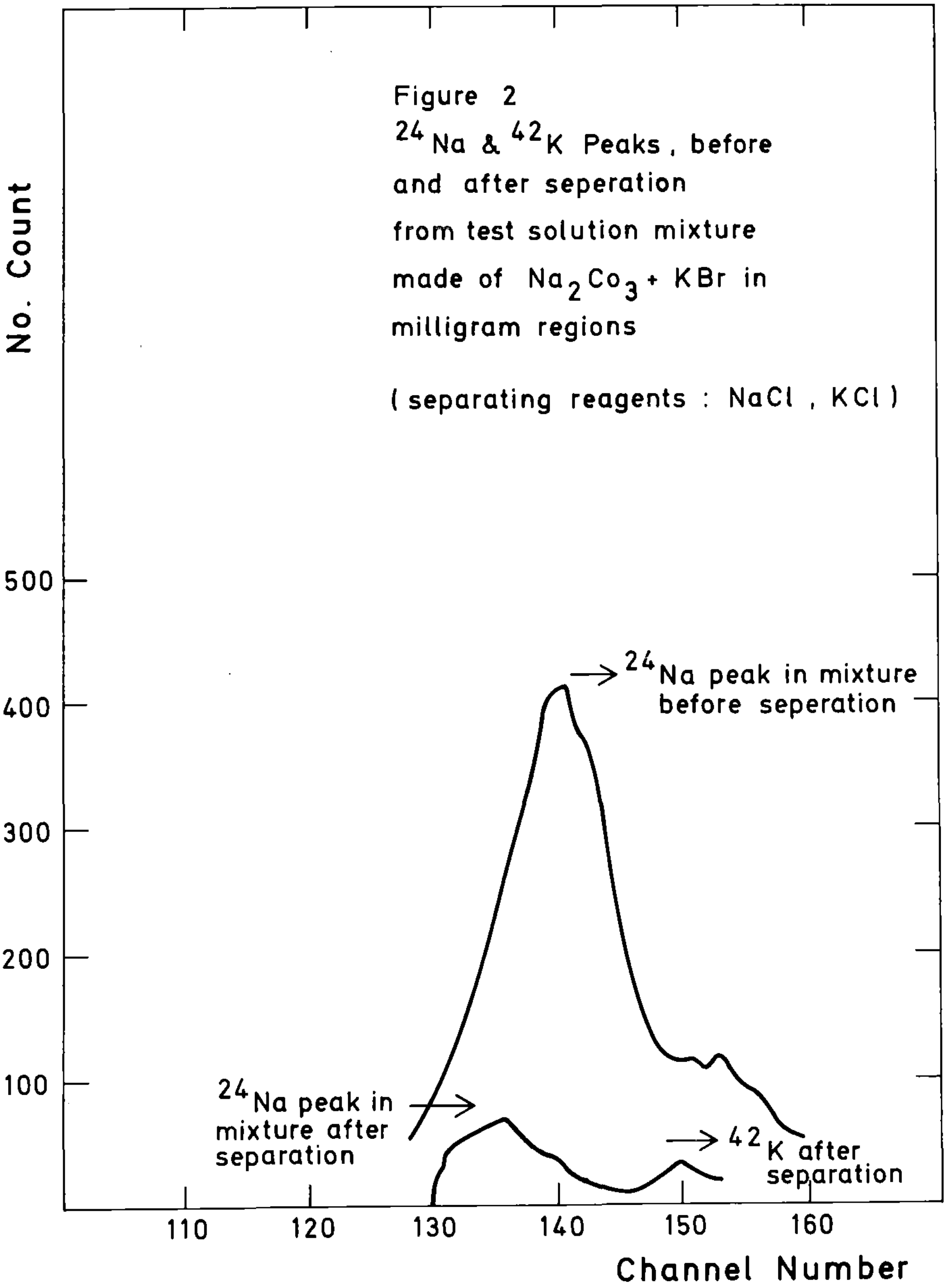

Figure 2
^{24}Na & ^{42}K Peaks, before and after seperation from test solution mixture made of Na_2Co_3 + KBr in milligram regions

(separating reagents : NaCl , KCl)

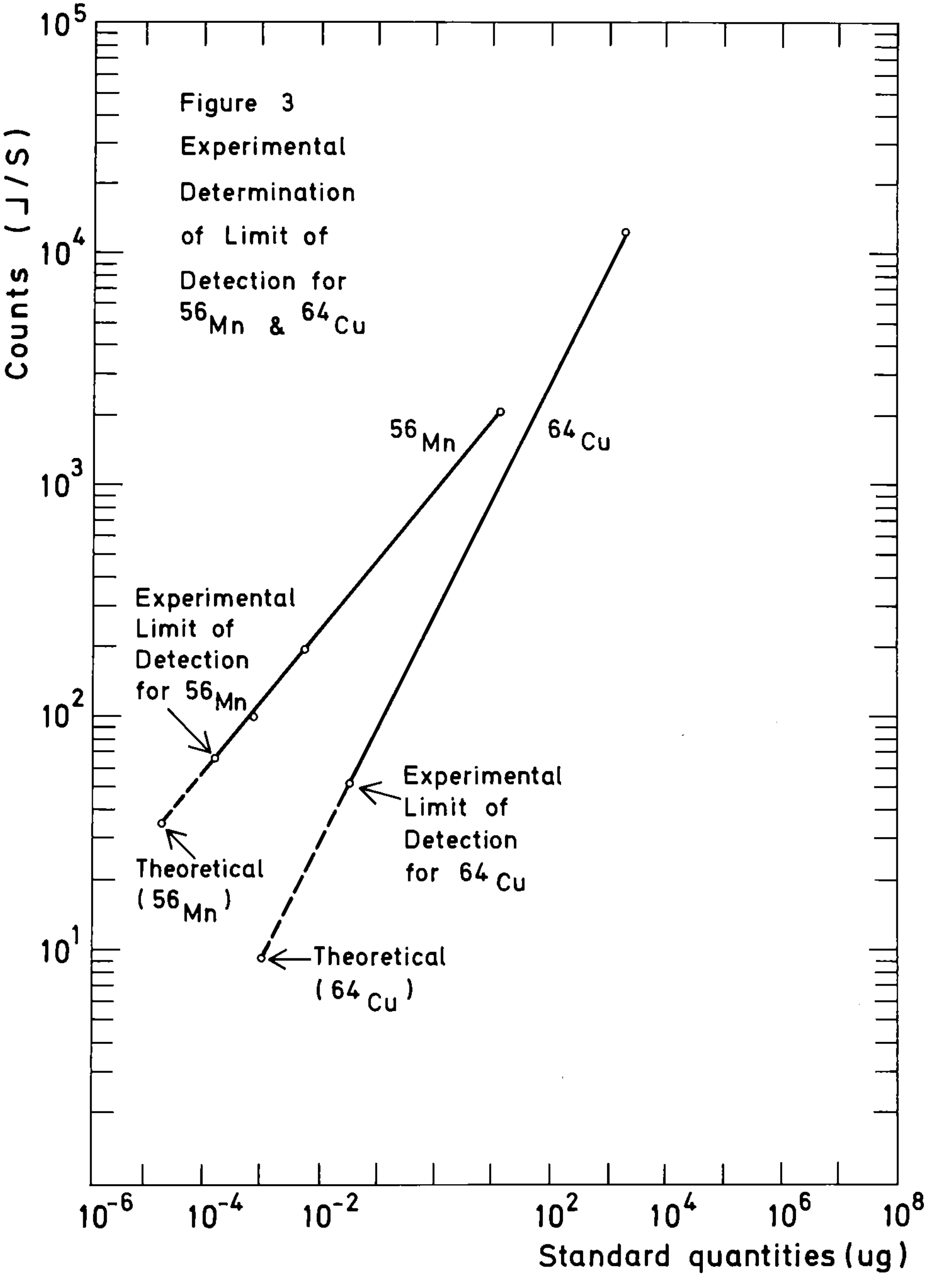

Figure 3 Experimental Determination of Limit of Detection for ^{56}Mn & ^{64}Cu

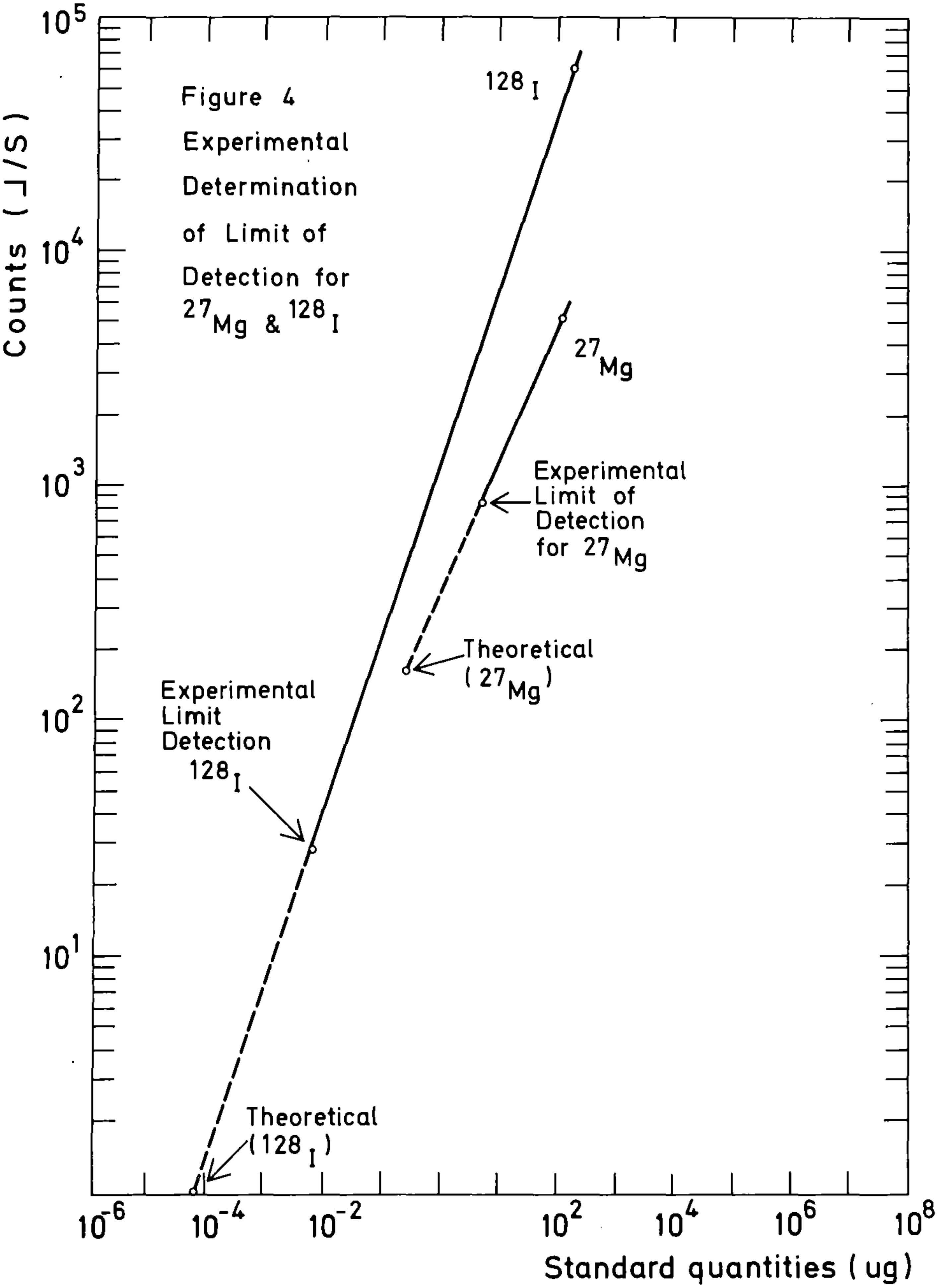

Figure 4 Experimental Determination of Limit of Detection for ^{27}Mg & ^{128}I

APPENDIX A

Spectra for standards of unknowns are diagrammed in the following pages:

Figures (5-16) present spectra for reactor products of standards:

^{37}S, ^{49}Ca, ^{27}Mg, ^{128}I, ^{38}Cl, ^{56}Mn,
^{31}Si, ^{42}K, ^{64}Cu, ^{24}Na, ^{76}As, ^{82}Br

Figures (17-25) present spectra for reactor products of Hot Springs Water sample:

^{37}S, ^{49}Ca, ^{27}Mg, ^{38}Cl, ^{31}Si, ^{42}K, ^{24}Na

Figures (26-33) present spectra for reactor products of Zamzam sample:

^{37}S, ^{49}Ca, ^{38}Cl, ^{31}Si, ^{42}K, ^{24}Na, ^{82}Br

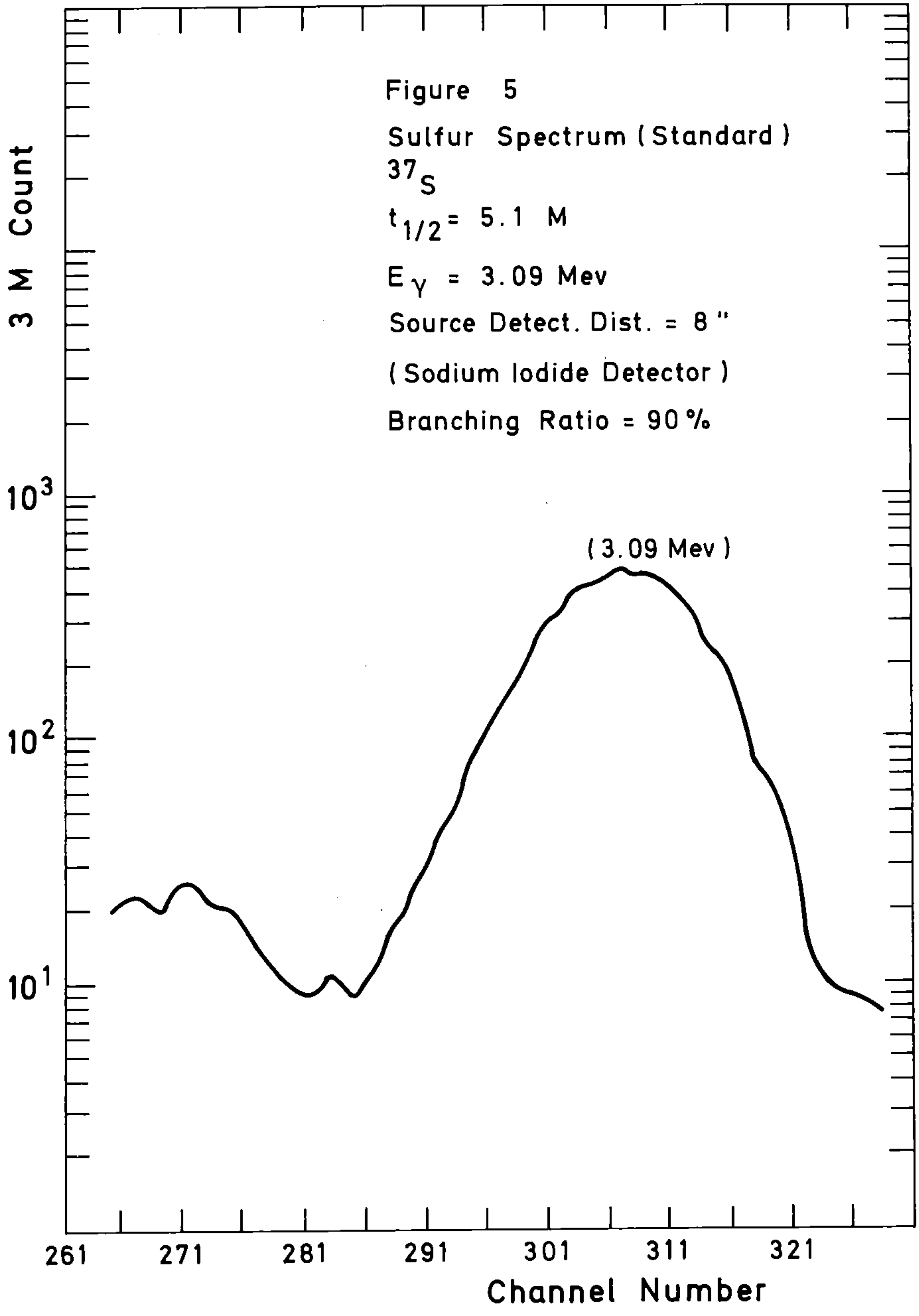

Figure 5

Sulfur Spectrum (Standard)

^{37}S

$t_{1/2}$ = 5.1 M

E_γ = 3.09 Mev

Source Detect. Dist. = 8"

(Sodium Iodide Detector)

Branching Ratio = 90%

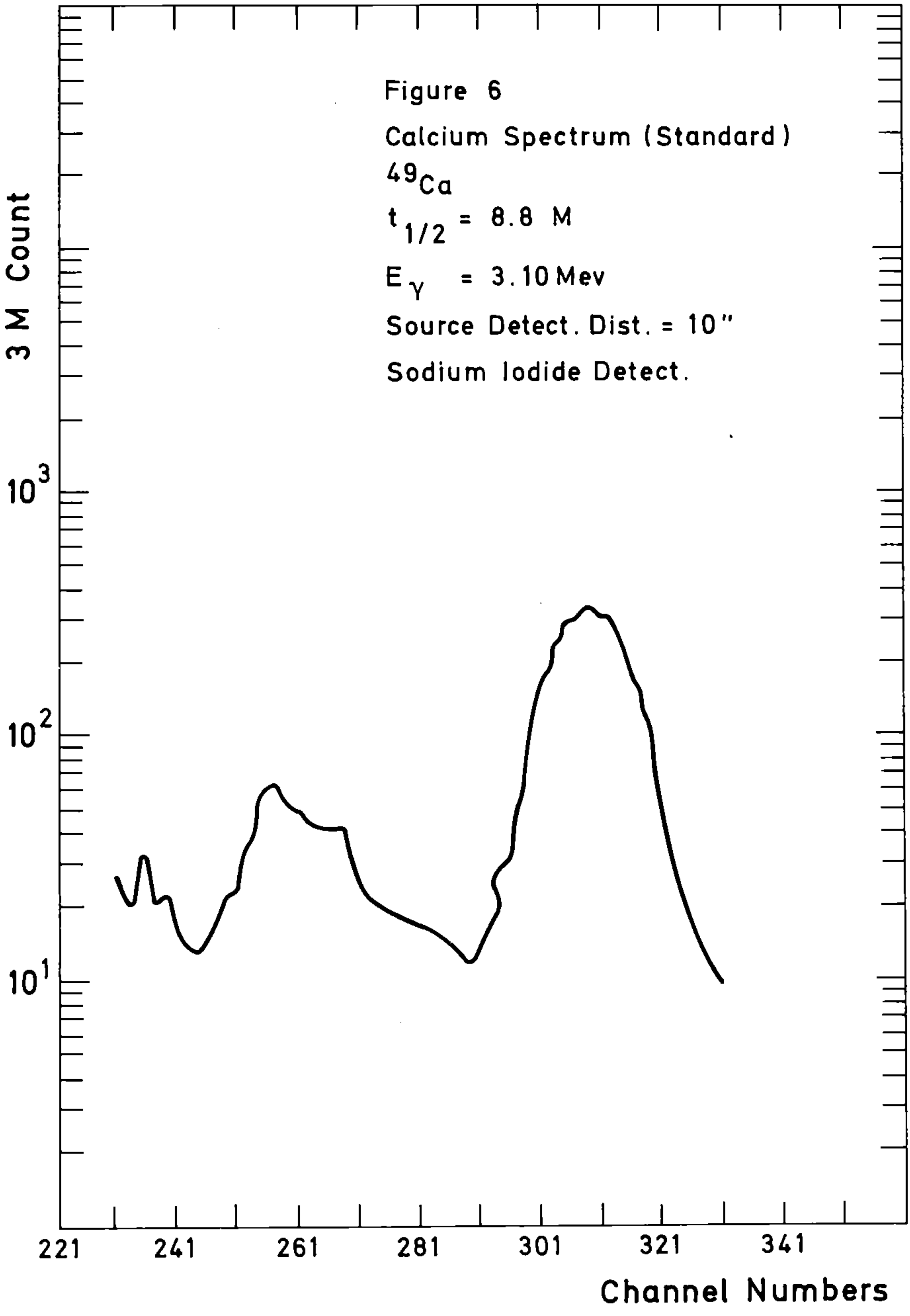

Figure 6

Calcium Spectrum (Standard)

^{49}Ca

$t_{1/2}$ = 8.8 M

E_{γ} = 3.10 Mev

Source Detect. Dist. = 10"

Sodium Iodide Detect.

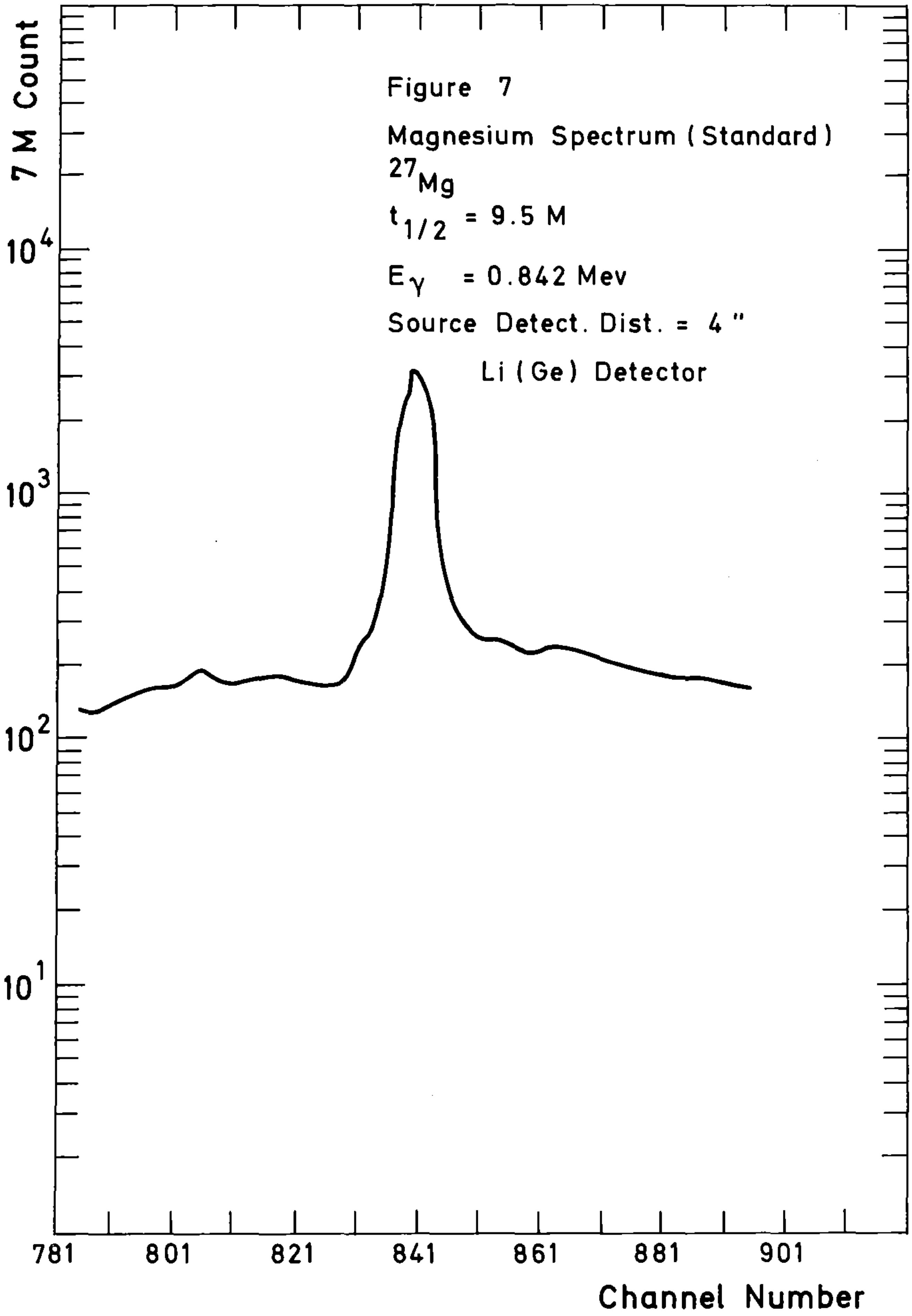

Figure 7

Magnesium Spectrum (Standard)

^{27}Mg

$t_{1/2} = 9.5$ M

$E_\gamma = 0.842$ Mev

Source Detect. Dist. = 4"

Li (Ge) Detector

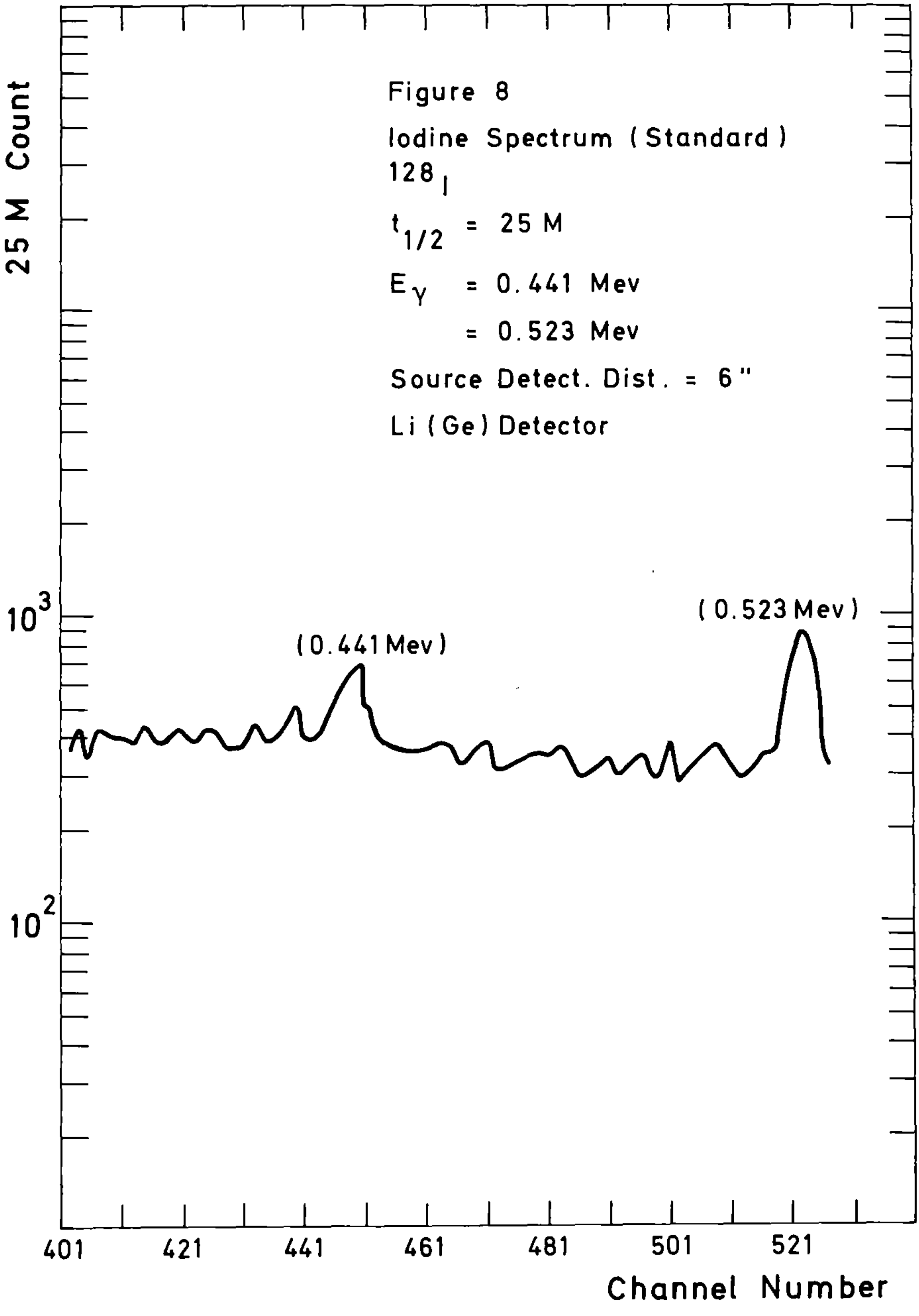

Figure 8

Iodine Spectrum (Standard)

^{128}I

$t_{1/2}$ = 25 M

E_{γ} = 0.441 Mev

= 0.523 Mev

Source Detect. Dist. = 6"

Li (Ge) Detector

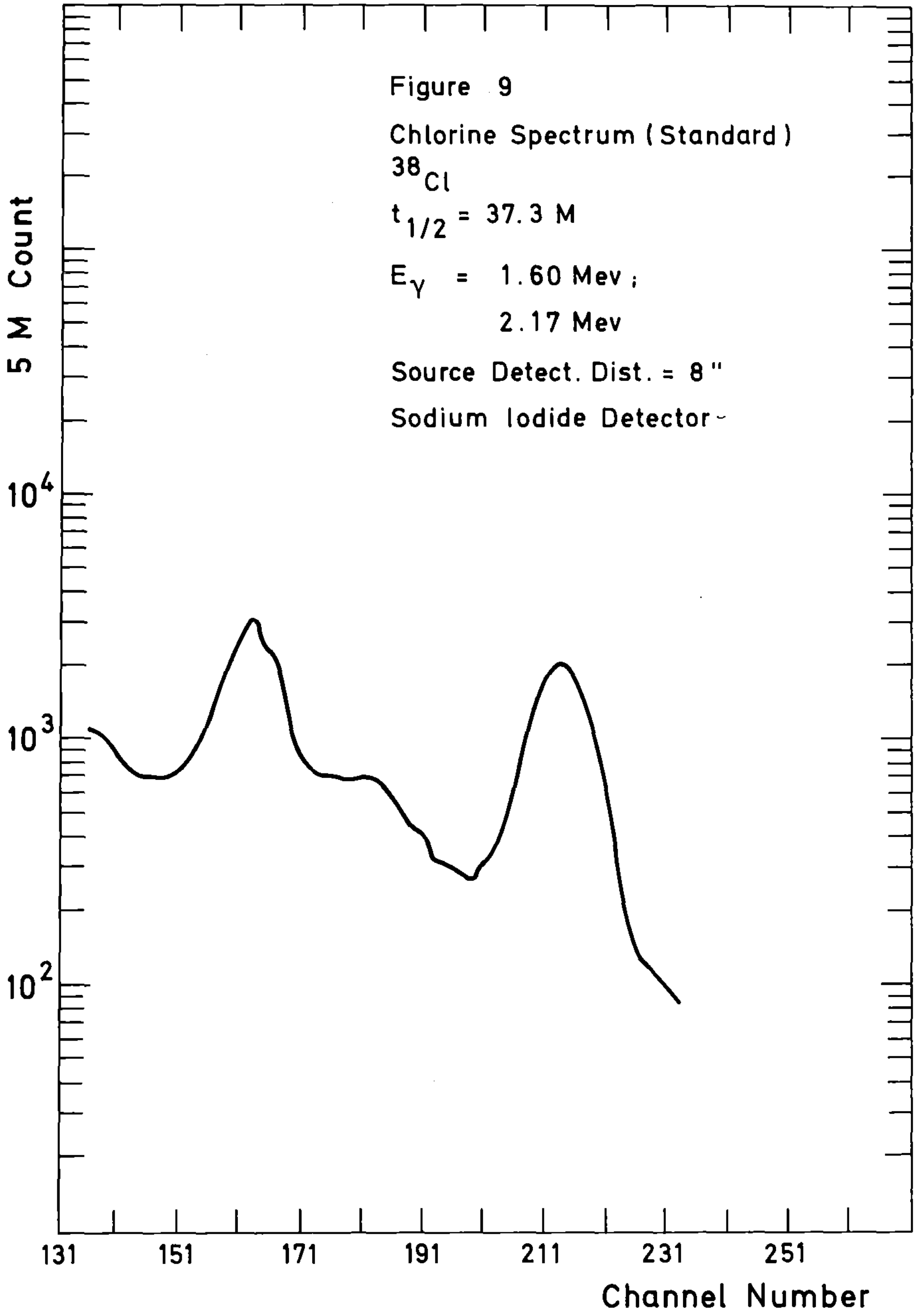

Figure 9

Chlorine Spectrum (Standard)

^{38}Cl

$t_{1/2}$ = 37.3 M

E_γ = 1.60 Mev ;
2.17 Mev

Source Detect. Dist. = 8"

Sodium Iodide Detector

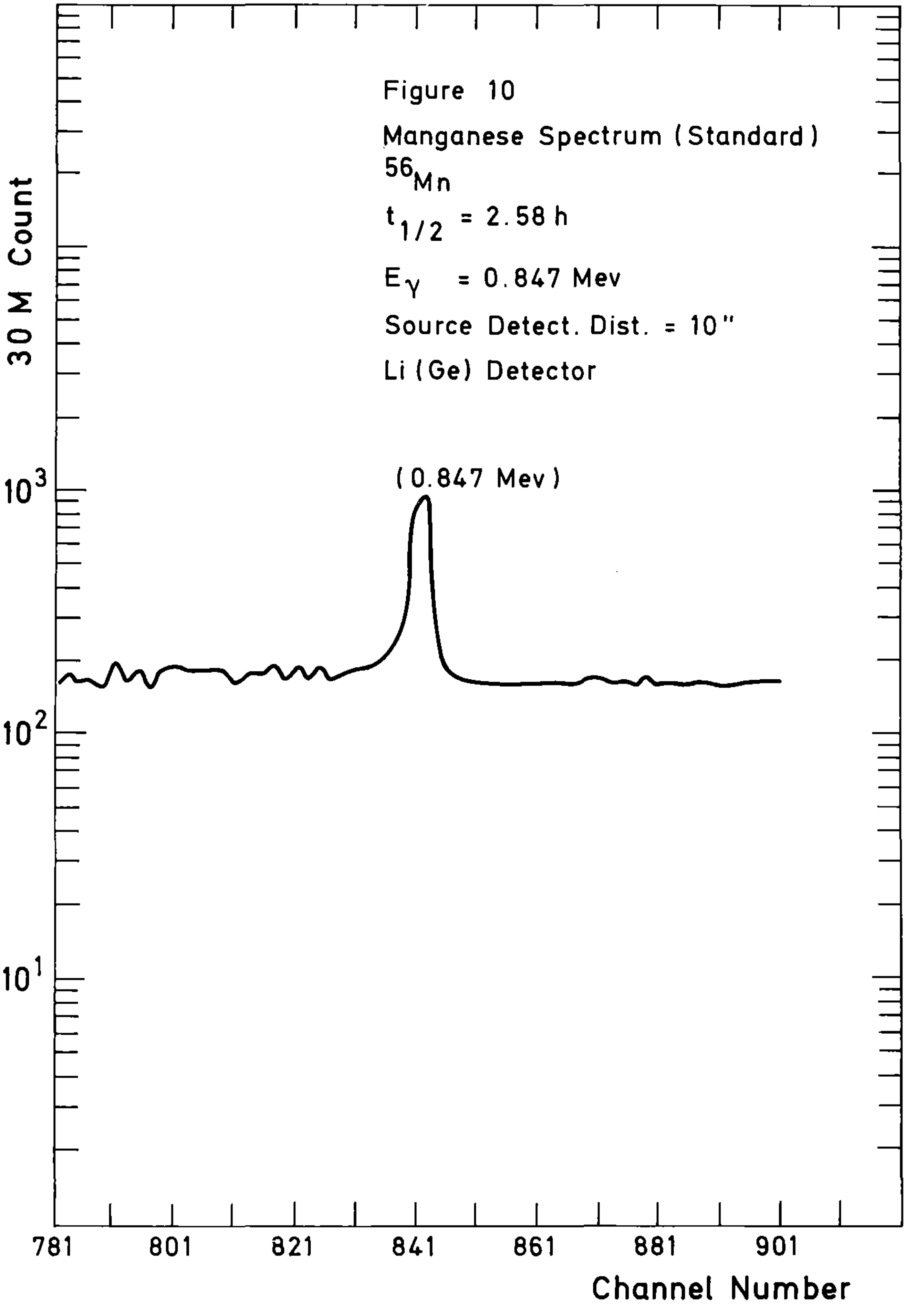

Figure 10

Manganese Spectrum (Standard)

^{56}Mn

$t_{1/2}$ = 2.58 h

E_γ = 0.847 Mev

Source Detect. Dist. = 10"

Li (Ge) Detector

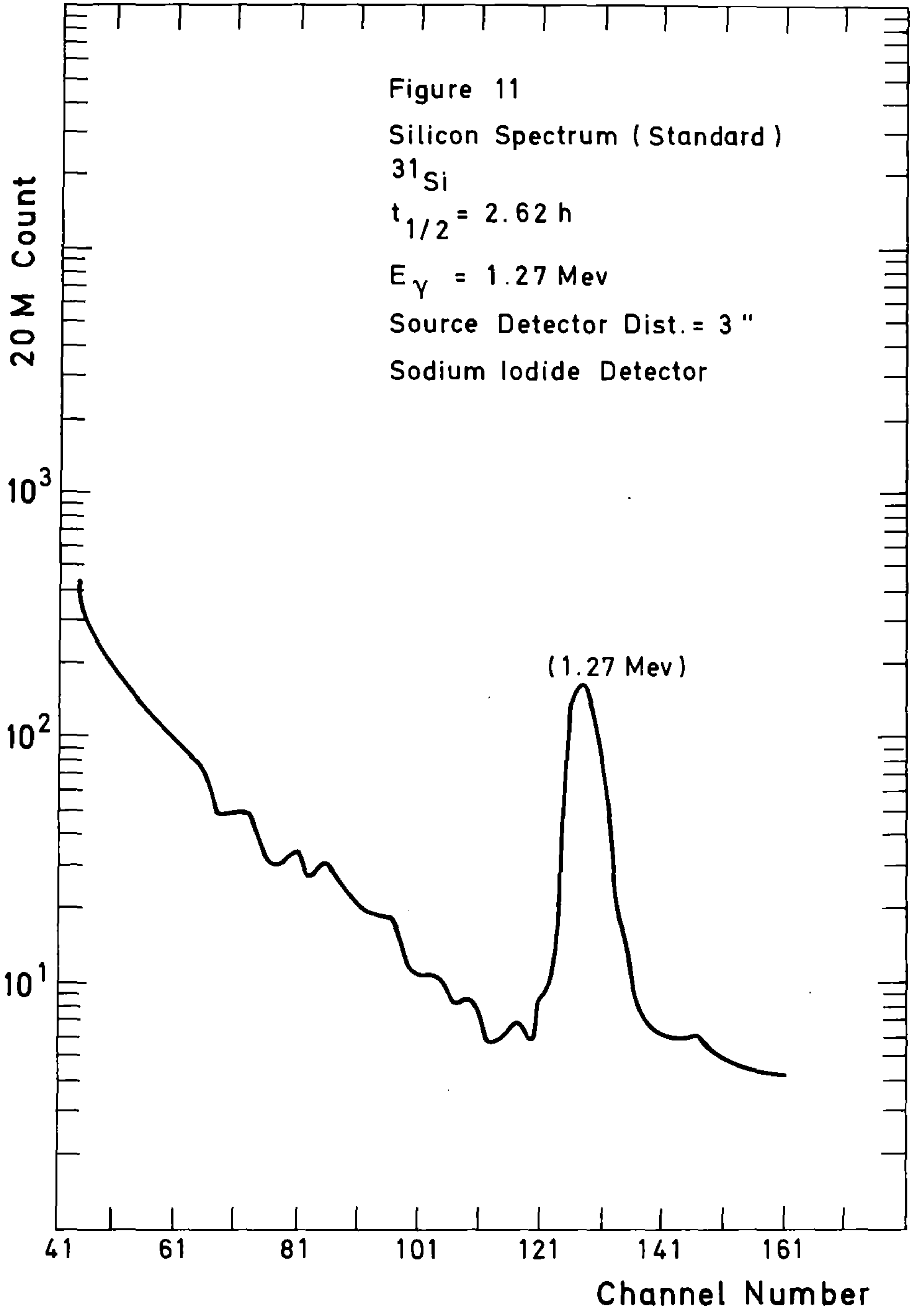

Figure 11

Silicon Spectrum (Standard)

^{31}Si

$t_{1/2} = 2.62$ h

$E_\gamma = 1.27$ Mev

Source Detector Dist. = 3 "

Sodium Iodide Detector

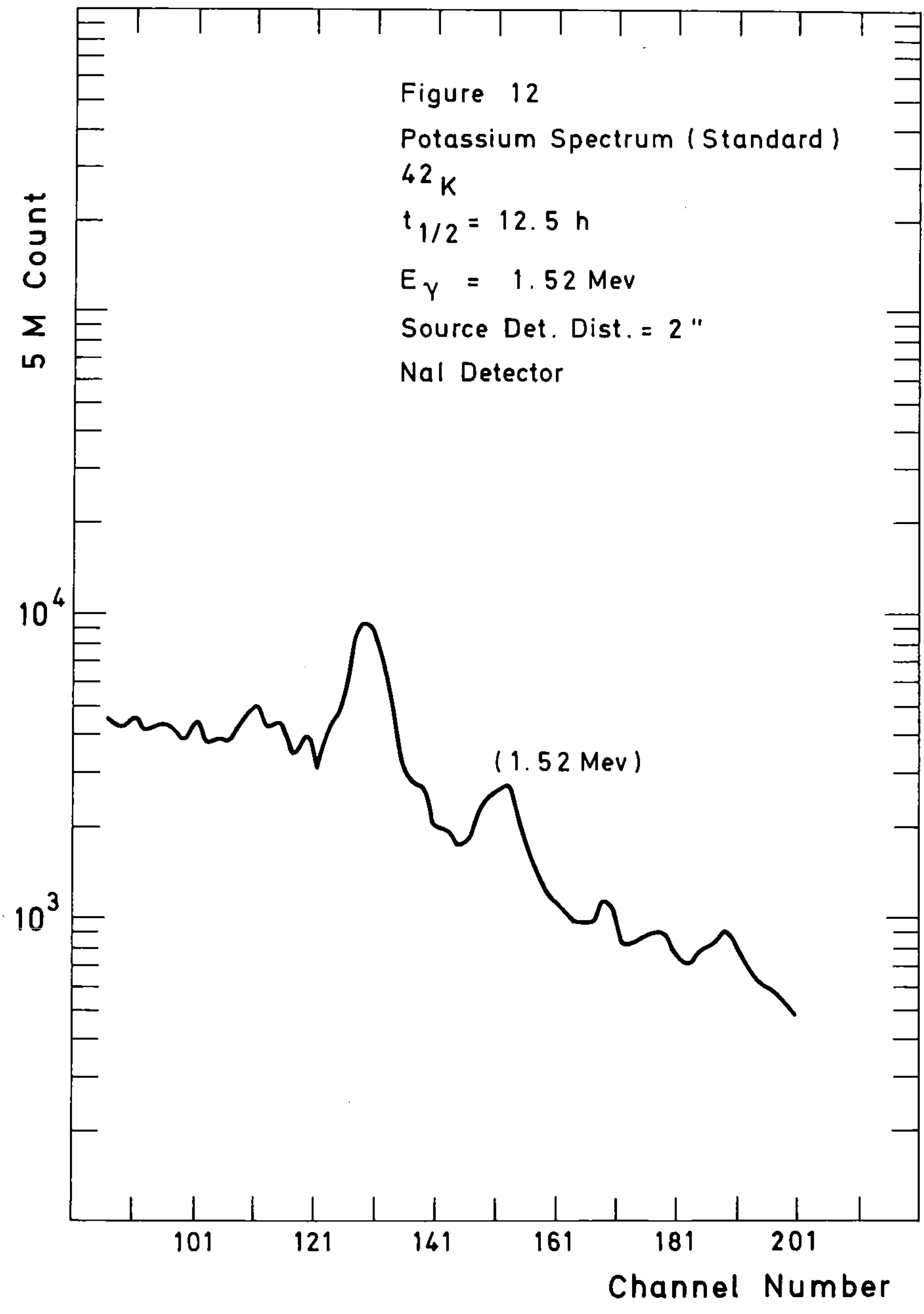

Figure 12

Potassium Spectrum (Standard)

^{42}K

$t_{1/2}$ = 12.5 h

E_γ = 1.52 Mev

Source Det. Dist. = 2"

NaI Detector

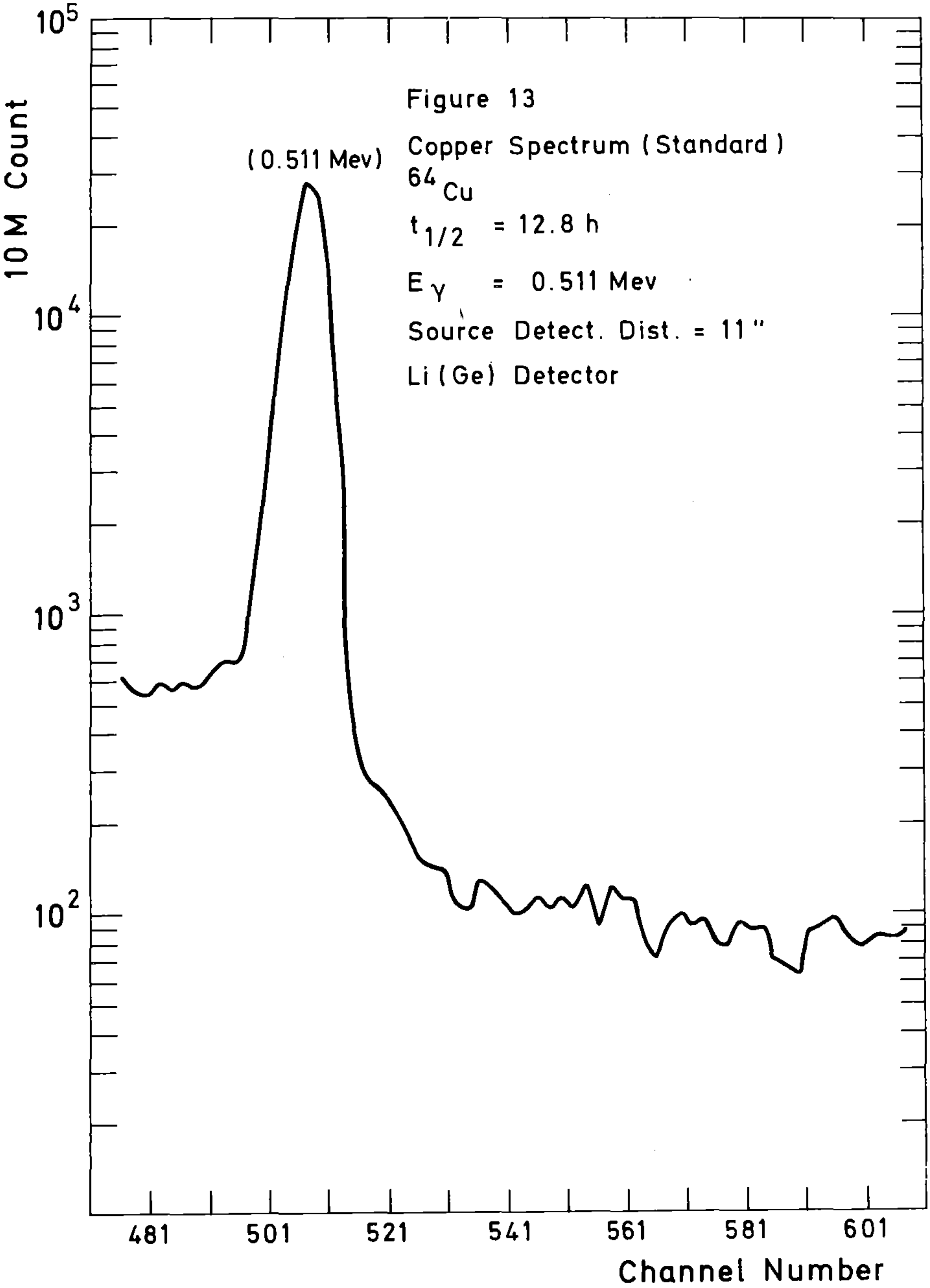

Figure 13

Copper Spectrum (Standard)

^{64}Cu

$t_{1/2}$ = 12.8 h

E_γ = 0.511 Mev

Source Detect. Dist. = 11"

Li (Ge) Detector

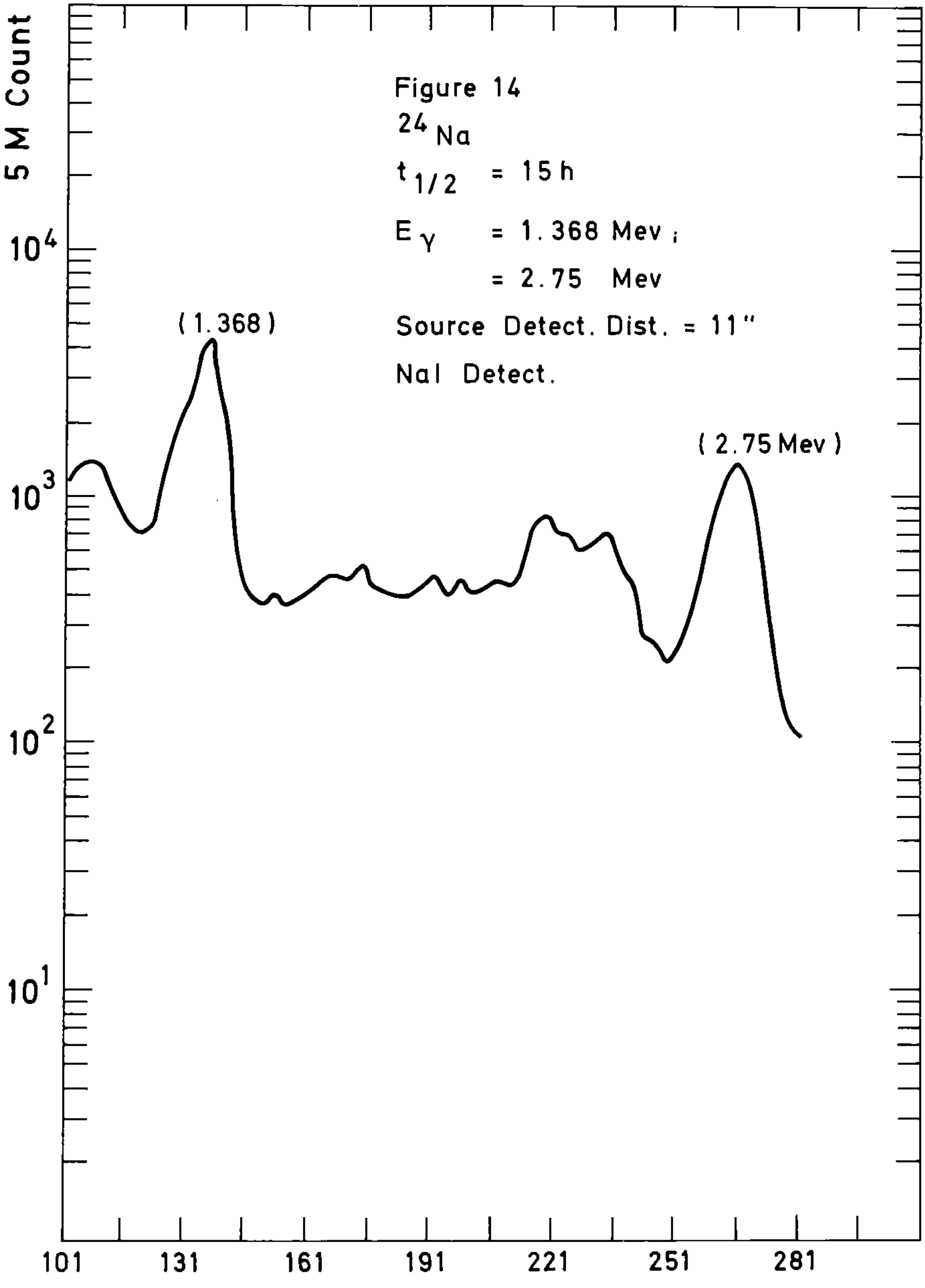

Figure 14
^{24}Na
$t_{1/2}$ = 15 h
E_γ = 1.368 Mev ;
= 2.75 Mev
Source Detect. Dist. = 11"
NaI Detect.

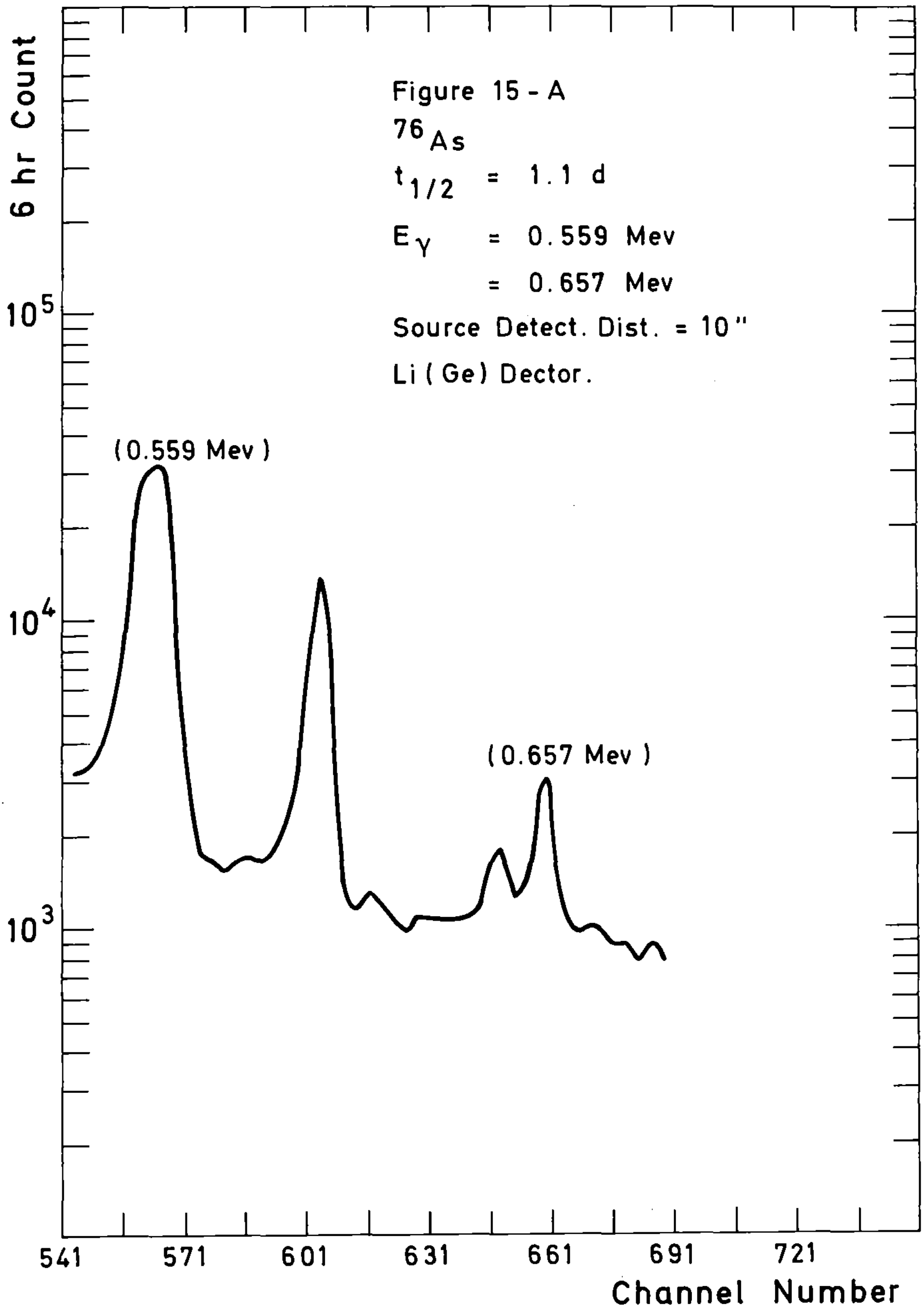
Figure 15 - A
^{76}As
$t_{1/2}$ = 1.1 d
E_γ = 0.559 Mev
= 0.657 Mev
Source Detect. Dist. = 10"
Li (Ge) Dector.
(0.559 Mev)
(0.657 Mev)
6 hr Count
10^5
10^4
10^3
541
571
601
631
661
691
721
Channel Number

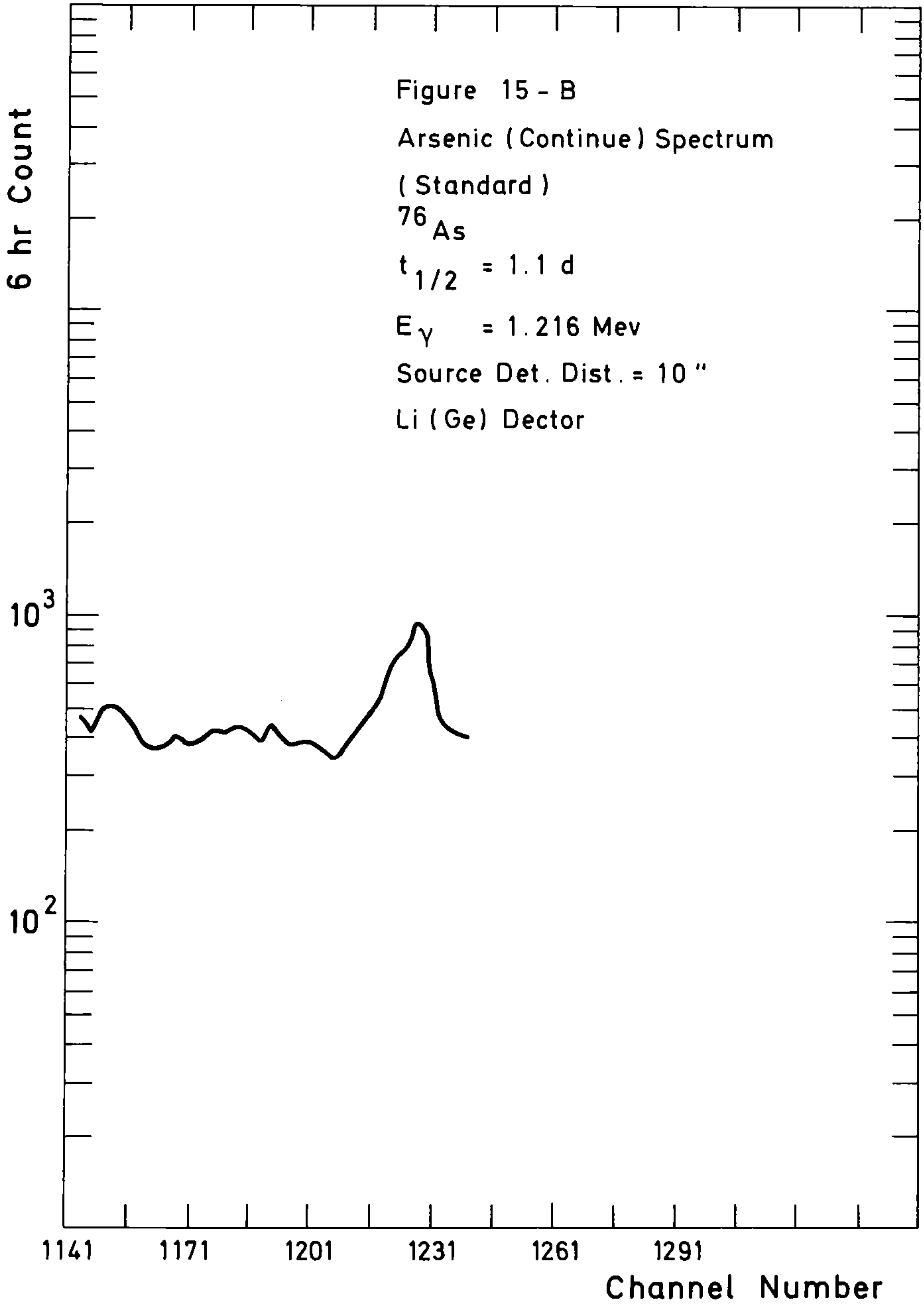
Figure 15 - B
Arsenic (Continue) Spectrum
(Standard)
76 As
t 1/2 = 1.1 d
E γ = 1.216 Mev
Source Det. Dist. = 10"
Li (Ge) Dector
6 hr Count
10^3
10^2
1141
1171
1201
1231
1261
1291
Channel Number

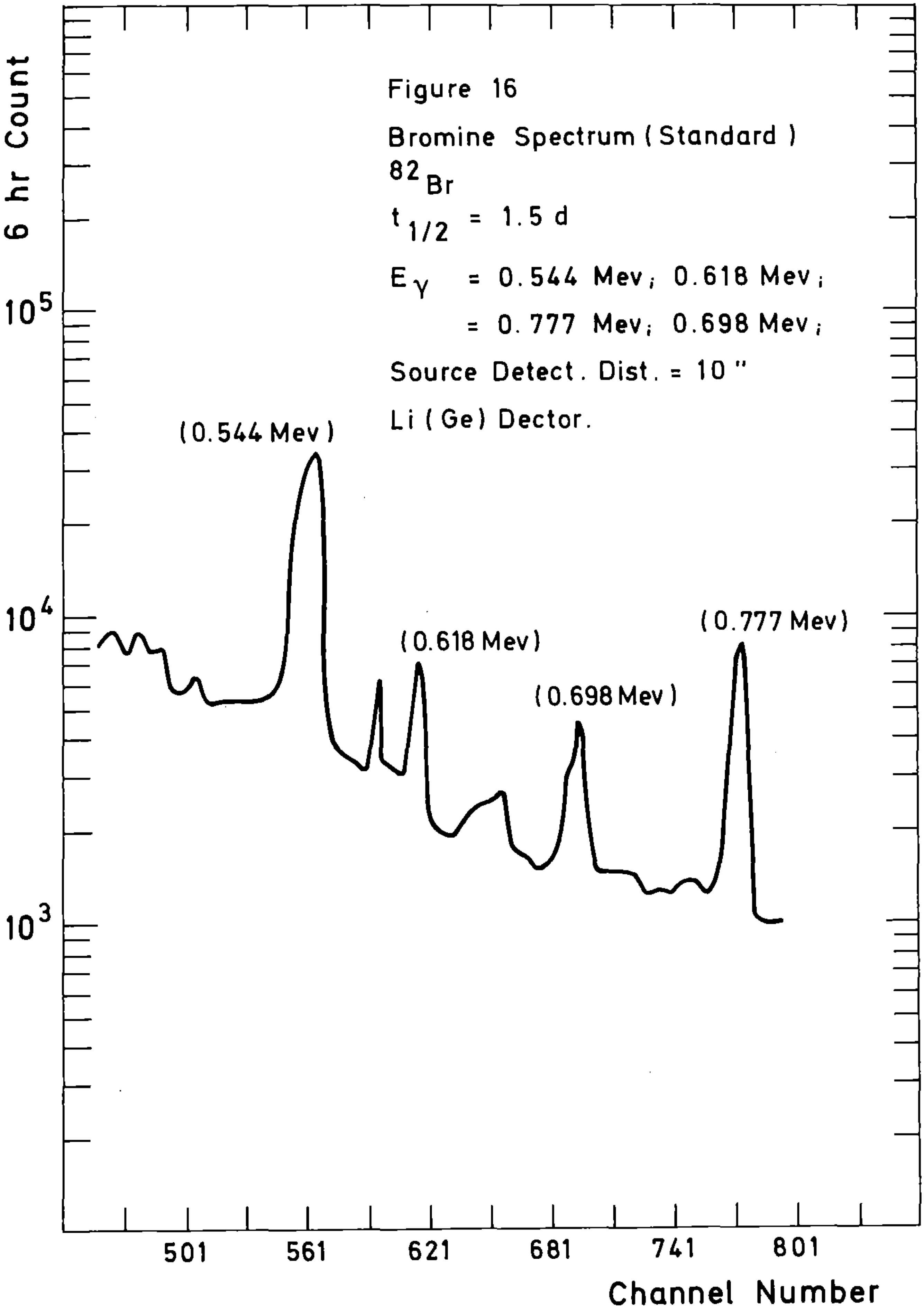

Figure 16

Bromine Spectrum (Standard)

^{82}Br

$t_{1/2}$ = 1.5 d

E_γ = 0.544 Mev; 0.618 Mev;

= 0.777 Mev; 0.698 Mev;

Source Detect. Dist. = 10"

Li (Ge) Dector.

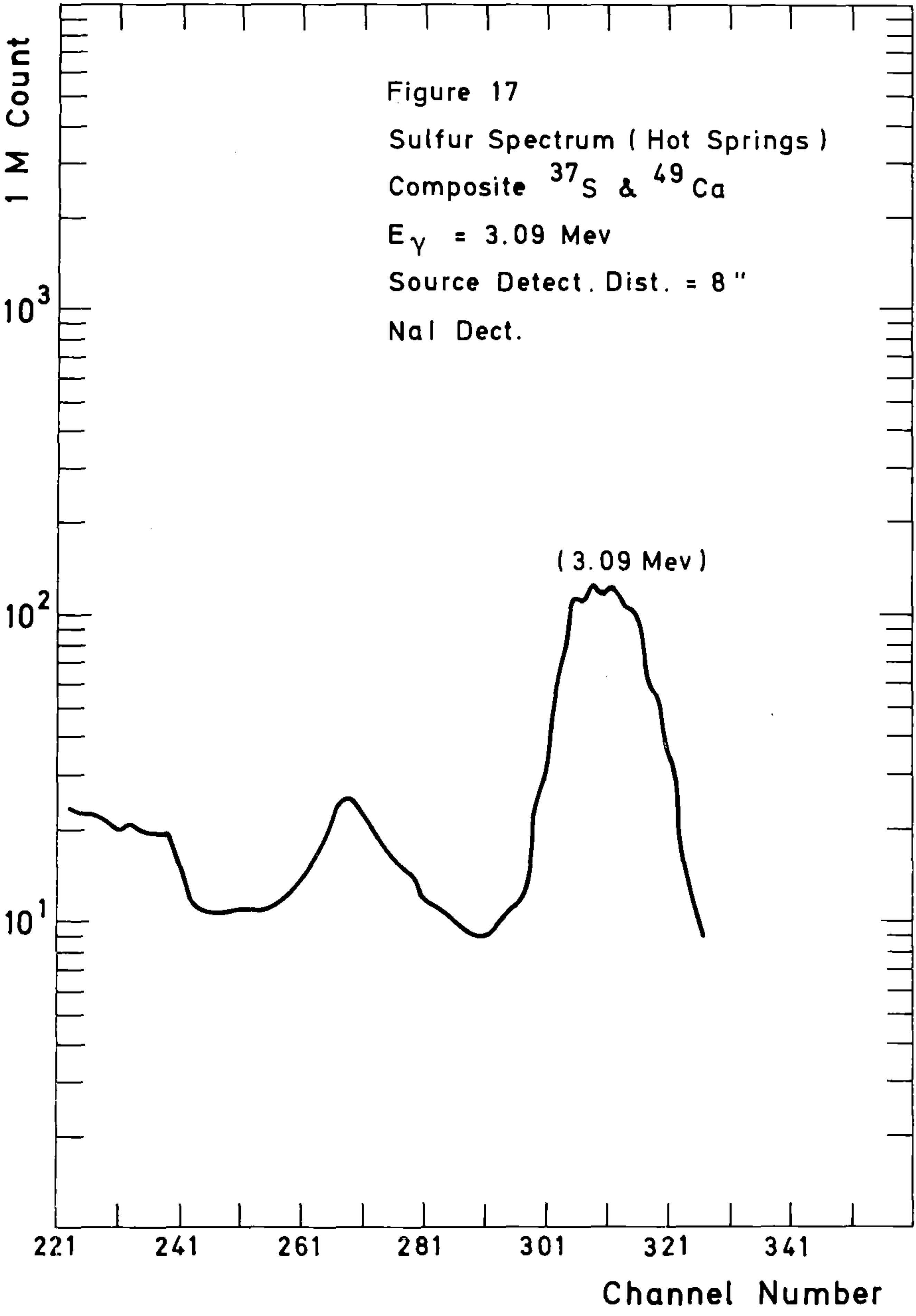

Figure 17

Sulfur Spectrum (Hot Springs)

Composite ^{37}S & ^{49}Ca

E_γ = 3.09 Mev

Source Detect. Dist. = 8"

NaI Dect.

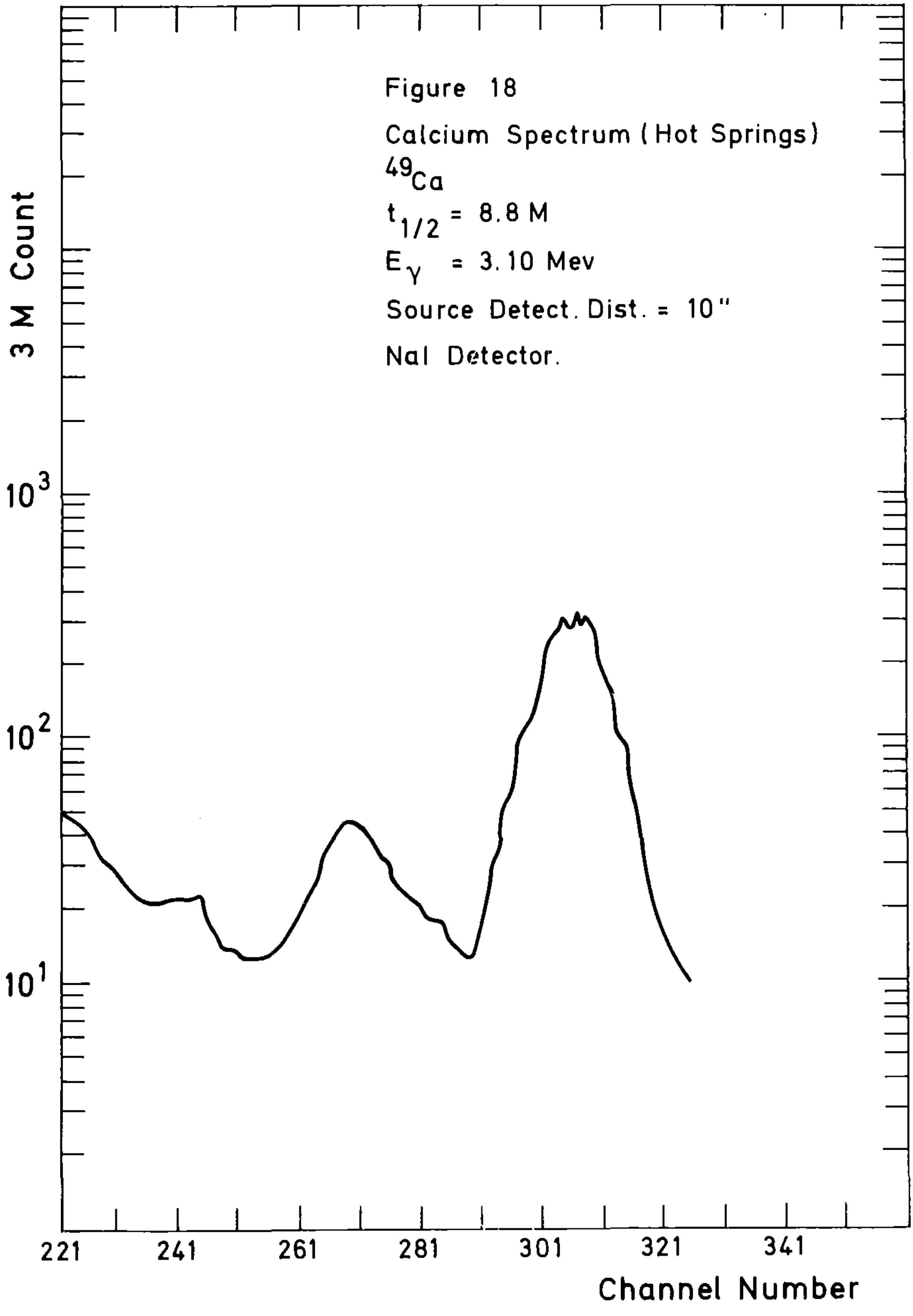

Figure 18

Calcium Spectrum (Hot Springs)

^{49}Ca

$t_{1/2} = 8.8$ M

$E_{\gamma} = 3.10$ Mev

Source Detect. Dist. = 10"

NaI Detector.

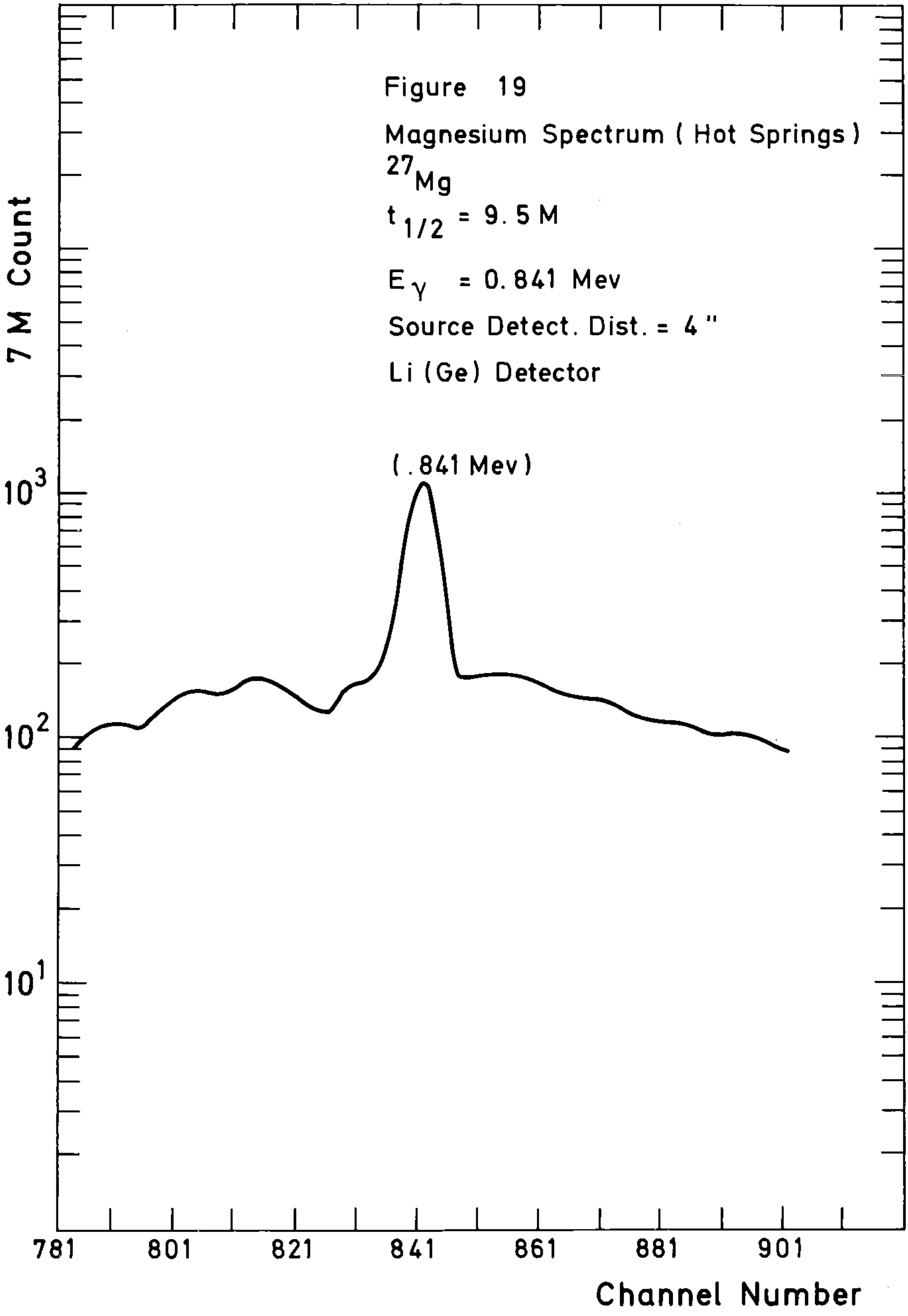

Figure 19

Magnesium Spectrum (Hot Springs)

^{27}Mg

$t_{1/2} = 9.5$ M

$E_\gamma = 0.841$ Mev

Source Detect. Dist. = 4"

Li (Ge) Detector

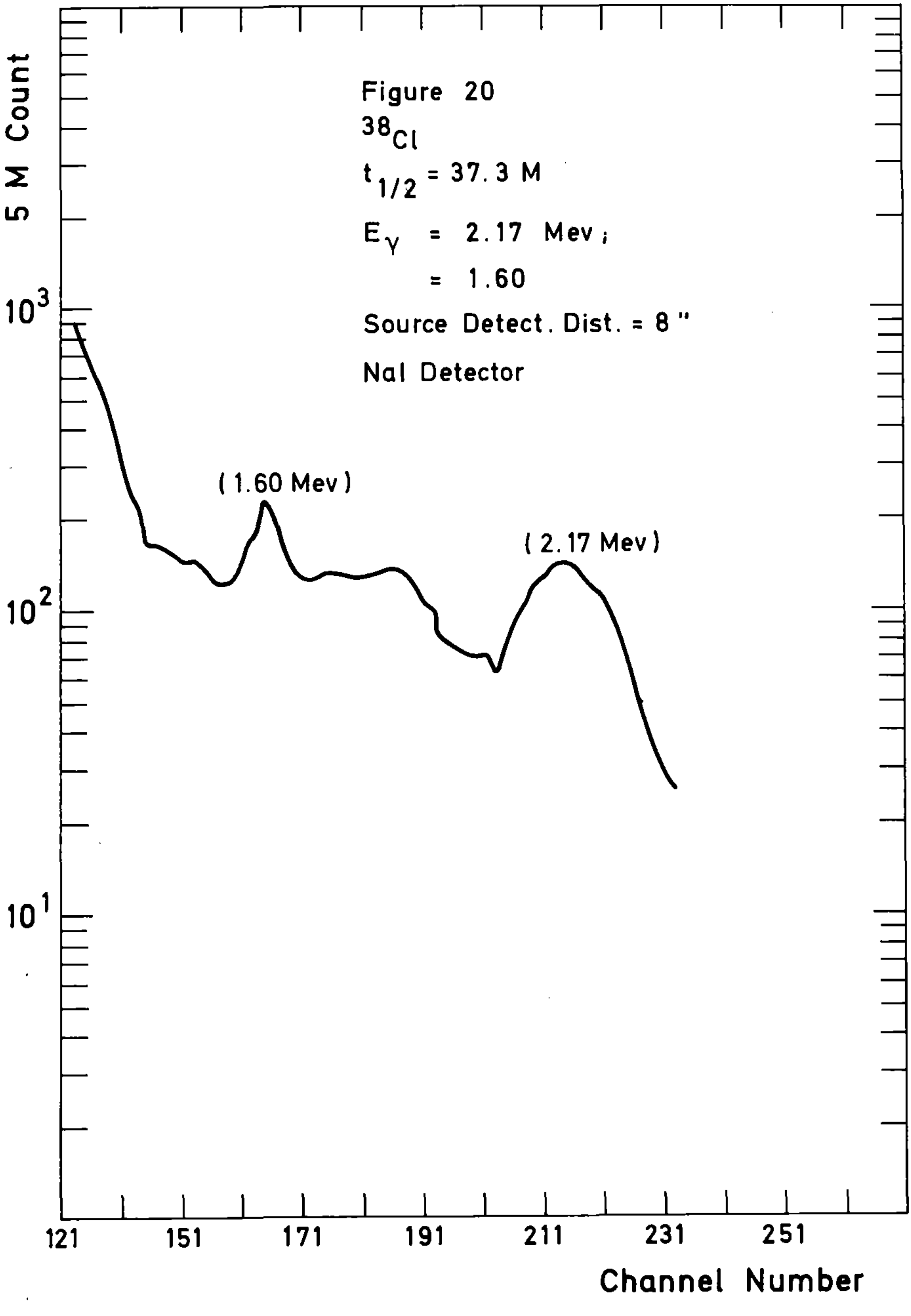
Figure 20
^{38}Cl
$t_{1/2} = 37.3$ M
E_γ = 2.17 Mev ;
= 1.60
Source Detect. Dist. = 8 "
NaI Detector
(1.60 Mev)
(2.17 Mev)
5 M Count
10^3
10^2
10^1
121
151
171
191
211
231
251
Channel Number

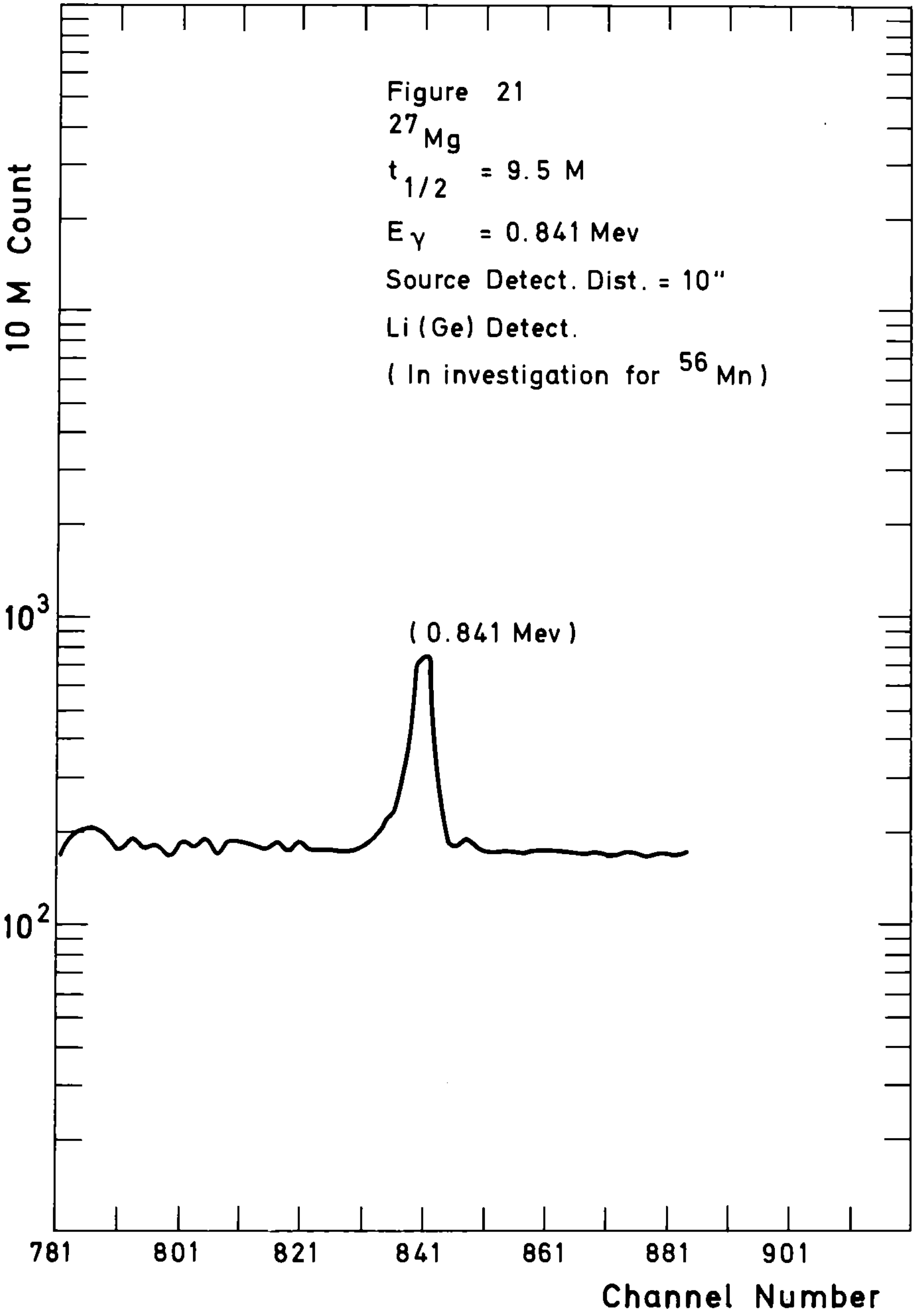

Figure 21

^{27}Mg

$t_{1/2}$ = 9.5 M

E_γ = 0.841 Mev

Source Detect. Dist. = 10"

Li (Ge) Detect.

(In investigation for ^{56}Mn)

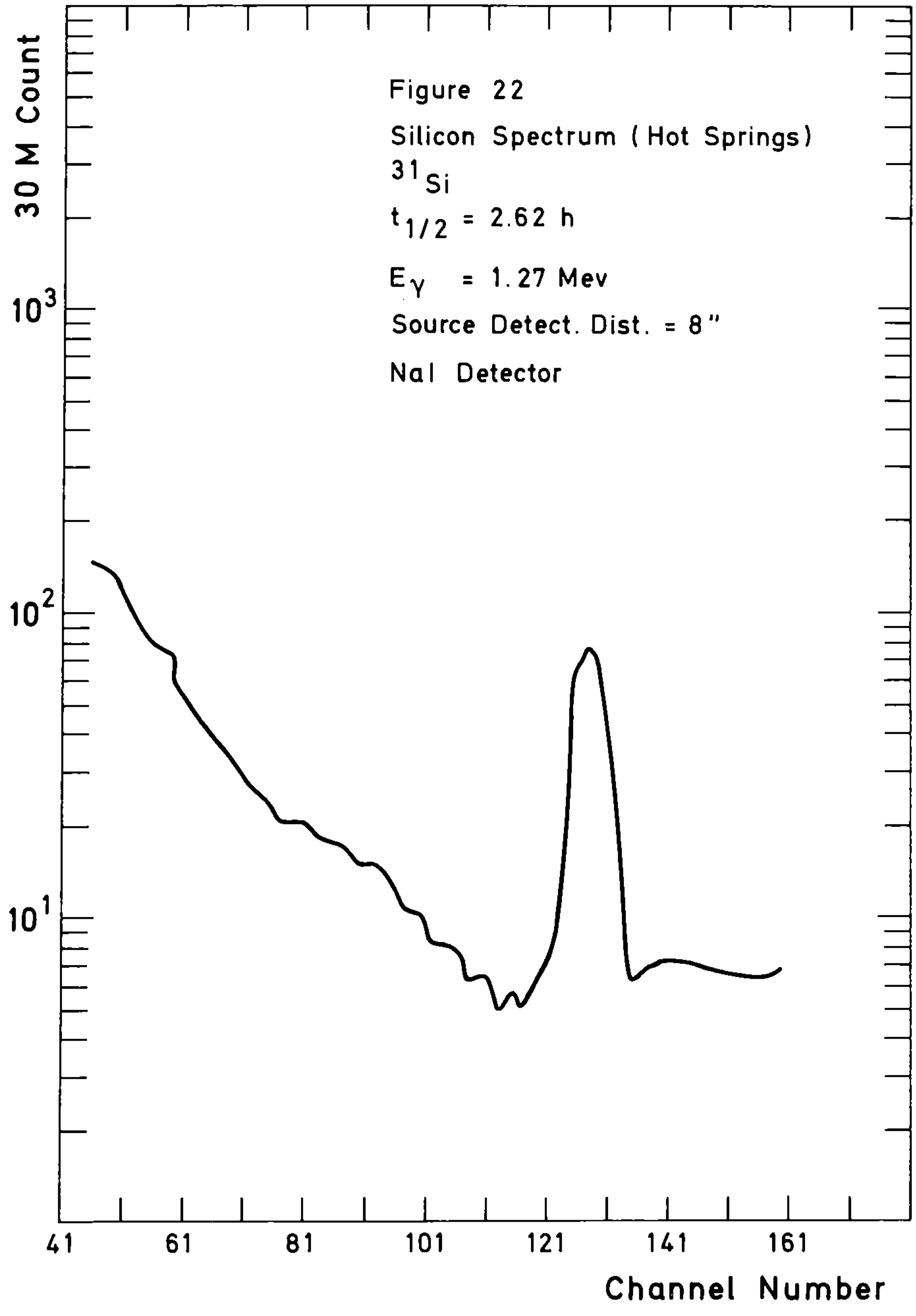

Figure 22

Silicon Spectrum (Hot Springs)

^{31}Si

$t_{1/2} = 2.62$ h

$E_{\gamma} = 1.27$ Mev

Source Detect. Dist. = 8"

NaI Detector

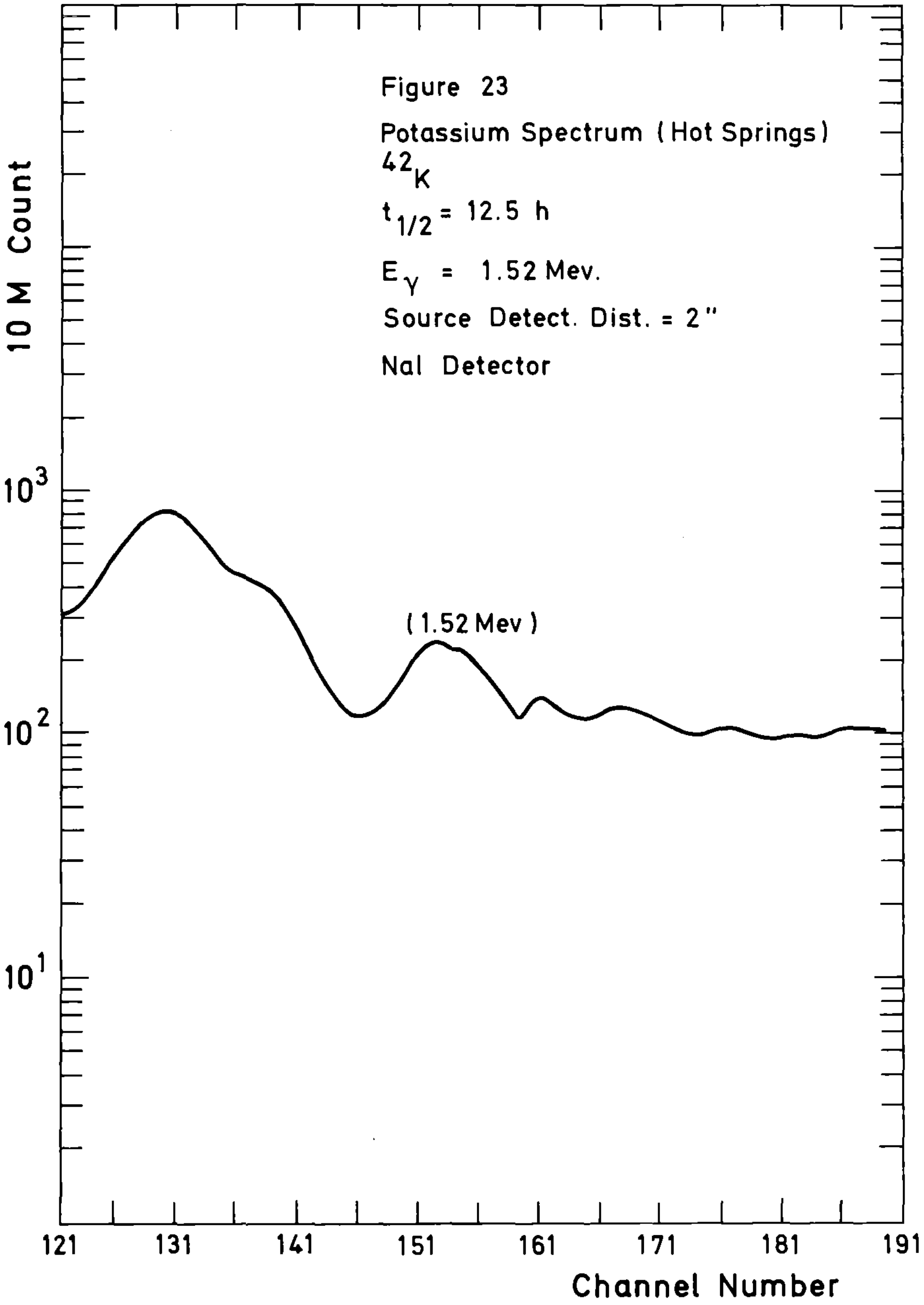

Figure 23

Potassium Spectrum (Hot Springs)

^{42}K

$t_{1/2} = 12.5$ h

E_γ = 1.52 Mev.

Source Detect. Dist. = 2"

NaI Detector

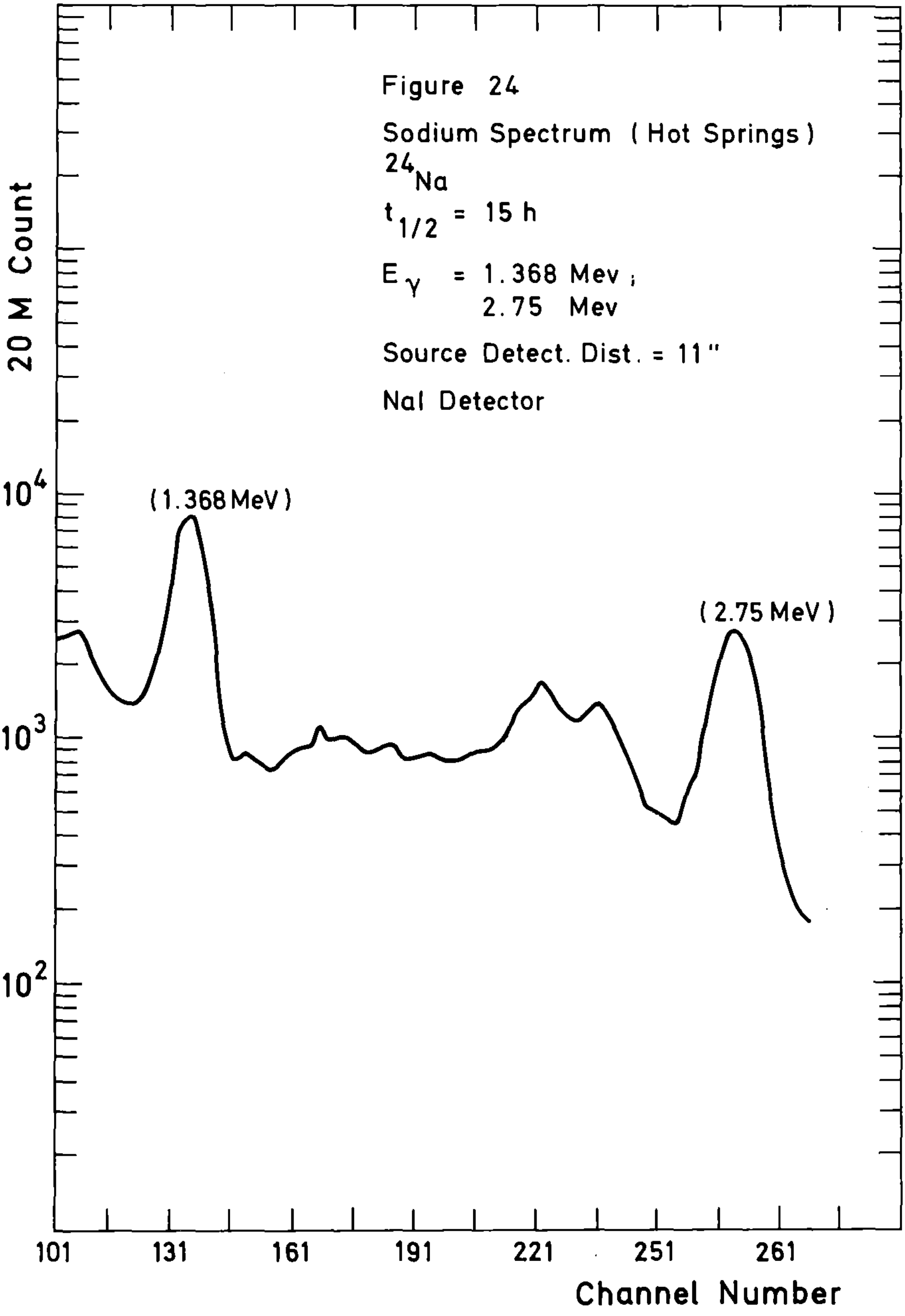

Figure 24

Sodium Spectrum (Hot Springs)
^{24}Na
$t_{1/2}$ = 15 h
E_{γ} = 1.368 Mev ;
2.75 Mev
Source Detect. Dist. = 11"
NaI Detector

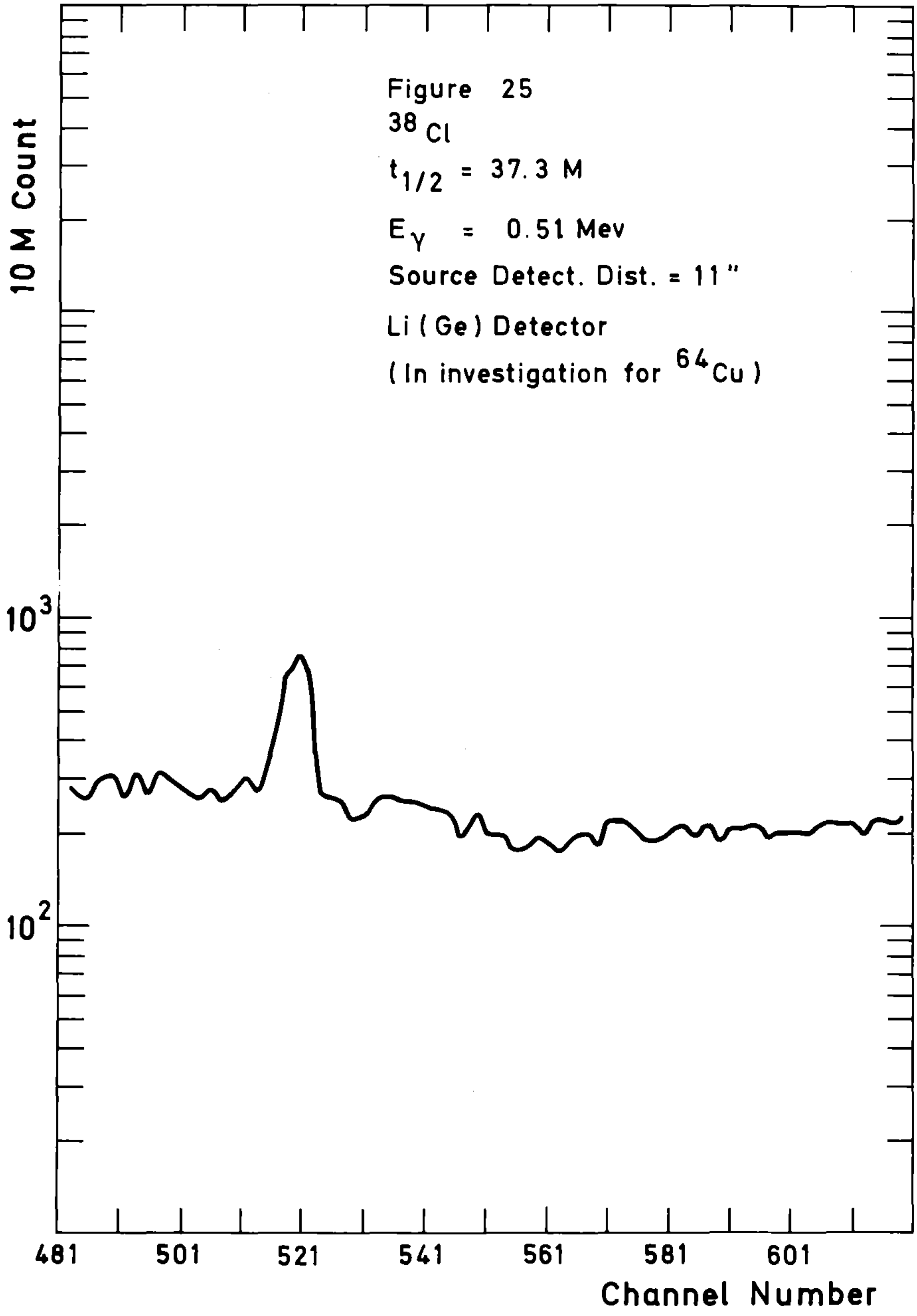
Figure 25
^{38}Cl
$t_{1/2}$ = 37.3 M
E_γ = 0.51 Mev
Source Detect. Dist. = 11"
Li (Ge) Detector
(In investigation for ^{64}Cu)
10 M Count
10^3
10^2
481
501
521
541
561
581
601
Channel Number

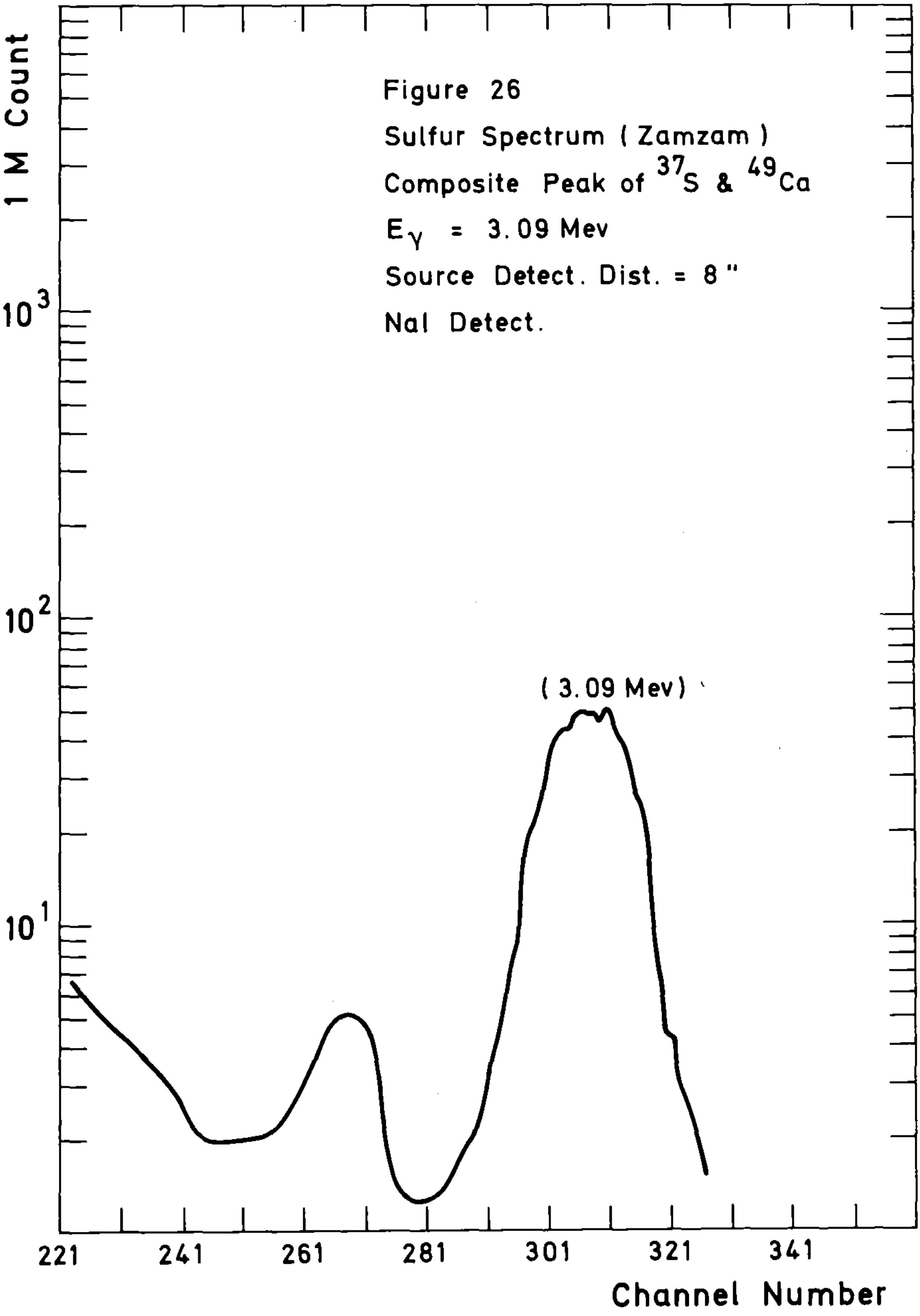
Figure 26

Sulfur Spectrum (Zamzam)

Composite Peak of ^{37}S & ^{49}Ca

E_{γ} = 3.09 Mev

Source Detect. Dist. = 8"

NaI Detect.

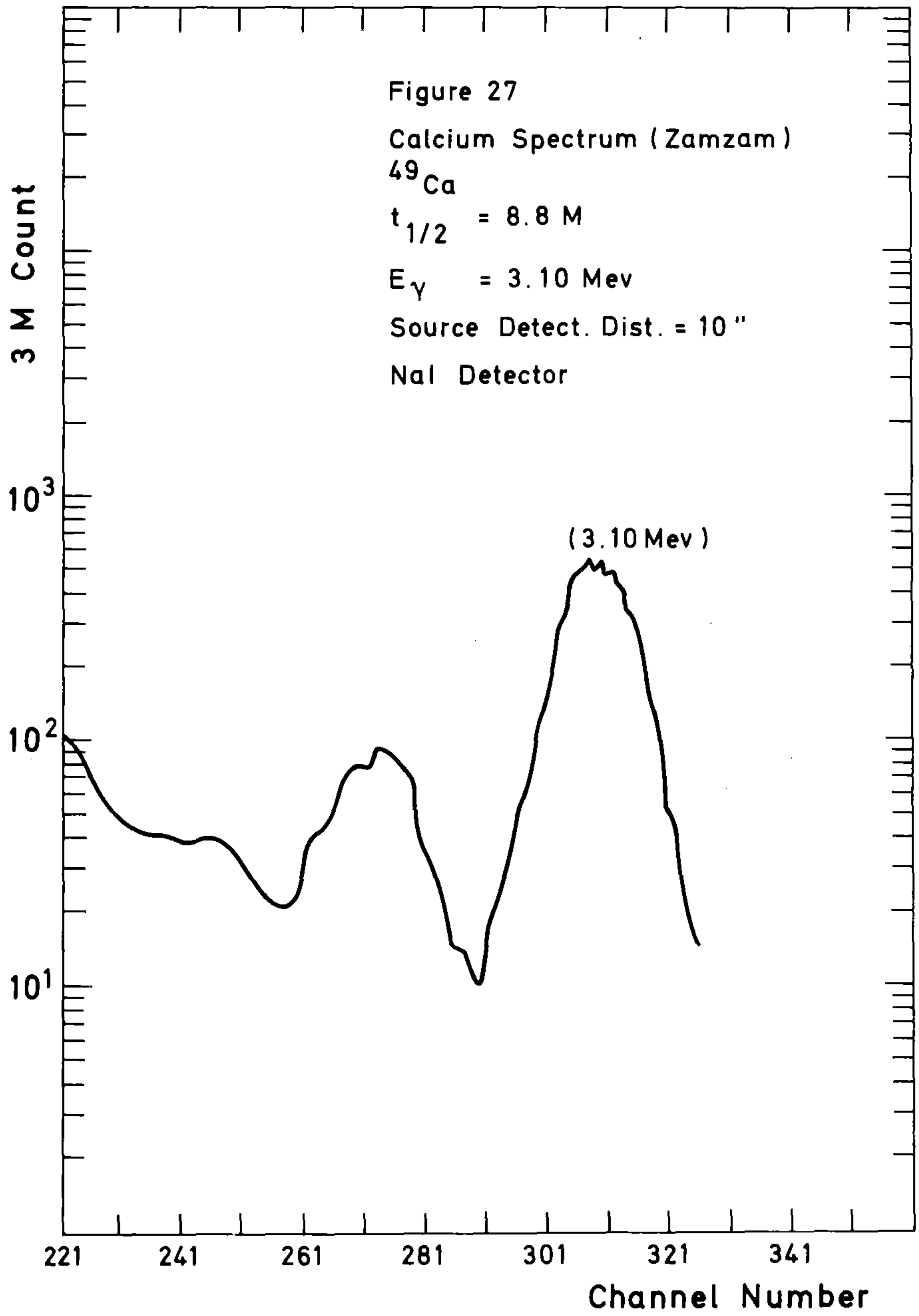

Figure 27

Calcium Spectrum (Zamzam)

^{49}Ca

$t_{1/2}$ = 8.8 M

E_{γ} = 3.10 Mev

Source Detect. Dist. = 10"

NaI Detector

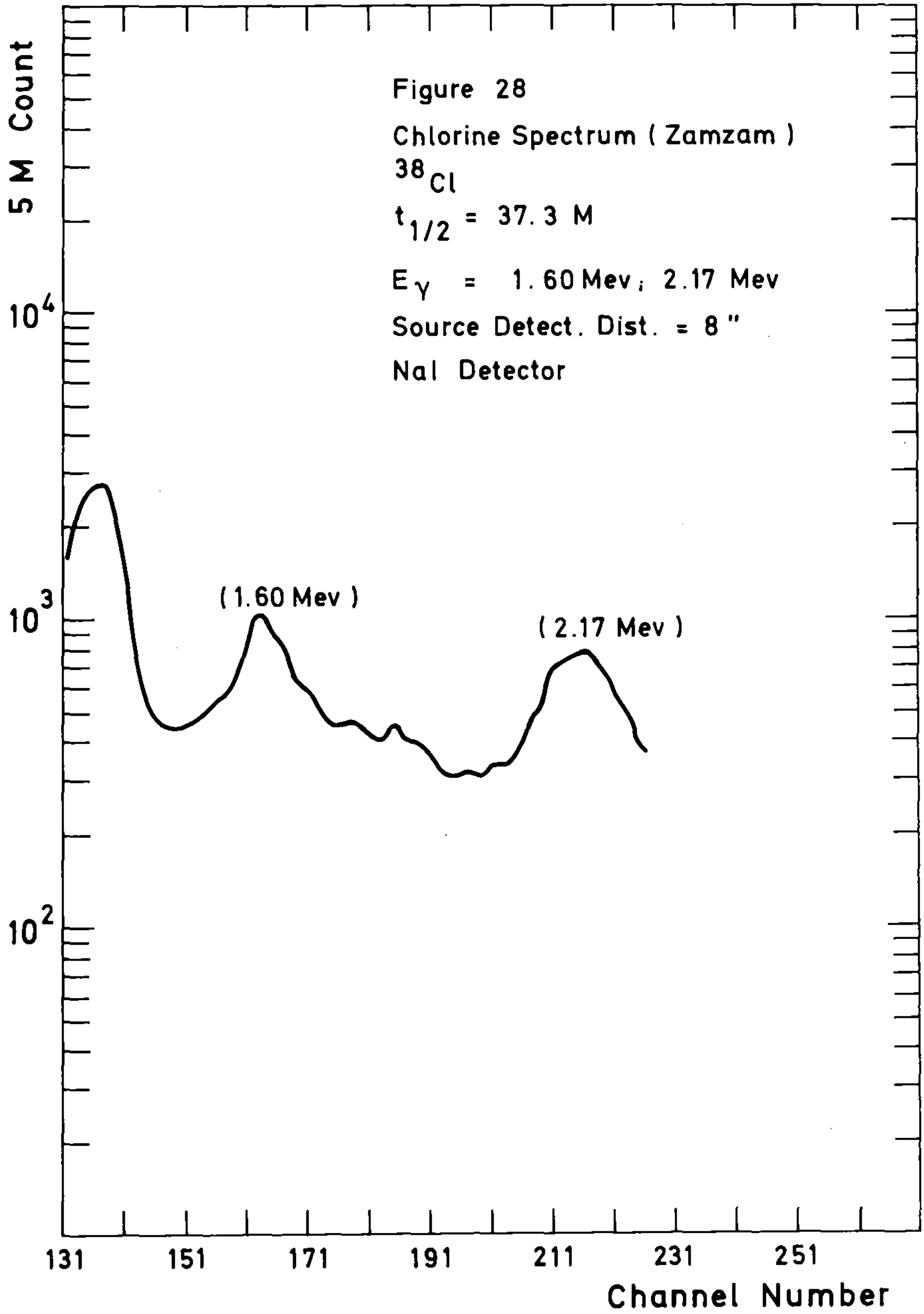

Figure 28

Chlorine Spectrum (Zamzam)

^{38}Cl

$t_{1/2}$ = 37.3 M

E_γ = 1.60 Mev ; 2.17 Mev

Source Detect. Dist. = 8"

NaI Detector

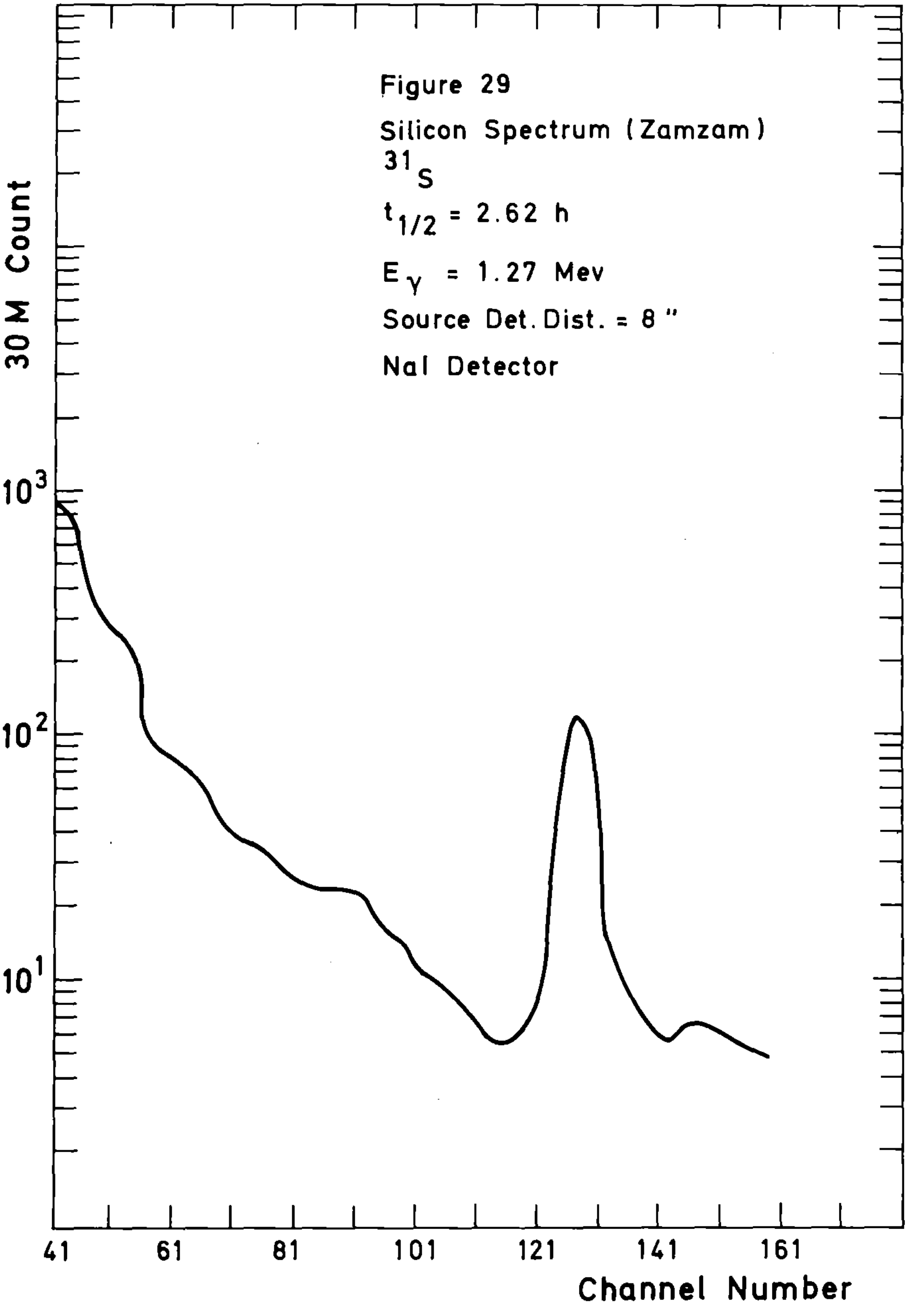
Figure 29
Silicon Spectrum (Zamzam)
^{31}S
$t_{1/2}$ = 2.62 h
E_{γ} = 1.27 Mev
Source Det. Dist. = 8"
NaI Detector
30 M Count
10^3
10^2
10^1
41
61
81
101
121
141
161
Channel Number

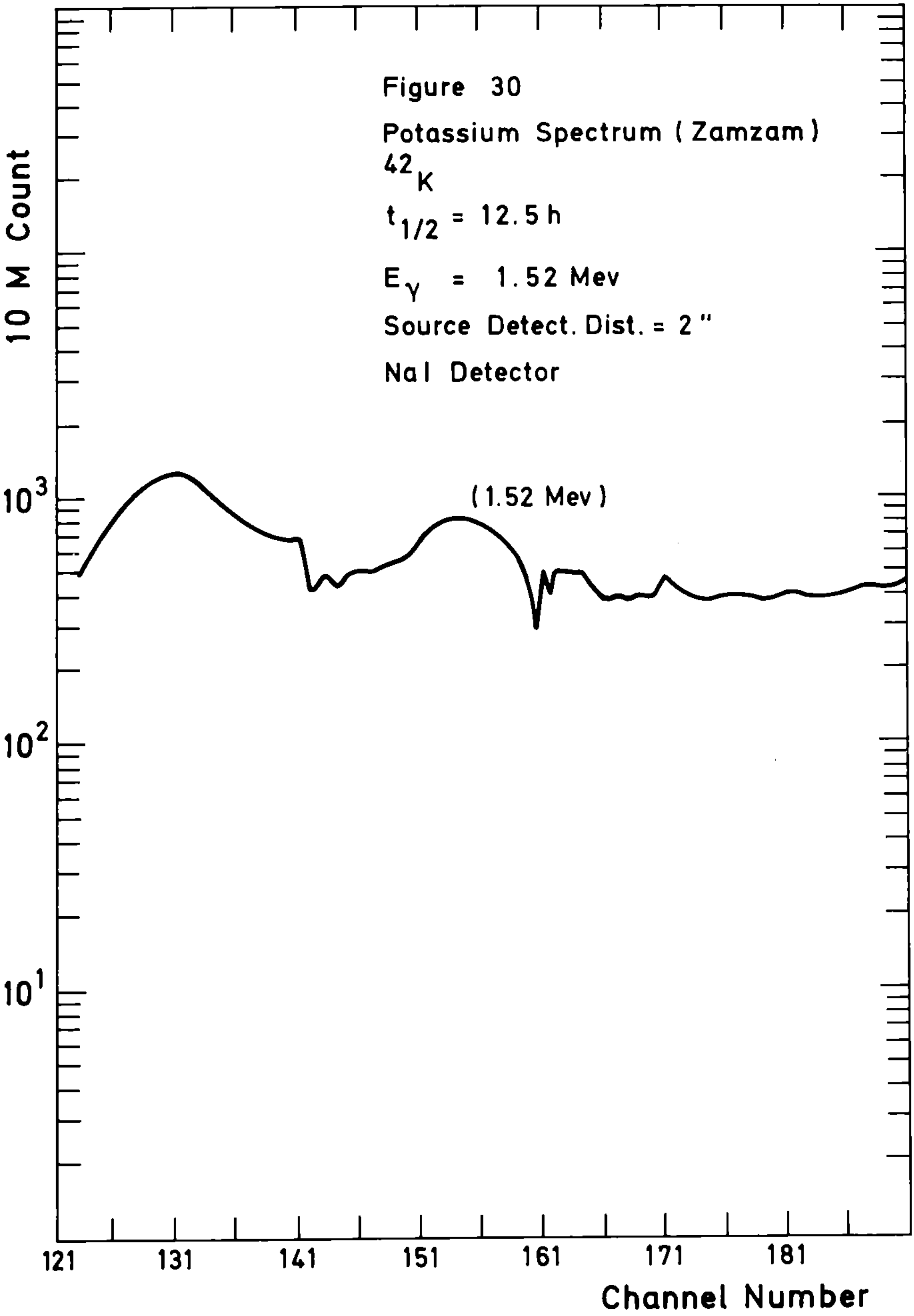
Figure 30
Potassium Spectrum (Zamzam)
^{42}K
$t_{1/2}$ = 12.5 h
E_{γ} = 1.52 Mev
Source Detect. Dist. = 2"
NaI Detector
(1.52 Mev)
10 M Count
10^3
10^2
10^1
121
131
141
151
161
171
181
Channel Number

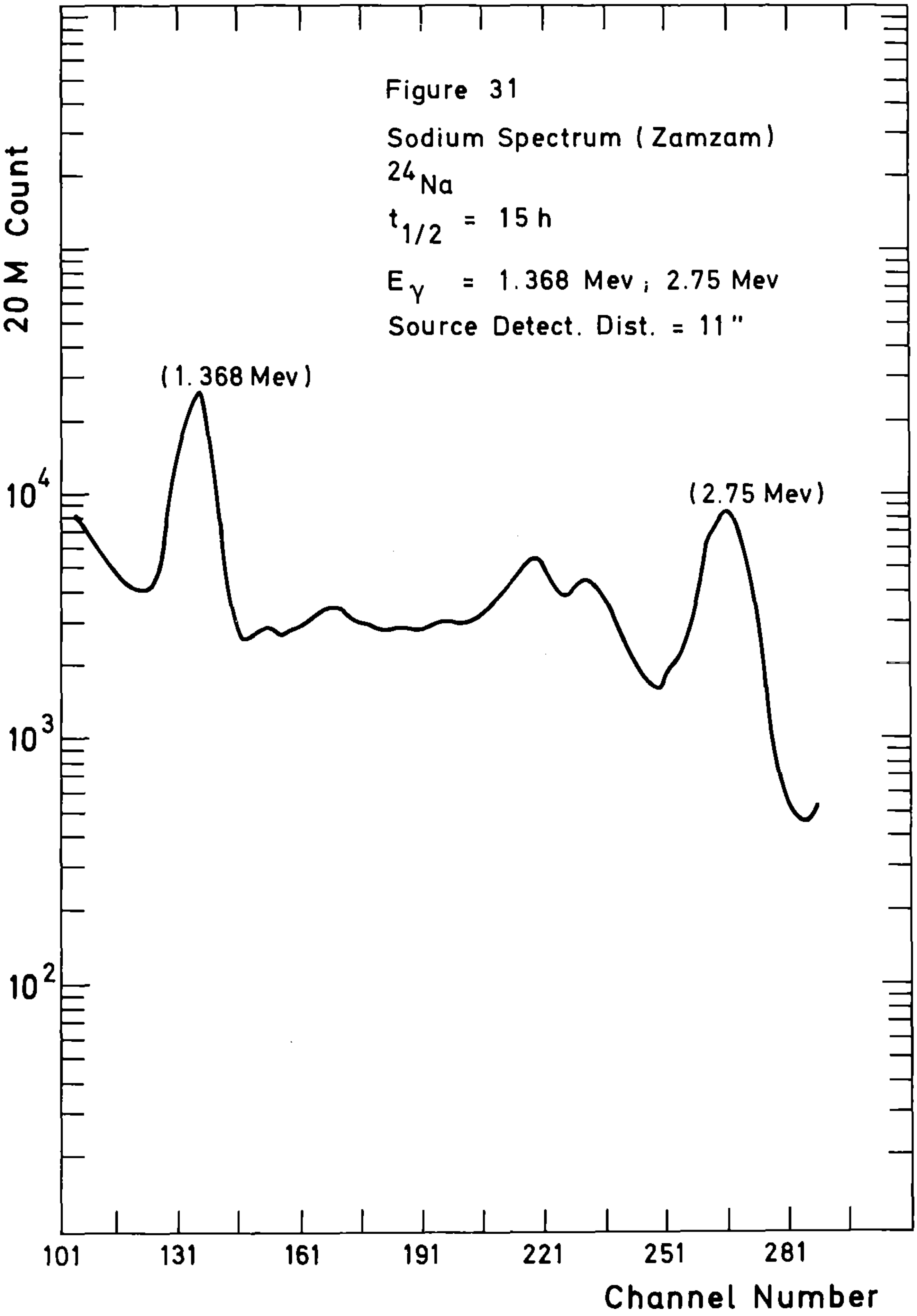

Figure 31

Sodium Spectrum (Zamzam)

^{24}Na

$t_{1/2}$ = 15 h

E_γ = 1.368 Mev ; 2.75 Mev

Source Detect. Dist. = 11"

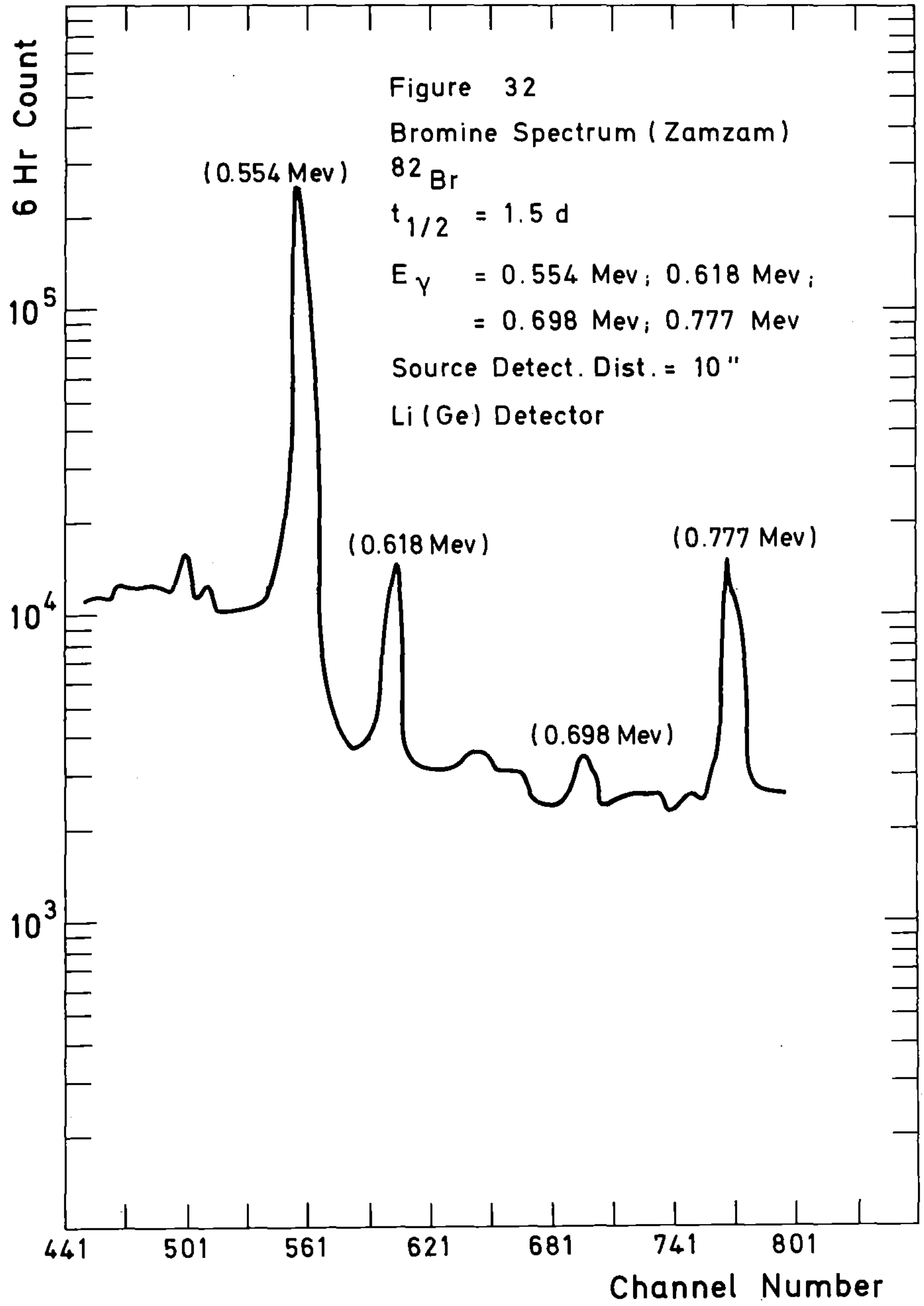

Figure 32

Bromine Spectrum (Zamzam)

^{82}Br

$t_{1/2}$ = 1.5 d

E_γ = 0.554 Mev; 0.618 Mev;
= 0.698 Mev; 0.777 Mev

Source Detect. Dist. = 10"

Li (Ge) Detector

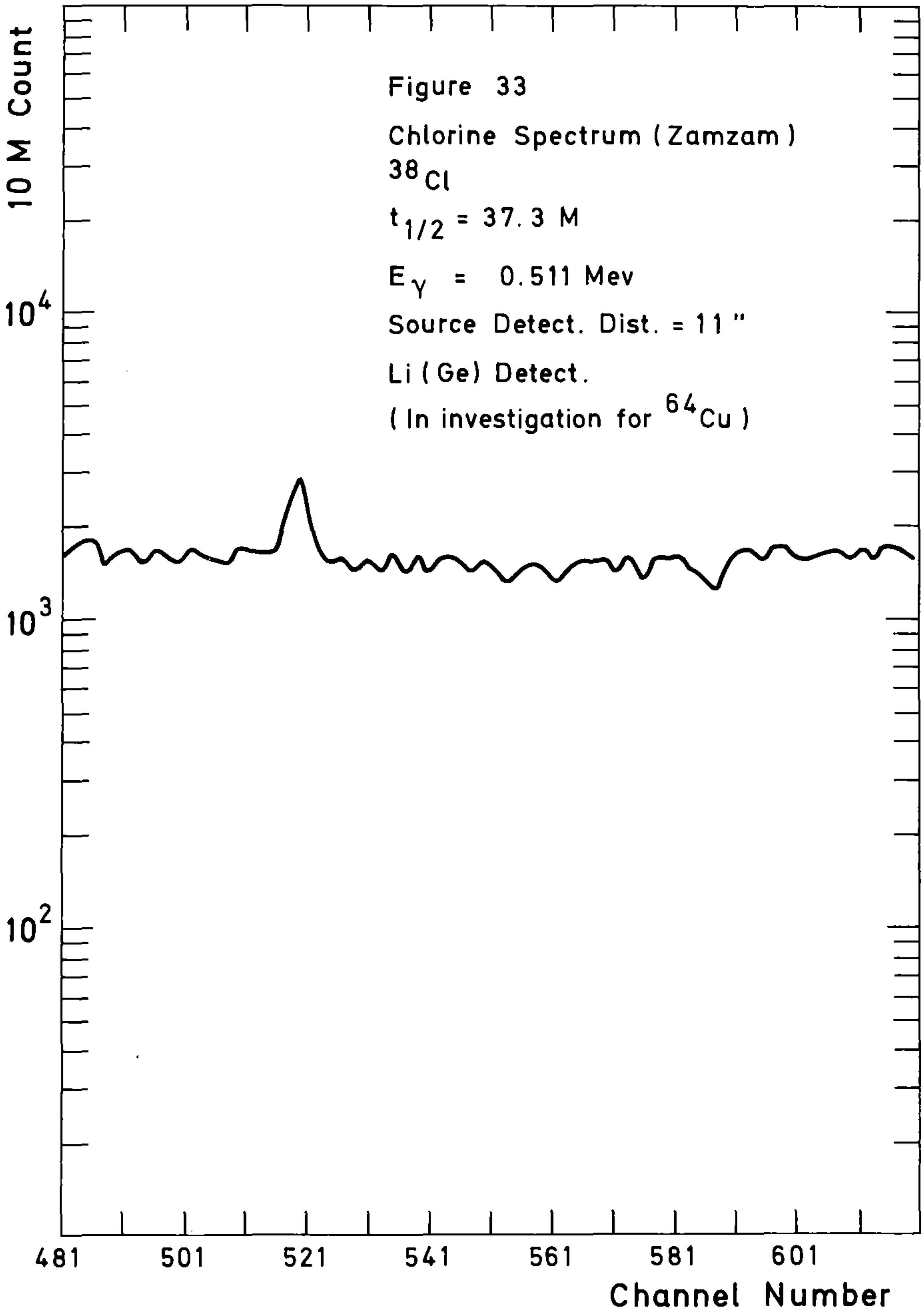

Figure 33

Chlorine Spectrum (Zamzam)

^{38}Cl

$t_{1/2} = 37.3$ M

$E_{\gamma} = 0.511$ Mev

Source Detect. Dist. = 11"

Li (Ge) Detect.

(In investigation for ^{64}Cu)

APPENDIX B

Officials at the Museum in Hot Springs, Arkansas hand out the following report:

Chemical Composition of the Hot Springs Waters (in part per million):

Silica (SiO_2) 45
Calcium (Ca) 45
Magnesium (Mg) 4.9
Sodium (Na) 4
Potassium (K) 1.6
Sulfate (SO_4) 7.8
Chloride (Cl) 2

Bicarbonate (HCO_3) 162
Fluoride (F) 0.1
Nitrate (NO_3) 0.3
Phosphate (PO_4) 0.1

Gases in cubic centimeters per liter at 0 ^{0}C and 76 millimeters pressure:

Nitrogen (N) 8.8
Oxygen (O) 3.8
Free Carbon Dioxide (CO_2) 6.9

Radioactivity through radon gas emanatron is 0.81 millimicro curie per liter.