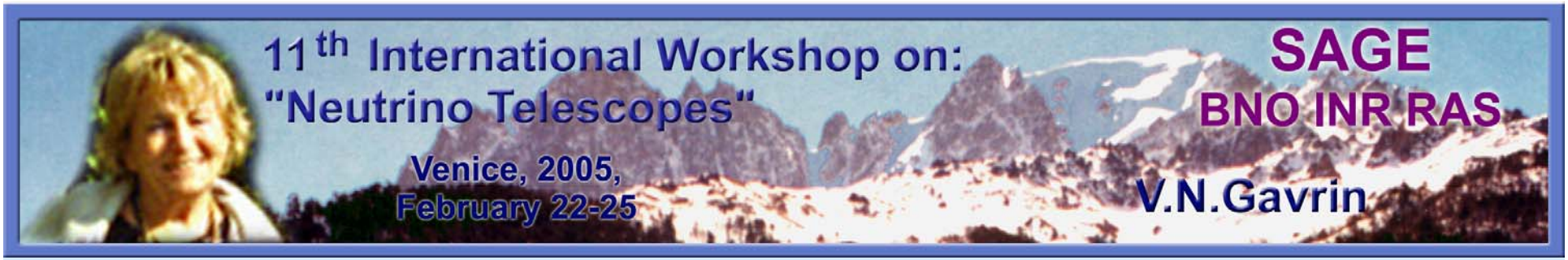

Present status of the SAGE ^{37}Ar neutrino source experiment

^{37}Ar as a calibration source for solar neutrino detectors

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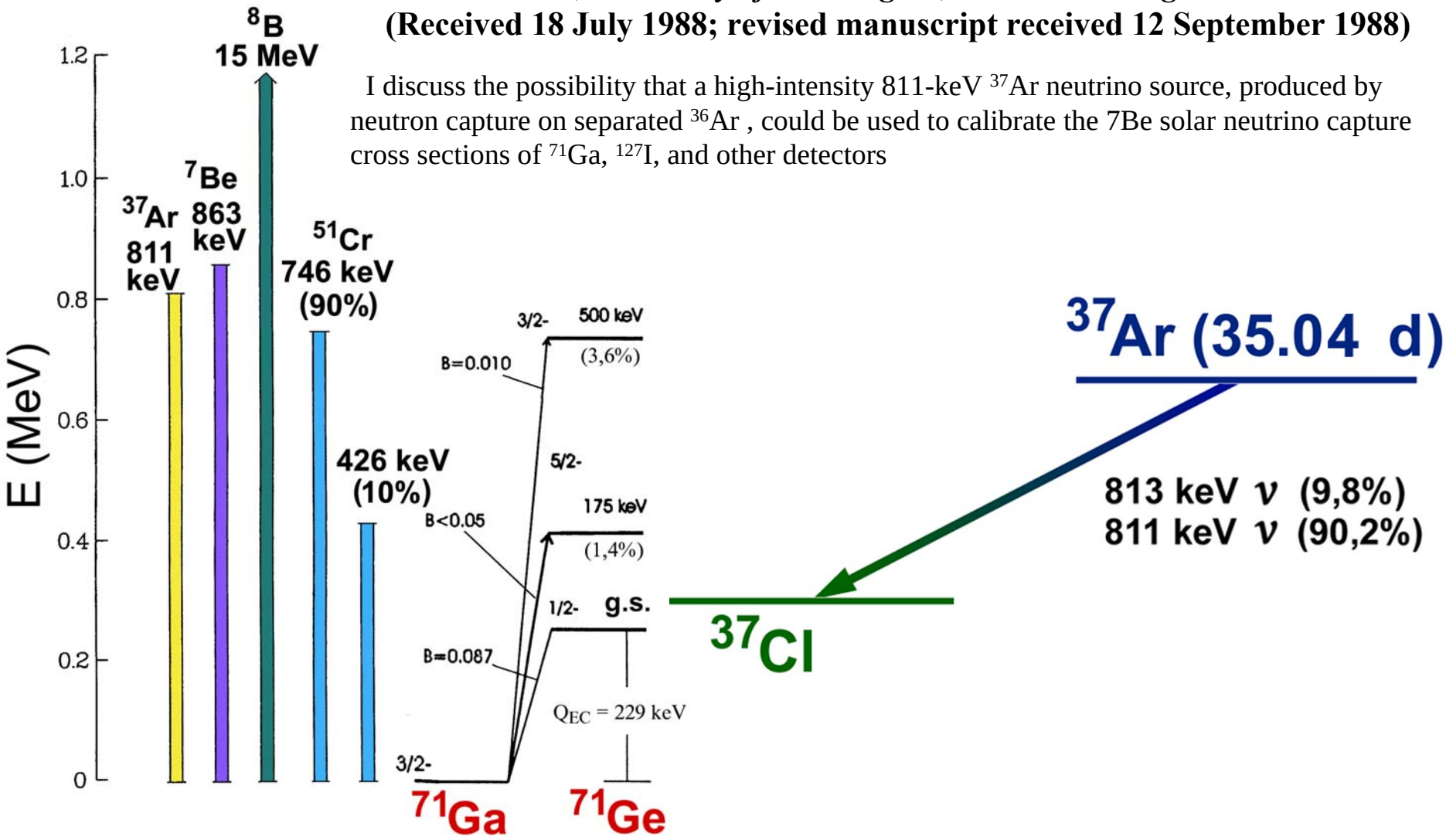
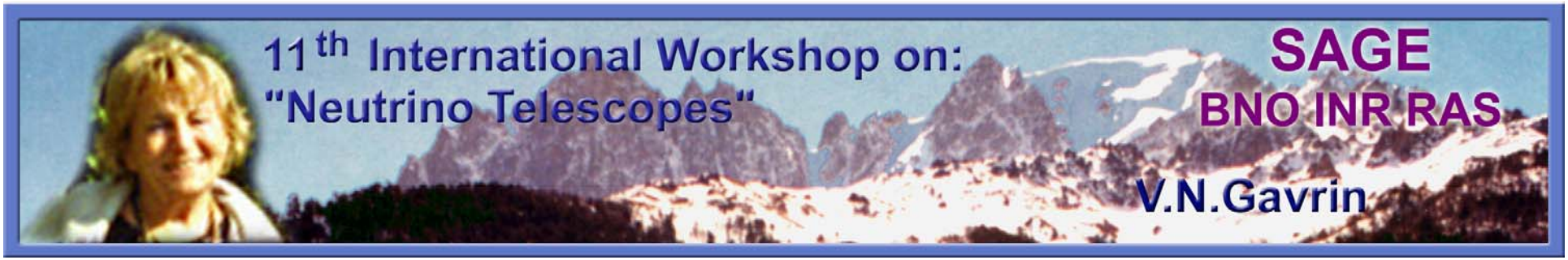
(Received 18 July 1988; revised manuscript received 12 September 1988)

I discuss the possibility that a high-intensity 811-keV ^{37}Ar neutrino source, produced by neutron capture on separated ^{36}Ar , could be used to calibrate the 7Be solar neutrino capture cross sections of ^{71}Ga, ^{127}I, and other detectors





The advantages of a ^{37}Ar source compared to a ^{51}Cr source

1. A major advantage is that the desired active isotope must be chemically separated from the target following irradiation. Thus a ^{37}Ar source, in contrast to a ^{51}Cr source can be made practically free of radioactive impurities.

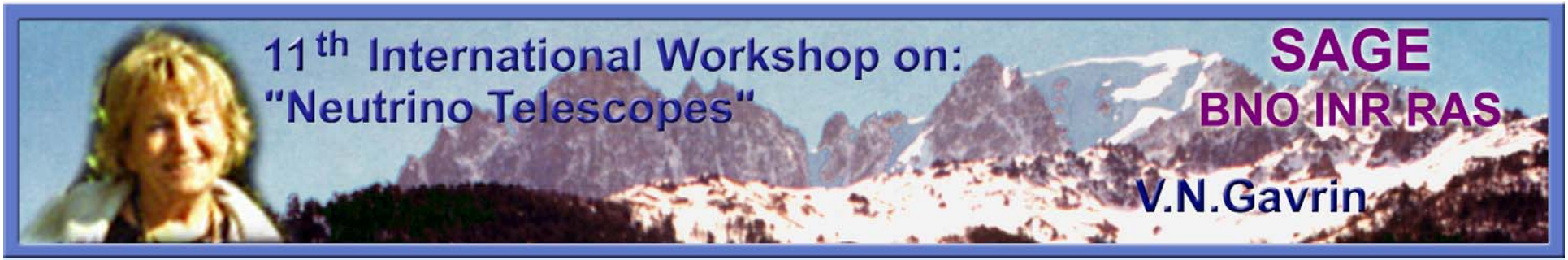
2. A ^{37}Ar compared to ^{51}Cr have the half-life longer (35 d compared to 27 d).

3. The neutrino energy is greater (811 keV compared to 747 keV), thus giving a higher cross section.

4. The decay is purely to the ground state (100% compared to 90%), thus giving a mono-energetic neutrino source, and that there are no accompanying gamma rays (except for inner bremsstrahlung), thus requiring little shielding and yielding a very compact source.

Decay modes of ^{37}Ar and the energy released

Decay mode	Atomic energy relese (keV)	Fraction of ^{37}Ar decays	Atomic energy per ^{37}Ar decay
K capture	2.8224	0.9017 ± 0.0024	2.5450 ± 0.0068
L capture	0.2702	0.0890 ± 0.0027	0.0240 ± 0.0007
M capture	0.0175	$0.0093^{+0.0006}_{-0.0004}$	0.0002
Int. brems.	325 (average)	~ 0.0005	$\sim 0.16 \pm 0.02$
Total			2.729 ± 0.021



Haxton' proposal immediately attracted our attention and we considered in detail a practical method to make an intense ^{37}Ar source by the (n, α) capture reaction on ^{40}Ca at a reactor with a high flux of fast neutrons.

РОССИЙСКАЯ АКАДЕМИЯ НАУК
ИНСТИТУТ ЯДЕРНЫХ ИССЛЕДОВАНИЙ

RUSSIAN ACADEMY OF SCIENCES
INSTITUTE FOR NUCLEAR RESEARCH

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On the Possibility ^{37}Ar Artificial Neutrino Source

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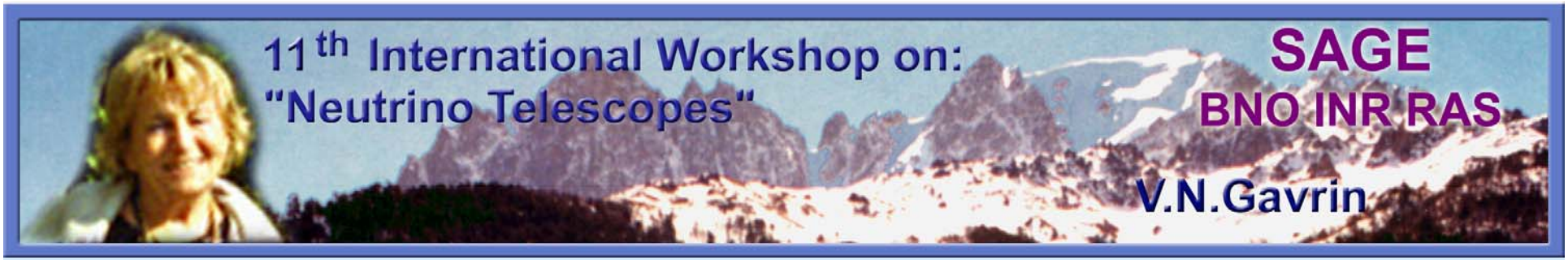
Project:

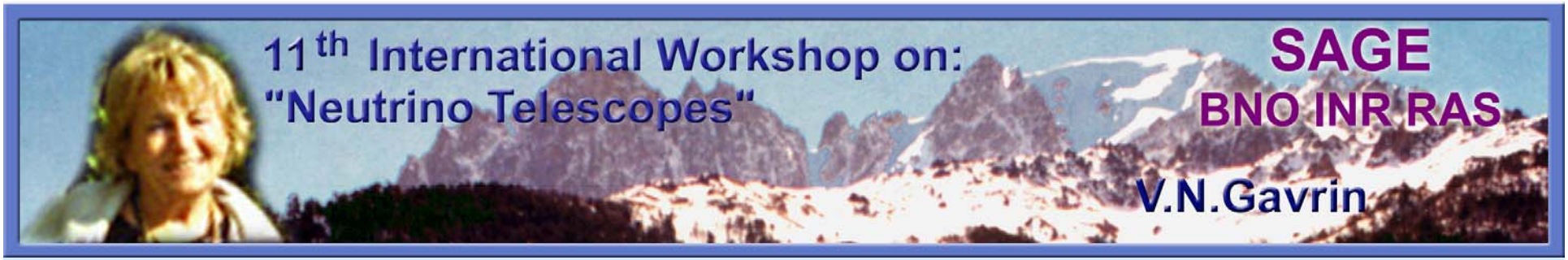
"Calibration and testing of the technology for the preparation of an intense neutrino source based on ^{37}Ar isotope as well as for the calibration of gallium detector of solar neutrinos"

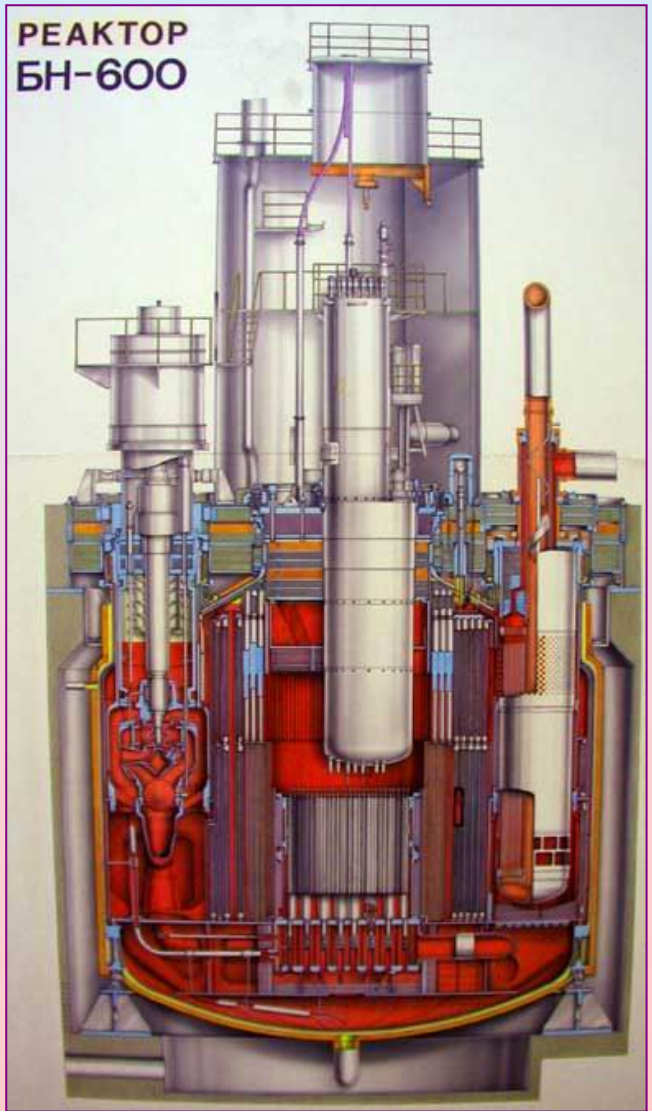


Source production

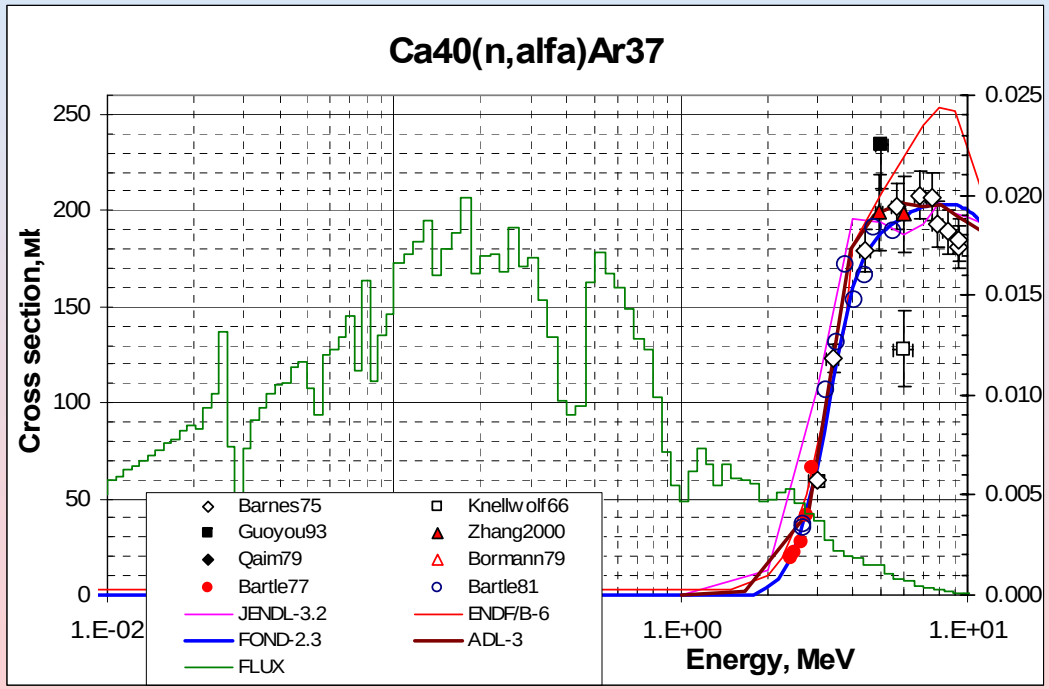
Beloyarsk Nuclear Power Plant
BN-600 Fast Neutron Reactor

The source was made by irradiating calcium oxide in the fast neutron breeder reactor BN-600 at Zarechny, Russia.



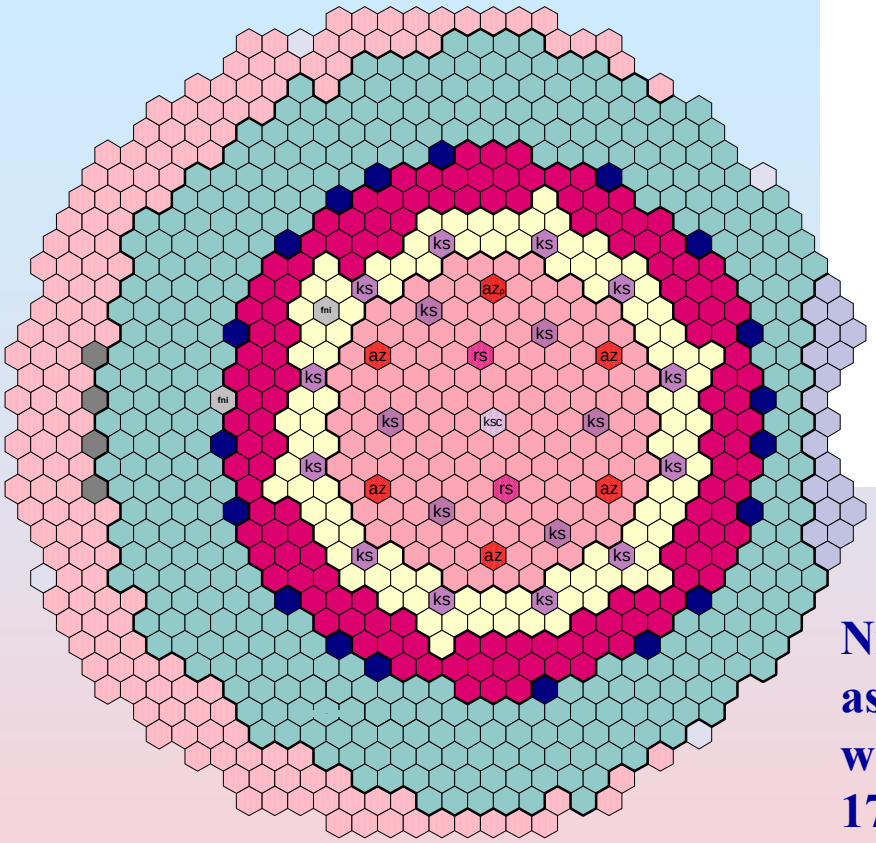


^{37}Ar production cross-section

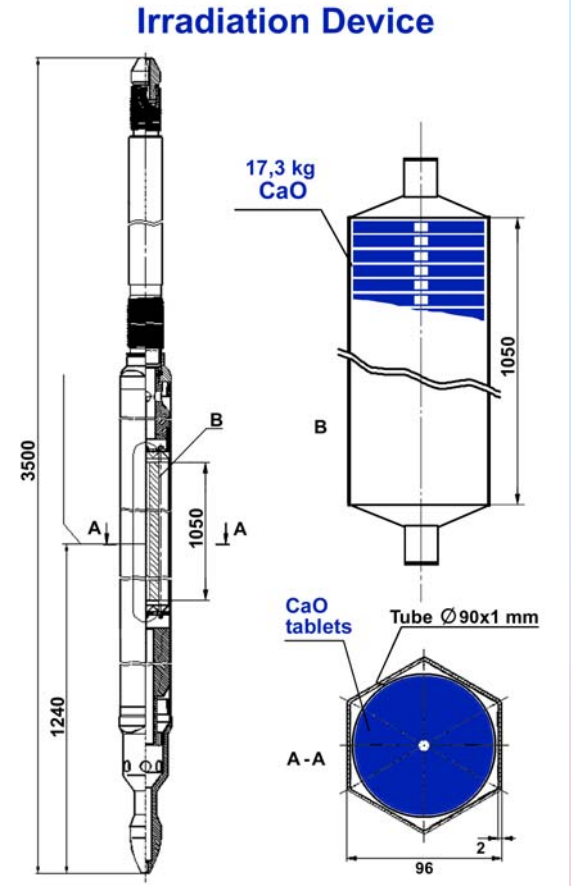


The total fast flux at this reactor is $2.3 \cdot 10^{15}$ neutrons/(cm^2 · s), of which $1,7 \cdot 10^{14}$ neutrons/(cm^2 · s) have energy above the 2 MeV threshold of the production reaction ^{40}Ca (n, α) ^{37}Ar.

Map of BN-600 Reactor



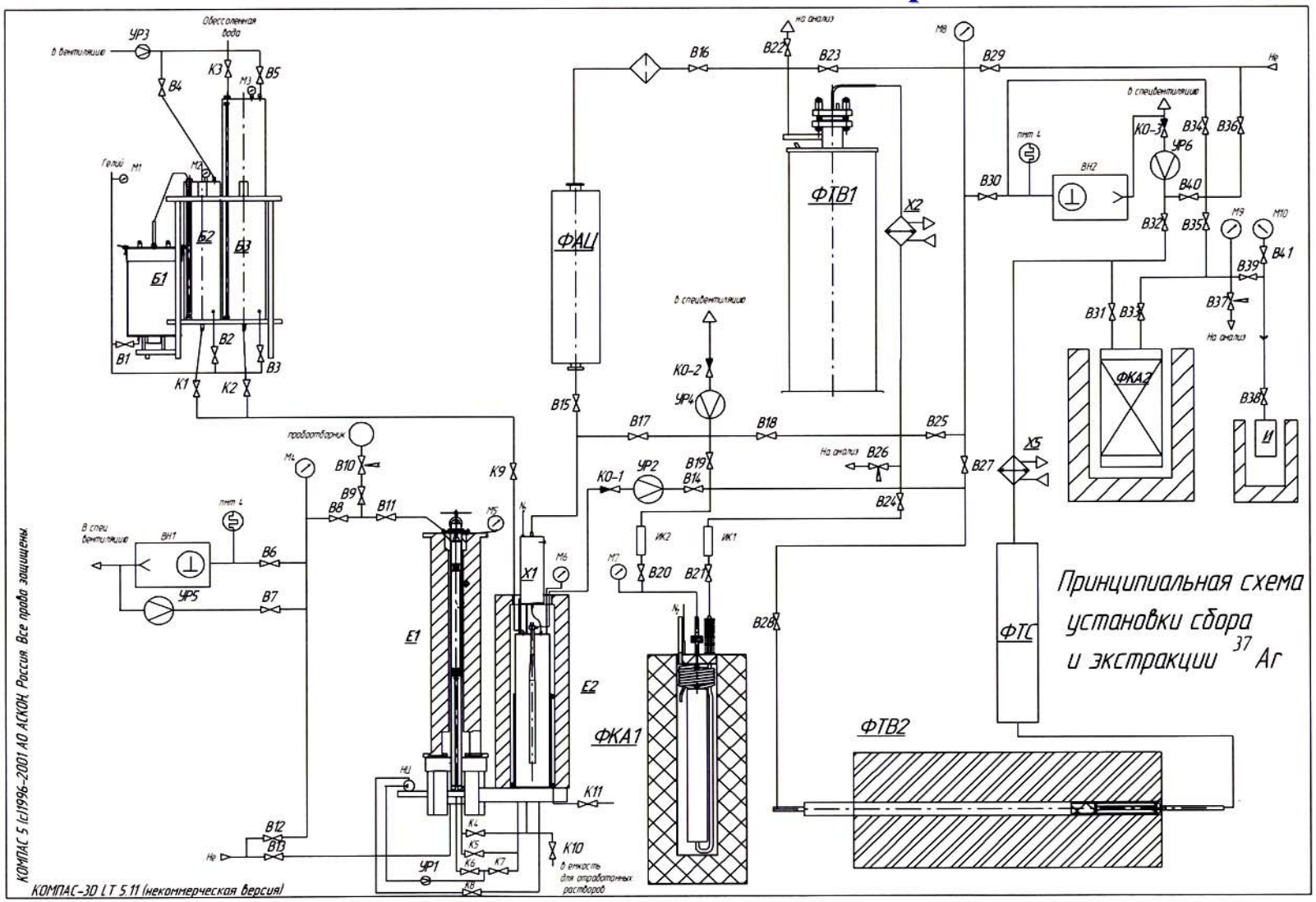
- low enrichment zone
- middle enrichment zone
- high enrichment zone
- side blanket
- storage assemblies
- control rod steel box
- steel assemblies
- CaO assemblies



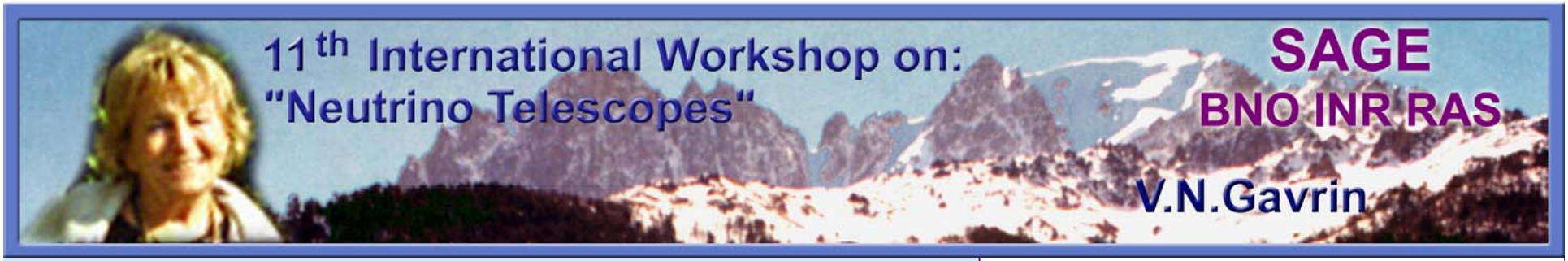
Nineteen irradiation assemblies, each of which contained 17.3 kg of CaO (12.36 kg Ca), were placed in the blanket zone of the reactor.

Irradiation began on 31 October 2003 and continued until 12 April 2004, the normal reactor operating cycle. After a cooling period of a week, the assemblies were removed from the reactor and moved to a hot cell of BNPP where ampoules with irradiated target were taken out from assemblies and moved to extraction facility of the Institute of Nuclear Materials, where each ampoule was cut open in a vacuum system and the CaO dissolved in nitric acid.

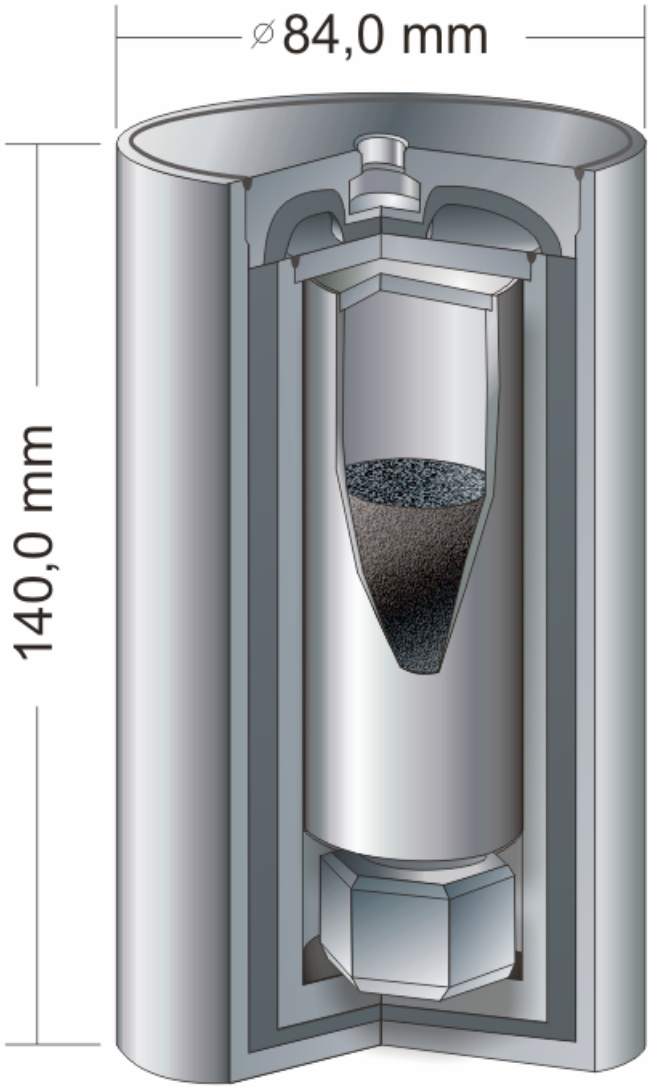
^{37}Ar was extracted from acid solution by a He purge and then stored on charcoal at LN2 temperature.

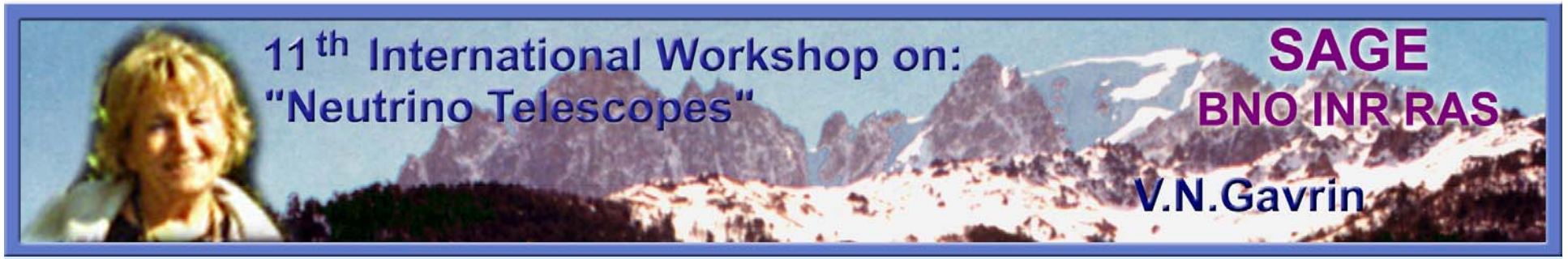


When the extractions from all the assemblies had been completed, the ^{37}Ar was purified by flowing over zeolite at room temperature, followed by two Ti absorbers, operating at 400-450°C and 900-950°C. The purified ^{37}Ar, whose volume was ~ 2.5 l, was then adsorbed on another charcoal trap and measurements of gas volume and isotopic composition were made.



As the last steps of source fabrication, the purified Ar was transferred to a pre-weighed source holder, which consisted of a stainless steel vessel with a volume of ~180 ml. Inside this vessel was 40 g of activated charcoal onto which the purified ^{37}Ar was cryopumped. When essentially all the ^{37}Ar had been adsorbed, the vessel was closed by compressing three separate knife-edge seals, two onto copper gaskets and one onto a lead gasket. The source holder was then weighed to determine the amount of ^{37}Ar contained within. To complete the source, the source holder was placed within two concentric stainless steel vessels with a Pb shield between them. These two vessels were welded shut and the heat output of the finished source was measured with a calorimeter. These procedures were completed on 29 April and the source was immediately flown by chartered plane to the Mineral Water airport, close to the experimental facility at the Baksan Neutrino Observatory in the northern Caucasus mountains.





S. Zlokazov, V. Gavrin, A. Korenkova, Z. Shakirov, V. Barsanov (left to right) with ^{37}Ar source before its delivery from the IRM


3 т
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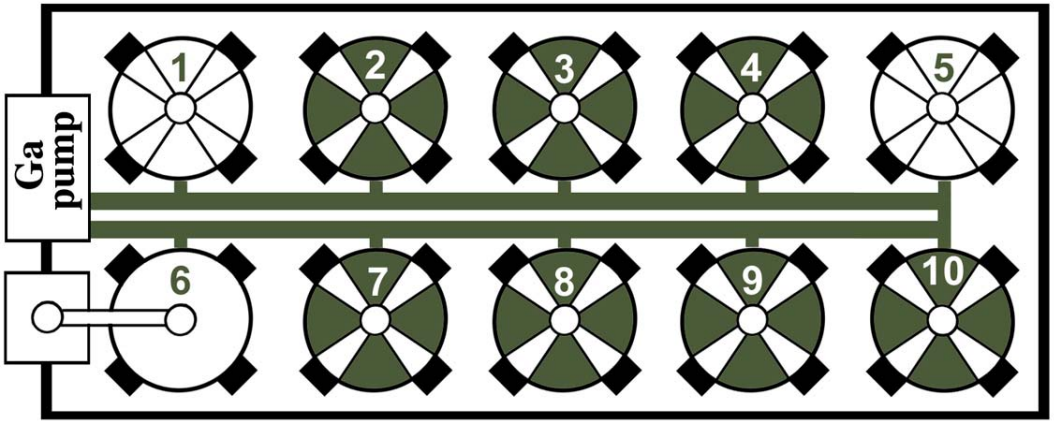
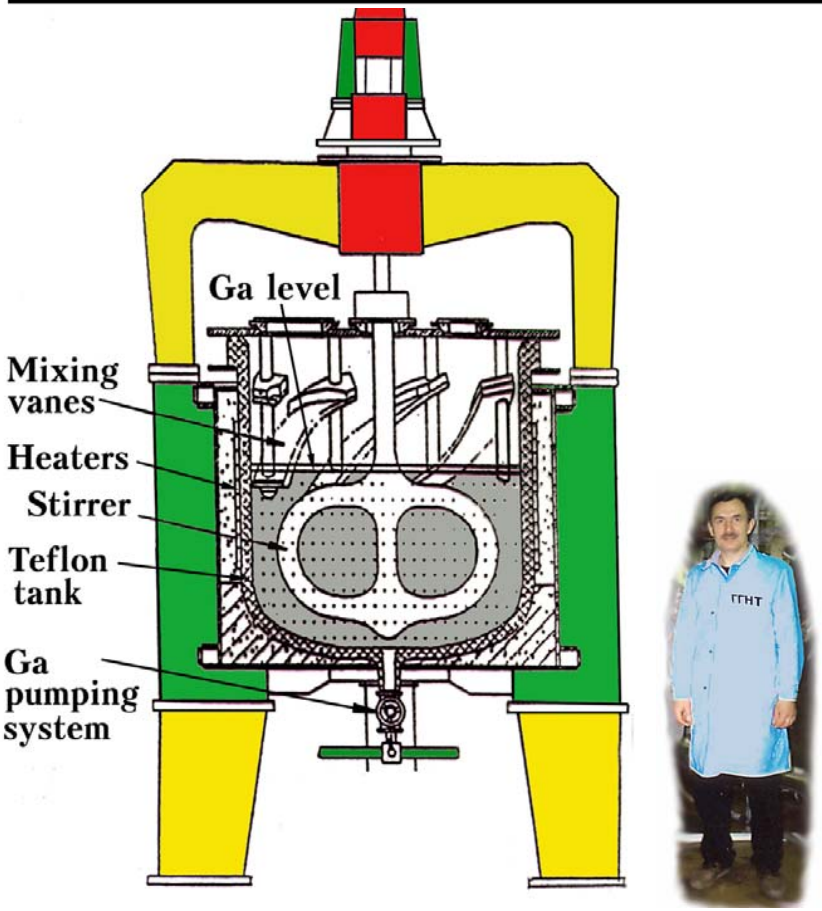
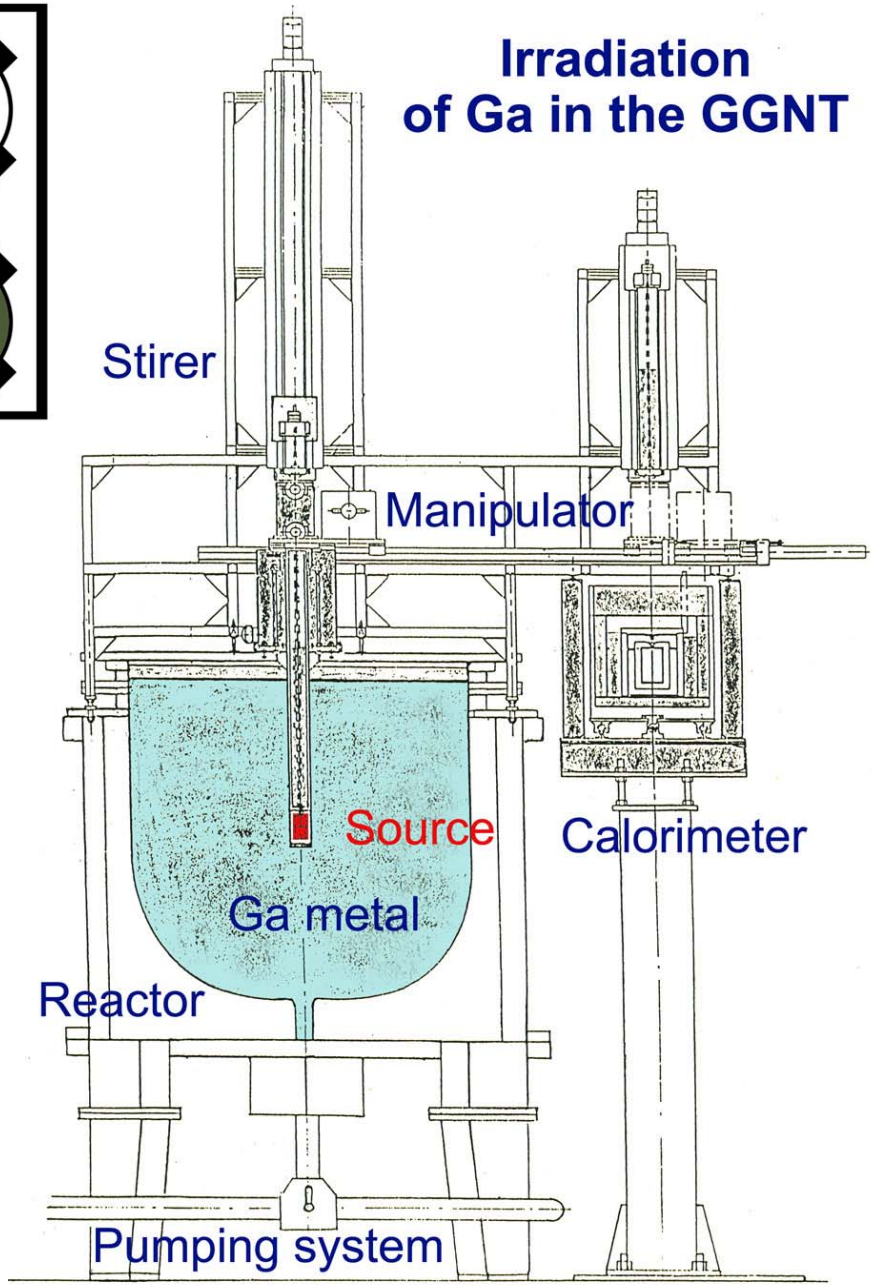
A plan view of the reactors layout in the laboratory



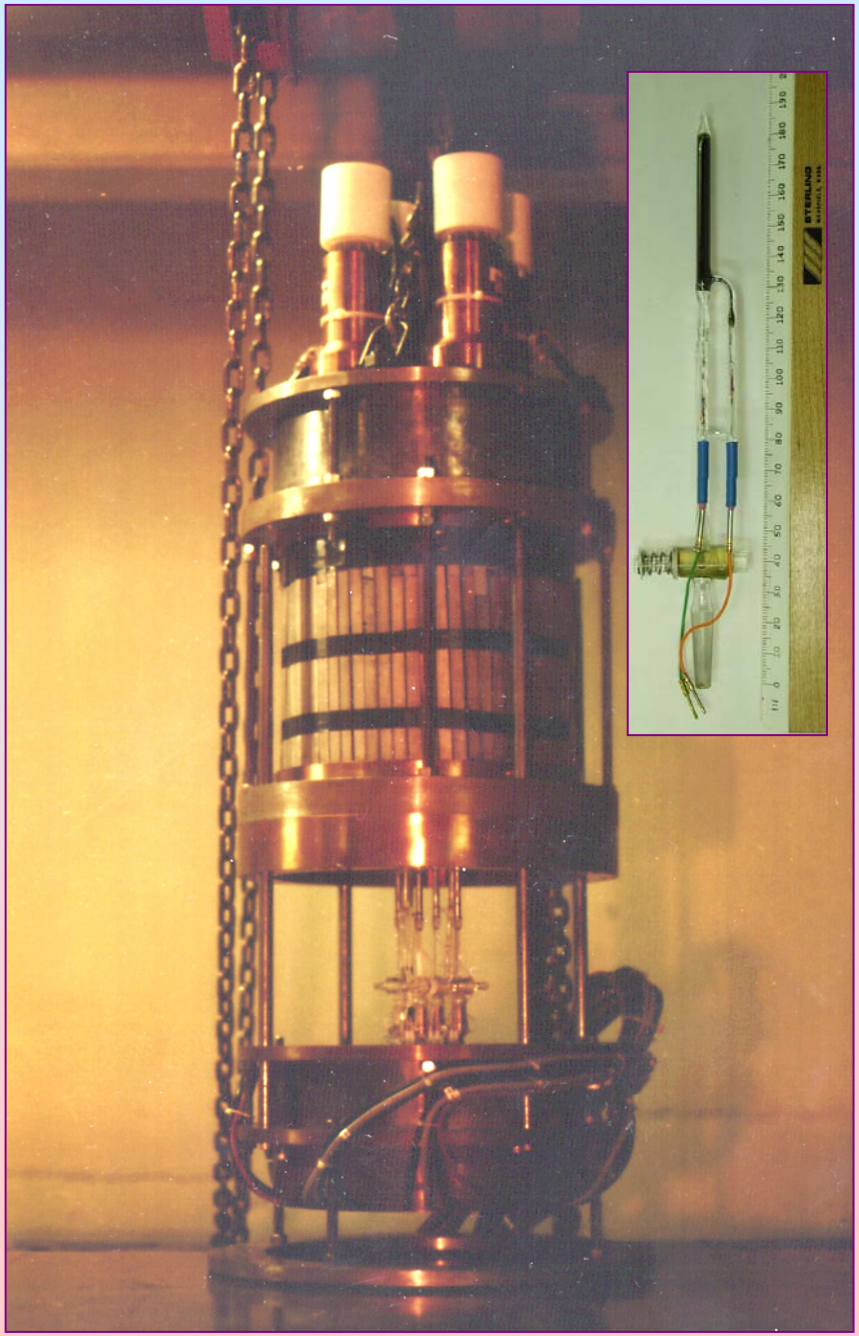


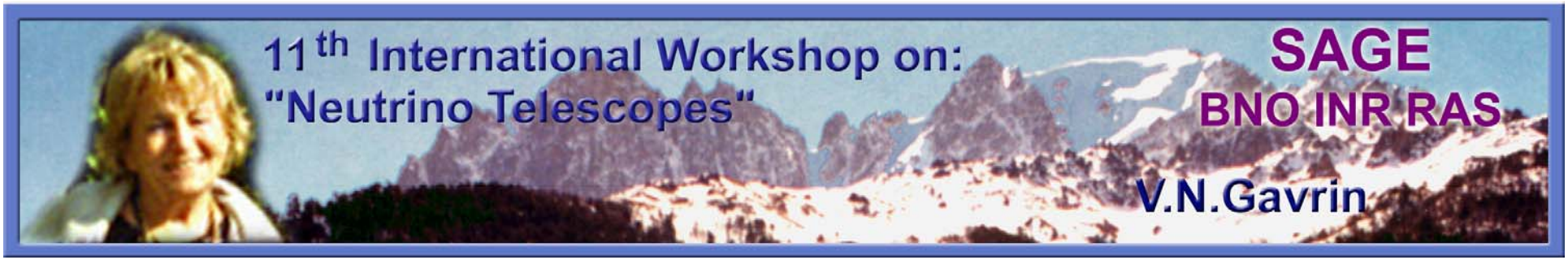


Measured production rate

Ten extractions were made. Exactly the same procedures were used to extract ^{71}Ge, to measure efficiency of extraction, to select candidates ^{71}Ge events as we use for solar neutrino runs.

The times of occurrence of the candidate ^{71}Ge events were analyzed with our standard maximum-likelihood program (Cleveland, 83) to separate the ^{71}Ge 11.4-d decay events from a constant rate background. This is the same program that we used to analyze the runs with the ^{51}Cr source and use to analyze all solar neutrino data.

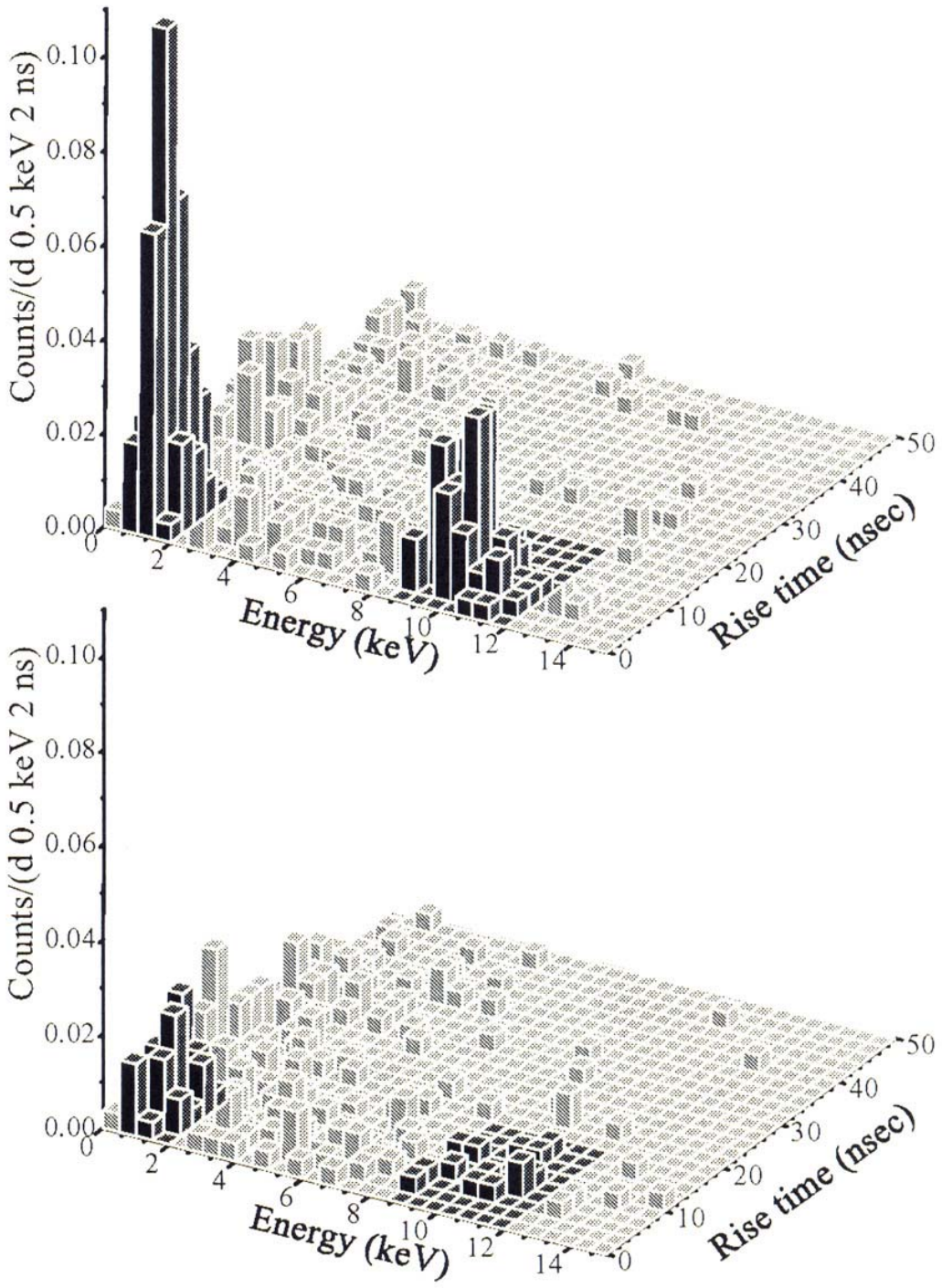




Results of analysis of L- and K-peak events. All production rates are referred to the time of the start of the first exposure. The parameter Nw2 measures the goodness of fit of the sequence of event times. The probability was inferred from Nw2 by simulation.

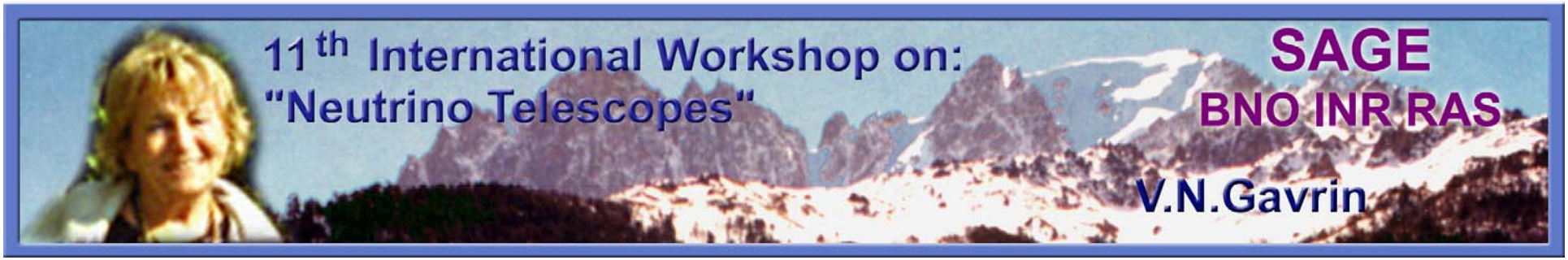
Extraction name	Extraction date (2004)	Number of candidate events	Number fit to ^{71}Ge	Number of events assigned to		^{71}Ge production rate by ^{37}Ar source (atoms/day)	Nw^2	Probability (percent)
				^{37}Ar source production	Solar ν production			
Ar1	6 May	28	20.1	19.4	0.7	$10.3^{+3.2}_{-2.8}$	0.065	60
Ar2	14 May	48	29.9	28.7	1.2	$10.5^{+2.5}_{-2.2}$	0.048	73
Ar3	29 May	69	52.9	51.3	1.6	$14.5^{+2.3}_{-2.1}$	0.110	35
Ar3-2	30 May	13	2.4	2.3	0.1	$0.8^{+1.0}_{-0.8}$	0.273	7
Ar4	13 Jun	45	25.4	23.8	1.6	$9.5^{+2.4}_{-2.2}$	0.142	13
Ar5	28 Jun	38	25.6	23.8	1.8	$11.5^{+2.9}_{-2.6}$	0.108	29
Ar6	13 Jul	34	11.6	9.7	1.9	$6.5^{+3.2}_{-2.7}$	0.042	81
Ar7	28 Jul	18	8.4	6.7	1.7	$6.1^{+3.3}_{-2.7}$	0.079	43
Ar8	12 Aug	29	12.8	11.2	1.6	$14.5^{+6.3}_{-5.5}$	0.055	68
Ar9	27 Aug	20	9.0	7.3	1.7	$12.1^{+6.6}_{-6.1}$	0.068	58
Ar10	11 Sep	34	6.7	5.1	1.6	$12.0^{+9.1}_{-7.3}$	0.293	3
Combined		363	203.4	188.0	15.4	$11.0^{+1.0}_{-0.9}$	0.063	55

For all runs combined the best fit rate is 11.0 +1.0/-0.9 atoms of ^{71}Ge produced by the source at the reference time. The uncertainty is purely statistical and is given with 68% confidence.



Upper panel: energy vs rise-time histogram of all events after time cuts observed in all ten exposures during the first 30 days after extraction. The live time is 263.1 days and 443 events are shown. The expected location of the ^{71}Ge *L* and *K* peaks based on the ^{55}Fe calibrations is shown darkened.

Lower panel: the same histogram for the 227 events that occurred during an equal live-time interval beginning at day 100 after extraction. The ^{71}Ge has decayed away and is absent. The number of events outside the peaks is about the same in both panels as these are mainly due to background.



Origin of uncertainty	Uncertainty (%)
Chemical extraction efficiency	
Mass of added Ge carrier	2.1
Amount of Ge extracted	3.5
Carrier carryover	0.5
Mass of gallium	0.5
Chemical extraction subtotal	4.1
Counting efficiency	
Calculated efficiency	
Volume efficiency	0.5
Peak efficiency	2.5
Simulations to adjust for counter filling	1.7
Calibration statistics	
Centroid	0.1
Resolution	0.3
Rise time cut	0.6
Gain variations	+0.5
Counting efficiency subtotal	+3.2, −3.1
Residual radon after time cuts	−1.7
Solar neutrino background	0.4
^{71}Ge carryover	0.0
Total systematic uncertainty	+5.2, −5.4

Summary of the contributions to the systematic uncertainty in the measured neutrino capture rate.

The quadratic combination of all these systematic uncertainties is +5.2/-5.4%. The measured production rate in the *K* and *L* peaks, including both statistical and systematic errors, is thus

$$p_{measured} = 11.0\ ^{+1.0}_{-0.9}\ (stat) \pm 0.6\ (syst)$$

Measurement of source activity

A. Measurements at Zarechny

(1) In the first method, carried out after argon purification, and while the gas was being put into the source holder, the total volume of gas and its isotopic composition were measured. The composition was determined with a mass spectrometer. The difference in pressure between before filling the source holder and after filling implied the volume of gas in the holder was 2.665±0.048 l at STP. Combining this with the isotopic composition and correcting for decay between the time of volume measurement and the reference time gives a source activity of 409±6 kCi at 04:00 on 30 April 2004. The stated uncertainty has 68% confidence and includes all known systematics.

Gas content of the ^{37}Ar source 47.5 h prior to the reference time in percent by volume. The uncertainty shown is statistical; there are additional systematic components whose sum is no more than 0.8%.

H_2	^{37}Ar	^{38}Ar	^{39}Ar	^{40}Ar
0.26 ±0.07	96.57 ±0.13	1.87 ±0.06	0.35 ± 0.03	0.95 ± 0.03

(2) In the second method, the source holder was evacuated and weighed before filling and then weighed again after filling with the extracted gas sample. The difference in mass was 4.400±0.042 g at the time of filling (06:25 on 28 April), from which the activity is calculated to be 412±4 kCi at the reference time.

A. Measurements at Zarechny

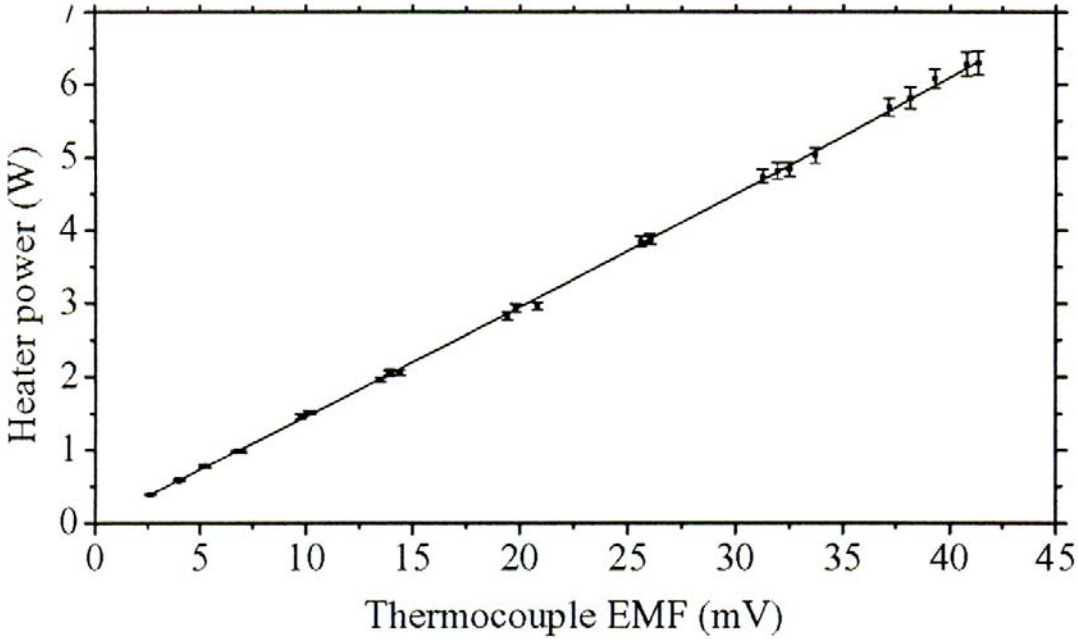
(3) In the third method, the heat output of the source was measured in a massive calorimeter specially built for this purpose. The calorimeter was calibrated using electrical heaters of known power and the thermocouple EMF over the range of (6-8) W (the expected source power) was found to have the constant value 0.1019±0.0002 W/mV. Stabilization of the calorimeter with the source required only 3 h and the measured thermocouple EMF was 65.9 mV at 22:00 on 28 April 2004. Applying a decay factor of 0.9740 gives a power of 6.54±0.04 W at our reference time. Using the conversion factor gives the source activity at this time as 405±4 kCi. The error estimate includes the calibration uncertainty, the errors in the calorimeter measuring circuits, and the uncertainties in both decay energy and ^{37}Ar half-life.

Summary of measurements at Zarechny

Measurement method	Activity (kCi 37Ar at 04:00 on 30 April 2004)
Volume of gas	409 ± 6
Mass of gas	412 ± 4
Calorimetry	405 ± 4

The weighted average value is 409 ± 3 kCi

B. Calorimetric measurement at Baksan

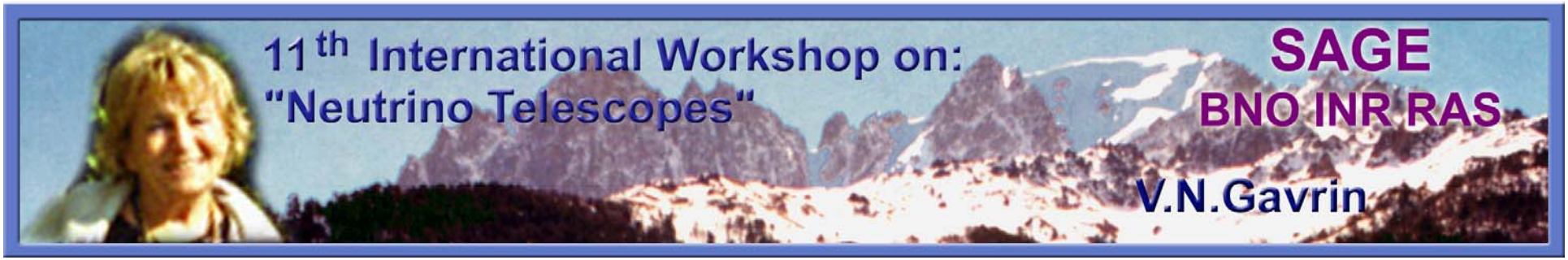


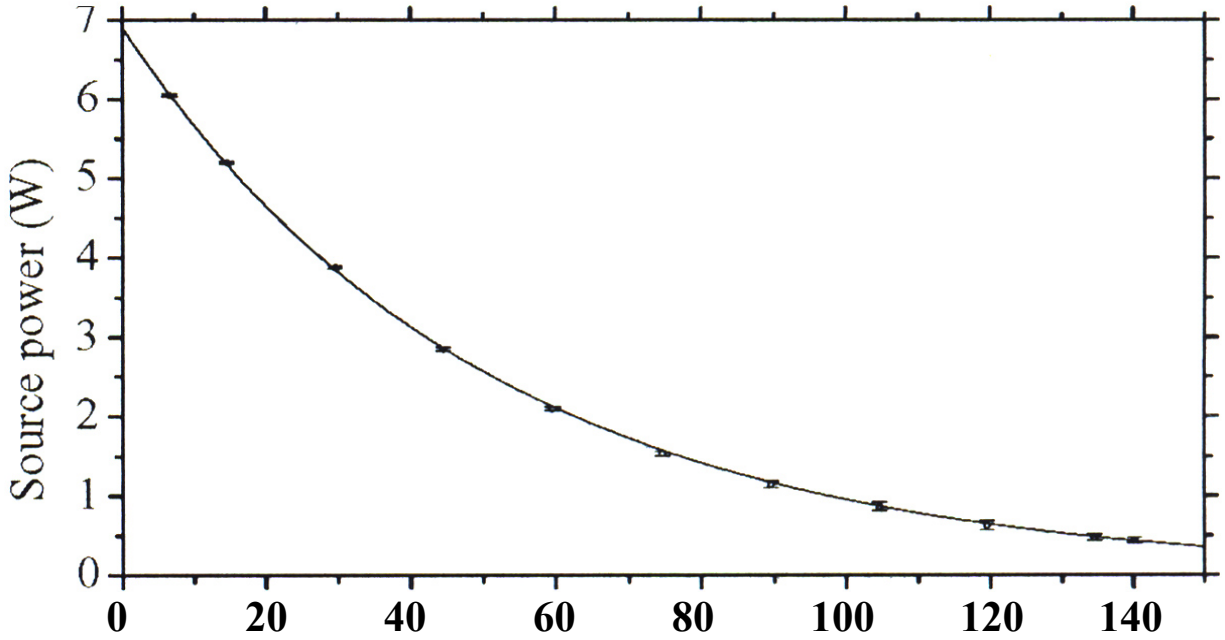
Calibration curve of the calorimeter at Baksan. The solid curve is a weighted least-squares fit to a 2nd-degree polynomial.

$p(v) = a+bv+cv2$ gives $a = 0.022\pm0.011$ W, $b = 0.1409 \pm 0.0022$ W/mV, and $c = 0.00028 \pm 0.00007$ W/mV^2. The uncertainties were used as weight factors in this fit and χ^2 is 19.5 with 26 degrees of freedom (probability = 81%).

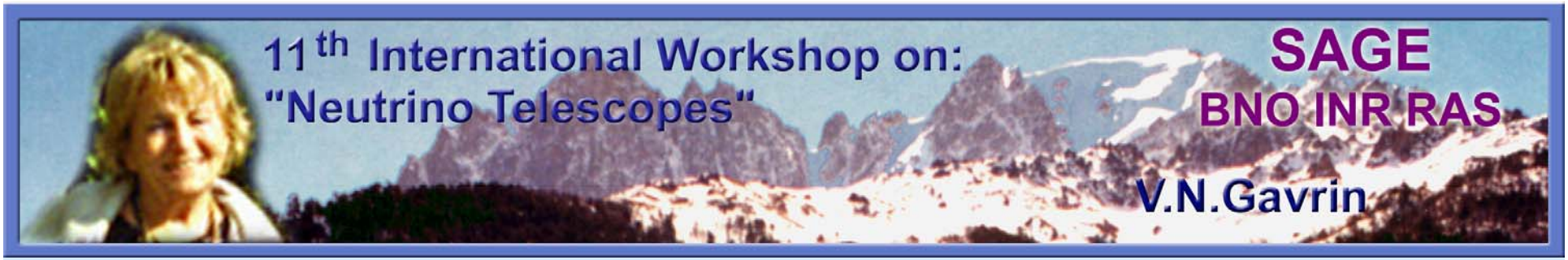
Measurements of the source power with the calorimeter at Baksan.

Days after 04:00 on 30 April 2004	Thermocouple voltage (mV)	Deduced power (W)	Power at 04:00 on 30 April 2004 (W)
6.55	39.661	6.051 ± 0.018	6.888 ± 0.020
14.55	34.415	5.203 ± 0.018	6.938 ± 0.024
29.55	26.024	3.878 ± 0.020	6.959 ± 0.037
44.55	19.338	2.851 ± 0.021	6.883 ± 0.051
59.54	14.290	2.093 ± 0.023	6.795 ± 0.074
74.55	10.490	1.531 ± 0.027	6.690 ± 0.119
89.54	7.841	1.144 ± 0.044	6.725 ± 0.261
104.54	5.928	0.867 ± 0.051	6.858 ± 0.404
119.55	4.268	0.628 ± 0.057	6.689 ± 0.610
134.55	3.189	0.474 ± 0.034	6.790 ± 0.488
140.21	2.942	0.439 ± 0.035	7.031 ± 0.567





If a weighted fit is made to this data with a decaying exponential whose half-life is fixed at 35.04 d (the half-life of ^{37}Ar), then the power at the reference time is 6.907±0.013 W. χ^2 for this fit is 11.2 with 10 degrees of freedom (probability = 34%). As a check, the same fit was made allowing the decay constant to be a free variable, along with the power at the reference time. The resultant best fit half-life is 34.80±0.20 d, in agreement with the known value. χ^2/DOF = 9.8/9 for this fit.



Using the energy release and the conversion factors and $3.7 \cdot 10^{10}$ decays of $^{37}Ar/(Ci \cdot s)$, the inferred source activity at the reference time was 426.9±0.8 kCi. The quoted uncertainty here of 0.2% is solely from the measurement errors. There are several additional systematic uncertainties that must be included in a full error estimate.

- the differences in thermal properties between the source and the calibration heaters;
- in the energy release;
- incomplete absorption of the IB component of energy release;
- in the ^{37}Ar half-life;
- in the capture of some of the gamma rays from the source in the other part of the calorimeter

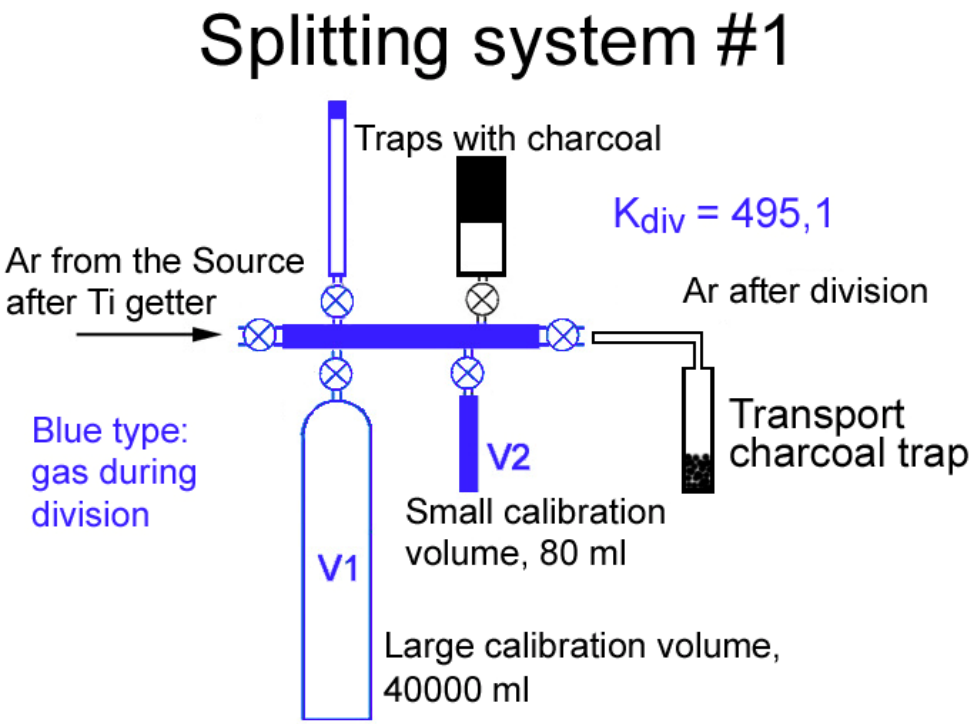
---

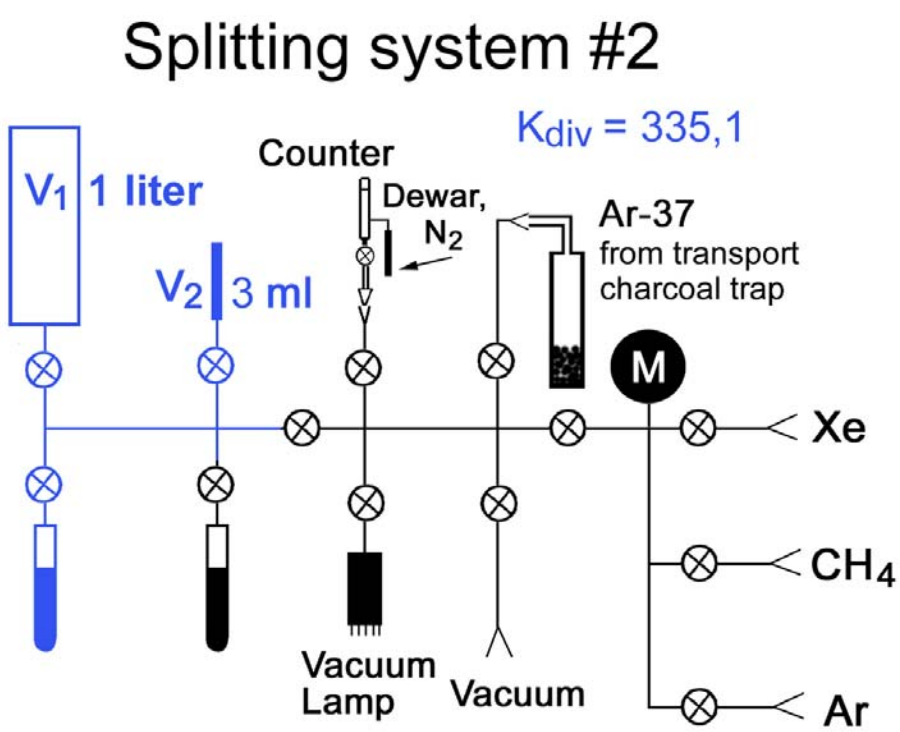
We assign a total error of ~ +/- 2% or +/- 9 kCi

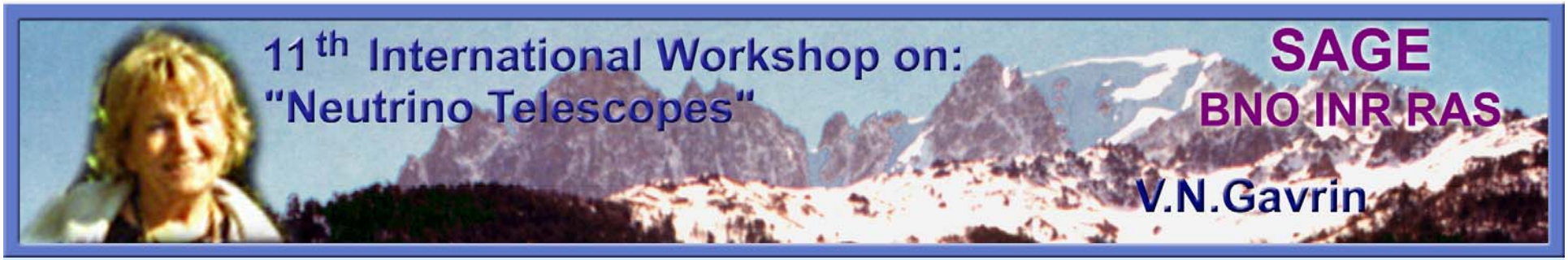
The final result of calorimetric measurements at Baksan is 426.9±9

C. Measurement by ^{37}Ar counting

The ^{37}Ar source was returned to the fabrication facility in December 2004. The source holder was cut open in a vacuum system, the entire gas sample was removed, and samples of the gas were taken for activity measurement in proportional counters. At this time the ^{37}Ar had decayed by a factor of 300. Because the specific activity was still very high, it was necessary to make several volume divisions to reduce the count rate to a value that was measurable in a proportional counter.







Four samples were taken in two proportional counters using different methods of volume division. Assuming an ^{37}Ar half life of 35.04 d, the inferred source strength at the reference time is 383.3 ± 4.3 kCi where the uncertainty includes all known systematic effects except for the half-life. Since the time delay from the reference time to the time of these measurements was 288 d, this result is rather sensitive to the value of the half-life that is used in the decay correction. The ^{37}Ar half-life uncertainty in the most recent data compilation for this nuclear mass is given as ±0.04 d, which leads to an additional uncertainty in the source strength of ±0.6%.

D. Measurements in progress

Another sample will have the ^{37}Ar content determined by the method of isotopic dilution.

E. Summary of source strength measurements

Summary of different activity measurements.
The stated uncertainty includes all known systematics.

Measurement method	Activity (kCi 37Ar at 04:00 on 30 April 2004)
Volume of gas	409 ± 6
Mass of gas	412 ± 4
Calorimetry at Zarechny	405 ± 4
Calorimetry at Baksan	427 ± 9
Proportional counter	383 ± 4

The five completed activity measurements are given in the Table. The three Zarechny measurements agree quite well, but the Baksan calorimetric measurement is distinctly higher and the proportional counter measurement is distinctly lower. The large spread among these measurements must be due to presently undetermined systematic effects; further work is underway to attempt to understand the causes of this disagreement. Until this disagreement is resolved, the weighted average value of the Zarechny measurements, 409 ± 3 kCi, will be used.

Predicted production rate

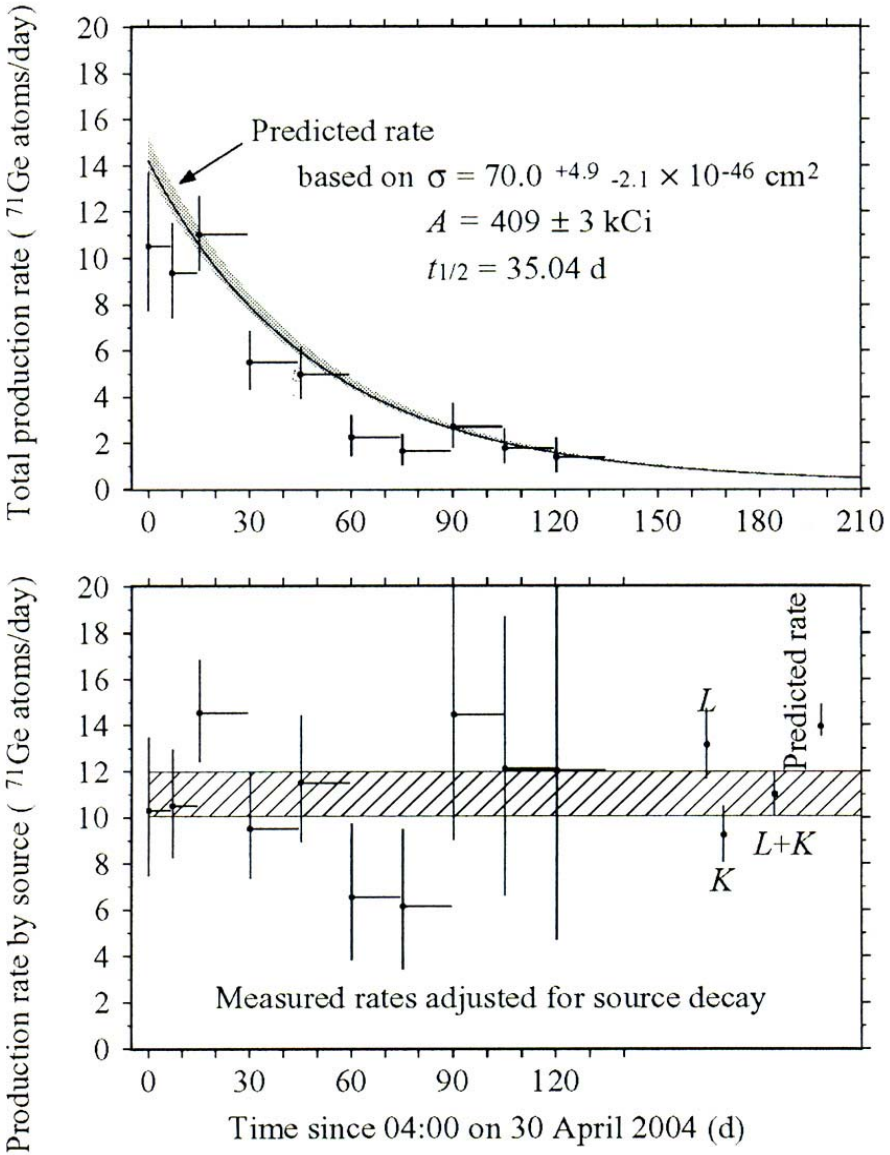
$$p = AD\langle L\rangle\sigma, \qquad \langle L\rangle = \frac{1}{4\pi V_S}\int_{\text{absorber}} dV_A \int_{\text{source}} \frac{dV_S}{r_{SA}^2}$$

Values and uncertainties of the terms that enter the calculation of the predicted production rate. All uncertainties are symmetric except for the cross section.

Term	Value	Uncertainty	
		Magnitude	Percentage
Atomic density $D = \rho N_0 f_I / M$			
Ga density ρ (g Ga/cm^3) [16]	6.095	0.002	0.033
Avogadro's number N_o (10^{23} atoms Ga/mol)	6.0221	0.0	0.0
^{71}Ga isotopic abundance f_I (atoms ^{71}Ga/100 atoms Ga)[17]	39.8921	0.0062	0.016
Ga molecular weight M (g Ga/mol) [17]	69.72307	0.00013	0.0002
Atomic density D (10^{22} atoms ^{71}Ga/cm^3)	2.1001	0.0008	0.037
Source activity at reference time A (10^{16} ^{37}Ar decays/s)	1.513	0.011	0.7
Cross section σ [10^{-46} cm^2/(^{71}Ga atom ^{37}Ar decay)] [6]	70.0	+4.9, −2.1	+7.0, −3.0
Path length in Ga $\langle L\rangle$ (cm)	72.6	0.2	0.28
Predicted production rate (^{71}Ge atoms/d)	14.0	+1.0, −0.4	+7.0, −3.1

Assuming a source activity of 409±3 kCi, and combining the uncertainty terms in quadrature, the predicted production rate is thus

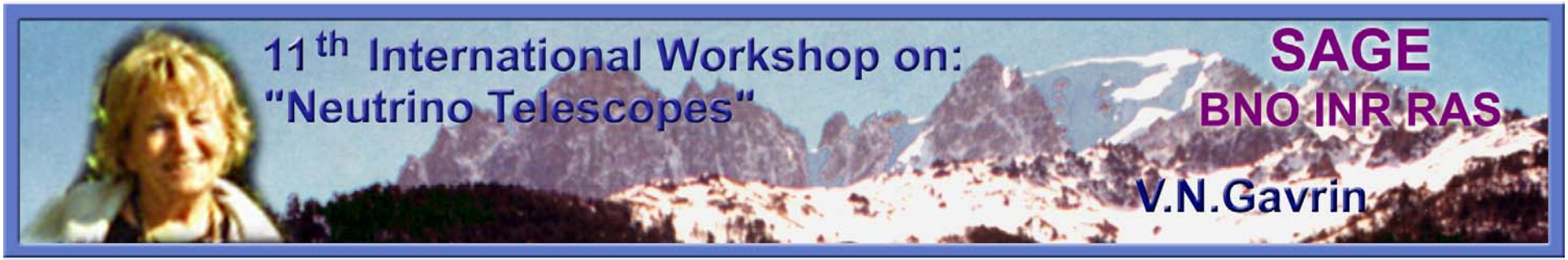
$p_{\text{predicted}}$ = 14.0 +1.0/ -0.4 atoms of ^{71}Ge produced per day.



Upper panel: **comparison of measured total production rate for each extraction with predicted rate.**

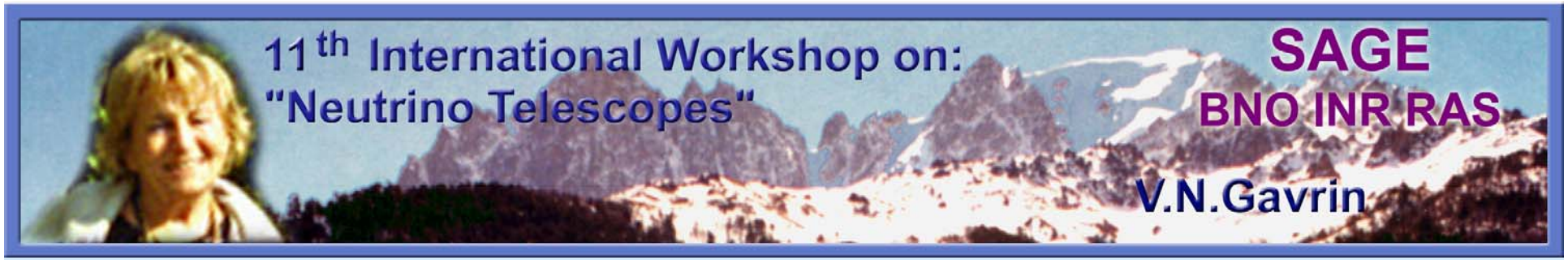
Lower panel: **measured rates from the ^{37}Ar source extrapolated back to the start of the first extraction. The combined results for events in the the *L*- and *K*- peaks and for all events are shown separately at the right and compared to the predicted rate.**

$$\frac{p_{\text{measured}}}{p_{\text{predicted}}} = \frac{11.0^{+1.0}_{-0.9}\ (\text{stat})\ \pm 0.6\ (\text{syst})}{14.0^{+1.0}_{-0.4}} = 0.79^{+0.09}_{-0.10}$$



Comparison of source experiments with Ga. Values for the ^{37}Ar source marked with a dagger (t) are preliminary. When two uncertainties are given, the first is statistical and the second is systematic. When one uncertainty is given, statistical and systematic uncertainties have been combined in quadrature.

Item	GALLEX Cr1	GALLEX Cr2	SAGE ^{51}Cr	SAGE ^{37}Ar
Source production				
Mass of reactor target (kg)	35.5	35.6	0.512	330
Target isotopic purity	38.6% ^{50}Cr	38.6% ^{50}Cr	92.4% ^{50}Cr	96.94% ^{40}Ca (natural Ca)
Source activity (kCi)	1714^{+30}_{-43}	1868^{+89}_{-57}	516.6 ± 6.0	†(409 ± 3)
Specific activity (kCi/g)	0.048	0.052	1.01	†93.0
Gallium exposure				
Gallium mass (tonnes)	30.4 ($GaCl_3$:HCl)	30.4 ($GaCl_3$:HCl)	13.1 (Ga metal)	13.1 (Ga metal)
Gallium density (10^{21} ^{71}Ga/cm^3)	1.946	1.946	21.001	21.001
Measured production rate p (^{71}Ge/d)	$11.9 \pm 1.1 \pm 0.7$	$10.7 \pm 1.2 \pm 0.7$	$14.0 \pm 1.5 \pm 0.8$	†$(11.0 \pm 1.0 \pm 0.6)$
$R = p_{measured}/p_{predicted}$	$1.01^{+0.12}_{-0.11}$	$0.84^{+0.12}_{-0.11}$	0.96 ± 0.12	(?)



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