

*Milano 21 e 22 Settembre 2017*

*The second radon-in-field international intercomparison for passive measurement devices: dwellings and workplaces*

**Metrological aspect of in-field radon intercomparison**



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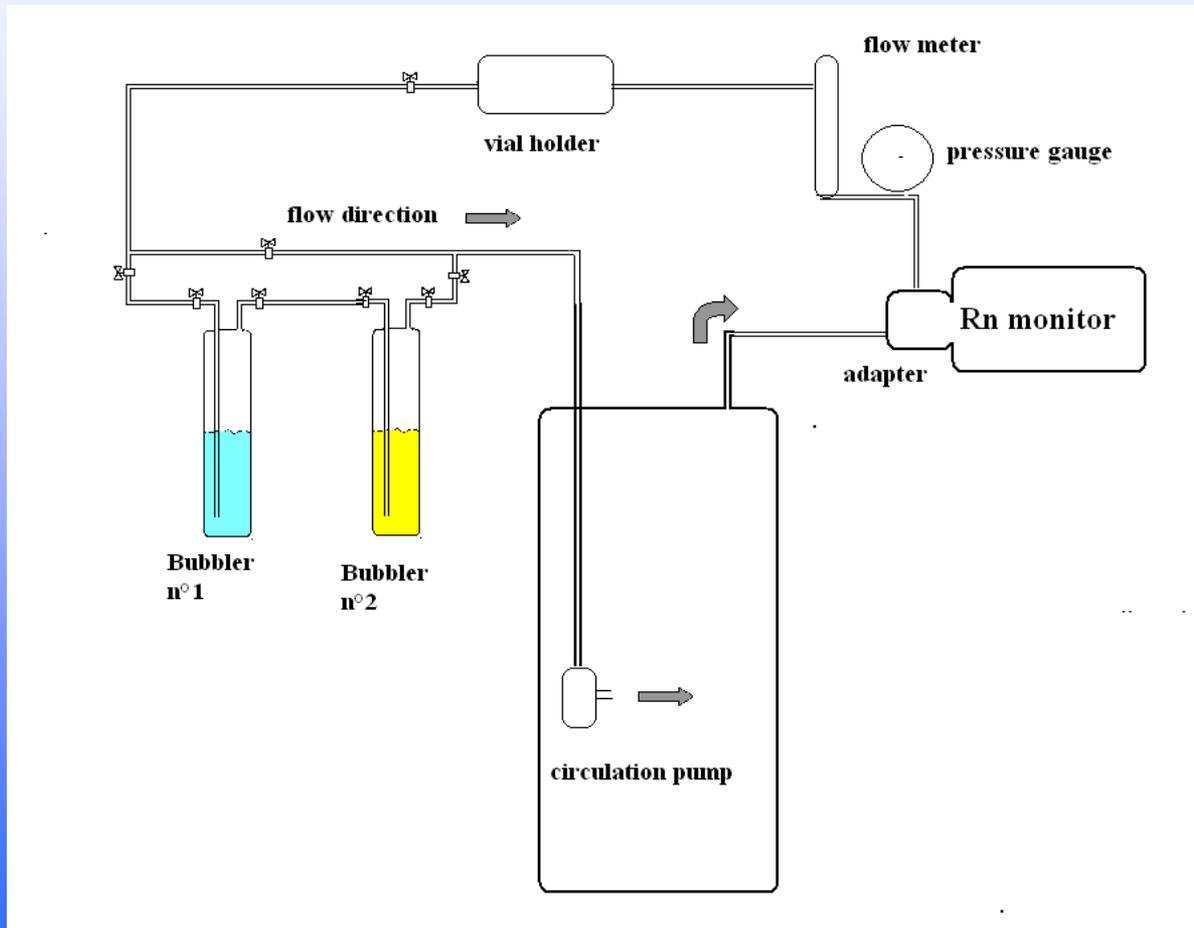
# Radon metrology and calibration facilities at the Italian Metrological Institute for Ionizing Radiation ENEA INMRI

## Main issues of the presentation

- Primary Radon standard for monitor calibration
- Reproducibility and linearity test
- Radon standard not based on radium solution
- Comparison with foreign Metrological Institutes
- Measurement of “blank indication” of radon monitors
- Influence of thoron on radon measurement.

# Radon Reference Measurement System (RRMS) for monitor calibration and measurement of radon sources.

Reference Rn-222 concentration is achieved by putting a known Rn activity in the known volume of the circuit. Volume of the circuit: 112,24 litre without bubblers



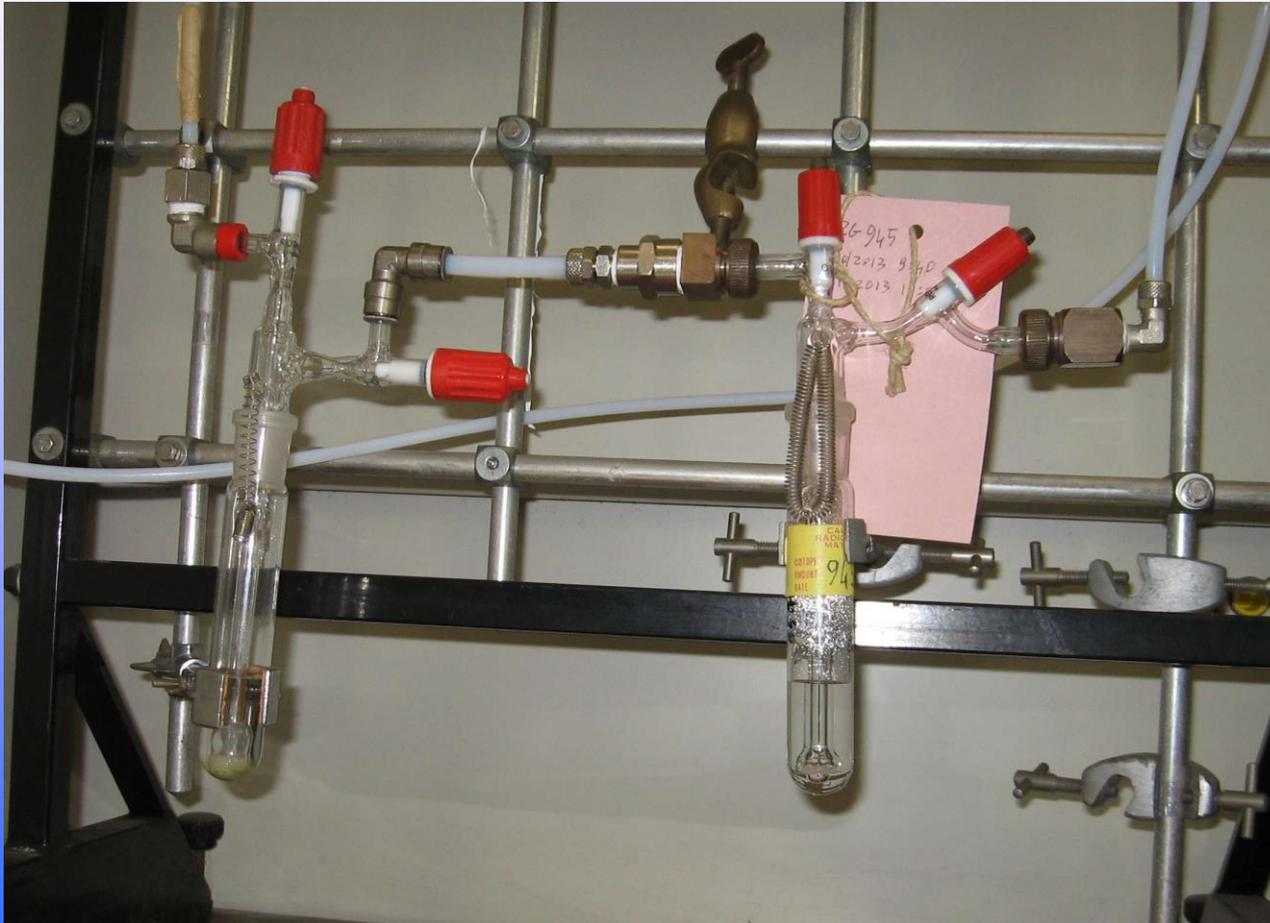
Details of the RRMS: Radon monitor based on scintillation cell, flow meter and power supply for the circulation pump, on the wall is fixed the hygrometer. Below you see the upper part of the main vessel.



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## Bubblers for Radon production in RRMS

Right bubbler contains a reference radium solution traceable to a NIST standard  
Activity  $\approx 1600$  Bq. Left bubbler contains only water to increase humidity of the air  
and avoid evaporation from Ra-226 solution. Air flows from left to right.



**Lucas Scintillation Cell of the radon monitor in RRMS.  
This device is very simple, reliable and sensible: nearly 76%  
 $\alpha$  emitted inside the cell are detected.**

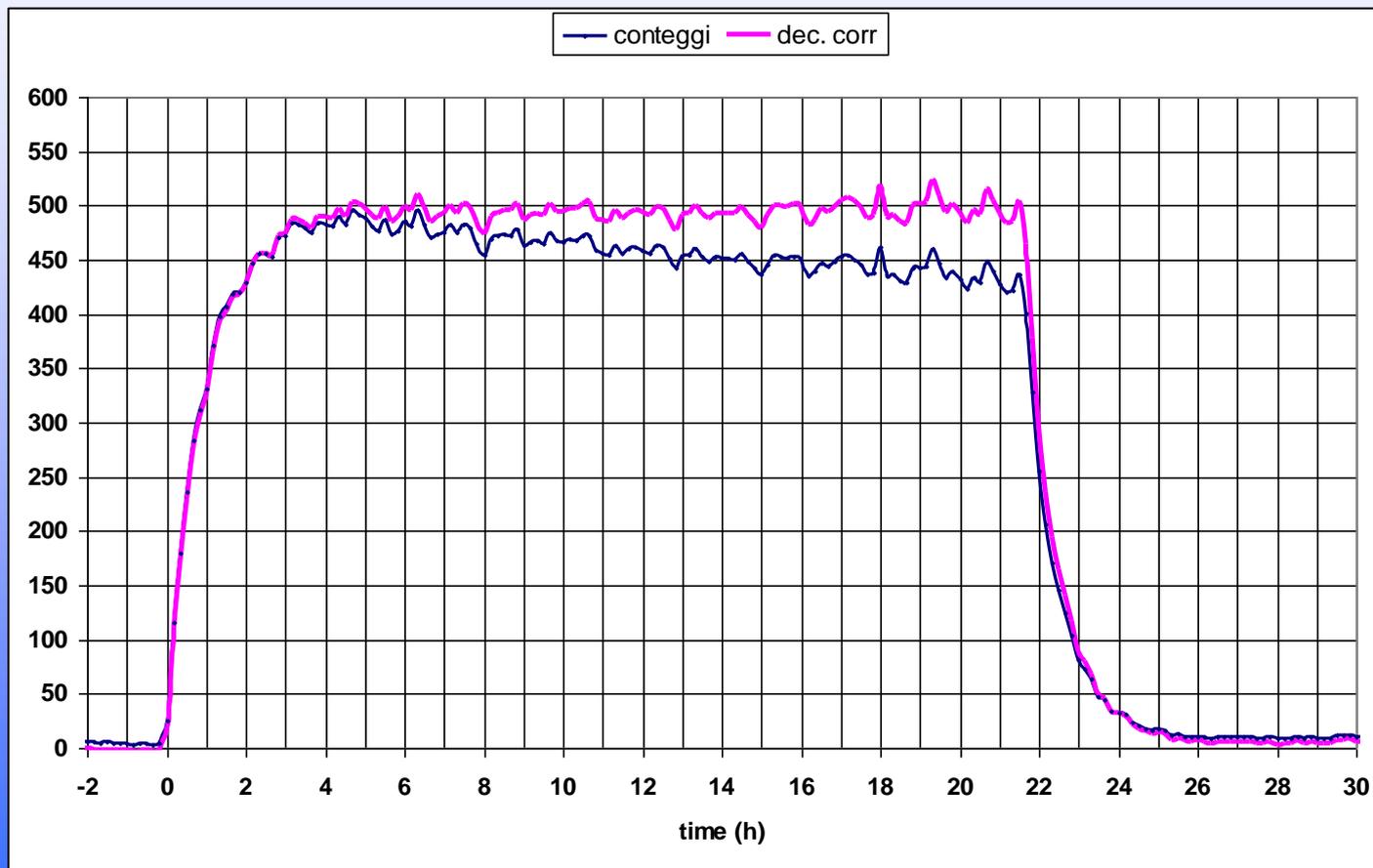


Counting rate of the monitor on the RRMS during a calibration test.  
Blue curve are the actual counts, pink curve are counts with decay correction.

Test data and result:

Source activity 1520 Bq; Rn activity conc. 13538 Bqm<sup>-3</sup>; count rate 457,8 cpm

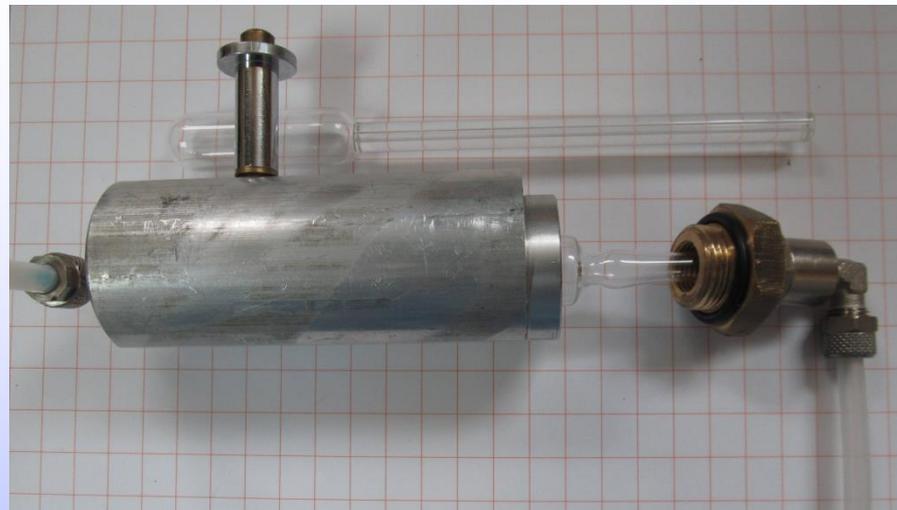
Monitor calibration factor measured in this test: 29.56 Bqm<sup>-3</sup> / cpm



# Reproducibility, linearity and stability of radon measurement system



Flame sealing of a glass vial



Device for Rn transfer from vial to RRMS

- Radon gas is sealed in a glass vial
- The vial is measured in ionization chamber (I.C.) and ionization current is recorded.
- Radon is then transferred from the vial to radon measurement system (RRMS).
- Ionization current is compared with counting rate in RRMS, the ratio must be constant.
- Proper correction are applied to account for radon decay in the time lapse between the two measurement.
- If time lapse between the two measurement is long, linearity of RRMS is checked

# 4 $\pi\gamma$ Gas Ionization Chamber      Centronic IG 11

- Argon gas 20 atm.
- Well diameter 25 mm, well length 320 mm
- Reproducibility better than 0,1%

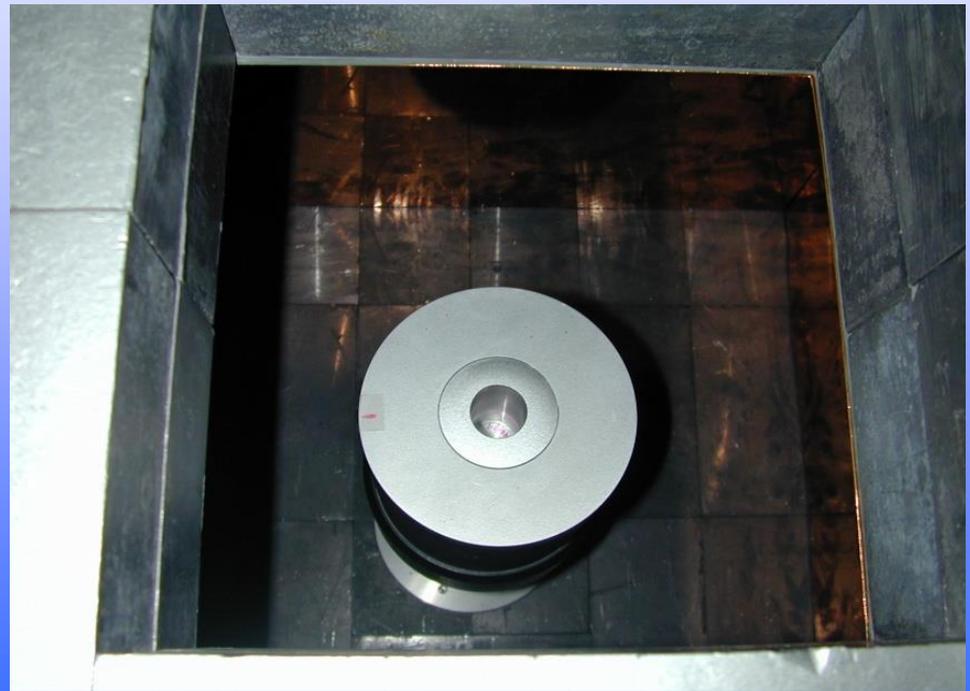


# Reproducibility, linearity and stability of radon measurement system

Sample	Date of measurement in IC (UTC)	Ionization current. nA	Date of measurement in RRMS (Local Time)	Counting rate in RRMS (cpm)	Radon activity in the vial (Bq)	IC calibration factor (kBq/nA)
PyI02	09/07/2009 12:16	1,503E-03	21/07/2009 08:10	848,6	23746	15796
PyI03	15/07/2009 15:09	4,338E-04	23/07/2009 09:00	508,7	6787	15646
Fil05	10/09/2009 14:30	1,949E-03	14/09/2009 16:55	4470,8	30815	15809
Fil06	16/09/2009 12:42	1,895E-03	22/09/2009 14:35	3005,8	29652	15649
Fil07	21/09/2009 13:16	1,007E-03	29/09/2009 11:40	1146,6	15833	15729
Fil10	15/10/2009 13:08	6,071E-04	20/10/2009 09:00	1205,0	9476	15607
Fil11	16/10/2009 08:40	1,156E-03	26/10/2009 14:30	848,4	17943	15528
<b>Small change in the vessel of RRMS</b>						
Fil12	14/12/2009 13:50	1,883E-03	16/12/2009 11:10	6346,8	29516	15673
Fil13	18/12/2009 08:21	1,640E-03	22/12/2009 09:00	3716,9	25470	15527
Fil17	21/01/2010 13:30	1,896E-03	22/01/2010 16:00	7320,0	29527	15576
Fil19	01/02/2010 14:52	1,788E-03	04/02/2010 11:30	5002,0	27739	15510
Fil21	06/02/2010 10:28	1,361E-03	12/02/2010 11:50	2138,2	21170	15555
Fil24	16/04/2010 12:40	1,408E-03	21/04/2010 09:20	2767,7	21894	15552
Fil25	02/12/2014 13:48	1,297E-03	09/12/2014 14:51	1728,0	20344	15687
<b>Statistics over all the measurement</b>					Average	<b>15632</b>
					Stand. dev.	<b>101</b>
					Stand. dev. %	<b>0,65%</b>
<b>Partial statistic after vessel change</b>					Average	<b>15583</b>
					Stand. dev.	<b>70</b>
					Stand. dev.%	<b>0,45%</b>

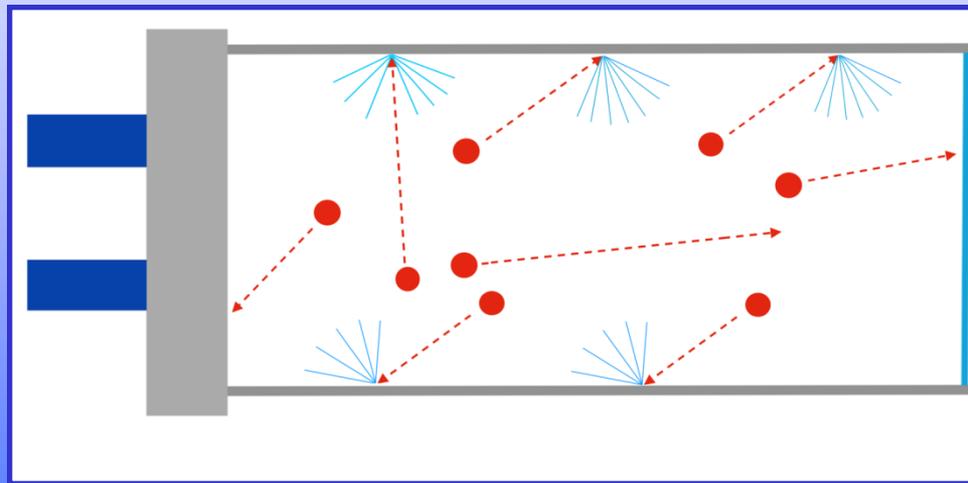
## Reference radon source with activity measured by absolute method

- Rn-222 gas (with air) is flame sealed in a glass vial (left picture).
- Gamma emission from the vials are measured in a well type NaI detector 5" x 5", geometry 4  $\pi$  (4  $\pi$   $\gamma$  integral counting) (right picture).
- Collected data are analysed by Giant3 Montecarlo code to compute the activity of the radon source.



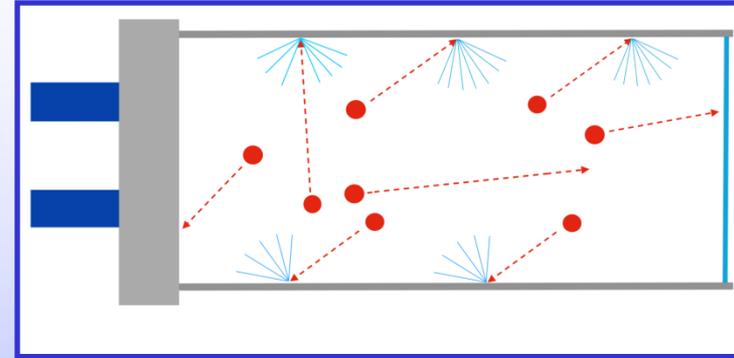
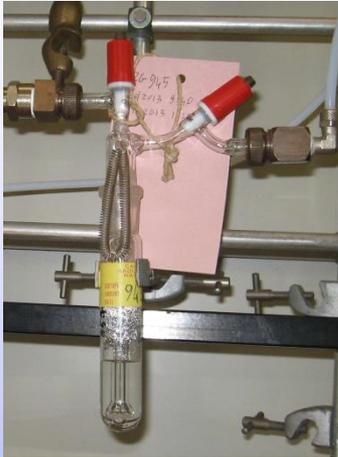
## Sensibility of Lucas Scintillation Cell was also computed by GEANT 4 Montecarlo code on the basys of simple assumptions.

- Rn-222 is distributed in the cell volume. Po-218 and Po-214 are plated on the cell walls
- $\alpha$  particles are emitted at random direction
- $\alpha$  particles travel in a strait line with a range fixed by their initial energy, thus by the emitting nuclide: Ranges are  $\approx 5.05$  cm and  $\approx 7.1$  cm for  $\alpha$  emission of Rn-222 and Po-214 respectively



Emitting nuclide and spatial distribution	$\epsilon$ [ENEA]
$^{222}\text{Rn}$ evenly distributed in cell volume	0.6541
$^{218}\text{Po}$ evenly distributed on wall	0.7233
$^{214}\text{Po}$ evenly distributed on wall	0.863

# Comparison of the calibration factor of LSC obtained with the three methods

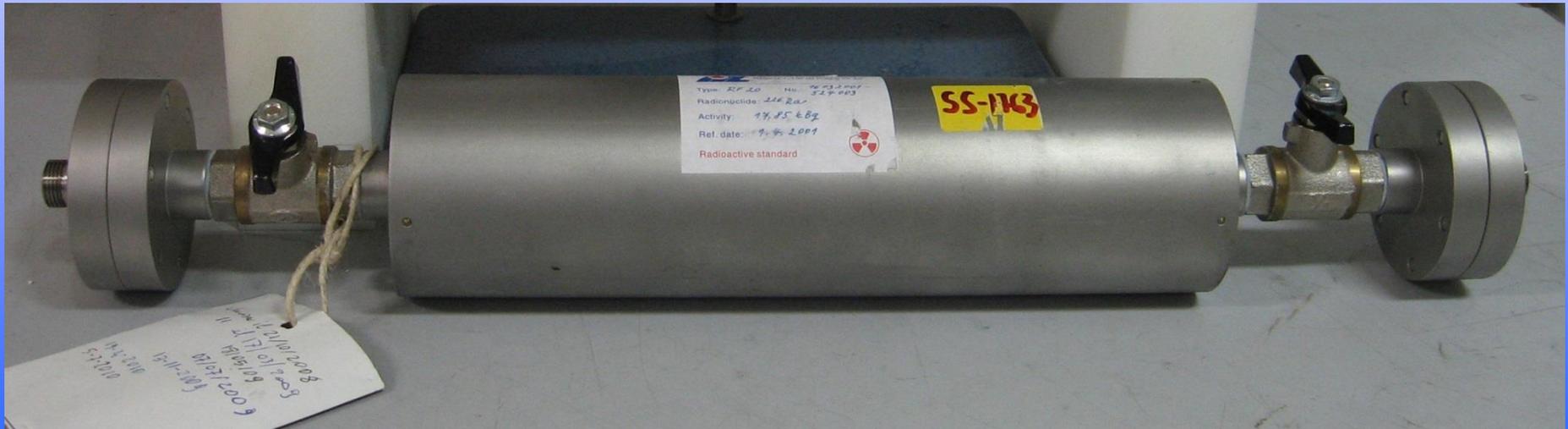


Reference Radium solution in bubblers  $FT = 29,45 \text{ (Bqm}^{-3}/\text{cpm)}$   
Rn in glass vials measured in NaI  $4 \pi \gamma$   $FT = 29,24 \text{ (Bqm}^{-3}/\text{cpm)}$   
Montecarlo simulation of scintillat. cell.  $FT = 29,23 \text{ (Bqm}^{-3}/\text{cpm)}$

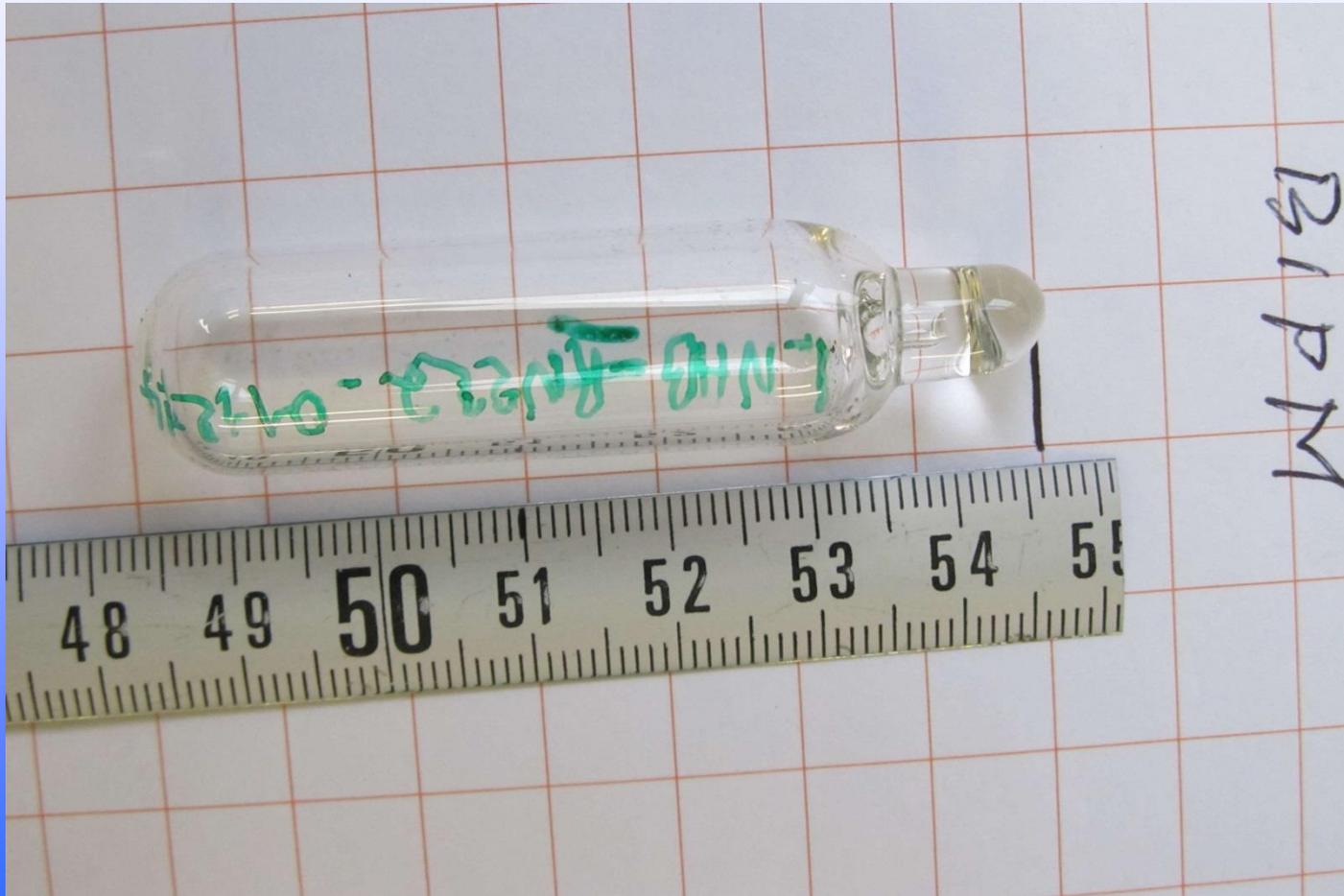
## Comparison with foreign Metrological Institutes:

Measure of Radon activity of reference source produced by Czech metrological Institute (CMI)

Radon source code	Rn activity certified by CMI (Bq)	Measure at ENEA INMRI (Bq)
Ceka RF 20	17742 ± 266 (1.5%)	17964 ± 215 (1.2%)
Ceka RF 180	187640 ± 2800 (1.5%)	187708 ± 2250 (1.2%)



On 5/12/2014 we received from BIPM this vial containing Rn-222 (500 kBq) for a SIR intercomparison. The result is positive within 1.3%.



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In June 2015 was held a new SIR intercomparison with 5 participants: Kriss North Korea, ENEA Italy , IFIN Romania, IRA Switzerland and LNHB France. Five samples in metallic cylinder previously measured at LNHB were sent to the 5 national laboratories.

Activity of our samples was  $105277 \pm 1180$  Bq al 29/06/2015 15:00 LT in Rome.



Official results are not yet available.

Three laboratories used the “defined solid angle” technique: Rn is frozen on the tip of a nail.

Two laboratories used different technique.

Calibration of radon monitor at constant radon concentration in one cubic meter radon chamber at ENEA INMRI.

In the first part of the experiment Rn was kept very low ( $\approx$ Bqm-3) for determination of the blank indication.

Main results of all the experiment are recorded for the quality system.

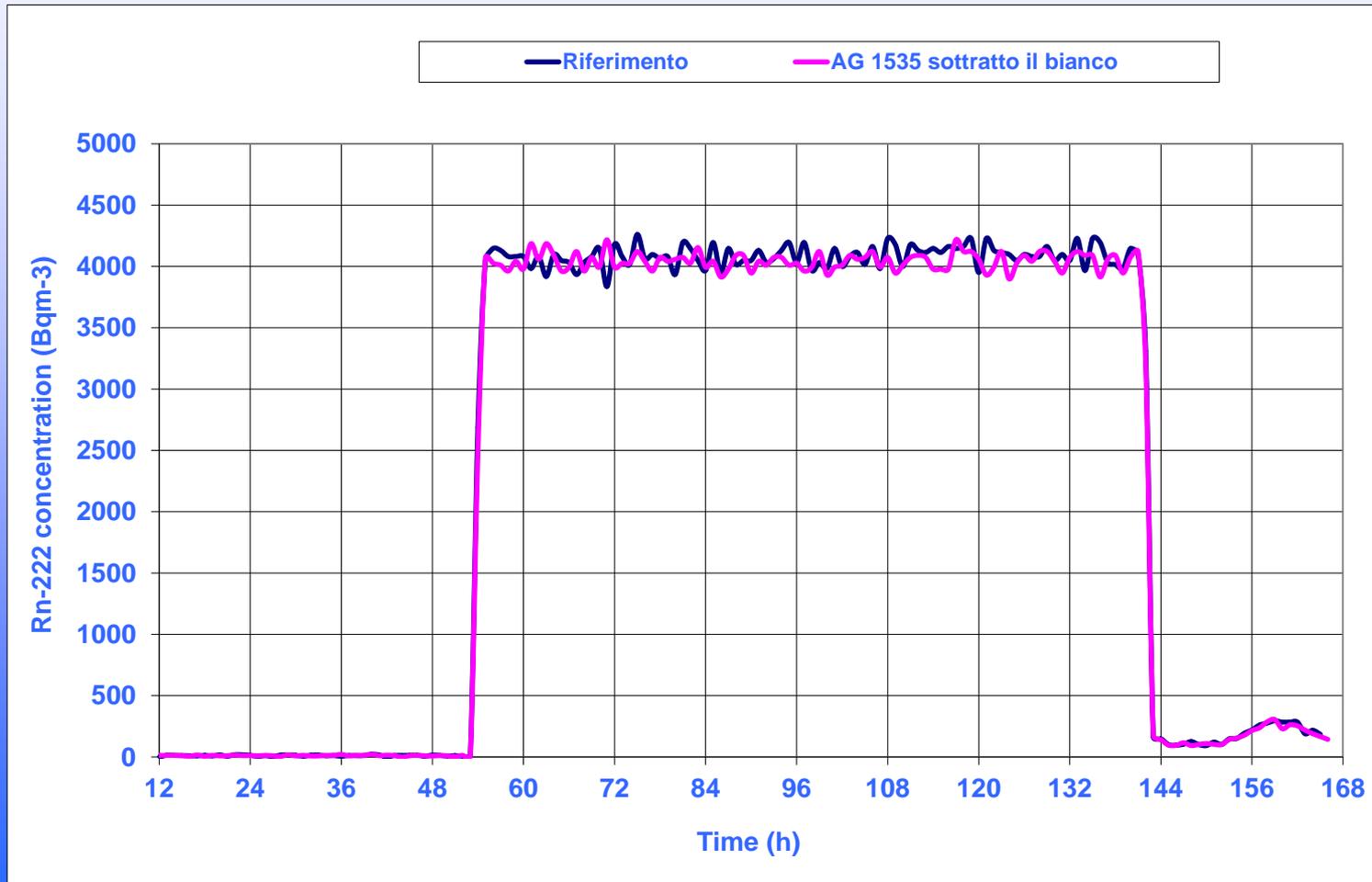


Table from quality system for AlphaGuard sn EF 1489 and Pylon source A273.  
 Same tables are recorded for 10 sources and for the three INMRI reference monitors.

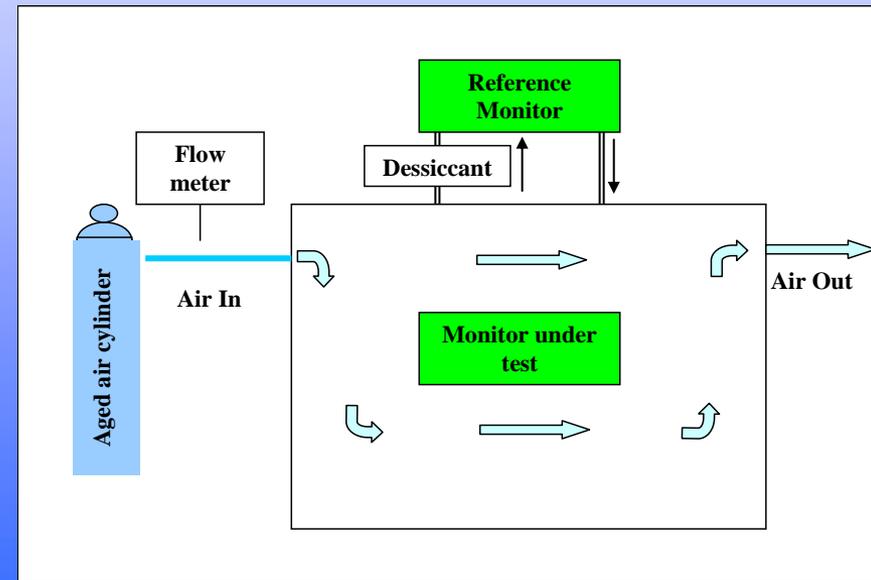
n° Exp	Date	Monitor	Rn Source	Average Monitor readings (Bqm-3)	dev.st of the series of measure	Readings corrected for blank and initial radon	Equivalent pressure at 22 °C
209	06/04/2006	AG INMRI 1498	Pylon A273	3813	1,9%	3817	987,3
221	13/10/2006	AG INMRI 1498	Pylon A273	3868	2,1%	3617	965,5
249	04/07/2007	AG INMRI 1498	Pylon A273	3941	2,0%	3926	984,4
341	10/07/2009	AG INMRI 1498	Pylon A273	4165	1,9%	3966	987,5
354	16/10/2009	AG INMRI 1498	Pylon A273	4148	2,0%	4112	1000,8
385	22/04/2010	AG INMRI 1498	Pylon A273	4191	1,9%	4028	989,2
394	18/06/2010	AG INMRI 1498	Pylon A273	4097	2,0%	3955	980,7
477	09/02/2012	AG INMRI 1498	Pylon A273	3939	2,3%	4097	993,6
483	12/03/2012	AG INMRI 1498	Pylon A273	4213	2,4%	4081	995,8
492	03/05/2012	AG INMRI 1498	Pylon A273	4186	1,9%	4013	987,9
504	28/06/2012	AG INMRI 1498	Pylon A273	4118	2,1%	3994	1009,8
656	26/06/2015	AG INMRI 1498	Pylon A273	4100	2,2%	4042	986,6
694	13/04/2016	AG INMRI 1498	Pylon A273	4057	2,1%	3990,4	985,6
698	01/06/2016	AG INMRI 1498	Pylon A273	4367	2,7%	4044	980,0
713	20/09/2016	AG INMRI 1498	Pylon A273	4085	2,0%	4052	984,9
728	01/12/2016	AG INMRI 1498	Pylon A273	4160	2,1%	4076	997,9
751	13/06/2017	AG INMRI 1498	Pylon A273	3987	2,1%	3953	980,3
			<b>AVERAGE</b>	<b>4062</b>		<b>4025</b>	
			<b>STD. DEV.</b>	<b>144</b>		<b>58</b>	
			<b>STD. DEV. %</b>	<b>3.5%</b>		<b>1.4%</b>	

Exp n°	data	Rn Bqm-3	AG1613	MR1sn50	AG tarato	MR1 tarato	AG/MR1
508	27/07/2012	4140	4076,9	138,64	4125,8	4081,1	1,011
509	28/09/2012	4140	4026,5	139,78	4074,8	4114,6	0,990
512	30/10/2012	9354	8889,5	311,50	8996,2	9169,6	0,981
513	06/11/2012	1536	1439,3	48,39	1456,6	1424,5	1,023
515	09/11/2012	21100	21119,0	721,1	21372,4	21227,0	1,007
516	16/11/2012	20627	20083,0	693,5	20324,0	20414,6	0,996
518	22/11/2012	4220	4126,2	141,0	4175,7	4150,6	1,006
519	28/11/2012	17352	16644,0	587,8	16843,7	17303,1	0,973
521	07/12/2012	45000	43696,0	1525,3	44220,4	44900,3	0,985
522	14/12/2012	9110	8890,0	308,24	8996,7	9073,7	0,992
528	22/02/2013	4000	3938,0	134,1	3985,3	3947,5	1,010
529	04/03/2013	1500	1422,0	45,84	1439,1	1349,4	1,066
531	15/03/2013	9200	9025,6	306	9133,9	9007,7	1,014
532	20/03/2013	21000	20797,6	719,5	21047,2	21179,9	0,994
533	04/04/2013	43300	42819,0	1530,8	43332,8	45062,2	0,962
534	12/04/2013	4150	4107,0	138,7	4156,3	4082,9	1,018
535	18/04/2013	76600	75768,0	2590,4	76677,2	76253,6	1,006
536	27/04/2013	105200	104127,0	3588,3	105376,5	105628,8	0,998
537	06/05/2013	1500	1418,0	47,9	1435,0	1410,0	1,018
539	10/05/2013	1052	8928,7	305,2	9035,8	8984,2	1,006
550	29/08/2013	30400	29674,0	1031,5	30030,1	30364,3	0,989
						media	0,999
						dev.st%	1,59%

Active radon monitors may exhibit non null blank indication (intrinsic instrument background) due to electronic noise and to detector poisoning or contamination by long lived radionuclides, e.g.  $^{210}\text{Pb}$ .

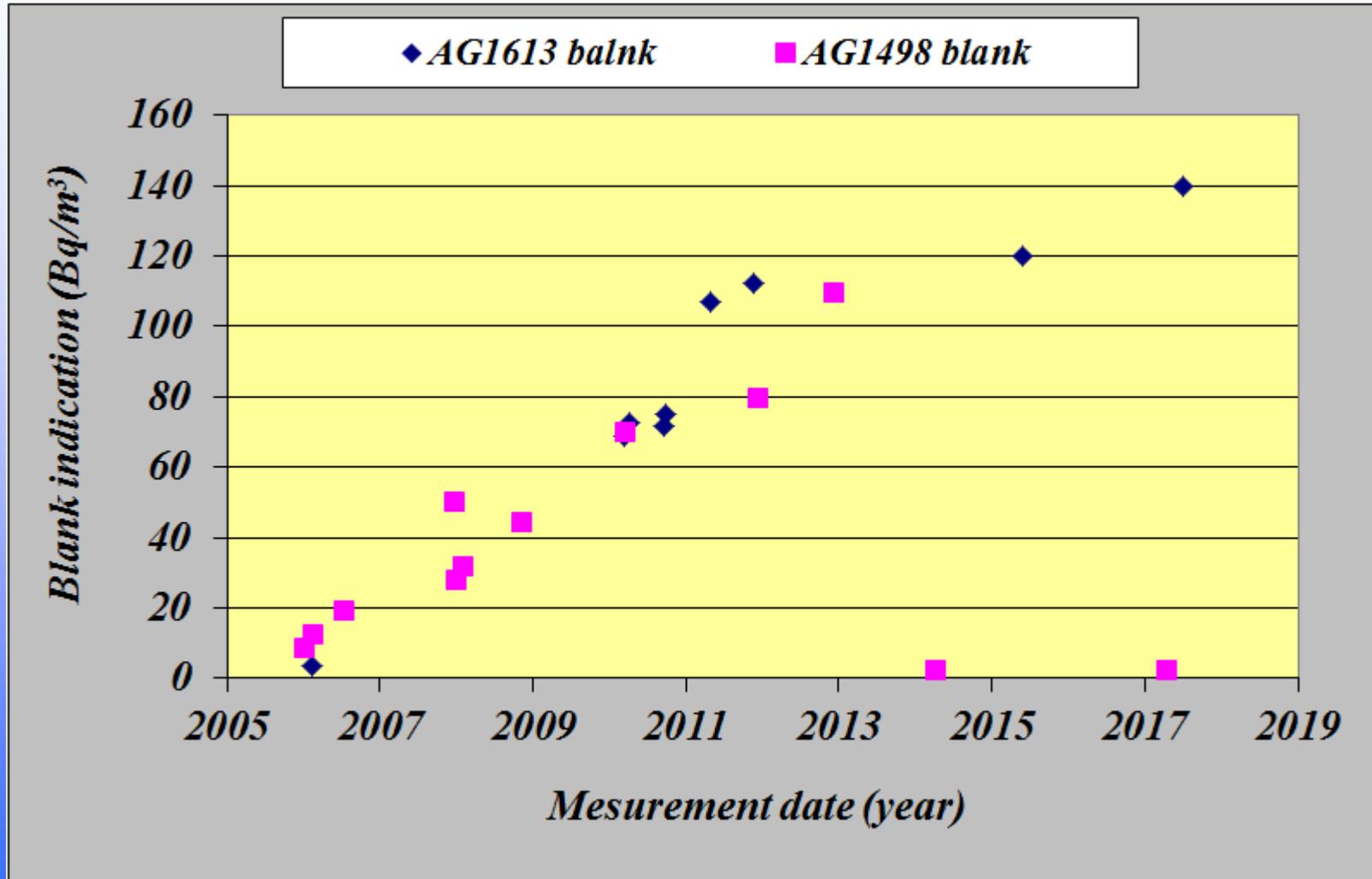
Correct determination of blank indication of radon monitors may have similar weight as determination of the instrument sensitivity.

For this reasons two new blank chambers (volume 78 and 141 L) were developed at ENEA-INMRI that allows blank determination with an uncertainty lower than a few  $\text{Bq}/\text{m}^3$ .

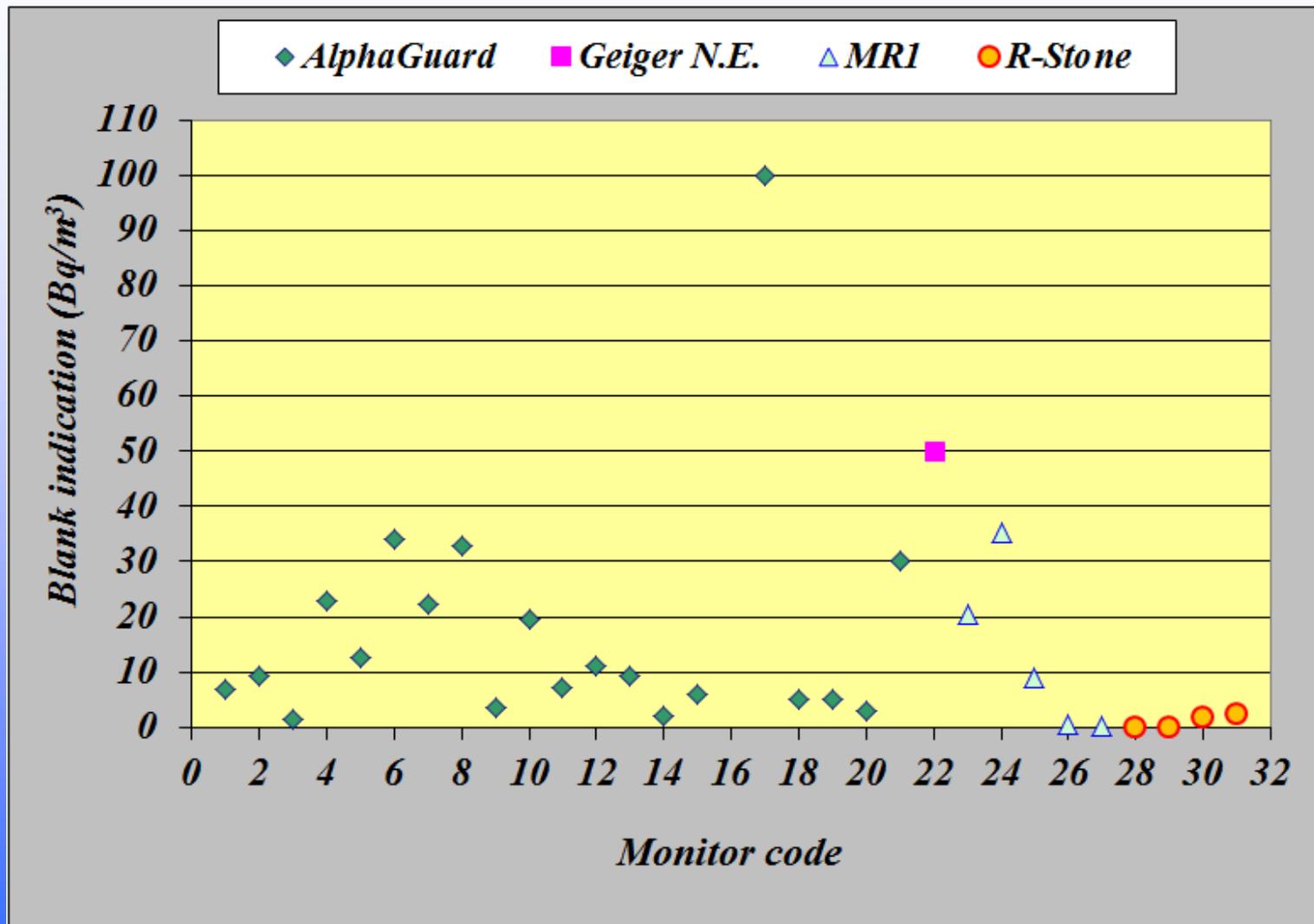


Blank indication of Rn monitors grow with time due to Pb210 deposition.

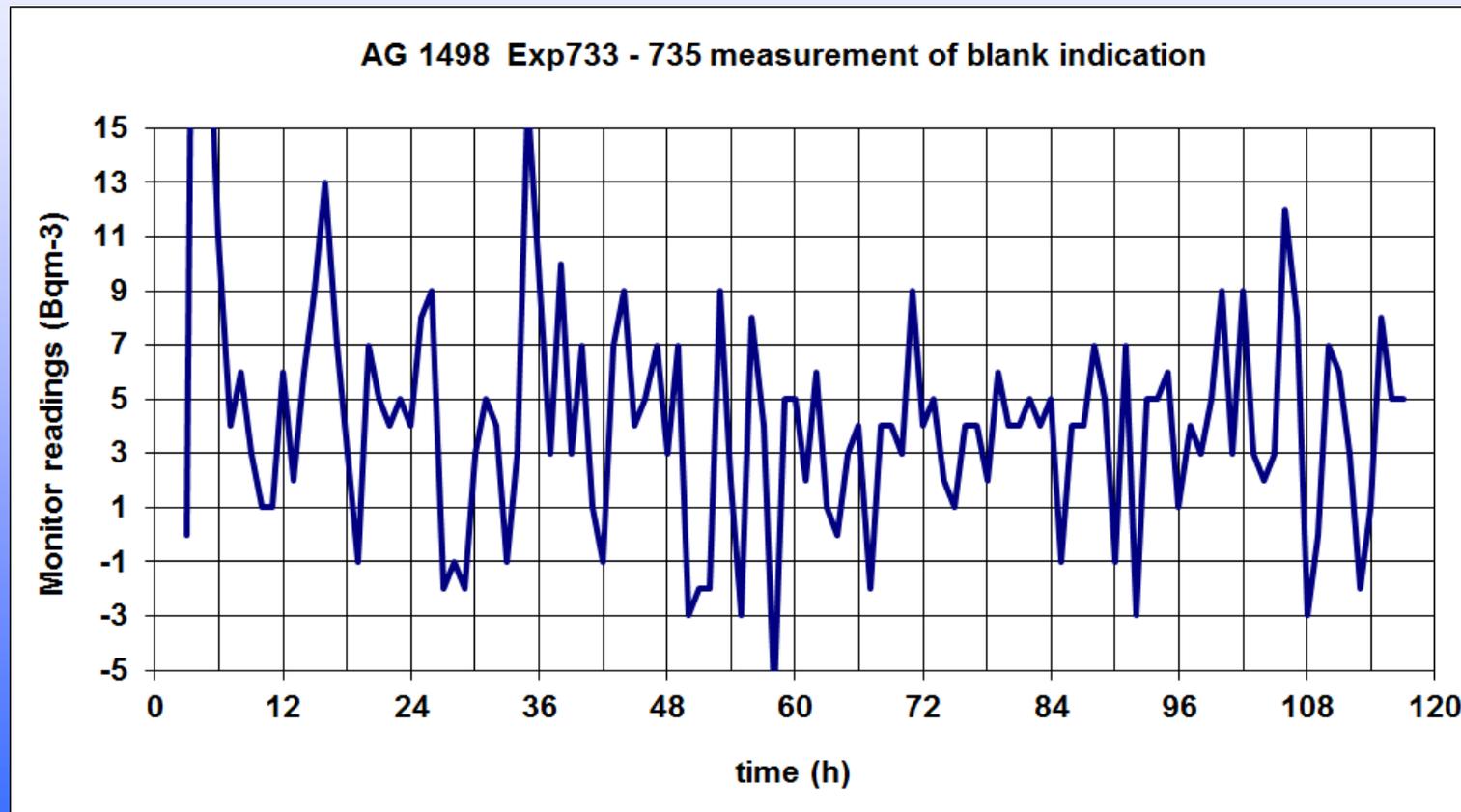
In the figure below variation of ENEA INMRI monitors is shown



## Blank indication of some radon monitors calibrated at ENEA INMRI laboratories.



In February 2017 ENEA INMRI and BFS Germany made a bilateral intercomparison for measurement of blank indication of an AlphaGuard monitor in the frame of Metro ERM European project. In the figure measurement at INMRI blank chamber. AlphaGuard readings:  $3,9 \pm 3,7$



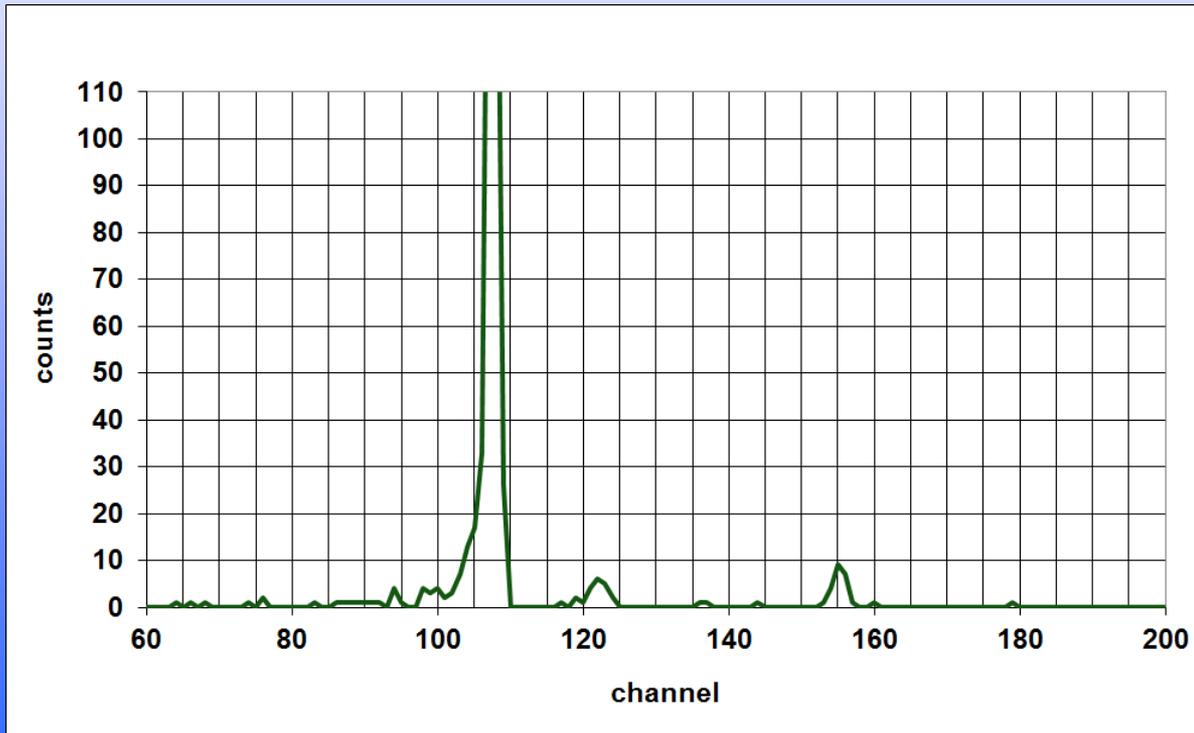
**Residual Rn activity in blank chamber was measured with DurrIDGE RAD7.**

**Rn activity in blank chamber was = 1.4 Bqm<sup>-3</sup> uncertainty 0.3 Bqm<sup>-3</sup>.**

**Intercomparison results:**

**Blank indication by INMRI 2.5 Bqm<sup>-3</sup> uncertainty 0.5 Bqm<sup>-3</sup>.**

**Blank indication by BFS 2.9 Bqm<sup>-3</sup> uncertainty 0.6 Bqm<sup>-3</sup>.**



**In the figure  $\alpha$  spectra of Rn progeny provided by RAD7.**

**Alpha peak from left:**

**1) Po 210 from Pb-210**

**2) Po 218**

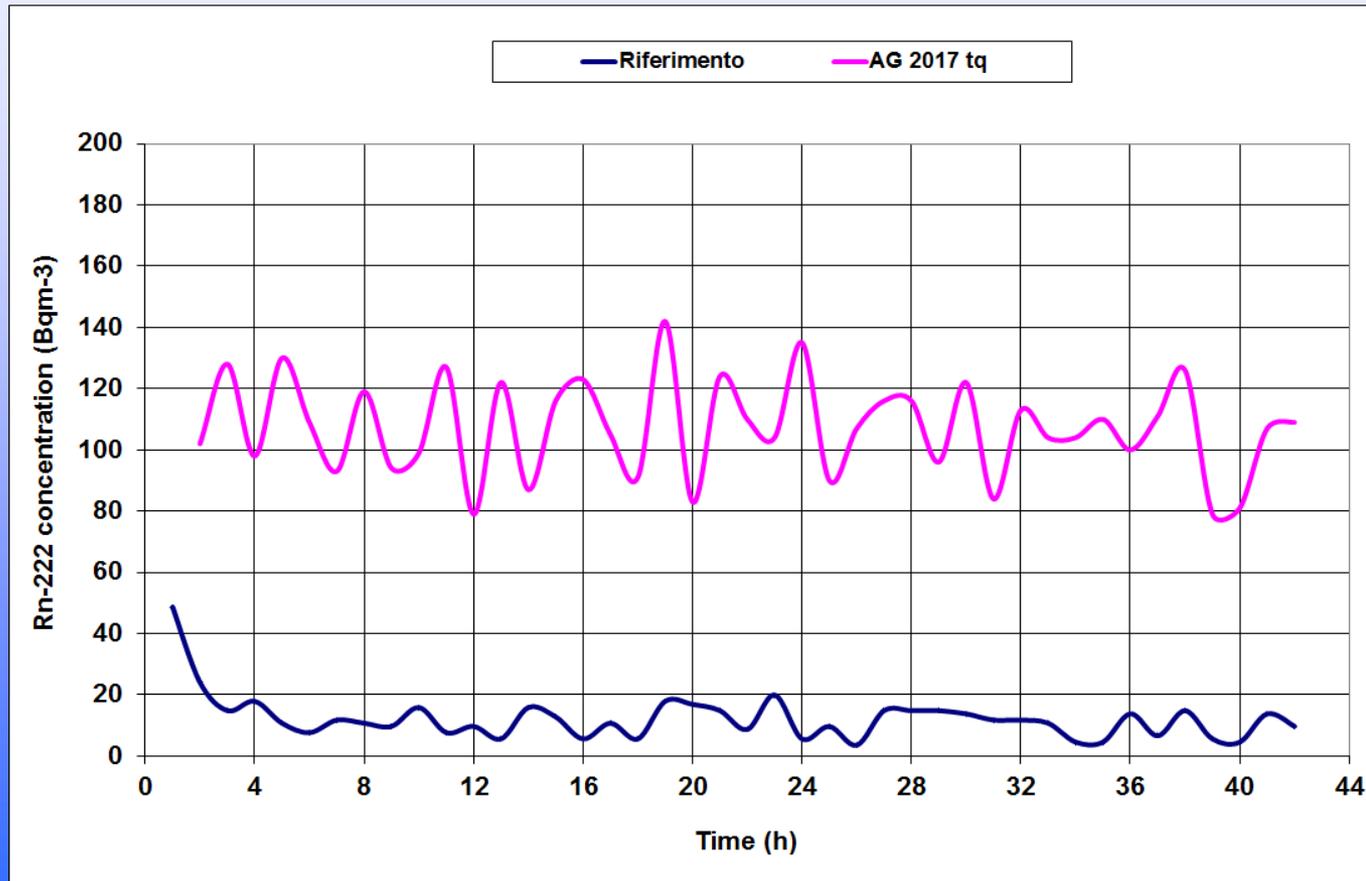
**3) Po 214**

**Blank indication of monitors may be measured at low radon concentration inside 1 m<sup>3</sup> radon chamber, reference concentration is provided by a suitable INMRI monitor with low blank. As an example see figure below.**

Reference activity:  $11 \pm 4$  Bqm<sup>-3</sup>

Readings of the minitor under calibration  $107 \pm 16$  Bqm<sup>-3</sup>

Blank indication of the monitor under calibration  $96$  Bqm<sup>-3</sup>



**Calibration at variable radon concentration of radon monitor  
ENEA INMRI “green house” inside walk in radon chamber.  
“green house” volume is 9 m<sup>3</sup>.**



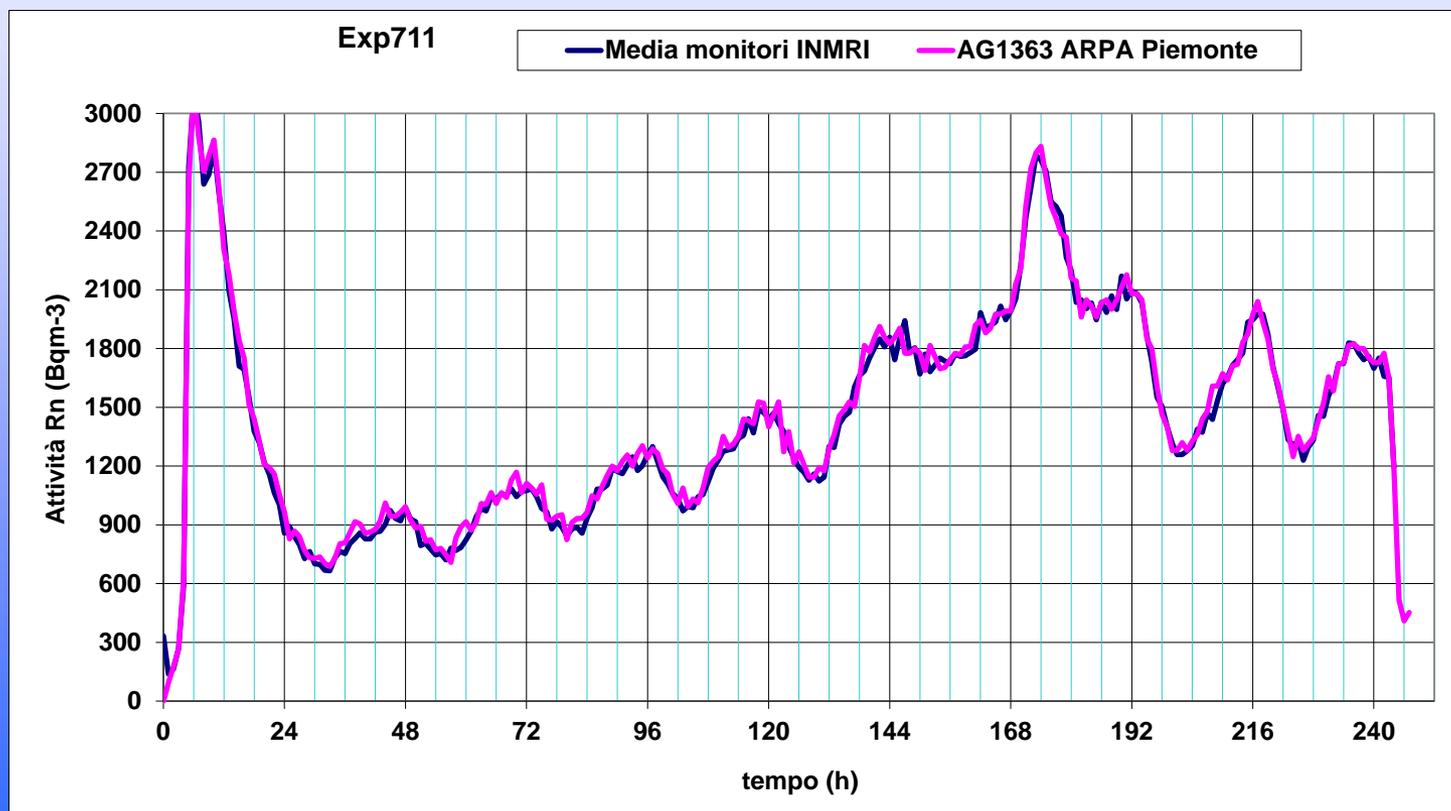
# Calibration of an AlphaGuard monitor at variable Rn activity concentration.

Measurement at variable radon concentration in walk in radon chamber are more realistic, but more difficult to be evaluated.

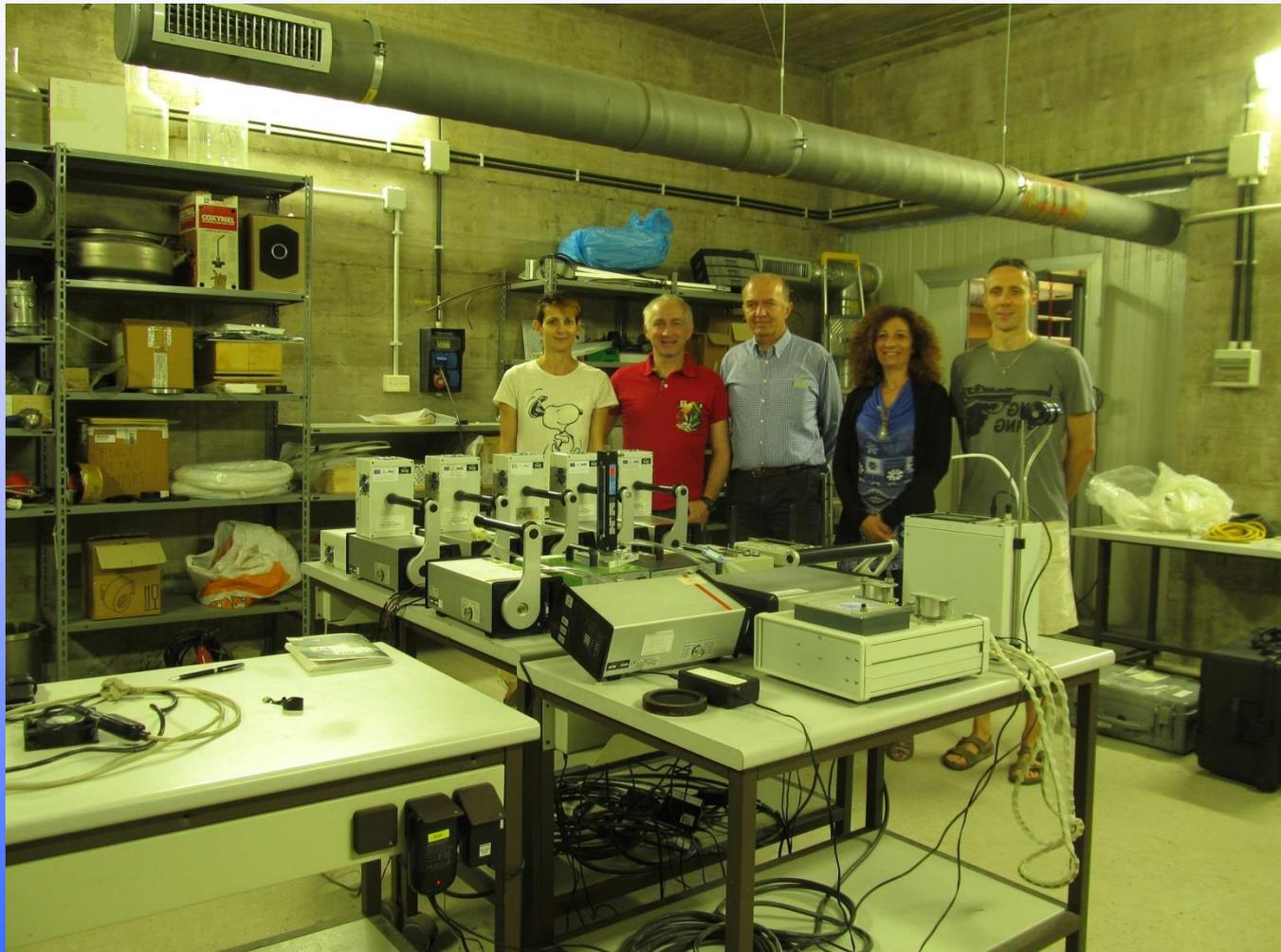
Some time the agreement between the different experiment is very good.

Sett. 2016 Constant Rn: FT= 1.008 – Variable Rn: FT= 1.01

June 2017 Constant Rn: FT=0.995

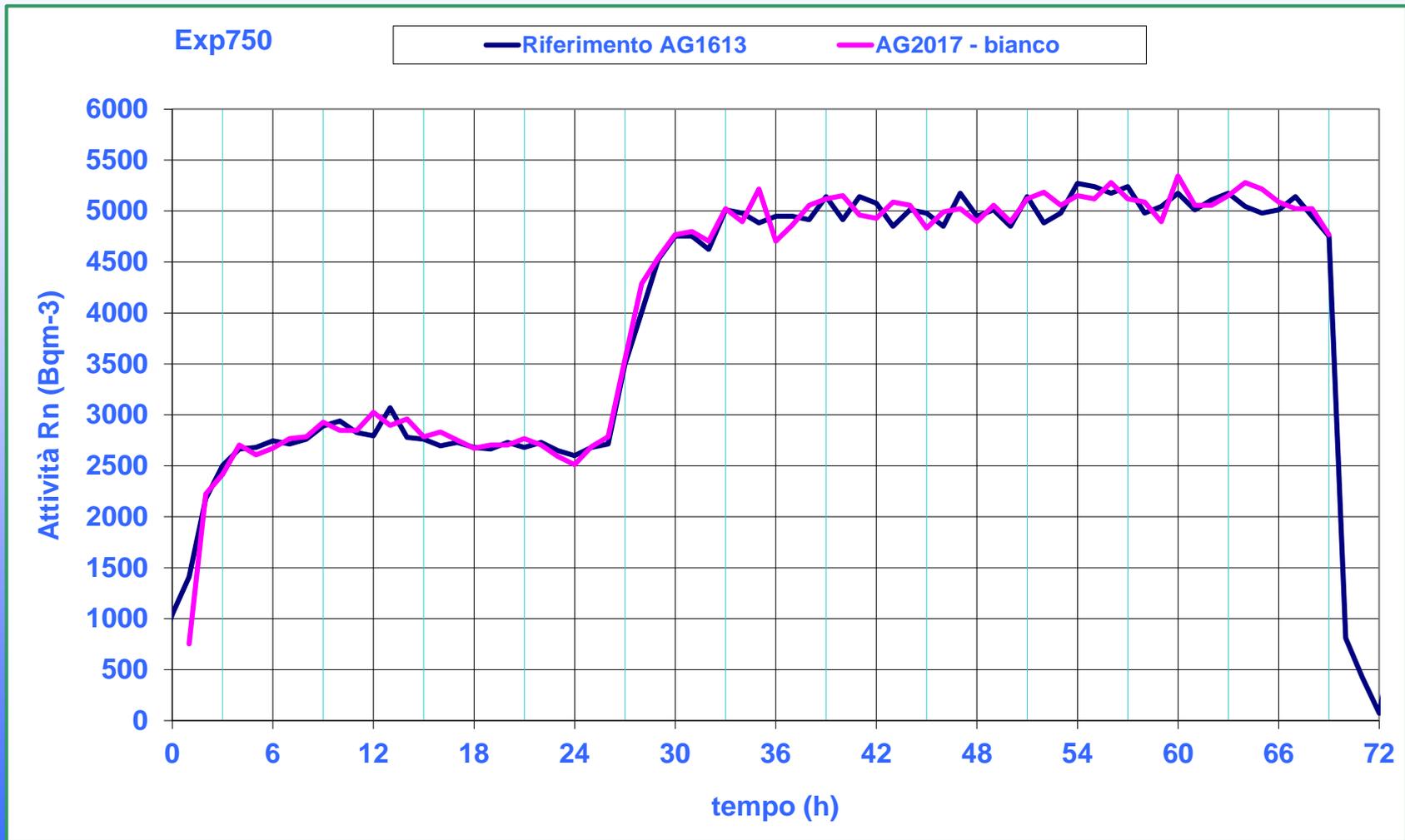


**ENEA INMRI walk in radon chamber, volume 150 m<sup>3</sup>.  
Calibration at variable radon concentration of the radon monitor  
used for the in field intercomparison**

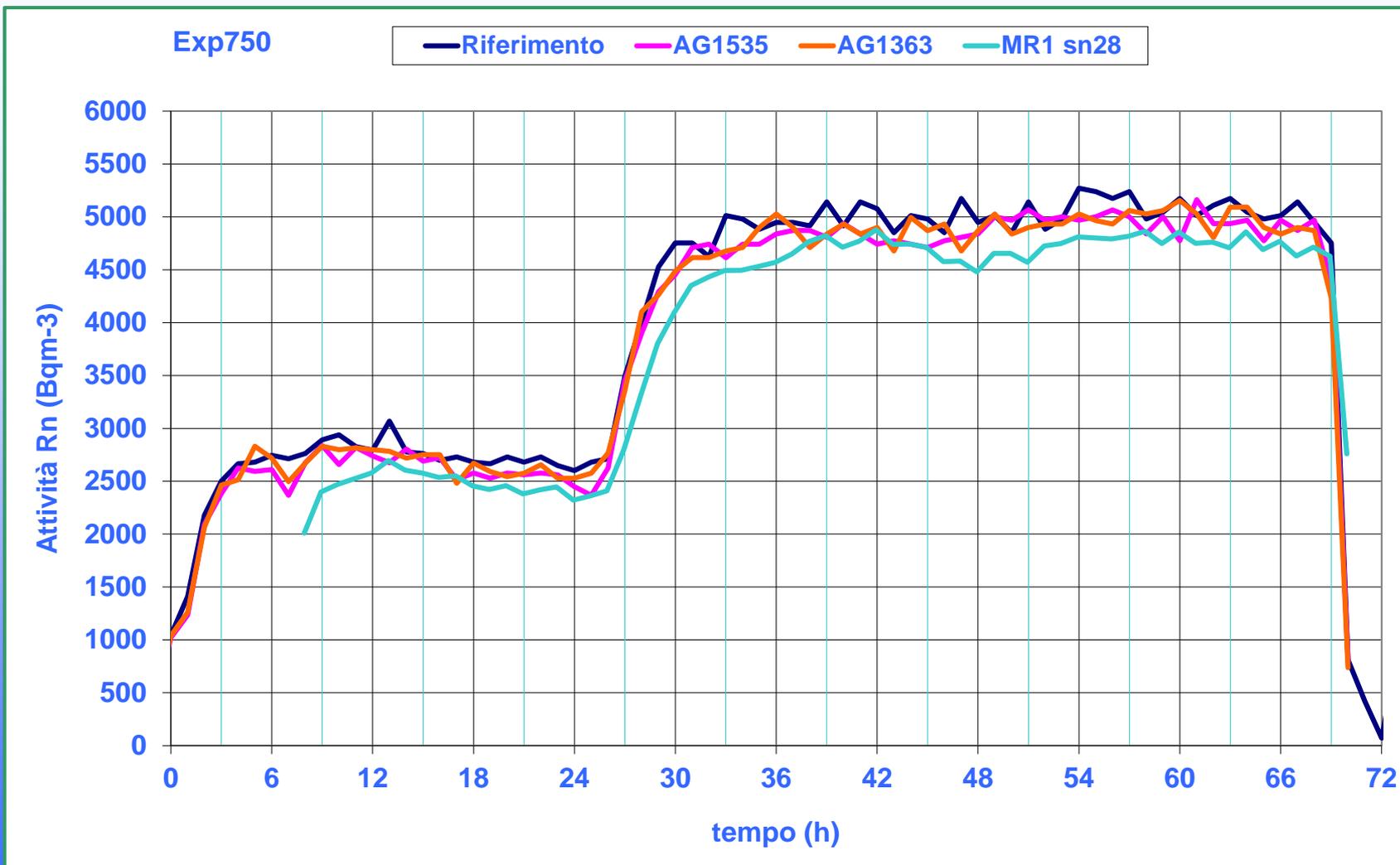


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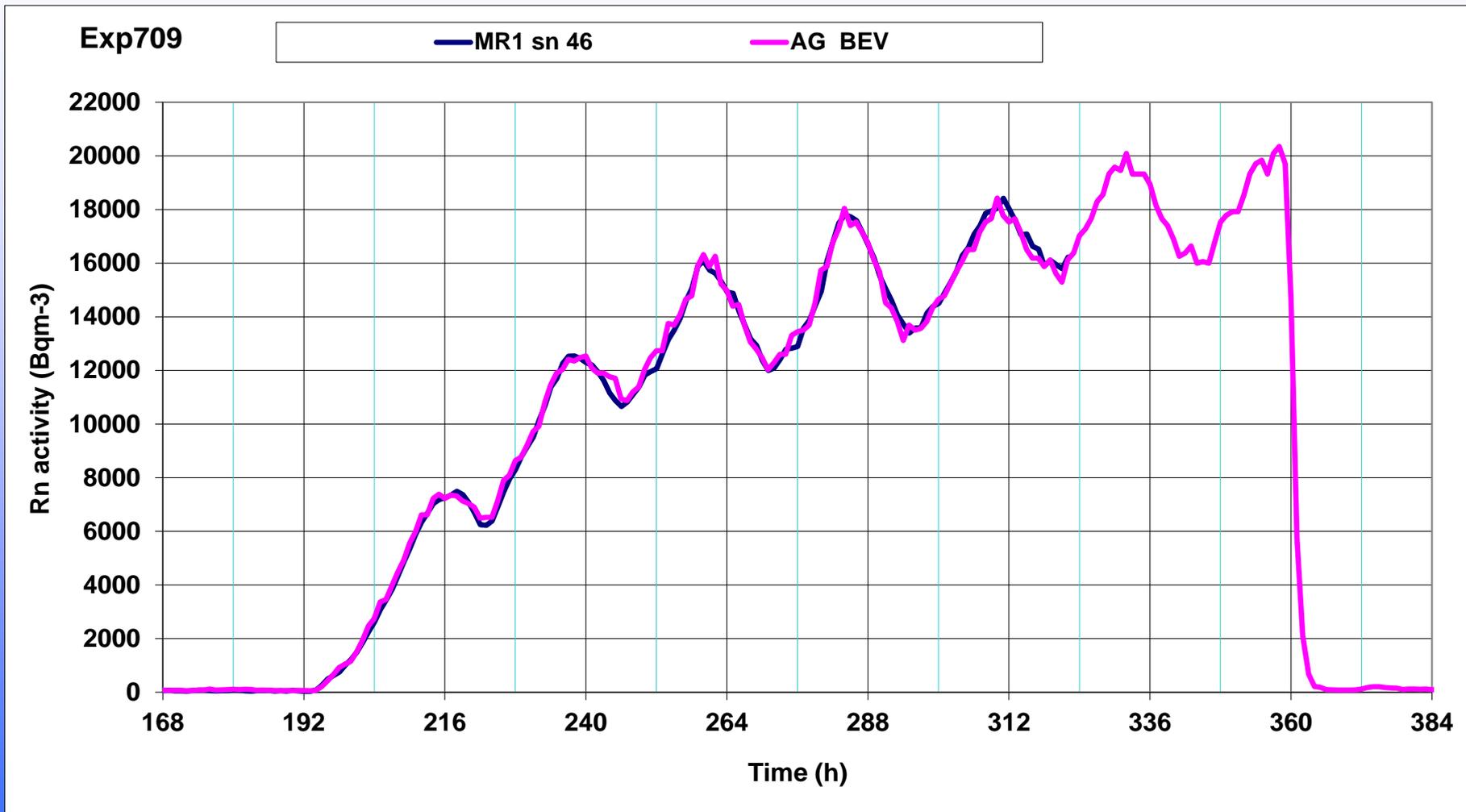
# Calibration at variable radon concentration in walk in radon. Some time the agreement between the instrument is excellent.



Some time the agreement between the instrument is “less” good.



# Comparison of MR1 scintillation cell and AlphaGuard ionization chamber during a calibration test in the walk in radon chamber.



## Calibration results of AlphaGuard and MR1 Tesys in different measurement condition:

at constant radon activity in 1 m<sup>3</sup> radon chamber or  
at variable radon activity in walk-in radon chamber.

AlphaGuard BEV	
Measurement condition	CF
Rn Constant	1,025
Rn Constant	1,025
Rn Variable	1,00
Rn Variable	1,024
Rn Variable	1,03
Rn Variable	1,006
<b>Average</b>	<b>1,018</b>
<b>Std. dev.</b>	<b>0,011</b>

AlphaGuard CISAM	
Measurement condition	CF
Rn Variable	1,020
Rn Variable	1,010
Rn Constant	1,006
Rn Variable	0,999
Rn Constant	1,012
Rn Variable	0,997
Rn Variable	0,995
<b>Average</b>	<b>1,006</b>
<b>Std. dev.</b>	<b>0,008</b>

MIAM MR1 sn 57	
Measurement condition	CF (Bqm-3/cpm)
Rn Variable	30,51
Rn Variable	30,28
Rn Constant	30,68
<b>Average</b>	<b>30,490</b>
<b>Std. dev.</b>	<b>0,164</b>
<b>Std. dev%</b>	<b>0.5%</b>

**Calibration facilities for radon measurement in ENEA INMRI are well assessed.**

**Main work in progress:**

- 1) Development of a radon standard not based on radium solution in bubblers.**
- 2) Standardisation of calibration procedures in walk-in radon chamber at variable radon concentration.**

**Thank you for your attention.**

