PRODUCTION OF MIXED RADIOACTIVE GAS ATMOSPHERE AND PROPOSITION OF SETUP TO TEST RN-220 AND RN-222 SEPARATION BY POLYMER FOILS

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CONCEPTION OF SETUP FOR THE PRODUCTION OF RADIOACTIVE GAS ATMOSPHERE

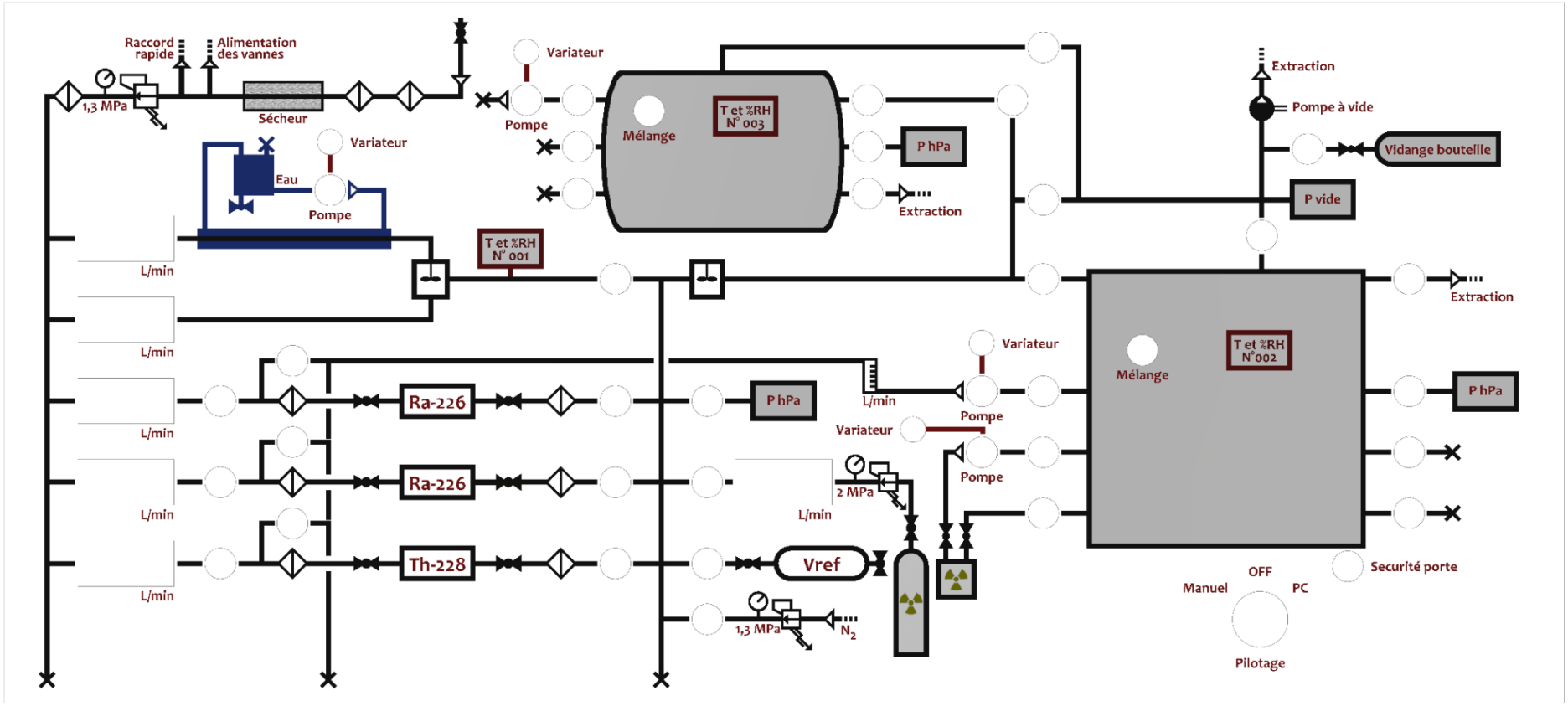
LNHB REQUIREMENTS FOR GAS ATMOSPHERES

- **LNE-LNHB is developing new measurements methods for radioactive gas at low level concentration mainly for CTBTO (Xe, Kr and Ar isotopes)**
- **As a results a new project was started in 2018 with the financial support of the LNE to create these new measurement methods and the gas atmosphere chamber for test purpose**
- **It was also interesting to extend the gas atmosphere chamber capacities :**
  - ✓ From 2012 to 2016 many development have been done on Rn-222 and Rn-220 at the LNE-LNHB
  - ✓ Other CEA labs have some interest with ^{3}H and ^{222}Rn
  - ✓ The LNHB decided to participate in the MetroRADON European project which required ^{222}Rn and ^{220}Rn atmosphere

➔ **RESULTS : we decided to build a new setup to create any atmosphere of radioactive gas, mixed or not, in order to: calibrate device, test new measurement systems, test new material for gas absorption, …**

A NEW SETUP HAS BEEN BUILT

- **Diagram of the setup**
  - ✓ Circle = Valves, Fan activation or pump activation with speed control
  - ✓ Square = Mass flow control and measurement



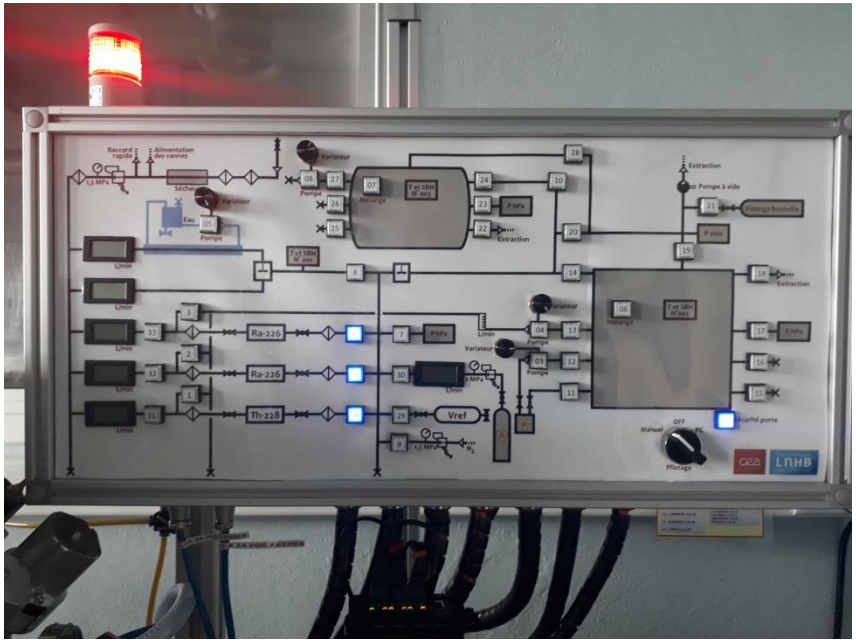
A NEW SETUP HAS BEEN BUILT

- **Picture**

  - ✓ NOTE : on this picture the Ra-226 and Th-228 are not installed.

DEVICE SPECIFICATIONS

- **Two chambers**
  - ✓ One that can be opened with a door and a volume of 125 L with high mixing using fans that can be moved inside the chamber to ensure good homogeneity
  - ✓ One that is smaller (42 L) that is kept for contaminating gas (as ^{3}H with some water in air), it has also 2 fans to ensure mixing
- **On both external device can be connected**
- **Pressure range is from vacuum up to 2 000 hPa, temperature is from 18 °C up to 50 °C (using stable heater), humidity from “ 0% RH ” up to “ 100% RH “**
- **Can operate manually using the command panel or with LabVIEW software**
- **Data acquisition by LabVIEW software for pressure, temperature, humidity and radon/thoron concentration**

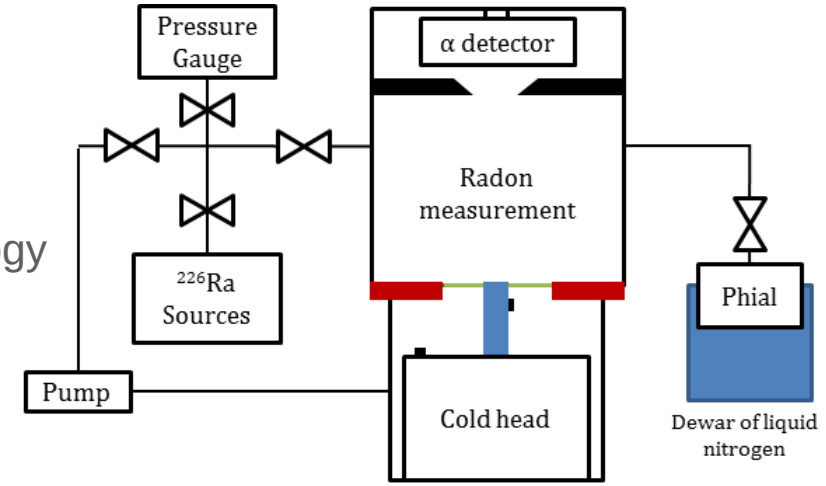


^{222}Rn REFERENCE ATMOSPHERE

^{222}Rn PRIMARY STANDARD

- ✓ Method created at the LNE-LNHB in 1995 with upgrade in 2012
- ✓ System under vacuum
- ✓ ^{222}Rn produced by a ^{226}Ra source and transferred on the cold finger at 80 K
- ✓ Measurement of the 6 mm diameter source by defined solid angle method with silicon detector (PIPS)

- ✓ Radon primary standard transferred in a metal vial which can be connected on circulation devices
- ✓ Activity of the primary standard 100 Bq up to 4 MBq with a relative standard uncertainty of 0,3 %
- ✓ International comparison with other national metrology institutes (CCRI-II)
- ✓ Publications :

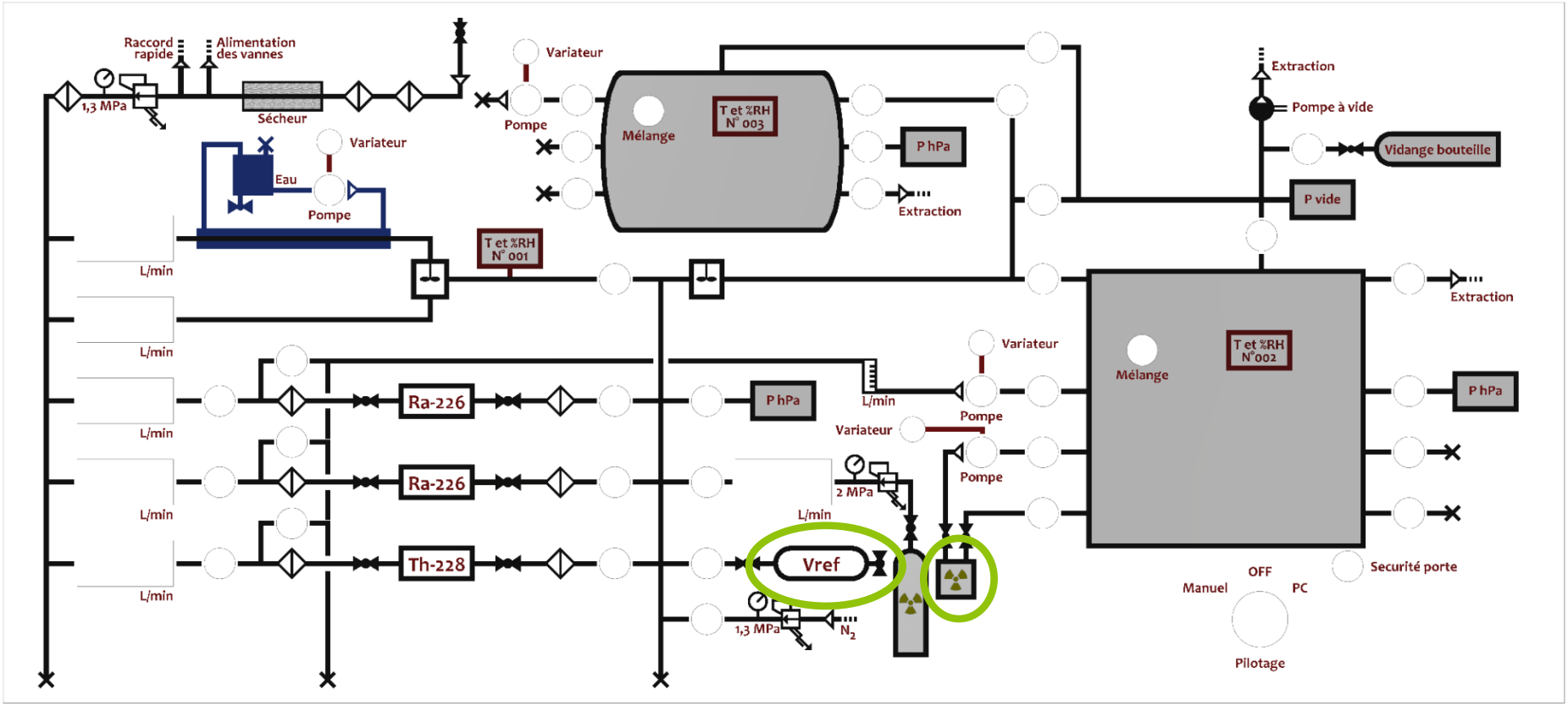


Picolo, J.L., Nuclear Instruments and Methods in Physics. Nucl.Instrum. Methods Phys. Res. A 369 (1996), 452–457

B. Sabot, et al., Applied Radiation and Isotopes 118 (2016) 167-174

USE OF THE ^{222}Rn PRIMARY STANDARD IN THE CHAMBER

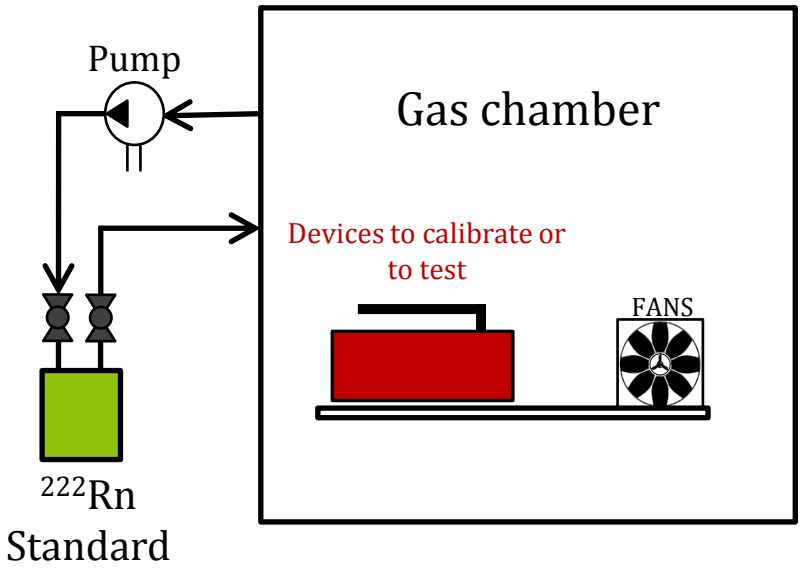
- With a tight chamber and a well know volume we can create well know ^{222}Rn atmosphere



- Measurement of the free volume using high precision pressure and temperature measurement and a certified volume given by the LNE (in this case 2 223,6 (10) cm^3)

EXEMPLE WITH THIS CHAMBER

- A device was placed in the chamber for its efficiency calibration with ^{222}Rn
- A ^{222}Rn standard vial was connected to the chamber



- Volume is measured and in this example we obtained : 1,2517 (27) 10^5 cm^3
- The ^{222}Rn standard is created and mixed with the air in the chamber

  ✓ For low activity it is better to used air from and old pressurised bottle as we have about 10 Bq/m^3 of radon in the pressurised air circuit from CEA

EXAMPLE AND RESULTS

- **In this example, using directly the primary standard we can create radon concentration between 800 Bq.m^{-3} and 32 MBq.m^{-3} with a relative standard uncertainty of 0,4 %**
- **With this closed and tight volume the ^{222}Rn concentration can be calculate at any time using half-life decay calculation which give similar uncertainty result as the ^{222}Rn half-life is well known : 3.8232 (8) days**
- **It is also possible to know the integrated activity concentration (I_A) for a time exposure *t* which can be expressed :**

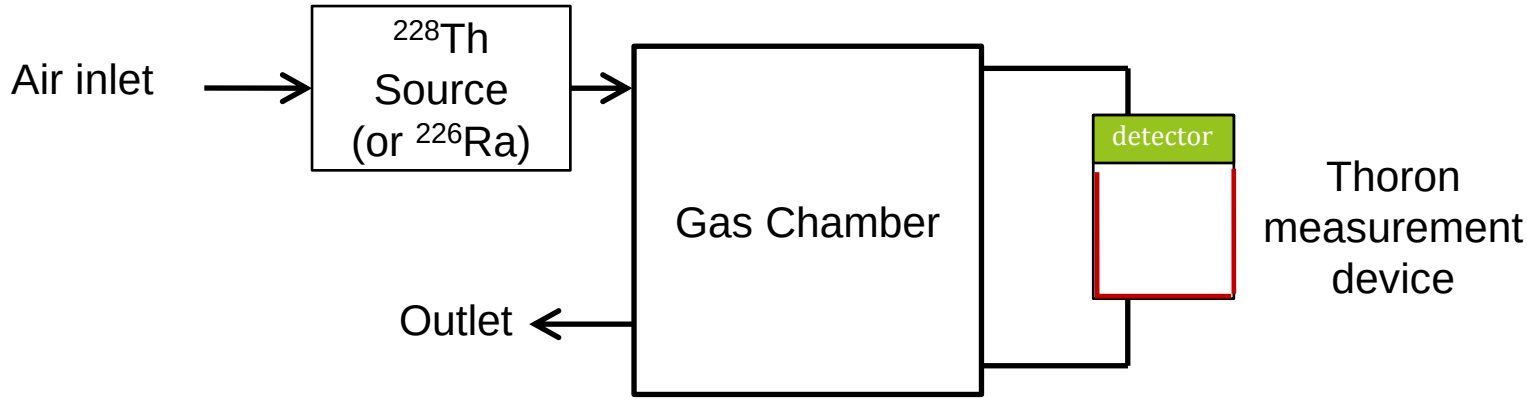
$$I_A = A\frac{1 - e^{-\lambda t}}{V\,\lambda}$$

- ✓ Where *A* is the given activity of the standard, *V* the volume of the chamber and λ the ^{222}Rn decay constant
- ✓ Related publications : J.L. Picolo et al., Applied Radiation and Isotopes 52, (2000) 427-434

^{220}Rn ATMOSPHERE OR ^{222}Rn WITH A STABLE ACTIVITY CONCENTRATION

ATMOSPHERE CREATION

- **This time the atmosphere is created using a solid source of Th-228 or Ra-226**

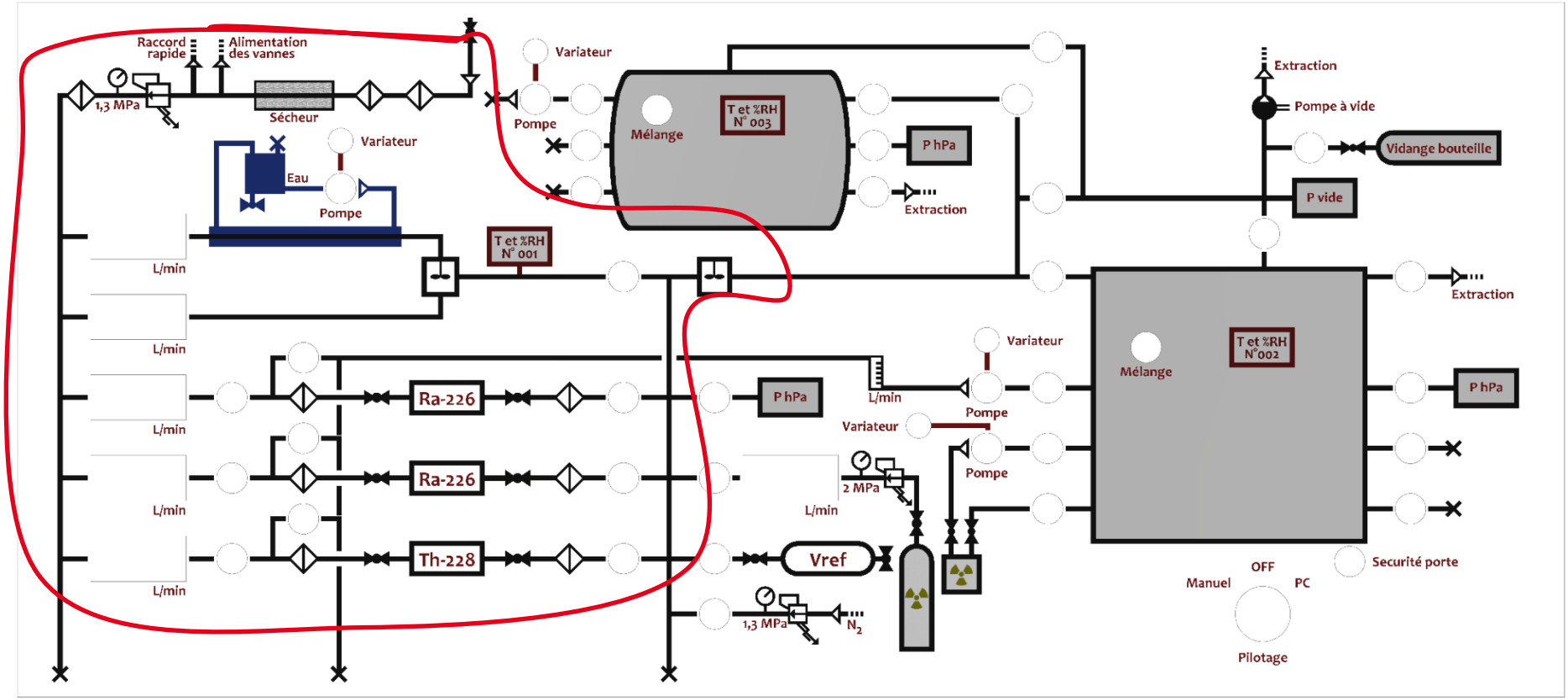


- ✓ With this chamber it's possible to do only one gas or both at the same time.
- ✓ Mixing and concentration is made using a constant flow rate circulation in the solid source with mass flow meter and compressed air circuit

- **The activity concentration is measured by a specific device developed at the LNE-LNHB that can detect both radon isotopes**

GAZ MIXING PART

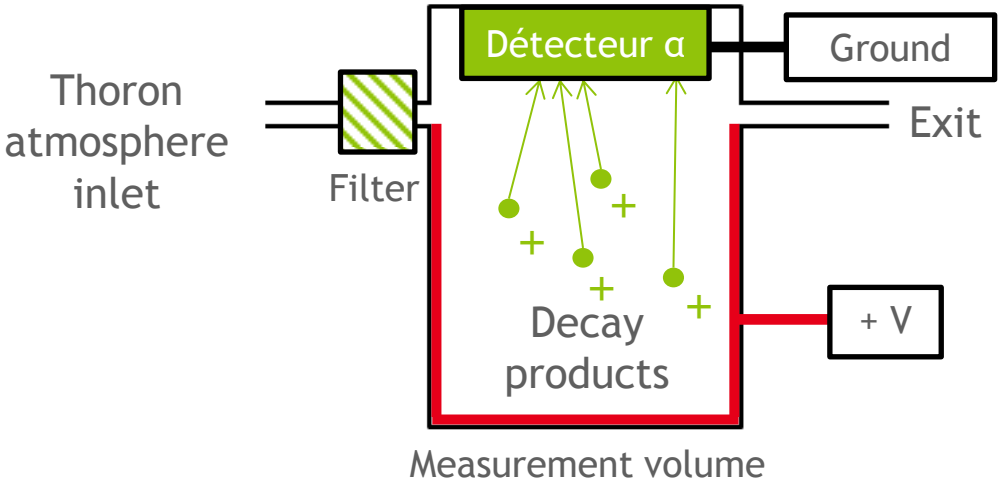
- It is possible to mix and make dilution by changing the flow rate of each line.
- Each flowmeter can be set between 0 and 20 L/min and with a relative standard deviation bellow 0,1%



MEASUREMENT OF Rn-220 AND Rn-222

- **The measurement of activity concentration of both gas can be done using the Rn-220 primary standard system that was developed in 2016.**

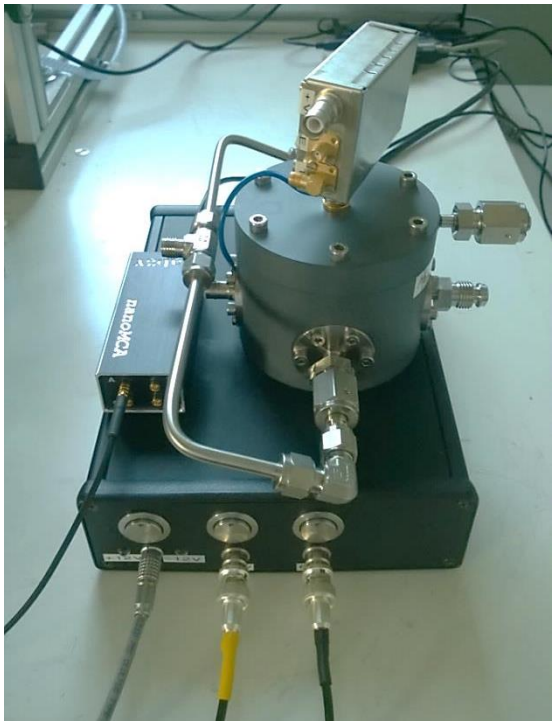
  - ✓ Special volume with alpha detector and electric field to catch decay product on the detector surface
  - ✓ The shape and size was optimized for best efficiency for gas detection and very high efficiency to catch the decay products on the detector surface for a flow rate around 1 L/min (optimized by multiphysics and monte-carlo simulations)



  - ✓ Works for both gas and give results with a relative standard uncertainty on the activity concentration of 1 %

DEVICE WITH ITS ELECTRONICS

- **The device is portable and can be plug on any gas chamber**
- **It has its own software to gather data and give direct results of activity concentration (LabVIEW)**
- **This device is currently used to qualify the emanation rate of the Ra-226 and Th-228 sources currently developed by the LNHB in the framework of MetroRadon**
- **The LNHB has 2 devices like that, a third one that has a smaller detector can be used to find Rn-222 and Rn-220 proportions in a mixed atmosphere**
- **Publications :**



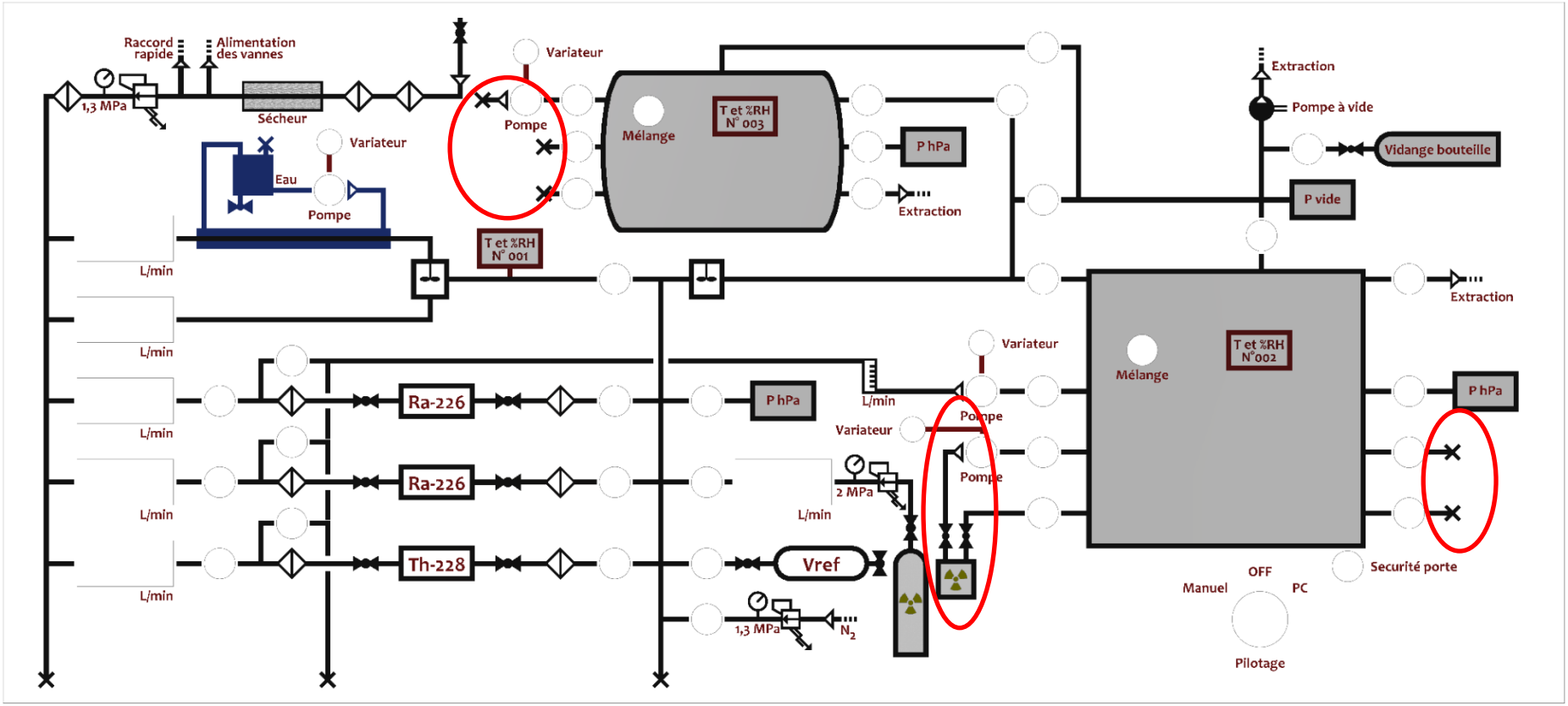
✓ B. Sabot, et al., Radiation Protection Dosimetry (2015), doi:10.1093/rpd/ncv221

✓ Publications : Benoît Sabot, thesis: Etalonnage des instruments de mesure de l'activité volumique du thoron (^{220}Rn) dans l'air https://www.theses.fr/2015SACLS122.pdf

✓ B. Sabot, et al., Applied Radiation and Isotopes (2015), http://dx.doi.org/10.1016/j.apradiso.2015.11.055

DEVICE ON THE CHAMBER

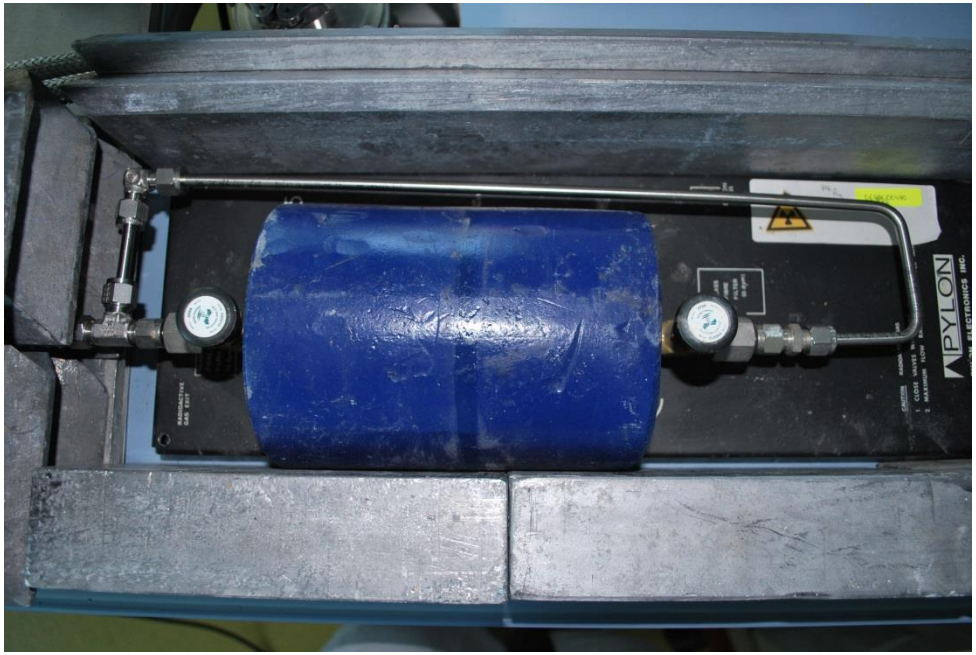
- The device with it's pump and flow meter is connected on the chamber by replacing the gas metal vial
- It is also possible to connect the device on other available inlet/outlet



THE Ra-226 AND Th-228 SOURCES

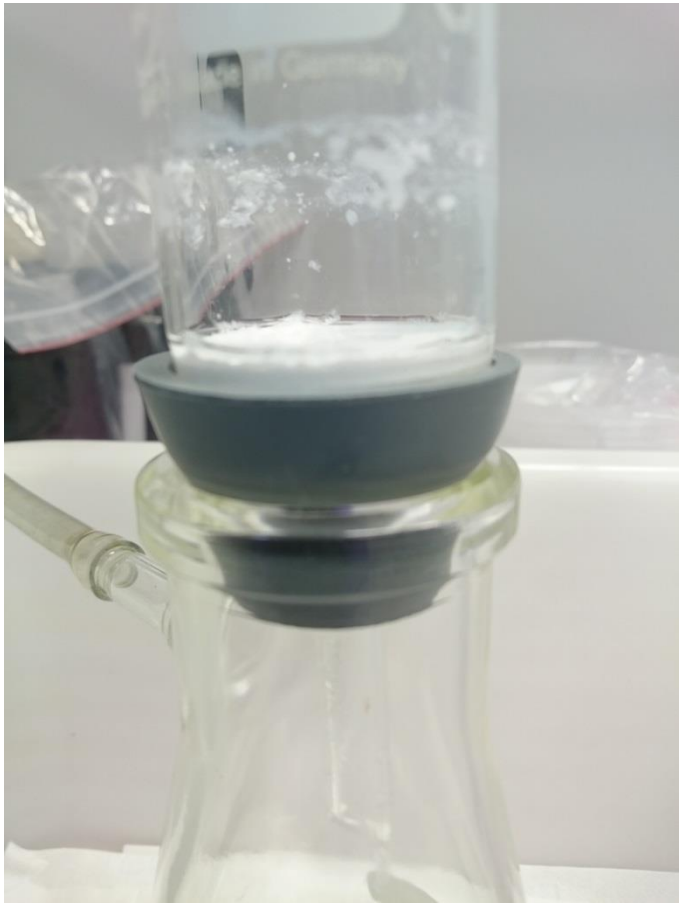
Ra-226 AND Th-228 SOURCES

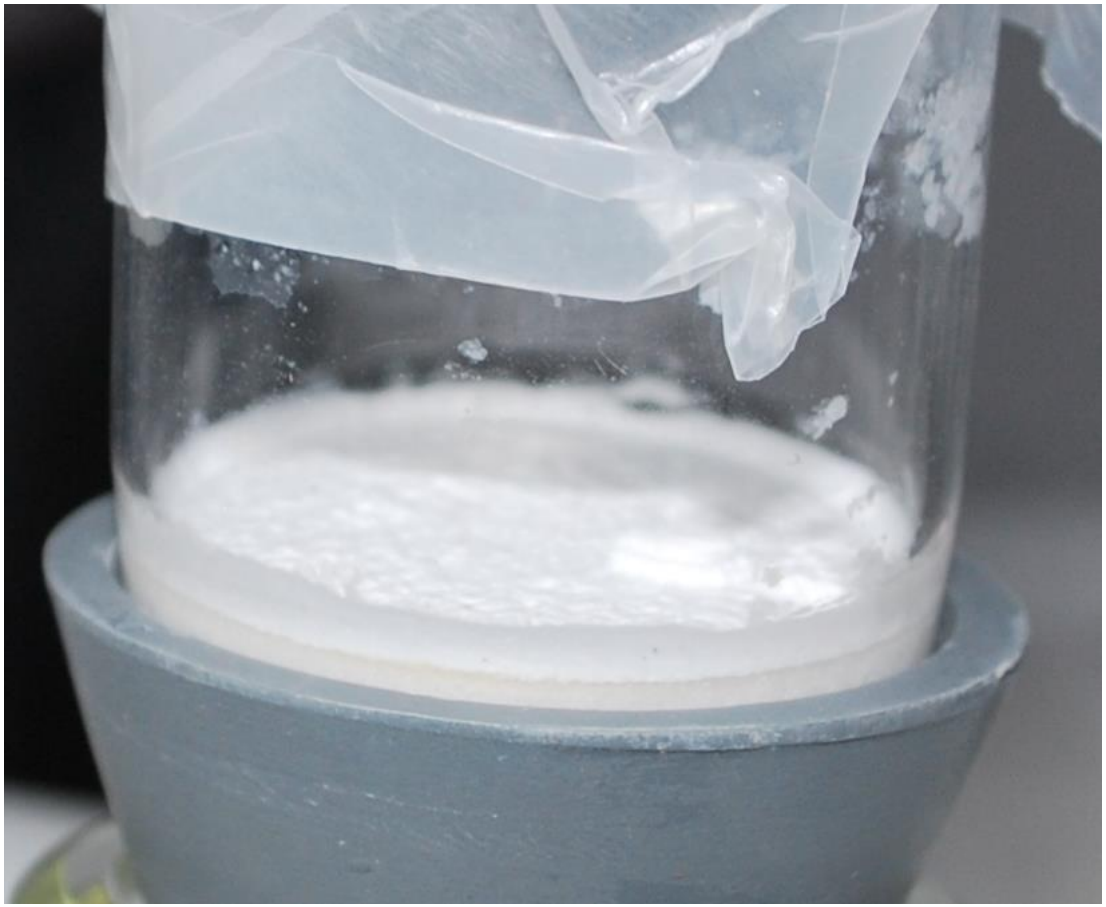
- **The biggest source is a commercial one from Pylon with a Ra-226 activity of 3,6 MBq.**
  - ✓ It is possible to evaluate approximately the radon production from the source using Pylon calculation factor
  - ✗ However these results are not certified by any standard : it is necessary to use the previous thoron/radon measurement system when we create the atmosphere



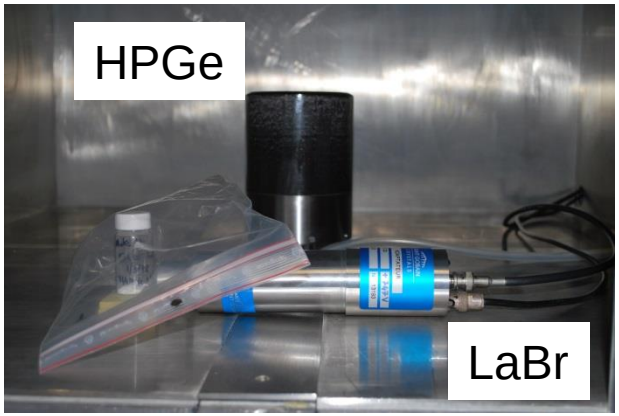
DEVELOPMENT OF $^{220}Rn/^{222}Rn$ EMANATION SOURCES

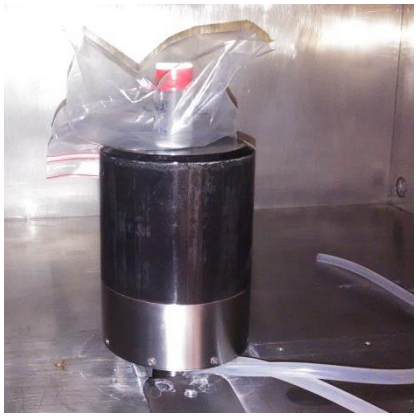
- Production of Ba-stearate to keep Th or Ra on a solid salt.





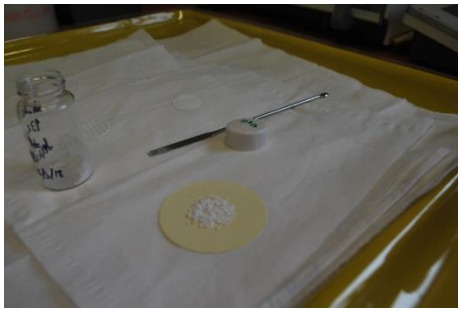
DEVELOPMENT OF $^{220}Rn/^{222}Rn$ EMANATION SOURCES

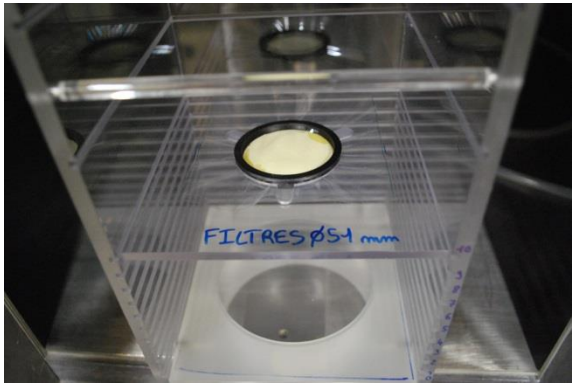




^{228}Th source (barium stearate) have been prepared at LNE-LNHB and measured with a HPGe (3 kBq)

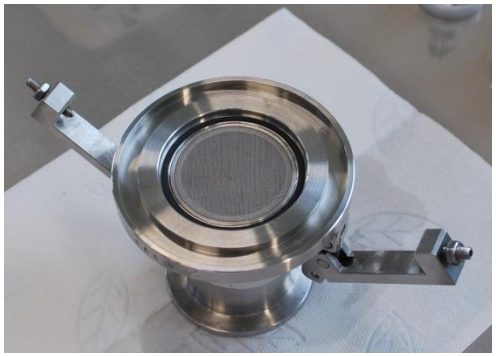
A sample of this 3 kBq powder was taken and deposited between 2 filters to prepare the emanation source of ^{220}Rn

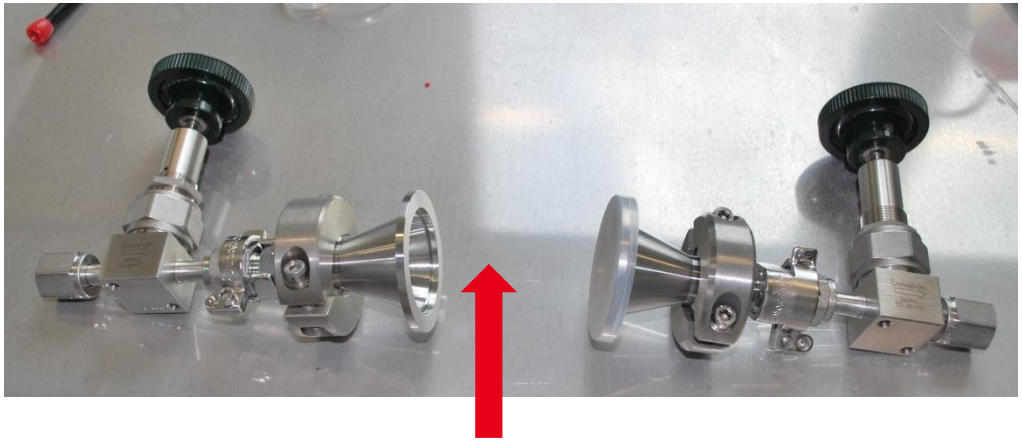




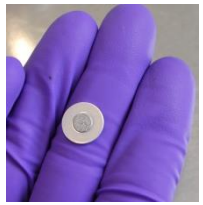
We measured 840 Bq of ^{228}Th (relative standard uncertainty: 2 %) with our HPGe

DEVELOPMENT OF ^{220}Rn EMANATION SOURCE PROTOTYPE





The emanation source is placed there

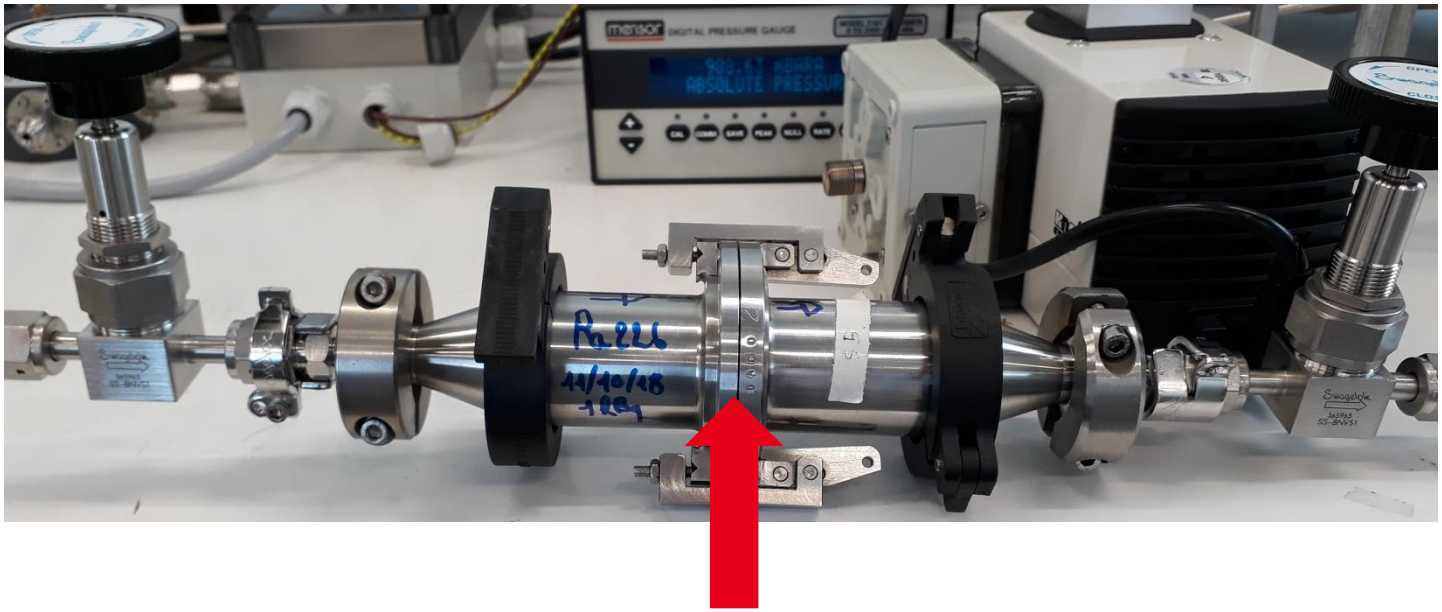


A security filter is added on both side.

DEVELOPMENT OF ^{222}Rn EMANATION SOURCE

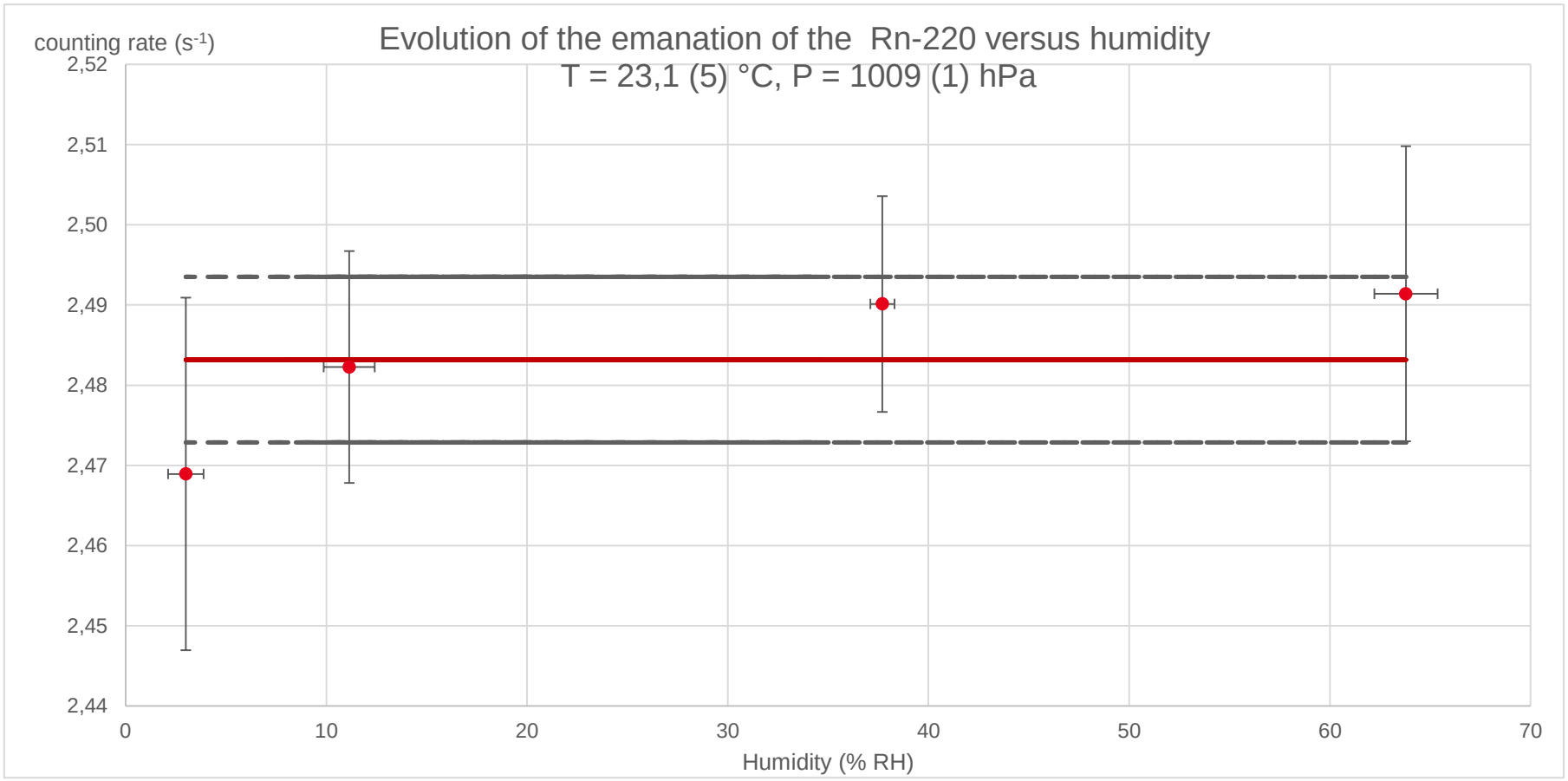
The same proceeding is followed to prepare a ^{226}Ra source.
1.059 kBq of ^{226}Ra (relative standard uncertainty of 1 %) is measured with our HPGe.

Each emanation source is places into our measurement loop:



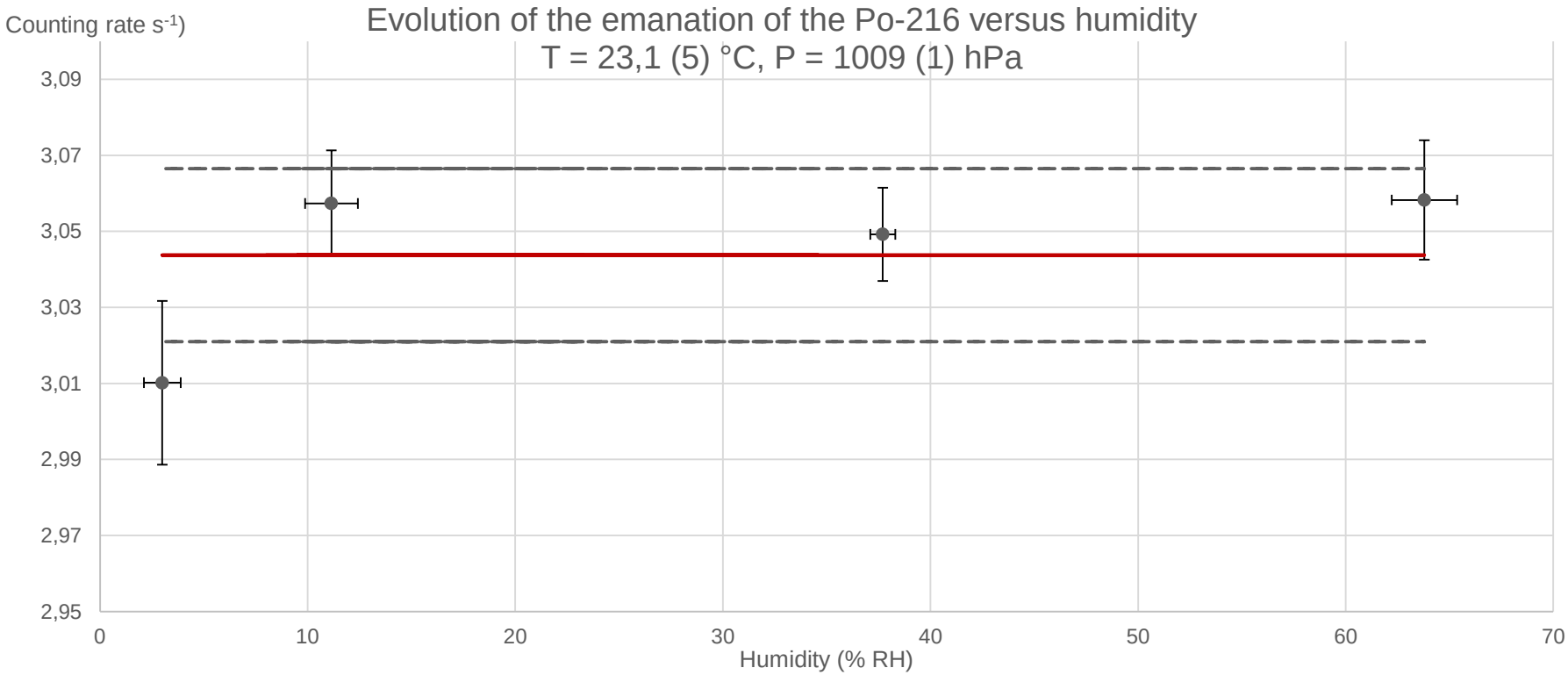
Filter with emanation source

INFLUENCE OF THE HUMIDITY FROM Rn-220 MEASUREMENT



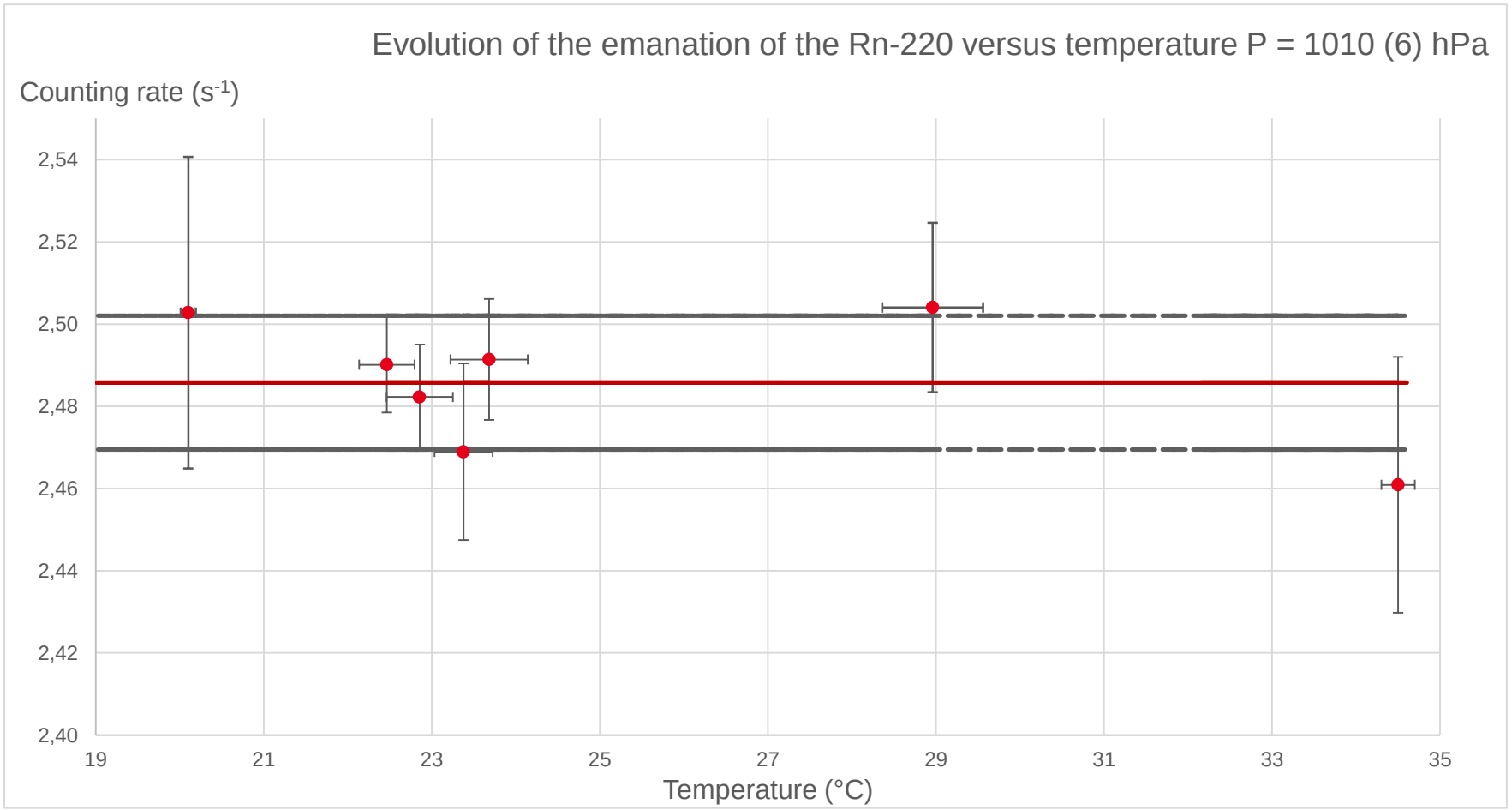
Average Counting rate (s^{-1})	2.48	Standard uncertainty (s^{-1})	0.01	**Relative standard uncertainty**	**0.4 %**

INFLUENCE OF THE HUMIDITY FROM Po-216 MEASUREMENT



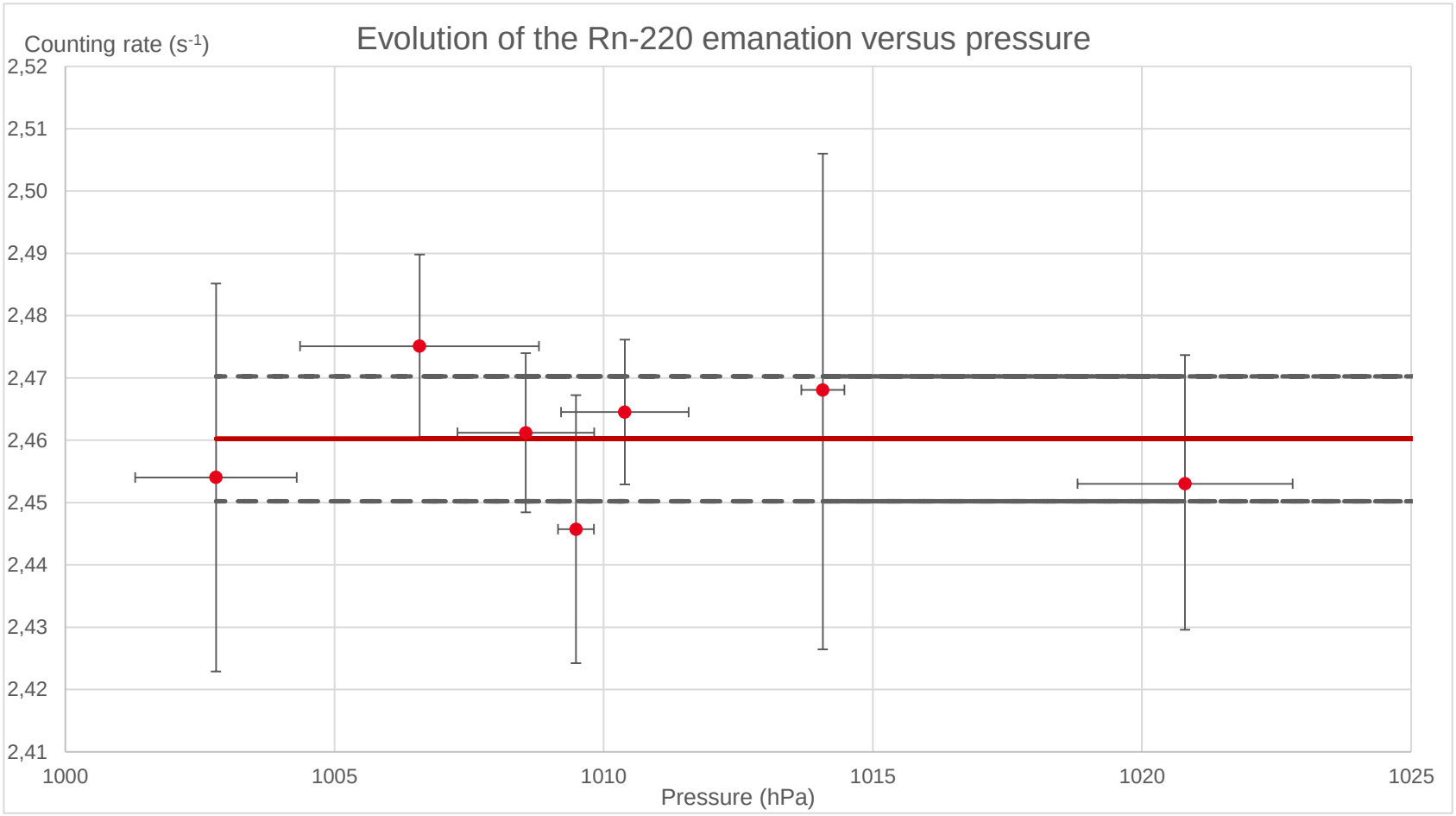
Average Counting rate (s^{-1})	3.04	Standard uncertainty (s^{-1})	0.02	**Relative standard uncertainty**	**0.7 %**

INFLUENCE OF THE TEMPERATURE



Average Counting rate (s^{-1})	2.49	Standard uncertainty (s^{-1})	0.02	Relative standard uncertainty	0.7 %

INFLUENCE OF THE PRESSURE



Average Counting rate (s^{-1})	2.46	Standard uncertainty (s^{-1})	0.01	**Relative standard uncertainty**	**0.4 %**

EMANATION RATE QUALIFICATION

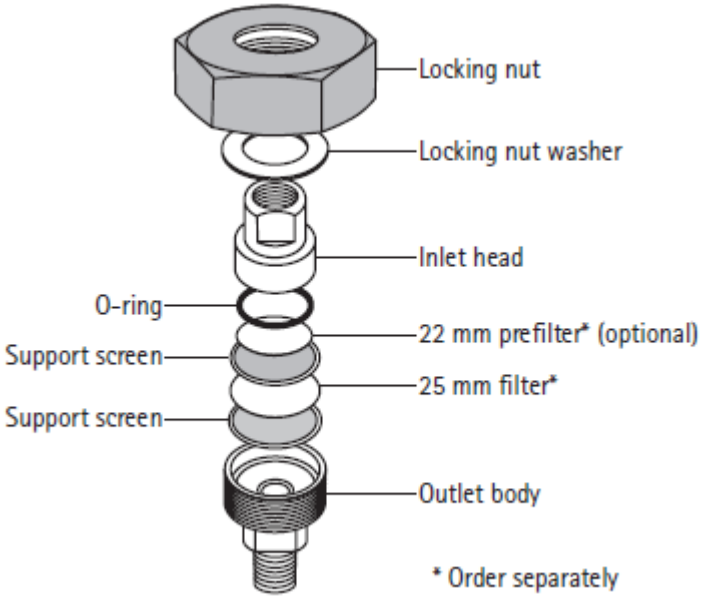
- **With combined measurement :**
  - ✓ Ra-226 or Th-228 activity with HPGe
  - ✓ Rn-222 or Rn-220 measurement with thoron measurement system and gas loop

➔ We obtain the emanation rate of the source.

- **Results for source with about 1 kBq of Ra-226 and Th-228 :**
  - ✘ 45,8 % with 3,5 % of relative standard deviation for Rn-222
  - ✓ 98,1 % with 2,5 % of relative standard deviation for Rn-220

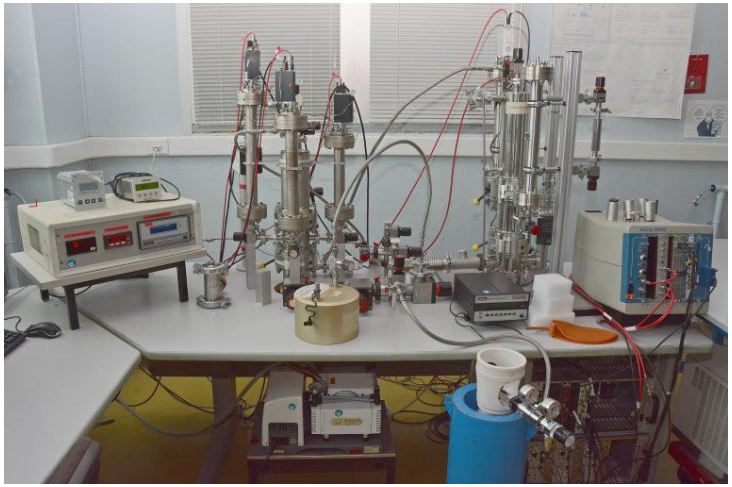
➔ Unknown reason for this difference

- **New sources are under preparation this time with flow rate tangential and perdicular to the source and with better filter to prevent charge lose in flow: test are done during March-April**
  - ✓ Activity arround 1 kBq for Ra-226 and Th-228 and also one source of Th-228 with 150 kBq.



OTHER RADIOACTIVE GAS

OTHER GAS

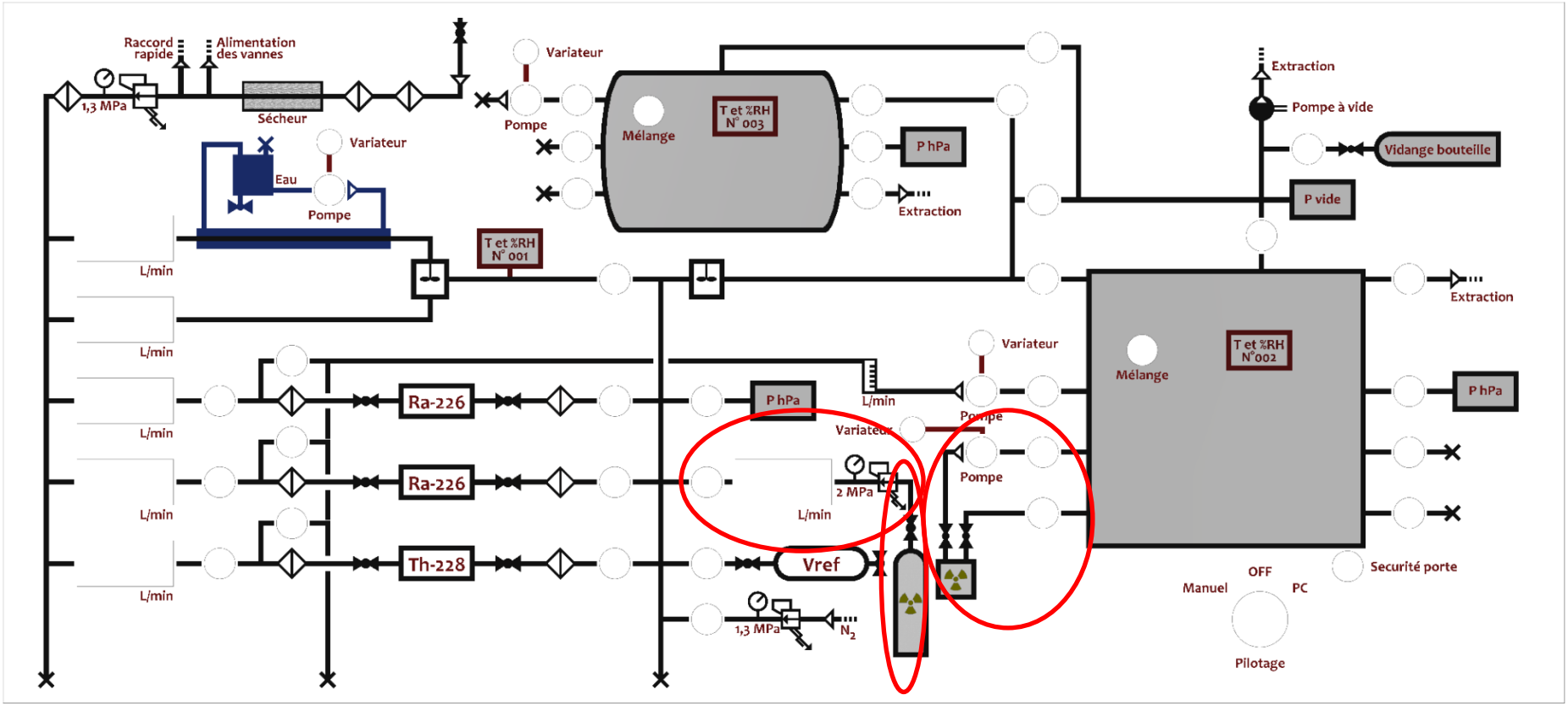


- **Main goal of the chamber was the other radioactive gas :**
  - ✓ Primary standard is the triple proportionnal counter methode
  - ✓ Activity concentration measure arround 200 Bq/cm^3 stored into a presurised bottle
  - ✓ Can used for Xe, Kr, Ar and H isotopes

- **Mixing on the gas chamber :**
  - ✓ One line with mass flow meter between 0 to 20 L/min that can be mixed with other line using the Th-228 and Ra-226 source or clean air to make a very high dilution
  - ✓ Other one is using the same vial than Rn-222 standard with a closed loop

OTHER GAS

- Possibilities for mixing :



CONCLUSION ON THE NEW SETUP

CONCLUSION ON THE SETUP

- **It took me time but finally I have a new tool to produce radioactive gas atmosphere with well know activity concentration**
- **Total control on the setup : I can add any new line easily, do any change and I know each pieces used to build the setup : nice filter older, liquid and tight small gas pump were hard to find…**

- **The interesting part, next steps :**
  - ✓ Many materials to test for radioactive gas absorption (3D printed material, porous scintillator and other…)
  - ✓ New devices to test using the porous scintillators in the framework of the LNE project for Xe, Kr, Ar and H
  - ✓ New devices to test with liquid scintillation system
  - ✓ Homogeneity studies for low half-life gas (Rn-220) : simulations + measurement (with aerogel and with multiple silicon detector and multi counter.

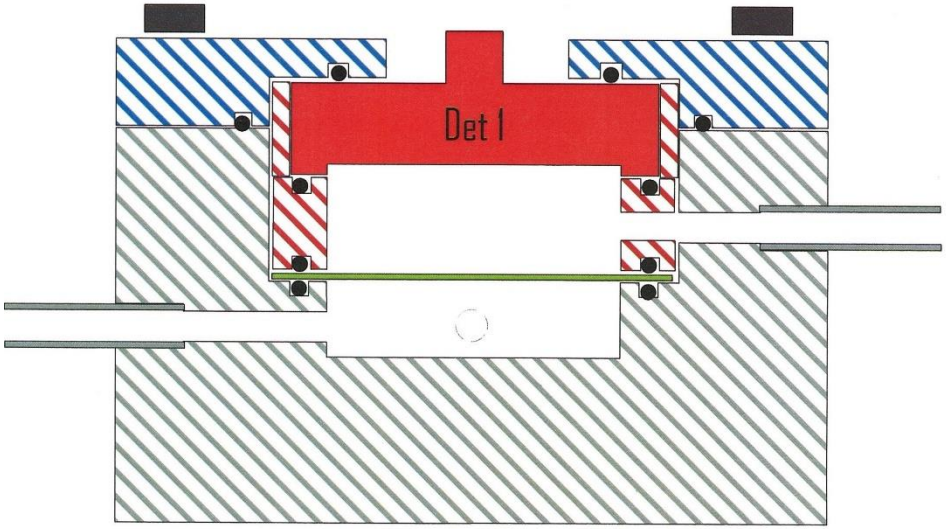
PLANED EXTENSIONS

- **Air/water line for radon in water production**
  - ✓ System already designed, one part is under construction, the last one I need to order it.
  - ✓ The purpose will be to produce a nice volume of standard “Radon in Water” : goal is to provide standard but also to give the possibility to other labs to participate in radon in water comparison
- **Increase the temperature range of the system**
  - ✓ Go bellow -20 °C will be nice and I have some idea how to do
- **Aerosol production for radon decay product attachment**
  - ✓ Quiet easy and fun to do with some ebay products ☺
  - ✓ Can be dry or wet aerosols

CONCERNING THE MEMBRANE FOR Rn-220 AND Rn-222 SEPARATION

IDEA FOR MEMBRANE TEST

- **Old prototype from my thesis with separation membrane for Rn-222 and Rn-220 :**
  - ✓ Never used but all pieces are here and can be used
  - ✓ 2 chambers can be separate by a membrane
  - ✓ It is possible to circulate gas in the chamber bellow and the chamber on the top have 1 inlet.
  - ✓ On the top chamber we can put a silicon detector and using metal grid we can apply electric field to perform alpha spectrometry on decay products and see if both gas are well separate



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