The University Of Sheffield.

DURRIDGE Radon Capture & Analytics

EPSRC Pioneering research and skills

# Molecular sieve-based gas recycling system with radon reduction for rare-event gaseous detectors

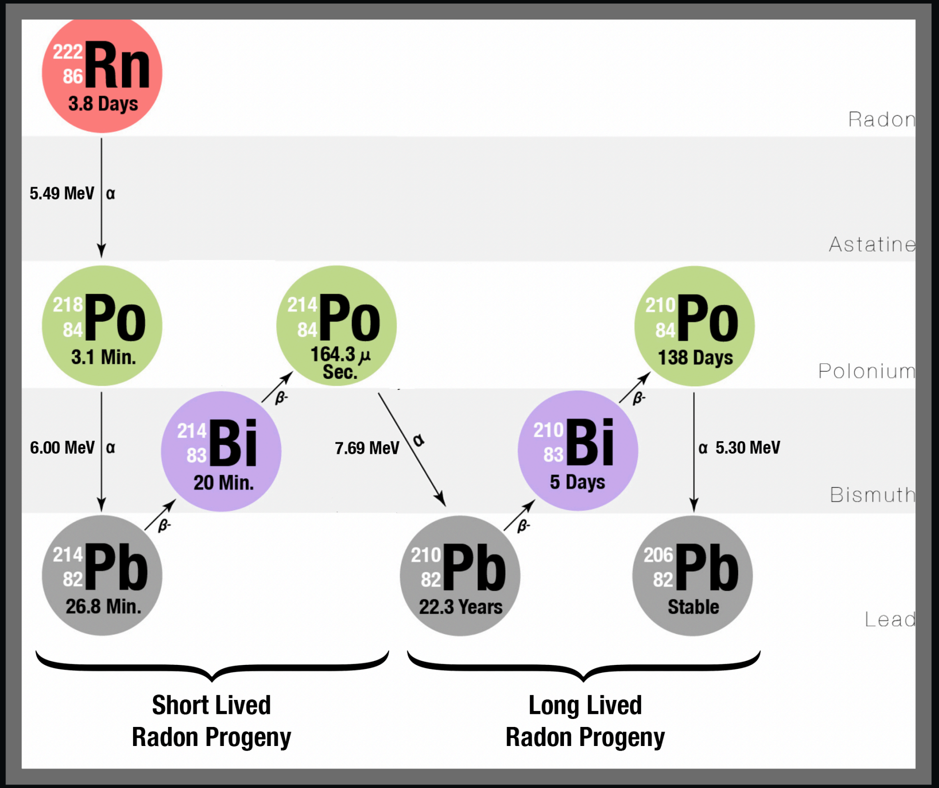

**Robert Renz Marcelo Gregorio**
University of Sheffield
robert.gregorio@sheffield.ac.uk
@askradonrob


CYGNUS online meeting
10-11 Jan 2022

# Introduction



- In ultra-sensitive gas-based experiments, it is crucial to use pure target gases
- Contaminants such as **radon** can **produce unwanted backgrounds**
- Other contaminants like **water, nitrogen** and **oxygen** can **suppress signals**



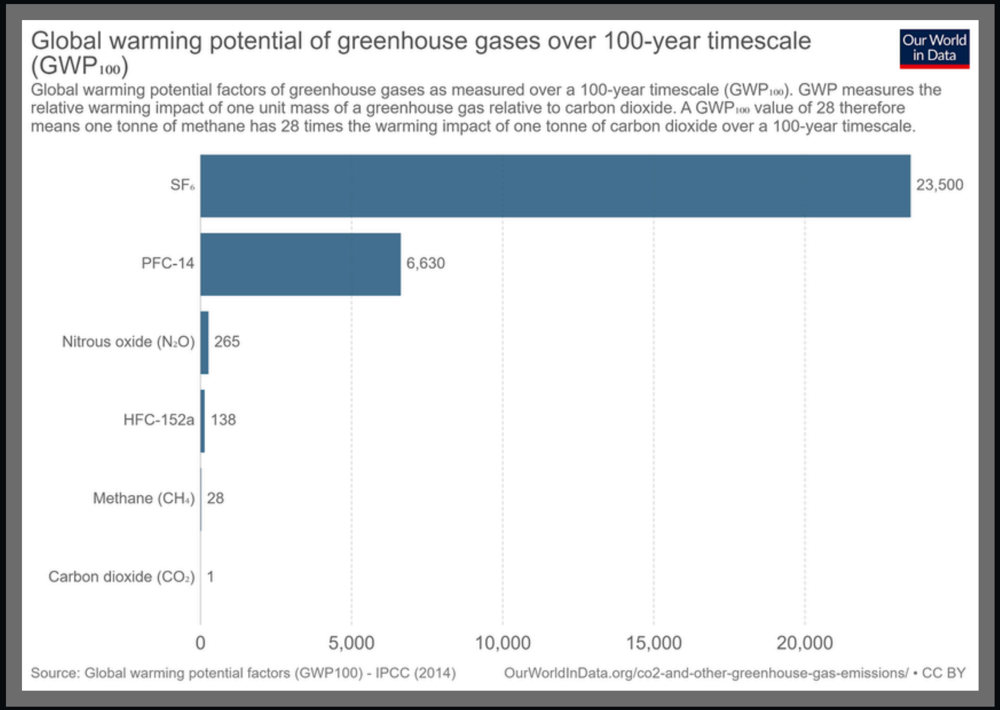

→ **Keep flowing *fresh* target gas**

B. R. Battat et al., Radon in the DRIFT-II directional dark matter TPC: emanation, detection and mitigation, JINST 9 (2014) P11004.
R. Guida et al., Effects of gas mixture quality on GEM detectors operation, J. Phys.: Conf. Ser. 1498 (2020) 012036

# Introduction



- $SF_6$ gas become of interest in directional dark matter searches
- Future large scale plans CYGNUS-1000 utilising 1000 $m^3$ of $SF_6$
- Continuously using *fresh* $SF_6$ gas is problematic due to strict regulations with the use F-gases

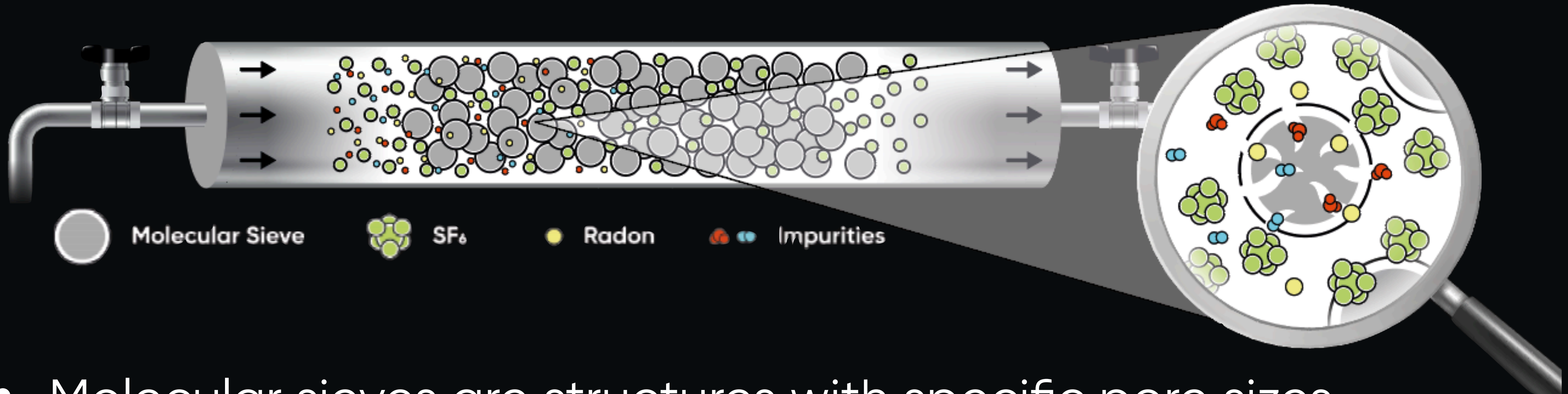



→ **We need an alternative method that *recycles* $SF_6$ gas**

S. E. Vahsen, C. A. J. O'Hare, W. A. Lynch, et al., Cygnus: Feasibility of a nuclear recoil observatory with directional sensitivity to dark matter and neutrinos, 2020.

# Molecular Sieves (MS)





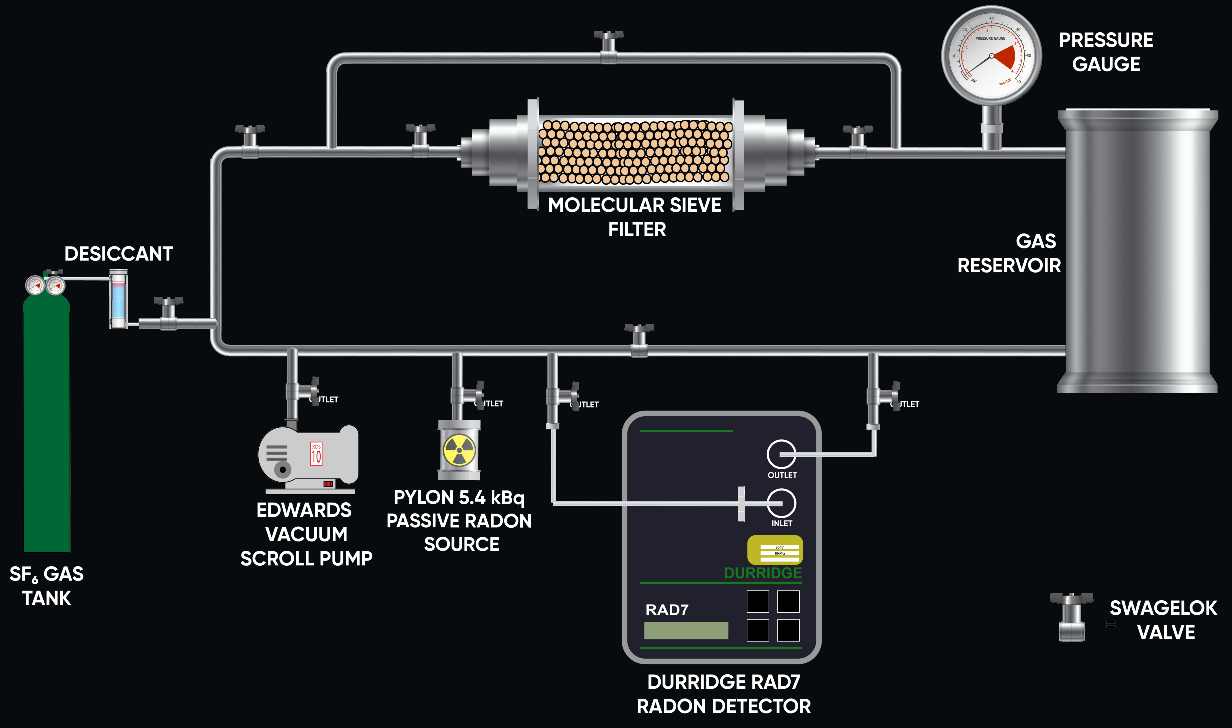

- Molecular sieves are structures with specific pore sizes (four different types: 3A, 4A, 5A and 13X)
- Pores allow molecules with the critical diameter equal or below to be adsorbed on to the structure
- Molecules with diameters larger than the critical diameters pass between the bead gaps

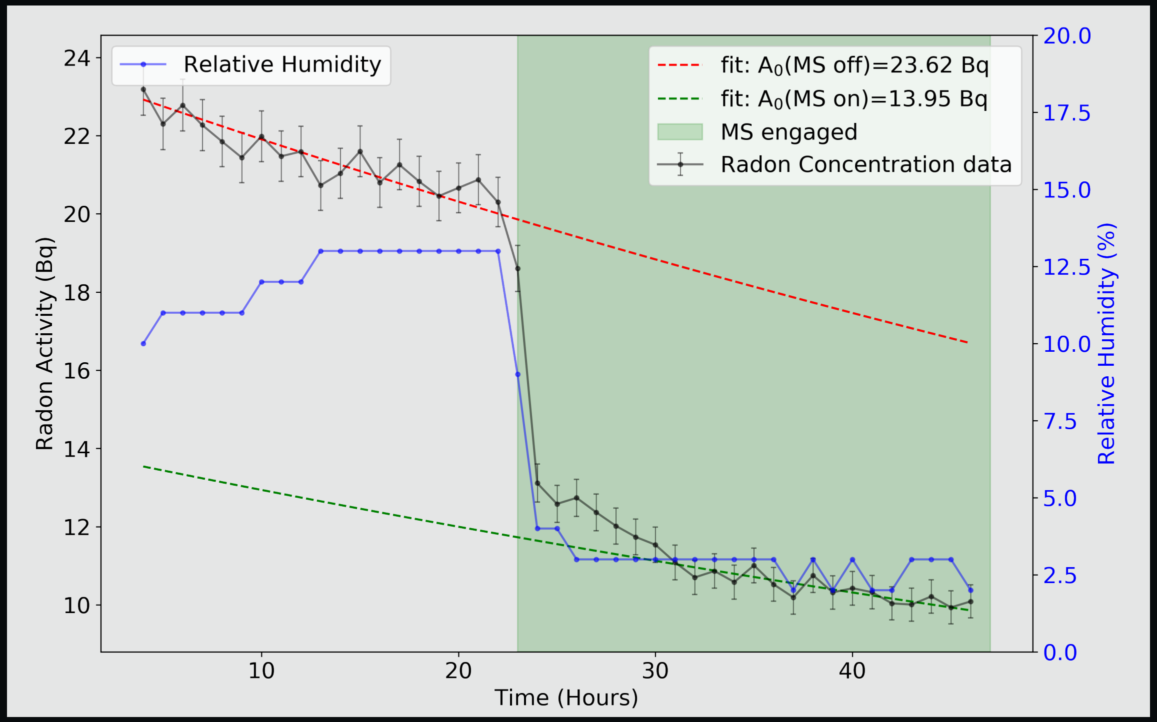
Sigma-Adrich, Molecular Sieves-Technical Information Bulletin, AL-143 Mineral Adsorbents, Filter Agents and Drying Agents (2020).

# Molecular Sieves (MS)



## Demonstration of radon removal from $SF_6$



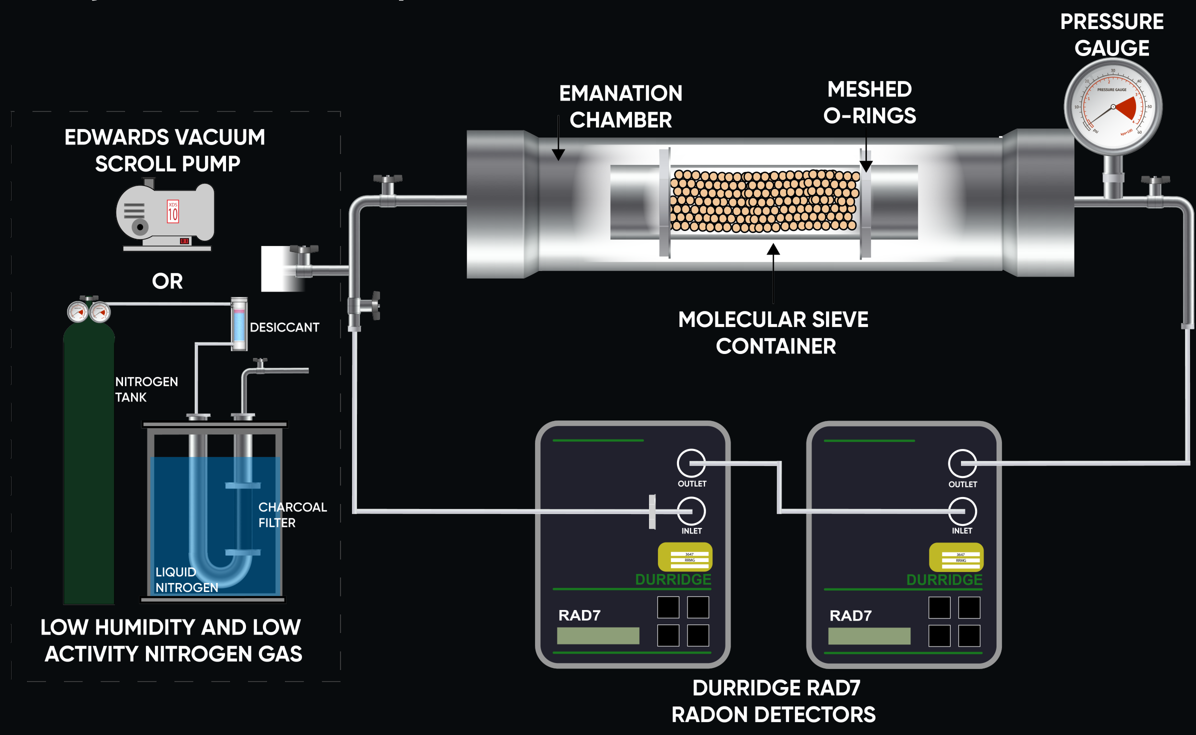

Radon removed by 5A type MS from $SF_6$
at room temperature **(97±1 Bq/Kg)**

R. R. Marcelo Gregorio et al., Demonstration of radon removal from SF6 using molecular sieves, JINST 12 (2017) P09025.

# Low Background MS



Commercial MS intrinsically emanate radon at levels unsuitable for ultra-sensitive rare-event physics experiments




Commercial 5A type MS: **525±37 mBq/kg**

Goal is to **maximise amount of MS allowed** by radioactive budget of an experiment ~ 1 mBq

R.R. Marcelo Gregorio et al., Test of low radioactive molecular sieves for radon filtration in SF6 gas-based rare-event physics experiments, 2021 JINST 16 P06024

# Low Background MS



Nihon University in collaboration with Union Showa K.K., has developed a method of producing low radioactive MS

Commercial
Sigma-Aldrich

Nihon Uni
MS V1

Nihon Uni
MS V2

To provide a complete comparison of the MS candidates, the results from the **emanation** and **filtration** tests were combined

H. Ogawa et al., Development of low radioactive molecular sieves for ultra-low background particle physics experiment, JINST 15 (2020) P01039.

# Low Background MS



| MS | Geometry | Rn emanated mBq/kg | Rn Captured Bq/kg |
|---|---|---|---|
| Sigma Aldrich (Commercial) | 8-12 mm uniform | 525±37 | 97±1 |
| Nihon-Uni (V1) | 1-2 cm Granules | 99±23 | 35±2 |
| | Fine Powder | 680±30 | 330±3 |
| **Nihon-Uni (V2)** | **Powder** | **<32** | **254±3** |

The NU-developed (V2) 5Å MS emanated radon **at least 98% less** per radon captured, compared to the commercial MS

R.R. Marcelo Gregorio et al., Test of low radioactive molecular sieves for radon filtration in SF6 gas-based rare-event physics experiments, 2021 JINST 16 P06024

# Overview so far



✓ Removes radon - **5A Type MS**

✓ Low intrinsic radioactivity MS **- NU V2 MS**

✓ Remove common impurities - **3A, 4A Type MS**

✓ Does not absorb target gas $SF_6$ - **3A, 4A, 5A Type MS**

Ideal for pure $SF_6$ experiment!

→ **How about other target gases?**

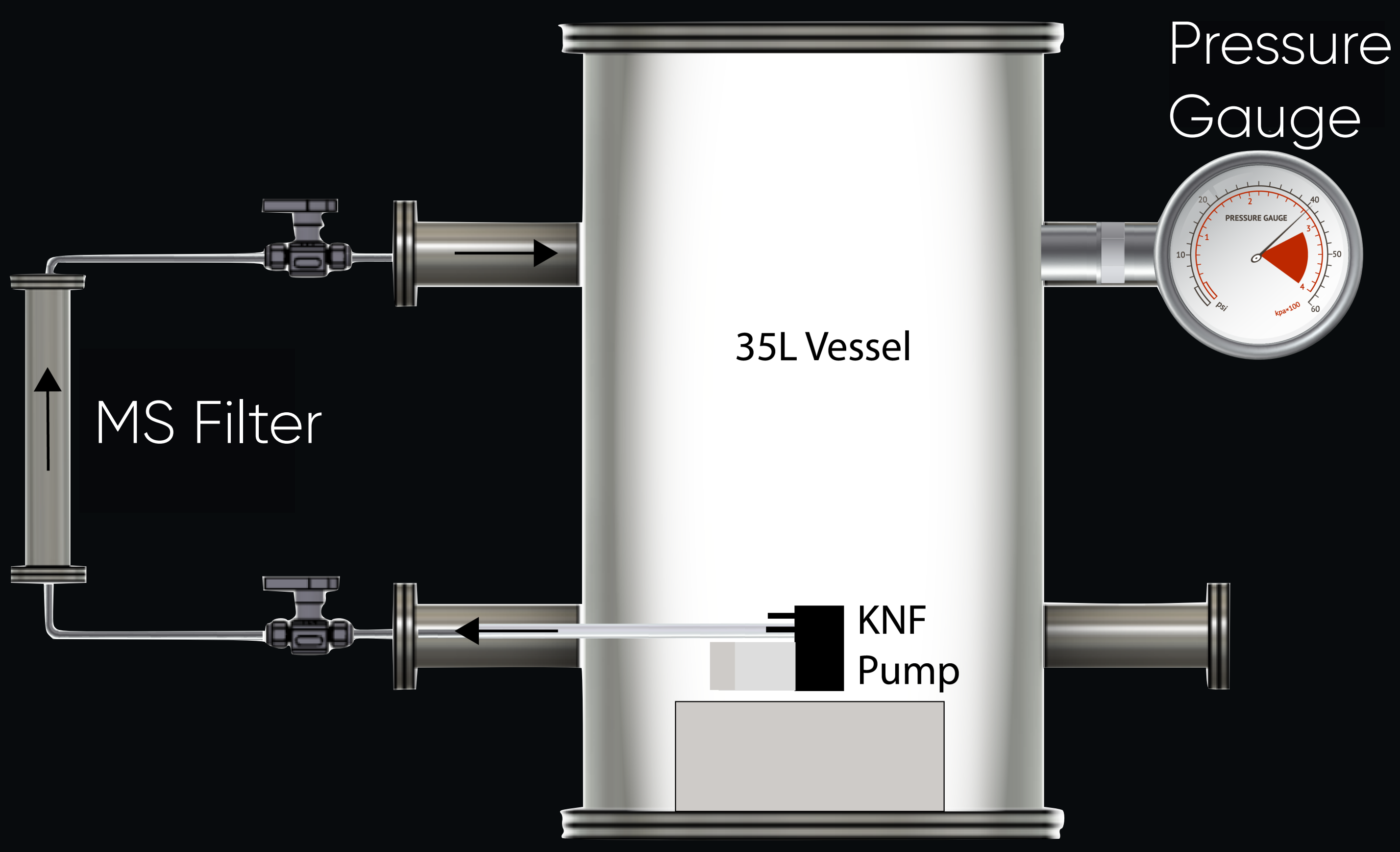
# Application to other gases



## Does the MS absorb the desired target gas?

**3A/4A**- removes $N_2$ ,$O_2$ and $H_2O$

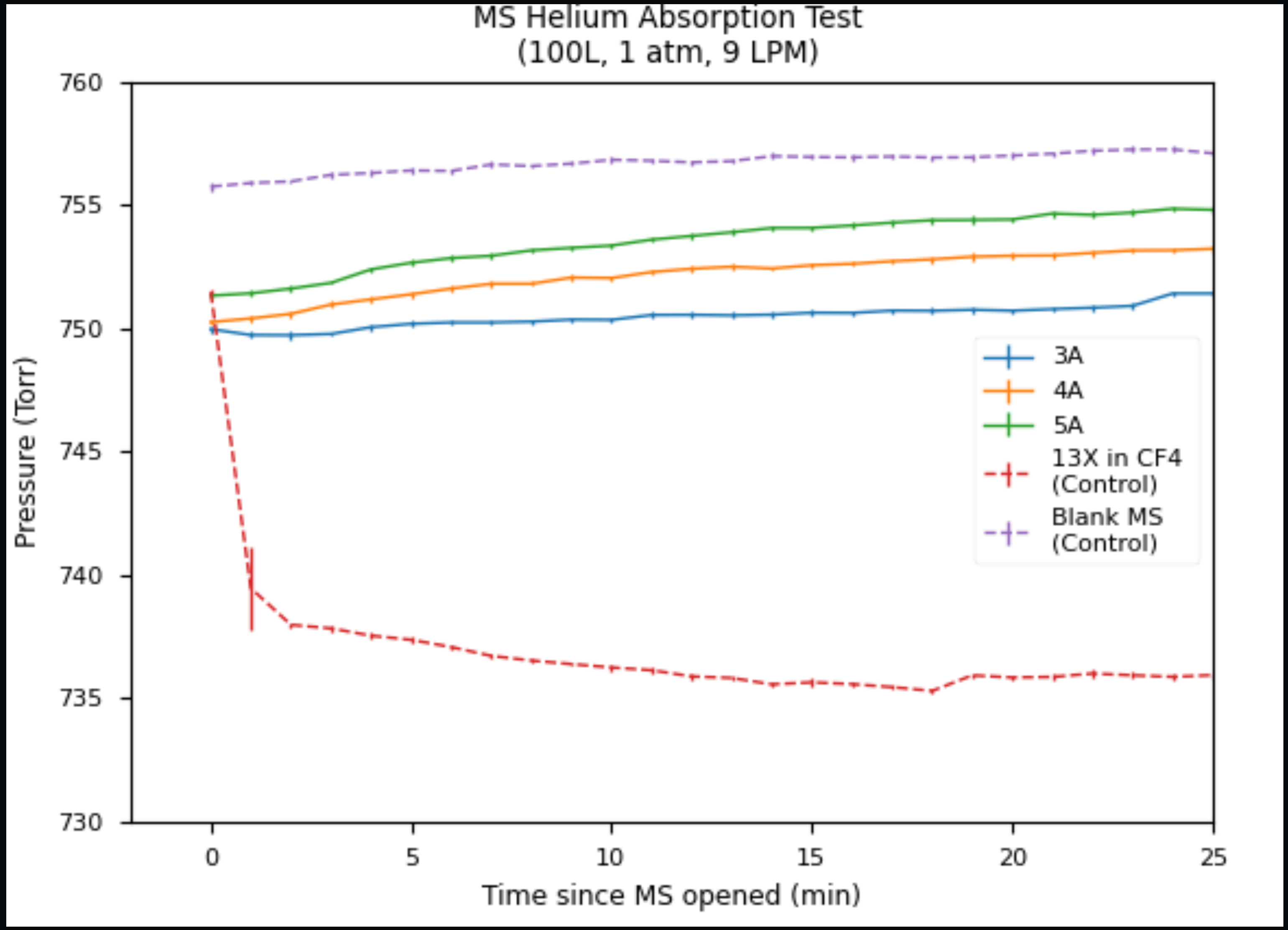
**5A** - removes radon



# Application to other gases: He



## Helium absorption test




**3A/4A**- removes $N_2$ ,$O_2$ and $H_2O$
**5A** - removes radon
**13X in $CF_4$**- absorption control

- ✓ 3Å, 4Å and 5Å type does not absorb helium
- ✓ Purification with radon removal suitable for helium in pure form and mixtures

# Application to other gases: $CF_4$



## $CF_4$ Absorption test

| MS | Pressure change after 20m (torr) | Absorption per mass of MS (torr/kg) | Notes |
|---|---|---|---|
| 3A | - | - | |
| 4A | 1±3 | -3±6 | |
| 5A | -39±3 | 87±7 | |
| 13X | -26±3 | 67±8 | Control |
| Activated Charcoal | -54±3 | 197±11 | |

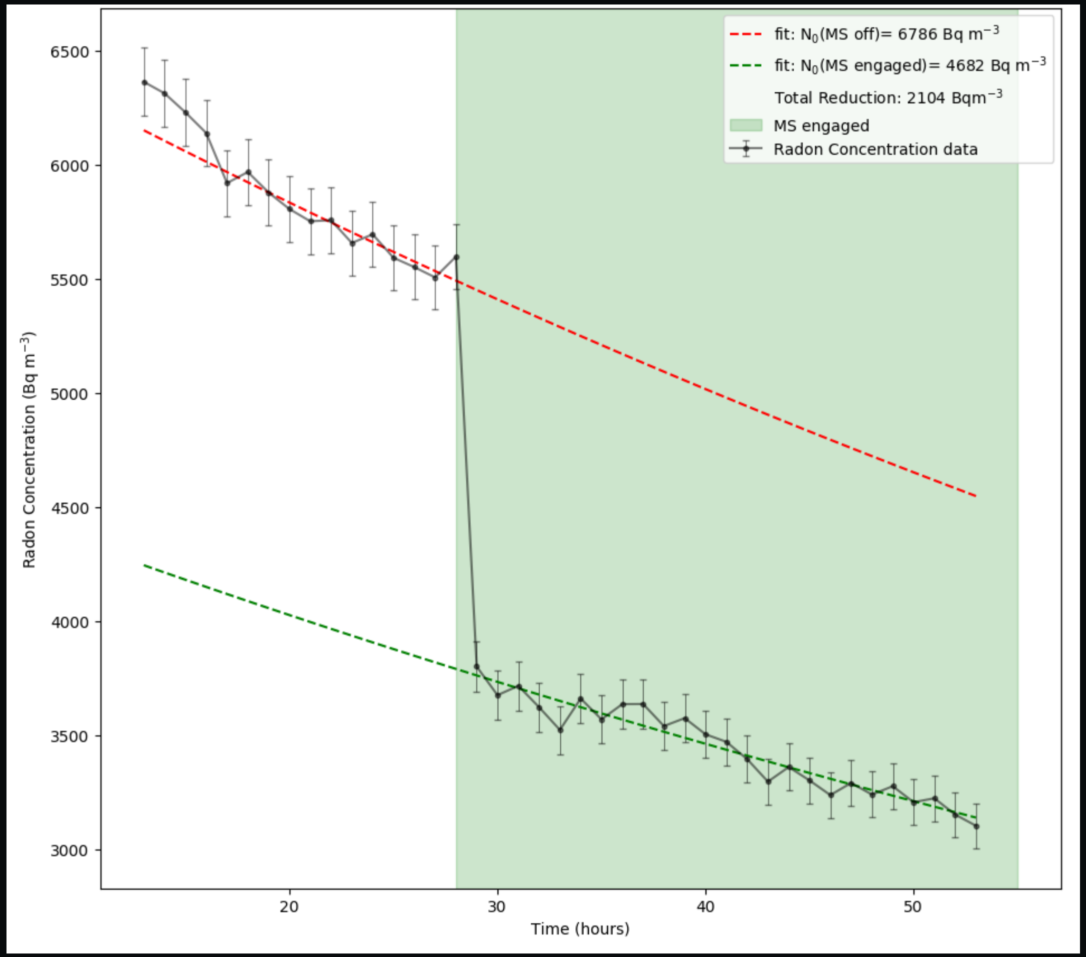
**3A/4A**- removes $N_2$, $O_2$ and $H_2O$
**5A** - removes radon
**13X** - absorption control

- ✓ 3Å and 4Å type does not absorb $CF_4$
- ✓ 5Å type absorbs $CF_4$ at 87±7 torr/kg

# Application to other gases: $CF_4$



## Radon removal from $CF_4$




100±1 Bq/Kg

Pressure drop: ~12 torr

- ✓ 5Å type absorbs $CF_4$ but still removes radon (c.f. $SF_6$ 97±1 Bq/Kg)
- ✓ Purification suitable for pure $CF_4$ but mixtures are problematic

# MS filters and target gases



| Experiment's Target Gas | MS Filter (5A) For removal radon only | MS Filter (3A:4A:5A) For removal of common impurities including radon | MS Filter (3A:4A) For removal of common impurities (no radon removal) |
|---|---|---|---|
| $SF_6$ | ✓ | ✓ | ✓ |
| $CF_4$ | ✓ | ✓ | ✓ |
| $SF_6$:He | ✓ | ✓ | ✓ |
| CF4:He | ✗ | ✗ | ✓ |
| $SF_6$:$CF_4$ | ✗ | ✗ | ✓ |
| $SF_6$:$CF_4$:He | ✗ | ✗ | ✓ |

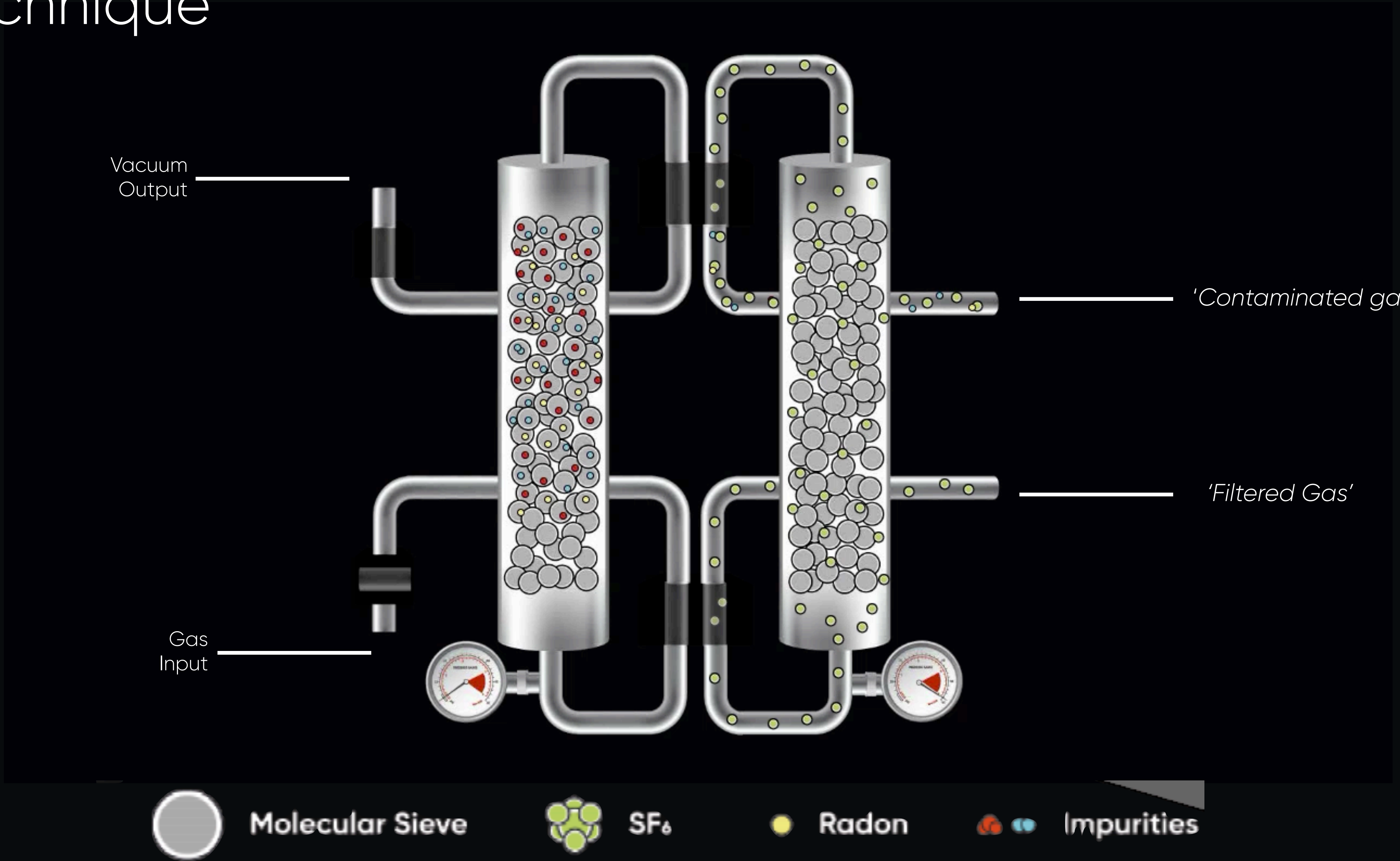
✓ **MS filter ready to use 'as is'**

✗ **Issues with conserving target gas mixing ratio**

# Gas System Design



Dual MS design utilises **Vacuum Swing Adsorption (VSA)** Technique

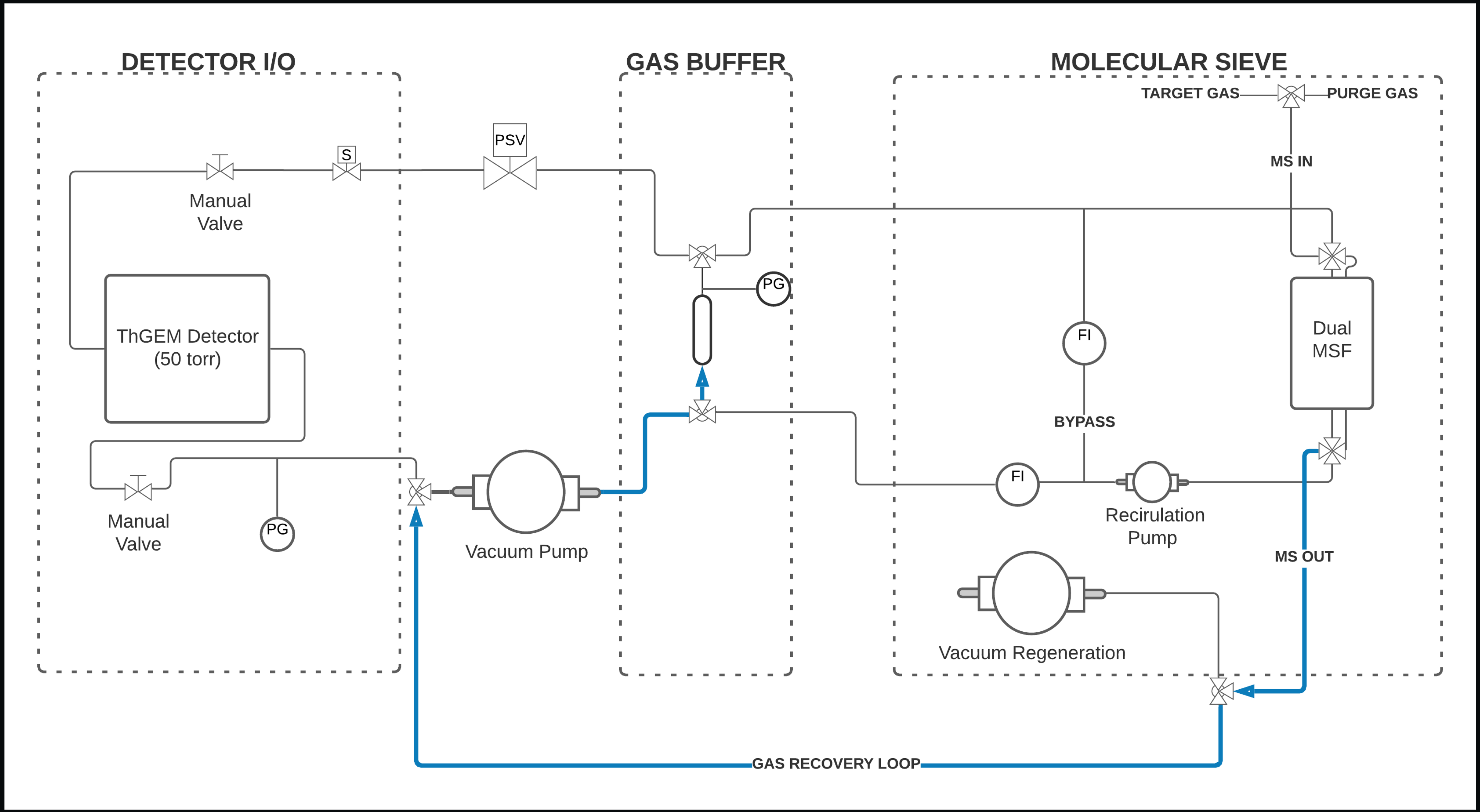



Notice small amount of our desired gas is lost during vacuum regeneration

# Gas System Design

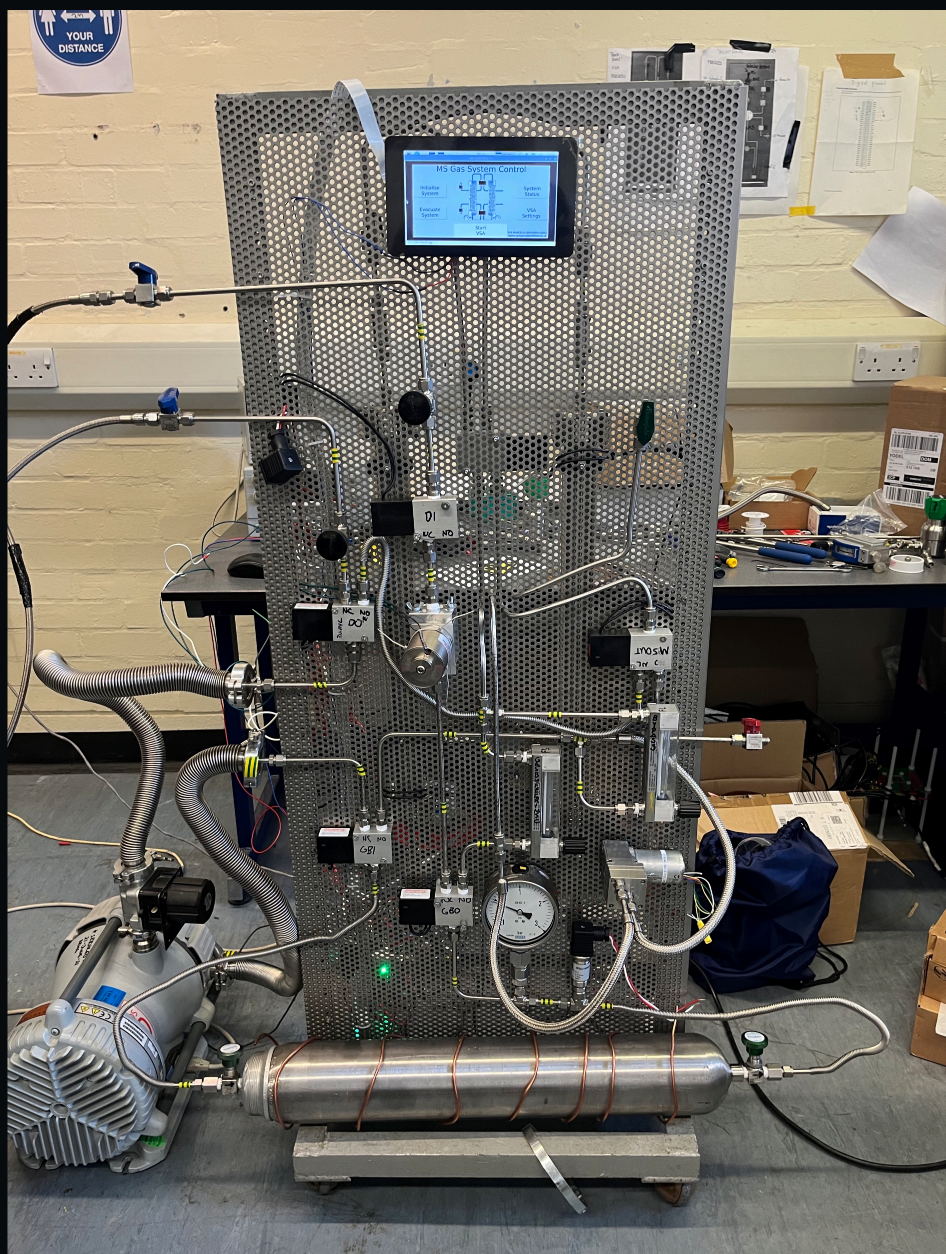


Unlike conventional VSA system we have a **gas recovery loop** allowing recovery of at least 99.99% of total gas used



# Current Status





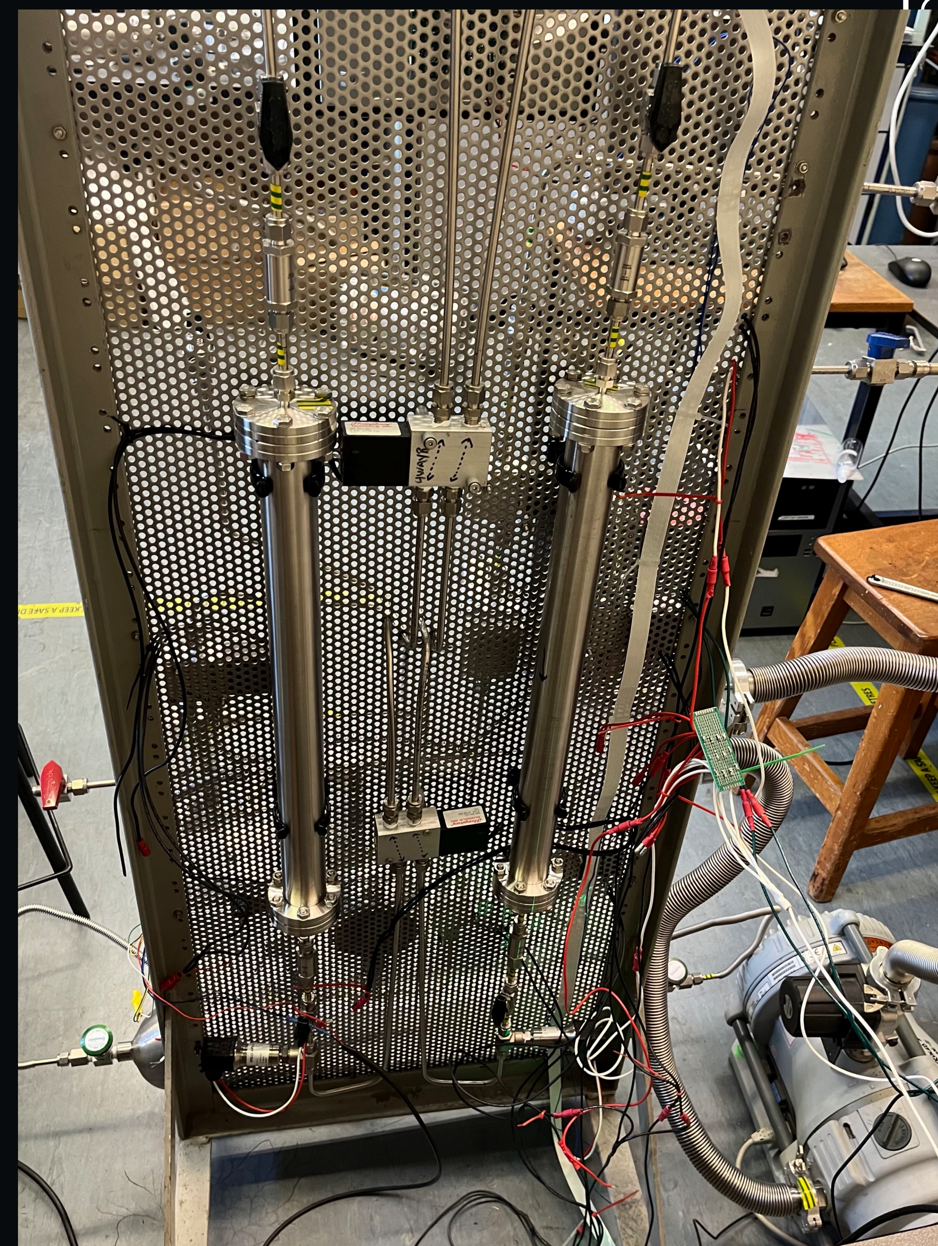
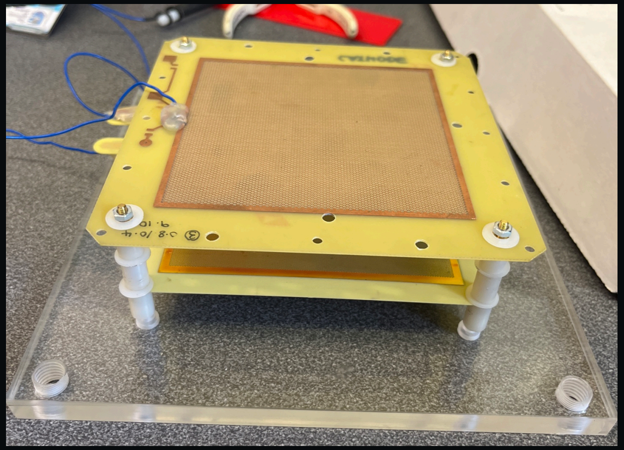


Photo of status
December 2021

# Performance Testing




ThGEM (10 x 10 cm)



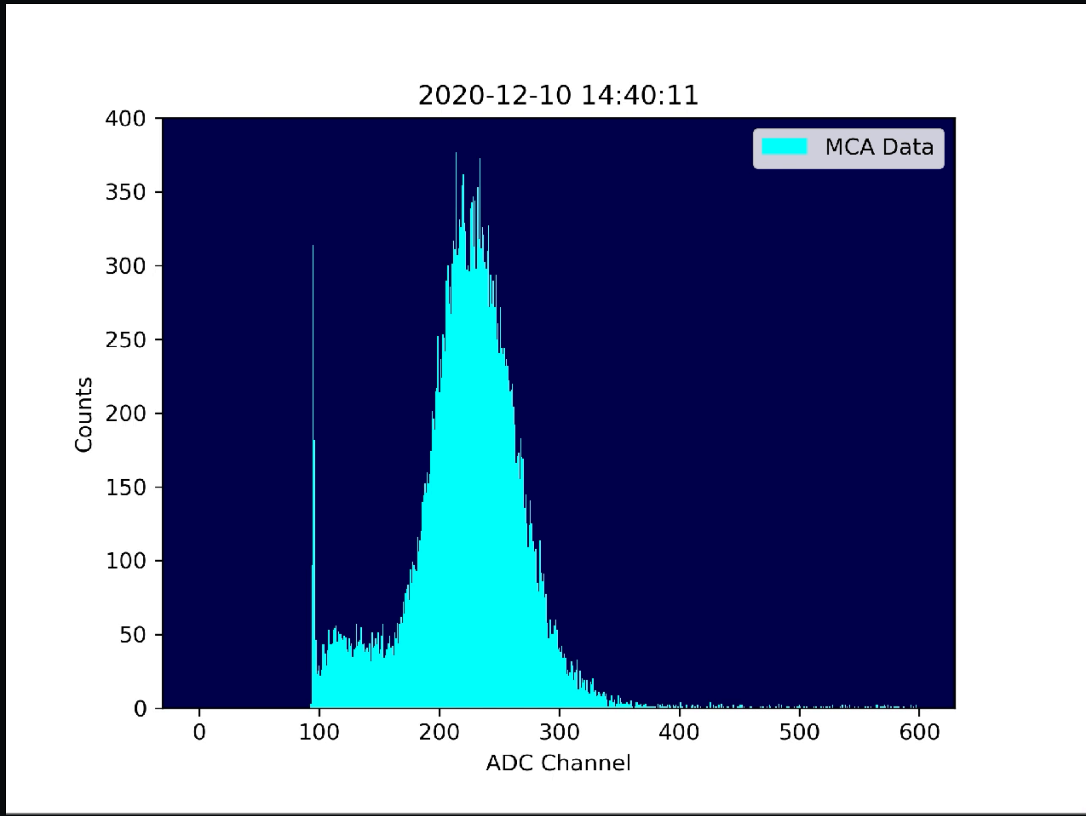
100L Vessel

# Performance Testing



Compare detector operation with and w.o. gas system by measuring:

1. Gas gain detector signal




Example of signal deterioration due to gas contamination in a ThGEM TPC $CF_4$ (Fe-55 calibration source)

2. Experiment's Intrinsic radon background

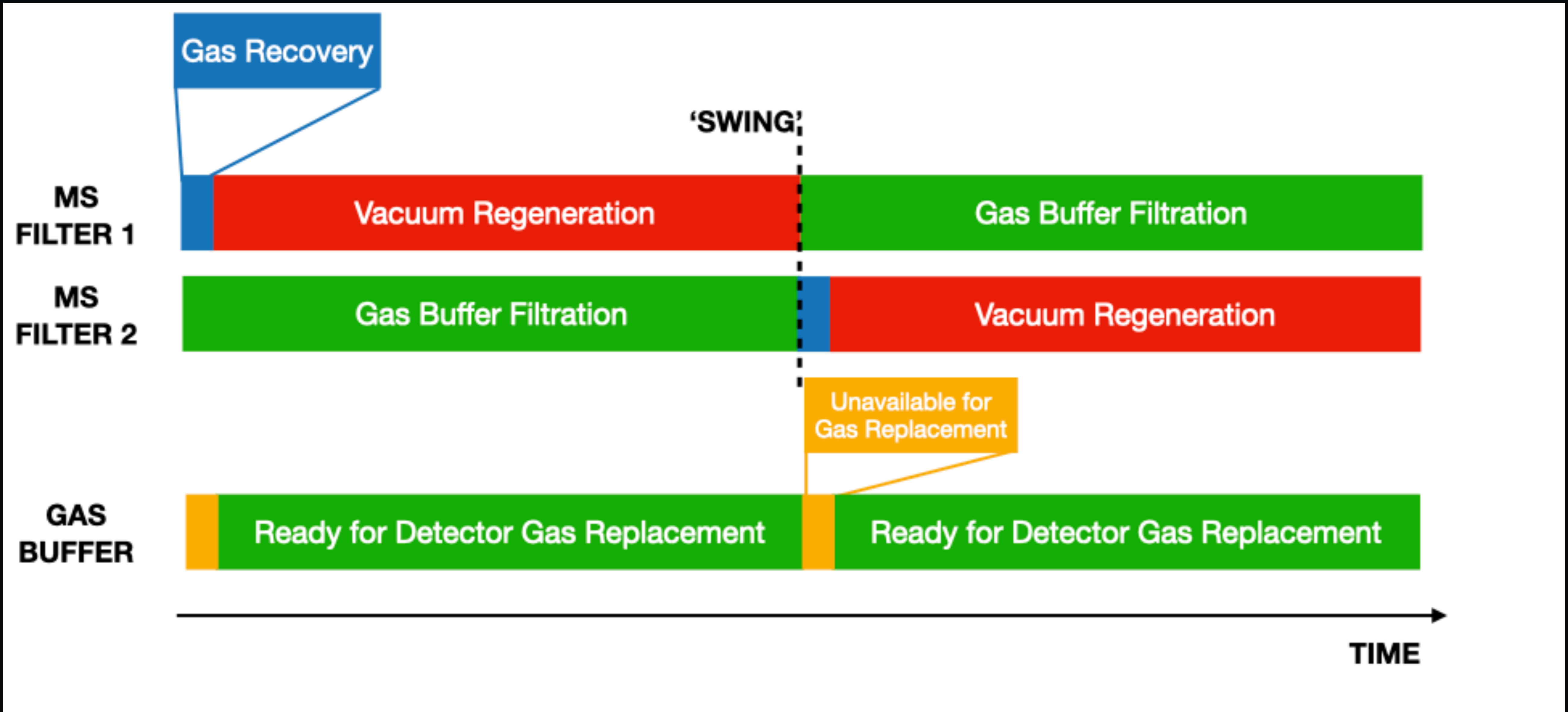
# Conclusions



- Devised an alternative method that recycles gas significantly reducing the total gas used in ultra-sensitive gas-based experiments
- Identified a suitable low background MS candidate
- Presented suitable MS Filters for $SF_6$ , $CF_4$ and He target gases in pure form and mixtures
- Working towards MS gas system demonstration with pure $SF_6$ , $CF_4$ and $SF_6$:He

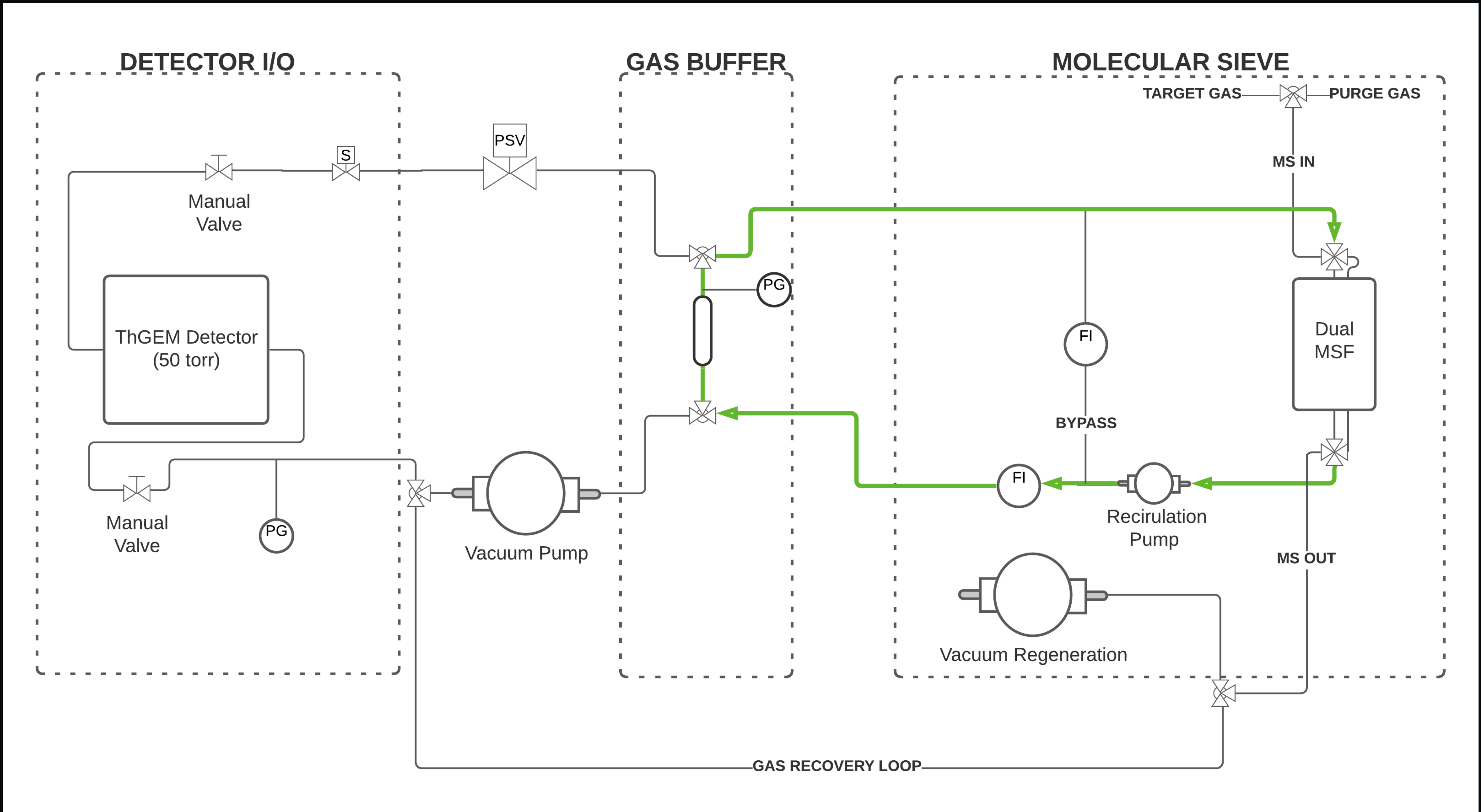
# VSA Operation





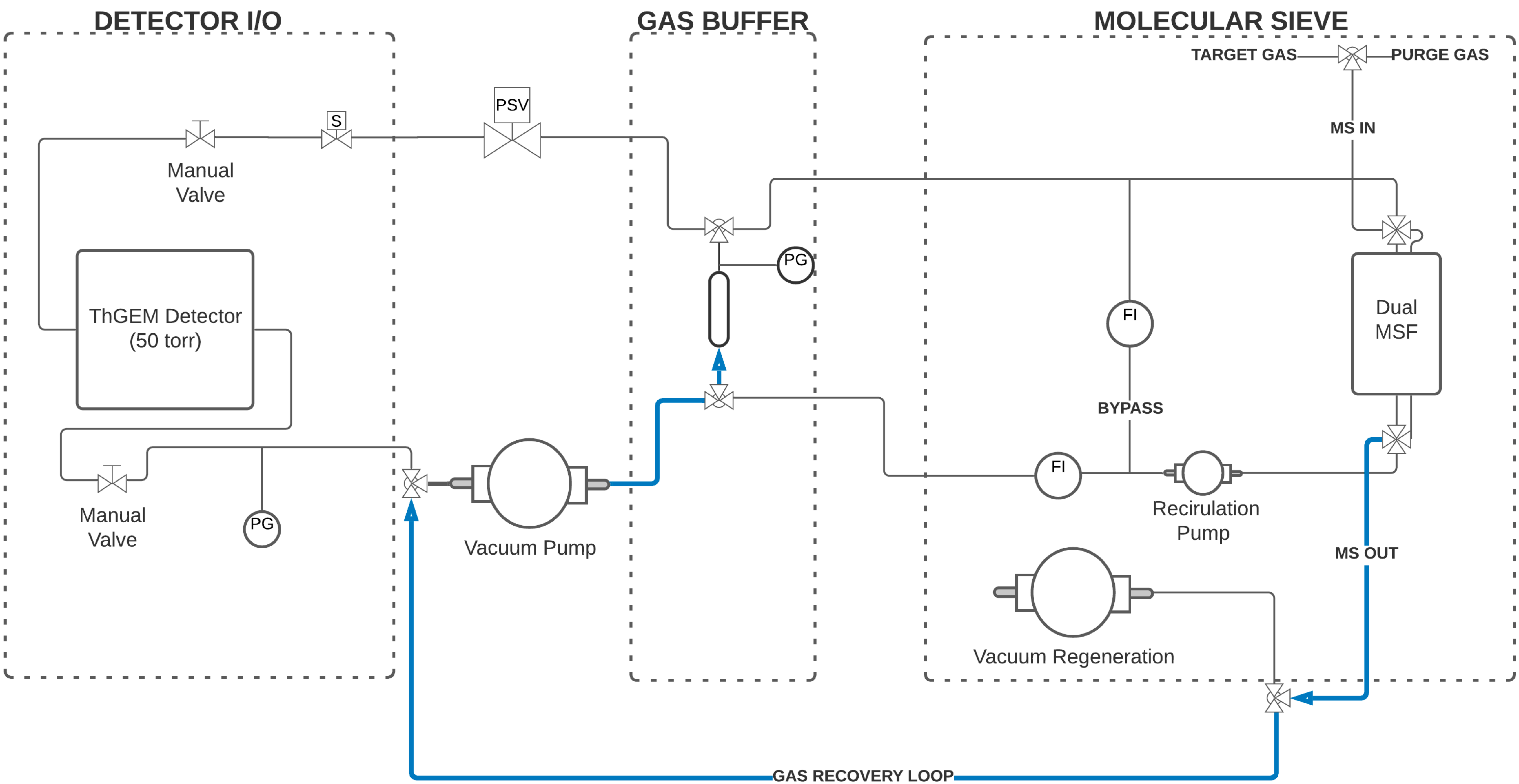
# VSA: Gas Filtration





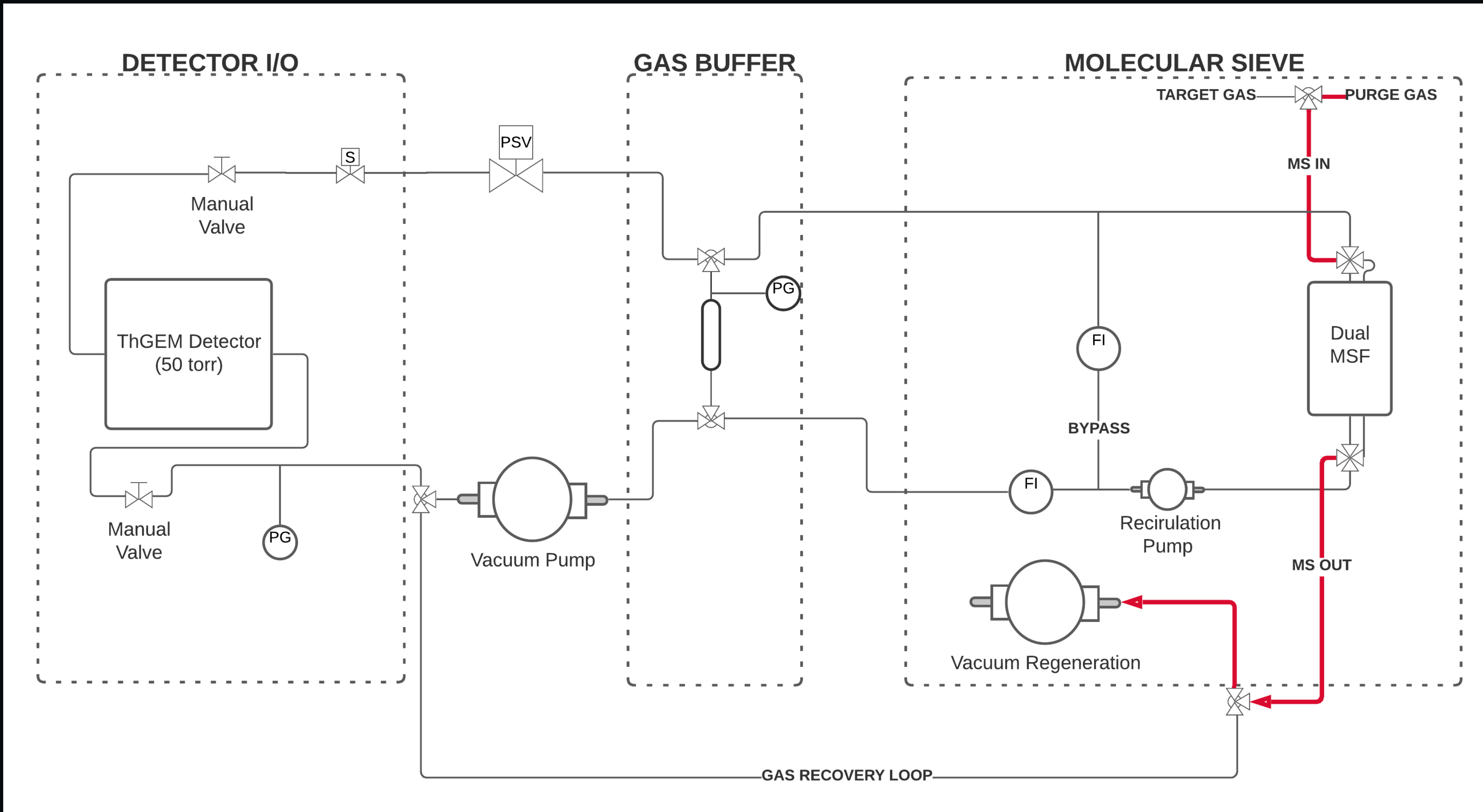
# VSA: Gas Recovery Loop





# VSA: Vacuum Generation





# VSA: Gas Replacement