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

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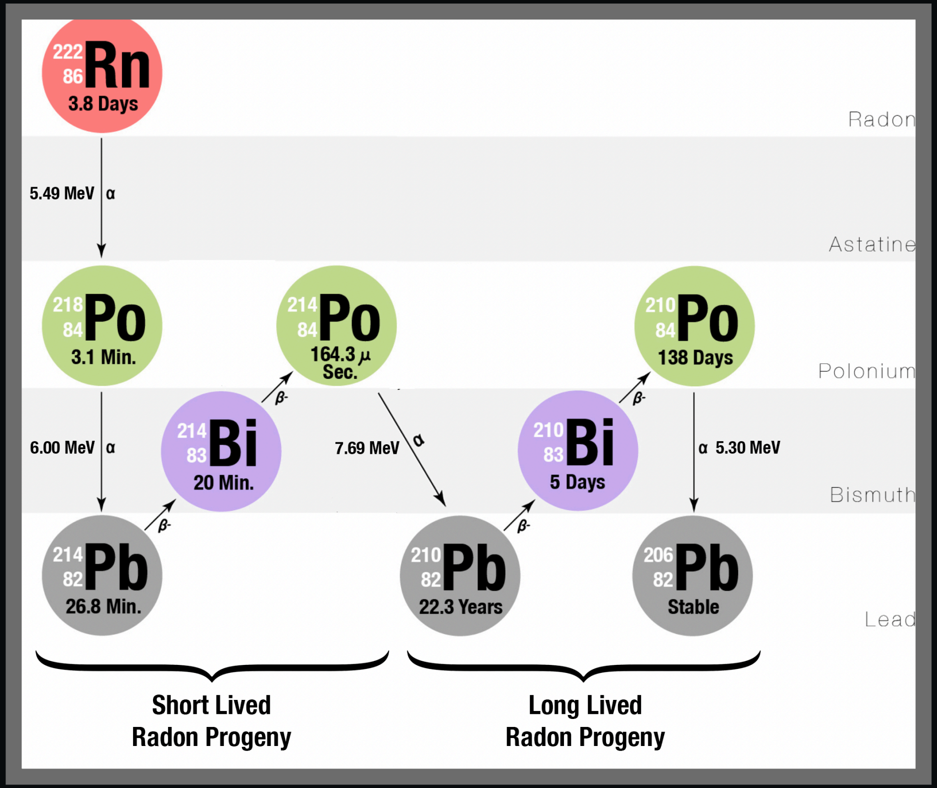
Introduction

- 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**

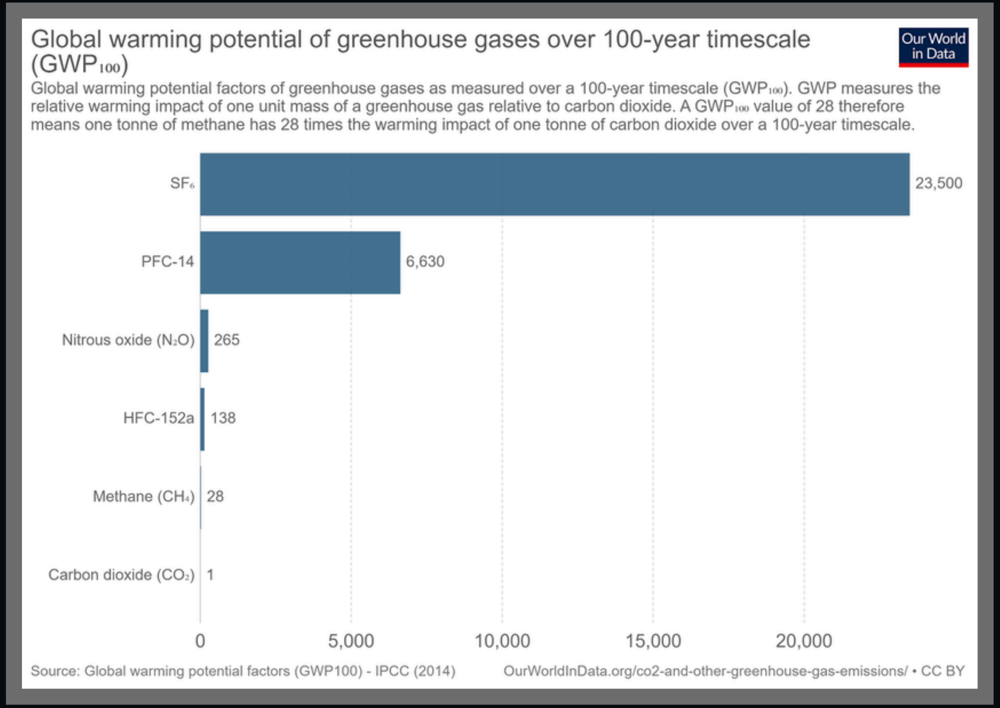


→ **Keep flowing *fresh* target gas**

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

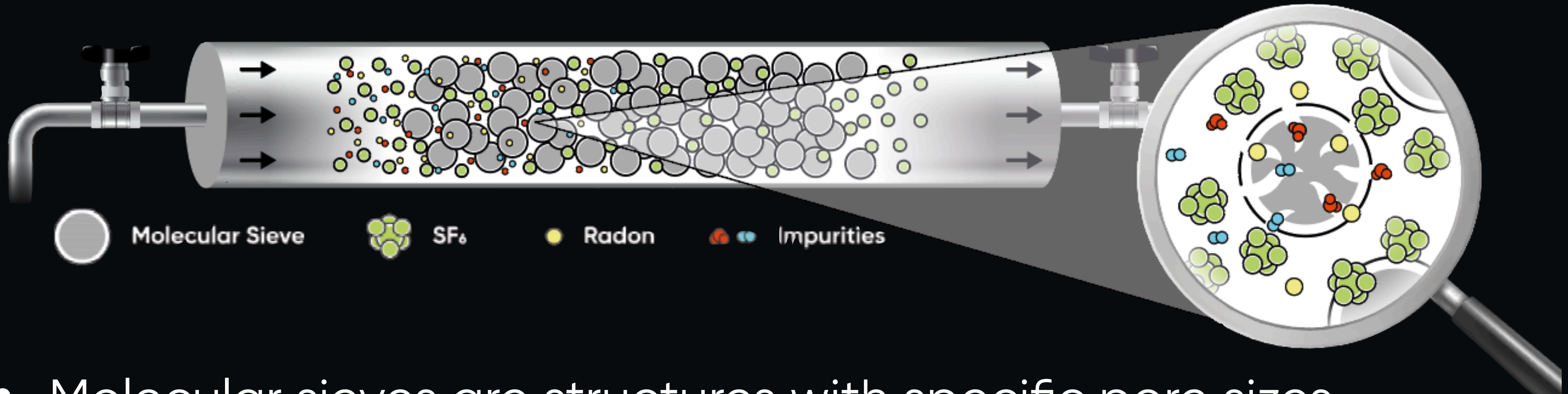
- SF_6 gas become of interest in directional dark matter searches
- Future large scale plans CYGNUS-1000 utilising 1000 m^3 of SF_6
- Continuously using *fresh* SF_6 gas is problematic due to strict regulations with the use F-gases



→ **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)

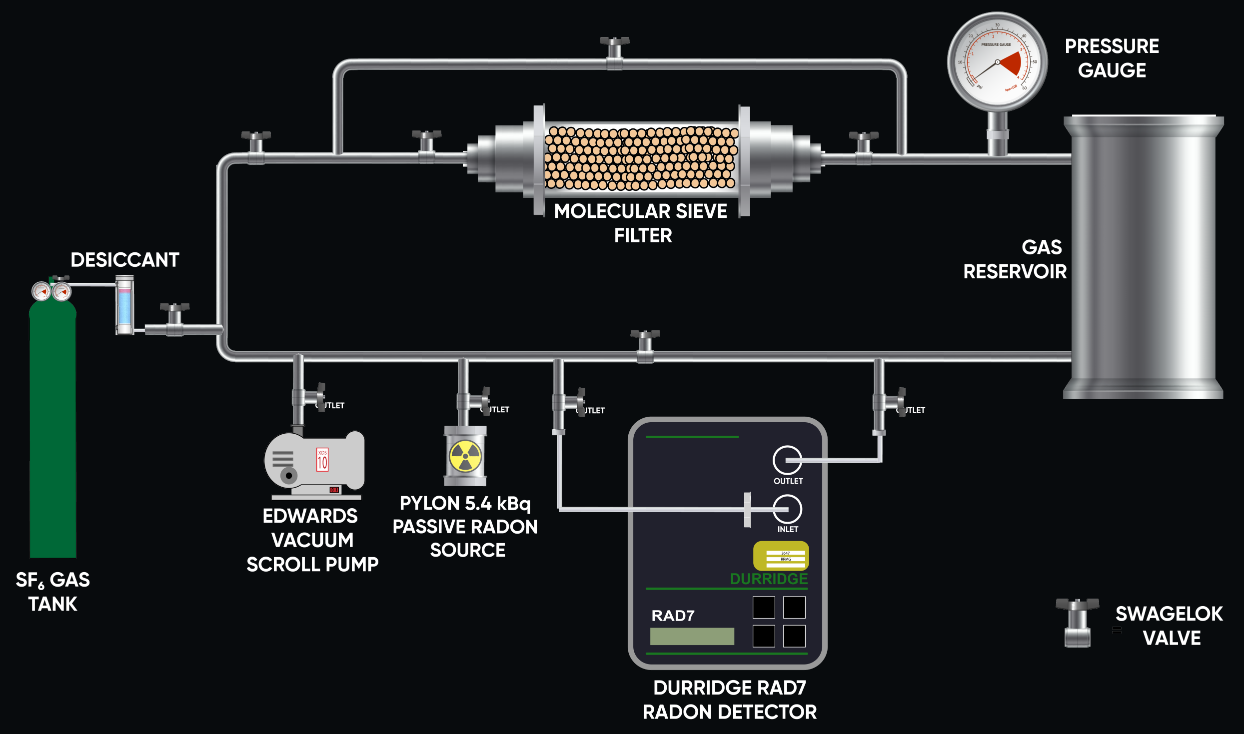


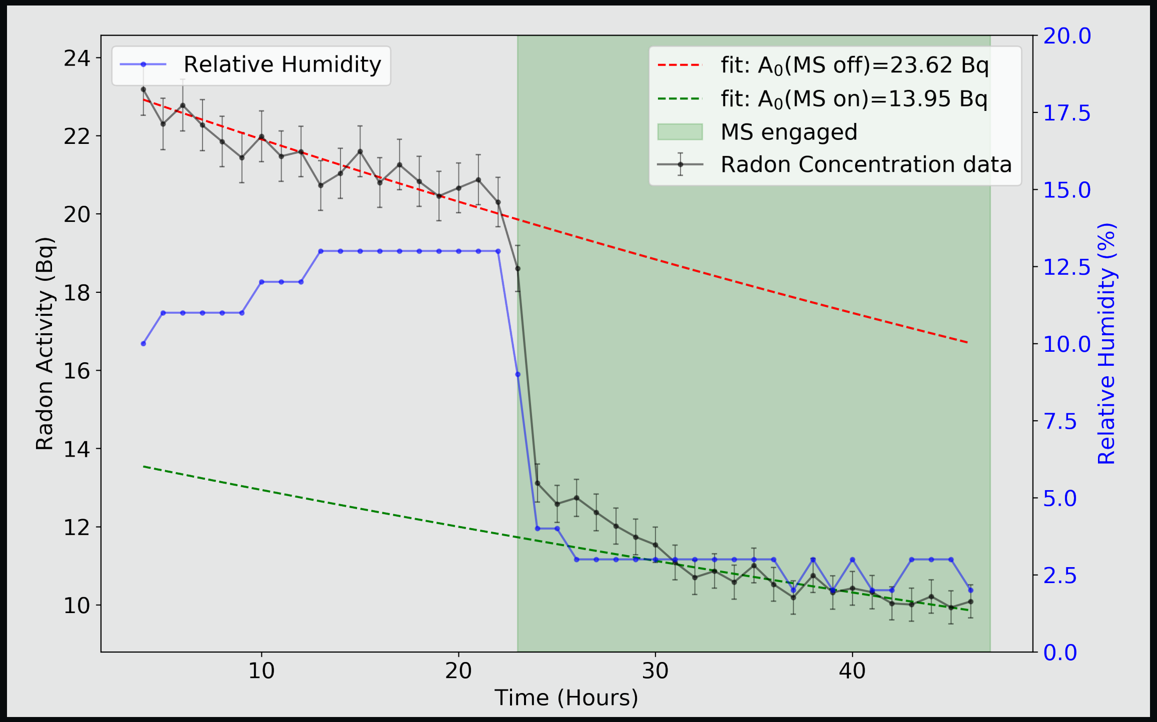
- Molecular sieves are structures with specific pore sizes (four different types: 3A, 4A, 5A and 13X)
- Pores allow molecules with the critical diameter equal or below to be adsorbed on to the structure
- Molecules with diameters larger than the critical diameters pass between the bead gaps

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



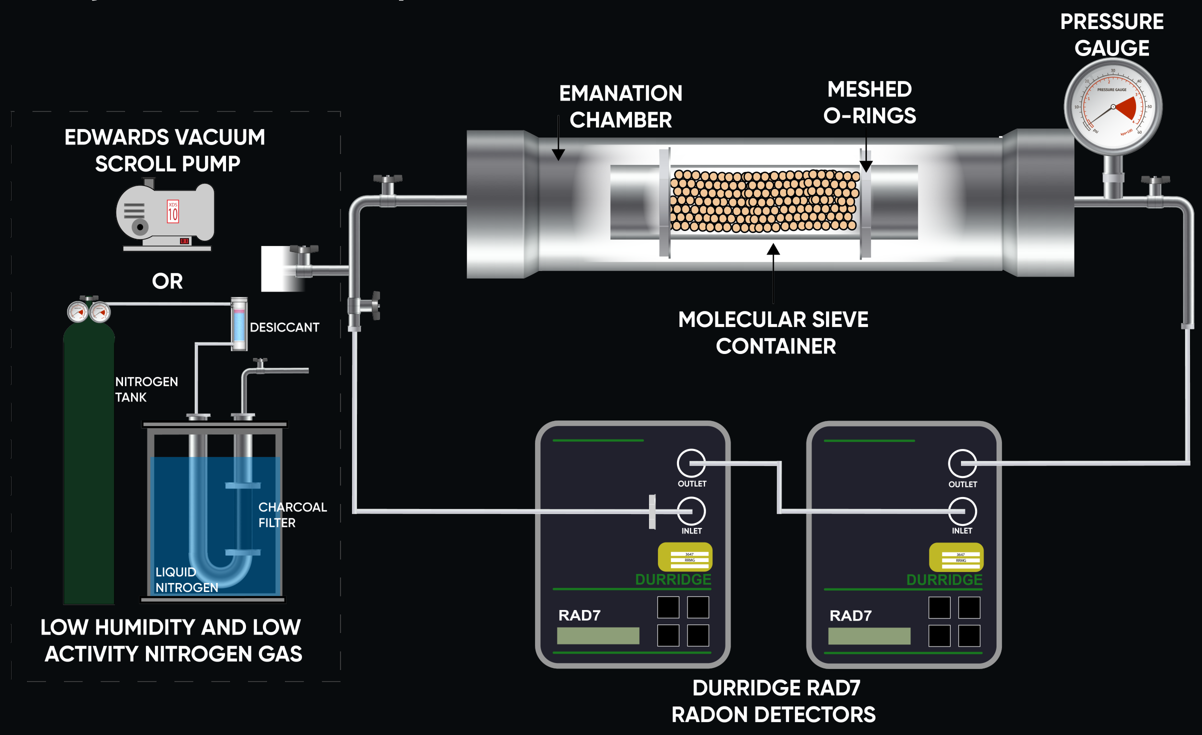


Radon removed by 5A type MS from SF_6 at room temperature **(97±1 Bq/Kg)**

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!

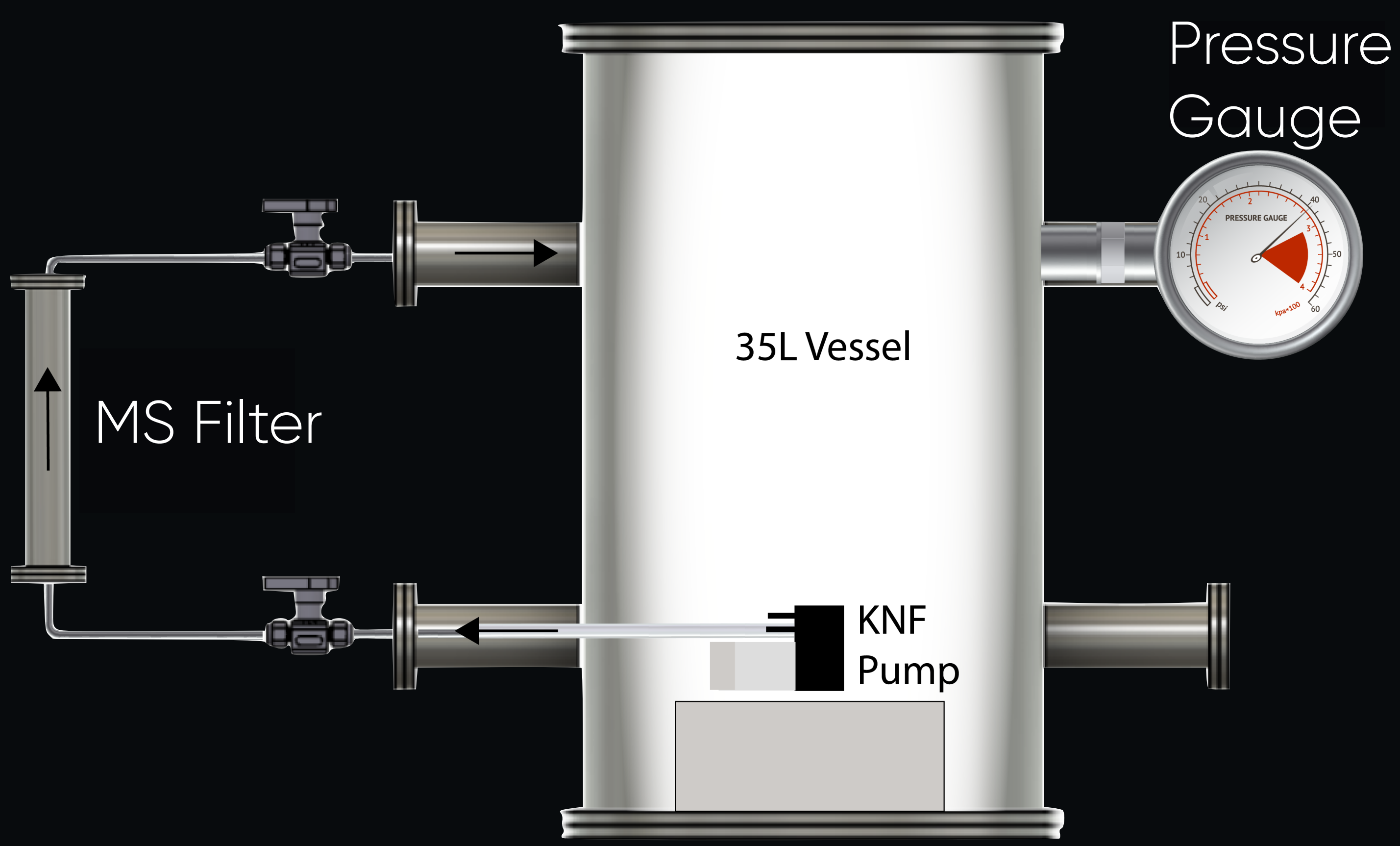
→ **How about other target gases?**

Application to other gases

Does the MS absorb the desired target gas?

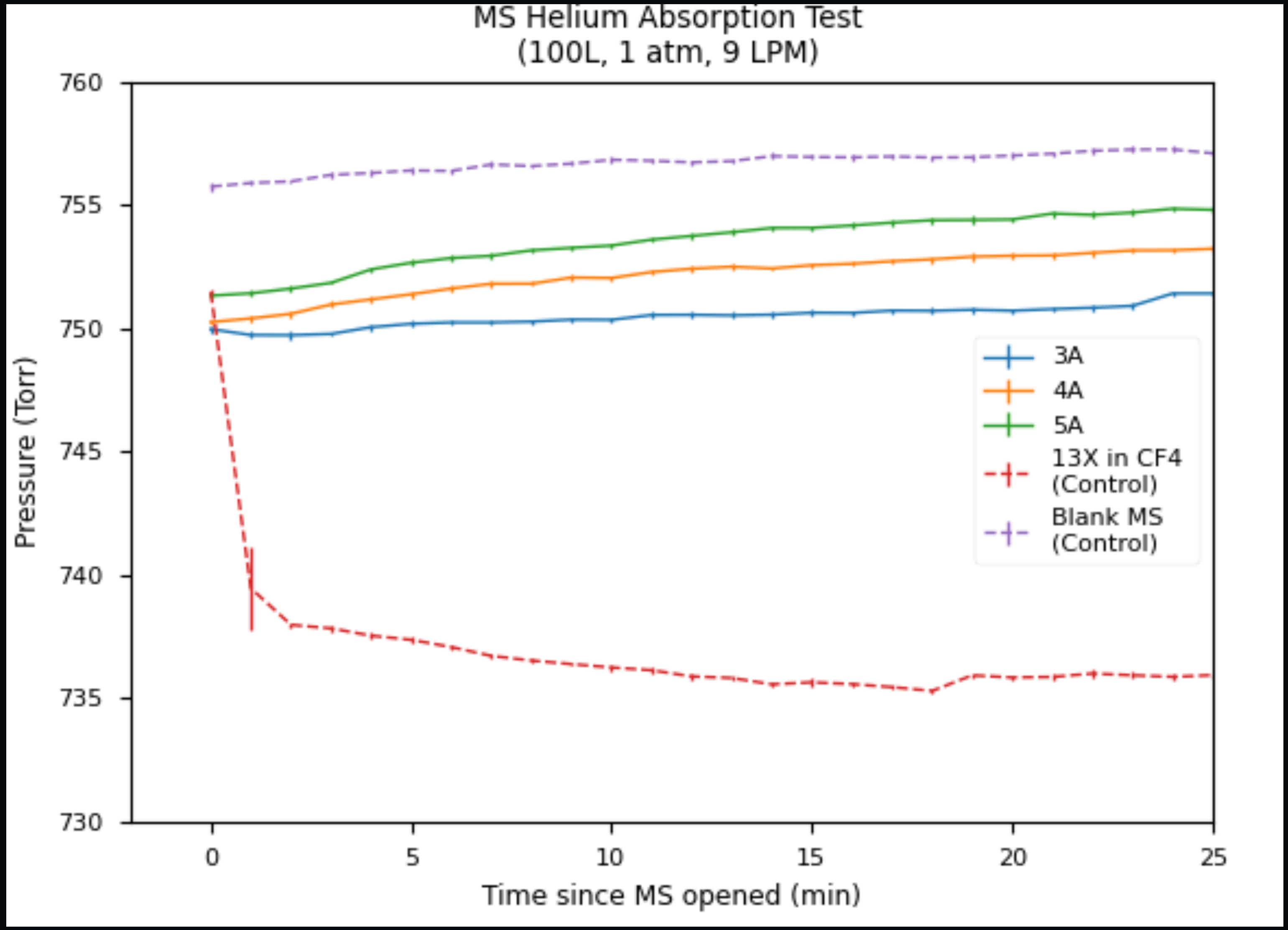
3A/4A- removes N_2, O_2 and H_2O

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

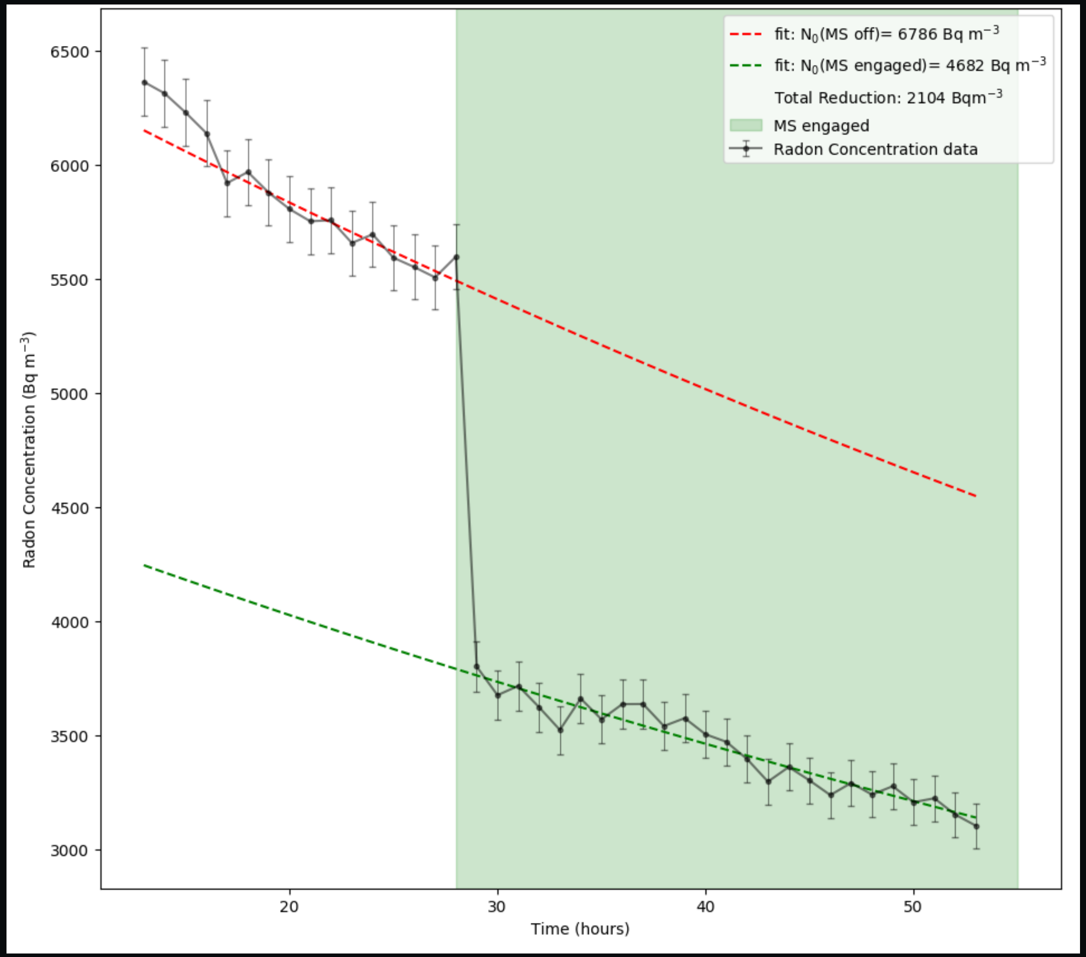
MS	Pressure change after 20m (torr)	Absorption per mass of MS (torr/kg)	Notes
3A	-	-	
4A	1±3	-3±6	
5A	-39±3	87±7	
13X	-26±3	67±8	Control
Activated Charcoal	-54±3	197±11	

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

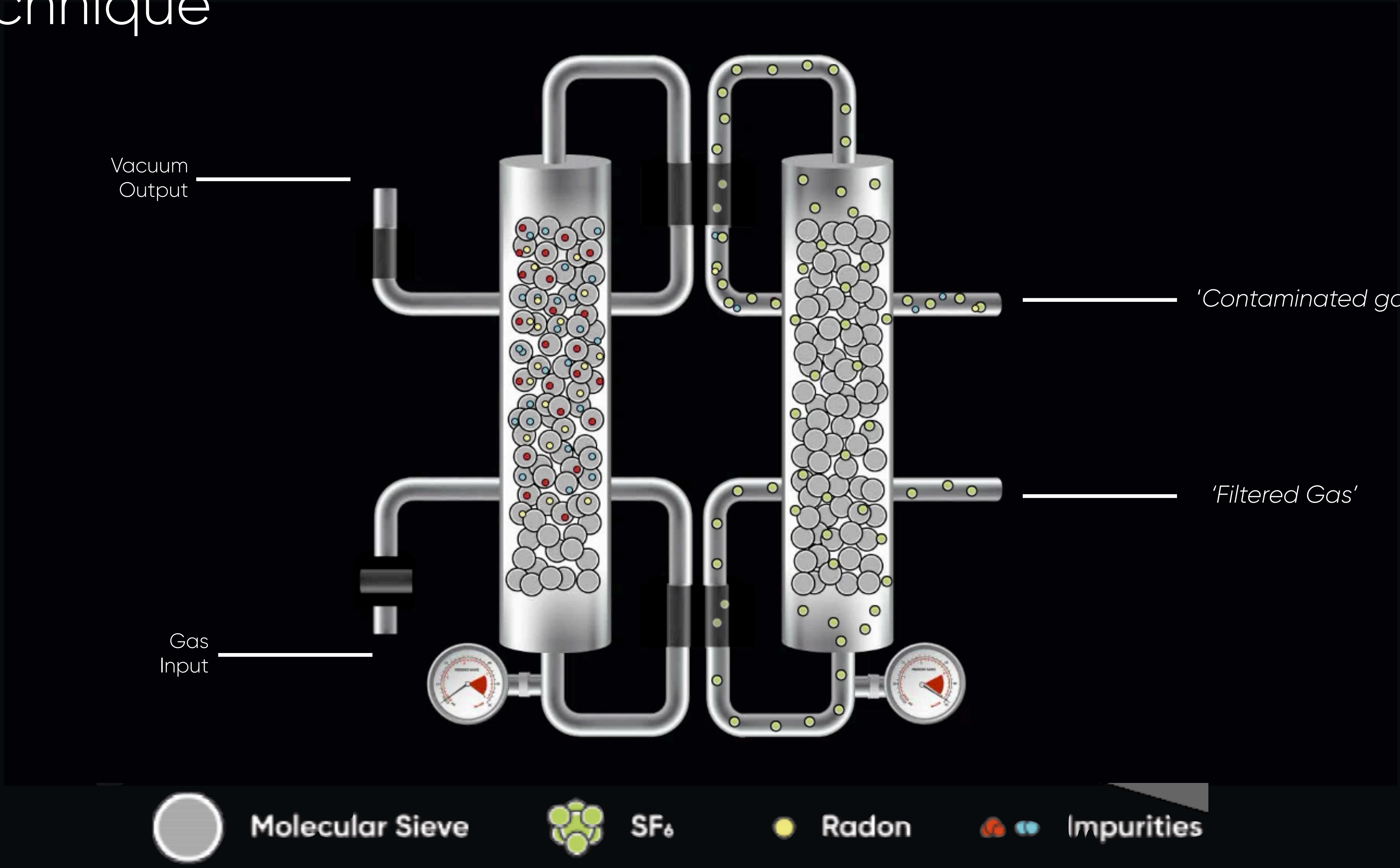
Experiment's Target Gas	MS Filter (5A) For removal radon only	MS Filter (3A:4A:5A) For removal of common impurities including radon	MS Filter (3A:4A) For removal of common impurities (no radon removal)
SF_6	✓	✓	✓
CF_4	✓	✓	✓
SF_6:He	✓	✓	✓
CF4:He	✗	✗	✓
SF_6:CF_4	✗	✗	✓
SF_6:CF_4:He	✗	✗	✓

✓ **MS filter ready to use 'as is'**

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

Gas System Design

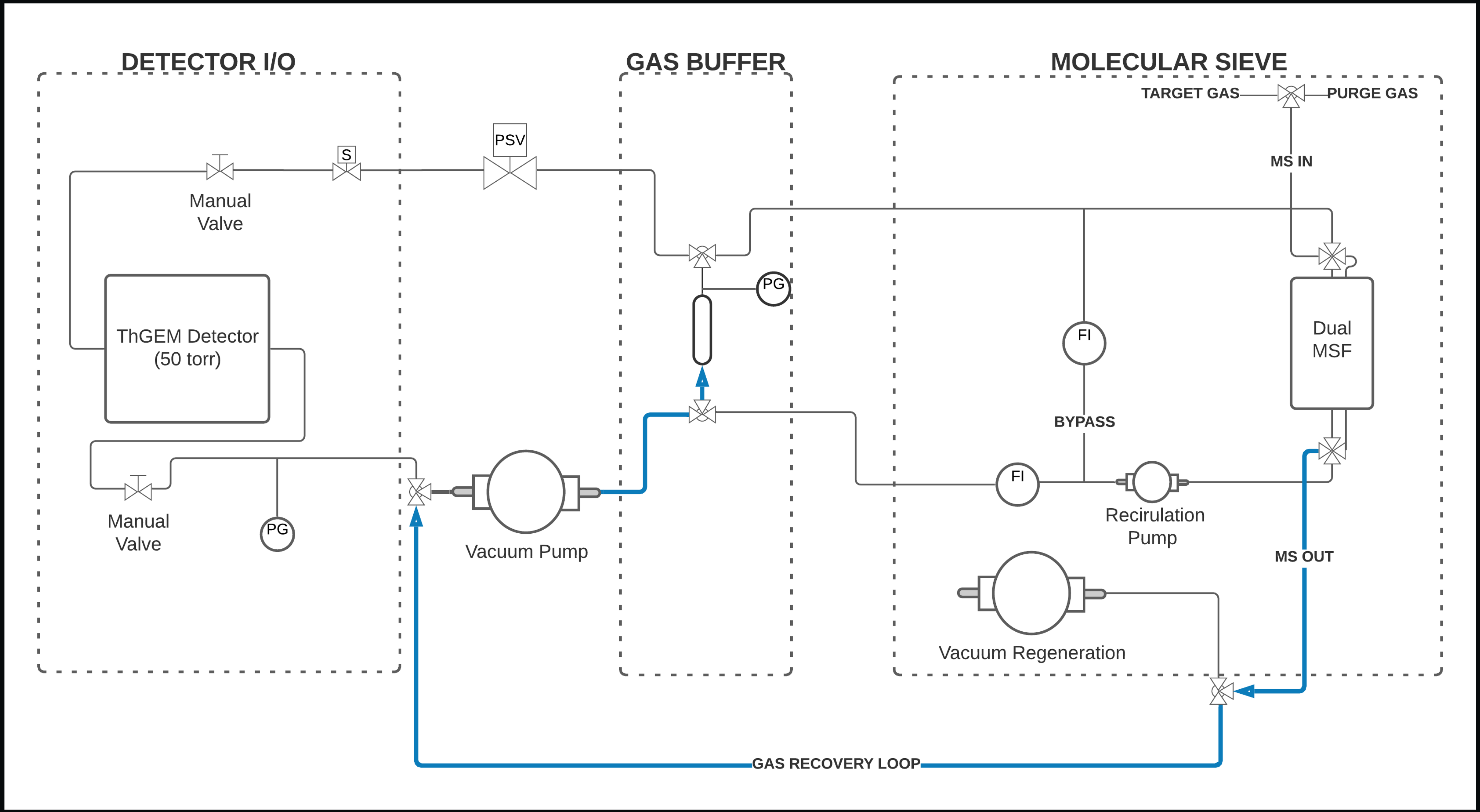
Dual MS design utilises **Vacuum Swing Adsorption (VSA)** Technique



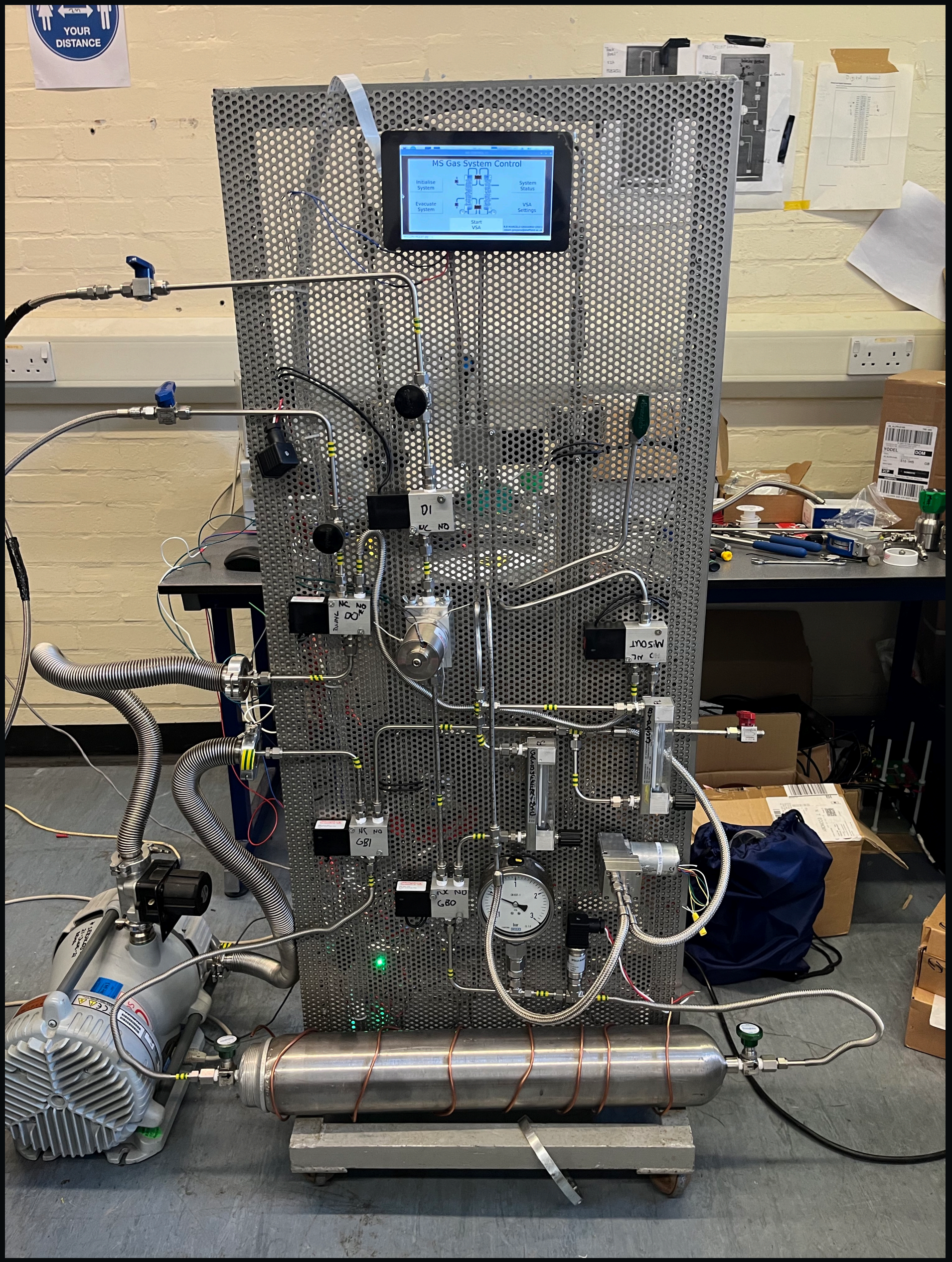
Notice small amount of our desired gas is lost during vacuum regeneration

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



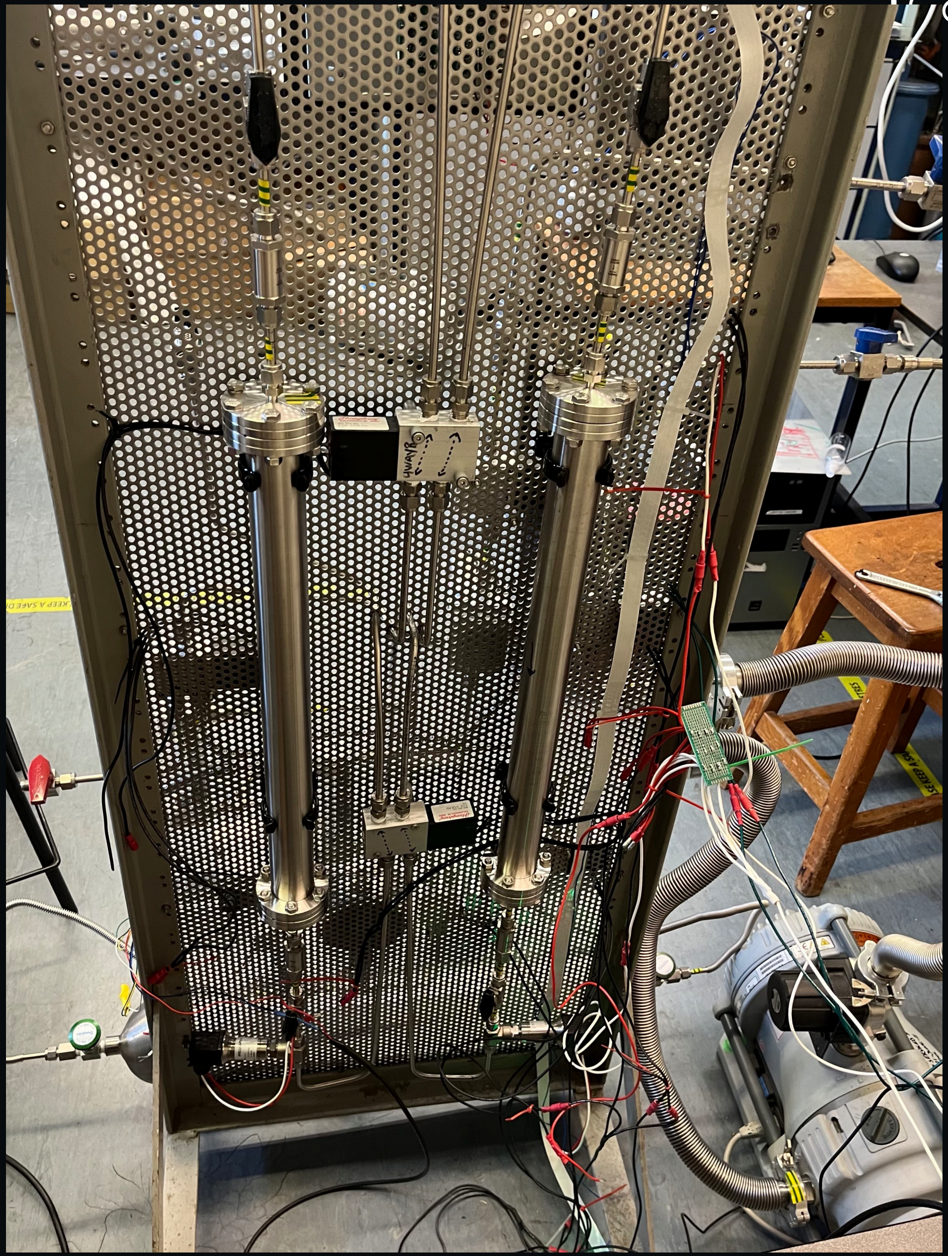
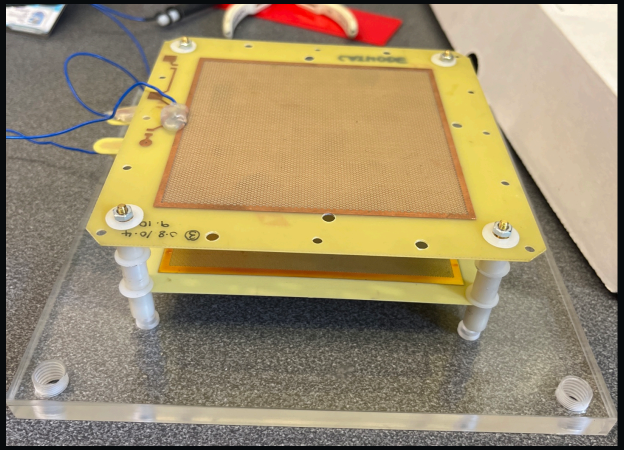


Photo of status
December 2021

Performance Testing

ThGEM (10 x 10 cm)

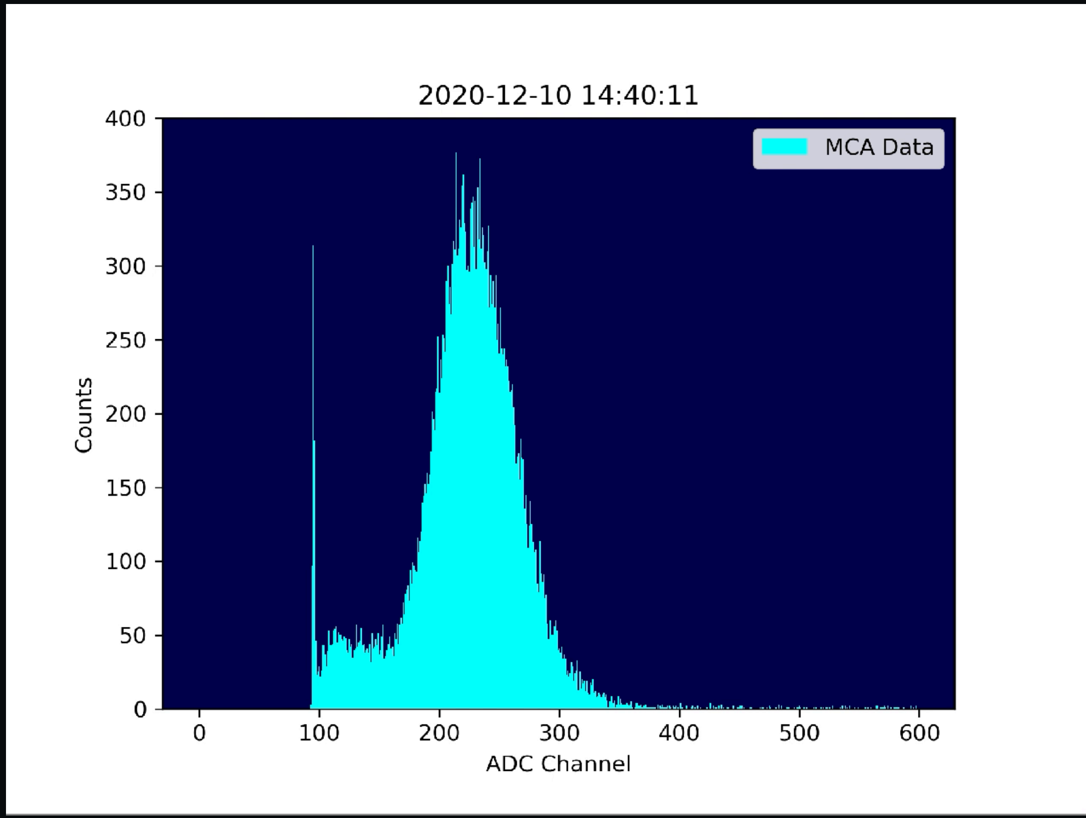


100L Vessel

Performance Testing

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

1. Gas gain detector signal



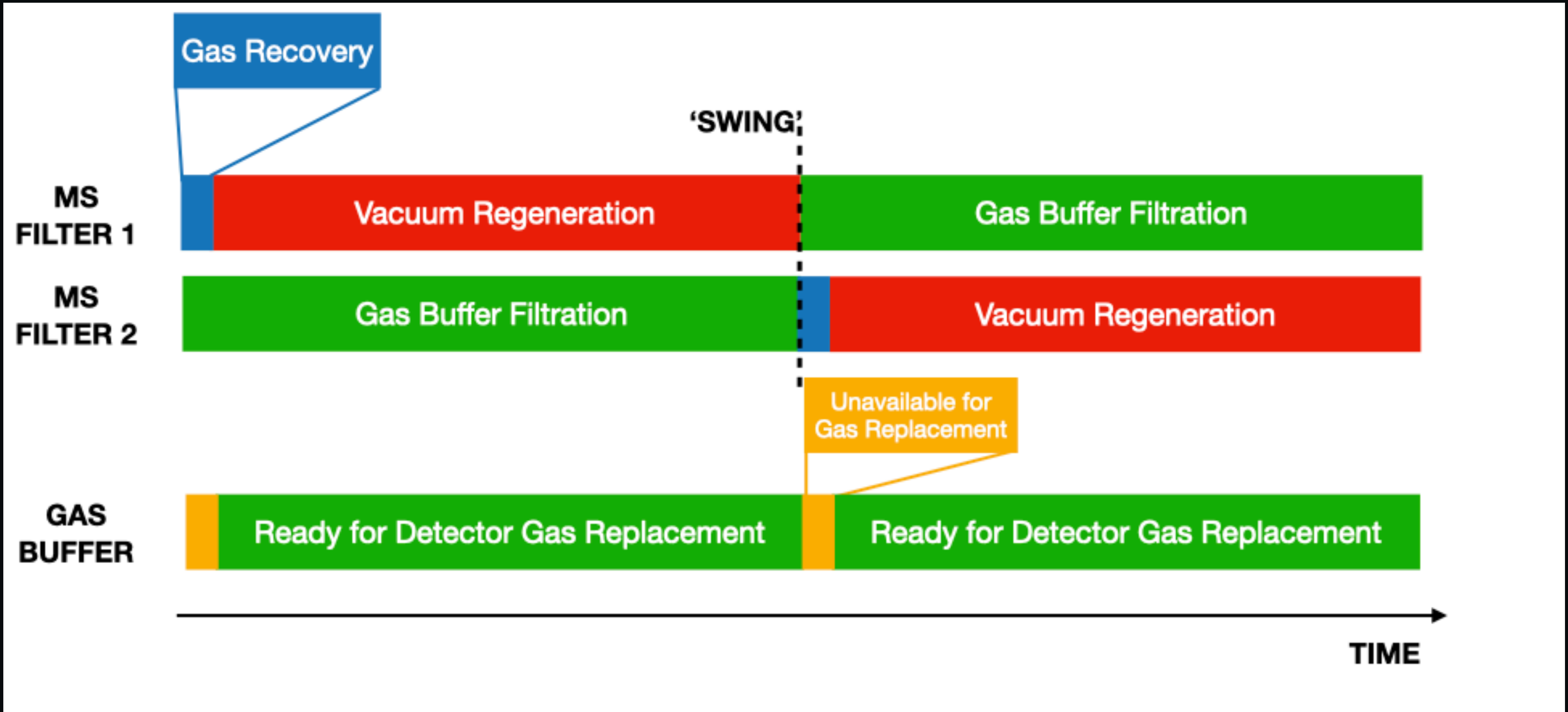
Example of signal deterioration due to gas contamination in a ThGEM TPC CF_4 (Fe-55 calibration source)

2. Experiment's Intrinsic radon background

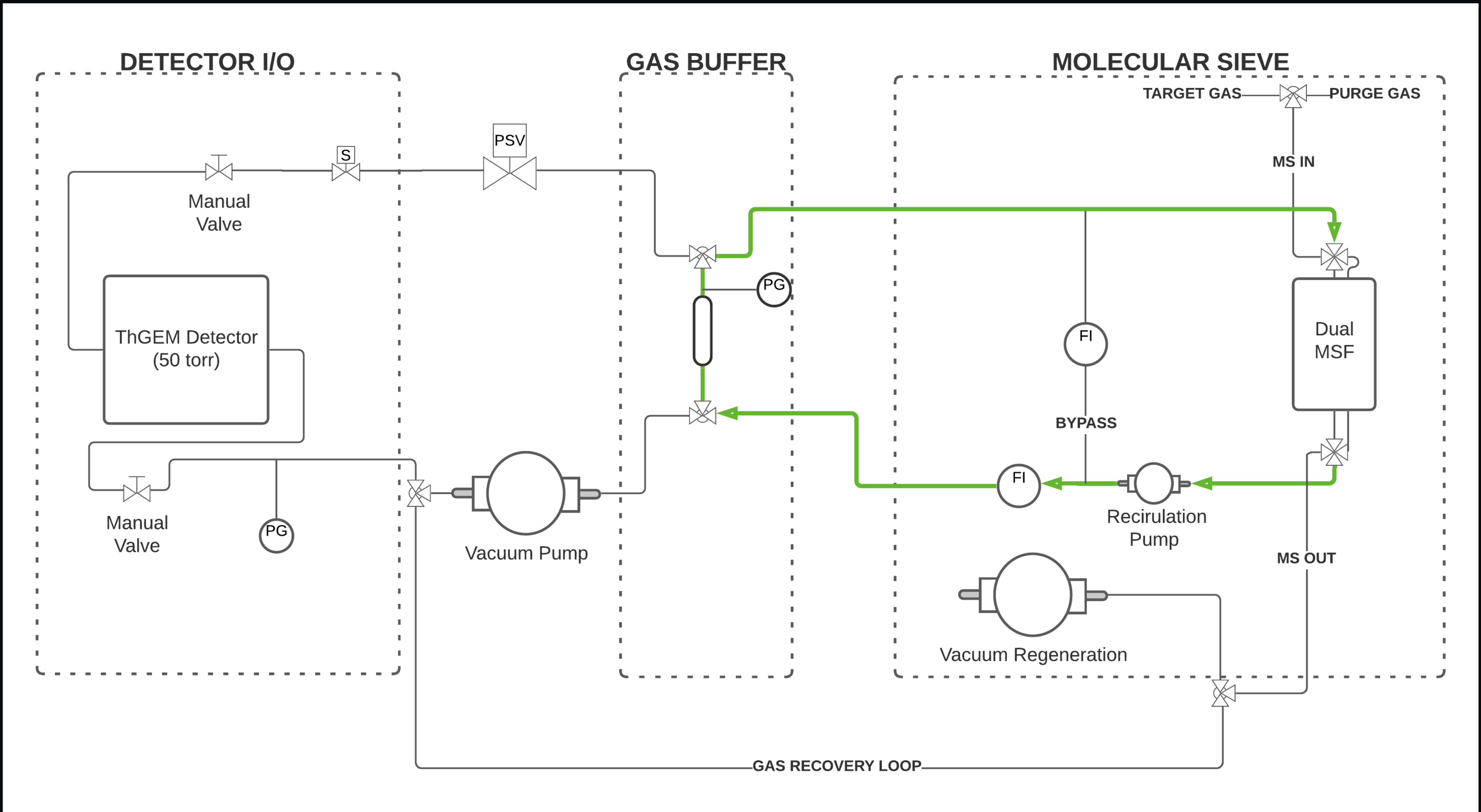
Conclusions

- 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

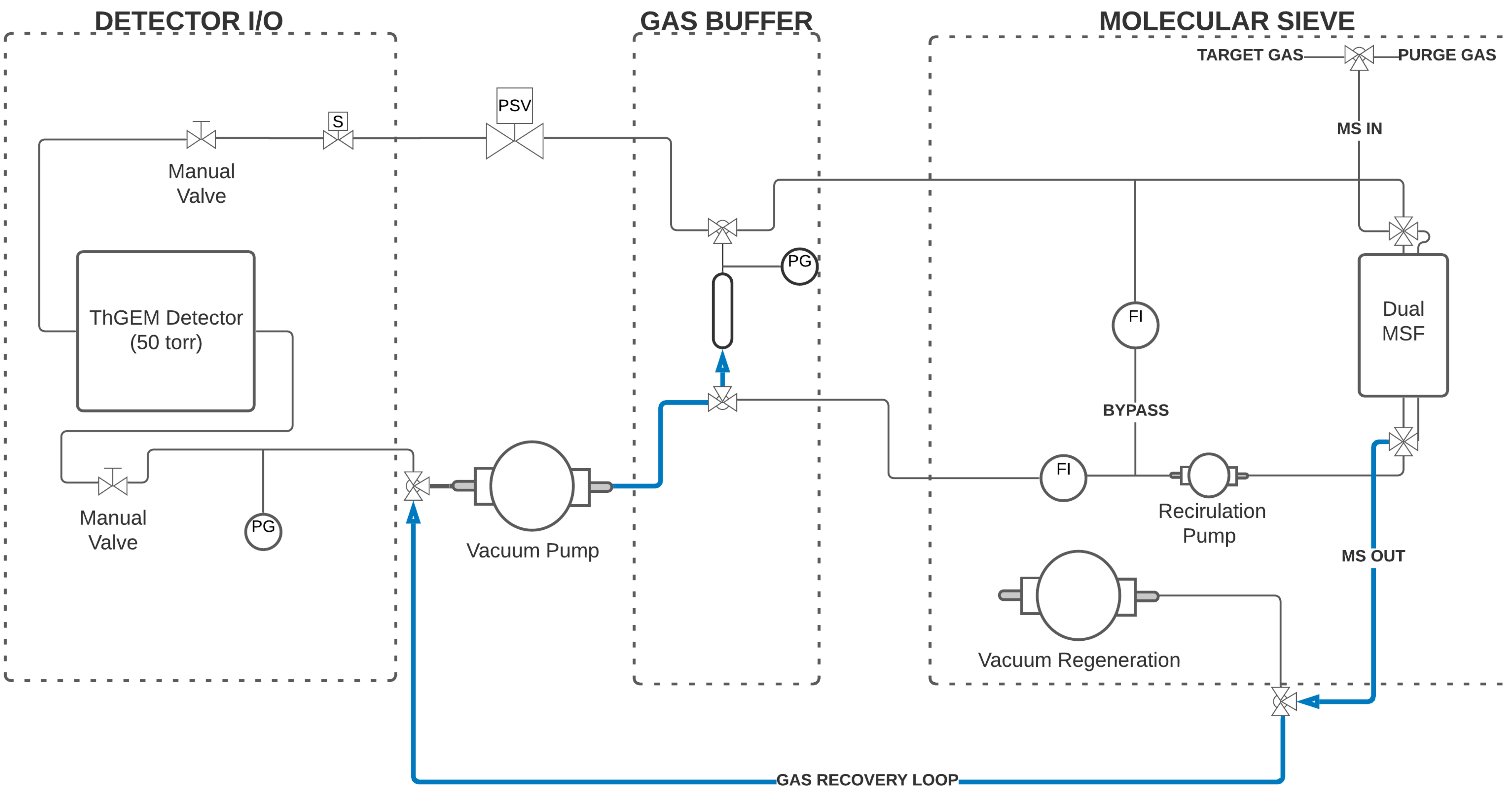
VSA Operation



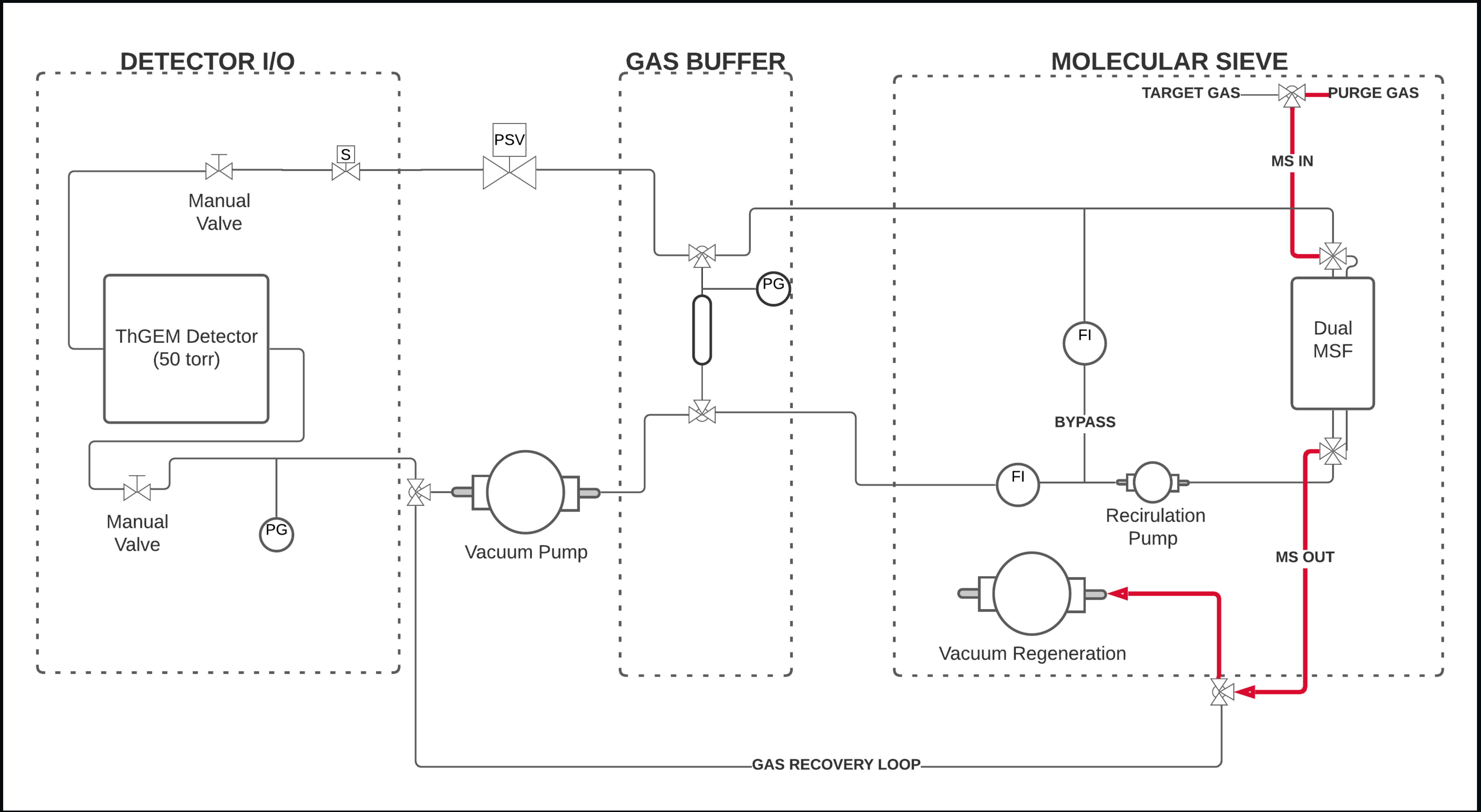
VSA: Gas Filtration



VSA: Gas Recovery Loop



VSA: Vacuum Generation



VSA: Gas Replacement