



EP-DT  
Detector Technologies

# **Eco-gas mixtures and mitigation procedures for GreenHouse Gases (GHGs)**

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Beatrice Mandelli

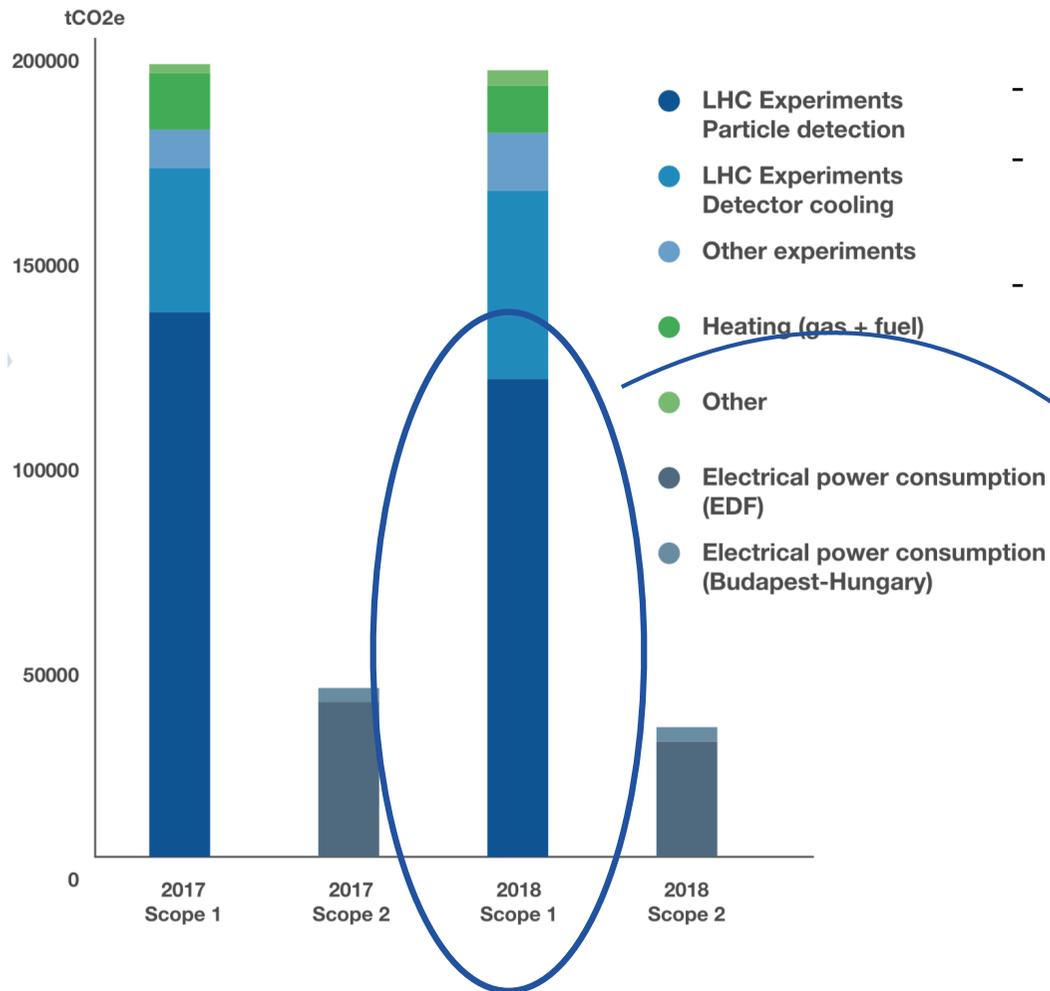
**CERN**

ECFA Detector R&D Roadmap Symposium of Task Force 1 Gaseous Detectors  
CERN, 29 April 2021

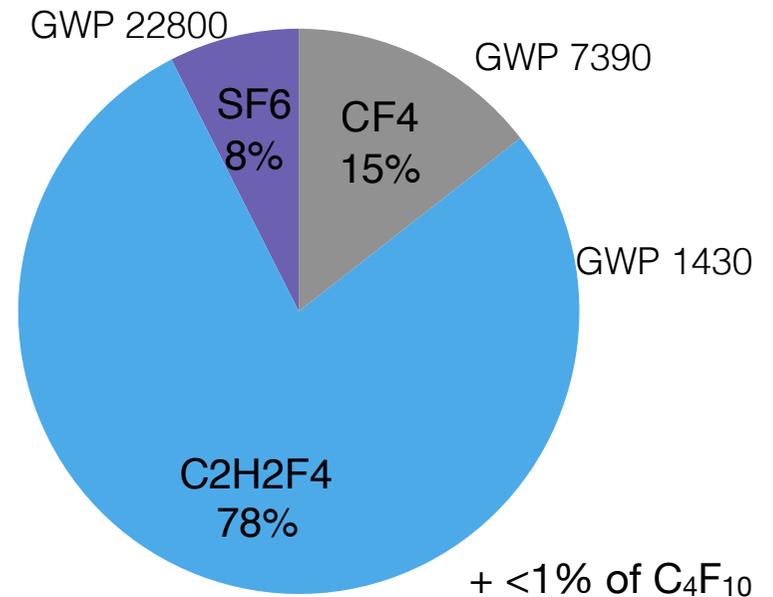
# CERN commitment to reduce GHG emissions

*Greenhouse gas emissions at CERN arise from the operation of the Laboratory's research facilities.*

*With climate change a growing concern, the Organization is committed to reducing its direct greenhouse gas emissions.*



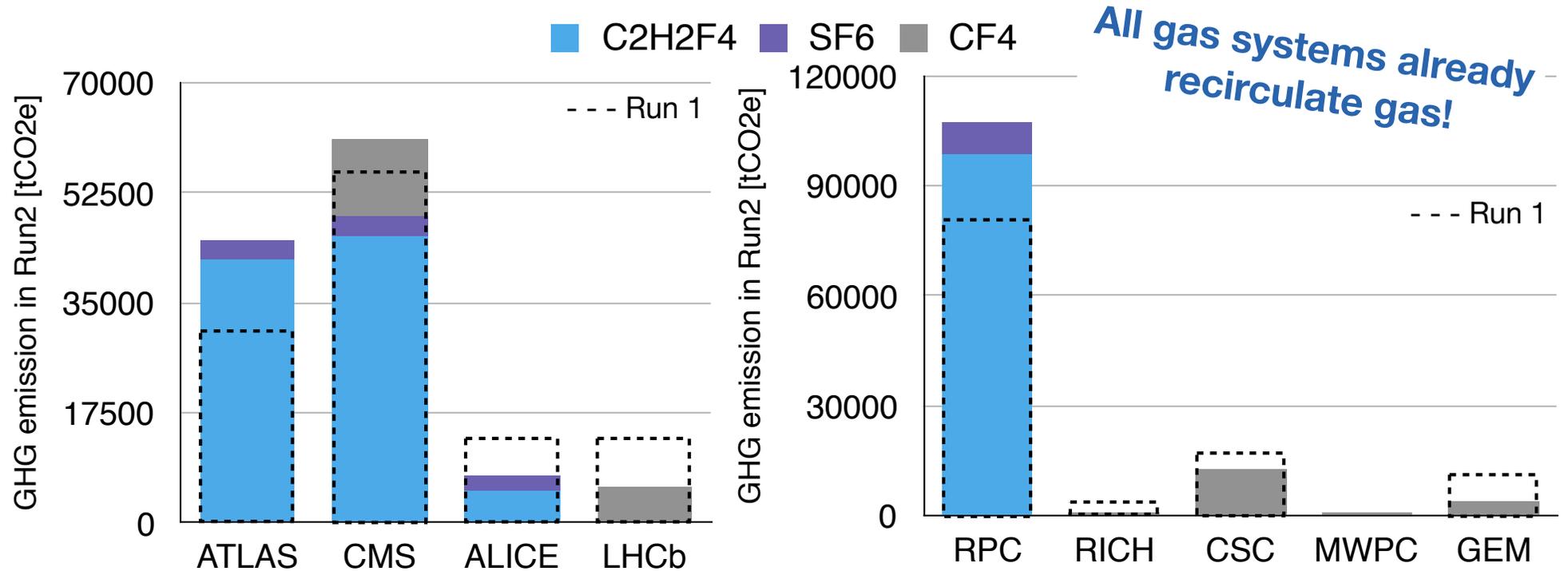
- 192.000 tCO<sub>2e</sub> in 2018
- 92% of emissions related to large LHC experiments
- Most emissions from particle detection



[https://e-publishing.cern.ch/index.php/CERN\\_Environment\\_Report/index](https://e-publishing.cern.ch/index.php/CERN_Environment_Report/index)

# GHGs for particle detection at LHC: Run 2

*GHGs are used in CERN experiments  
mainly due to their properties necessary for good detector operation*



*All gas systems already recirculate gas!*

**Leaks in detectors**  
(ATLAS and CMS)

**Permeation to Air**  
(CMS)

**Upgrade to gas recirculation**  
(LHCb)

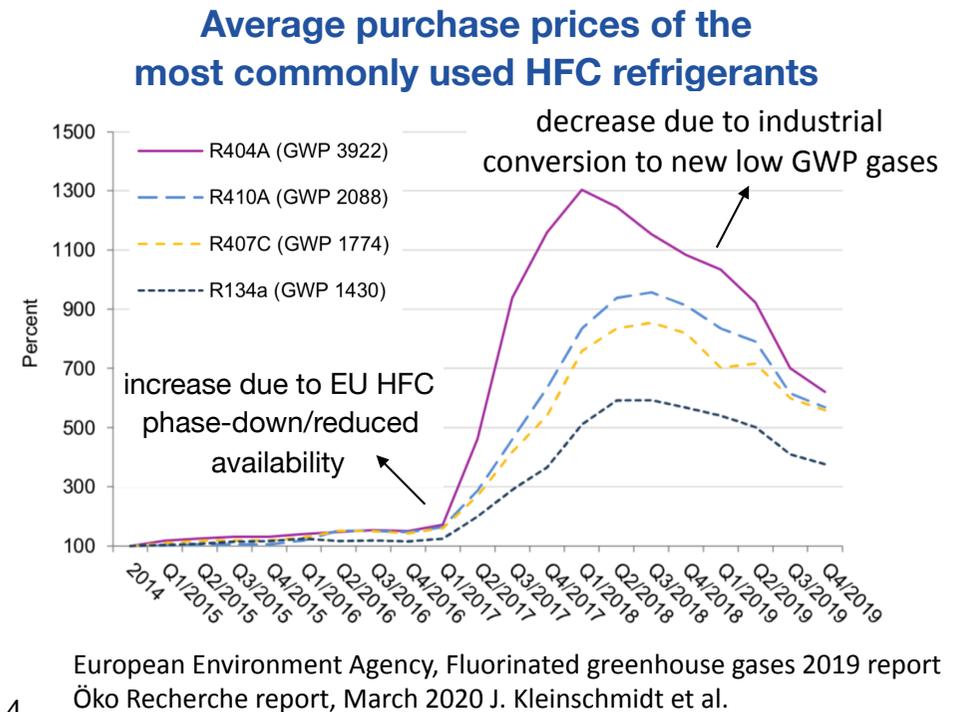
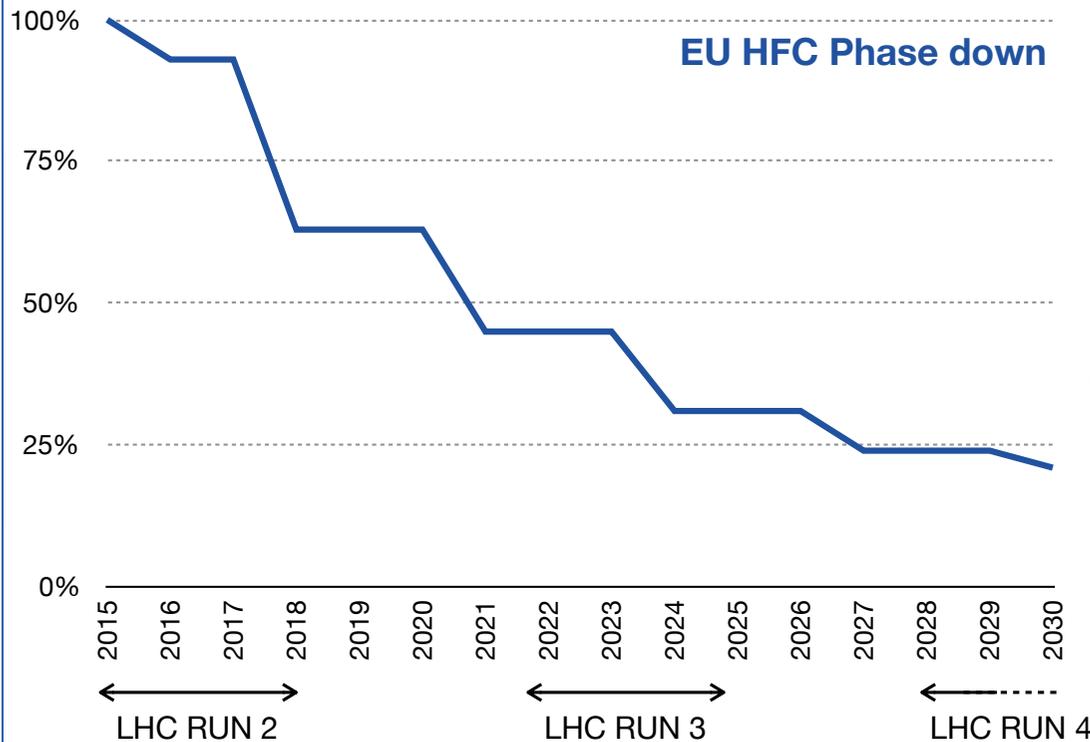
Leaks are concentrated in the gas inlets, polycarbonate gas connectors and Polyethylene pipes

Big leak search campaign on-going in LS2: fundamental to have access to chambers for repairing

# EU HFC phase-down policy

## European Union “F-gas regulation”:

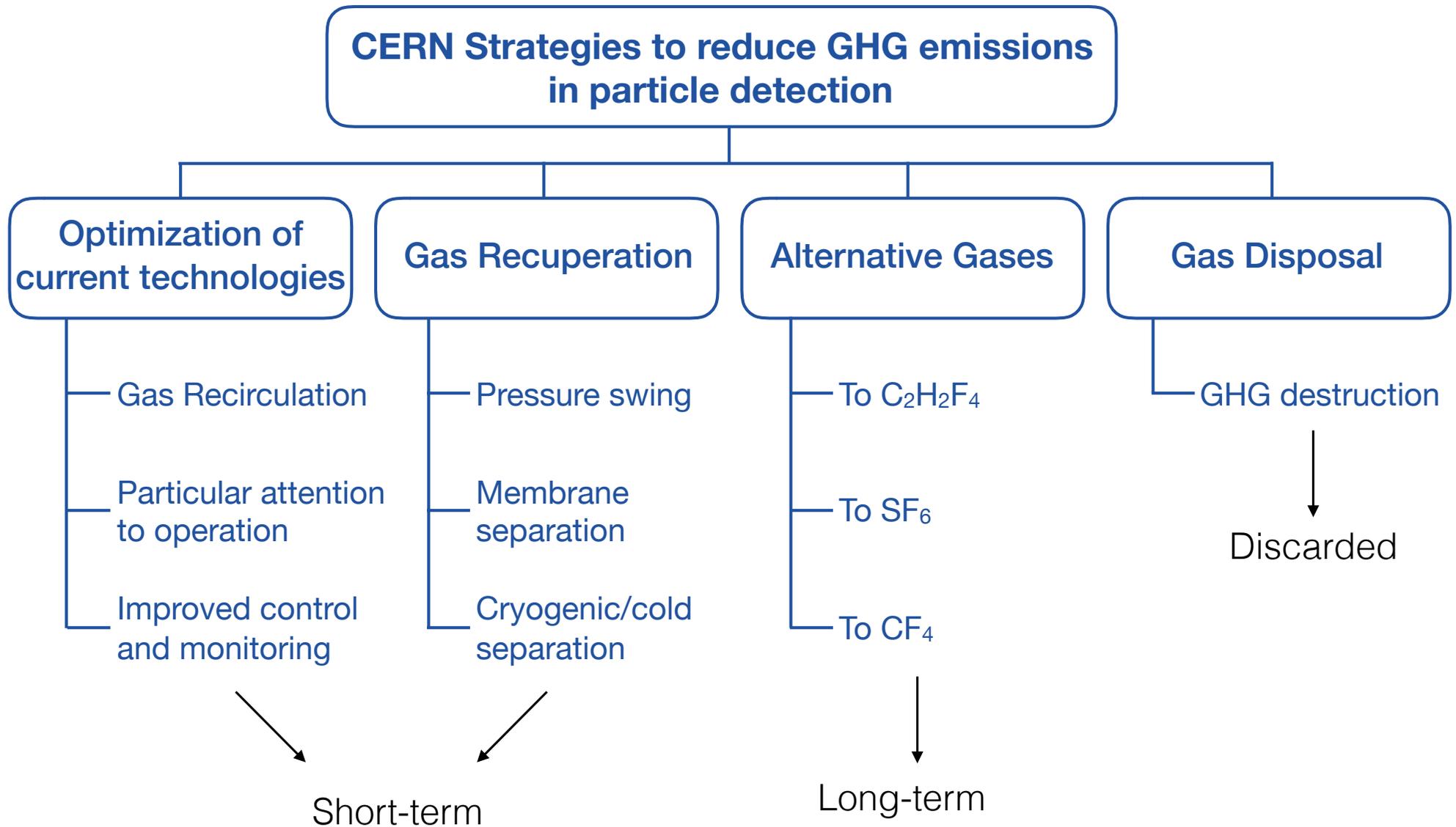
- **Limiting the total amount** of the most important F-gases that can be sold in the EU from 2015 onwards and phasing them down in steps to one-fifth of 2014 sales in 2030.
- **Banning the use** of F-gases in many new types of equipment where less harmful alternatives are widely available.
- **Preventing emissions** of F-gases from existing equipment by requiring checks, proper servicing and recovery of the gases at the end of the equipment's life.



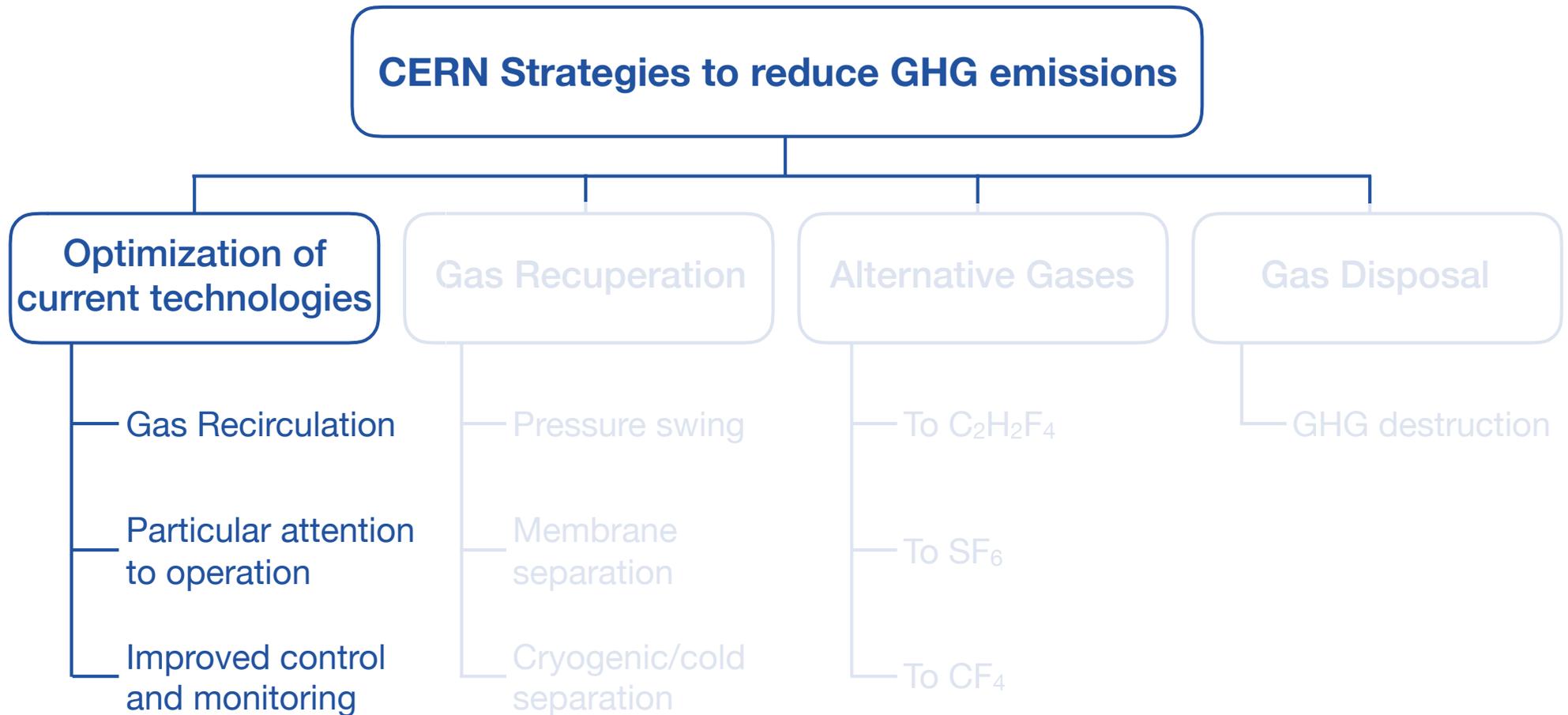
European Environment Agency, Fluorinated greenhouse gases 2019 report  
 Öko Recherche report, March 2020 J. Kleinschmidt et al.

**Prices could increase in EU and availability in the future is not known.  
 Reduction of the use of F-gases is fundamental for future particle detector applications**

# CERN strategies for GHG reduction



# CERN strategies for GHG reduction



# Gas recirculation

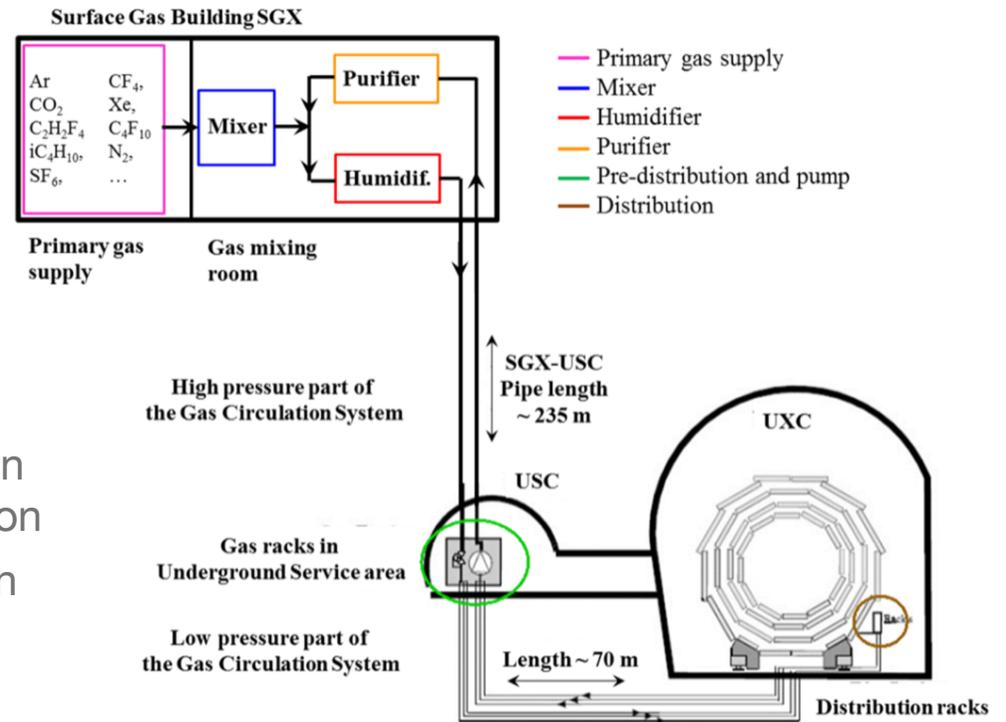
*Thanks to gas recirculation GHG emission already reduced by > 90% wrt to open mode systems!!!*

## Advantages

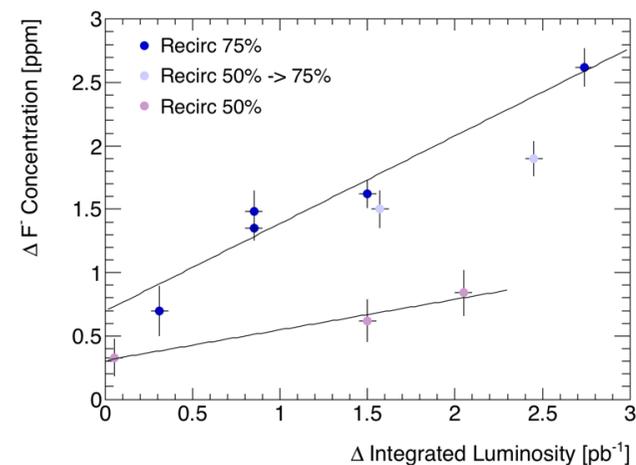
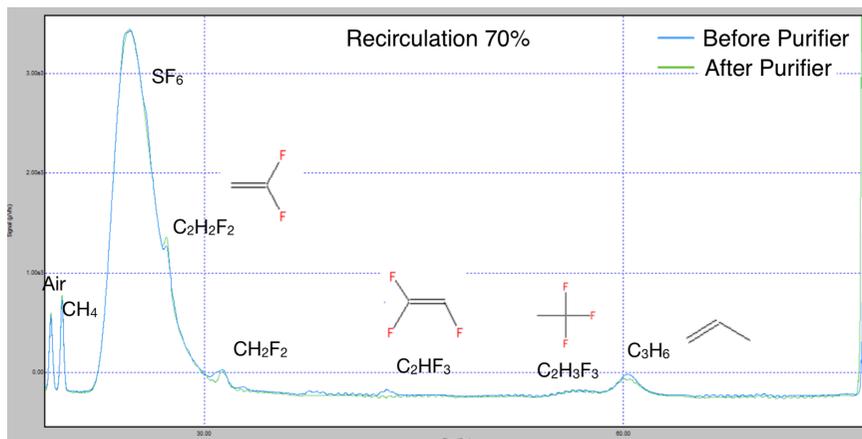
- Reduction of gas consumption

## Disadvantages

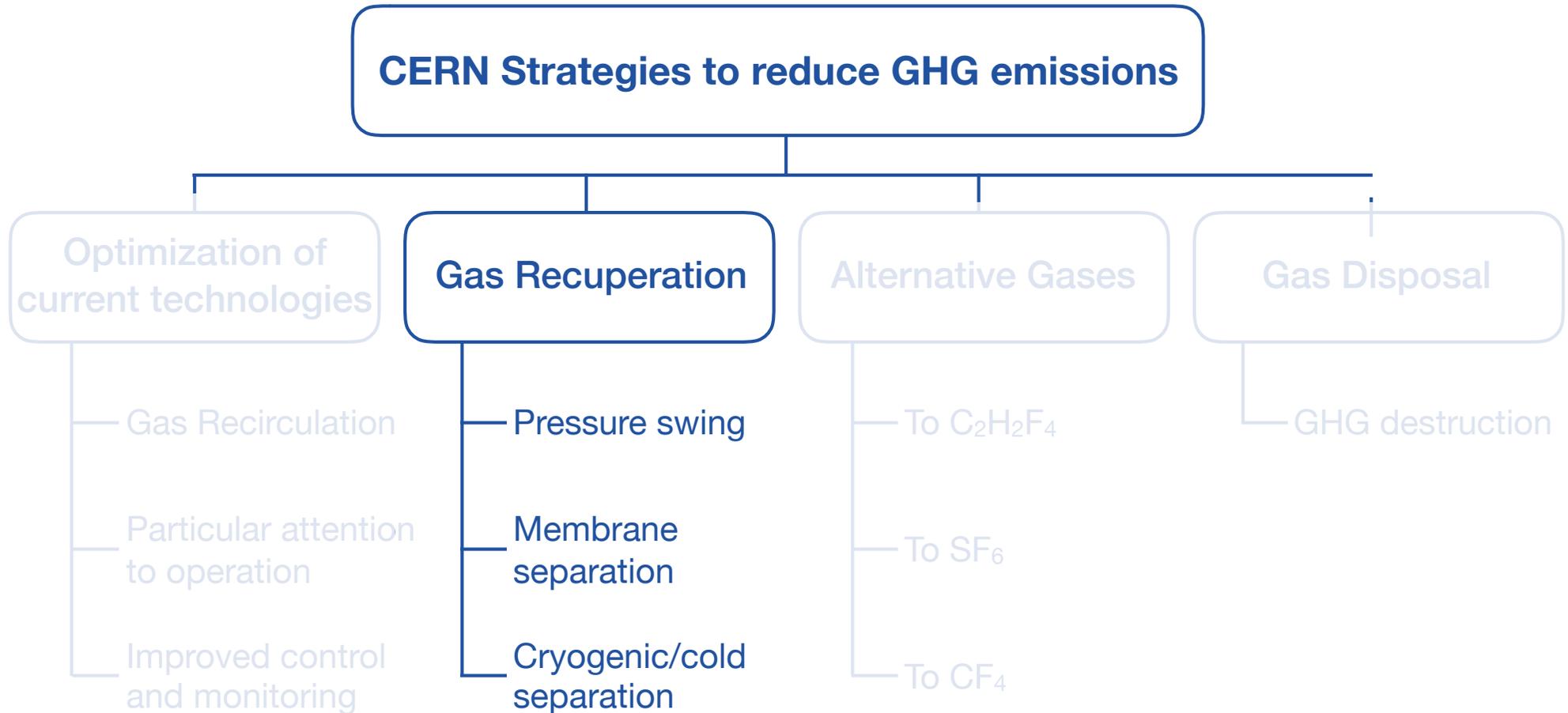
- Complexity in operation
  - Pressure and flow fluctuations, etc.
- Creation of impurities
  - Especially when F-gases present
  - Accumulation in the gas mixture, concentration depends on luminosity and recirculation fraction
  - They could affect long-term detector operation
- Gas purifying techniques
  - Needed to absorb impurities



## Example of studies performed for ALICE MTR in LHC Run 2

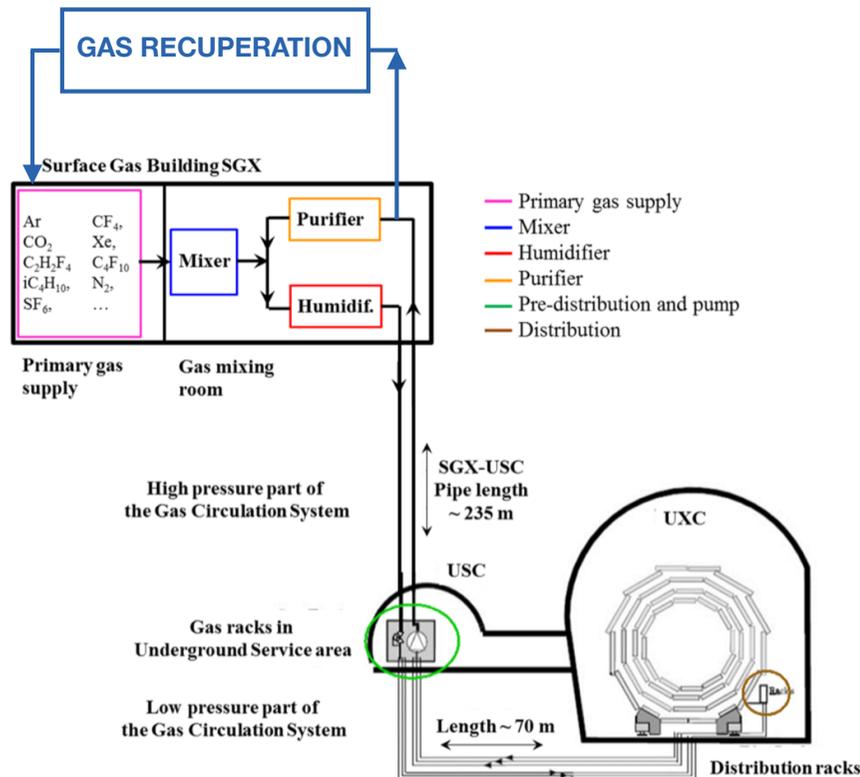


# CERN strategies for GHG reduction



# Gas Recuperation systems at LHC experiments

- Sometimes it is not possible to recirculate 100% of the gas mixture and a fraction cannot be re-used and therefore it would have been sent to atmosphere
  - Detector permeability, detector requirements (max recirculation fraction tested), impurities, etc.
  - To keep lower  $N_2$  concentration
- This fraction of gas mixture is sent to a recuperation plant where the most valuable component is extracted, stored and re-used
  - Often challenging to extract a single component
  - The quality of recuperated gas is fundamental



## Many LHC gas systems with gas recuperation

### Advantages:

- further reduction of gas consumption

### Disadvantages:

- higher level of complexity
- dedicated R&D
- gas mixture monitoring

# Gas recuperation: CMS CSC CF<sub>4</sub>

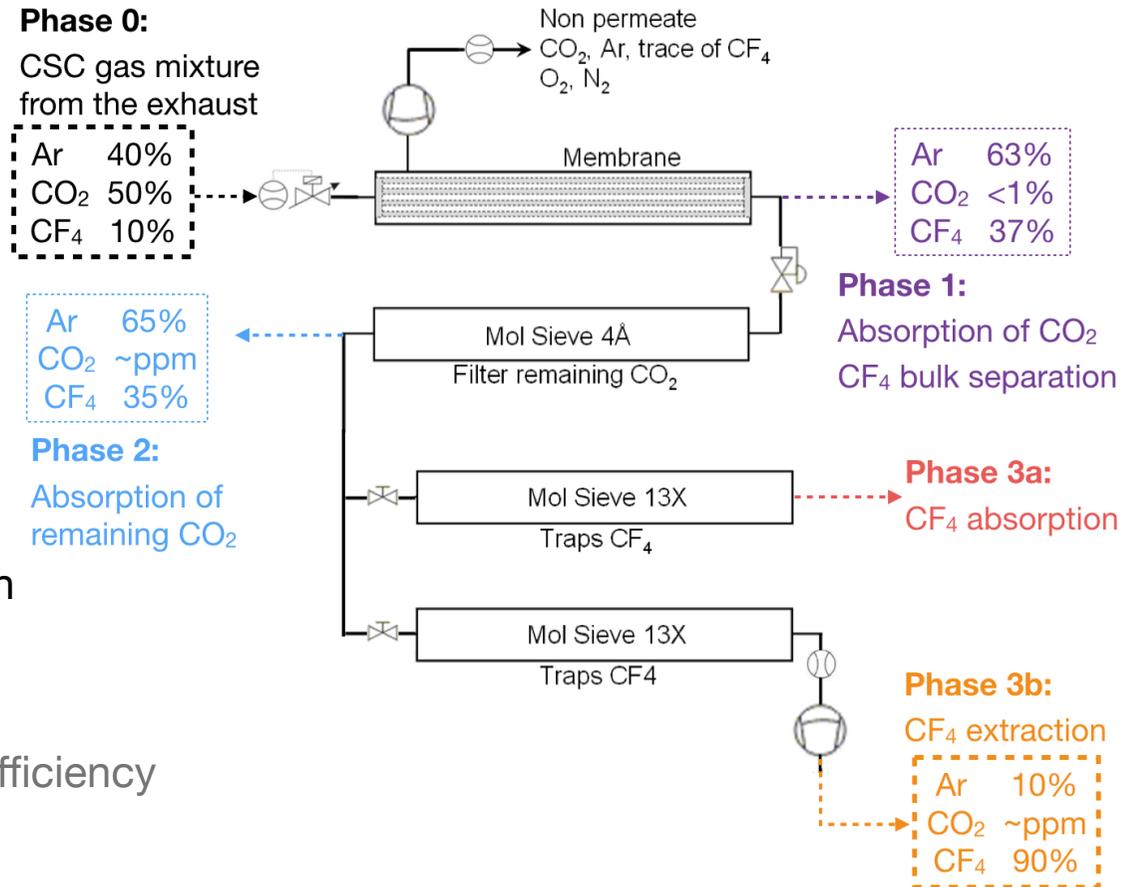
GHG reduction from Run1 to Run2 up to **45%**

## CSC Gas System

- Detector volume ~90 m<sup>3</sup>
- Gas mixture: 50% CO<sub>2</sub>, 40% Ar, **10% CF<sub>4</sub>**
- Gas recirculation: 90%
- No possible to increase due to detector permeability to Air
- ~600 l/h at exhaust -> 60 l/h of CF<sub>4</sub>

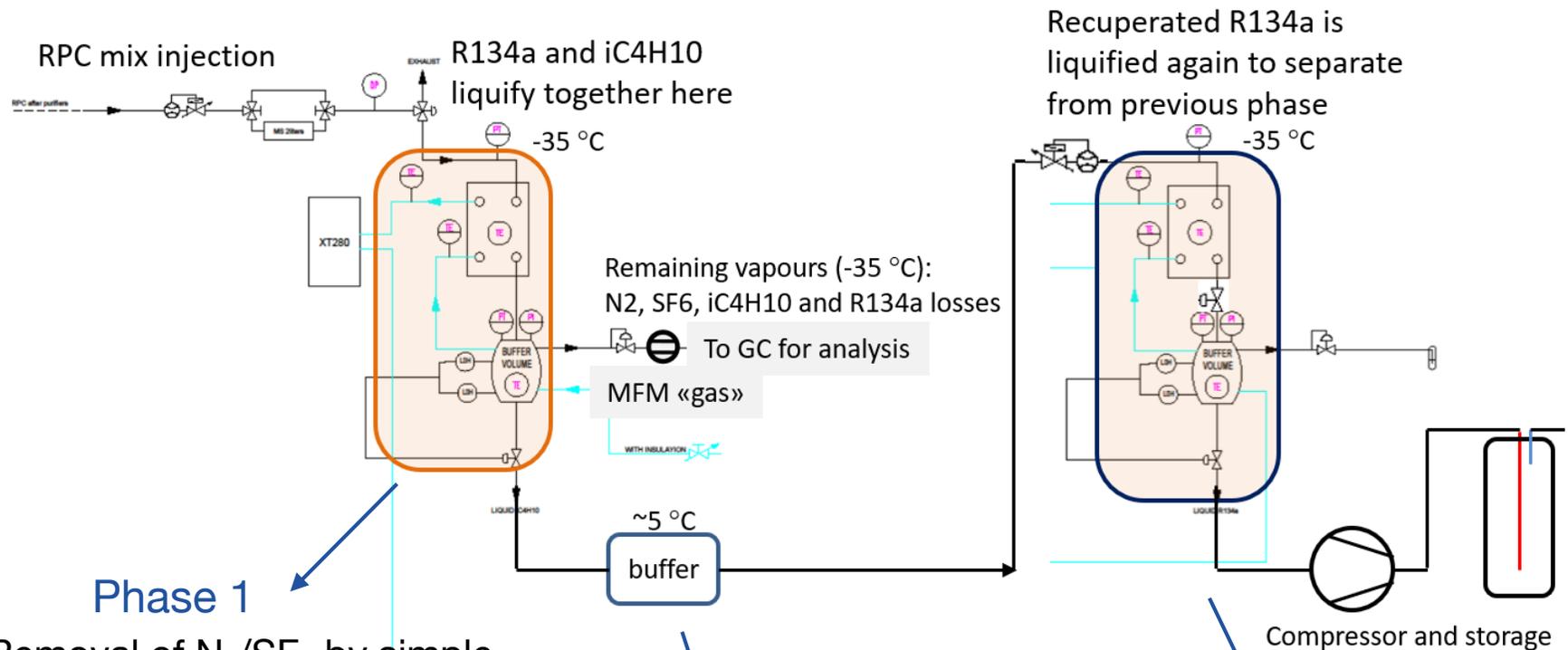
## CSC Recuperation System

- Recuperation of CF<sub>4</sub> with warm separation
- 3 phases needed
- Current recuperation efficiency ~65%
- Several parameters affect recuperation efficiency
- CF<sub>4</sub> quality satisfactory
- Recuperated CF<sub>4</sub> quality to monitor
- CSC detectors operated with recuperated CF<sub>4</sub> during Run 2
- No change in the CSC performance observed



# Gas Recuperation: C<sub>2</sub>H<sub>2</sub>F<sub>4</sub> for RPC detectors

New C<sub>2</sub>H<sub>2</sub>F<sub>4</sub> recuperation prototype under study/test since 2019



Phase 1

- Removal of N<sub>2</sub>/SF<sub>6</sub> by simple distillation
- Gas mixture in buffer 1 cools down at -35 °C
- N<sub>2</sub>/SF<sub>6</sub> in vapour phase

Phase 2

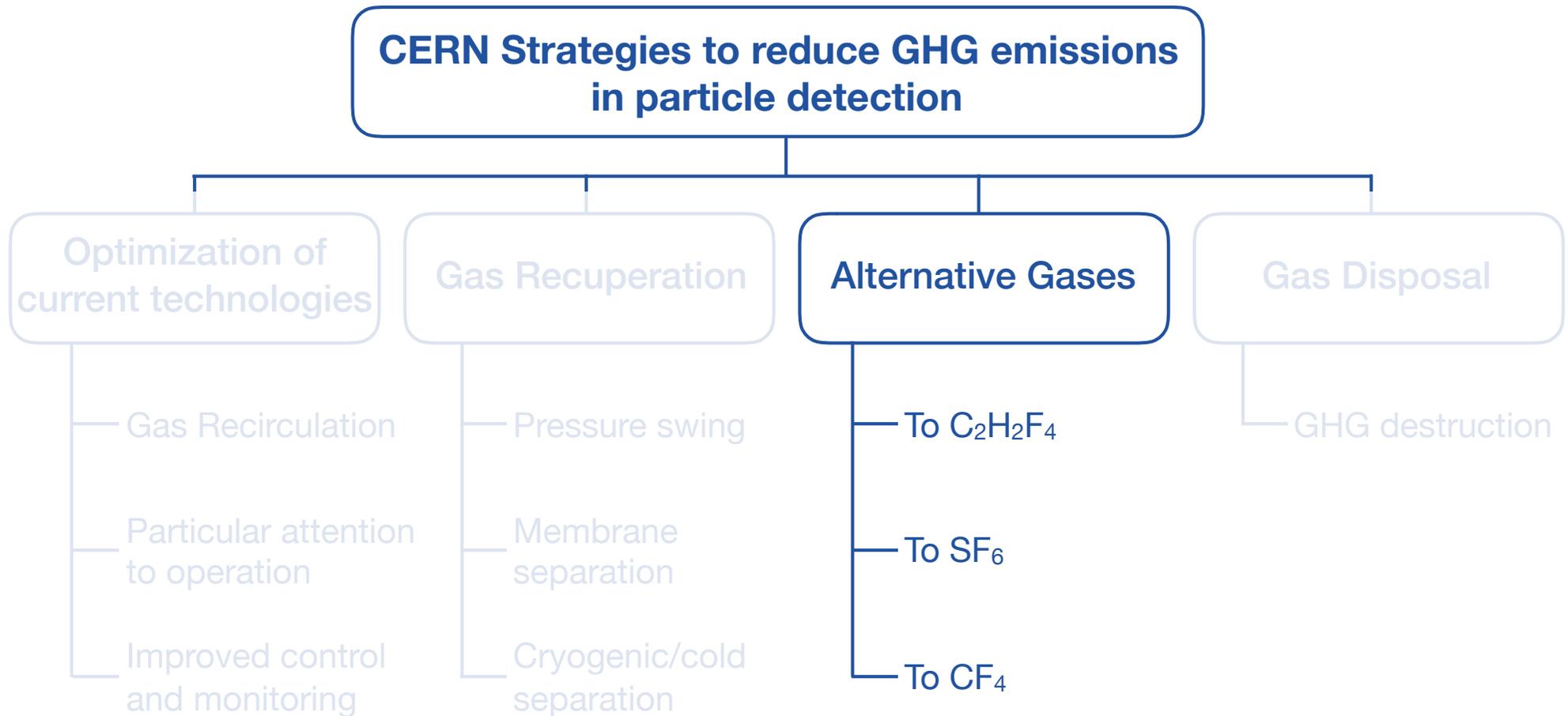
- Detachment of R134a from iC<sub>4</sub>H<sub>10</sub>
- Liquid heats up and vapour is made of azeotrope
- Vapours go back in buffer 1
- Liquid R134a go in buffer 2

Phase 3

- Compression of R134a
- Vapour is compressed in liquid storage

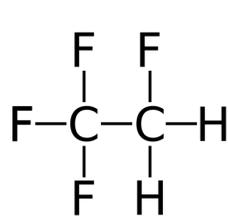
Recuperation efficiency ~80%

# CERN strategies for GHG reduction

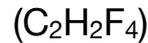


# Possible alternatives to GHG gases

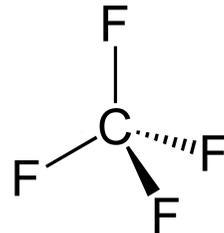
*New eco-friendly liquids/gases have been developed for industry as refrigerants and HV insulating medium...  
ionisation properties in particle detection not well known*



R134a



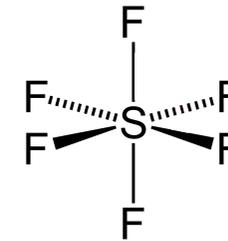
**GWP 1430**



R14



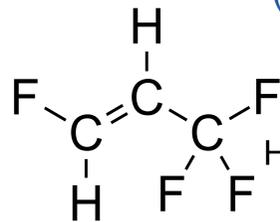
**GWP 5700**



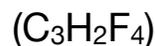
SF<sub>6</sub>

**GWP 23900**

Not in industry  
(tried by CSC)



HFO-1234ze



**GWP 6**

Hydro-Fluoro-Olefin (HFO)

C=C double bond

fluorine-containing

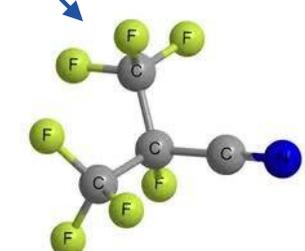
hydrogen-containing



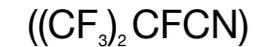
3M™ Novec™ 5110



**GWP <1**



3M™ Novec™ 4710



**GWP 2100**

Mini-workshop on gas transport parameters for present and future generation of experiments

<https://indico.cern.ch/event/1022051/overview>

# HFO-based gas mixtures

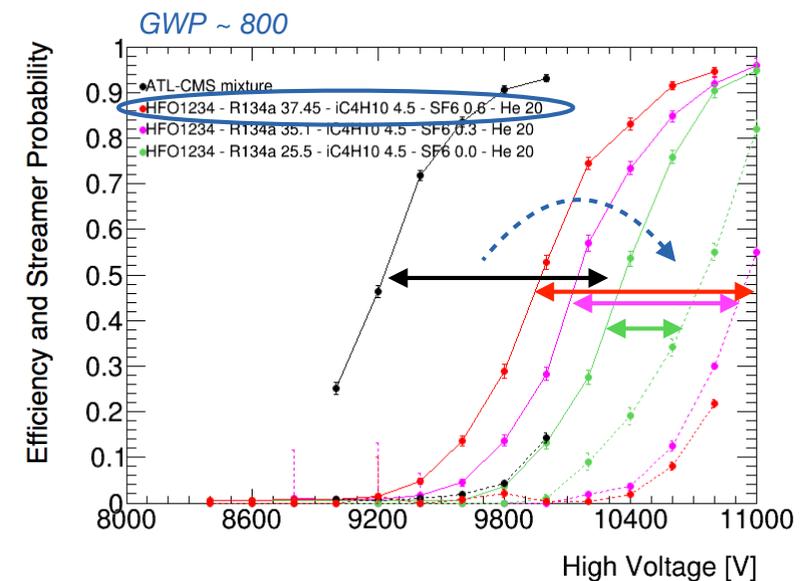
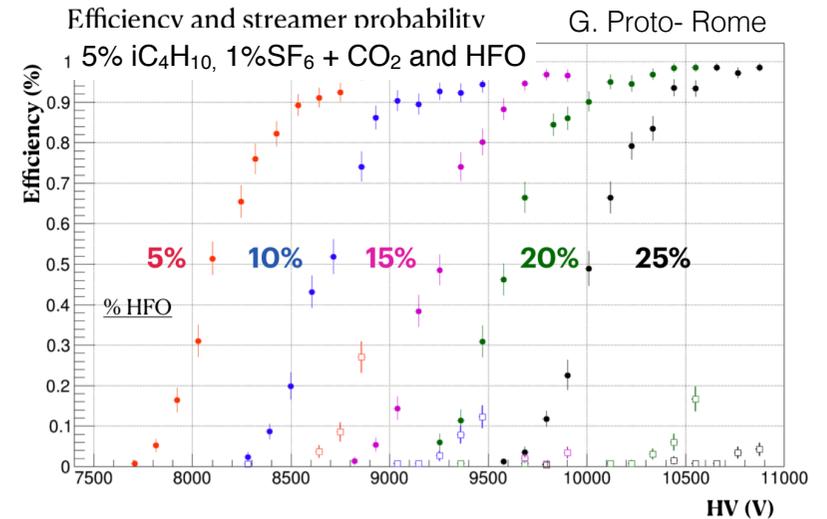
*In these slides: search for an eco-friendly gas mixture that is compatible with the current ATLAS and CMS RPC systems (i.e. no change in HV cables, FEB electronics, gas system, etc.)*

HFO cannot directly replace  $C_2H_2F_4$

- Higher applied voltage necessary (>12kV)
  - One C more with a double bond
- Small avalanche signal

Addition of He or  $CO_2$  to lower the HV working point

- Helium helps in reducing the HV working point
  - +10% He  $\rightarrow$  1 kV
  - in first approximation it doesn't take part in the avalanche processes
- $CO_2$  helps in reducing the HV working point
  - + 10%  $CO_2 \rightarrow$  0.8 kV
  - $CO_2$  is used as quencher gas in gaseous detectors but it has different quenching properties wrt  $iC_4H_{10}$  (different absorption coefficient)
- With both He and  $CO_2$  in HFO-based gas mixture: higher streamer probability
  - Also higher charges
  - Necessary to increase  $SF_6$  concentration
  - Keep a small fraction of  $C_2H_2F_4$  helps in reducing the signal charge



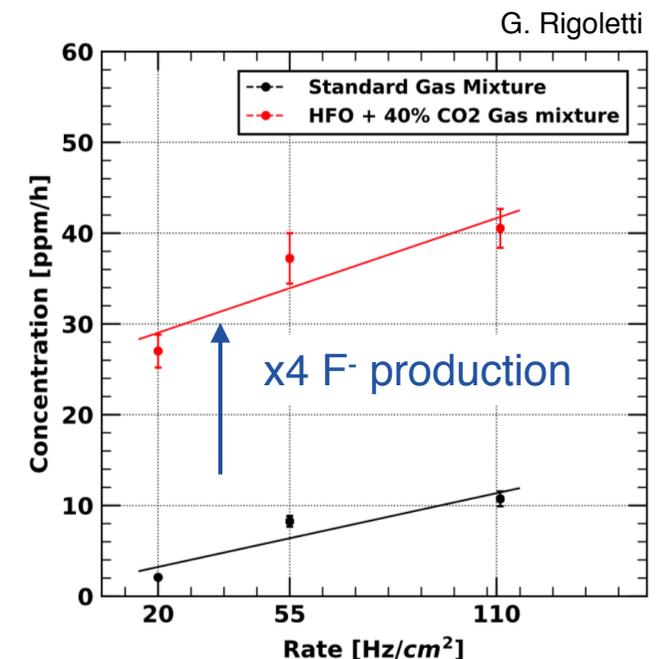
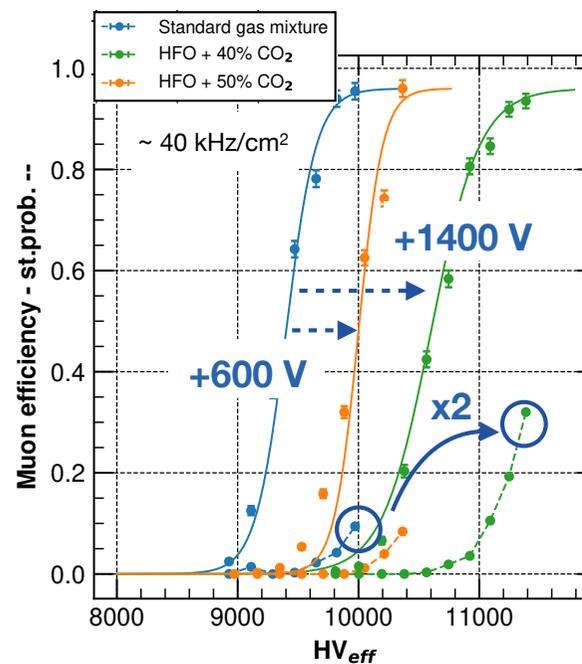
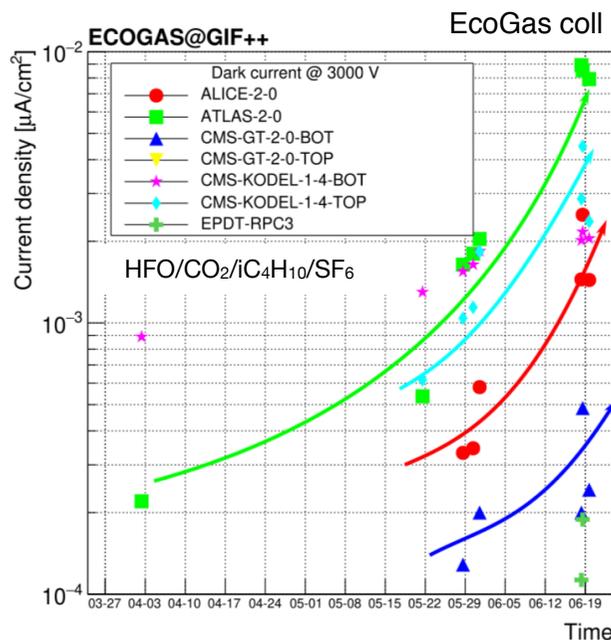
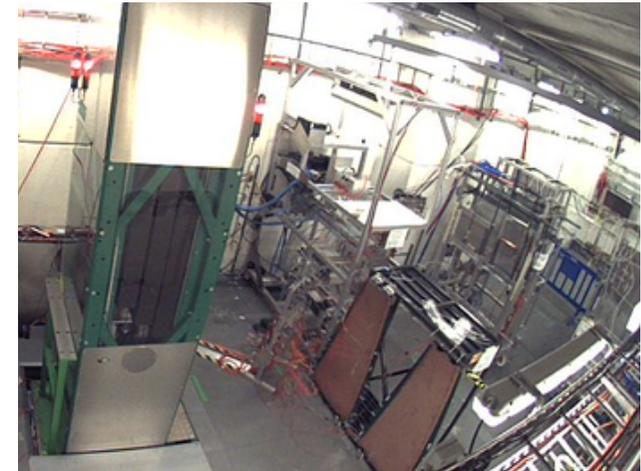
# Long-term studies with HFO gas mixtures

*Performance studies of several eco-friendly gas mixtures for RPCs operated at different background conditions*



EcoGas collaboration

- GIF++: 12.2 TBq  $^{137}\text{Cs}$  + H4 SPS beam line
- Long-term studies (aging-test)
  - Fundamental for the validation of new eco-friendly gas mixtures. Accumulation of high integrated charge
- Studies on detector performance
  - In presence of LHC and HL-LHC like background radiation
- Studies on creation of impurities
  - HFO breaks easier than  $\text{C}_2\text{H}_2\text{F}_4$  during detector operation



# Possible SF<sub>6</sub> replacements

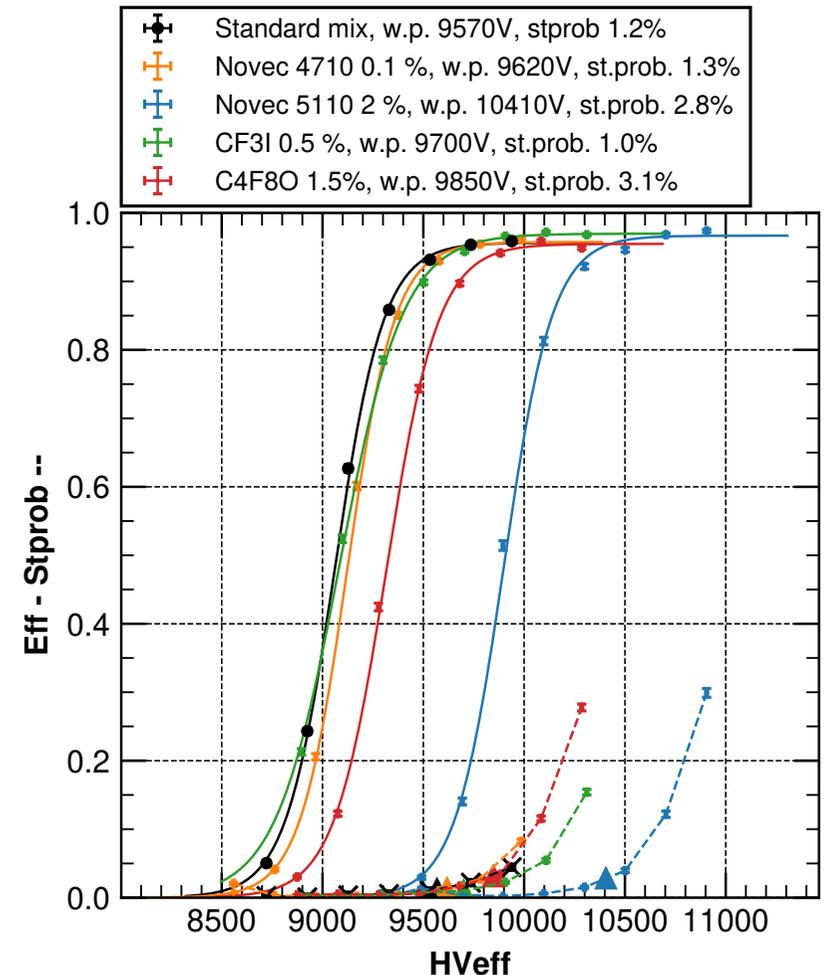
SF<sub>6</sub> has a very high GWP and it contributes for ~5% in the GWP of RPC gas mixture

## 3M™ Novec™ Dielectric fluids

- Very good alternative to SF<sub>6</sub> for arc quenching and insulation applications
  - Developed few years ago
  - Dielectric breakdown strength approximately 1.4-2 times that of SF<sub>6</sub>
  - Especially used in HV industrial plants
- Novec 4710 (GWP 2100)
  - Very good performance but...
  - It may react with water
- Novec 5110 (GWP <1)
  - Very low GWP but..
  - RPC performance not optimal
  - sensitive to UV radiation

## Other alternatives

- Looks for other gases not used only for HV plants
  - Other electronegative gases could work
- CF<sub>3</sub>I (GWP 0.4)
  - Good performance but...
  - Toxic, mutagenic, ODP 0.008
- C<sub>4</sub>F<sub>8</sub>O (GWP ~8000)
  - Good performance at 1.5%
  - 1.5% C<sub>4</sub>F<sub>8</sub>O GWP equivalent to 0.5% SF<sub>6</sub>



# Possible CF<sub>4</sub> replacements

CF<sub>4</sub> is used in different types of particle detectors to prevent aging, to enhance time resolution or because of its scintillation photon emission

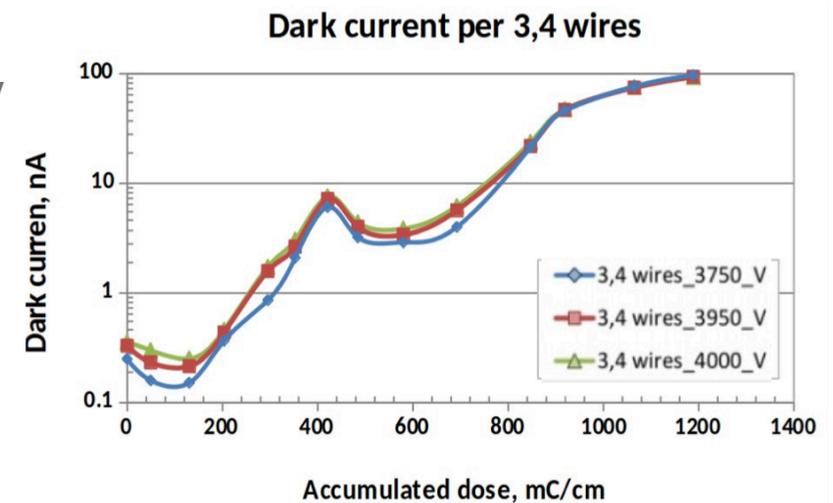
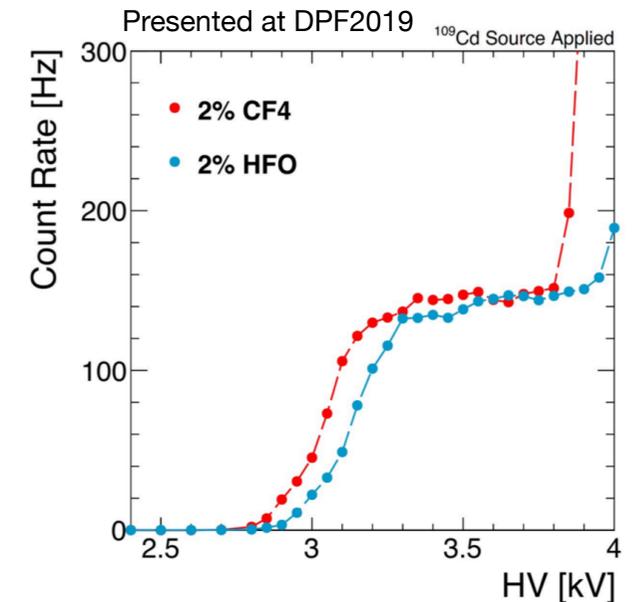
## CMS CSC studies

- CF<sub>4</sub> is a source of fluorine radicals to protect against anode ageing
  - Now 10% CF<sub>4</sub> in CSC gas mixture
- Two possible approaches to reduce GHG consumption (beyond the recirculation and recuperation systems)
  - Decrease the CF<sub>4</sub> concentration: preliminary results show that 5% could be safe for operation
  - Look for alternatives to CF<sub>4</sub>
- Tried CF<sub>3</sub>I and HFO1234ze
  - F/C ratio very important
  - CF<sub>3</sub>I excluded: too electronegative and toxic
  - HFO: promising operation performance, further longevity studies are needed
- Several other gas candidates are considered for investigation
  - HFO-1336mzz(E), HFE-245fa1, HFE-143m
  - Studies not yet started

With 2% of HFO1234ze

+ no gain degradation up to 1.2 C/cm

- significant increase in dark current after 0.6 C/cm



Physics of Atomic Nuclei, Vol83(10), 2020

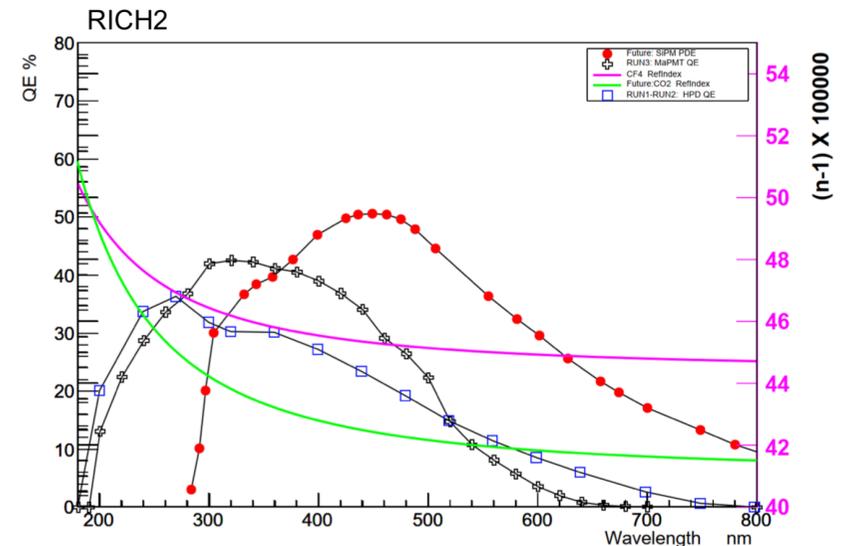
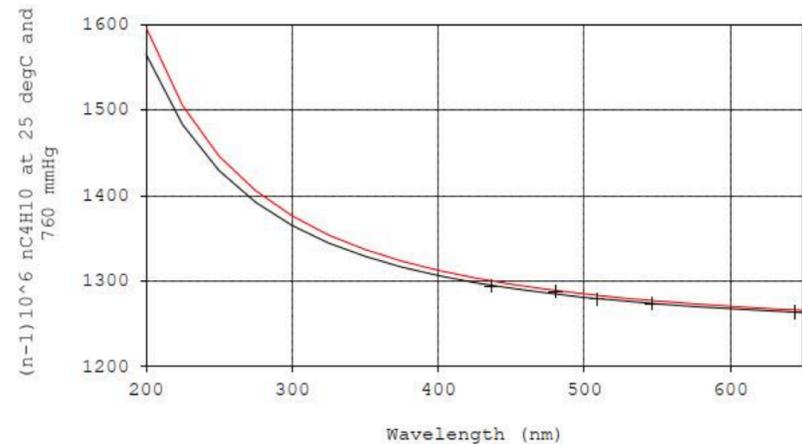
# Possible CF<sub>4</sub> replacements

CF<sub>4</sub> is used in different types of particle detectors to prevent aging, to enhance time resolution or because of its scintillation photon emission

## LHCb RICH studies

- RICH detectors use either CF<sub>4</sub> or C<sub>4</sub>F<sub>10</sub>
  - Necessary for good refractive index
- Replacement of C<sub>4</sub>F<sub>10</sub> with C<sub>4</sub>H<sub>10</sub>
  - Refractive index matches very well
  - But C<sub>4</sub>H<sub>10</sub> flammable
- Replacement of CF<sub>4</sub> with CO<sub>2</sub>
  - Under investigation
- Use of SiPM to reduce the chromatic error and increase the yield

C<sub>4</sub>F<sub>10</sub> vs C<sub>4</sub>H<sub>10</sub> : RICH1



S. Easo and O. Ullaland

# Conclusions

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*With climate change a growing concern,  
CERN is committed to reducing its direct greenhouse gas emissions*

## Optimization of current technologies

- Gas recirculation systems are the best way to reduce GHG consumption
- Nowadays upgrades of gas systems beyond original design

## Gas recuperation plants

- Used when not possible to recirculate 100% of the gas
- Very complex and different technologies depending on the GHG to recuperate

## Alternative gases

- A lot of work especially in RPC community to search for alternative to  $C_2H_2F_4$
- Not an easy task to find new eco-friendly gas mixture for current detectors

## GHG Disposal

- Very last alternative: only if previous strategies will not work

# Why it is so difficult to find alternatives

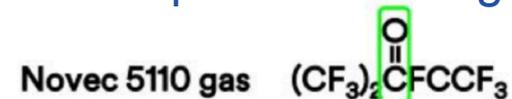


## Several key factors to take into account

- Environment → GWP
  - Related to IR absorbance over time
- Performance → Atmospheric lifetime
  - Water solubility → Rain out
  - OH reactivity → Oxidation
  - UV absorbance → Photolysis
- Safety
  - Non toxic
  - Non flammable (depending on detector/experiment)

$C_2H_2F_4$  and  $SF_6$  have good performance also because of their stability in atmosphere

## Example for Novec gases



Rain Out → Water Solubility  
very low water solubility (1 ppmw)

Oxidation → Reactivity with •OH  
unreactive w/ •OH radicals

Photolysis → UV Absorbance  
strong absorbance in near UV (wavelengths ≥ 300 nm)



Rain Out → Water Solubility  
very low water solubility (272 ppbw)

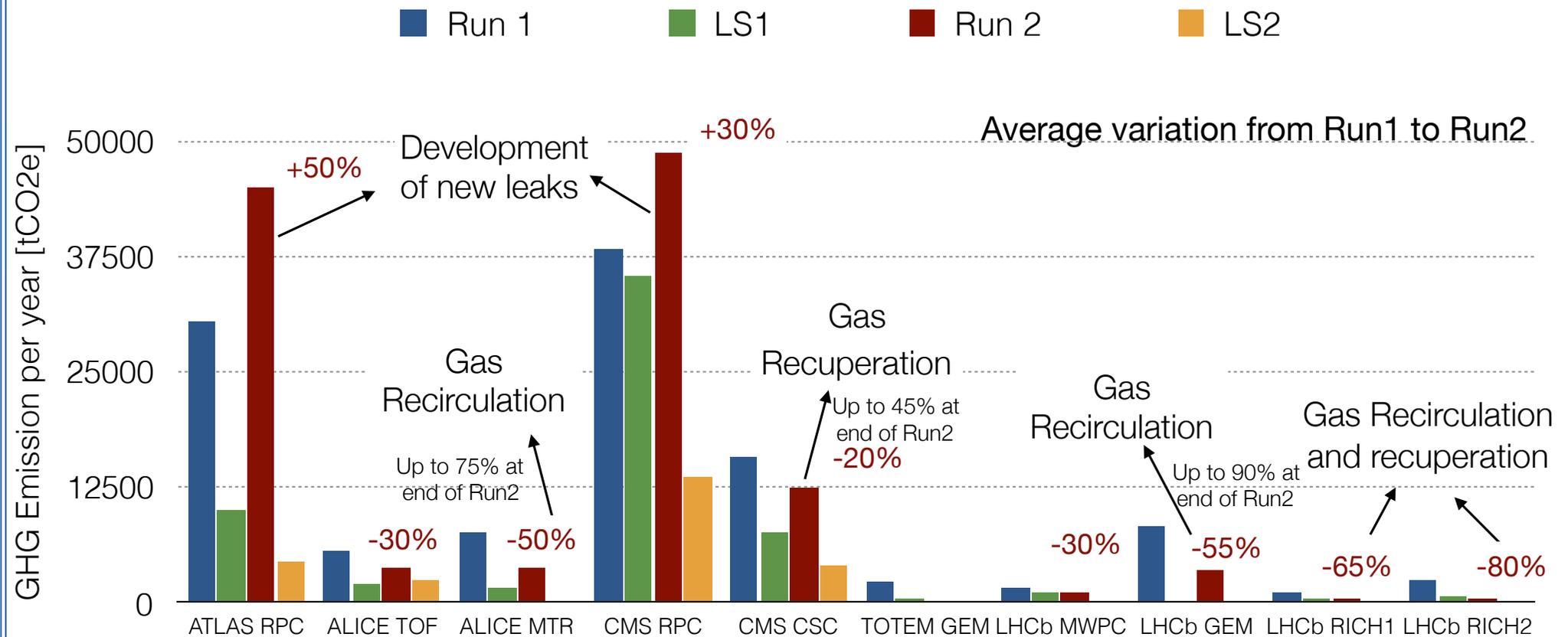
Oxidation → Reactivity with •OH  
reactive w/ •OH radicals

Photolysis → UV Absorbance  
transparent in near UV

# Back-up slides

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# GHGs for particle detection at LHC: Run1 vs Run2



- From Run1 to Run2 only increase of emissions is ATLAS and CMS RPC due to development of new leaks at detector level
- All other detector systems had a decrease of GHG emission from -20% to 80%
- Thanks to the different gas system upgrades performed and a major attention on GHG use

# Gas Systems at the LHC experiments

*The gas systems are complex apparatus that extend over several hundred meters and have to ensure an extremely high reliability in terms of **stability** and **quality** of the gas mixture delivered to the detectors*

At LHC Experiments we have 30 gas systems for a total of ~300 modules interconnected with ~90 km of pipes and controlled/monitored with PLCs and > 1000 sensors

## Reliability

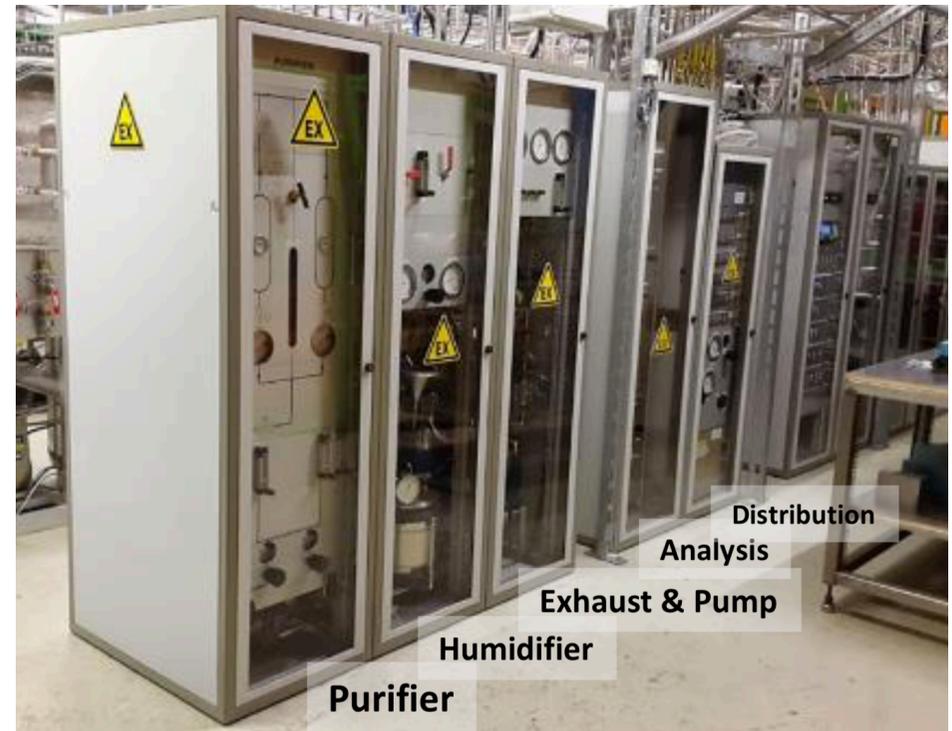
- LHC experiments are operational 24/24 7/7
- Gas systems must be available all time

## Automation

- Large and complex infrastructure
- Resources for operation
- Repeatability of conditions

## Stability

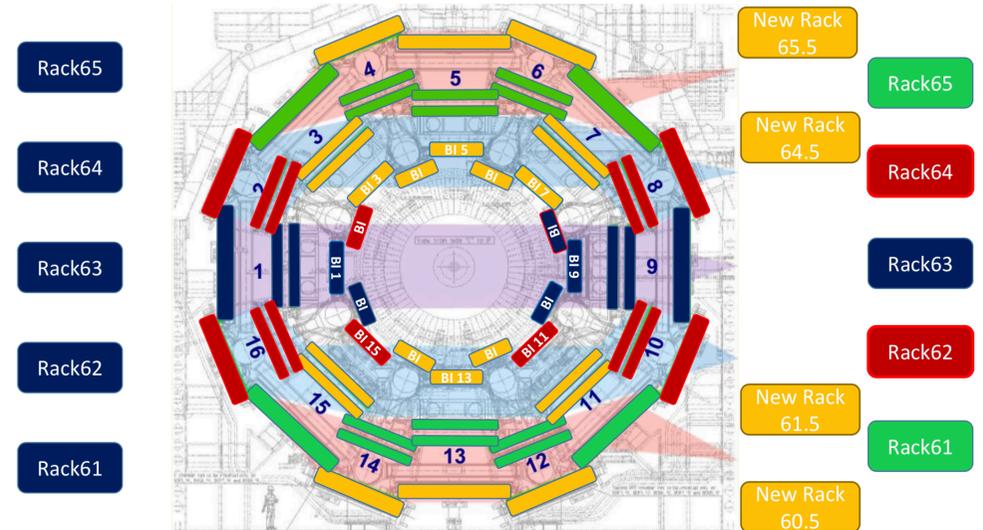
- Detector performance are strictly related with stable conditions (mixture composition, pressures, flows, ...)



# Optimization of distribution systems: ATLAS RPC

**Goal: to minimize the hydrostatic pressure on the detectors**

- The RPC gas mixture has a high hydrostatic pressure:  $\sim 0.3$  mbar/m
- The gas distribution racks are located in the cavern on different levels
- The addition of 4 new distribution racks will allow a better pressure equalisation between the chambers (total 9 racks)

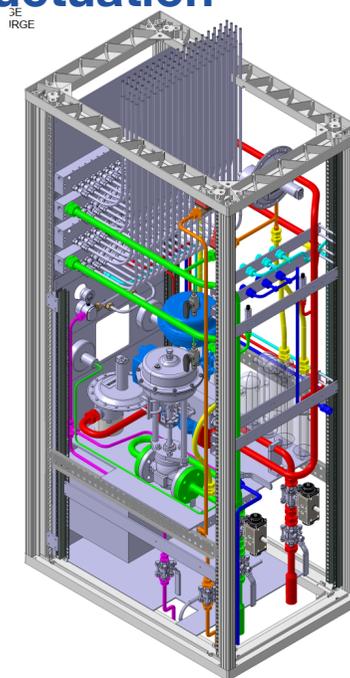


**Goal: to minimize any chamber pressure/flow fluctuation from some 0.1 to  $\sim 0.1$  mbar**

**Addition of regulation valves:** to better regulate and smooth the input pressure going to the flow distribution

**Reference chamber:** to have a good reference for the regulation of the detectors pressure

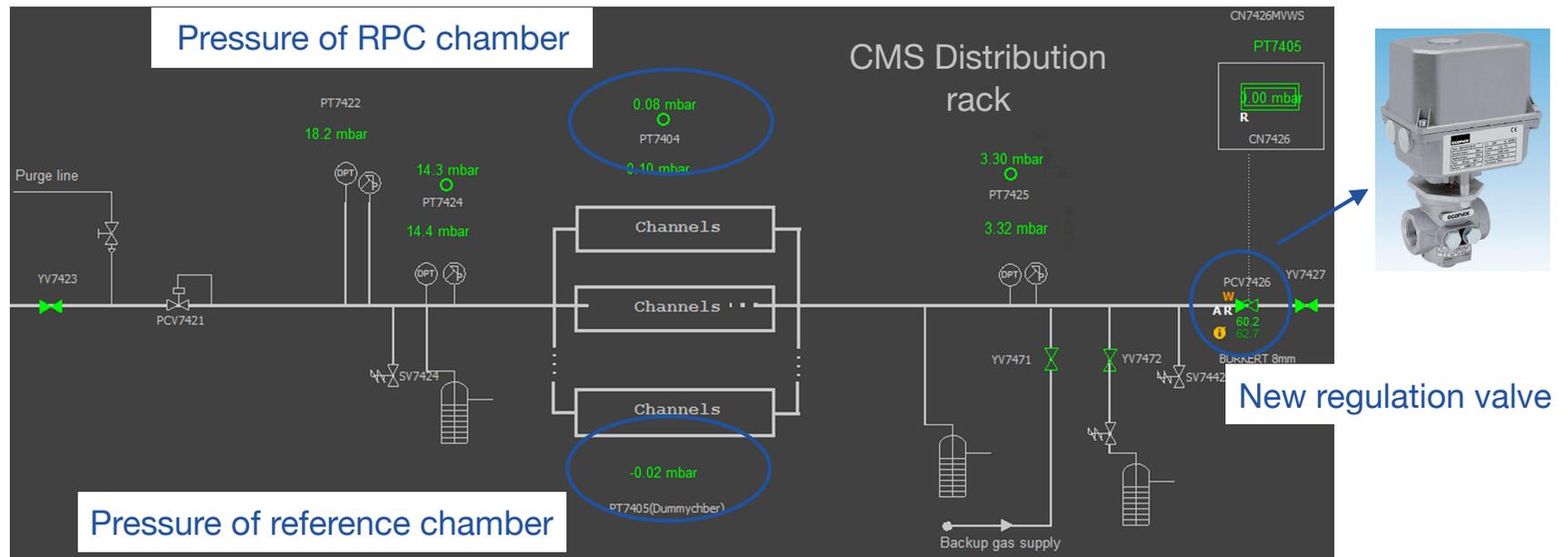
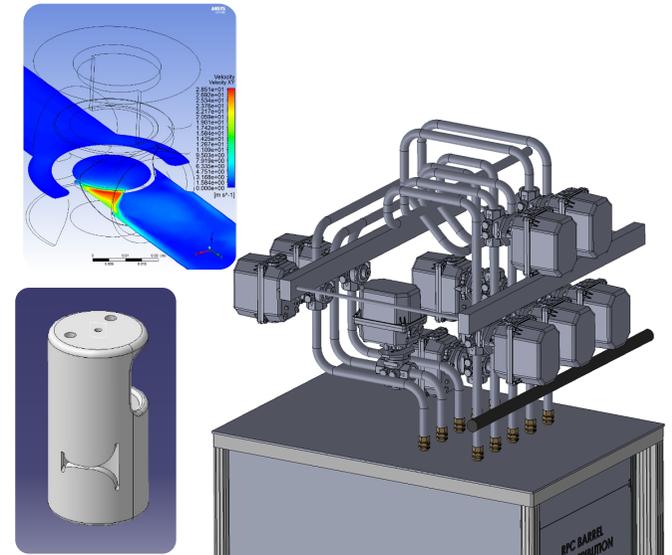
**Gas impedance:** to smooth pressure and flow fluctuations at the output of distribution system, i.e. pressure and flow seen by the detectors



# Optimization of distribution systems: CMS RPC

**Goal: to try to minimise as much as possible any fluctuation of pressure and flow at the detector level**

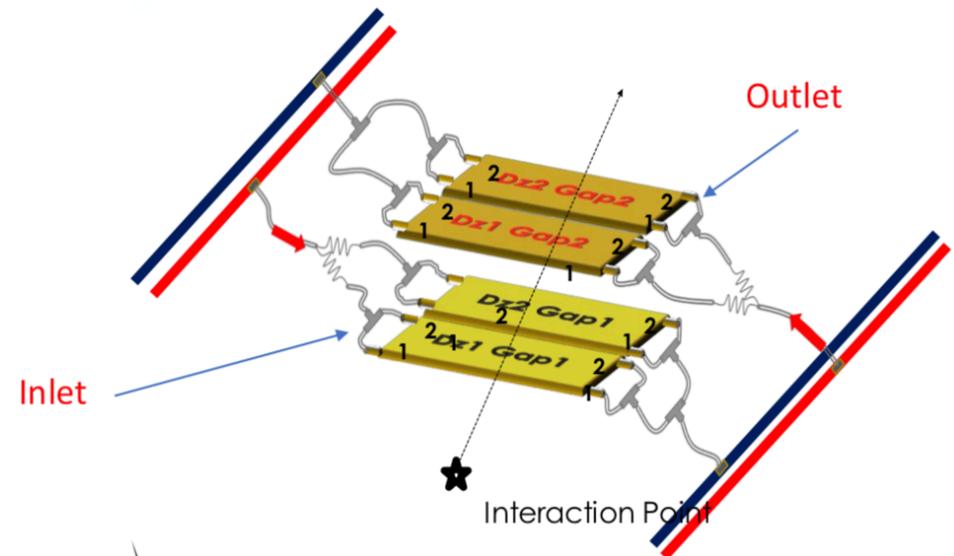
- New automated regulation valves on the return of each distribution rack to minimize any pressure changes
- To decrease the risk of developing new leaks at the detector level
- 30 distribution racks for Barrel and Endcap divided into top and bottom
- Different valve seats depending on pressure, flow, etc.
- Installation of 30 reference volumes
- To have a good reference for the regulation of the detectors pressure



# Causes of the leak: ATLAS case

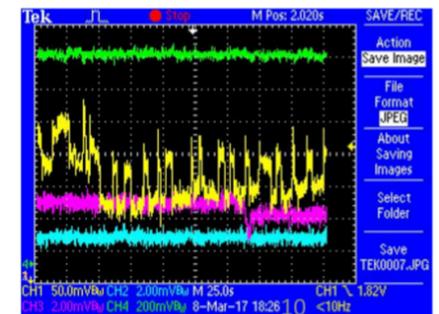
The ATLAS RPC leaks are **concentrated in the gas inlets and outlets.**

**Typical new cracked inlet**



The following main causes were individuated:

- A **lower than expected quality in the original polycarbonate** moulded inlet and outlet production
- A stress applied to the gas inlets through the gas pipes
- The gas system is generating a constant stress in form of **fast propagating flow changes (order of 1-2 %)**
- The **former purge procedure** (happening at each YETS) was **producing a very large shock** with a consequent large system damage.



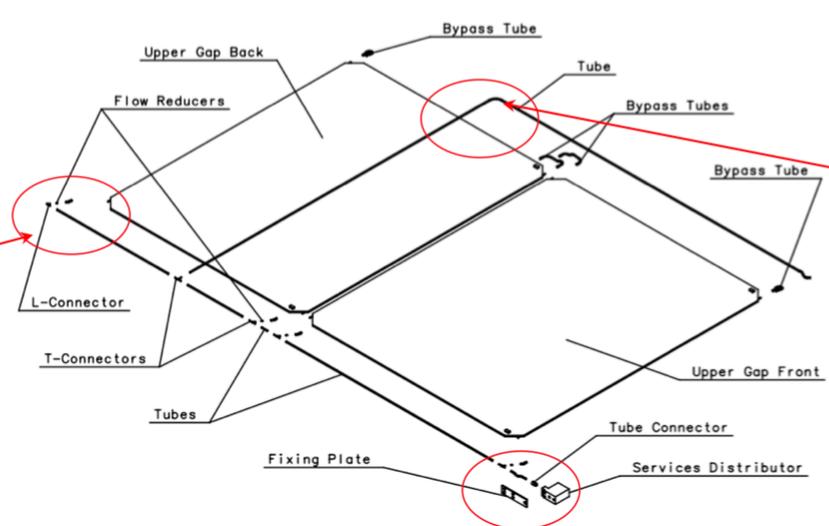
# Causes of the leak: CMS case

The CMS RPC leaks are mainly caused by:

1. **T or L polycarbonate gas connectors** break due to too much stress applied through the gas pipes.
2. **Polyethylene LD pipes** brittle/deteriorated or cut. This problem is mainly present in the last two station where two chambers are internally connected in parallel.
  - One “bad” batch of pipes was identified. Cracked pipes are all coming from the same batch.
  - Environmental cavern Humidity can accelerate this process.



Broken L



Cut bypass pipe RB3/RB4



# Gas recuperation: LHCb RICH2 CF<sub>4</sub>

## RICH2 Gas System

- Detector volume ~100 m<sup>3</sup>
- Gas mixture: **92% CF<sub>4</sub>**, 8% CO<sub>2</sub>
- Gas recirculation: ~100%
- Small quantity lost in leaks or for gas system operation

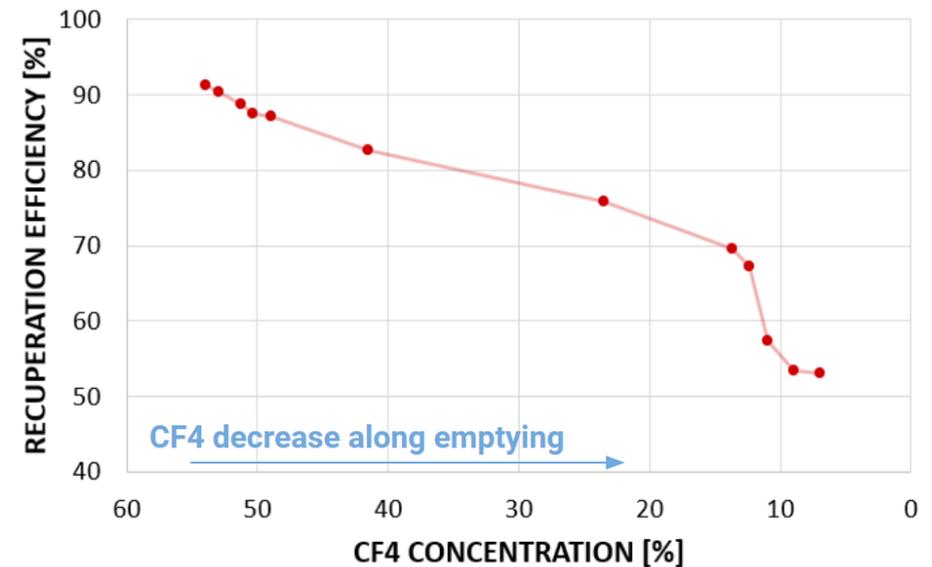
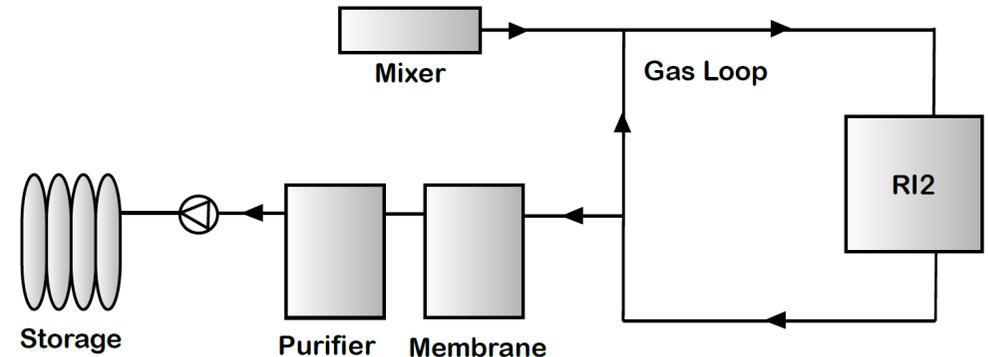
## RICH2 Recuperation System

- Two recuperation modes (warm separation)
- During long shutdown: emptying detector
- During Run: recuperation of small quantities otherwise lost in gas modules
- New system implemented in LS2
- Upgrades on-going

## Performance

- Recuperation efficiency ~60%
- About 30 m<sup>3</sup> of CF<sub>4</sub> recuperated in LS2
- CF<sub>4</sub> quality satisfactory
- CF<sub>4</sub> recuperated will be re-used for Run 3 operation

GHG reduction from Run1 to Run2 up to **60%**



# Gas Recuperation: azeotrope $C_2H_2F_4/iC_4H_{10}$

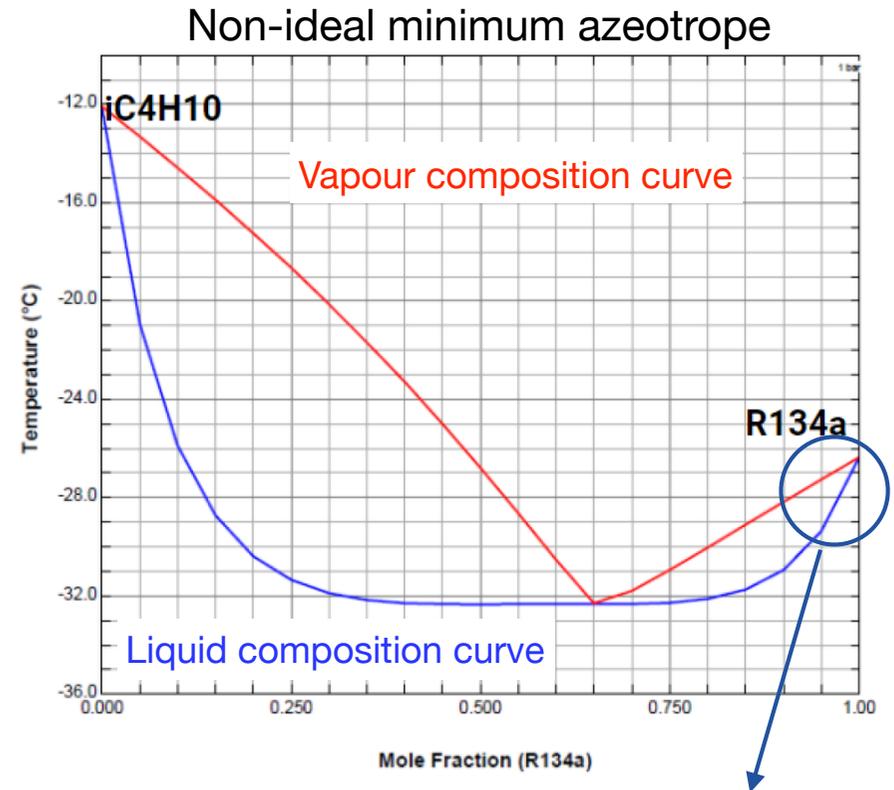
## ATLAS and CMS RPC Gas Systems

- Detector volume  $\sim 15 \text{ m}^3$
- Gas mixture:  $\sim 95\% C_2H_2F_4$ ,  $\sim 5\% iC_4H_{10}$ ,  $0.3\% SF_6$
- Gas recirculation:  $\sim 90\%$ 
  - maximum recirculation validated for RPC detectors
- Fundamental to repair detector leaks
  - To have the gas at the exhaust (600-1000 l/h)

## RPC Recuperation System

- Not convenient to recuperate the gas mixture
- Cold separation for R134a
  - Thermodynamic phase transitions
- R134a and  $iC_4H_{10}$  form an azeotrope
  - A mixture of liquids whose proportions cannot be altered or changed by simple distillation
  - Intramolecular force of same-species is much higher than the reciprocal attraction separation by quasi-static increase of temperature

*Slow heating of the liquified azeotrope allows to enrich the liquid of R134a and the vapour of  $iC_4H_{10}$ , obtaining the separation*



# Gas disposal

***Abatement plants are employed when GHGs are polluted and therefore are not reusable***

In case all studies on recuperation will not bring to efficient recuperation plants, industrial system able to destroy GHGs avoiding their emission into the atmosphere have been considered

Quite heavy infrastructure required:

- CH<sub>4</sub>/city gas + O<sub>2</sub> supply + N<sub>2</sub> supply
- Waste water treatment
- PFC/HFC are converted in CO<sub>2</sub> + HF acid dissolved in water
- disposal of remaining waste/mud
- To have the gas at the exhaust (600-1000 l/h)

*Joint CMS and EP-DT gas team is studying the feasibility*



Found also companies available to take PFC/HFC based mixture for disposal: but extremely expensive