Environmentally friendly gas mixtures for Resistive Plate Chambers

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EP-DT
Detector Technologies

66th ELOISATRON WORKSHOP

Outline

- RPCs at LHC
  - R-134a and SF6 usage
  - Key aspect for finding new environmentally friendly gas mixtures
- Performance with RPCs operated with eco-friendly gas mixtures
  - R-1234ze as a R-134a alternative for the long term
  - CO2 addition to standard gas mixture for the short-mid term
  - SF6 alternatives
- Performance with RPCs operated with eco-friendly gas mixtures
  - HF production studies
  - Environmental chemistry studies
- Conclusions

GHG consumption of RPCs at LHC

HPL RPCs at LHC are operated with 90-95% of *R-134a* + 0.3% SF6 → **95% GWP** contribution due to **R-134a**, **5%** due to **SF6**

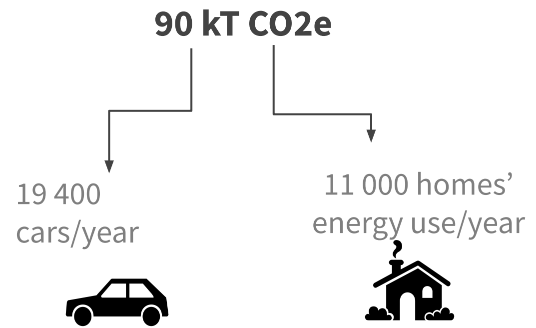
Glass RPCs at LHC operated with 93/7 R-134a/SF6 → **37% GWP** contribution to **R-134a**, **63%** due to **SF6**

LHC 2022-2023 consumption estimates

Gas	Consumption	Yearly consumption	CO2e consumptions	Relative GHG contribution
R-134a	1550 ln/h	61.8 t	88.3 kTon	89%
SF6	8.3 ln/h	474 kg	10.8 kTon	11%

Key aspects in GHG reduction

- **Environmental** → high GHG consumption due to leaks
- **Economical** → increasing costs of high-GWP gases
- **Availability** → decreasing on the market due to EU phase out



https://www.epa.gov/energy/greenhouse-gas-equivalencie-calculator

Goal

Replace or reduce R-134a without changing the current RPC infrastructure (no change in FEB, HV, Gas system)

Key aspects for the search of new gas mixtures for LHC

RPC short term performance

- Working point limit for HV system
- Sufficient rate capability for HL-LHC

RPC long term performance

- Sustained rate capability with reasonably contained currents

Human safety

- Flammability limits
- Toxicity limits

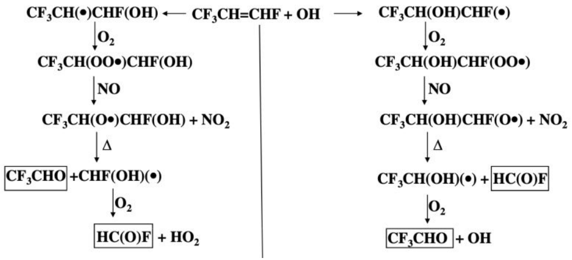
Environmental properties

- Chemical stability
- Side effects on environment

Engineering for gas systems

- Components validation in case of new gases
- Purifying system
- Compatibility with recuperation plant

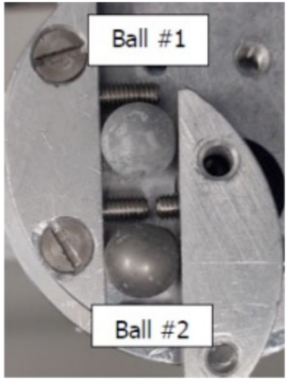
R-1234ze degradation products in atmosphere



R-1234ze flammability with i-C4H10

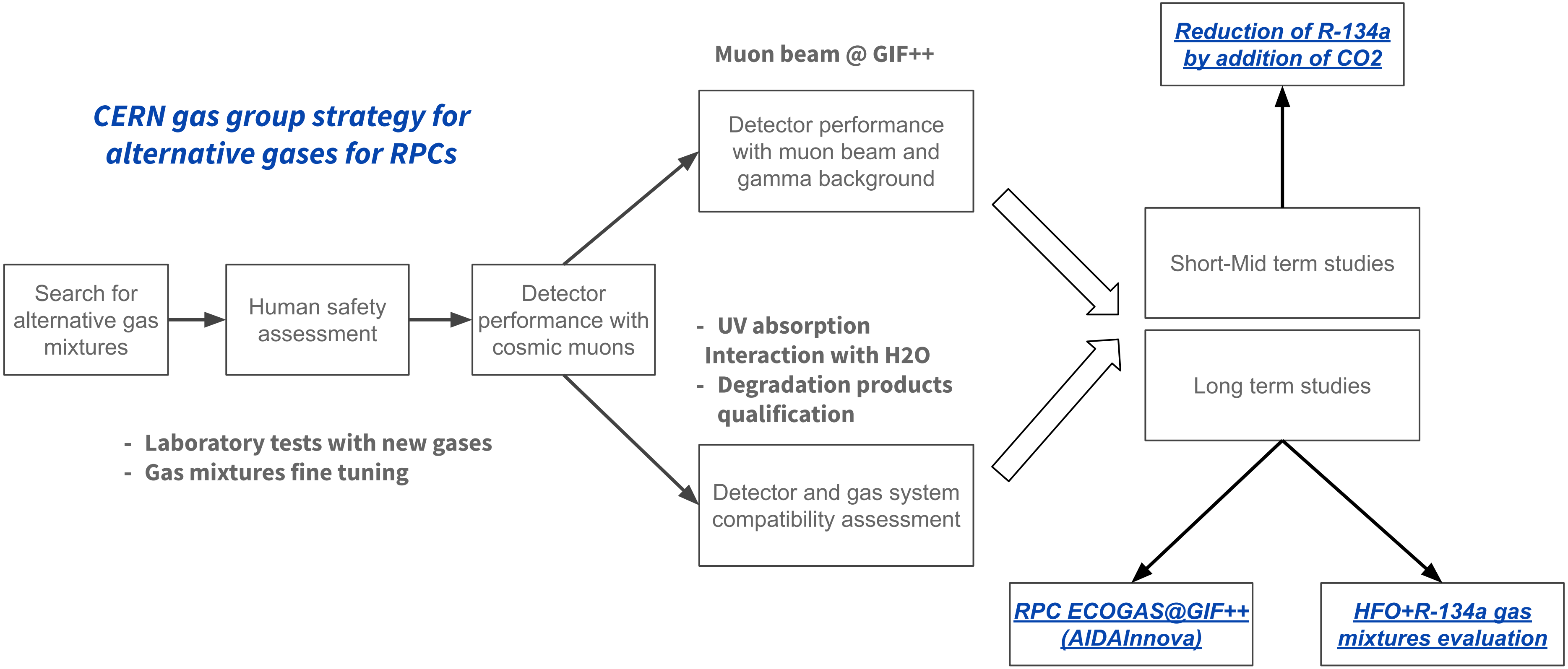
test no.	Iso-butane fraction in test gas mixture in mol%	fraction of test gas mixture of iso-butane and HFO1234ze in mol%	fraction of air including 2.25 mol% water in mol%	reaction
9	**6.2**	**15.0**	**85.0**	**+**
10	6.0	20.0	80.0	-
11	**4.2**	**13.0**	**87.0**	**+**
12	**3.1**	**10.0**	**90.0**	**+**
13	**2.2**	**13.0**	**87.0**	**+**
14	1.1	13.0	87.0	-
15	**1.0**	**10.0**	**90.0**	**+**
16	0.0	12.0	88.0	-
17	0.0	11.0	89.0	-
18	0.0	10.0	90.0	-
19	0.0	9.0	91.0	-

Presence of Chlorine in CMS-RPC



Research lines in eco-friendly gas mixtures for RPCs at LHC

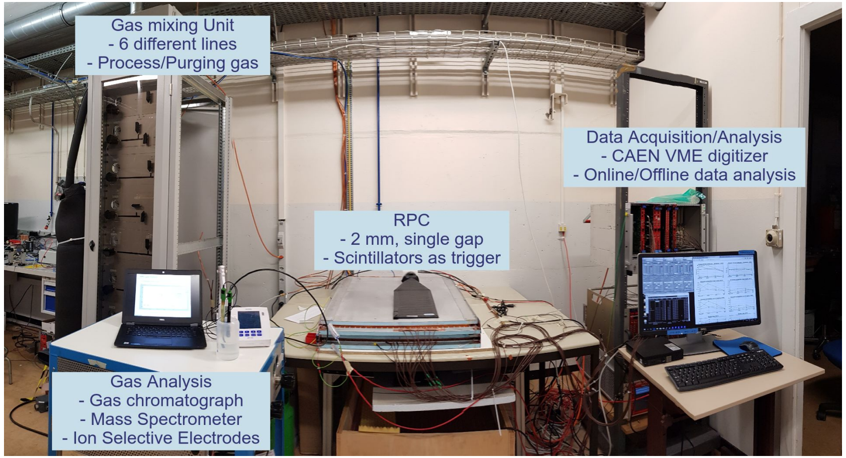
CERN gas group strategy for alternative gases for RPCs



Experimental setup: detectors

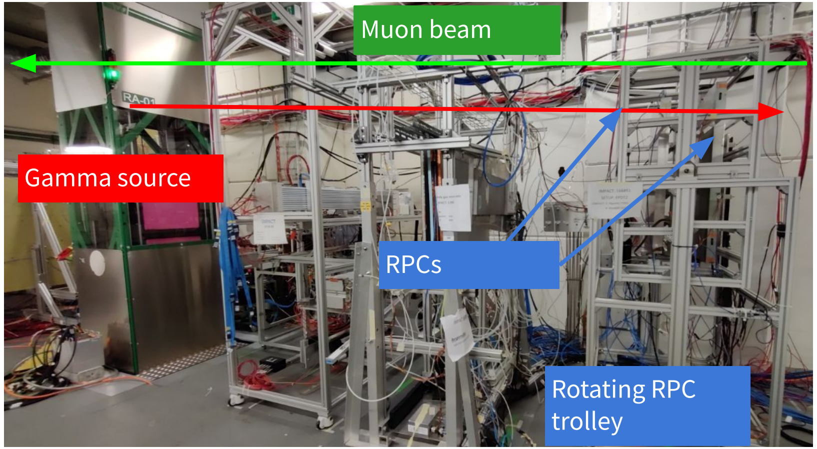
Laboratory setup

- Single gap, 2 mm electrodes + 2 mm 80x100 cm^2 gap HPL
- Tests of new gases
- Gas mixtures fine tuning: up to 6 components, 0.01% precision
- Low rates, cosmic muons → short term performance
- Raw waveform analysis: efficiency, st. prob., cluster size, time resolution, prompt charge



GIF++ setup

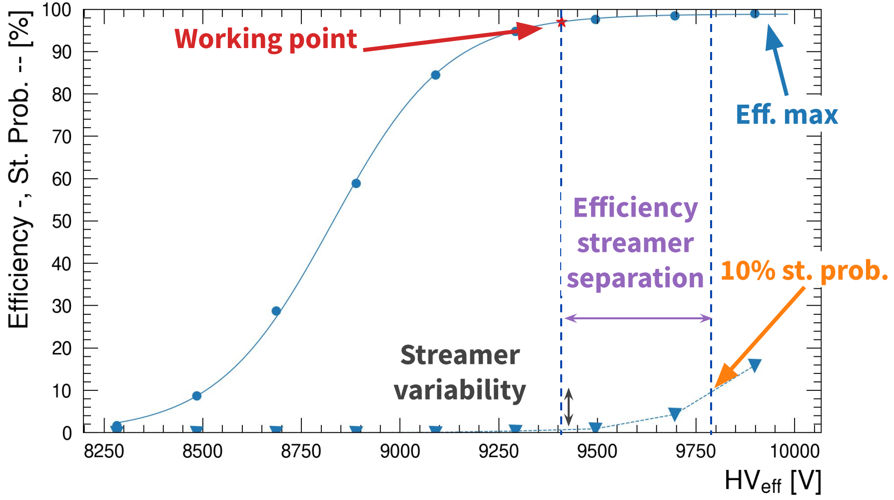
- Muon beam + ^{137}Cs gamma source
- Gas mixtures validation:
  - Muon beam at different background rates (ABS filters) → short term performance
  - Currents, resistivity stability under irradiation → long term performance



Data acquisition and analysis

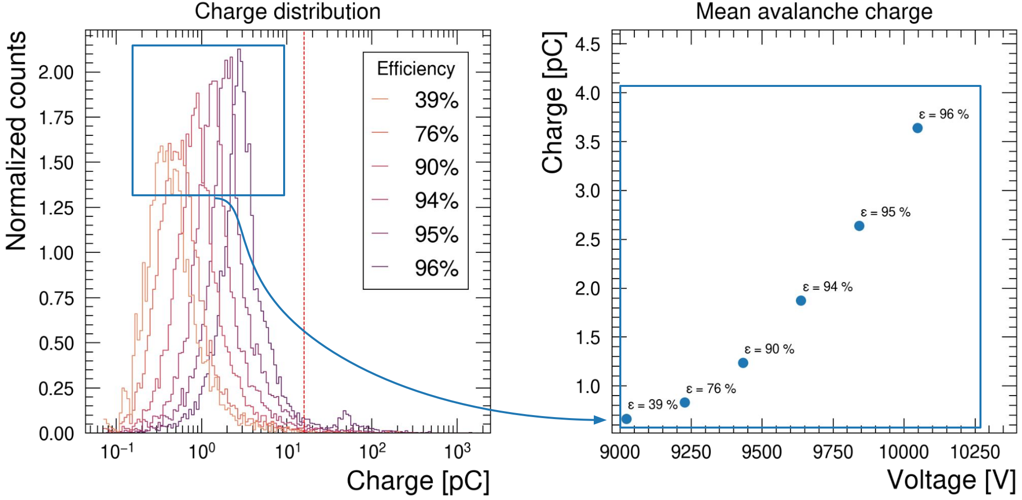
Data acquisition

- **Raw waveform** digitizing: efficiency, charge, shape, time analysis of signals
- 7 strips readout / RPC
- 2-3 RPC for **result consistency**
- HV scans: ~ 10 HV points, 10^4 waveforms for each HV point → **O(10^5-10^6)** waveforms analyzed per run



Data analysis

- Efficiency fitting with sigmoid function
- Working point definition: **HV(95% of ε_{max}) + 150 V**
- Avalanche / Streamer threshold: **10^8 electrons** ~ 16 pC
- **Efficiency-streamer separation**: $\Delta V_{w.p}$ - $\Delta V_{10\%\ st.prob.}$, **streamer variability:** (w.p. ± 50V)
- **GIF++ tests**: foremost parameters evaluated at working point **for each ABS filter**
  - Tested up to 500-600 Hz/cm²



Alternatives to R-134a

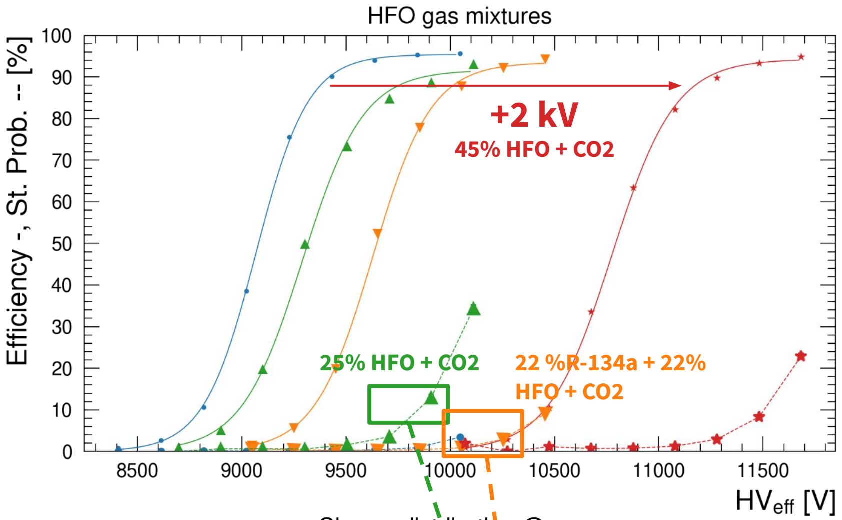
Alternative to R-134a: R-1234ze

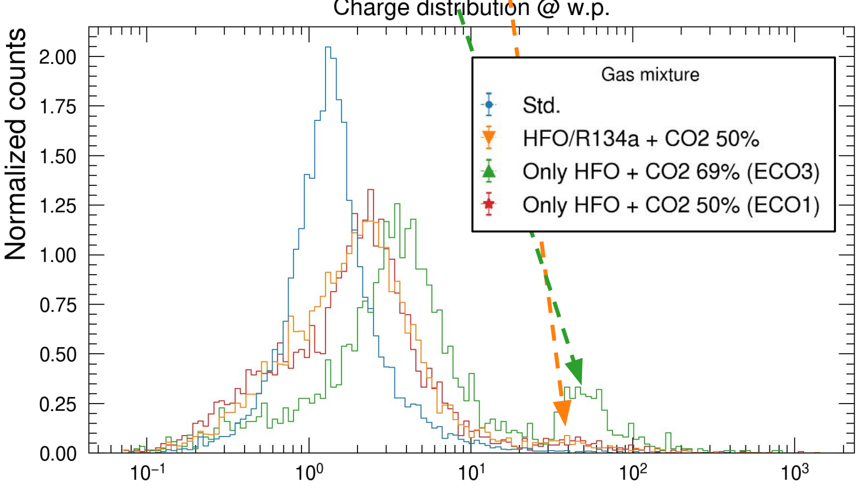
R-1234ze identified as possible replacement to R-134a

- Extremely **low GWP (~ 7)**
- Increasingly **wider adoption** in refrigerant industry
- However, **market price and availability** not yet comparable to R-134a → *Honeywell patented*
- **Cannot replace 1:1 R-134a** → w.p. too high → CO_2/He required to lower w.p.
- **Long term effects** still under investigation

R-1234ze performance with CO_2 (+ R-134a) with cosmic muons

- **45% HFO + CO_2 (ECO1) ⇒** w.p. too high (~11.6 kV)
- **25% HFO + CO_2 (ECO3) ⇒** low GWP, high charge content → higher currents. Currently being tested by *RPC ECOGAS collaboration*
- **22% HFO + 22% R-134a + CO_2 ⇒** higher GWP, lower charge content than HFO only. **Possible compromise** between performance and environment



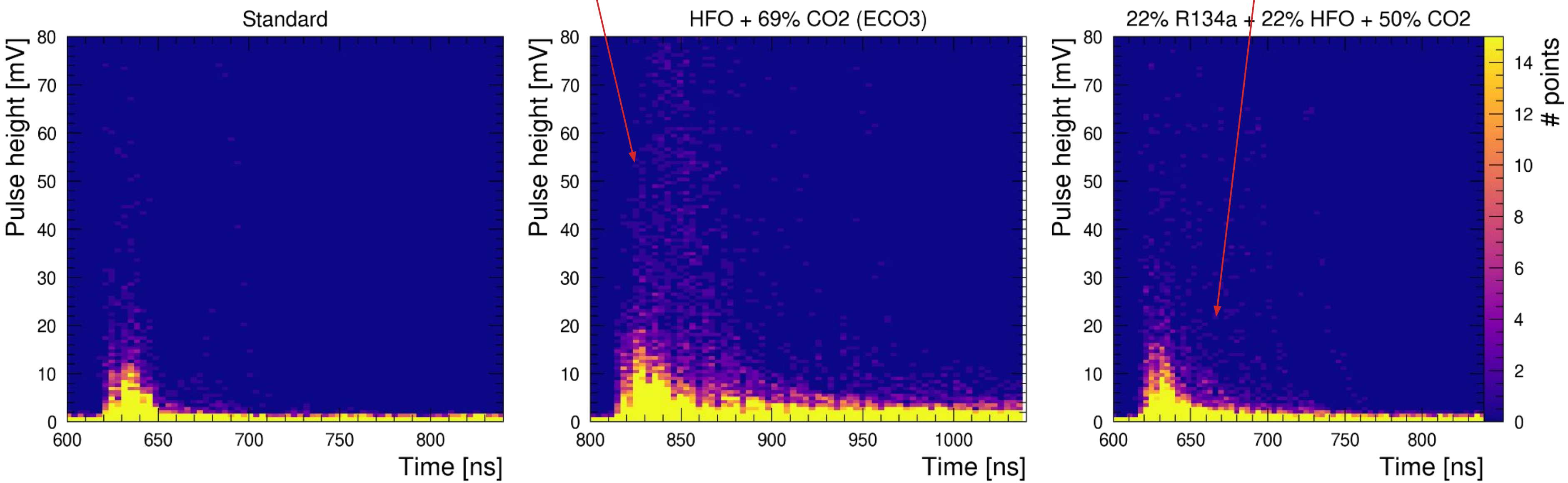


Waveforms of Std vs. HFO vs. HFO + R134a gas mixtures

HFO only → higher charge content: bigger and longer signals

HFO + R-134a only: lower charge content and faster signals decay times

Waveforms with cosmics muons @ w.p.



ECOGAS Collaboration: test beam results

Two HFO/CO2 gas mixtures identified

- higher HFO → lower currents but higher w.p.
- higher CO2 → lower w.p., higher currents, lower max. eff.

Irradiation campaign ongoing

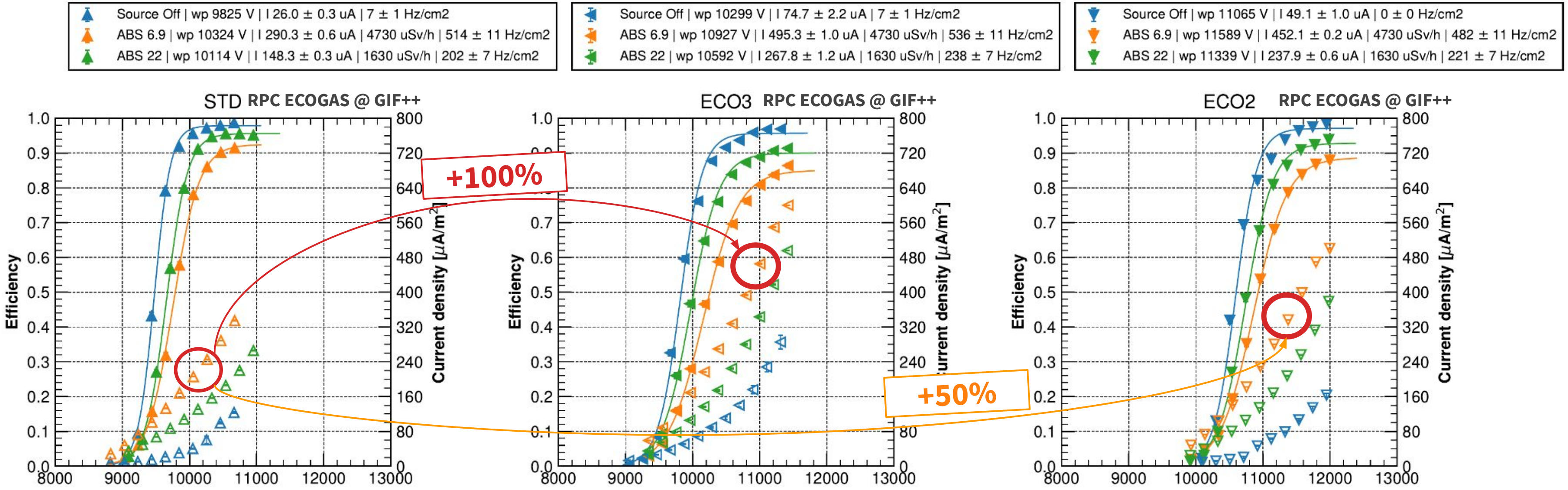
- **ECO2** selected for long term tests

Joint collaboration between CERN Gas group, ALICE, ATLAS, CMS, LHCb-SHiP

STD = 95.2% R134a + 4.5% ISO + 0.3% SF6

ECO2 = 35% HFO + 60% CO2 + 4% ISO + 1% SF6

ECO3 = 25% HFO + 69% CO2 + 5% ISO + 1% SF6



R-134a + R-1234ze + CO2/He gas mixtures @ GIF++

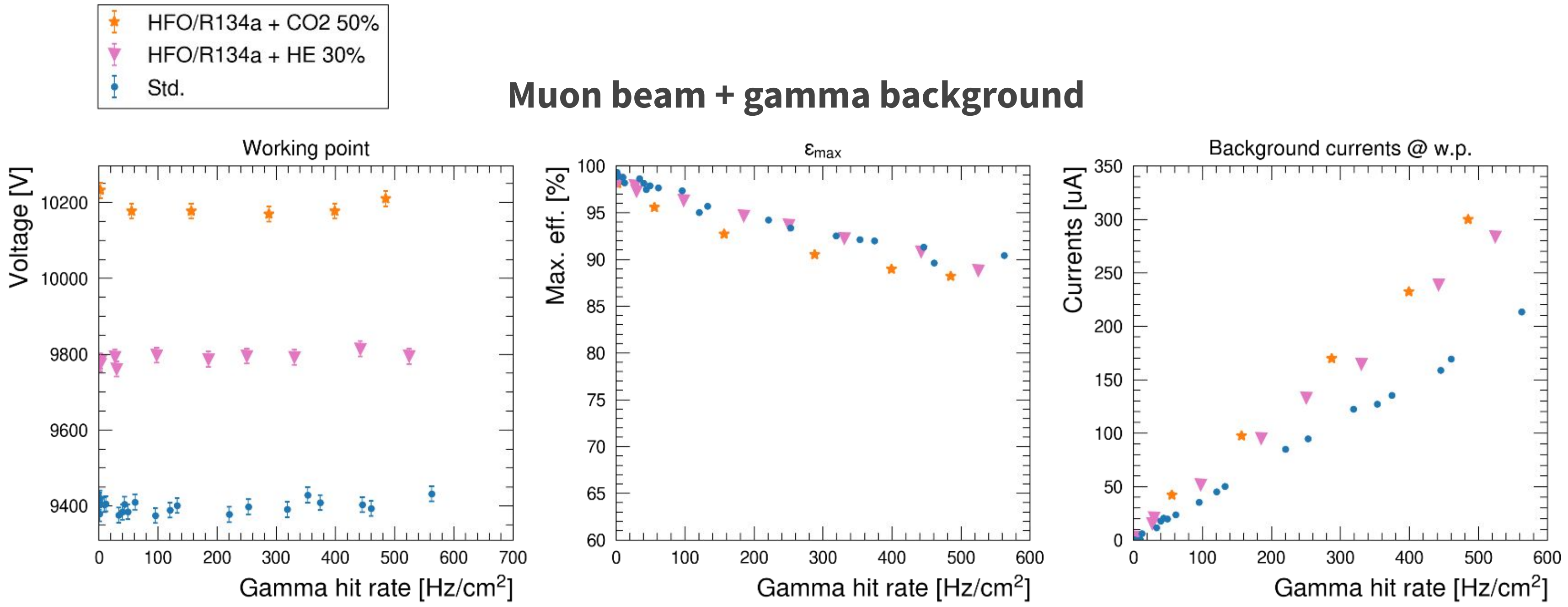
R-134a + R-1234ze: two gas mixtures at high rates (1 **CO_2 50%**, 1 **He 30%**):

He gas mixture has lower working point than **CO_2** one

CO_2 + R-1234ze gas mixtures have slightly higher **efficiency drop** (-2 %)

He gas mixture has slightly lower currents than **CO_2** equivalent (**+30% He**, **+50% CO_2**)

Muon beam + gamma background



Short-Mid term: R-134a reduction by addition of alternative gases

Reduction of R-134a in the standard gas mixture by addition of a 4th, non-fluorinated gas

O2: good performance but highly reactive → lower **flammability limit**, higher currents due to oxidation reactions

Ne: good performance but **no availability** on the market

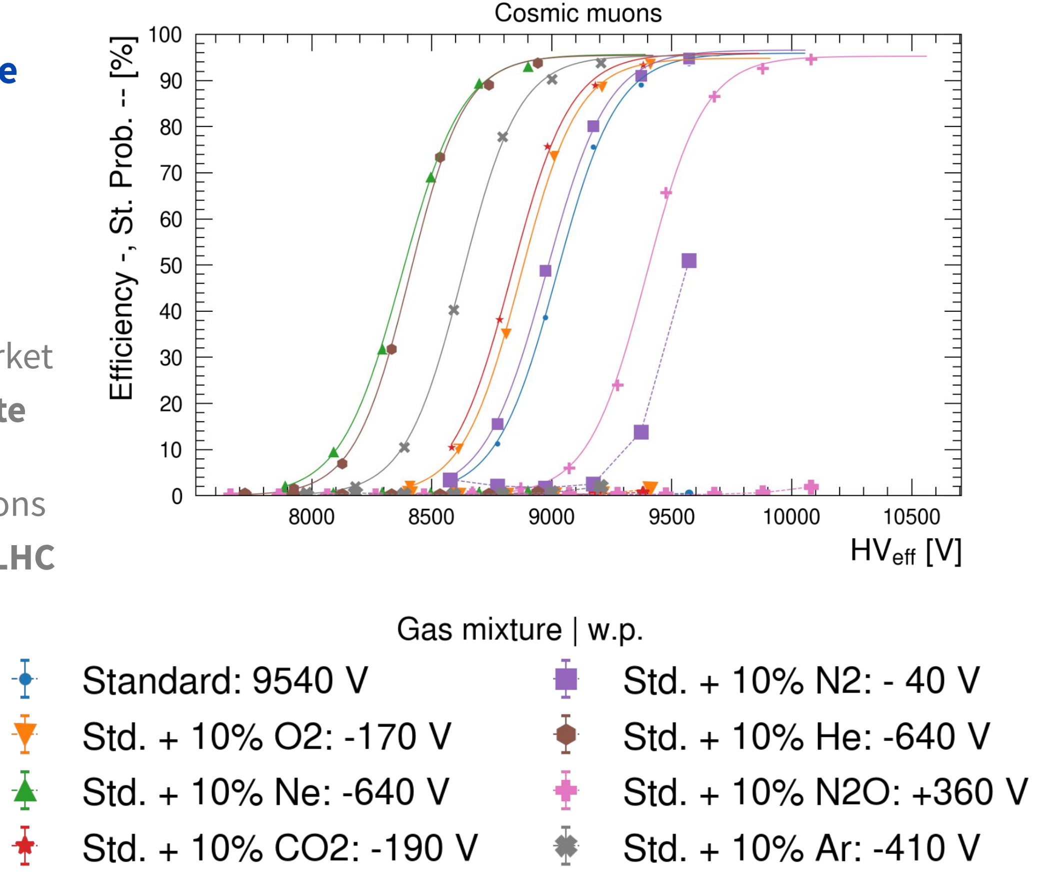
CO2: good performance → **selected as main candidate** for GIF++ tests

N2: **high streamer** contamination at low concentrations

He: good performance but **problematic for PMTs in LHC** caverns

N2O: discrete performance but **increased working point** of ~ 300 V

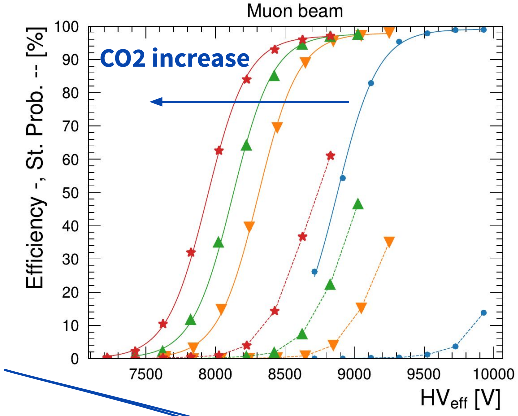
Ar: slightly **high streamer probability**

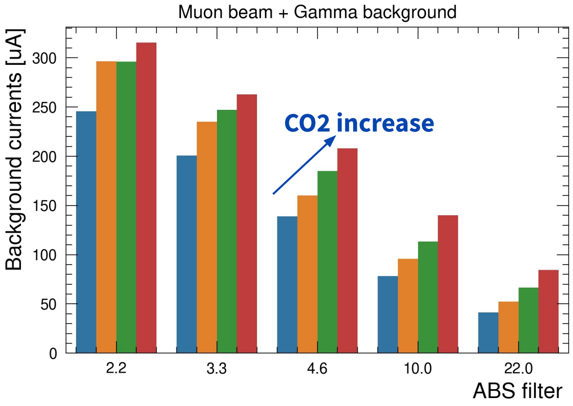


CO2 usage to mitigate R-134a consumption

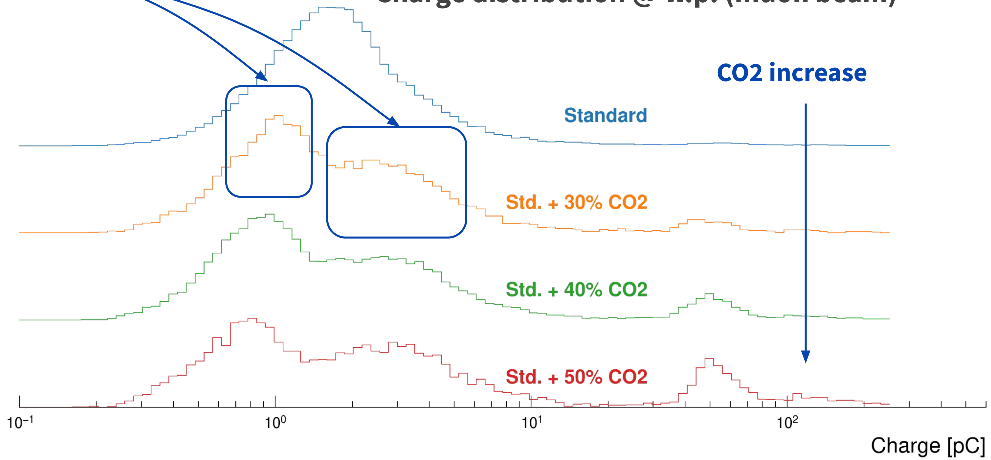
Studies on CO_2 impact when added to the standard gas mixture: 30%, 40%, 50%

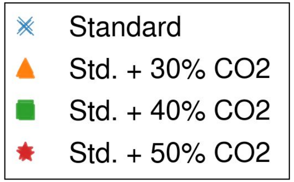
- Tests performed with **muon beam** and **gamma background**
- w.p. decreases of **~ 190 V / 10% CO2**
- **GWP** reduction of **30-50%**
- **Current** increase of **+10-15%** @ 500 Hz/cm2
- **Streamer** fraction **increases**
- **Two** avalanche **populations** when using CO2 and R-134a → under investigation





Charge distribution @ w.p. (muon beam)



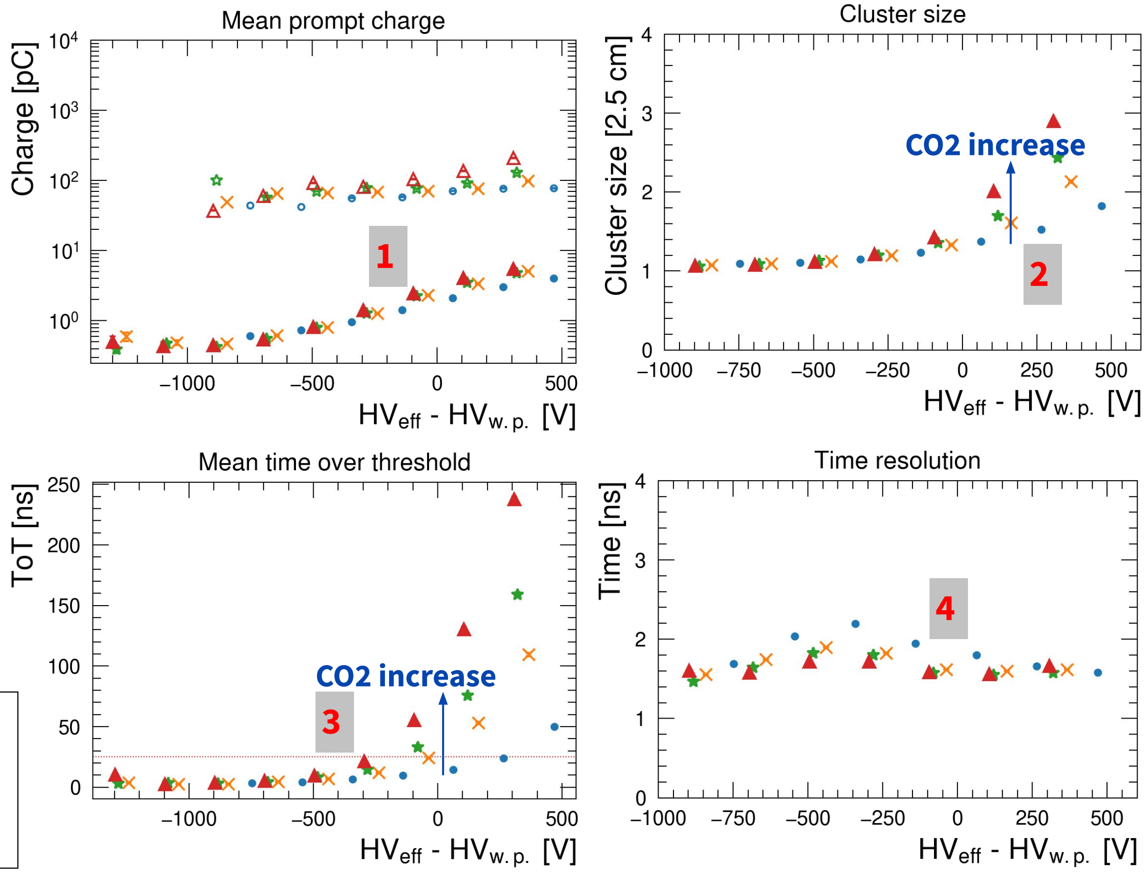


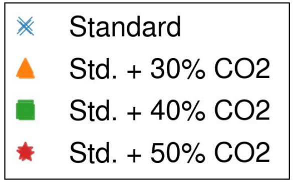
CO2 usage to mitigate R-134a consumption

Addition of 30%, 40%, 50% of CO_2 gas mixture as mid-term solution to mitigate R-134a. Muon beam studies

1. Average prompt charge slightly increasing (+10-15%)
2. Cluster size increases with CO_2 amount
3. Average signal times over threshold increase with CO_2
4. Time resolution of CO_2 gas mixtures is lower than std. one

Adjustment of **SF6** needed to further suppress **streamers**



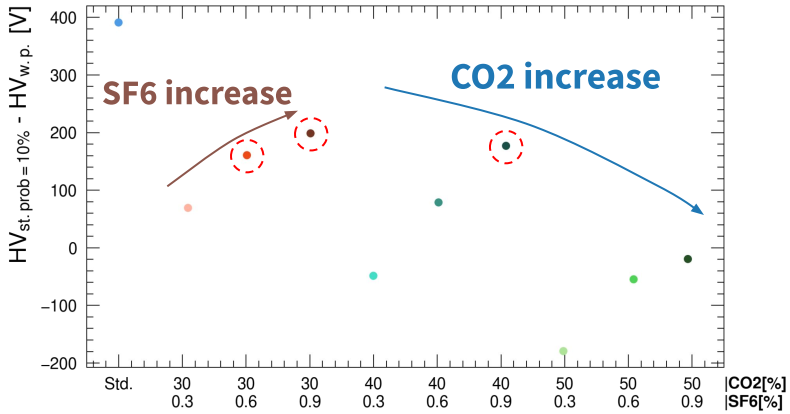


SF6 adjustment in CO_2 + R-134a gas mixtures

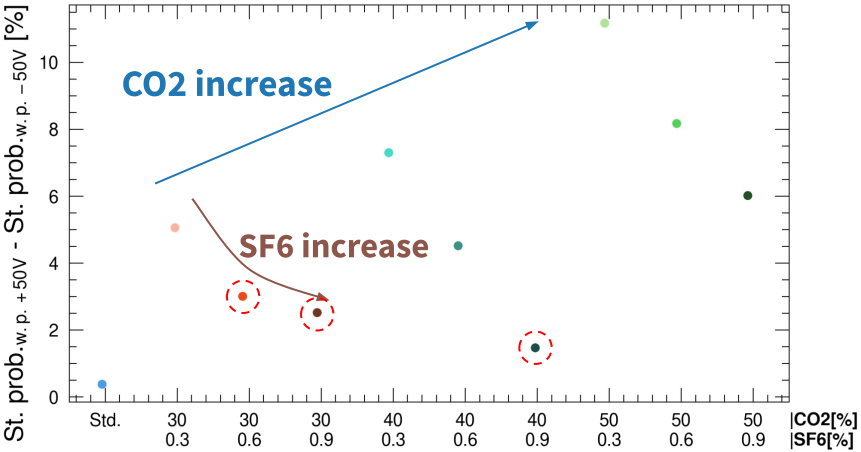
Combination (30%, 40%, 50%) **CO2** x (0.3%, 0.6%, 0.9%) **SF6**

- Higher efficiency-streamer separation for 30%/40% CO2 **+ 0.9% SF6** or 30% CO2 **+ 0.6% SF6** → possible candidates
- Lower variation of streamer probability for the same gas mixtures

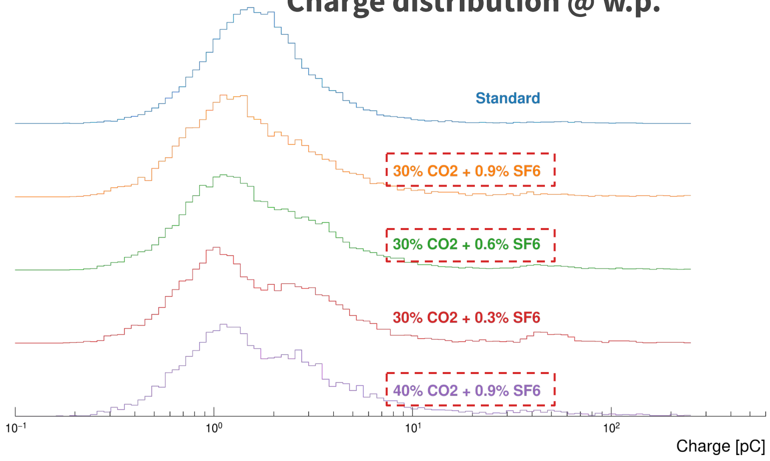
Efficiency - Streamer separation. Muon beam only



Streamer variability. Muon beam only



Charge distribution @ w.p.



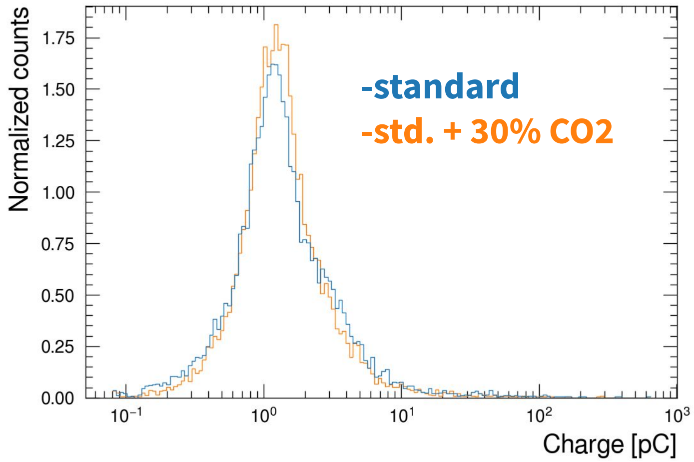
Selected CO2-based gas mixtures

Selected CO2/SF6 gas mixture for long term tests:

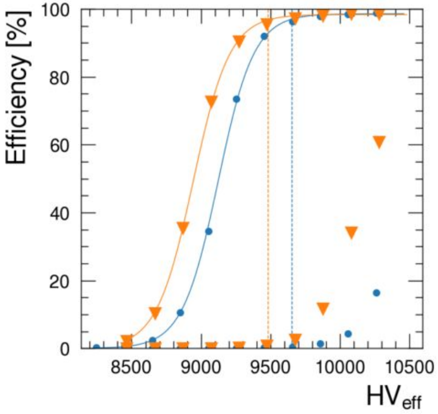
64% R-134a + 30% CO2 + 5% i-C4H10 + 1% SF6

- **30% CO2** → conservative approach
- **1% SF6** chosen to lower charge content
- **5%** i-C4H10 chosen to be compatible with **ATLAS RPC** requirements
- Currents ~15% higher → under investigation

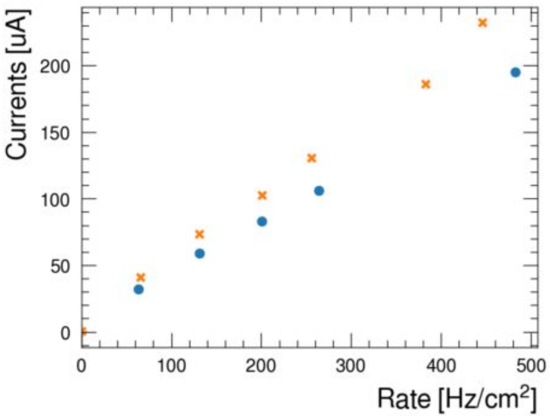
Prompt charge distribution - muon beam



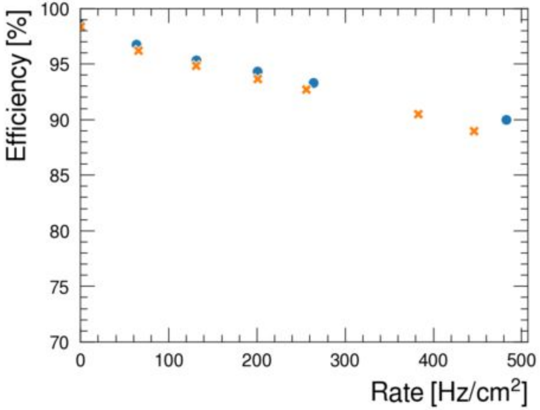
Eff. - St. prob.



Gamma currents



Maximum efficiency



Alternatives to SF6

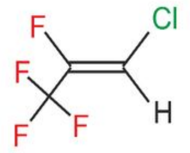
Alternatives to SF6: possible candidates

SF6 is used in power plants as an electrical insulator.
Energy industry engineered some alternatives:

- Ketones: **C4F8O**, **Novec^{TM} 5110**, other Novecs
- **Novec^{TM} 4710**
- Chlorinated HFOs: **HFO-1224yd**, HFO-1233zd, HFO-1336mz
- **CF3I**

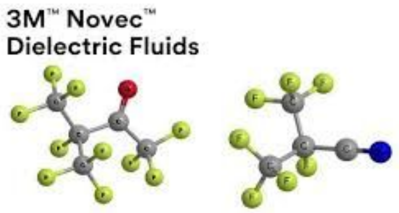
Key factors in the search of SF6 alternatives:

- Human safety → toxicity
- Low GWP and ~0 ODP
- Vapour pressure → sufficient gas phase
- Presence of new elements in the molecules → Cl, I, Br



HCFO-1224yd

GWP < 1

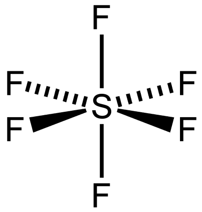


Novec 5110

GWP < 1

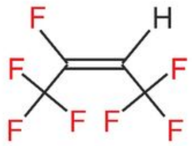
Novec 4710

GWP 2100



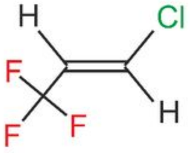
SF6

GWP 23900



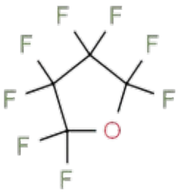
HFO-1336mzz(Z)

GWP ~ 1



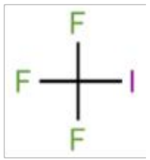
HCFO-1233zd

GWP ~1



C4F8O

GWP 8700



CF3I

GWP < 1

Alternatives to SF6

SF6 in the standard gas mixture **replaced** with its alternative:

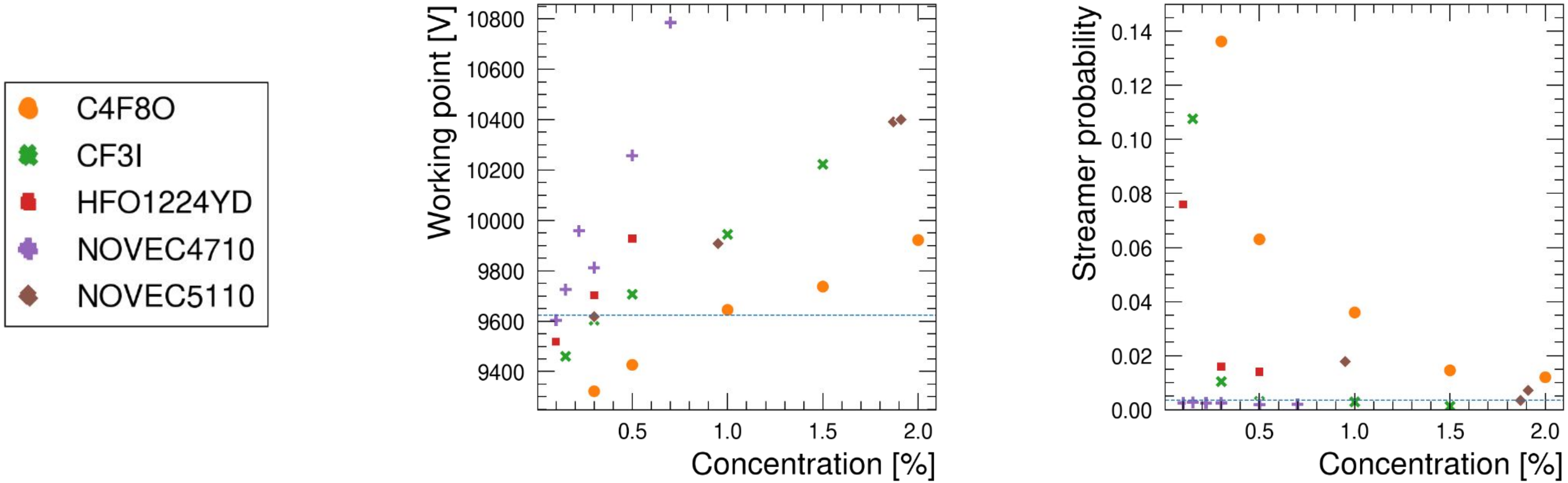
C4F8O → Discrete performance at 1.5%, **high GWP** (~8000)

CF3I → Good performance at 0.3% but **mutagenic toxicity**

Novec 5110 → Discrete performance at 2% but almost **liquid**

Novec 4710 → **Excellent** performance at 0.1% but it may react with **water** (under investigation) → selected for beam studies

Amolea 1224yd → **Good performance** at 0.3% → selected for beam studies



SF6 alternatives at GIF++

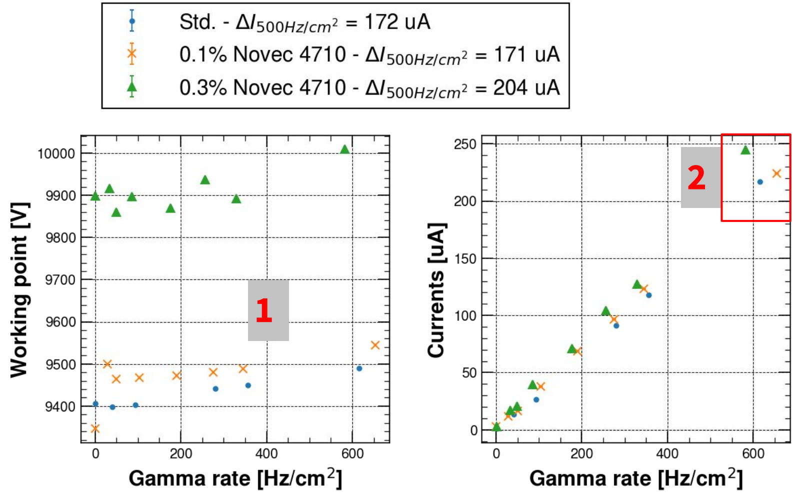
Two selected gases tested with muon beam and gamma background: **Novec 4710, Amolea 1224yd**

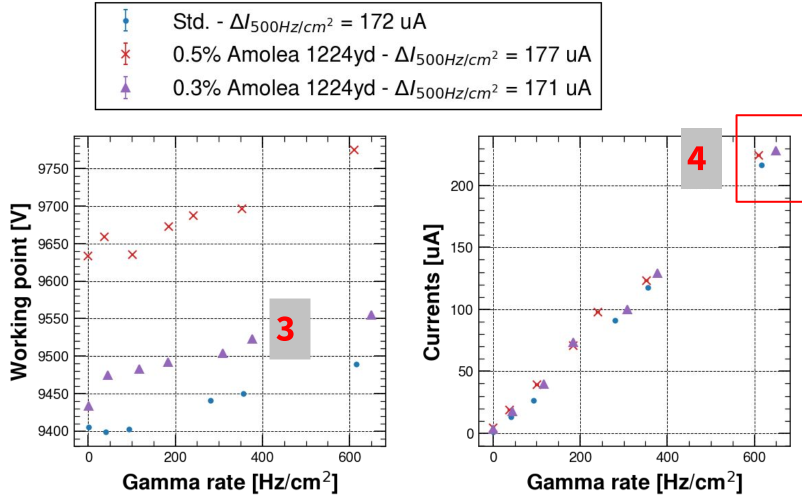
Novec 4710 **0.1%**, **0.3%**:

1. Working point similar to std. when used at **0.1%**
2. Currents slightly lower for **0.1%** → possibly due to lower charge per count at higher concentrations

Amolea 1224yd **0.3%**, **0.5%**:

3. Working point similar to std. gas mixture at **0.3%**
4. Currents at **0.3%** and **0.5%** similar to std. gas mixture





Impurities studies: setup and methodology development

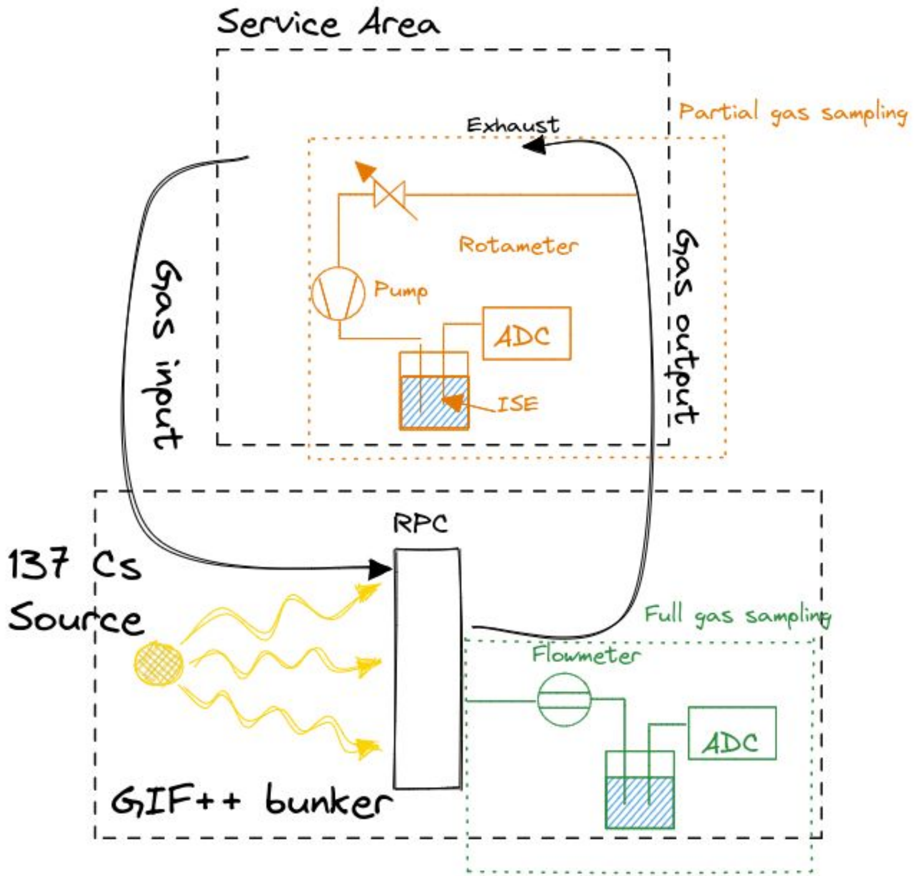
R-134a and **R-1234ze** break under electric field and gamma irradiation → **HF** production → detector inner surface possible damage

Studies on HF production

- **Gas** analyzed at the **output** of the detector irradiated and operated at working point with different gas mixtures
- **Ion Selective Electrode** technique employed: gas is sampled into a F- capturing solution, the **concentration** of **HF** is measured

Setup and methodology development

- **Partial** gas sampling or **full** gas sampling → Both have pros and cons
- **Optimization** of the existing **methods**: increase the **accuracy** of measurement by improving parameter **monitoring** and measurements **procedure**
- **Tests** on **hardware** components for optimal measurements: long lasting electrodes, mass flow meters, stirrers, etc.



Impurities studies

Impurities studies: HF production of different gas mixtures

Standard vs R-1234ze based gas mixture

- Comparison between **standard** and **R-1234ze/R-134a + CO2** gas mixture
- Detector operated at **w.p.** and **different** background **rates**
- **R-1234ze** gas mixture produced around 4 times more HF than std. gas mixture

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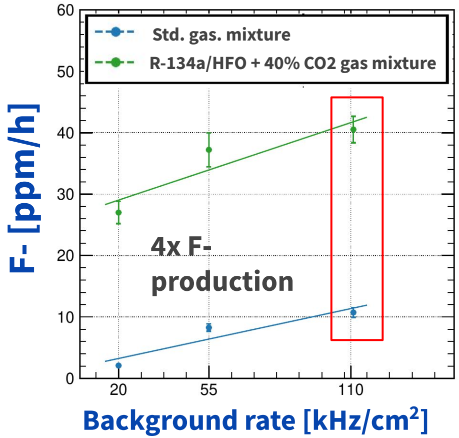
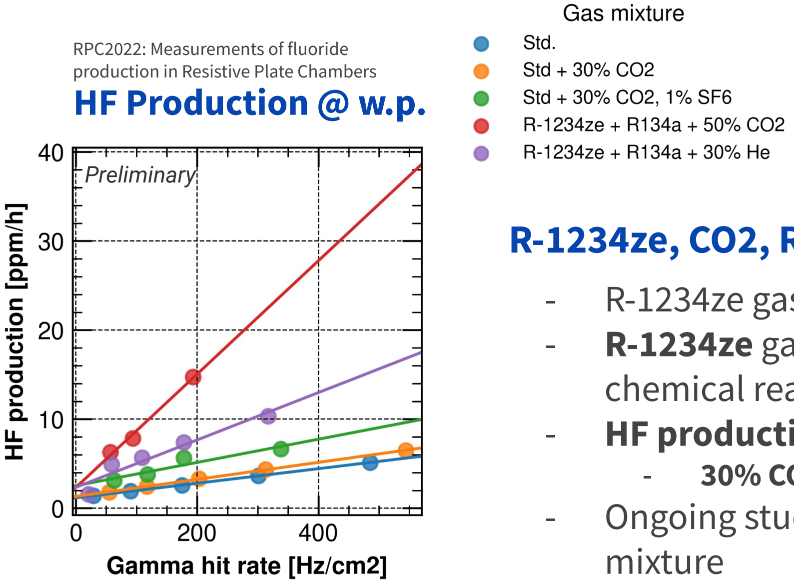
HF Production @ w.p.



RPC2022: Measurements of fluoride production in Resistive Plate Chambers

HF Production @ w.p.



R-1234ze, CO2, R-134a contributions to HF production

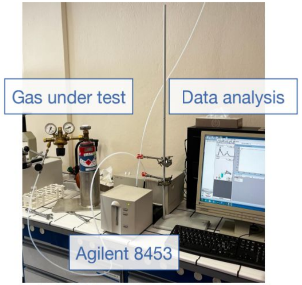
- R-1234ze gas mixtures and Std. + CO2 gas mixtures
- **R-1234ze** gas mixtures have the **highest HF production** → R-1234ze higher chemical reactivity
- **HF production** is **not proportional** to amount of **F-gases** in the mixture:
  - **30% CO2** + R-134a produces the **same amount** of HF as the **std.** gas mixture
- Ongoing studies to understand **correlation** between **HF** and gases in the mixture

UV absorption and water solubility studies

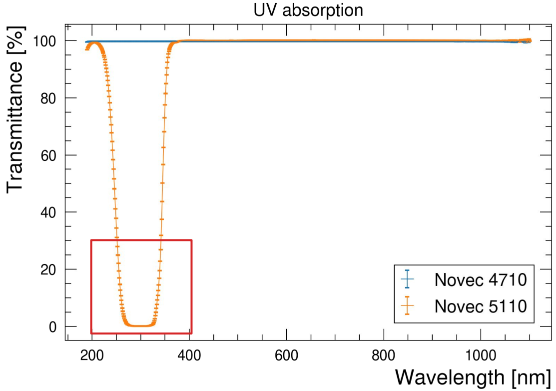
UV absorption of Novec 5110 and Novec4710

- Measurements done together with CERN Chemistry group
- **Novec 5110** shows a significant UV absorption in the UVB-UVA region (300-400 nm)
- **Novec 4710** doesn't show any significant absorption in UV region
- Further studies needed at lower wavelengths

UV-spectroscopy (CERN chemistry lab)



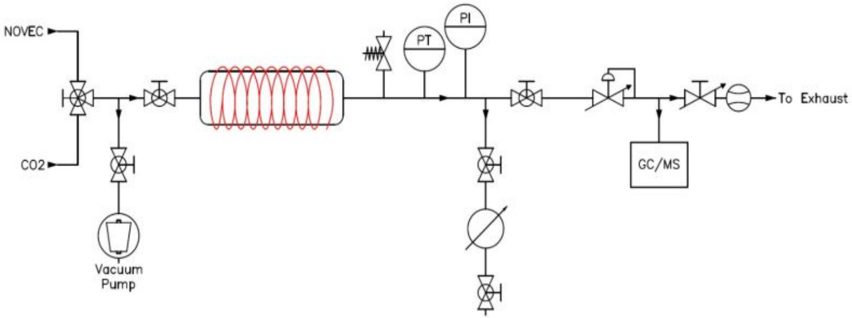
Thanks to B. Teissandier for helping in the measurements and providing the instrument



UV absorption of Novec 5110 and Novec4710

- Novec 4710 low GWP is due to its water solubility
- Bakelite RPC uses 40% RH humidity
- Novec 4710 + H2O → amide formation
  - ppb concentration
  - Solid at room temperature
  - Significant vapour pressure at 60 °C
- Ongoing tests in laboratory
  - Qualification of sub products formation
  - Quantification of amide formation at the temperatures used in RPC systems

Novec 4710 + humidified CO2 setup



Conclusions

Alternatives gas mixtures for RPCs at LHC

- **R-1234ze** studied in RPCs with different gas mixtures, **muon beam and gamma background**
- **Performance** are **not matching** the ones of the std. gas mixture → compromise between environment/safety/performance might be required
- **R-1234ze** effects still needs to be better addressed on the long term due to **higher currents** and **HF production**
- Environmental **subproducts** of **R-1234ze** to be better understood

Non fluorinated alternatives

- Several gas tested: N2, N2O, O2, Ne, He, CO2, Ar
- They cannot replace R-134a but **mitigate** its **consumption**
- **CO2 selected**: availability, price, good performance, known effects on other detectors
- **64% R-134a + 30% CO2 + 5% iso + 1% SF6** selected for LHC

Alternatives to SF6

- Lots of candidates that can be used as SF6 alternatives
- Most of **low-GWP** candidates are **highly reactive** in atmosphere → RPCs performance might be affected
- Tests ongoing to carefully evaluate the **environmental chemistry of new gases**
  - UV absorption
  - Reactivity with humidity
- **Novec 4710** and **Amolea 1224yd** showed good performances with muon beam and gamma background
- **Amolea 1224** contains Cl atom: may severely affect gas systems and detector operation
- **Novec 4710** not mean to be used in wet environments → dedicated setup to quantify the production

Thank you

GWP calculation

GWP for a single gas is well defined: it is a measure of how much energy the emissions of **1 ton** of a gas will absorb over a given period of time, relative to the emissions of 1 ton of carbon dioxide (CO2)

Gas mixture is expressed in fractions of normal volume → proportional to number of moles → molecular weight

GWP of gas mixture: $(\Sigma GWP_i * M_i * f_i) / M_{CO2,}$ where M is molecular mass and f the amount of the gas in the mixture

Example:

Suppose RPCs are operated with 1000 ln/h of CO2. After one year the tons of CO2 are:

1000 ln/h * 8760 h / 22.4 l/mol * 44 g/mol = **17.2 tons**

Suppose RPCs are operated with 70% R-134a and 30% CO2. If we simply do the proportion we would get: 1430 * 0.7 + 1* 0.3 = **1001** of GWP → *wrong estimation*

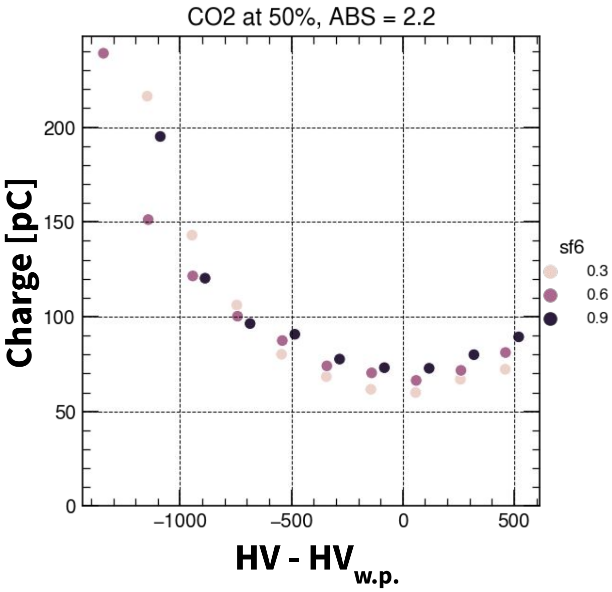
After one year the equivalent tCO2e are:

- **CO2**: 300 ln/h * 8760 h / 22.4 l/mol * 44 g/mol = 5.2 tons
- **R-134a**: 300 ln/h * 8760 h / 22.4 l/mol * 102 g/mol * 1430 = **39.9 kTons**

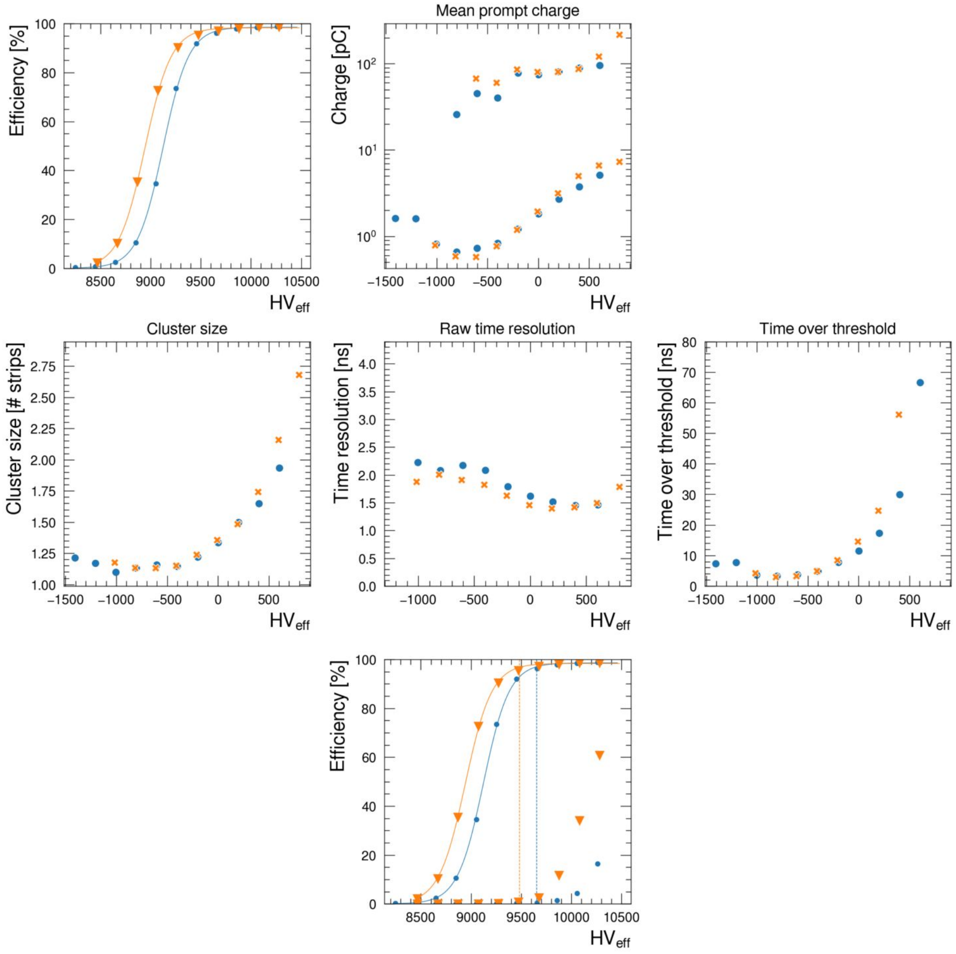
⇒ 39.9 ktCO2e ⇒ GWP_e = 39.9 ktons / 17.2 tons = **2320 GWP_e**

This results are because detectors and gas systems are operated using **normal volume units** and not mass of the gases

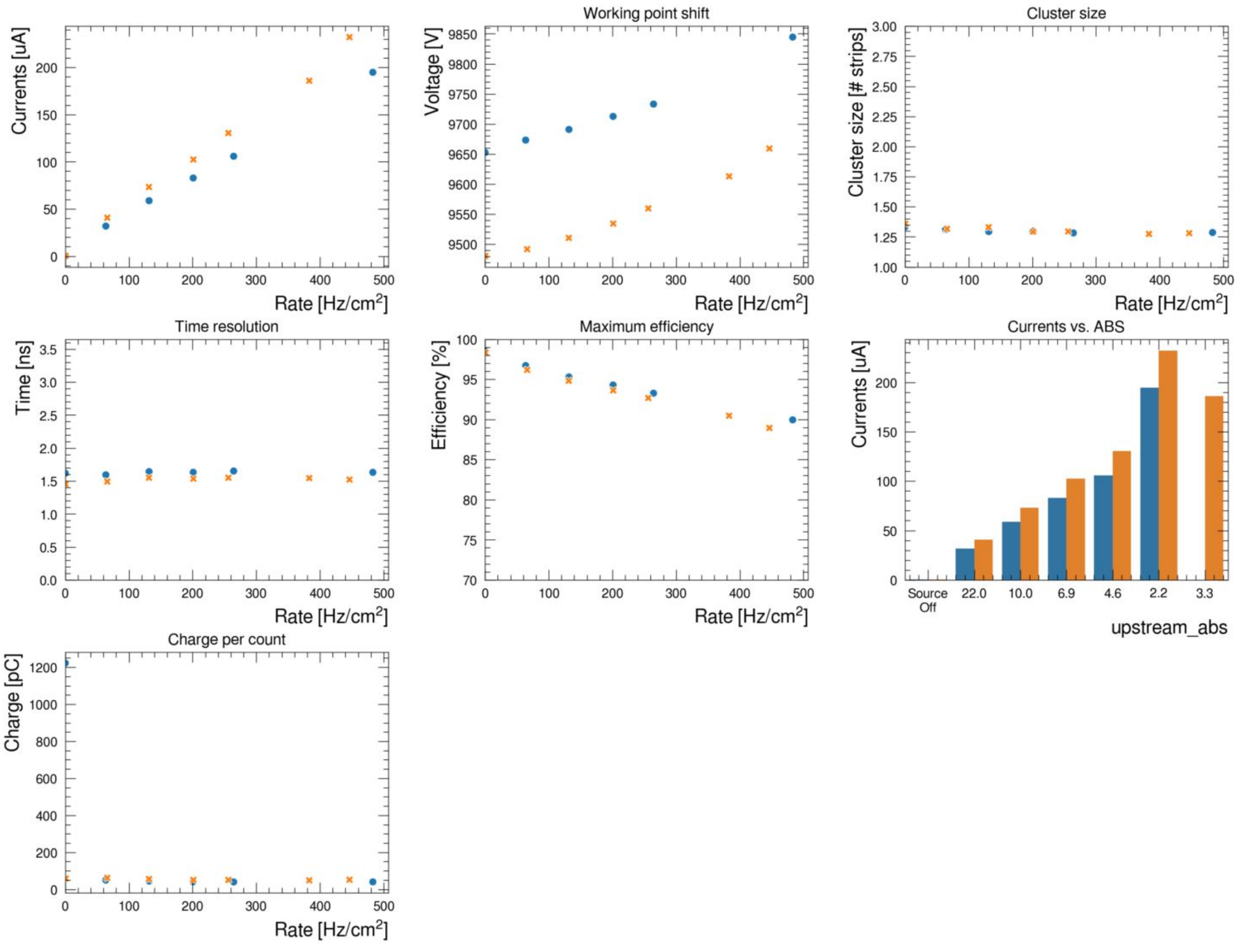
Gamma charge per count



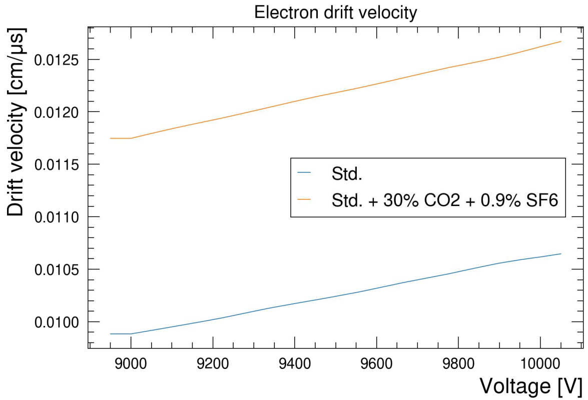
Std vs Std + 30% CO_2 selected gas mixture: source off

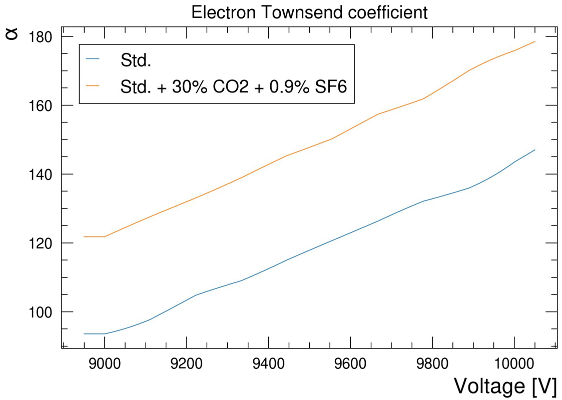


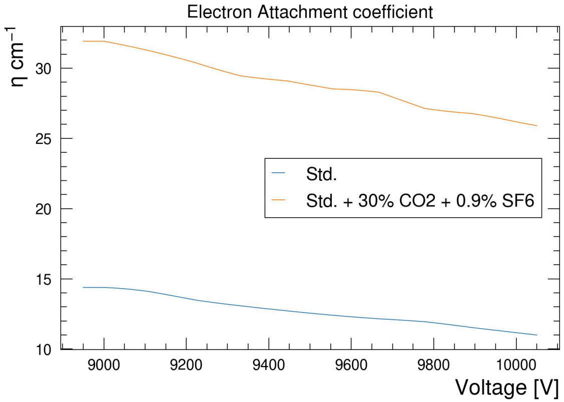
Std vs Std + 30% CO_2 selected gas mixture: source off

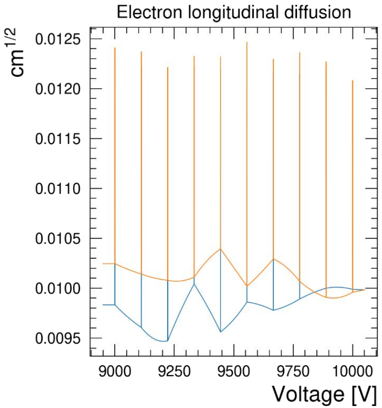


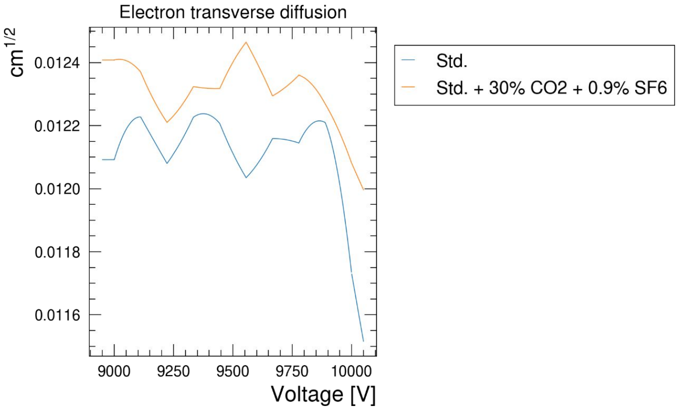
Gas coefficients from simulation











HFO flammability tests

Safety concerning HFO usage

- R-1234yf classified as mildly flammable → Focus on R-1234ze

R-1234ze + i-C4H10 + 40% RH flammability test conducted:

ISO 1056 standard flammability test (detachement + flame propagation criteria) performed by external company

Results

- *Mixture with **1% i-C4H10** + R-1234ze **is flammable***
- Water vapour plays an important role

HFOs alone + i-C4H10 is flammable → Effects of the CO2 on the mixtures to be understood/checked

illustration of a flame detachment with flame propagation over a distance of at least 100 mm as criterion for flammability

test no.	Iso-butane fraction in test gas mixture in mol%	fraction of test gas mixture of iso-butane and HFO1234ze in mol%	fraction of air including 2.25 mol% water in mol%	reaction
9	**6.2**	**15.0**	**85.0**	+
10	6.0	20.0	80.0	-
11	**4.2**	**13.0**	**87.0**	+
12	**3.1**	**10.0**	**90.0**	+
13	**2.2**	**13.0**	**87.0**	+
14	1.1	13.0	87.0	-
15	**1.0**	**10.0**	**90.0**	+
16	0.0	12.0	88.0	-
17	0.0	11.0	89.0	-
18	0.0	10.0	90.0	-
19	0.0	9.0	91.0	-

https://edms.cern.ch/document/2463340/1