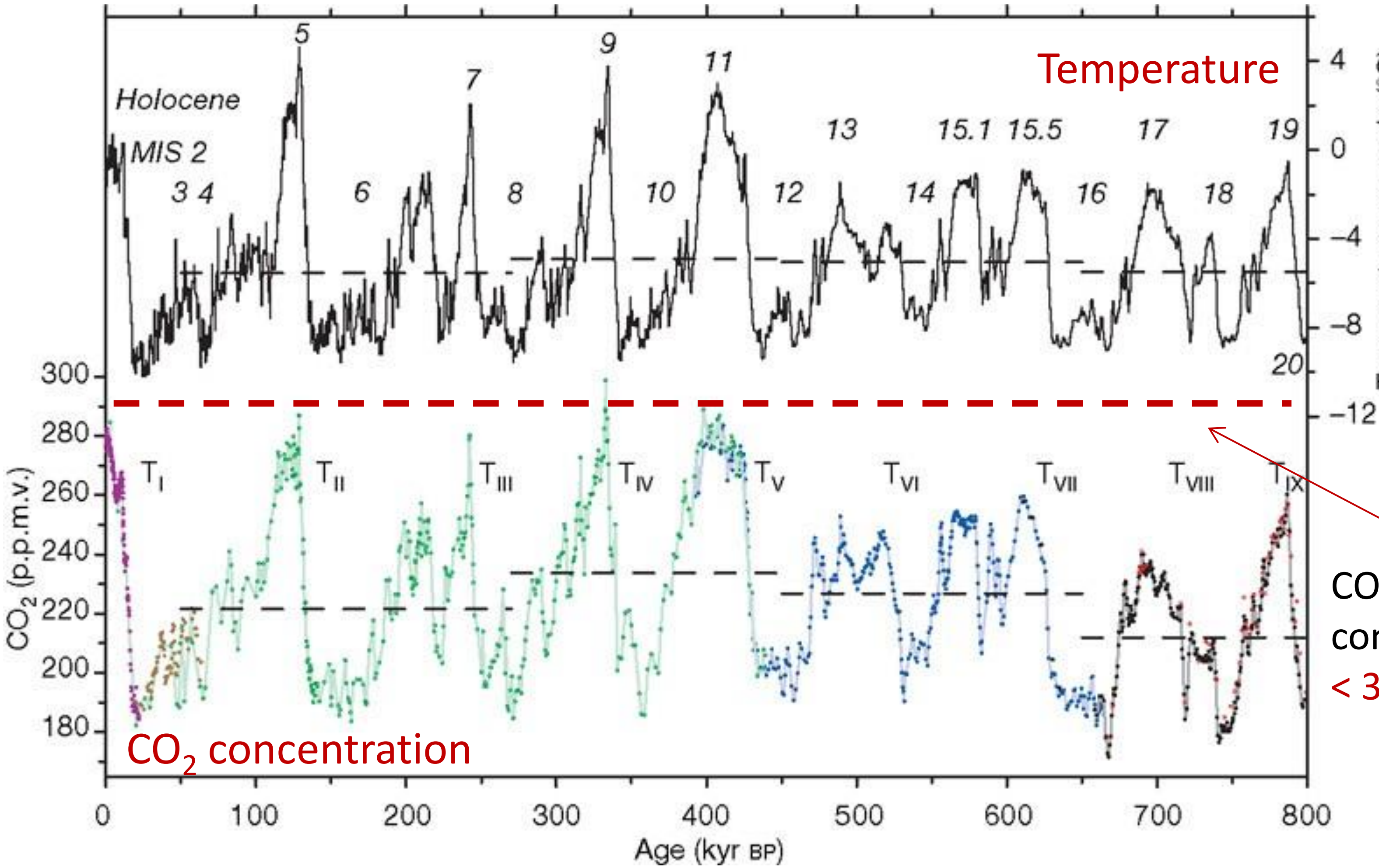
Istituto Nazionale di Fisica Nucleare

Effetto serra provocato dai gas, strategie di mitigazione, e studio di gas eco-sostenibili

Marcello Abbrescia

Corso di formazione INFN: preparazione ed utilizzo delle miscele gassose per i rivelatori di particelle
Sezione di Roma Tor Vergata, 21-23 Ottobre 2024

Temperature and CO_2 concentration: last 800.000 years

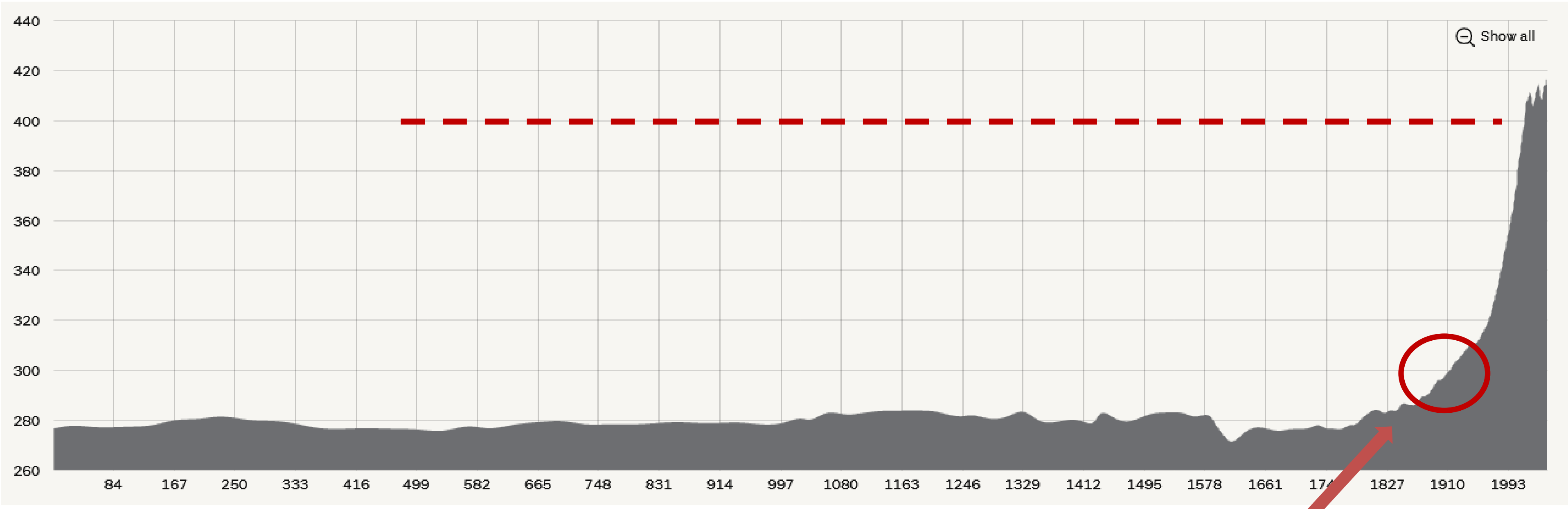


CO_2 concentration
< 300 ppm

Lüthi, D., *et al.* High-resolution carbon dioxide concentration record 650,000–800,000 years before present. *Nature* **453**, 379–382 (2008). https://doi.org/10.1038/nature06949

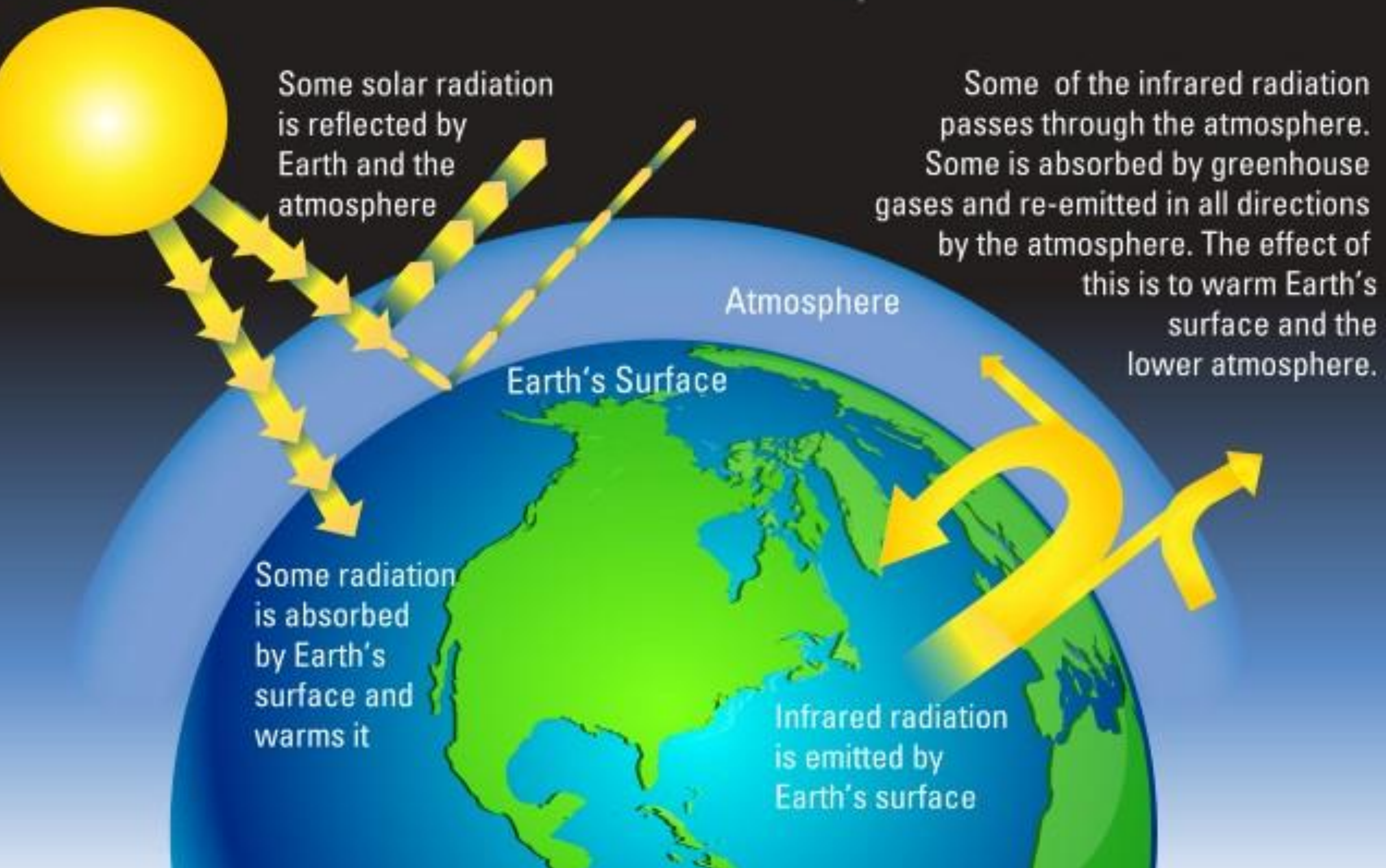
CO_2 concentration: last two thousand years

We know that atmospheric CO_2 has ranged between 172 and 300 part per million (ppm) for the past 1 million years.



The first time in human history that atmospheric CO_2 exceeded 300 ppm was about the time the **Titanic sank (1912)** in the North Atlantic Ocean. Now, the concentrations of CO_2 stays constantly above 400 ppm.

THE GREENHOUSE EFFECT



The Global Warming Potential

- Not only CO2 contributes to the greehouse effect. There are other gases (and families of gases) that contribute: H_20, CH4, CFC, HCFC, NO_x, SO_x

To compare various gases, the Global Warming Potential was introduced:

- it is a measure of how much energy the emission of 1 ton of a gas will absorb over a given period of time, relative to the emission of 1 ton of carbon dioxide (CO_2).
- The larger the GWP, the more that a given gas warms the Earth compared to CO_2 over that time period.
- The time period usually used for GWPs is 100 years(*).

(*) Some gases, once arrived in the high atmosphere, dissociate, producing by-products that may or may NOT be greenhouse gases: therefore the lifetime a a gas species MUST be considered when computing its GWP

GWP for various gases

Table 8.4 Greenhouse gas properties related to global warming.

Gas	Residence time / y[a]	Relative instantaneous radiative forcing	Global warming potential (GWP)[c]
CO_2	50–200[b]	1	1
CH_4	12	43	23
N_2O	115	250	296
CFC-11	45	15 000	4 600
CFC-12	102	19 000	10 600
HCFC-22	12	13 000	1 700
CCl_4	35		1 800
C_2F_6	10 000		11 900
SF_6	3 200		22 200

[a] Most of the atmospheric lifetime values are taken from Additional Reading 1.

[b] Reported residence time values for carbon dioxide are highly variable. Differences are associated with the way in which oceanic uptake is measured, particularly whether the surface layer or the entire ocean is considered in the calculation.

[c] GWP values are obtained by integration over a 100 y period. Obtained from Blasing, T.J. and S. Jones, *Current greenhouse gas concentrations*, <http://ediac.esd.ornl.gov/pns/current_ghg.html> (February 2004). These values relate to direct effects; interactions of CFCs with ozone in the lower stratsophere may reduce the amount of radiation into the lower atmosphere, contributing to a cooling effect. The GWP values would be correspondingly reduced.

The Relative instantaneous radiative forcing, is independent of the lifetime of the gas in the atmoshpere.

The GWP take also into account the lifetime of the gas in the atmoshpere.

GWP for gas mixtures

Note that the GWP refers to **equal masses of different gases** (for instance 1 kg of SF_6 with respect to 1 kg of CO_2).

In HEP we usually refer to volumes of gases: for instance, «we are flushing the gas at 1 liter/hour»

A gas mixture often used in gaseous detectors of HEP is:

$C_2H_2F_4$ (TFE)/SF_6 98/2 in volume!

So, how do we compute the GWP of such gas mixture? We have to use the gas densities, which, in this case, are:

$C_2H_2F_4$ (TFE): 4,68 g/l (GWP=1430)

SF_6: 6.61 (g/l) (GWP=22800)

1 liter of this mixture has:

- 0.98 liters of TFE → 4,5864 g of TFE

- 0.02 liters of SF_6 → 0,1322 g of SF_6

GWP for gas mixtures

So, the total weigth of 1 liter of this mixture is 4,7186 g.

Its GWP, for 4,7186 g is:

4,5864 x 1430 (TFE) + 0,1322 x 22800 (SF_6) = 9572,512 for 4,7186 g

So, 1 gram (unit of mass) of this mixture has a

GWP = 9572,512/4,7186 = 2028

The GWP is always referred to the unit of mass.

The CO_2 equivalent

There is a way to compute the total amount of contribution to the greenhouse effect of a given quantity of gas injected in the atmosphere: the CO_2 equivalent.

- the GWP is an intensive quantity

- the CO_2e equivalent is an extensive quantity.

Let's imagine to inject into the atmoshpere 2 liters of the previous gas mixture:
2 liters of this mixture (using the densities) has:

1.96 liters of TFE = 9,1728 g of TFE

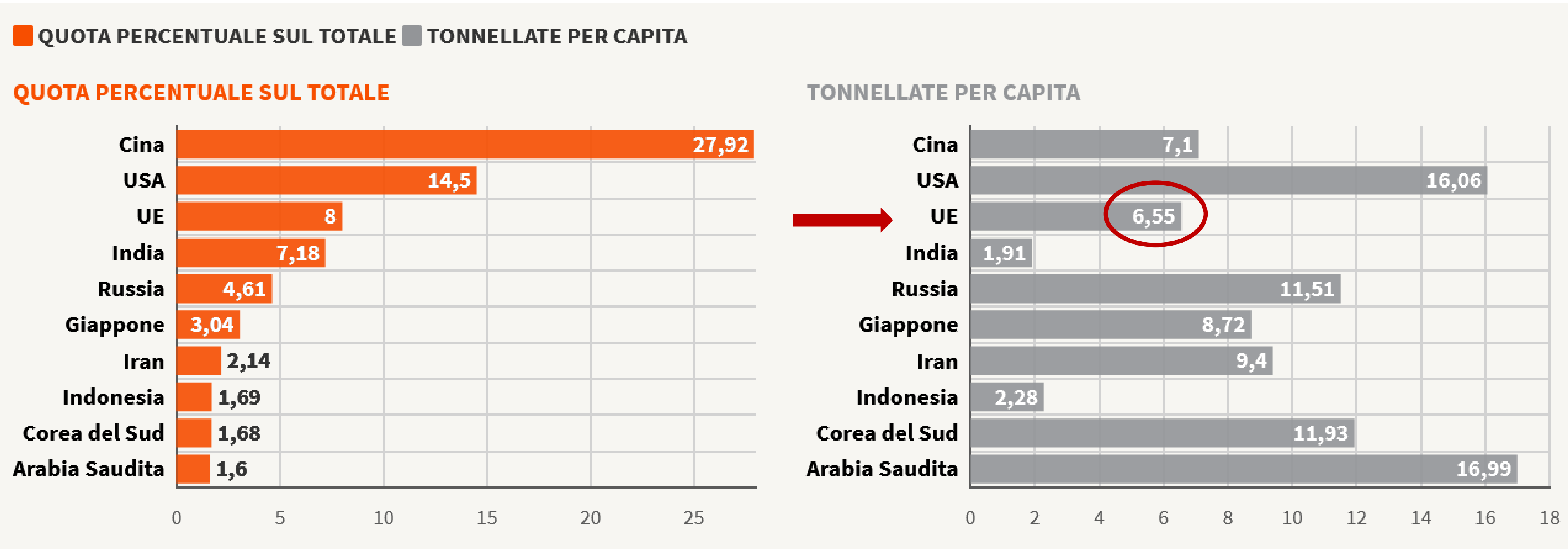
0.04 liters of SF6 = 0,2644 g of SF6

The CO_2e therefore is:

9,1728 x 1430 (TFE) + 0,2644 x 22800 (SF_6) = 19145 g

Injecting into the atomosphere 2 liters of this mixture is equivalent to inject about 19 kg of CO_2!

Amount of CO_2 produced



An EU citizen injects about 6.55 tons/year of CO_2 equivalent in the atmoshpere

An inhabitant of Geneve injects about 13 tCO_2e per year → 2.600.000 tCO_2e per year in total (fonte: www.geneva.ch)

Global CO_2 emission from human activity

Most human-caused emissions of CO_2 into the atmosphere are from burning fossil fuels that had long been stored in the crust of the Earth. A small part of the fossil fuel total is from new cement usage.

86%

34.4 $GtCO_2$/yr

Fossil fuel emissions

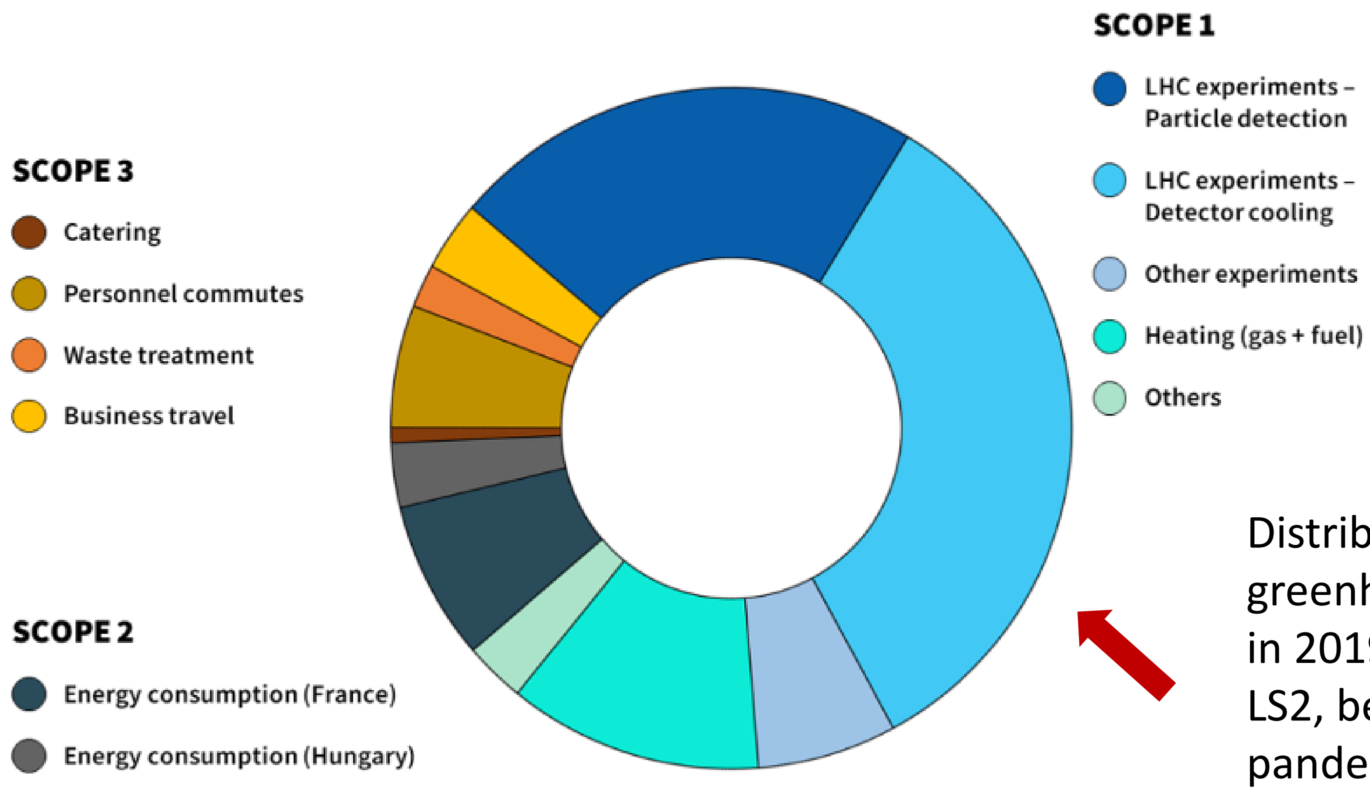
14%

5.7 $GtCO_2$/yr

Emissions from land use change

(mostly deforestation)

CERN emissions of GHGs



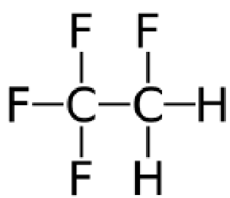
Distribution of CERN's greenhouse gas emissions in 2019 (representative of LS2, before the COVID-19 pandemic) (Image: CERN, 10 January 2022)

	2021	2022
Scope 1	123174	184173
Scope 2	56382	63161
Scope 3	105843	113930

Emissions in tCO_2e

GHGs for particle detection at LHC experiments

GHGs are used in several gaseous particle detectors due to their characteristics suitable for optimal detector performance AND long term operation

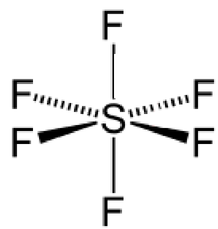


$C_2H_2F_4$

GWP 1430

Containment of charge
Rate capability

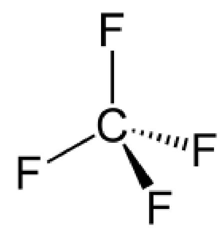
Resistive Plate Chamber (RPC)



SF_6

GWP 22800

- **Electronegative: limiting charge development**



CF_4

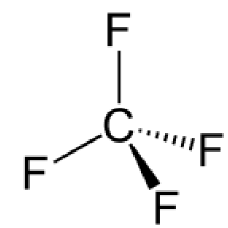
GWP 7390

- **time resolution**

Gas Electron Multiplier (GEM)

- **Mitigation of aging phenomena**

Cathode Strip Chamber (CSC)
Multi Wire Proportional Chamber (MWPC)



C_4F_{10}

GWP 8860

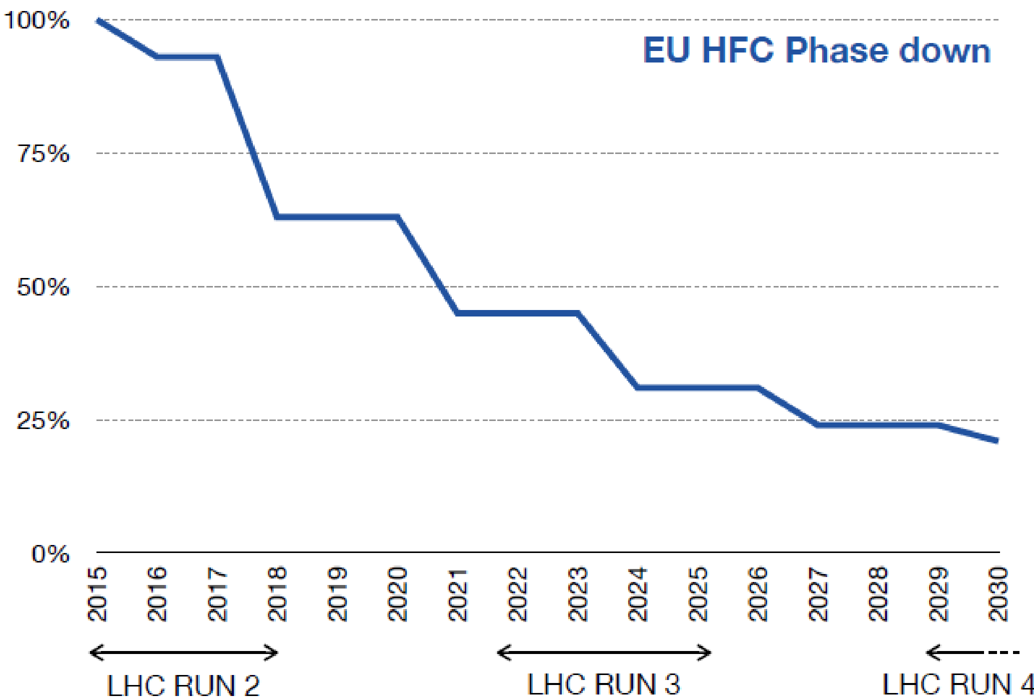
- **Cherenkov radiator**

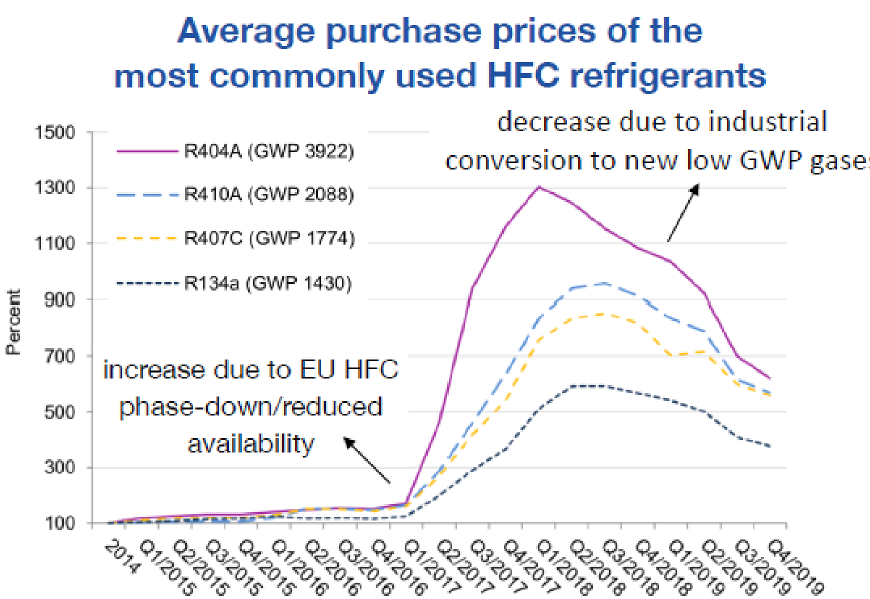
Ring-imaging Cherenkov detector (RICH)

- 30 years ago we had to get rid of Ozone Depletion gases
- There was not the feeling about GHGs → many detectors were designed from the beginning to be used with GHGs

The EU HFCs phase down policy

- **Limit 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.
- **Ban the use** of F-gases in many new types of equipment where less harmful alternatives are widely available.
- **Prevent 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.

A green choice

The problem of GHGs is being addressed worldwide.

EU is progressively banning greenhouse gases:

- but they are still allowed for research applications (like in HEP)
- nevertheless the green choice of the INFN, CERN communities (and others) was to switch **NOW** to ecofriendly gas mixtures

The problem: use of Greenhous gases in HEP

We need to replace:

- ✓ $C_2H_2F_4$ = R134a = TFE mainly used in RPCs
- ✓ SF_6 mainly used in RPCs
- ✓ CF_4 used in CSCs, GEMs, RICH, etc.

It's not a problem concerning just the RPC community

with more ecological gases, namely with a much lower Global Warming Potential.

Difficult problem: gases are the core of gas-filled detectors.

We also need:

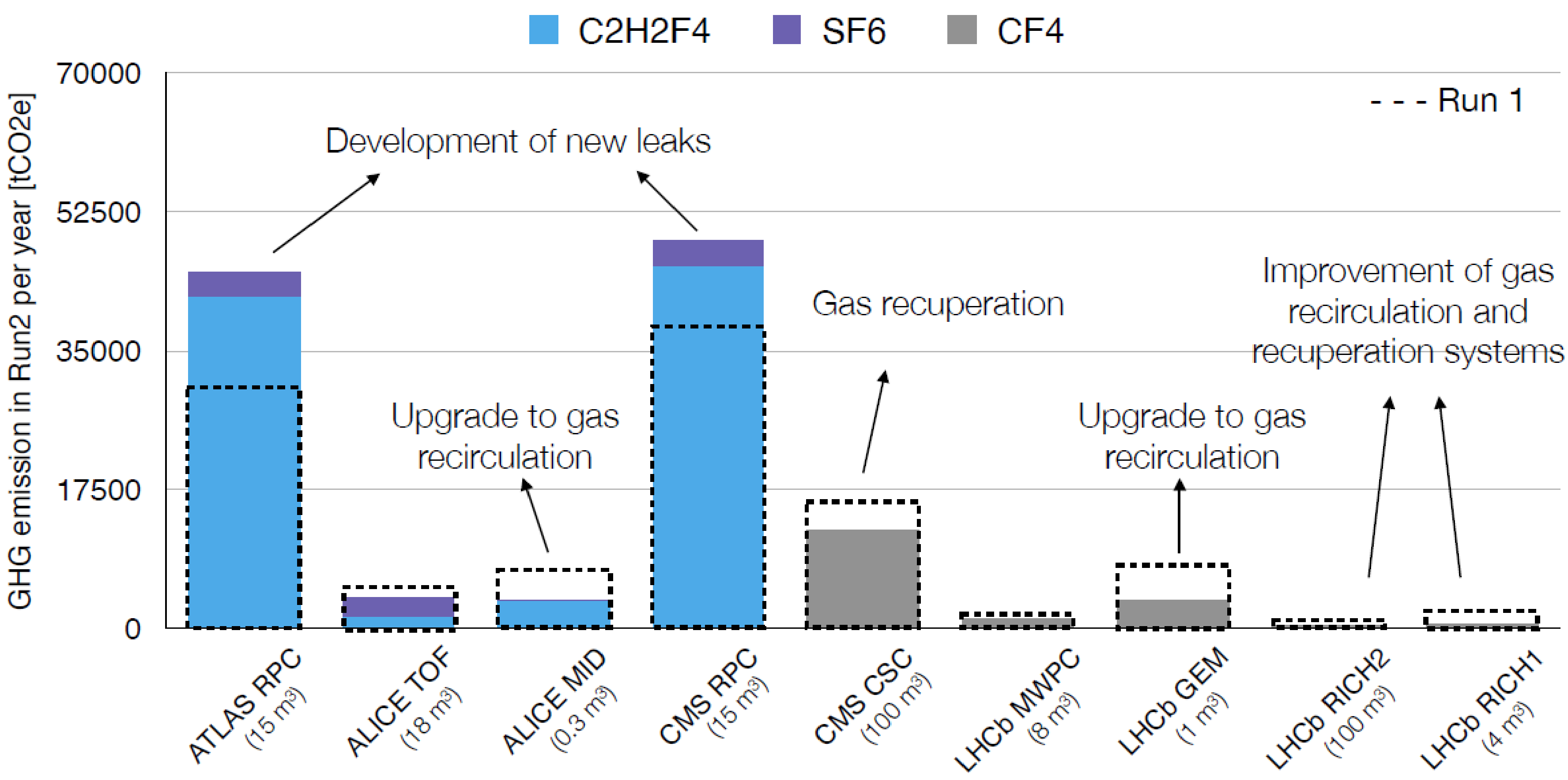
- to get the same performance
- not to change the electronics and HV (for existing systems)

➢ HEP experiments, present and future, last several (dozens) of year
→ A good performance must be mantained for an adequate period of time
→ Aging tests are needed as well.

Of course we can also re-circulate the gases used, after purifying them, and reduce leaks

See talk by R. Guida

The use of GHGs at the LHC experiments



- 40% GHG emissions from Run 1 to Run 2 excluding from the calculations ATLAS and CMS RPC systems

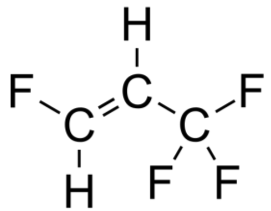
- ATLAS and CMS RPC systems: +35% increase of GHG emissions due to development of new leaks

The importance of collaborative effort

- All high energy experiments (ALICE, ATLAS, CMS, LHCb, etc.) and the CERN gas group (CERN EP-DT) started, already several years ago, an intense R&D program to find suitable gas mixtures.

- Practically all research trendlines concentrate around the idea of replacing:
  $C_2H_2F_4$ (GWP=1430)→ $C_3H_2F_4$ze (GWP=4)
  \+ CO_2 (GWP=1) or He

EEE



- ✓ $C_3H_2F_4$ (here indicated as HFO for short) is the molecule most similar to TFE but with low GWP
- ✓ CO_2 (or He) are essentially added to reduce the operating voltage.

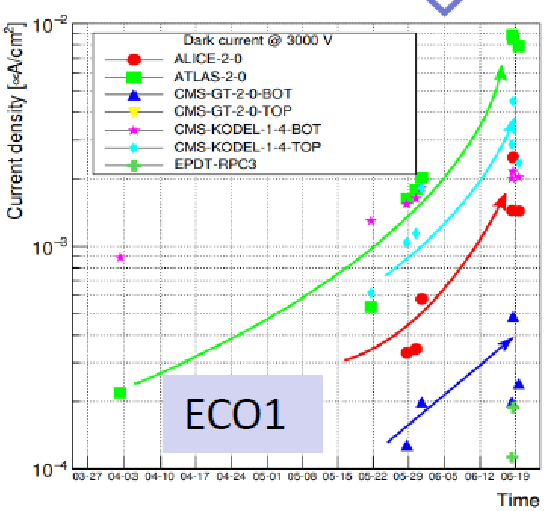
The RPC EcoGas@GIF++ is a Collaboration **transversal to ALICE, ATLAS, CERN EP-DT, CMS**, **and LHCb** willing to put together expertise and resources in order to test potential candidates of eco-friendly gas mixtures with different detectors and electronics.

The RPC ECOGas@GIF++ timeline

2018 — RPC EcoGas@GIF++ Collaboration

Assessment of the possibility of using HFO in place of TFE
→ ECO1, ECO2 and ECO3

Studies in different laboratories
Setup of the system at GIF++ and first HFO/CO_2 based gas mix. under irradiation



ECO1 discarded due to high current increase after ~20 mC/cm^2 integrated charge

AidaInnova Startup

2021

2021 testbeam — 2022 testbeam — 2023 testbeam — 2024 testbeam

Irradiation campaign for aging studies

Performance baseline

Performance comparison

Std, **ECO2** and **ECO3** mixtures tested under irradiation. First results in papers *[1][2][3]*.

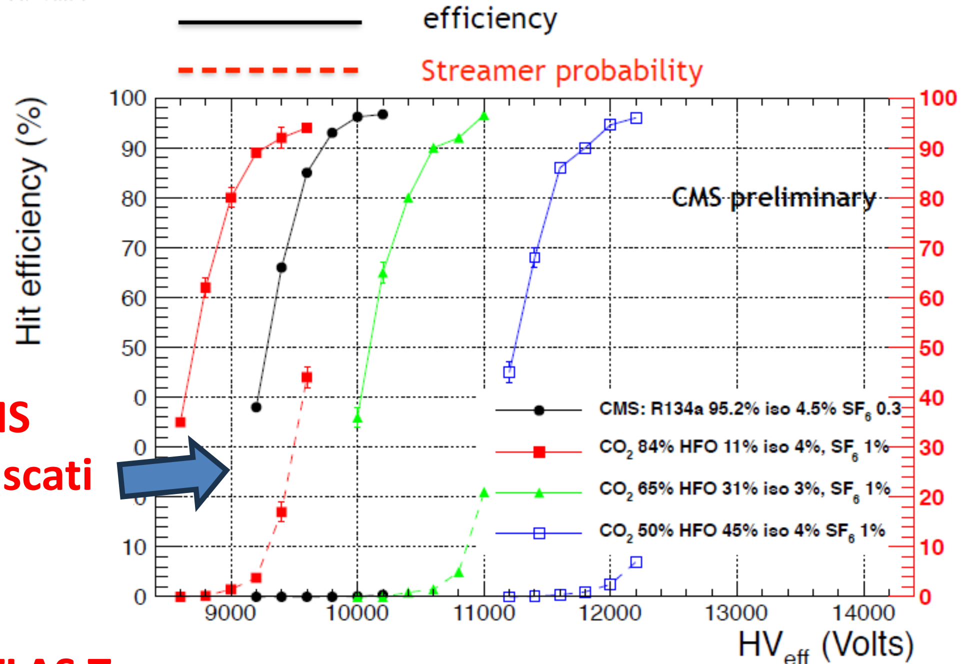
[1] "High-rate tests on resistive plate chambers operated with eco-friendly gas mixtures", 2024 Eur. Phys. J. C.

[2] "Performance of thin-RPC detectors for high rate applications with eco-friendly gas mixtures", 2024 Eur. Phys. J. C.

[3] "Preliminary results on the long term operation of RPCs with eco-friendly gas mixtures under irradiation at the CERN Gamma Irradiation Facility" – submitted to EPJplus.

Tests at the various home-labs

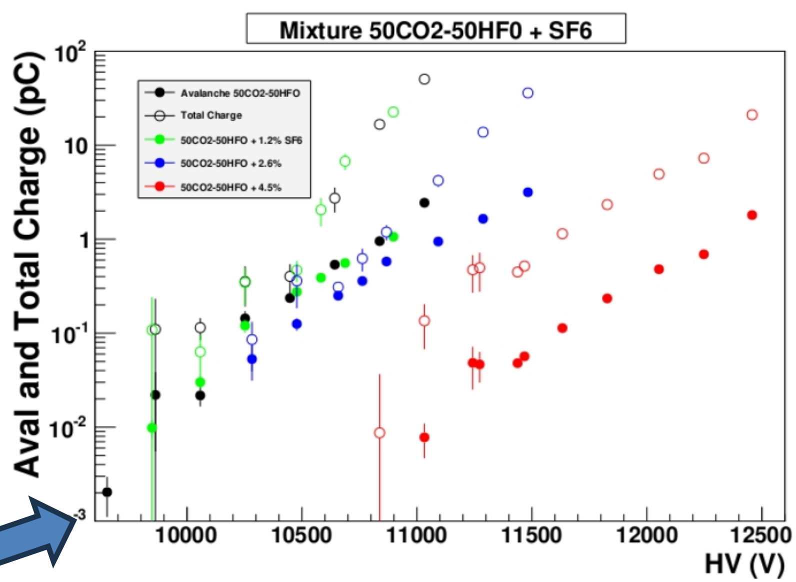
CMS Frascati

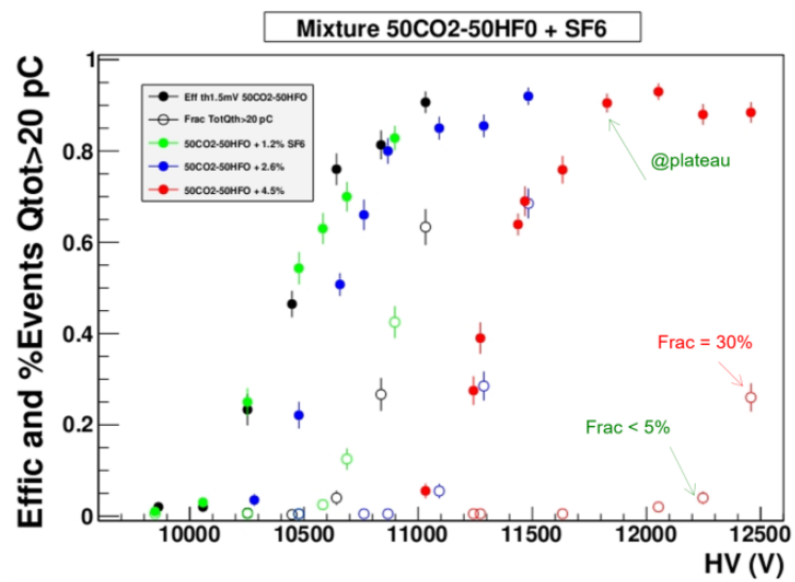


Preliminary results of Resistive Plate Chambers operated with eco-friendly gas mixtures for application in the CMS experiment, JINST 11 C09018 (2016)

ATLAS Tor Vergata

B. Liberti, “Recent results on environmental-friendly gas mixtures for ATLAS RPC”, 66th INFN Eloisatron workshop: New gas mixtures for RPC and MRPC detectors

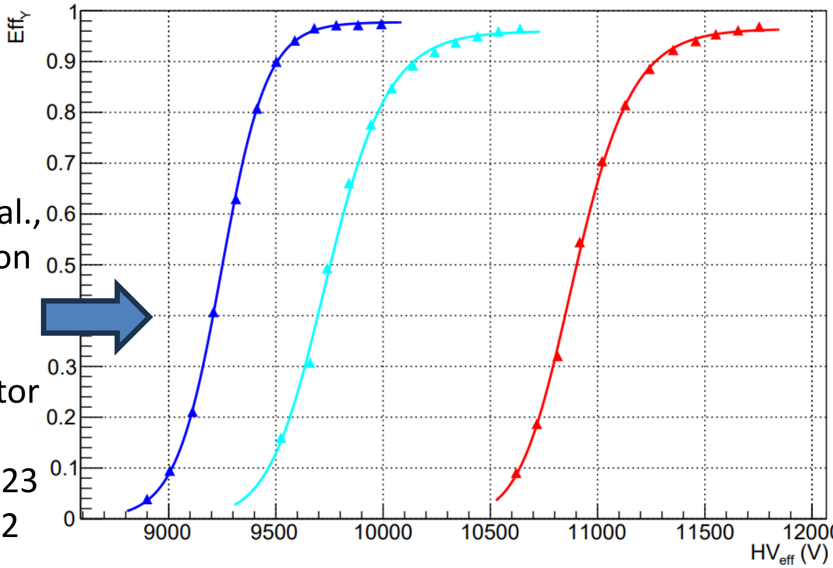




Tests at the various home-labs

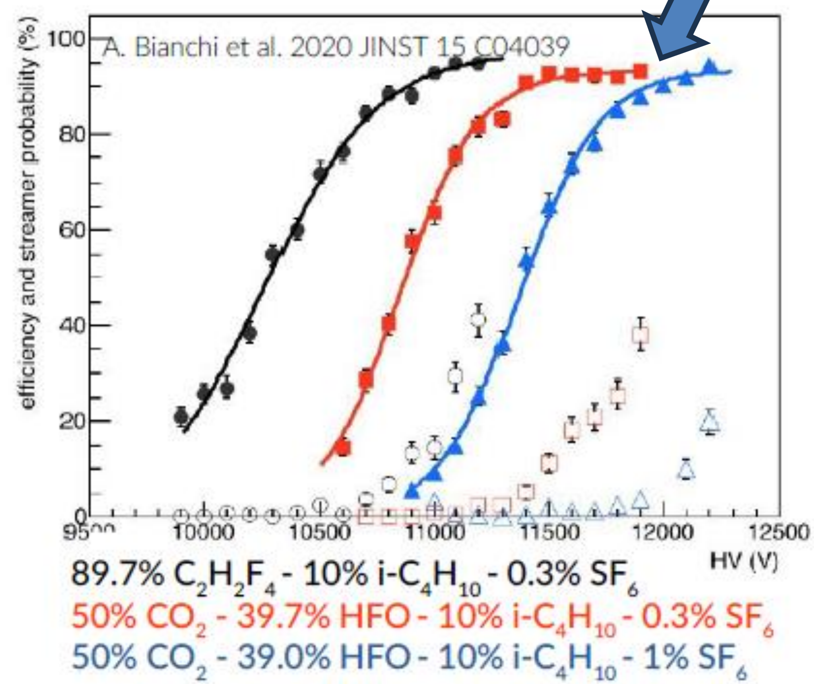
LHCb/SHip Bari

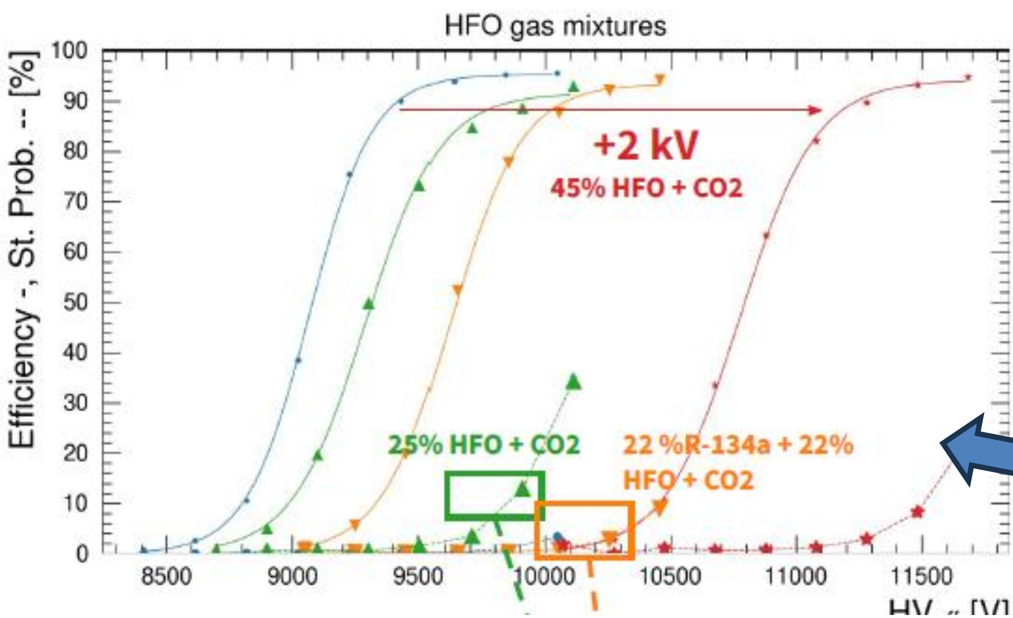
R. Albanese et al., RPC-based Muon Identification System for the neutrino detector of the SHiP experiment, 2023 JINST 18 P02022



- ▲ 95.2% R134a/ 4.5% iC_4H_{10} / 0.3% SF_6 (standard)
- ▲ 60% CO_2/ 35% HFO/ 4.5% iC_4H_{10} / 0.5% SF_6
- ▲ 69.5% CO_2/ 25% HFO/ 5% iC_4H_{10} / 0.5 % SF_6

ALICE Torino





CERN EP-DT

From: G. Rigoletti, "Environment friendly gas mixtures for Resistive Plate Chambers", 66th INFN Eloisatron workshop: New gas mixtures for RPC and MRPC detectors

Experimental set-up @GIF++

- Three gas mixtures identified, with various concentrations of HFO and CO_2.

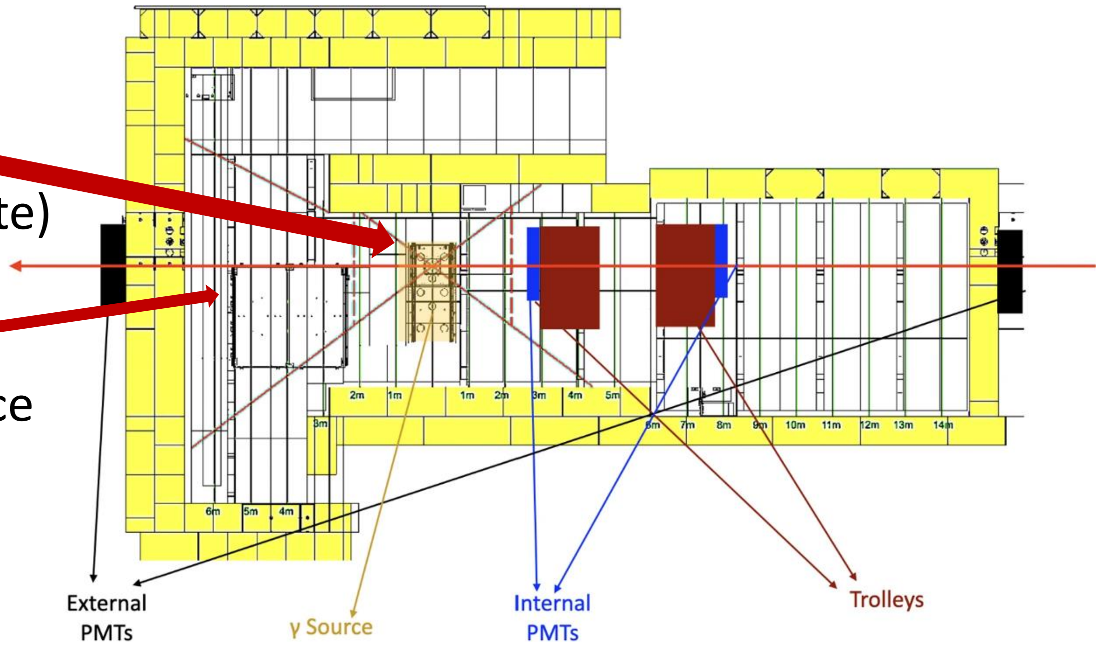
ECO1: 45% HFO / 50% CO_2 / 4% iC_4H_{10} / 1% SF_6
ECO2: 35% HFO / 60% CO_2 / 4% iC_4H_{10} / 1% SF_6
ECO3: 25% HFO / 69% CO_2 / 5% iC_4H_{10} / 1% SF_6

GWP reduced by 1/3 w.r.t. the std mixture!

Attention focussed on ECO2 and ECO3 because of the good stability and performance demonstrated in home-labs tests.

Aging tests performed at GIF++

- 12.5 TBq 137Cs source
- ✓ to generate background (high rate)
- ✓ to accelerate aging processes
- 100 GeV muon beam
- ✓ to measure detector performance



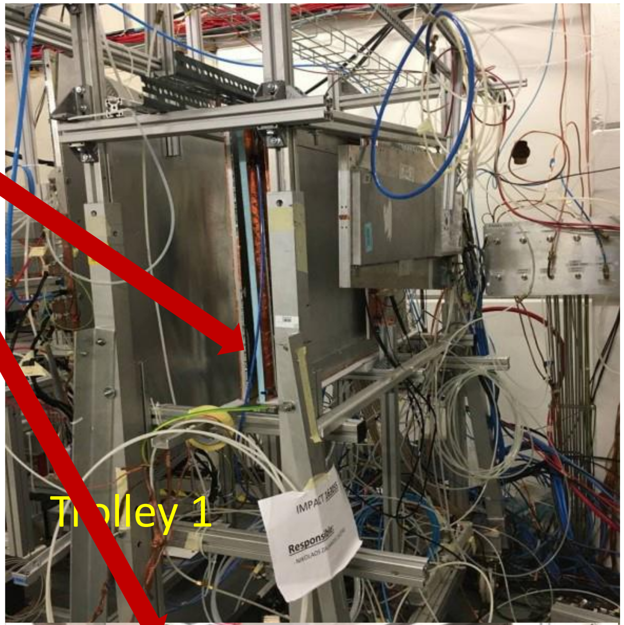
Detectors @ GIF++

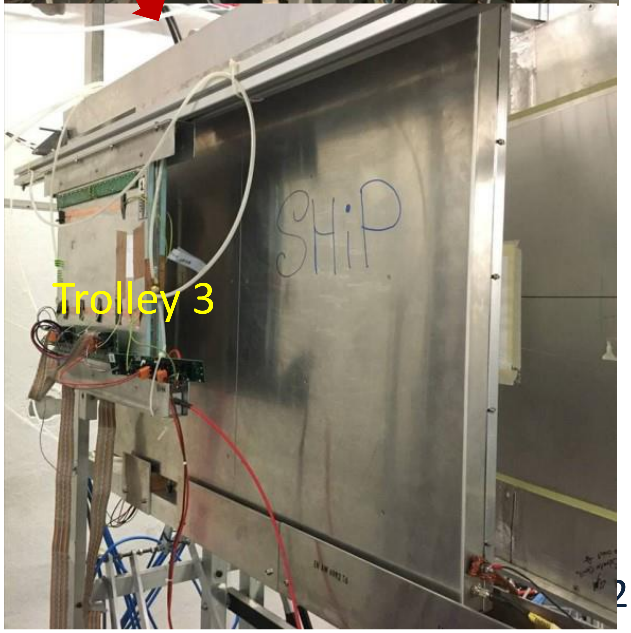
- Various detectors, mounted on two trolleys, equipped with various electronics.

→ Help in disentagling common observed effect from effects specific of ONE detector

RPC	Gap thickness	Electronics
ALICE	2mm	FEERIC + TDC
ATLAS	2mm	Digitizer
CMS	2mm – double gap	CMS FEB + TDC
CMS upgrade	1.4mm – double gap	CMS FEB + TDC
EP-DT	2mm	Digitizer
LHCb/SHiP	1.6mm	FEERIC + TDC

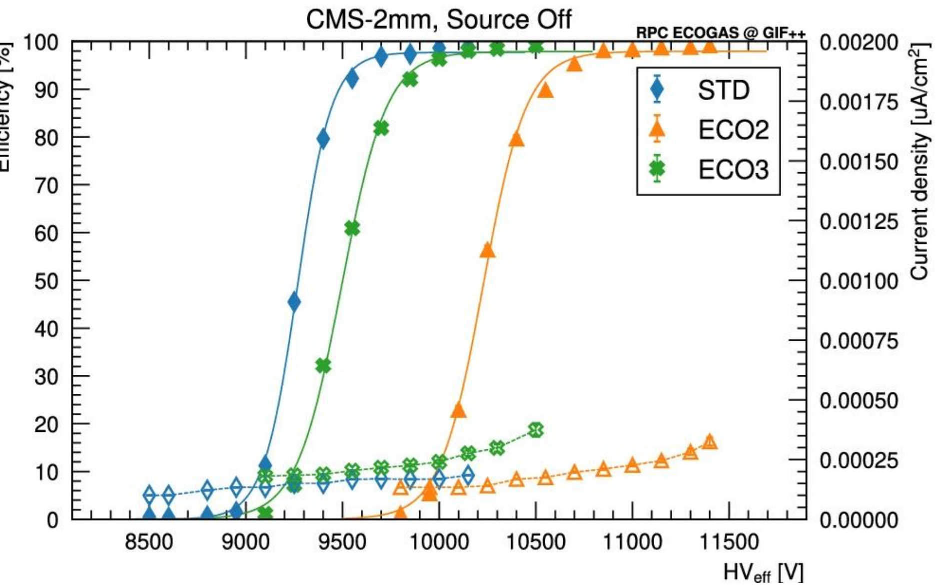
- The results presented here refer particularly to the detectors equipped with TDC

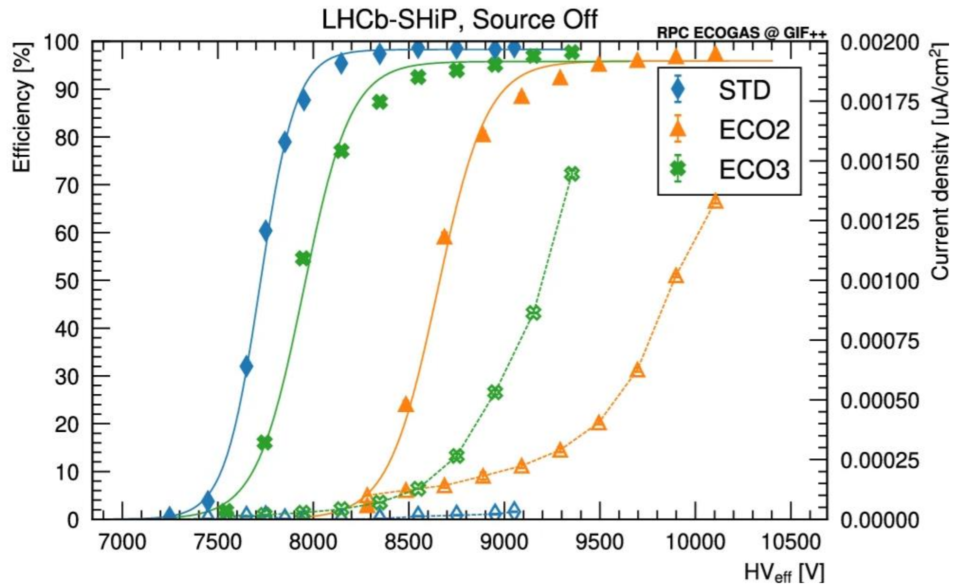




Determination of baseline perfomance

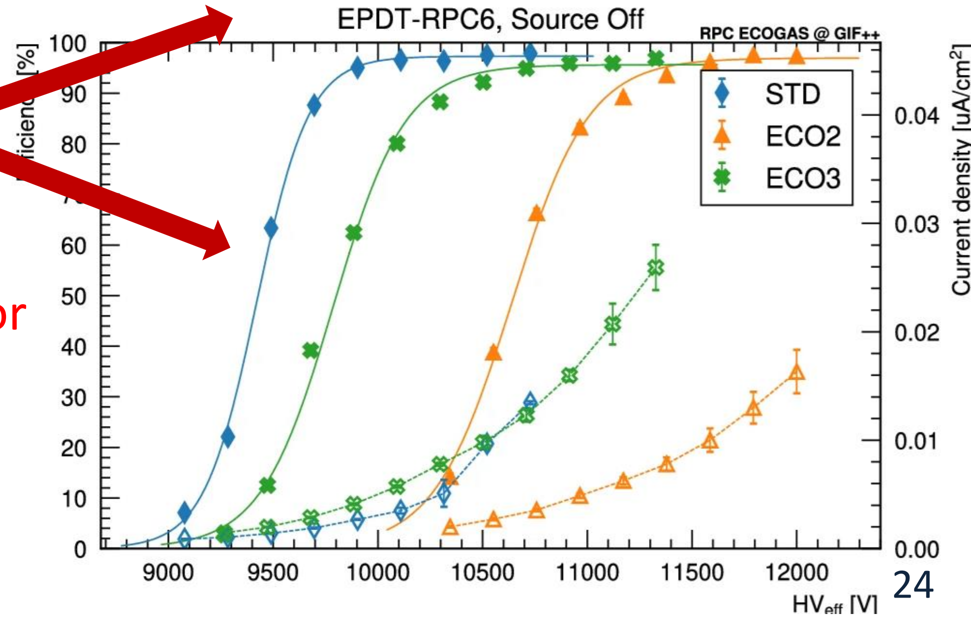
Source OFF, 2021 data





Fit of efficiency curves with a sigmoid:

$$\mathcal{E}(HV_{\text{eff}}) = \frac{\mathcal{E}_{\max}}{1 + e^{-\beta(HV_{\text{eff}} - HV_{50})}}$$



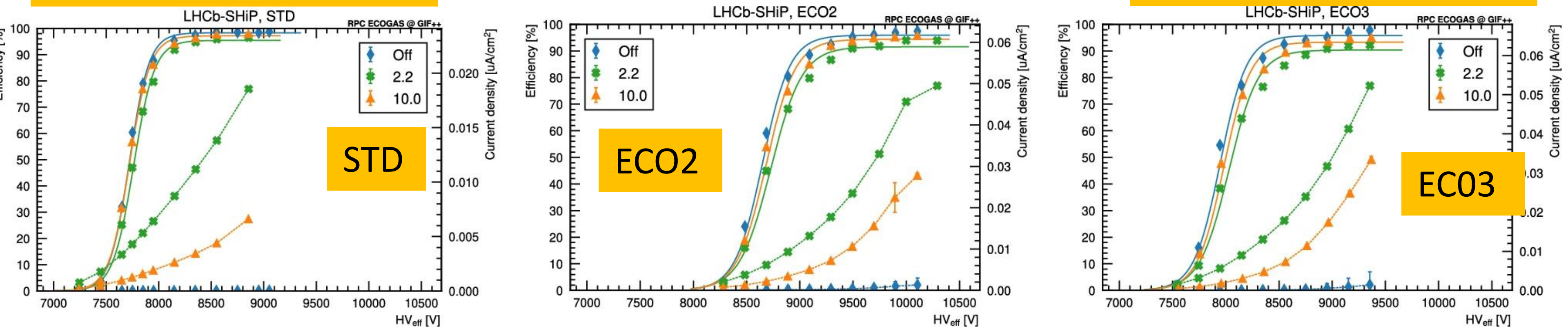
- ➢ Efficiency at plateau basically similar for STD, ECO2 and ECO3 > 95%
- ➢ Efficiency curves shifted at higher voltages depending on fraction of HFO

→ WP(ECO2) > WP(ECO3) > WP(STD)

Efficiency and counting rate with irradiation

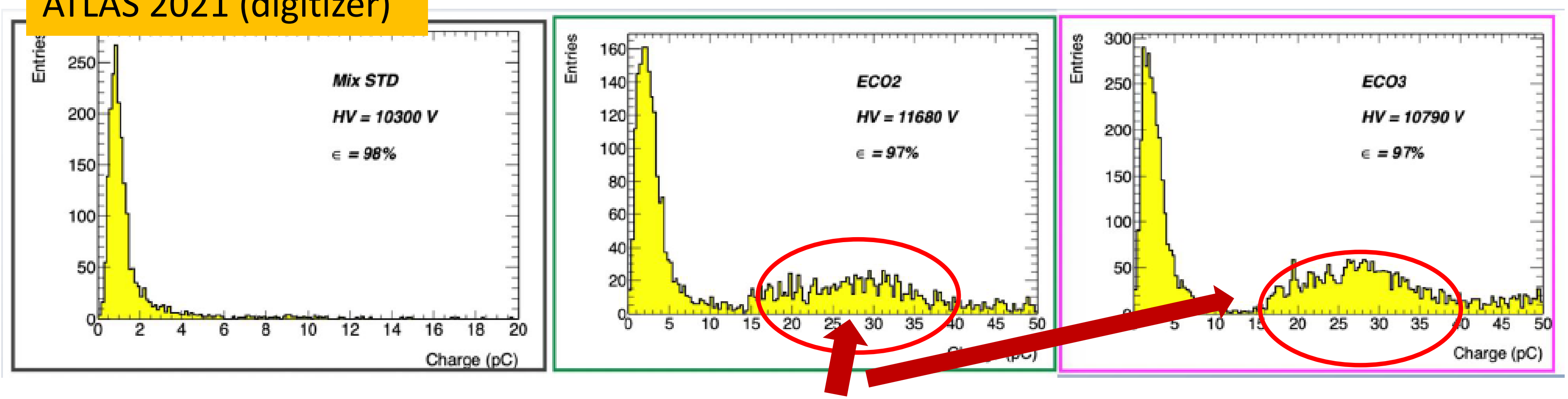
Source ON, 2021 data

LHCb/SHiP (SG 1.6 mm)



- Efficiency > 90% at the highest rate for all detectors

ATLAS 2021 (digitizer)



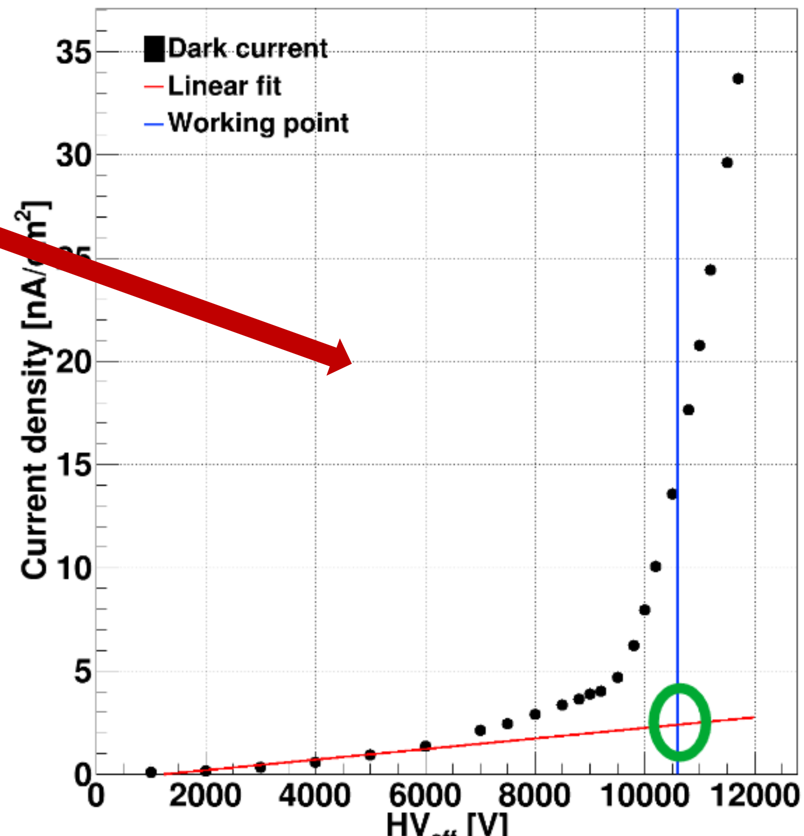
With ECO2 and ECO3 the presence of larger charge events (not streamers) observed

Coherent with larger current density at WP, and larger efficiency drops at high rates for ECO2 and ECO3 under irradiation

Aging tests: methodology

- All the detectors under test are flushed with the ECO2 gas mixture, while kept at fixed HV suitably chosen by the various groups: (irradiation voltage)
- They are irradiated so that, depending on their position, they absorb a dose typically between ≈ 1 and 5 mGy/h
  → they are subject to a background γ rate between 400 and 1000 Hz/cm^2
- The HV and absorbed current are continuously monitored and data stored every 30 s

- ❑ Weekly HV scans are performed to monitor the absorbed current <u>without irradiation</u>
- ❑ Both the ohmic and the total current are measured (the ohmic current by means of a linear fit in the low voltage range).

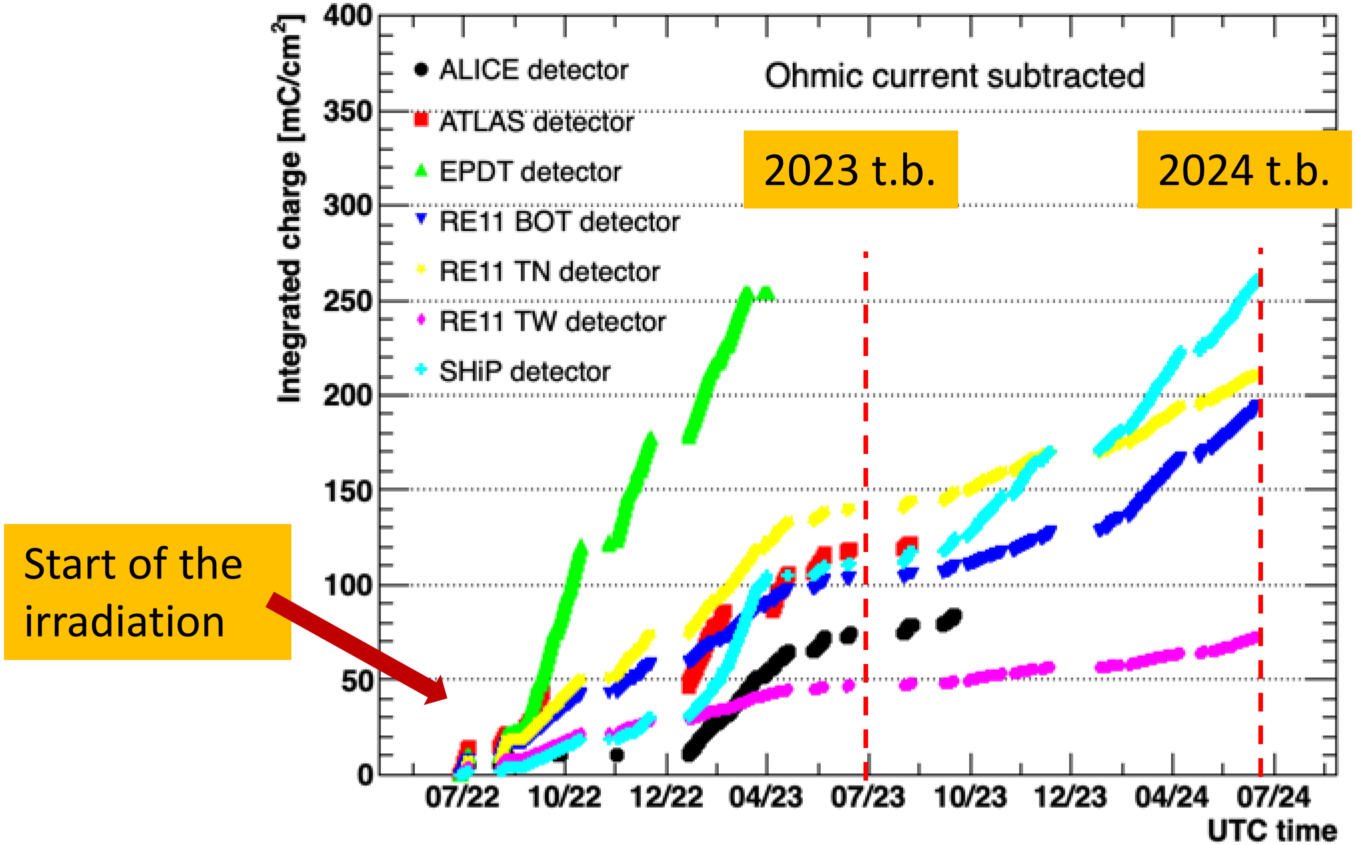


✓ Resistivity is measured by the Ar method 2-3 times/year

✓ Detectors performance is measured during dedicated beam tests 2-3 time/year

Causes for aging in RPCs

- Generally, the charge integrated along a certain elapsed time is considered the most important factor for aging in RPCs;
- The targets of integrated charge are different for various experiment: for instance, for ALICE is ≈ 100 mC/cm^2, for CMS is ≈ 1 C/cm^2 CMS, including a safety factor of 3.



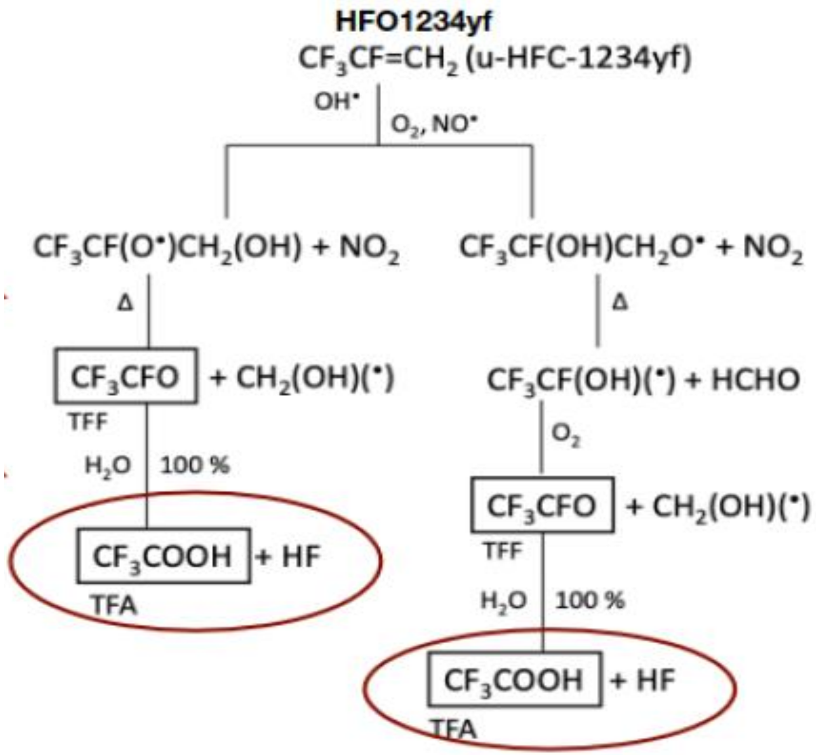
Further considerations about aging

Caveat: The importance of the integrated charge derives from the fact that production of HF was measured to be proportional to the integrated charge

→ Direct damage of the detector

- However, HFO typically dissociates producing TriFluoroAcetic acid

How TFA causes aging in RPCs and affects their performance on the long term is still to be investigated.



Aging is also caused by irradiation itself
→ Chemical modifications in the HPL electrodes

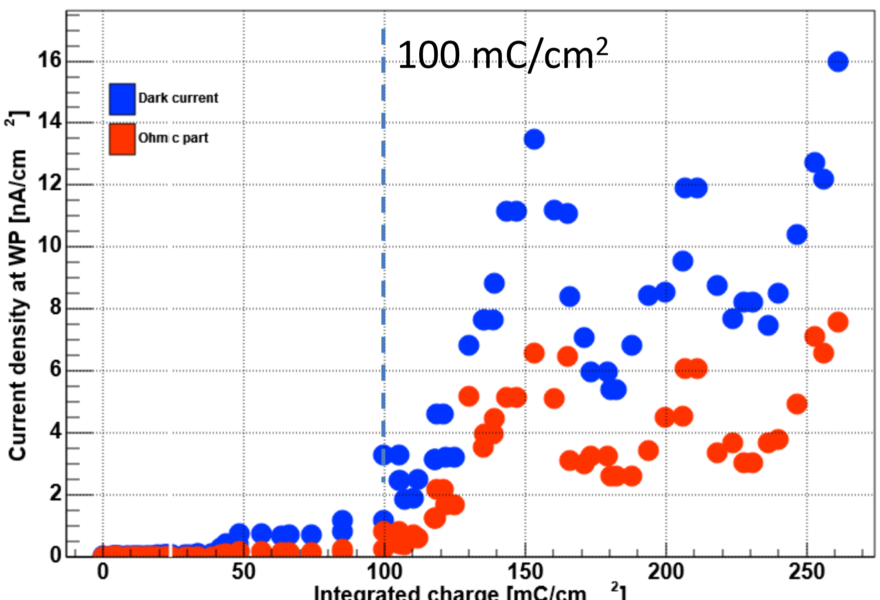
Aging is also caused by time itself
→ e.g. changed in HPL resistivity because of drying up.

Current density vs. integrated charge

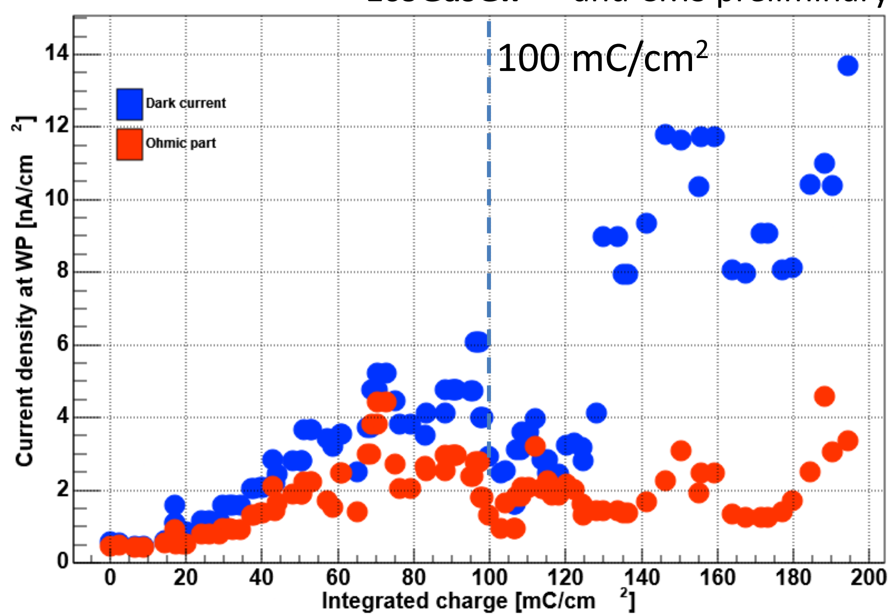
- Up to ≈ 100 mC/cm^2 of integrated charge (almost) all detectors present currents basically stable with time.
  - → CERN EP-DT detector 6 replaced by detector 25 in 2022 because of high currents, present already from the beginning (old detector).
- After ≈ 100 mC/cm^2 of integrated charge most detectors show the current fluctuations and slow rise with time.
  - → Behaviour similar in all detectors under test

Source OFF

EcoGasGIF++ and CMS preliminary

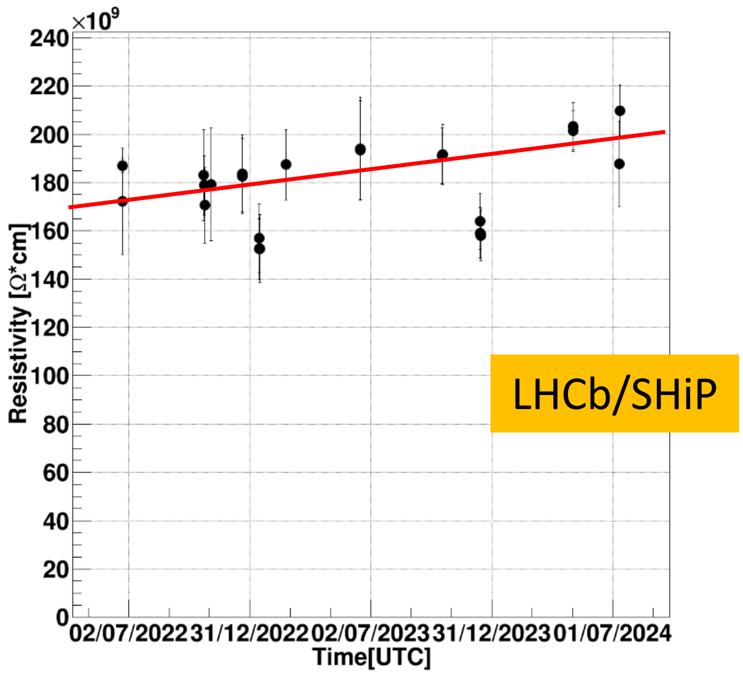


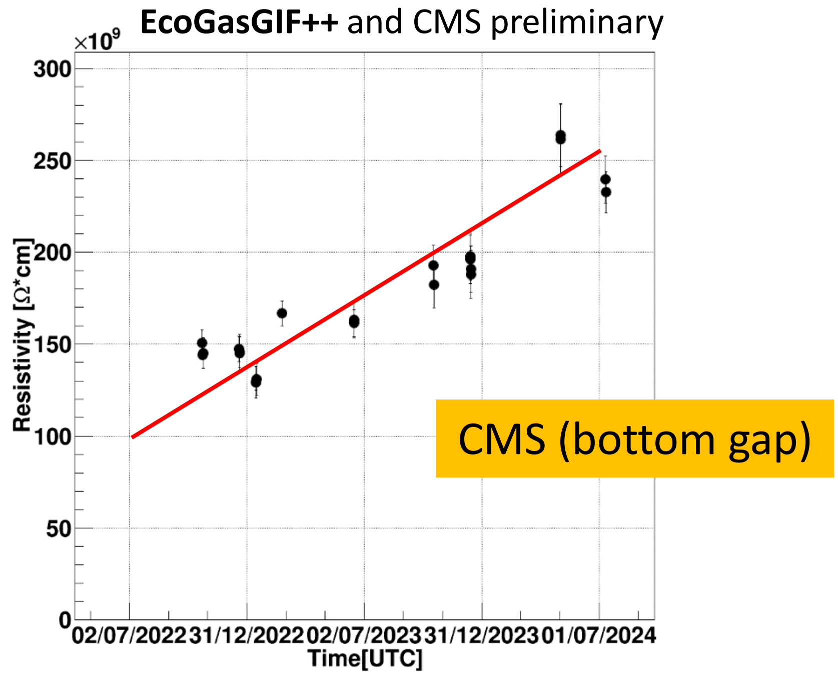
LHCb/SHiP
HV=8.7 kV



CMS (bottom gap)
HV=10.6 kV

Resistivity measurement campaign





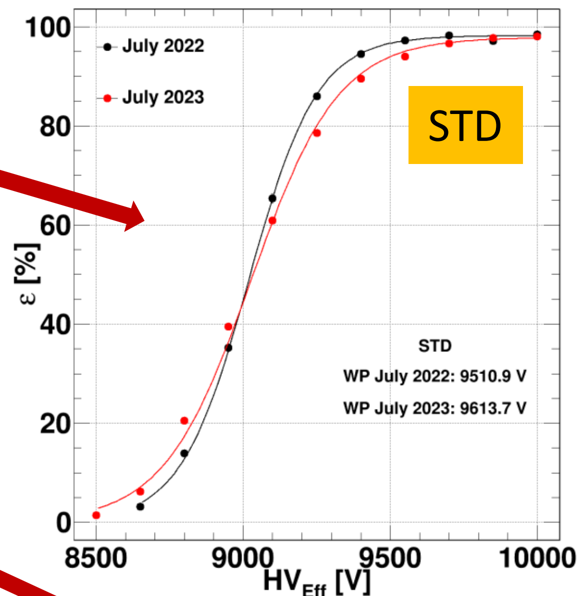
- The shift of the operating voltage observed might be related to the increase of bakelite resistivity and/or current observed.
- Indeed an increase of resistivity is observed when measured with the Ar method, with some differences across the detectors under test

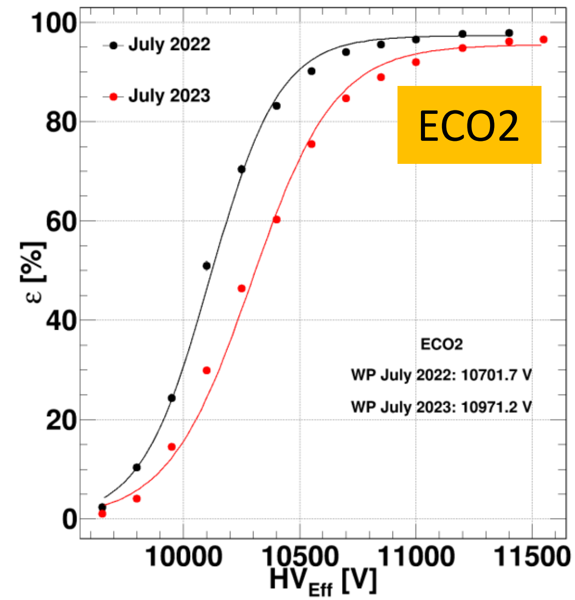
→ A study to quantify these effects on WP and current will be done in the future.

Efficiency before and after irradiation campaign

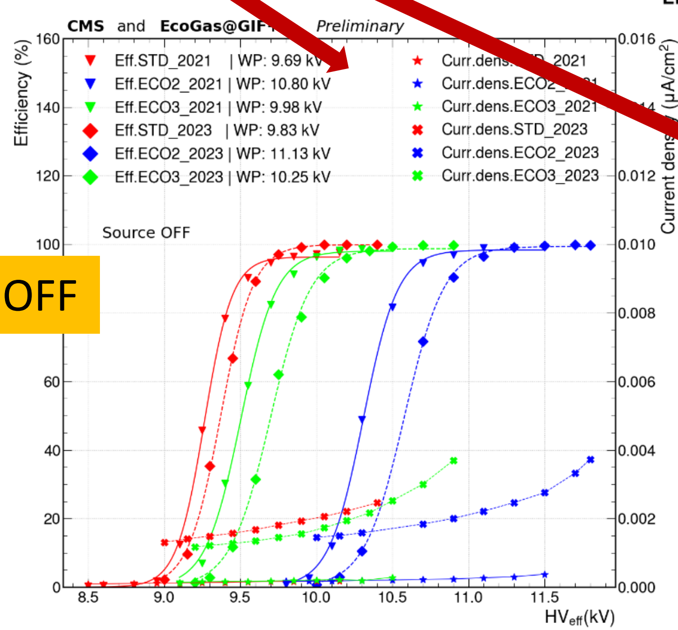
ALICE, comparison
2022 – 2023
SOURCE OFF

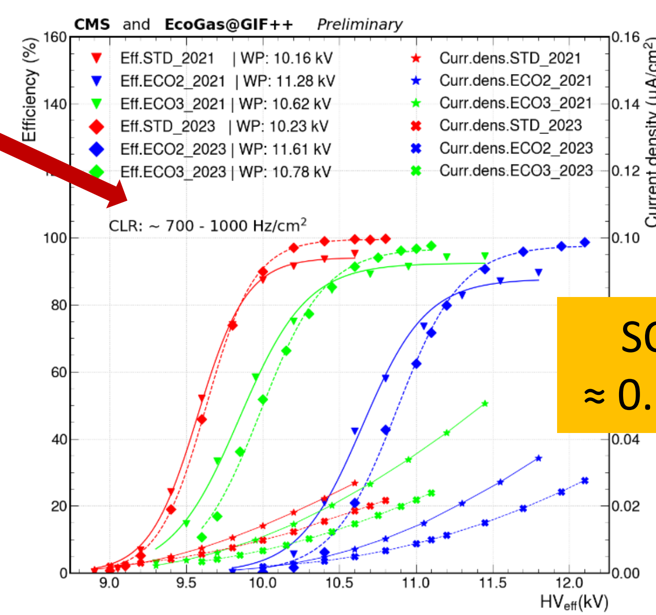
CMS (double gap),
comparison 2021 – 2023
Chamber OFF in 2022
because of low voltage
connector





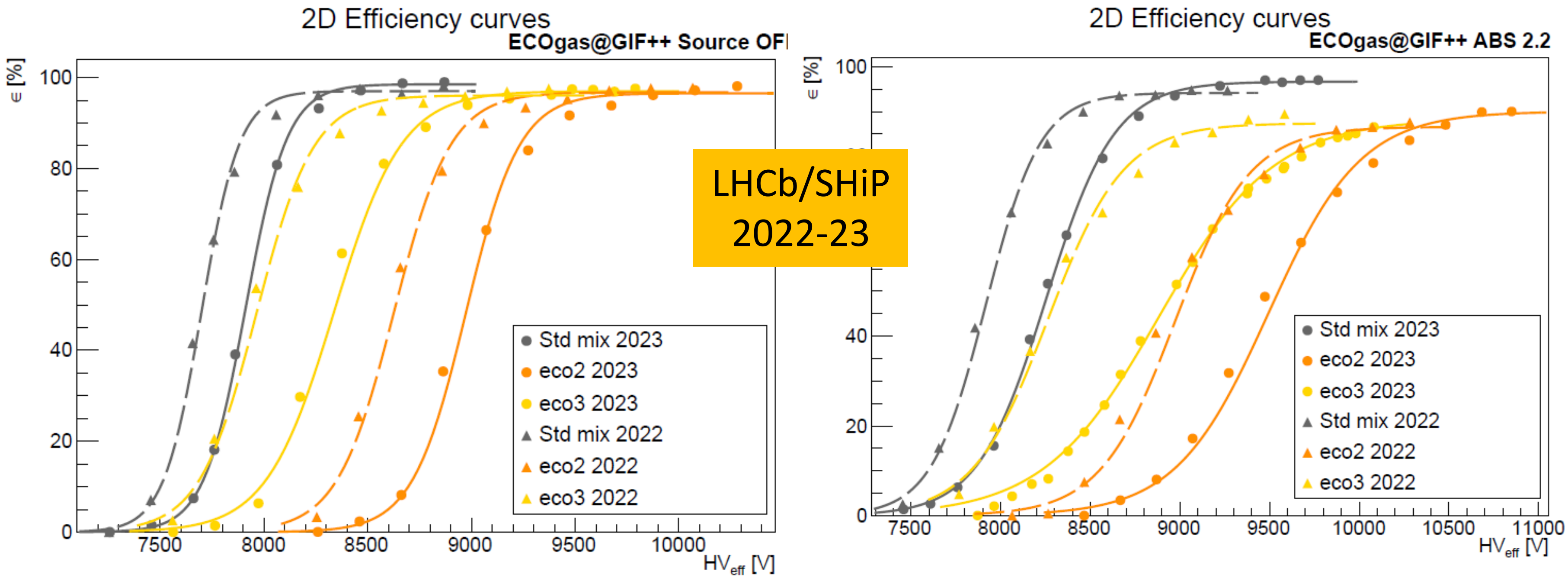
SOURCE OFF





SOURCE ON
≈ 0.7-1 kHz/cm^2

Efficiency before and after irradiation campaign

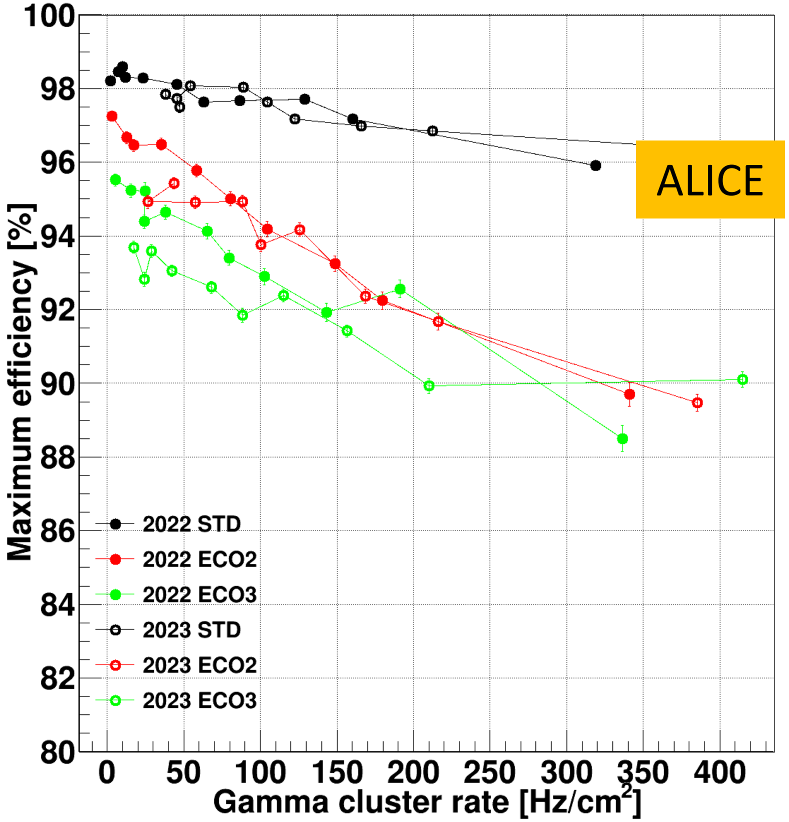


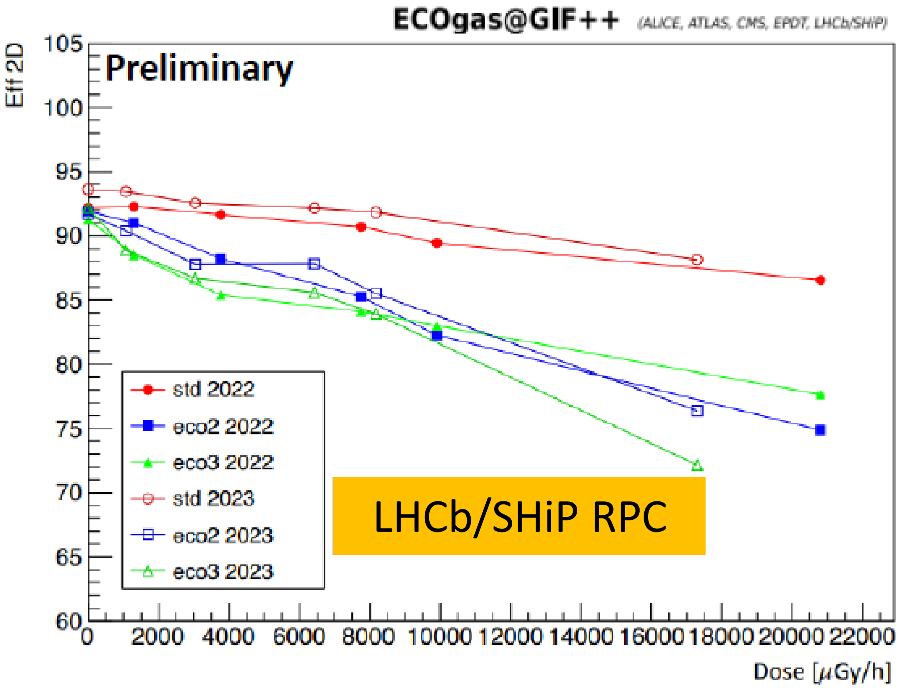
In general, for all detectors:

- A shift of the efficiency curves (few hundreds V) towards larger HV is observed
  - → For ALL gas mixtures used (so not directly caused by the gas)
  - → Smaller for STD with respect to ECO2 and ECO3
  - → Might be caused by changes in the HPL resistivity?
- Plateau efficiency remains approximately stable after the irradiation

Plateau efficiency before and after irradiation

Comparison 2022-2023 data
(2024 data still under analysis)





- The usual decrease of plateau efficiency with rate (or dose) is observed.
- Nevertheless, there seems NOT to be any efficiency degradation in the time lapse 2022-23

The other piece of the puzzle

- The replacement of TFE is just **part of the problem**; in ECO2 and ECO3 the residual GWP is **almost ALL due to the presence of SF6.**

- Gas mixture replacement is generally done at constant number of gas volumes → CO2e is the parameter to consider when evaluating the reduction of the impact on greenhouse effects

Mixture	GWP (100 y)	CO2e (g/l)
Standard	1485	6824
ECO2	475	1522
ECO3	527	1529

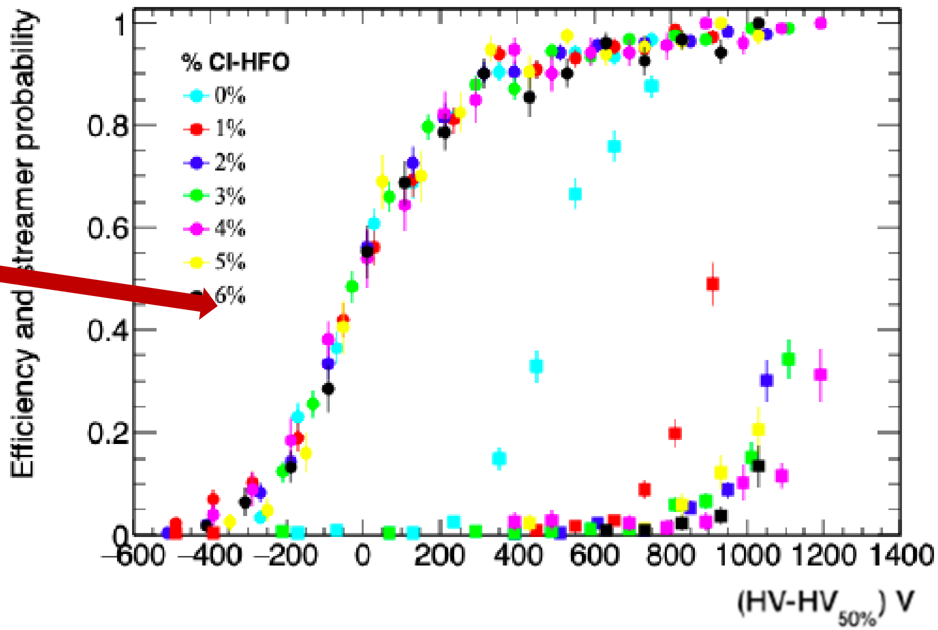
- With ECO2 and ECO3 achieved a reduction of 4 times the CO2e wrt. STD
- The residual CO2 is ALL due to SF6

→ Need to find replacement for SF_6, with low GWP and CO_2e, which could reduce the fraction of large charge events when in combination with HFO.

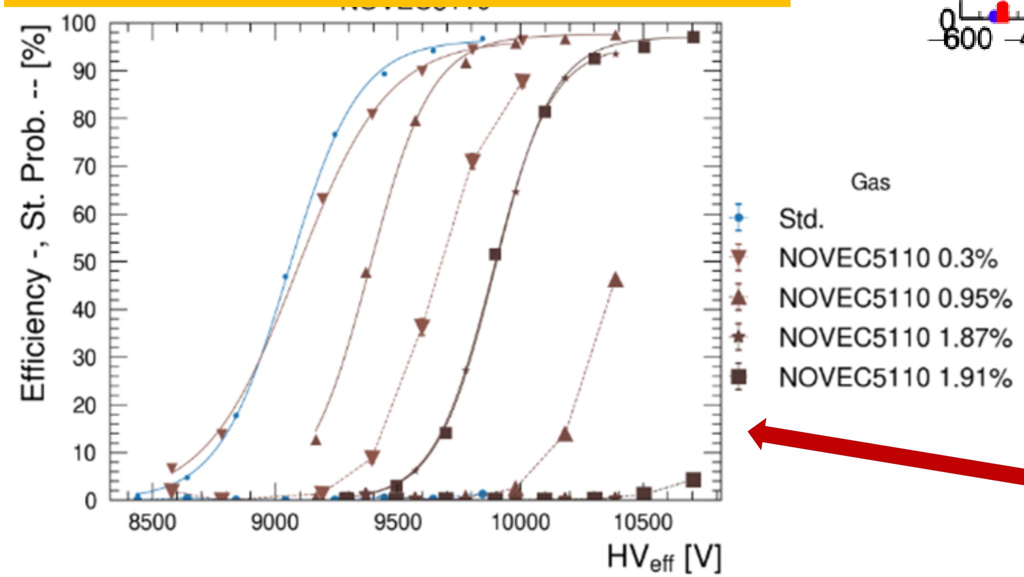
Looking for replacements for SF_6

One proposed solution is Chloro-trifluoropopene $C_3H_2ClF_3$ (HFO1233zd)
Avalanche/streamer separation ("useful" plateau) **larger than 400V**

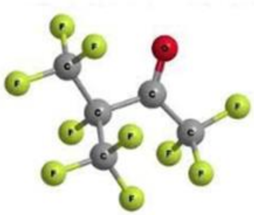
G.Proto et al, 2022 *JINST* **17** P05005



B. Mandelli, Possible alternatives to SF6 for Resistive Plate Chambers RPC2022



NOVEC 5110



- GWP < 1
- Atm. lifetime = 15 days
- Application in industry

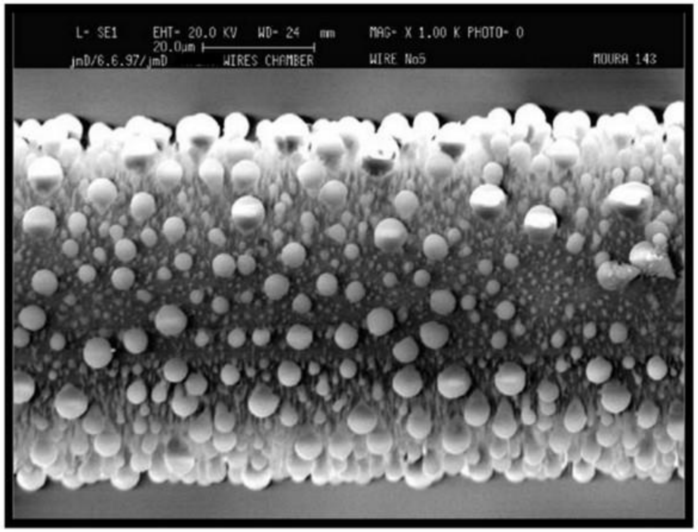
What about CF_4?

Used in CSCs and GEMs

For instance, gas mixture used in CSCs of CMS:

- **40% Ar + 50% CO2 + 10% CF4**

- The main purpose of CF4 in the gas mixture – protection against anode wire aging : Si + 4 F → SiF4 (also breaking C-chains in polymer formation)



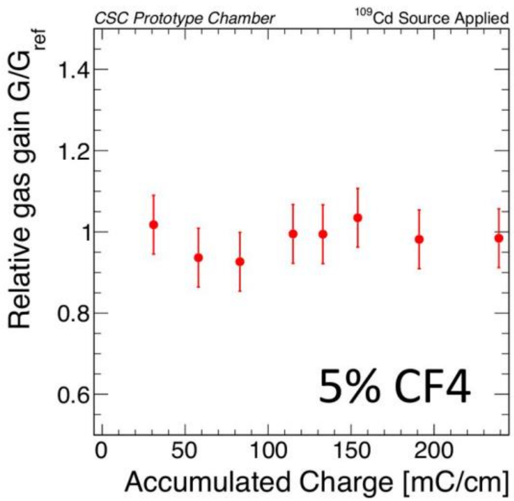
Used in GEMs basically to increase drift velocity → better time resolution. Anyhow without CF_4 time resolutions till within requirements,

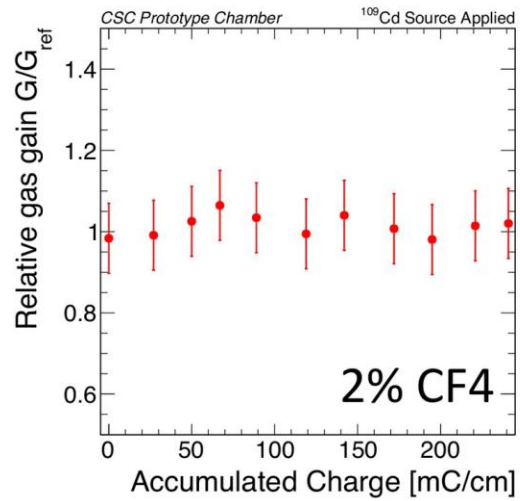
Main ideas:

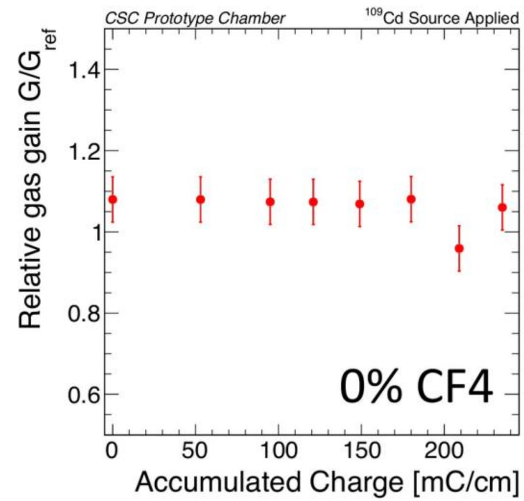
✓ Reduce (or eliminate) CF_4

✓ HFO to replace CF_4, but this implies an increased HV → more studies needed

Reduction of CF_4 in CSC

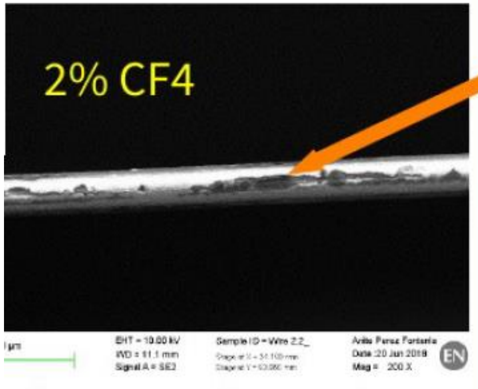


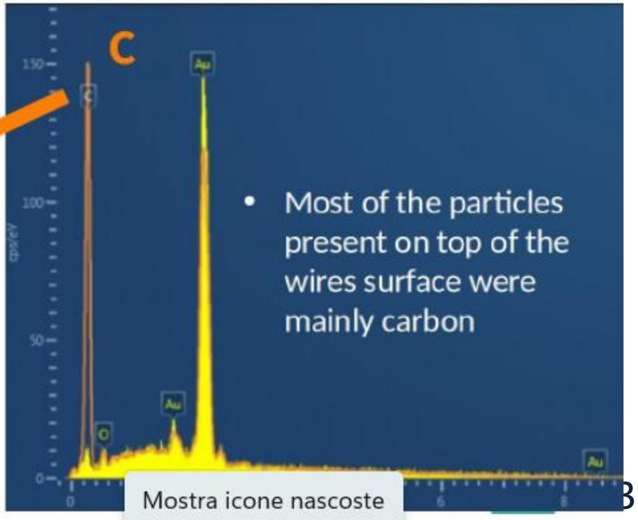




No significant degradation seen, in terms of performance, in all longevity tests

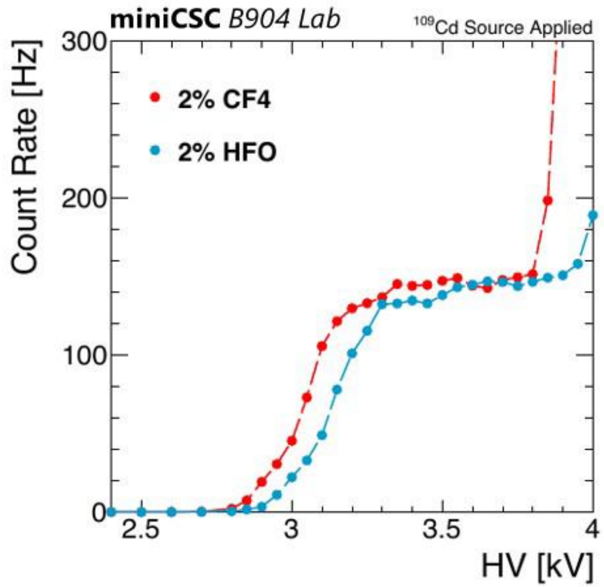
- However cathode modifications were seen in all cases.
- Anode surface depositions are seen with 0 and 2% CF4 even with naked eye.

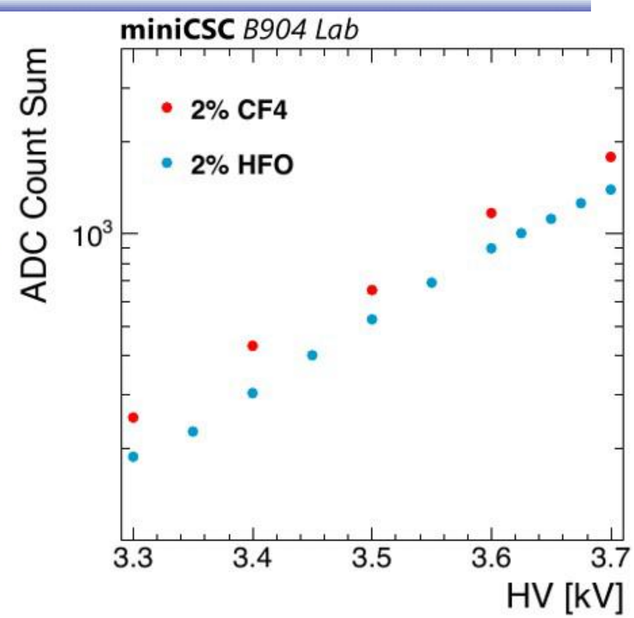


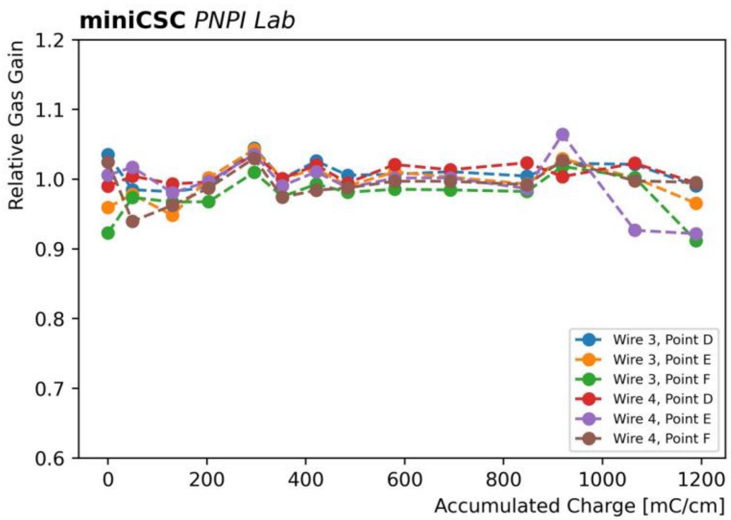


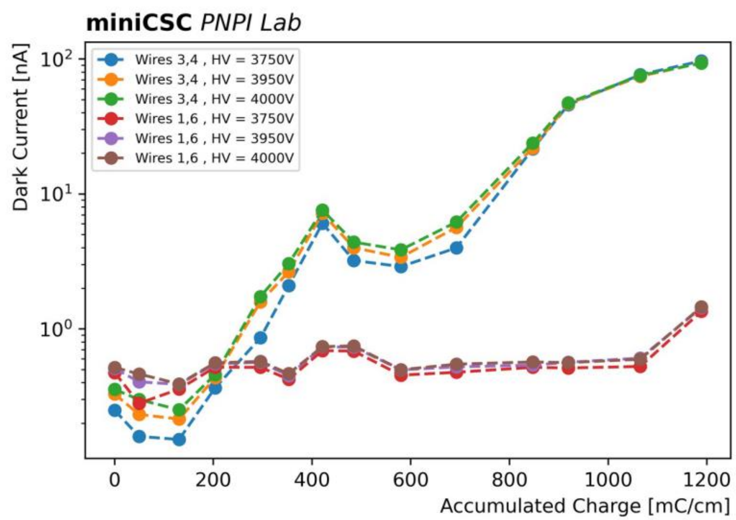
Replacement of CF_4 with HFO1234ze

Just 1000V increase in the operating voltage, good efficiency, reasonable plateau length









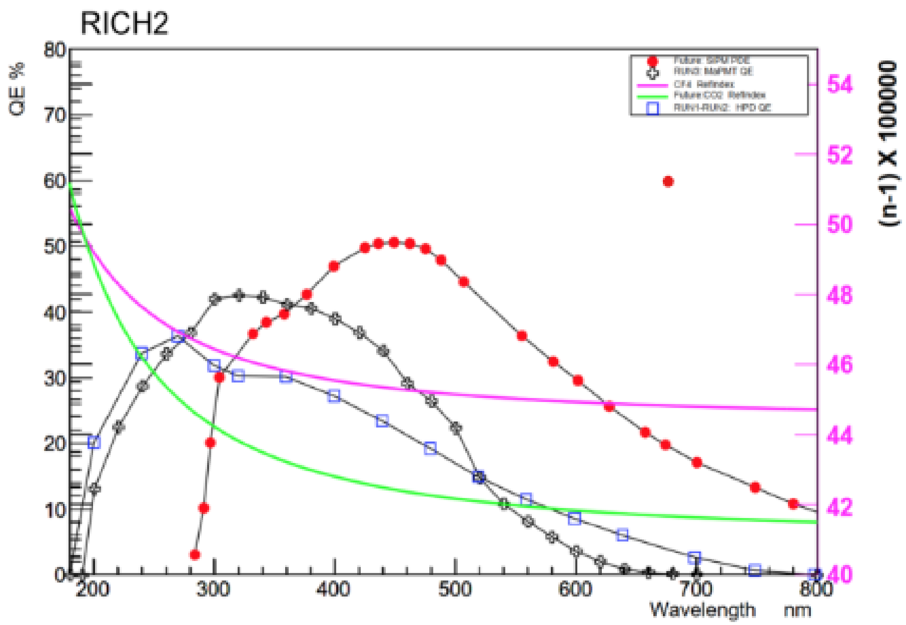
No gain reduction up to 1 C/cm, but significant increase in the dark current in first irradiation tests

Addendum: use of GHG in RICH detectors

- C_4F_{10} used in the LHCb RICH
- CF_4 used in the COMPASS RICH

LHCb RICH studies

- RICH detectors use either CF_4 or C_4F_{10}
  - Necessary for good refractive index
- Replacement of C_4F_{10} with C_4H_{10}
  - Refractive index matches very well
  - But C_4H_{10} **flammable**
- Replacement of CF_4 with CO_2
  - Under investigation
- Use of SiPM to reduce the chromatic error and increase the yield



Conclusions

- In general the idea of replacing TFE with HFO (+CO_2 to reduce the operating voltage) seems to work.
  - ECO2 and ECO3 might be good candidate gas mixtures
- Interpretation of the effects observed not trivial
- Replacement of TFE is not the only issue here
  - →check the performance of fully eco-friendly gas mixtures
- Collaborative effors of paramount importance at this stage.