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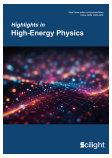

Article

Reducing the Greenhouse Gas Usage from Particle Detection Systems: Status and Perspectives

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Abstract: The gaseous particle detector community is nowadays facing the problem of minimizing usage and, in the long term, finding replacement for fluorinated gases used in several detector technologies. Fluorinated gases like $C_2H_2F_4$, SF_6, CF_4, C_4F_{10}, ... are used because they allow achieving detector performance needed for data taking in presence of hostile radiation background like the one present at the Large Hadron Collider experiments. However, fluorinated gases are nowadays subject to increasingly stringent regulations that aim to ban their use in the industrial world as soon as new technologies or ecological alternatives are developed. Unfortunately, the application of new ecofriendly gases developed as refrigerant fluid or high voltage insulation medium to particle detectors is not straightforward. To optimize the usage of fluorinated gases and with the idea of preparing the long-term operation of gaseous detectors different strategies have been developed at CERN. These strategies are based on extensive use of gas recirculation systems (both for large detector systems and laboratory applications), the development of gas recuperation plants and the search for new ecofriendly mixtures for existing and future gaseous detectors.

Keywords: gaseous detectors; environmentally friendly gases; gas recirculation and recuperation systems; greenhouse gases; low GWP gases; HFO

1. Introduction

A wide range of gases is used for the operation of different gaseous detectors for particle physics research. Among them there are greenhouse gases (GHG) with high Global Warming Potential (GWP) like $C_2H_2F_4$ (also known as R134a, GWP 1430), CF_4 (also known as R14, GWP 7390), C_4F_{10} (also known as R610, GWP 8600) and SF_6 (GWP 22800), which are needed to achieve specific detector performance required in particular for data taking in a harsh environment in terms of radiation and particle rate like the one today present at the LHC experiments (i.e., long term stability, time resolution, rate capability, etc.). Due to their environmental impact, these gases are increasingly subject to regulations. For example, the revised version of the European regulation (EU517/2014 revised in 2023) [1] calls for the total elimination of Hydrofluorocarbons (HFC) by 2050. Moreover, it bans the use of F-gases where less harmful alternatives are available, and it requires adequate checks, maintenance and recovery of F-gases to avoid emissions from existing equipment. Under the pressure of a total phase out policy, the HFC market started to be affected with consequences on price and availability for these products.

Considering this context, it is beyond question that future detectors will have to attentively minimize or even exclude the use of GHGs.

With the aim of reducing GHG usage, securing the operation of current detectors and of preparing the future generation of particle physics detectors CERN has elaborated a strategy based on four action lines: implementation of gas recirculation systems, development of gas recuperation systems, search for eco-friendly replacements of currently used GHGs, and GHGs disposal [2].

2. Gas Recirculation Plants

CERN is today the largest accelerator complex and together with its experiments represent a natural place where to learn from past experiences and prepare the future detector generations. At the CERN Large Hadron Collider (LHC) experiments ALICE (A Large Ion Collider Experiment), ATLAS (A Toroidal LHC Apparatus), CMS (Compact Muon Solenoid) and LHCb (Large Hadron Collider beauty) about 30 gas plants are delivering the proper gas mixture to the corresponding detectors [3]. Gas plants extend from the surface building where the primary gas supply is located to the service caverns up to the single detector following a route a few hundred meters long. In total, the gas system infrastructure (pipes excluded) corresponds to about 300 euro-rack sized modules, which if placed one on top of the other are equivalent to about 2.5 times the height of the Eiffel tower. Particle detectors at the LHC experiments are very often characterized by large detector volume (up to several hundred cubic meters) and by the use of very specific gases often expensive and/or GHGs. For these reasons, since the design phase, gas consumption optimization was considered an important criterion for reducing operational costs. Indeed, most of the LHC gas systems were already designed to operate in re-circulation mode. In this layout, after being used, the gas mixture passes through specific gas purification units. A small fraction of fresh gas is added before resending the mixture to the detector system. The maximum recirculation rate is fixed by detector leak or by the need to control impurities that cannot be filtered (for example N_2). Figure 1 shows a schematic view of a gas recirculation system. Figure 2 shows a view of the gas recirculation modules present in the surface gas building for one detector.

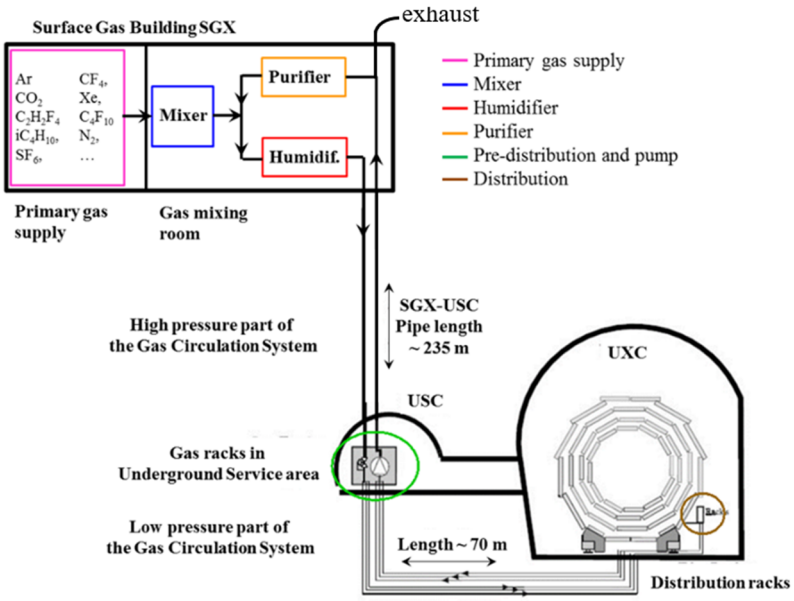

Figure 1. Schematic view of the gas recirculation system. After being used in particle detectors, the gas mixture passes through dedicated purification modules, and it is then re-used in large fraction. The maximum recirculation fraction is fixed by detector requirements or filtering capacity for specific impurities.

Figure 2. View of gas systems modules present in surface for one detector at the LHC experiments.

The advantage of the gas recirculation system is that gas consumption can be drastically reduced. However, gas recirculation systems are complex to operate: constant monitoring of mixture composition and concentration of impurities is mandatory. A dedicated module for gas mixture purification is needed to avoid any impurity accumulation. In addition, detector pressure and flow stability are crucial requirements.

The use of gas recirculation plants should be a must for all detector systems using GHGs. Since any intentional release of F-gases is forbidden by the F-gas regulation, the recirculation fraction should be as close as possible to 100%. This implies that gaseous detectors should be absolutely leak tight and their operation should be certified from the beginning in conditions as close as possible to the real ones in terms of background radiation and of any other source that can lead to deterioration of detector components and gas molecules. Concerning the latter, production of impurities, filtering techniques and test with gas mixture recirculation system should be addressed since the design phase [4].

Already during Run3, all detectors using GHGs were operated with gas recirculation systems. The next step was an optimization aiming at ensuring flow and pressure regulation stability beyond original requirements to cope with new detectors' needs like enhanced stability, increase of recirculation fraction and flows [5]. At gas systems level these requirements implied an improvement of the detector pressure regulation from fluctuation of the order of few 0.1 mbar down to 0.1 mbar or below. Particularly critical is the situation of the ATLAS and CMS Resistive Plate Chamber (RPC) detector systems where plastic connectors and pipes used for gas mixture distribution inside detectors are degrading with time due to aging effects or subject to mechanical vibrations. For achieving this enhanced stability needed to compensate for the observed detector fragility, the gas distribution systems were redesigned. An intense R&D activity started since the gas system complexity is often a consequence of specific detector requirements that do not find equivalent in industrial applications (low pressure, limited space, use of refrigerant in gas phase, …). New regulation valves and pressure sensors were installed. The latter are mounted on reference volumes simulating RPC detectors at the highest point of each distribution sector. Moreover, new distribution modules are equipped with regulation valves at the input allowing an opening as smoothly as the detector might need. Also, fresh mixture injection and purifier module operational phases were subject to substantial improvements. However, it was not possible to correlate the improved stability with a decrease in leak development rate.

The lesson to be learned from these experiences is that the risk of gas leak development should be carefully evaluated during detector design phase not only if expensive or flammable gases are used, but also in case of GHGs usage. The use of plastic pipes or plastic connectors should be reduced to minimum. If necessary to prevent the transmission of mechanical vibrations, soft plastic pipes should be chosen considering gas mixture versus material properties.

Nowadays the objective is to extend the use of gas recirculation plants to small and medium size experiments as well as to laboratory application. Indeed, a considerable contribution to GHG emissions is due to setups used for detector upgrade projects, R&D activities, detector production tests and longevity studies. Furthermore, in some university and research institutes the purchase of GHGs is already forbidden or subject to strict rules. A recirculation system will allow to continue tests in laboratory. To minimize such emissions, a compact and flexible recirculating gas system was developed [6]. The small recirculating system was basically a small replica of a LHC gas system and it was conceived as a single euro-rack containing the essential gas systems modules such as humidifier, pump, distribution, gas analysis, purifier, and control PLC. The system was designed to operate several detectors with volumes up to several tens of litres and circulation flow up to 200 l/h. Figure 3 shows the front and rear view of a small recirculation unit. Several of these recirculation systems have been built in the last years and are used for RPC, Cathode Strip Chamber (CSC), Gas Electron Multiplier (GEM) detectors in laboratory set-ups, test-beam set-ups and at the CERN Gamma Irradiation Facility (GIF++) [7].

Table 1 compares construction, operation and maintenance costs as well as GHG emissions for different gas recirculation plants. Three examples are presented for large detector systems at the LHC experiments: a typical RPC, the CMS-CSC the LHCb-RICH1 (Ring-Imaging Cherenkov 1 (The LHCb experiment has two RICH detectors called respectively RICH1 and RICH2)) detector systems. A gas recirculation plant costs between 50% to 100% more with respect to a simple open mode gas distribution system. However, for such large detector systems, the gas recirculation plant cost is paid back in few months of operation. The number of distribution channels, i.e., the gas mixture distribution granularity at detector level, accounts for more than 30% of the total plant cost. This explains the higher cost of the RPC and CSC gas recirculation plants, which have respectively about 400 and 162 channels with respect to the RICH1 gas recirculation plant cost where there is only one channel.

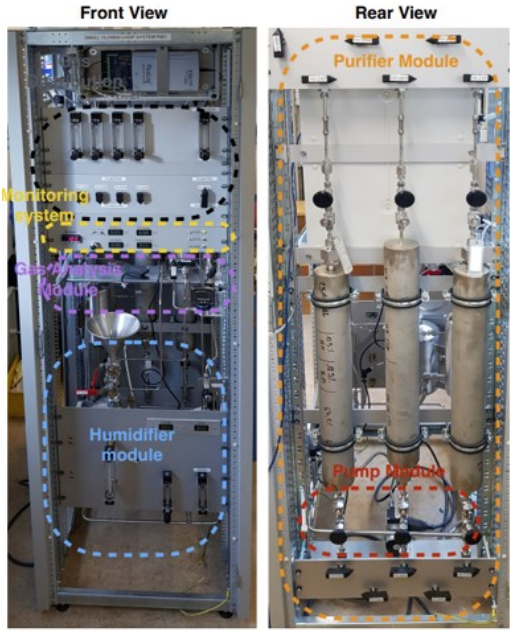

Figure 3. Front and rear view of the small gas mixture recirculation unit. Together with development and commissioning of small recirculating system, an even smaller and simpler recirculating unit (micro gas recirculation system) was designed with the goal of having a portable recirculation system for few detectors as simple and economically suitable for small institutions such as schools, universities, and CERN laboratories [8]. The system should also be kept as simple as possible, and the operation should require the least amount of manual intervention possible.

The maintenance and operational costs are about equally subdivided between material and personnel. Gas recirculation plants developed at CERN are made of industrial components available worldwide. Components have been selected according to severe requirements on material and cleanliness. Mainly (if needed only) stainless steel pipe and components are used. Before their use, all components are subjected to a validation test to exclude that they contain chemical compounds (oils, greases, ...) which can produce aging effects in detectors and finally a deterioration of detector performance. In some cases, components have been adapted to specific detector's needs. However, the modifications are documented and relatively easy to reproduce.

The operation of such large gas recirculation plant is clearly more complex than an open mode gas distribution system. However, few months of training are enough to achieve good understanding of the plant allowing to being able to solve most of the issues that can appear during operation.

Table 1 shows also the situation for small and micro gas recirculation systems. Considering the use of these systems for RPC detector, the gas recirculation is paid back in about two years of operation.

Concerning GHG emission, a 90% gas recirculation implies a 90% reduction of emissions. When GHG are used is important to consider potential consequences coming from increase of price and limited availability. Figure 4 shows the increase of price for R134a in Europe after the entry in force of the European regulation EU517/2014 [9,10]. In about 10 years the R134a cost has increased by a factor of 10. By limiting by 90% or more the gas needed for operation, a gas recirculation plant is limiting the risks due to price increase and reduced availability due to market crisis. An important point to underline for detector development is to consider long-term validation tests at the highest possible recirculation factor.

Considering that limiting factors to gas recirculation are development of gas leaks at detector level and the use of material permeable to air a particular attention should be paid during detector and related infrastructure design phase. All material used should be tested in their final or realistic condition of use. Also very important is to consider the past experiences.

Table 1. Comparison between construction, operation and maintenance costs as well as GHG emissions for different gas recirculation plants. Three examples are presented for large detector systems at the LHC experiments. The cases of small and micro gas recirculation systems applied to RPC detectors are also shown. The numbers in the table are calculated considering systems without presence of gas mixture leaks.

	RPC	CSC	LHCb-RICH1	Medium Recirculation for RPC	Micro Recirculation for RPC
mixture cost (CHF/m^3)	60.1	18.0	2125	60.1	60.1
Gas mixture flow through detector system (m^3/h)	8	6.5	0.1	0.03	0.005
operational cost without recirculation—gas supply only (kCHF/year)	3462	842	1530	13	2
Open flow gas distribution system—construction costs (kCHF)	500	350	180	15	1
recirculation fraction (%)	91	89	99.6	90	90
gas operational cost with recirculation system (kCHF/year)	312	91	6	1.3	0.2
gas recirculation system—construction cost (kCHF)	≈750	≈600	≈300	45	5
gas recirculation system maintenance and operation per year (kCHF)	60	40	20	1	0.5
Return of investment for gas recirculation system (years of operation)	≈0.3	≈0.7	≈0.2	≈1.7	≈1.7
tCO_2e emission without gas recirculation system (tCO_2e)	382,745	145,845	67,779	1435	239
tCO_2e emission with gas recirculation system (tCO_2e)	34,447	15,706	266	144	24

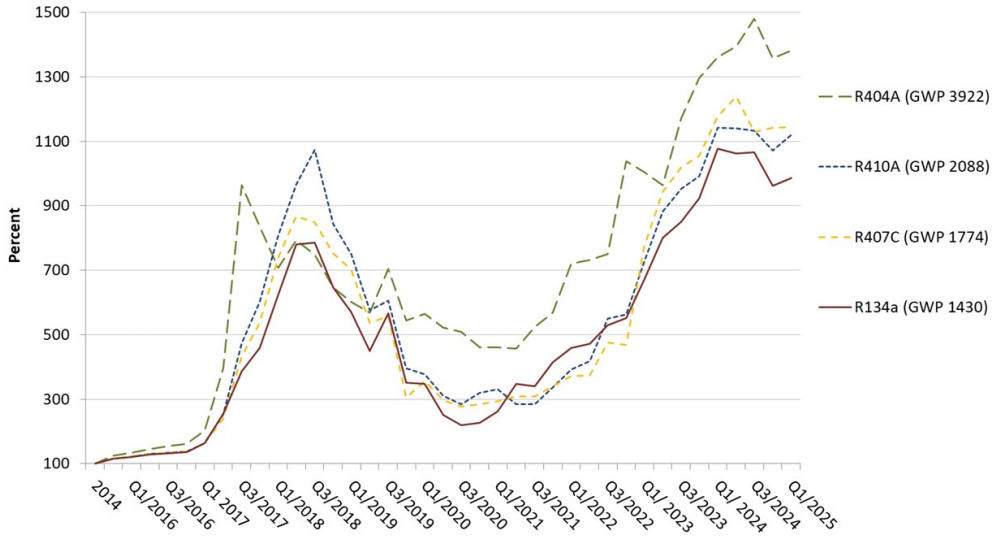

Figure 4. R134a price increase in Europe after the entry in force of the European regulation EU517/2014 [7,8]. In about 10 years the R134a cost has increased by a factor of 10 [10]. The decrease observed between 2020–2022 was probably due to illegal imports.

3. Gas Recuperation Plants

In some cases, depending on the material used during construction, particle detectors can be gas tight but at the same time permeable to impurities that cannot be filtered by gas purification modules (for example N_2). Also, the fresh gas flow injection may be needed by detectors for safe long term operation following results of long-term validation tests. In these circumstances the gas recirculation rate is limited by the need to avoid an excessive

accumulation of impurities in the gas stream. The mixture can be collected from the exhaust of the gas recirculation plant and sent to dedicated gas recuperation plant able to extract a specific mixture component.

Gas recuperation plants allow to reduce the usage of fresh gas injected into gas recirculation plants and therefore to further reduce the GHG emissions. However, they introduce a second level of complexity in addition to gas recirculation plants, and they require dedicated quality monitoring systems for checking the purity of recuperated gas. Moreover, given the specificities of the gas mixture used by particle detector systems, no industrial apparatus currently exists to address these specific needs. Dedicated R&D studies are required for the design of a recuperation plant and similarly for the definition and tuning of gas mixture monitoring tools.

The objective is finding the best separation technique that maximise purity and efficiency for recuperated gas without introducing additional chemical compound that can affect detectors operation even at long term. The best approach was found to be the use of physical separation techniques: membrane separation processes, adsorption through Pressure and Thermal Swing cycles (PSA and TSA) using different types of molecular sieves and distillation [4]. More techniques can be combined in recovery plants to achieve the most efficient process, depending on gaseous compound to separate and contaminants present in the mixture.

Each recovery system installed at the LHC experiments required intense R&D activity for various reasons, including:

- Very specific gas mixtures not used in other industrial processes and consequent lack of information in literature.
- Selection of separation processes that did not cause formation of undesirable reaction by-products, in order not to contaminate the detectors.
- Choice of inert materials capable of interacting only physically with gas mixture without altering its chemical structure.
- Evaluation of construction and operational costs with respect to the objective of GHG emissions reduction in relation to the initial gas mixture.

Recuperation plants have been developed for CF_4, R134a and C_4F_{10}. New R&D are nowadays ongoing for SF_6.

The first CF_4 recuperation plant was installed in 2012 [11,12]. It was developed for the CSC detectors of the Compact Muon Solenoid (CMS) experiment. The CMS-CSC muon detector uses a three components gas mixture made of Ar, CO_2 and CF_4. The detector is tight (only about 60 l/h of gas mixture are lost due to leak and gas analysis compared to a total detector volume of 90 m^3 and a circulation flow of 6.6 m^3/h). However, it has been discovered that due to gas diffusion mechanism the gas mixture gets contaminated by air. Oxygen and water concentration are kept under control using standard purification modules, but unfortunately Nitrogen cannot be easily filtered by the gas stream. The recuperation plant is a fully automated system for warm gas separation. CF_4 is separated from the detector return mixture in three stages: first, a membrane is used to remove the bulk of the CO_2, then molecular sieve 4 Å is used for removing the residual CO_2 (from hundreds ppm down to almost zero), and, finally, CF_4 is extracted by means of pressure swing absorption cycles on molecular sieve 13X. The CF_4 plant has now a recuperation efficiency of about 70%. During a typical year of run, it allows to recuperate about 1400 kg of CF_4 out of the 2200 kg needed. This produces a positive impact on three important aspects: operational budget, greenhouse gas emissions and CF_4 availability. Indeed, the plant was paid back in about 5 years of operation. It reduced by 10,000 tCO_2e the emission from particle detectors at CERN (about 8% of the total). Finally, the CSC detector operation is less subject to market crisis affecting price and availability: for example, during the worldwide CF_4 crisis of 2022, the CSC detector was operational, and therefore the CMS experiment was able to take data, only thanks to the recuperated CF_4. Indeed, in addition to the standard gas analysis module (common to all gas systems for experiments at the CERN Large Hadron Collider, LHC) a dedicated infrared analyser has been installed for continuously monitoring CO_2 and CF_4 concentrations, gas chromatographic analyses are performed one-two times per week on the gas mixture from the gas mixer and a gas monitoring system based on single wire detectors has been implemented.

A second example is related to the development of a R134a recuperation plant. The RPC detector gas mixture is composed of about 95% R134a (the remaining 5% is made of iC_4H_{10}, SF_6 and water vapour). During earlier tests the detector was validated for operation in high radiation environment like the one of the LHC experiments with a gas mixture recirculation fraction of about 90%. This means that 10% of fresh gas mixture is needed and, therefore, 10% of gas mixture will have to be exhausted from the gas recirculation loop. Greenhouse gas emissions related to the operation of the ATLAS and CMS RPC detectors represent about 80% of the total emissions due to particle detector operation at CERN. The first R134a recuperation plant developed and built at CERN is extracting R134a from the RPC mixture. The plant is installed at the CMS experiment [13] (Figure 5). For R134a, the gas

separation process is complicated by the fact that R134a and $i C_4H_{10}$ form an azeotrope, i.e., a mixture that cannot be separated by simple distillation. To overcome this complication, firstly the azeotrope is totally liquefied and then it is sent to a warm buffer kept at few degrees Celsius above 0 ◦C where the R134a separation from $i C_4H_{10}$ occurs. The R134a recuperation efficiency is about 80% with a good quality of the recuperated gas (the only extra-component is $i C_4H_{10}$ in concentration of about 0.3%).

Unfortunately, both RPC detector systems at the ATLAS and CMS experiments suffer of important gas leaks at detector level due to fragility of plastic pipes used for the mixture distribution in the detector or due to inborn fragility of plastic gas inlet/outlet. Today, since part of the fresh mixture flow injected is needed to compensate for detector leaks, the use of the R134a recuperation plant is limited only to the CMS experiment, moreover, below its full capacity. Despite this limitation, during a typical run year, about 12 t of R134a are recuperated out of 24 t needed. Here the impact on budget and reduction of greenhouse emissions is even more important: the plant is paid back in less than three years of operation, and GHG emissions are reduced by about 18,000 tCO_2e per year.

Figure 5. R134a recuperation plant installed at the CMS experiment and connected to the RPC detector gas recirculation system.

Other recuperation plants have been developed and are already in operation for CF_4 used by the LHCb-RICH2 (Ring-imaging Cherenkov 2) detector and for C_4F_{10} used by the LHCb-RICH1 detector (these plants are fundamental also for the detectors filling in preparation of run periods). A new R&D for the development of an SF_6 recuperation plant is ongoing.

Given the important role played by F-gases for particle detectors operation and the current context where the use of these gases starts to be strictly regulated due to an increasing environmental concern related to the climate change, the use of recirculation and recuperation plants must be extended also to laboratory setups, detector R&D and characterization facilities. The cost/benefit ratio must not be calculated considering a short time frame (in the past typically about one year), but the application of recirculation and recuperation plants must be considered feasible even if they have a cost that can be only partially paid back over the duration of the experiment or facility. Indeed, considering the instability of the F-gases market in the years to come, all systems that can reduce the F-gas consumption also strengthen the stability of particle detectors operation by making them less subject to market crises.

Table 2 compares construction, operation and maintenance costs as well as GHG emissions for different gas recuperation plants. Three examples are presented for large detector systems at the LHC experiments: $C_2H_2F_4$ plant for RPC, CF_4 plant for CSC and C_4F_{10} plant for LHCb-RICH1. For the first two plants the advantages are clear: the costs are paid back in very few years especially if compared with the duration of the experiments. Also concerning GHG emissions reduction, the advantage is clear: 75% for $C_2H_2F_4$ and 65% for CF_4. Different is the LHCb-RICH1 C_4F_{10} recuperation plant case: the return of investment requires about 35 years of operation, and it seems to indicate that the construction of the plant is not cost effective. However, C_4F_{10} is not produced anymore since long time and the experiment decided to buy a stock for the full duration of the data taking. So, given that it is basically impossible to find more C_4F_{10} (especially of the quality required by the RICH1 detector) the use of the recuperation plant is well justified.

Table 2. Comparison between construction, operation and maintenance costs as well as GHG emissions for different gas recuperation plants. Three examples are presented for large detector systems at the LHC experiments: $C_2H_2F_4$ plant for RPC, CF_4 plant for CSC and C_4F_{10} plant for LHCb-RICH1. The numbers in the table are calculated considering systems without presence of gas mixture leaks.

	RPC ($C_2H_2F_4$)	**CSC (CF_4)**	**LHCb-RICH1 (C_4F_{10})**
Gas recuperation plant efficiency (%)	80	65	85
Operational cost with gas recirculation and recuperation (kCHF/year)	65	33	1
gas recuperation system—construction costs (kCHF)	400	300	200
gas recuperation system maintenance and operation (kCHF)	40	40	10
Return of investment for gas recuperation system (years of operation)	≈1.6	≈4.1	≈35.2
tCO_2e emission with gas recirculation system (tCO_2e)	34,447	15,706	266
tCO_2e emission with gas recirculation system (tCO_2e)	8738	5500	40

4. New Ecofriendly Gases

The third research line is based on the long-term replacement of currently used gases for the operation of gaseous detectors [14].

As it was already the case in the past when Ozone Depletion Gases were banned, under the challenges represented by the F-gas regulations, several alternative gases have been developed by industry as alternative to GHGs in refrigerant and HV insulation medium applications. However, their behavior as particle detection medium is not straightforward.

New recently developed gases are part of the Hydrofluoro-Olefine (HFO) and Novec families. They are characterized by longer Carbon chains and, in some cases, by the presence of double Carbon-Carbon bound. Their GWP is very low, close to 1 which makes them very interesting. However, the lower the GWP and the lifetime in atmosphere, the easier these molecules are going to react by creating sub-products and disappearing. The three factors that determine the atmospheric lifetime are: rain out (water solubility), oxidation (reactivity with OH) and photolysis (UV absorbance). These three mechanisms are present also in most gaseous detectors and their effects must be considered for detector operation when eco-friendly gases are used.

Additional challenges related to HFOs use in particle detectors are related to the flammability of many of these low GWP gases, to their low vapor pressure at ambient temperature and, in the longer term, to their classification as PFAS substances.

HFOs are classified in the ASHRAE 34 Safety group (American Society of Heating, Refrigerating and Air-Conditioning Engineers) as A2L which means they are refrigerants with low toxicity and low flammability. However, mildly flammable might still be an issue and require specific components and design for installation.

The low vapor pressure (below 1 barg) might lead to condensation in devices used to control the gas flow or other instability in the mixture.

PFAS are per- and polyfluoroalkyl substances, which contain at least one fully fluorinated methyl (CF3-) or methylene (-CF2-) carbon atom (without any H/Cl/Br/I attached to it). In 2023 the European Chemicals Agency (ECHA) released a proposal regarding restrictions on PFAS. The proposal envisages covering over 10,000 different PFAS, which are considered environmental pollutants with links to harmful health effects. Following these considerations, the 3M company, that developed the Novec gas family, decided to stop their production from 2025. However, for the moment, these gases are still produced by other companies.

In 2025 ECHA released an updated version of the PFAS restriction proposal [15] and it plans to launch a consultation in spring 2026 inviting participants to respond to questions on the potential impacts of restricting the use of PFAS across various sectors. Participants will also be asked to provide specific information about the availability and feasibility of alternatives to these widely used chemicals [16]. However, the impact on the use of PFAS gases for particle detectors is not clear at the moment. The updated ECHA restriction proposal contains a potential 12 years derogation and it will probably cover the full LHC operational period. What will come after, it is even more difficult to predict and a lot will depend on how the industrial sector will react, especially if non-PFAS alternatives will be found. Refrigerant industry is now reconsidering the use of CO_2 for refrigerant plants. Indeed, CO_2 systems were originally discarded due to their higher operating pressures compared to other

commercial refrigerants, but now this limiting factor is solved thanks to technological developments. This might limit the interest in developing new refrigerant families in the far future.

To replace GHGs two research lines are ongoing. The first challenge is represented by the GHGs replacement in detector systems currently in operation at the LHC experiments. Here constraints are coming from the fact that detectors and infrastructure are already installed: for example, it is not easy to replace high voltage connectors or cables or Front-End electronics (FEE), but the new ecofriendly mixture has to work with the current system as it is.

The second research line should look at how to design new detectors for future experiments. Here the design process should balance correctly detector performance and related infrastructure. A new detector design with new FEE might make easier the use of ecofriendly gases. However, it remains important to have reliable systems for the gas mixture distribution that minimize the risk of developing any gas leak.

Since large part of the GHGs emission (about 80%) from current large experiments is coming from the ATLAS and CMS RPC detector systems at the CERN LHC, a particular effort has been injected into the search for new ecofriendly gas mixtures for the RPC detectors. Studies are performed in laboratories where new gas mixtures are characterized, and, after, at the CERN Gamma Irradiation Facility (GIF++) [7] where detector performance (instantaneous and long term) can be evaluated in presence of high radiation simulating the background expected during High Luminosity LHC operation.

4.1. Addition of Inert Gas to GHG Based Mixtures

A mitigation strategy based on the addition of inert gas to the standard RPC gas mixture has been pursued to reduce the GHGs emissions from the RPC detector systems at a short time scale without changing completely the gas mixture and maintaining the detector performance. The first inert gas considered was Helium, but due to presence of photomultipliers in the experiments and presence of leaks at the detector level it was considered too risky and therefore discarded. As alternative to Helium, CO_2 was considered. Since 2023, the ATLAS RPC detector system has reduced the GHG impact of the standard RPC gas mixture made of 94.7% R134a, 5% $i C_4H_{10}$ and 0.3% SF_6 by replacing part of R134a with 30% CO_2 [17]. The addition of CO_2 increases the streamer probability, and it broadens the charge distribution, as it is visible in Figure 6. The addition of 1% SF_6 brings the streamer probability to 2.9% (with 0.6% SF6, the streamer probability is 3.9%) compared with 0.8% for the standard mixture.

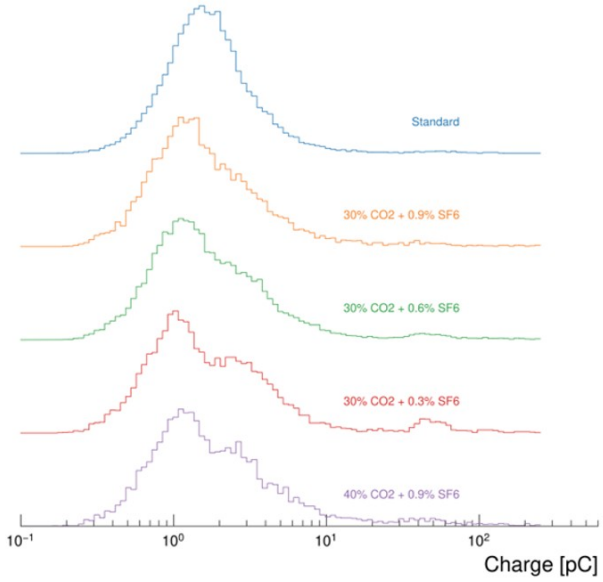

Figure 6. Prompt charge distribution for the standard gas mixture compared with different additions of CO_2 and SF_6 at detector's working point.

During tests where the detectors were exposed to high intensity radiation background at the GIF++ facility, it was also observed that detectors operated with the CO_2-based gas mixture with 1% SF_6 have higher current with respect to the standard gas mixture (about 15% more as visible in Figure 7). Moreover, with the CO_2-based gas mixture the High Voltage working region with low streamer probability is about half with respect to the standard RPC mixture (Figure 8). In conclusion, the CO_2-based mixture is a compromise between single detector

performance and reduction of GHG emissions (indeed, the performance of the RPC systems as whole were never affected). In addition, long-term performance studies are necessary to evaluate any possible effect.

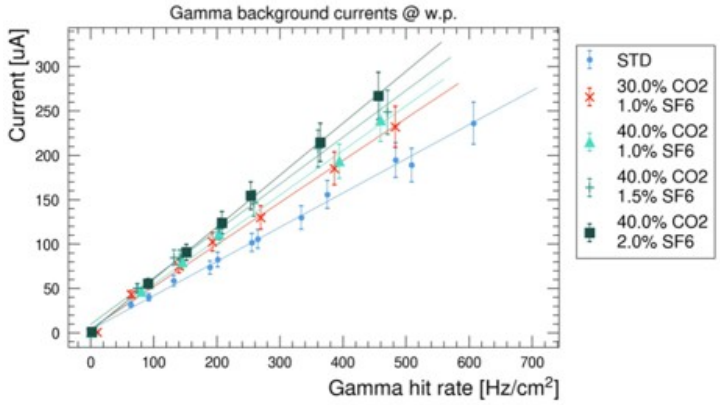

Figure 7. RPC detector current operated with different CO_2 and SF_6 concentrations as a function of the gamma background radiation.

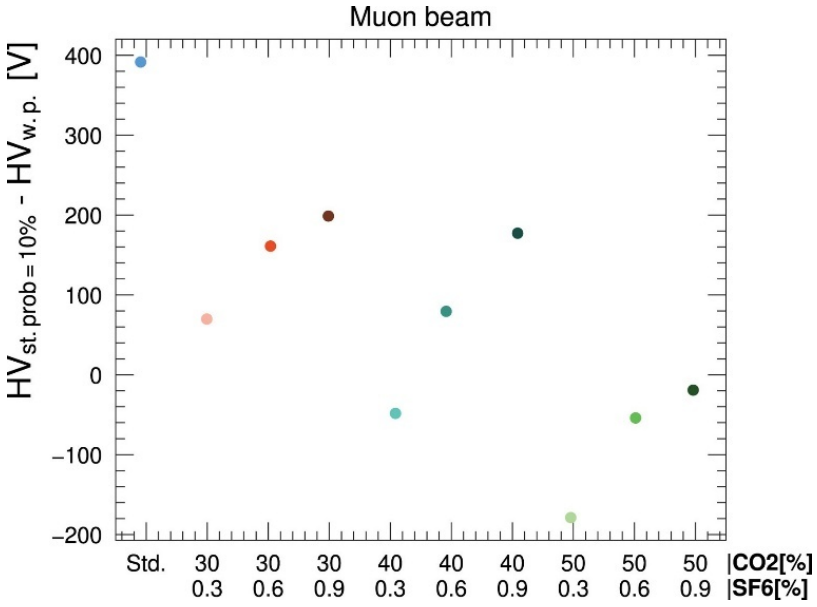

Figure 8. Width of RPC detector High Voltage working region with streamer probability below 10% as a function of CO_2 and SF_6 concentrations.

By using the new CO_2-based gas mixture a 15% reduction of GHG emission was achieved by the ATLAS-RPC detector system. In addition, the reduced R134a consumption brought a cost saving of 100 kCHF per year. From 2025, the SF_6 concentration was reduced to 0.5%, which brings to a total 25% reduction of GHG emissions.

The strategy of adding some CO_2 to the gas mixture was also adopted by LHCb-RICH2 detector where 8% CO_2 was used to partially replace CF_4.

4.2. R134a Replacements

Following the development of new ecofriendly gases, driven by the refrigerant industry, the best candidate for replacing R134a seemed to be one refrigerant of the HFO family, particularly the HFO1234ze. However, when HFO1234ze is used in place of R134a, the high voltage working point is shifted towards too high values for the current ATLAS and CMS RPC systems. To bring back the detector efficiency range towards usual applied high voltage values (compatible with the current high voltage infrastructure), the addition of Helium or CO_2 was tested. Also maintaining some R134a in the mixture together with HFO helps lowering the detector dark current and high voltage working point. Good results have been obtained with mixtures where the R134a was only partially replaced by HFO-1234 (Figure 9) [18]. In particular, the five components mixture HFO-1234 37.45%–R134a 37.45%–$i$$C_4H_{10}$ 4.5%–SF_6 0.6%–He 20% shows an efficiency plateau without streamers almost equivalent to the standard

RPC R134a based mixture. Increasing the HFO-1234 concentration with consequent reduction of R134a produces a reduction of the operation region without streamers.

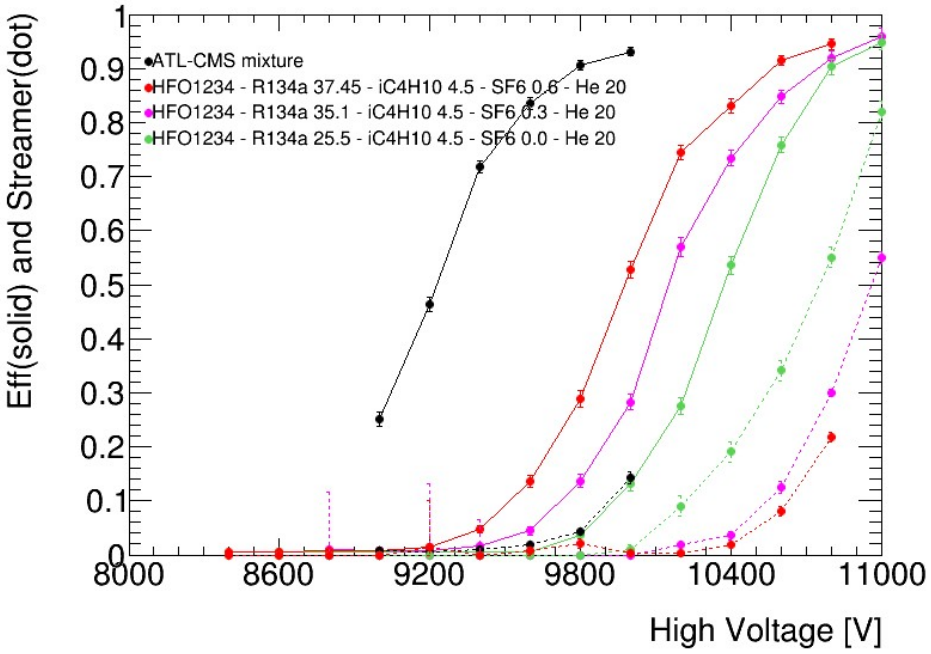

Figure 9. Comparison of RPC detector efficiency and streamer probability for HFO-1234-R134a based mixture with the addition of Helium and standard R134a based mixture.

Unfortunately, due to the presence of important leaks, the use of Helium was considered too risky for the photomultipliers used in the calorimeters. Different CO_2 concentrations were added (always maintaining 5% iC_4H_{10} and 1% SF_6). Most relevant mixture tested are the so called ECO1 (45% HFO), ECO2 (35% HFO) and ECO3 (25% HFO). Figure 10 shows efficiency and charge distribution. ECO1 has clearly a too high voltage working point. ECO2 and ECO3 have higher currents (almost double), a charge distribution containing a significant fraction of large signals and a high streamer probability (at least a factor 10 higher for ECO3) compared with the standard mixture. ECO2 has been selected for long-term aging studies. A possible compromise between performance and reduction of GHG emissions, can be a mixture where a fraction of R134a is still maintained. For example, Figure 10 shows also results obtained for a mixture containing 22% HFO, 22% R134a, 50% CO_2, 5% iC_4H_{10} and 1% SF_6: the working point is acceptable, and streamer probability is similar to the one of standard gas mixture. Obviously the GWP of this gas mixture is higher with respect to the other two but it could represent a mid-term solution.

Before any application in the experiments, a long-term aging study for RPC operated with eco-friendly gas mixtures under high background radiation is mandatory. This study is on-going at CERN-GIF++ in the framework of the EcoGas@GIF++ collaboration [19]. Detector currents under irradiation are monitored every week. Detectors' performances are measured during test-beam periods (typically 3 times per year). A total integrated charge of about 200 mC/cm^2 has been collected up to now by all detectors. The expected integrated charge for High Luminosity LHC is 1 C/cm^2 for CMS-RPC, safety factors included. Preliminary results show an increase in current and a shift towards higher high voltage values (about + 200 V) of the working point. Further analysis is ongoing to find any possible explanation. Concerning gas mixture quality and production of impurities, studies performed under high radiation background and in gas recirculation show that HFO-1234 breaks more easily than $C_2H_2F_4$ creating several impurities and Hydrofluoric acid (HF) [20,21].

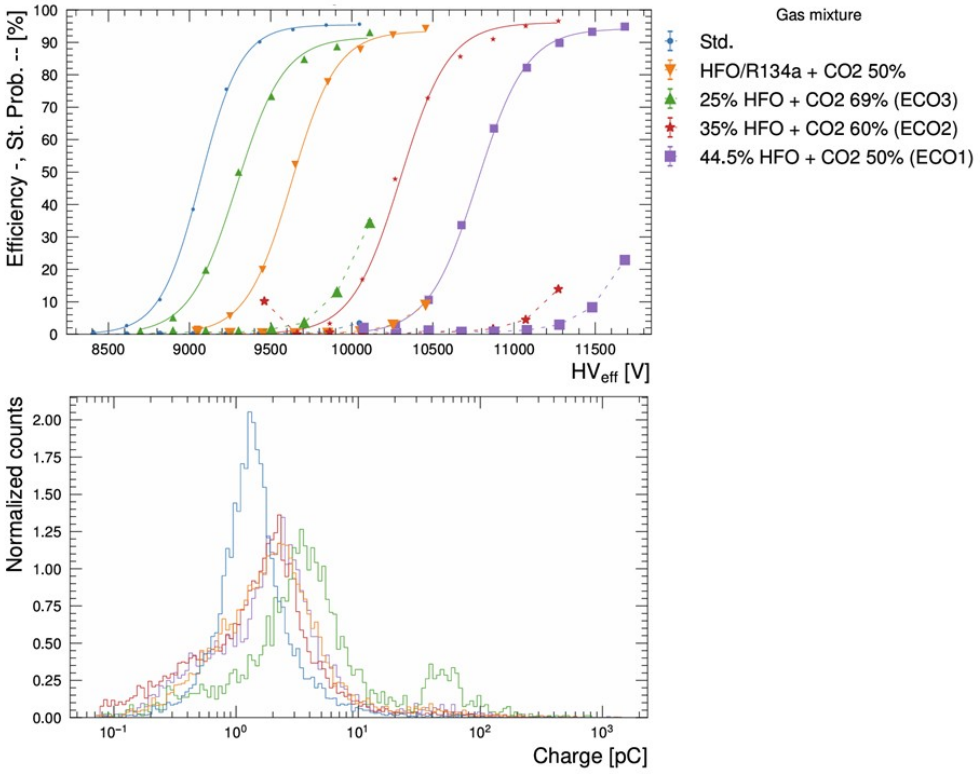

Figure 10. Efficiency and charge distribution for different HFO/CO_2 based mixtures and for a mixture where R134a is only partially replaced by HFO and CO_2.

4.3. SF_6 Replacements

SF_6 is used in relatively low concentrations (between 0.3% up to 7%) but, given its very high GWP, and the fact that the updated EU F-gas regulation now includes SF_6 in the phasing-out policy, it is important to look for possible alternatives. SF_6 is mainly used by the electrical power industry (about 80% of the total production), mostly as a gaseous dielectric medium. So, it is natural to look for alternatives in new ecofriendly gases developed for such application. Novec 4710 and Novec 5110 are perfluorinated compound developed by 3M as a high-dielectric gas for high-voltage switchgear and promoted as an alternative to SF_6 for interruption and insulation applications, as they have better insulation properties than of SF_6 and a relatively low GWP (2100 for the 4710 and about 1 for 5110). Novec 4710 showed excellent performance already at very low concentration when used to replace SF_6 in RPC detectors like the ones installed in the ATLAS and CMS experiments. Figure 11 shows charge distribution, efficiency curve and streamer probability for mixtures containing Novec 4710 in different concentrations. Despite the good performance, Novec 4710 has two weak points: it has still a relatively high GWP and, most critical, it is a PFAS. In addition, Novec 4710 reacts with water and, even if at very low concentrations, it produces an amide which is solid at room temperature [22,23], while Novec 5110 is sensitive to UV radiation. Water vapor and UV radiation and both present in RPC detectors during operation.

Other alternatives to SF_6 considered are the Hydrochlorofluoro-Olefine (HCFO) 1224yd and 1233zd, both having GWP lower than 1. These gases were developed by the AGC Chemical company as next-generation environmentally friendly solvent and refrigerant. They are commercialized under the name of AMOLEA$_{TM}$. Both HCFO1224yd (Figure 12) and HCFO1233zd [24] show good performance. Studies are ongoing to evaluate long term performance and to exclude any risk deriving from the presence of Chlorine and the potential production of HCl during detector operation (a mechanism very similar to the one which leads to the formation of HF from Fluorine). Since AMOLEA$_{TM}$ gases contain Chlorine, they have the advantage of not being part of the PFAS family and therefore not subject to potential restrictions.

Other gases tested are CF_3I (good performance, but toxic and mutagenic) and C_4F_8O (too high GWP, about 8000).

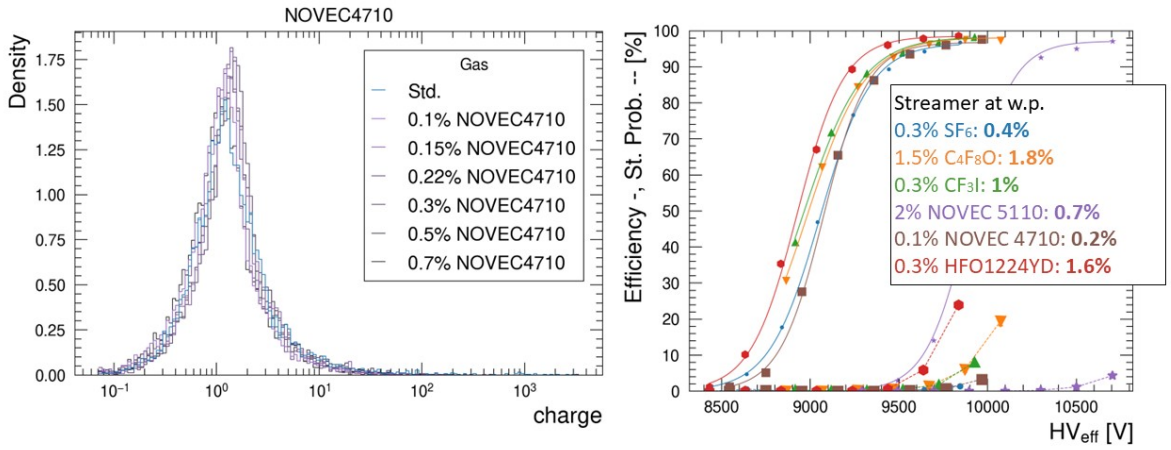

Figure 11. Charge distribution, efficiency curve and streamer probability for mixtures containing Novec 4710 in different concentrations and other possible ecofriendly replacement for SF_6 (NOVEC 5110, AMOLEA$_{TM}$ HCFO1224yd, C_4F_8O, CF_3I).

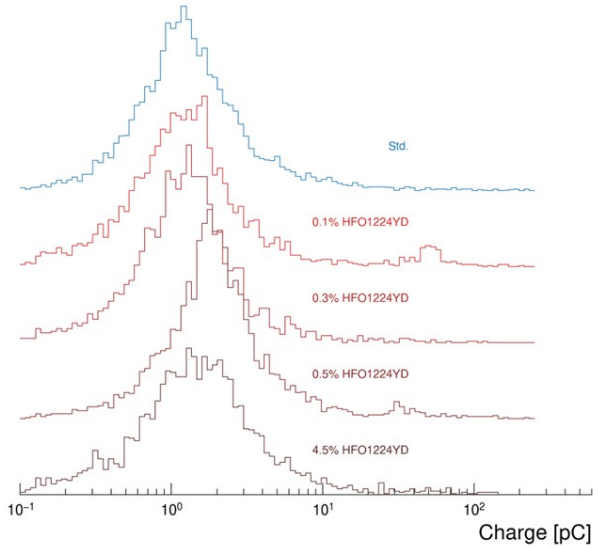

Figure 12. Charge distribution for mixtures containing AMOLEA$_{TM}$ HCFO1224yd in different concentrations.

Table 3, Figures 13–15 show working point, streamer probability at working point and voltage working region width for several mixtures as a function of GWP and $kgCO_2e/m^3$. For particle detector applications, where the concentrations are expressed in volume, it might be more representative to consider the equivalence in $kgCO_2e/m^3$ rather than GWP since the latter is considering concentration in weight.

Unfortunately, Figures 13–15 clearly show how better detector performance are associated with mixtures characterized by higher GWP or $kgCO_2e/m^3$. This is probably related to the need of having stable molecules (which implies high GWP) with strong electron affinity (i.e., high number of Fluorine or other Halogen atoms). However, looking for a possible compromise which does not contain GHG, the ECO2 mixture seems to be confirmed as a reasonably good choice to be explored.

Table 3. Summary of working point, streamer probability at working point and voltage working region width for several mixtures as a function of GWP and $kgCO_2e/m^3$. Negative values for the voltage working region width mean that 10% of streamers are already present below the working point.

	GWP	$kgCO_2e/m^3$	HV Working Point (V)	Streamer Probability (%)	HV Working Region Width (V)
Standard RPC mix 94.7% R134a 5% iC_4H_{10} 0.3% SF_6	1482	6612	9600	0.8	500

Table 3. *Cont.*

	GWP	$kgCO_2e/m^3$	HV Working Point (V)	Streamer Probability (%)	HV Working Region Width (V)
CO_2 30%–SF6 0.9% 30% CO_2 64.1% R134a 5% iC_4H_{10} 0.9% SF_6	1491	5512	9300	2.9	200
CO_2 30%–SF6 0.6% 30% CO_2 64.4% R134a 5% iC_4H_{10} 0.6% SF_6	1378	5086	9100	3.9	160
ECO1 49% CO_2 45% HFO1234ze 5% iC_4H_{10} 1% SF_6	434	1496	11,600	18	−100
ECO_2 60% CO_2 35% HFO1234ze 4% iC_4H_{10} 1% SF_6	478	1494	11,000	4	150
ECO3 70% CO_2 25% HFO1234ze 4% iC_4H_{10} 1% SF_6	530	1493	9900	15	−100
HFO-R134a-CO_2 50% CO_2 22% HFO1234ze 22% R134a 5% iC_4H_{10} 1% SF_6	887	2924	10,400	6	125
HFO-R134a-He 20% He 37.45% HFO1234ze 37.45% R134a 4.5% iC_4H_{10} 0.6% SF_6	886	3338	10,450	1.8	400
NOVEC 4710 0.1% NOVEC 4710 95.4% R134a 4.5% iC_4H_{10}	1394	6230	9600	0.2	400
NOVEC 5110 2% NOVEC 4710 93% R134a 5% iC_4H_{10}	1316	6056	10,400	0.7	450
AMOLEA 1224yd 0.3% AMOLEA 1224yd 94.7% R134a 5% iC_4H_{10}	1382	6167	9500	1.6	225

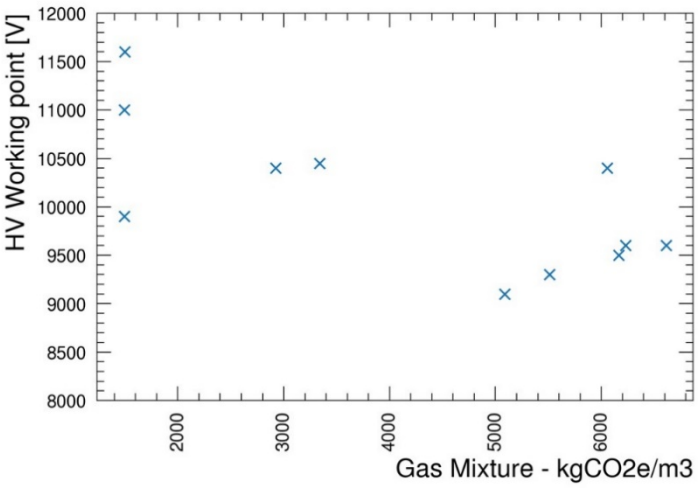

Figure 13. High Voltage working point as a function of gas mixture type expressed in $kgCO_2e/m^3$.

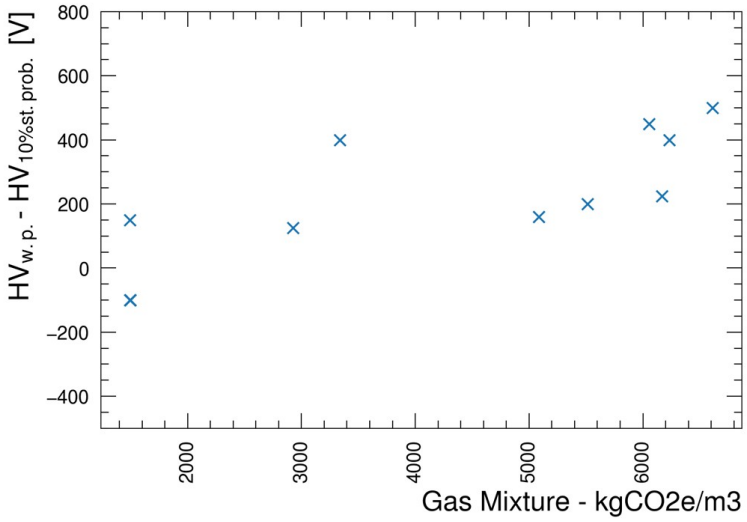

Figure 14. Width of RPC detector High Voltage working region with streamer probability below 10% as a function of gas mixture type expressed in $kgCO_2e/m^3$.

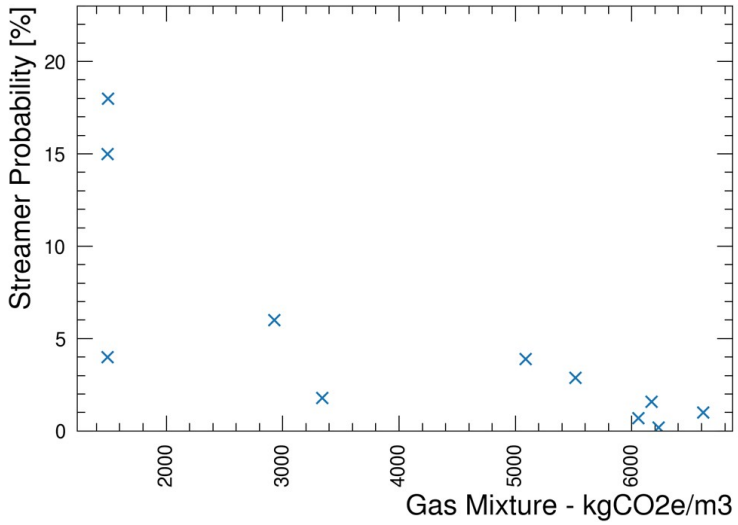

Figure 15. Streamer probability at working point as a function of gas mixture type expressed in $kgCO_2e/m^3$.

4.4. Other GHGs Used in Particle Detectors

CF_4 is another GHG quite extensively used in current detectors and in new detector technologies prototypes for future experiments. It is currently used to mitigate aging effects in Cathode Strip Chambers (CMS-CSC) and Multi Wire Proportional Chambers (LHCb-MWPC). In the latter and in Micro pattern Gaseous detectors it is used to increase the drift velocity and therefore to improve the time resolution.

CF_4 as well as other Perfluorocarbons (for example C_4F_{10}, C_3F_8, C_2F_6) are used as Cherenkov radiator in current detectors (LHCb-RICH2, LHCB-RICH1) as well as tested for future detector applications (for example, the dRICH detector of the ePIC experiment [25] or the new Picosec detector concept [26]).

In all cases, it seems quite difficult to find an ecofriendly replacement in the short term. C4H10 has been considered as radiators in place of C_4F_{10}, but it is flammable and the currently installed RICH1 detector cannot use flammable gas (both electronics and leak tightness need to be revised). The use of CO_2 is under investigation in place of CF_4, but since it has a lower refractive index, it could affect particle identification resolution.

No clear candidate has been identified for replacing CF_4 in micropattern gas detector technologies where the time resolution needs to be fully exploited.

In general, new gases are still not fully characterized. Fundamental parameters for HFO1234ze have been measured only very recently [27].

In addition, the observed gas properties important for detector operation have never been closely linked to the fundamental chemical structure. A better understanding of this link could probably help in identifying potential ecofriendly gases.

5. GHGs Disposal

Gas abatement systems are industrial solutions available on the market for the on-site disposal of GHG [28,29]. They are adopted when GHGs used in industrial process are polluted to a level where recuperation for re-use is not possible. In particle detector applications gases pollutants might be present but only at relatively low concentrations. Therefore, gas recirculation and recuperation should be considered as first solutions. In any case, this approach limits the amount of gas to dispose of. Gas disposal might be needed in the future given the increasing importance of the CO_2 footprint for companies' supplying gases and the subsequent request to go to zero emissions.

A feasibility study for the implementation of an exhaust management system was performed by the CMS experiment in 2018 [30]. In the context of the study, two running systems were visited at the French Alternative Energies and Atomic Energy Commission (CEA) laboratory in Grenoble. They were both used for treating exhaust from semiconductor processes.

Perfluorocarbons and hydrofluorocarbons are very stable compounds and therefore very difficult to abate. Their stability is also responsible for the long lifetime of these products in the atmosphere. A big push is required to break the C-F bond and therefore large quantity of fuel (CH_4 or city gas) and O_2 are needed. In the case of the RPC detector mixture, the reaction produces Hydrofluoridric acid and sulfur dioxide. Since water is used to trap the produced volatile components, the waste product of the abatement systems is water contaminated with hydrofluoridric, sulphurous and sulphuric acids. To treat wastewater produced, a dedicated plant is needed. Nowadays companies exist that collect containers for GHG disposal. However, despite the apparent simplicity of abatement systems, a complex and heavy infrastructure is needed for their operation. Safety aspects and costs related to the transport of compressed exhaust gas or heavily polluted wastewater produced by abatement plants should be carefully evaluated. For this reason, the abatement systems should be left as the last option after having implemented all the others (gas recirculation and recuperation) and therefore after having reduced to the minimum the gas volume to dispose of.

6. Conclusions

With the growing concern on GHG emissions and the implementation of F-gas regulation, it is fundamental for current and future experiments to minimize the usage of GHG and, in the long term, to find ecofriendly gas mixtures. Mitigation strategies for a short-term reduction of GHG usage are based on gas recirculation and gas recuperation plants. Gas recirculation can basically bring down the GHG usage almost to zero. Only limitations could come from presence of gas mixture leaks at detector level or specific detector requirements. In any case, reductions up to 90–95% are easily achieved. The use of gas recirculation system should not be limited to large experiments but extended to medium experiments as well as laboratory and detector test facilities. In general, whenever a GHG is used, the mixture distribution system should be based on gas recirculation or a system capable to collect the return mixture should be put in place to avoid any intentional release of GHG in atmosphere.

For the remaining quantity that cannot be recirculated (between 5–10%) the use of gas recuperation systems should be considered. Recuperation plants able to extract CF_4, R134a, C_4F_{10} have been developed for specific gaseous mixture used by particle detectors at the LHC experiments. After a challenging R&D phase, the operation of these systems has brought several remarkable positive effects. First, a total reduction of almost 30000 tCO_2e/y has been achieved which represents about 25% of total emissions from CERN particle detection activities [31]. Moreover, thanks to the budget saved by using recuperated gas, the plants are paid back in few years of operation. Last, but more important, decreasing GHG consumption makes detector operation less subject to crisis affecting price and availability, already experienced in the past for CF_4, R134a. For example, during the worldwide CF_4 shortage in 2022 the CMS experiment was able take data only thanks to recuperated CF_4.

Since gas recuperation plants might be really specific for a given mixture, their application should be considered case by case. If a recuperation plant has been already developed for the mixture used, then the use of the recuperation plant should be evaluated over the full experiment or facility operational period: even if initial investment and operational costs are paid back only in the full duration of the facility, it might be worth to use it. Indeed, it must be considered that for GHG availability and price are going to be a challenge in the years to come. Therefore, any system able to limit their consumption and emission should be put in place considering the expected duration of the facility and not only the return of investment in few years. Of course, it is more complicated if a

recuperation plant does not exist for the specific mixture used. In this case, the cost for R&D phase should be considered.

At long-term, it is clearly necessary to look for ecofriendly replacement of currently used GHGs. The role played by gas recirculation and recuperation plants will remain always important because they allow to minimize the operational budget in case expensive gases are used. Ecofriendly alternatives have been developed by the refrigerant industry or as high voltage insulation medium, but their application to particle detectors is not straightforward. This is particularly true for the current systems where additional constraints come from the high voltage systems or from the front-end electronics. New detector technologies under development in view of future experiments should look at detector design that might facilitate the use of ecofriendly gas mixtures. For future detectors, not only the limitations imposed by the F-gases regulations should be considered, but also the increasing concern on the use of PFAS, since some of the current ecofriendly alternatives unfortunately are PFAS.

For the future of gaseous detector technologies, it is important to support all research aiming in reducing GHGs usage and to share results and developed technologies among institutes. It is also fundamental to support these activities in developing countries.

Author Contributions

R.G. is leading the gas systems project at CERN. He participated to the development of gas systems for the experiments at the LHC since the early phase. R.G. proposed the strategies for minimizing the greenhouse gas usage outlined in the present article to the gaseous detector community. R.G. designed the CF4, R134a recuperation plants and guided the development of the other plants. R.G. and his team are a reference in the search and test for new ecofriendly alternative. The author has read and agreed to the published version of the manuscript.

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Data Availability Statement

Data will be made available on request.

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Conflicts of Interest

The author declares no conflict of interest.

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