September 22, 2023

Non-confidential comments for Annex XV restriction report: Alternatives to fluorinated gases

Dear Madame or Sir,

We welcome the opportunity to provide input on the restriction proposal regarding per- and polyfluoroalkyl substances (PFAS). This submission was prepared by Dr. Juliane Glüge (ETH Zürich) with input of members of the Global PFAS Science Panel (GPSP). The GPSP is a collaborative partnership among academic researchers, regulatory scientists and policy analysts dedicated to enhancing understanding of PFAS and to protecting human and environmental health from potentially adverse effects associated with PFAS through better informed decision-making. The scientific work of the GPSP in the last five years has considerably advanced the understanding of PFAS and the GPSP would like to take the opportunity to also give input to the PFAS restriction proposal.

We see the restriction proposal as a very important initiative and good way forward to counteract the ongoing emissions of PFAS in Europe (and in part even worldwide). Our submissions should therefore be seen as a support of the restriction proposal to make it even stronger and more fully complete. The current submission is on Alternatives to fluorinated gases.

1) Alternatives to fluorinated gases as propellants for technical aerosols

A potential derogation in the PFAS restriction proposal marked for reconsideration is ‘propellants for technical aerosols for applications where non-flammability and high technical performance of spray quality are required’. A propellant is a substance used in the form of a pressurized gas for the production of energy that is subsequently used to create the movement of a fluid (Mosil 2021). There are two main types of aerosol can propellants. One is compressed gas aerosol can propellants and the other liquid gas aerosol can propellants.

The most common form of aerosol can propellant is a liquefied gas (liquid gas aerosol can propellants) such as butane, isobutane, propane or a fluorinated gas (Justrite 2023). The gas is turned into liquid by pressurizing it within the can, providing constant pressure levels throughout the life of the can. A disadvantage is that aerosols with liquid gases as a propellant have a relatively low quantity of product concentrate and more of liquified gas (PURmate 2023). Additionally, large temperature changes affect the pressure in the can (Mosil 2021). Also, hydrocarbons that are used as liquid gas propellants are flammable and can only be stored in limited quantities (PURmate 2023).

Compressed gas aerosol can propellants include for example CO_2 or nitrogen which are both very cheap and not flammable. Also, temperature changes have little effect on the pressure. However, as the product within the can is used, the pressure within the can decreases, and the can will have less dispersing power because of it (Mosil 2021). Also, compressed gas propellants act like pistons and contain large wet particles (Mosil 2021). In addition, CO_2 is incompatible in pressurized cans with water or traces of water (PURmate 2023).

This shows that no single solution works for all applications. A derogation for ‘propellants for technical aerosols for applications where non-flammability and high technical performance of spray quality are required’ might therefore be justified; however, it would be important to define the derogation more precisely, stating in which applications exactly fluorinated propellants may continue to be used. The current wording is too vague and could be misused for cases where non-fluorinated alternatives are actually suitable and available.

2) Alternatives to fluorinated gases as refrigerants – proposed derogations

2.1) Refrigerants in low temperature refrigeration below -50 °C

The restriction proposal proposes a derogation for 5 years after EiF for ‘refrigerants in low temperature refrigeration below -50 °C’. It is stated in the restriction proposal that ‘Alternatives for the use are available and technically feasible. However, according to stakeholder input alternatives may be less flexible with regards to operating temperature ranges.’

We have conducted a market search and can confirm that various ultra-low and low temperature freezers from different manufacturers are on the market that use hydrocarbons as refrigerants.

Available systems on the market include e.g., ultra-low freezer (−86 °C to −40 °C) from Tecnomara that use a cascade system of propane (first level) and a mixture of propane and ethane (second level) (Tecnomara 2023), and ultra-low temperature freezers with propane from Liebherr (Liebherr 2023). Other manufacturer that offer ultra-low temperature freezers with hydrocarbons as refrigerants include Meling (Meling 2023a, 2023b), VWR (VWR 2023), and Carlo Erba Reagents (CarloErba 2023). Cryogenic ultra-low temperature freezers with hydrocarbons are also on the market, e.g., from PHCBI (PHCBI 2023), Haiers Biomedical (HaierBiomedical 2023), and Linde (Linde 2023). Even cold rooms can be operated with non-fluorinated refrigerants. The Odense University Hospital in Denmark inaugurated a cold-storage room with –80°C for storing tissue samples in 2012. The refrigeration system is built as an indirect cascade system using ethane and ammonia (Johnson_Controls 2015).

However, fluorinated gases are also used in centrifuges that require cooling (EFCTC 2023) and in specific test systems that require heating and/or cooling (Weisstechnik 2023). Some of these systems might be sold in very low numbers (e.g., specific centrifuges) so that it has not been cost-effective so far to develop new systems with non-fluorinated refrigerants.

Based on this information we think that a derogation for low temperature refrigeration might be justified for very specific use cases such as centrifuges or specific test systems. However, the derogation suggested in the PFAS restriction proposal 'refrigerants in low temperature refrigeration below -50 °C' is too broad in our point of view and should be made more specific.

2.2) Refrigerants in transport refrigeration other than in marine applications

The restriction proposal proposes a derogation for 5 years after EiF for 'refrigerants in transport refrigeration other than in marine applications'. It is stated in the restriction proposal that 'Some non-PFAS alternatives are already in use in trucks, in trawlers and in reefer containers. However, they are not currently widespread. Alternatives may have lower energy efficiency, and safety is of concern as people are carried together with the goods to be cooled. Specific barriers affect the sector, for example, size limitations are problematic for the use of active CO_2 systems given the layout of existing trucks. Further design work would be needed to provide viable alternatives that are widely applicable across the market.'

We do not entirely agree with this assessment. It might be true that the alternatives are not yet widespread, but they exist and provide viable alternatives that can and should be used in the future. For trucks, a roof-mounted refrigeration unit has been developed by the German company ECOOLTEC that applies propene and CO_2 (R-744) (Ecooltec 2022). Propene is used inside the active outdoor refrigeration loop, while the air inside the cargo compartment is either heated or cooled by CO_2. The cargo-air/CO_2 heat exchanger is operated indirectly by the heat pipe principle, whereby CO_2 is utilized as the working fluid between the cargo and the refrigeration unit.

Also, the Austrian company PBX has tested a propane-based refrigeration unit, called M24, for light commercial electrical vehicles (Atmo 2023). Container refrigeration systems applying CO_2 as working fluid have been on the market since 2010 (Carrier 2023). The NaturaLINE® unit was the world's first container technology that used non-fluorinated refrigeratns. In the meantime, additional transport refrigeration systems not based on fluorinated gases have been developed and are summarized in Cooltechnologies (2023) and EIA (2021).

It is also stated in the restriction proposal that 'Refrigeration systems used under transport especially need high energy efficiency to travel long distances. Trucks drive from the Arctic Circle to southern Spain and are exposed to great fluctuations in outside temperature, while safety of the refrigeration system is also crucial as it carries people and various materials. There is no non-PFAS alternatives yet available to meet the required performance, energy efficiency and safety.' Again, we do not entirely agree with this statement. Hydrocarbons behave thermodynamically the same or better as hydrofluorocarbons and therefore have no other (severe) limitations than those given by their flammability. In the ECOOLTEC system, the use of CO_2 excludes the formation of a flammable mixture in the closed structure, thus minimizing the risk of the application. The roof-mounted refrigeration unit developed by ECOOLTEC is therefore a viable alternative.

We therefore do not see the need for a derogation for 'refrigerants in transport refrigeration other than in marine applications' and request that this derogation is withdrawn from the restriction (proposal).

2.3) Refrigerants in HVACR-equipment in buildings where national safety standards and building codes prohibit the use of alternatives

The restriction proposal proposes a time-unlimited derogation for 'refrigerants in HVACR-equipment in buildings where national safety standards and building codes prohibit the use of alternatives'. While we understand that ECHA and the EU Commission cannot really influence the national safety standards and building codes, we would like to suggest a) to introduce reporting obligations for this derogation and b) to introduce an obligation for the member states using the derogation to update their national safety standards and building codes every 5 to 10 years to base the standards on the latest knowledge and practice.

3) Alternatives to fluorinated gases as refrigerants – general information

We have seen from the discussion in the media that some industry stakeholders see fluorinated gases as critical components of our daily life. We have therefore prepared a scientific publication on alternatives to fluorinated gases as refrigerants. The publication is currently under review with the journal Environmental Science: Processes and Impacts. The version of the publication that we submitted to the journal is attached to our comment to the public consultation as confidential attachment. The following paragraphs summarize the findings from the publication and draw some conclusions for the PFAS restriction proposal.

3.1) General points

Refrigeration, air conditioning, heating and other uses of heat-transfer fluids make up 74% of all uses (by mass) of F-gases (EEA 2021) and a significant increase in the demand for refrigerants is expected (due to the heat pump boom and increased comfort requirements for air conditioning) in the next years (McLinden et al. 2020). The use categories in the attached publication are therefore important areas for reducing the use of F-gases. This is significant for two reasons: (1) F-gases represented 2.3% of the total EU greenhouse-gas emissions in 2019 (EEA 2021) and reducing their emissions is an important element in efforts against global warming (Bai et al. 2023; Velders et al. 2015). 90% of the F-gases in the EU are HFCs (EEA 2021) and many of them are included in Annex I of the current F-gas Regulation and subject to the quota system. However, some of the other fluorinated greenhouse gases such as R-1234yf (GWP 0.5) or R-1234ze (GWP 1.37) are not covered by the quota system and are used as alternatives to HFCs with a higher GWP. Reducing their emissions is also important, because (2) certain F-gases form trifluoroacetic acid (TFA). Given the increasing levels of TFA in the last decade, emission reductions for compounds that degrade into TFA are urgently required. Reducing the amount in use of R-1234yf (forming 100% TFA), R-1234ze(E) (forming up to 10% TFA) and R-134a (7–20% TFA) (Behringer et al. 2021) is therefore also urgently needed.

In addition to the emissions from the F-gases themselves, it has also been shown that atmospheric abundances and emissions of five CFCs (CFC-13, -112a, -113a, -114a and -115) increased between 2010 and 2020 (Western et al. 2023). In terms of impact on climate, the emissions of these five CFCs estimated for 2020 are equivalent to 47 ± 5 Tg CO_2 equivalent yr^{-1}, which is the same amount as the 2020 CO_2 emissions of Switzerland (PHYSORG 2023). These five CFCs are allowed under the Montreal Protocol in the production of other chemicals. Previous work has linked emissions of CFC-113a and CFC-115 to the production of R-125 (Adcock et al. 2018; Laube et al. 2014; UNEP 2018; Vollmer et al. 2018); CFC-113a and CFC-114a are intermediates in a production pathway of R-134a. Replacing R-134a and R-125 with non-fluorinated refrigerants in air conditioners and other refrigerant applications would therefore very likely also reduce these highly problematic CFC emissions.

The following subsections summarize the use categories where fluorinated gases are still used as refrigerants and show which non-fluorinated alternatives are available.

3.2) Refrigeration

Various refrigeration systems are on the market. In analogy to the current F-gas Regulation (EP 2014), they are divided here into domestic refrigeration, commercial refrigeration (stand-alone systems and multipack centralized systems), industrial refrigeration, transport refrigeration of goods, refrigeration in ice rinks and ultra-low and low temperature freezers.

Table 1 gives an overview of which non-fluorinated refrigerants are already used in commercially available refrigeration systems. The list is not exhaustive. Specific brand names and links to references are provided in the attached publication.

Table 1: Overview of which non-fluorinated refrigerants are already used in commercially available refrigeration systems. The list is not exhaustive. An 'x' indicates the availability of the non-fluorinated refrigerant in a commercially available refrigeration system.

	CO_2	Ammonia	Propane	Isobutane	Others
Domestic refrigeration				x	
Stand-alone refrigeration systems in commercial stores	x	x	x	x	x
Multipack centralized refrigeration systems in commercial stores	x	x	x		
Industrial refrigeration	x	x			
Transport refrigeration of goods	x		x		x
Refrigerants in ice rinks	x	x			
Ultra-low and low temperature freezers		x	x		x

Our search showed that non-fluorinated refrigerants are widely available for the mentioned refrigeration systems even if there is no single refrigerant that would be the perfect solution for all systems.

3.3) Air conditioning

Currently only a few percent of European residential and commercial buildings have air conditioning systems installed. The annual sales in 2016 were around 12 million units and thus still represent an important market. Air conditioning systems are subdivided here into stationary air conditioning systems that are moveable in the room, single and multiple split stationary air conditioning systems, mobile air conditioning in vehicles on the ground, and mobile air conditioning in aircrafts.

Table 2 gives an overview of which non-fluorinated refrigerants are already used in commercially available air-conditioning systems. The list is not exhaustive. Specific brand names and links to references are provided in the attached publication.

Table 2: Overview of which non-fluorinated refrigerants are already used in commercially available air conditioning systems. The list is not exhaustive. An 'x' indicates the availability of the non-fluorinated refrigerant in a commercially available air conditioning system.

	CO_2	Ammonia	Propane	Isobutane	Others
Stationary air conditioning – moveable in the room			x		
Stationary air conditioning – single and multiple split	x		x		
Mobile air conditioning in vehicles on the ground	x		x		x
Mobile air conditioning in aircraft	x				x

For the systems listed in Table 2, non-fluorinated refrigerants are available for almost every capacity ranges. An exception are single-split stationary air conditioning systems that are not available with non-fluorinated refrigerants above 5 kW. However, it might be possible to use water circuits loops (chillers) for these applications. Large propane chillers for industrial applications are readily available in Europe although their energy demand is somewhat higher than the demand of air conditioning systems (BAuA et al. 2023).

3.4) Chillers

A chiller, typically a vapor compression or absorption unit, removes heat from a secondary fluid circulated towards the equipment to provide any kind of cooling. The heat from the chiller is either transferred towards a secondary loop, which enables heat recovery, or directly transferred to the ambient air via a heat exchanger. In addition to maintaining the temperature of various industrial devices and laboratory instruments, equipment and apparatuses at a constant level, chillers are also used for air conditioning in buildings and factories.

Chillers are offered nowadays in various specifications. The non-fluorinated refrigerants used include CO_2, ammonia, isobutane, propene, propylene and water. Specific brand names and links to references are provided in the attached publication.

3.5) Heat pumps

Heat pumps provide heating or cooling to a building or process by transferring thermal energy between the outside and the inside of the building, using a refrigeration cycle. Heat pumps can use various media as heat source. According to the media, we divided them into air-source heat pumps, water-source heat pumps, geothermal heat pumps, (industrial) heat pumps with waste heat as energy source, and district heating network. Table 3 gives an overview of which non-fluorinated refrigerants are already used in commercially available heat pumps. The list is not exhaustive. Specific brand names and links to references are provided in the attached publication.

Table 3: Overview of which non-fluorinated refrigerants are already used in commercially available heat pump. The list is not exhaustive. An 'x' indicates the availability of the non-fluorinated refrigerant in a commercially available heat pump.

	CO_2	**Ammonia**	**Propane**	**Isobutane**	**Others**
Air-source heat pumps	x		x		
Water-source heat pumps		x	x		
Geothermal heat pumps	x	x	x		x
(Industrial) heat pumps with waste heat as energy source	x	x			
District heating network	x	x			

It has often been stated that split-heat pumps are not available with non-fluorinated refrigerants. However, this is not true. Split heat pumps have been developed with CO_2 and are widely in use in industrial applications as well as residential water heaters in countries such as Japan and Australia (SmallPlanetSupply 2023). CO_2 has even become the new norm for domestic hot-water heat pumps in Japan, where more than 5 million heat pumps with CO_2 are already installed to produce hot water for apartments (Hafner and Ciconkov 2021).

Additionally, the first manufacturers are planning to launch residential split heat pumps applying propane in Europe already this year (Hydrocarbons21 2022). Considering the current standards (EN378) and the relatively low cost of these systems, a fast market introduction can be expected. The Fraunhofer Institute for Solar Energy Systems in Germany is also working on developing low-charge propane heat pumps for multi-family housing (Hydrocarbons21 2023).

3.6) Devices for dehumidification and drying

Devices for dehumidification and drying include heat pump tumble dryers, refrigerant air dryers and mobile dehumidifiers. For all three types of devices, there are solutions on the market that use propane as refrigerant. Specific brand names and links to references are provided in the attached publication.

3.7) Conclusions for the PFAS restriction proposal

A transition to non-fluorinated refrigerants is possible and has happened in many sectors already. Technically, there are no barriers to replacing the fluorinated refrigerants in almost all newly developed systems by applying the existing standards. In some cases, trade-offs must be accepted in terms of costs, operating pressure, and/or capacity; however, compared to the improved environmental compatibility of the non-fluorinated alternatives, these trade-offs are worth making.

The transition to non-fluorinated working fluids is also a great opportunity for European companies to be frontrunners and it will create increasing demand within the heating and cooling sector. Many jobs within the chemical companies will also stay, as they will continue to serve and manage the aftermarket for the next decades.

We think therefore that the derogations in the PFAS restriction for F-gases as refrigerants should be as few as possible and not more than currently proposed. Otherwise, PFASs emissions related to the heating and cooling sector will continue to grow with negative impacts for human health and the environment.

4) Specific comments on Annex A of the restriction proposal

4.1) Volumes and identities of fluorinated gases

i) It needs to better be defined which substances are fluorinated gases and which ones are not. Or at least, for the substances registered under REACH, it should be clearly stated which substances that are liquid under normal conditions are gases in the atmosphere. There are seven substances that are given in REACH as liquids or solids but that are included in Table A.70 (Annex A) as gases. These are CAS Registry Number (RN) 80793-17-5, 355-04-4, 756-13-8, 335-27-3, 1800-91-5, 3709-71-5 and 15290-77-4.

ii) In Table A.70 of Annex A, there are a few substances that are not counted as fluorinated gases although there are gaseous. These include CAS RN 42532-60-5, 1187-93-5, 428-59-1.

iii) It is also important to note that substance CAS No 29118-24-9 is registered under REACH under the (incorrect CAS No) 1645-83-6.

iv) There are additional gaseous substances that are registered under REACH with a full registration but that are no included in Table A.70 (which forms the bases for the volumes). These include CAS No. 29118-25-0, 382-10-5, 66711-86-2, 111512-60-8, 431-89-0, 76-16-4, and 10493-43-3. The tonnages, names and other information can be found in the Supporting Material 2 of Rudin et al. (2023).

v) For a few midpoints in Table A.70, 450 (instead of 550) tonnes/year or 4500 (instead of 5500) tonnes/year were used. It is unclear why and should be corrected in our point of view.

vi) Given these points, we would like to ask the dossier submitter to go again over Table A.70, correct the entries and update the total volume of the fluorinated gases.

4.2) Volumes of PFAA and PFAA precursors

i) In Annex A, the last paragraph of Section A.2.1.3 states that Table A.71 and A.72 provide the REACH registered substances for PFAAs and PFAA precursors. However, Table A.71 contains non-polymers with unsaturated bonds and Table A.72 perfluoroether non-polymers with saturated bonds. Even if the perfluoroether would degrade to PFAAs (which might not be the case for all of them), there are more PFAAs and PFAA precursor in the ECHA database. These include (PACF-based) CAS No. 756-12-7, 756-13-8, (PASF-based) CAS No. 67584-59-2, 34455-00-0, 34454-97-2, 67584-55-8, PFAAs (375-73-5, 21615-47-4, 29420-49-3, 220689-12-3, 25628-08-4). For more information see the Supporting Material 2 of Rudin et al. (2023).

ii) In Table A.71, CAS RN 1187-93-5 is a gaseous substance

iii) In Table A.71, for 1,1,2,2,3,3-hexafluoro-1-trifluoromethoxy-3-trifluorovinyloxypropane, the EC number is given under CAS No. The correct CAS RN would be 40573-09-9.

iv) So similar to the fluorinated gases, we would like to ask the dossier submitters to also go again over the PFAAs and PFAA precursor and update the total volume of for the PFAAs and PFAA precursor.

5) References

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