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STUDY OF THE METHODOLOGY FOR ESTIMATING FUGITIVE EMISSIONS IN HYDROFLUOROCARBON REFRIGERANTS (HFC's)

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Abstract. Refrigerants are substances used in the refrigeration of air conditioners of various equipment today. With each passing moment, new products are created with less ozone depletion potential and less global warming potential to reduce the effects on climate change. In this way, companies need to evaluate their equipment and carry out an inventory of greenhouse gas emissions to quantify the impact they cause and how to minimize them. In this work we used the estimation method of fugitive emissions developed by the "Australian Government Department of Environment and Heritage" in a simulated process line of 57 kg cylinders of R-134a. With the quantified data, it was observed that all equipment, no matter how small its emission rate is, end up emitting a percentage to the atmosphere, but compressors and pressure relief valves are the largest emitters. In the emission ratio of this equipment, it was possible to quantify the volume lost in operating cost also the impact caused in the atmosphere converted into equivalent Carbon Dioxide. Thus, inventories are extremely important to manage industrial processes of gaseous fluids and their fugitive process losses, in addition to considering other emissions generated as Scope I and II to improve industrial processes.

Keywords. *Emissions, Fugitive, Emission Factor, GHG; Estimate; Inventory; Refrigerant Fluid; HFCs*

Introduction. Refrigerants are chemical substances responsible for transporting energy in the refrigeration cycle, where heat from one place is absorbed by the refrigerant and rejected in another environment (RAMIREZ, J. A. S, 2021)

When the first refrigeration system was patented in 1834, the refrigerants used were those of natural origin. That is, they could be found in nature, such as ammonia, CO2 and hydrocarbons (propane and isobutane, for example). They were the only fluids used for nearly a century. The expense of collecting, transporting and maintaining these gases no longer paid off, which is why in the early 20th century research began to develop synthetic gases. (ARCONDITIONED WEB, 2019)

Natural fluids were the only option available until 1929, when the first synthetic fluid was developed, R-12, which was a CFC (chlorofluorocarbon) patented exclusively for use in air

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conditioning. In this way, the use of synthetic fluids grew more and more in the industry, until it completely replaced natural fluids there in the 1970s. (WEB ARCONDICIONADO, 2019)

Around the 1980s, it was found that CFCs caused damage to the ozone layer of the Earth's atmosphere, which is the protection we have against the ultraviolet (UV) rays emitted by the sun. (ARCONDITIONED WEB, 2019)

For a while, the industry has been trying to bet on HCFCs (hydrochlorofluorocarbons), such as R-22, as they cause only 5% of the environmental damage that CFCs do. (ARCONDITIONED WEB, 2019)

With the worldwide concern about Global Warming, a new treaty, the Kyoto Treaty, is signed in 1997 and in 2019 the Kigali Amendment that defines a schedule for reducing the production and consumption of hydrofluorocarbons (HFCs) used in refrigeration equipment and air conditioning, which will be replaced by HFOs (hydrofluorolefin). (ARCONDITIONED WEB, 2019)

Briefly, the most common refrigerants in mechanical vapor compression refrigeration cycles are "hydro-chloro-fluoro-carbon - HCFC" (eg, R-22, R-123), "hydrofluor-carbon - HFC" type. (e.g., R-134a, R-152a, R-23) and ammonia (R-717), since the older ones like "chloro-fluorocarbon - CFC" (e.g., R11, R-12 and R- 13) and ammonium became extinct. (RAMIREZ, J. A. S, 2021)

According to Ramirez (2021) the authors claim that refrigeration has been making life easier for human beings for a long time. The process of taking heat from a body and pouring it into the medium or another with a higher temperature. Refrigeration performs the individual's thermal comfort, food storage, conservation of products of various types, whether residential or industrial, among other diverse attributes.

According to Ramirez (2021) the refrigeration system has two basic cycles: the vapor compression refrigeration cycle and the absorption refrigeration cycle. Vapor compression systems are the most used today, as they are composed of five basic components, which are: compressor, condenser, expansion valve, evaporator and refrigerant.

When understanding steam, the refrigerant fluid is an important participant in the performance of the refrigerant cycle, hence the importance of performing a performance analysis of the coefficient of performance of the cycle or coefficient of performance (COP), because according to Salvador (1999), this coefficient is a very important parameter in the analysis of refrigeration installations. The COP is a function of the refrigerant properties and, consequently, depends on the condensing and vaporizing temperatures. The coefficient is the ratio of the cooling effect to the compression work. In this way, it is possible to indicate which fluid is most comfortable to use. It is usually measured using the coefficient of performance (COP). Measurements are calculated by dividing the value of the heating capacity in kW to obtain the COP, by the nominal electrical consumption (kW) is given by the expression in Eq.1. (RAMIREZ, J. A. S, 2021)

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$$COP = \frac{Heat\ rejected\ by\ the\ cycle}{necessary\ work} = \frac{Q}{W} = \frac{h1-h4}{h1-h2} \quad \text{Eq 1}$$

Fluids must meet criteria for thermodynamic properties (entropy, enthalpy, temperature, pressure), flammability, toxicity, environmental impact, moderate price, and easy availability. One of the explorations and innovations proposed by the refrigeration industry is the use of fluids with less environmental impact. Manufacturers of air conditioners place equipment on the market with greater efficiency and performance in use and with less impact on environmental degradation. (RAMIREZ, J. A. S, 2021)

Among the fluids that are being used in the market are R-134a, used in automotive air conditioners.

The term "climate change" is defined as change that is directly or indirectly attributable to human activity that alters the composition of the global atmosphere, that is, observed over comparable periods of time, in addition to the natural variability of the climate (Framework Convention of the United Nations on Climate Change) – UNFCCC, 2014). The Intergovernmental Panel on Climate Change (IPCC) defines climate change as a statistically significant change and its variability over a long period of time (decades or more). In this definition, the cause can come from internal natural processes or external forces or human actions. The main human activities that affect climate change include the burning of fossil fuels, industrial and transport activities, changes in land use and deforestation. The realization of the Greenhouse Gases (GHG) inventory is an initial action by entrepreneurs who want to contribute to the issue of climate change in order to minimize its negative impacts. With the information generated by the inventory, entrepreneurs will be able to obtain relevant data on the management of greenhouse gas emissions. Through this action, organizations can attract new investments, obtain business opportunities with large clients that work in the area and also plan their processes to ensure greater economic, energy and/or operational efficiency. Many companies are required by legislation to carry out an annual GHG inventory, but they do not have the technologies and equipment to do so. (ABNT, 2016)

The objective of this work is to show the importance of carrying out an inventory of greenhouse gas emissions, as well as presenting the method of estimating fugitive emission according to the Australian Government Department of Environment and Heritage together with the GWP (Warming Potential global) simulating a process line of 57Kgs cylinders of R-134a.

Theoretical Foundation. Refrigerant Fluid: Refrigerants are chemical substances responsible for transporting energy in a refrigeration cycle, where heat from one place is absorbed by the refrigerant and rejected in another environment. The most common refrigerants used in mechanical vapor compression refrigeration cycles are "chlorofluorocarbons - CFCs" (e.g. R-11, R-12 and R-13), "hydrochlorofluorocarbons - HCFCs" (e.g. R-22, R- 13) -123), "hydrofluorocarbon-HFC" (e.g. R-134a, R-152a, R-23) and ammonia (R-717), according to Stoeker (2002) and ASHRAE (1997)). "CFCs" help to deplete the ozone layer, and their supply has virtually disappeared from the market. These refrigerants have the highest levels of ozone depletion "ODP" (Ozone Depletion Potential) and also have the biggest cause of global warming "GWP" (Global Warming Potential). Refrigerants "HCFCs" (R-22) and "HFC" (R-134a) are chlorine free and this gives them low

"ODP" indices, however, these refrigerants have high "GWP" indices. Different refrigerant fluids that are being used today are refrigerant mixtures called "azeotropic mixtures" and "non-azeotropic mixtures". "Azeotropic mixtures" exhibit behavior similar to pure substances during phase change from liquid to vapor and vice versa. The "non-azeotropic" mixtures present an abnormal behavior, as their evaporation and condensation temperatures change during the phase change process, even though the pressures are kept constant. These mixtures most often have good thermodynamic characteristics and low "ODP" indices, although they have high "GWP" indices. (Santos, W.D.D, 2020)

Types of Refrigerant Fluids: If we consider the diversity of fluids in the current market, we would need to carry out research just to reference them through their molecule and application. Thus, we present only a few types of refrigerant fluids. (FERRAZ, GOMES 2008);

CFCs - They are molecules formed by the element's chlorine, fluorine and carbon. (Examples: R-11, R-12, R-502, etc.), this type of gas is used in automotive air conditioning, commercial, industrial and domestic refrigeration; CFCs contribute to the depletion of the ozone layer. The national chemical industries have ceased the production of CFCs and the importation of these virgin substances is controlled. To convert or replace equipment operated with CFCs, two types of alternative refrigerants were created: HCFC's and HFC's. (FERRAZ, GOMES 2008)

HCFC - Some chlorine atoms are replaced by hydrogen (Examples: R-22, R-141b, etc.). These are used in: window air conditioners, split, self, cold rooms, etc. (FERRAZ, GOMES 2008)

HFC - All chlorine atoms are replaced by hydrogen (Examples: R-134a, R-404A, R-407C, etc.). Being used: automotive air conditioning, commercial refrigeration, domestic refrigeration (refrigerators and freezers), etc. (FERRAZ, GOMES 2008)

Considering that we present some of the types of refrigerant fluids, we will direct our study to what really interests our research, which is to present results regarding the fluid R-134a.

R-134a (1,1,1,2-dichlorodifluormethane): R-134a is one of the main refrigerants to replace R-12 in many of your applications. R134a is non-flammable and non-explosive which has zero ODP and a low GWP, since in its molecule it is an HFC, having no chlorine (Cl) in its molecule, responsible for the degradation of the ozone layer and global warming. Regarding toxicity and environmental aspects, according to the Material Safety Data Sheet (FISPQ), there were no perceptible indications of teratogenicity, mutagenicity and carcinogenicity. Due to the absence of chlorine atoms in the R-134a molecule, this alternative offers excellent chemical and thermal stability, being even better than R-12. This has been proven in several tests. R-134a is compatible with all metals and alloys normally used previously with CFCs and the use of Zinc, Magnesium, Lead and Aluminum should be avoided, which was already valid for CFCs. R-134a has good compatibility with elastomers. However, fluorinated rubbers of the FKM or FPM (Viton) types are not recommended. Due to the existence of different types of rubber, it must be analyzed case by case. Also in this case, the compatibility of the lubricating oil with the materials used must be evaluated. R 134a and mineral oils or alkyl benzenes used with current R-12 and R-22 refrigerants are not miscible. The lubricants that showed better miscibility were synthetic compounds with higher polarity. Initially, Poly Alkylene Glycols (PAG's), with viscosities lower than 30 cSt at 40ºC, are almost completely miscible between -40 and 80ºC. However, its use ended up being limited due to being highly hygroscopic and presenting some compatibility problems, for example, with chlorine residues. They are not suitable for use in hermetic compressors. (PARANÁ-UFPR, 2010)

Greenhouse Gases – GHG: A gaseous component of the atmosphere, both natural and anthropogenic, that absorbs and emits radiation at specific wavelengths within the spectrum of infrared radiation emitted by the Earth's surface, atmosphere, and clouds. GHGs include carbon dioxide (CO2), methane (CH4), nitrous oxide (N2O), hydrofluorocarbons (HFC), perfluorocarbons (PFC) and sulfur hexafluoride (SF5). (CETESB 2021)

Australian Government Department of Environment and Heritage: The Australian Government's Department of Environment and Heritage is an Australian regulatory body whose mission is to improve Australia's agriculture, environment, heritage and water resources through regulation and partnership. Among its objectives are to provide advice to the Australian Government; deliver programs and administer legislation among various topics, in particular on the use of pollutants and hazardous substances, in which we can classify various topics as. (AWE, 2022)

Mass balance: A form of monitoring that consists of quantifying the input, output, accumulation, generation or destruction of the substance of interest, calculating, by difference, its emission to the environment, according to equation 2 below. (AUSTRALIA, 2004 Modified).

$$\sum \text{input} = \sum \text{output} \qquad \text{Eq 2}$$

$$\sum \text{output} + \sum \text{emission} = \sum \text{input}$$

Fugitive Emission: According to Australia (2004), fugitive emissions include the diffuse release of any form of solid, liquid or gaseous matter into the atmosphere from sources that do not have a means designed to direct or control their flow. Fugitive sources are those that produce emissions that do not enter directly into a chimney, exhaust pipe, vent or other equivalent device. According to Wallace (1979), fugitive emissions are characterized by the diffuse release of pollutants into the environment, so that they do not pass through equipment designed to direct their flow. Siqueira (2007) cites storage piles, cooling towers and components of industrial production lines as examples of sources of fugitive emissions. See examples in the 3 images below showing where fugitive emissions occur in flanges, valves, connections, line openings. The emissions shown in Figure 1 show the fugitive emissions that normally occur in equipment due to the internal pressure of the system, exactly where they have seals which cannot seal 100% due to pressure.

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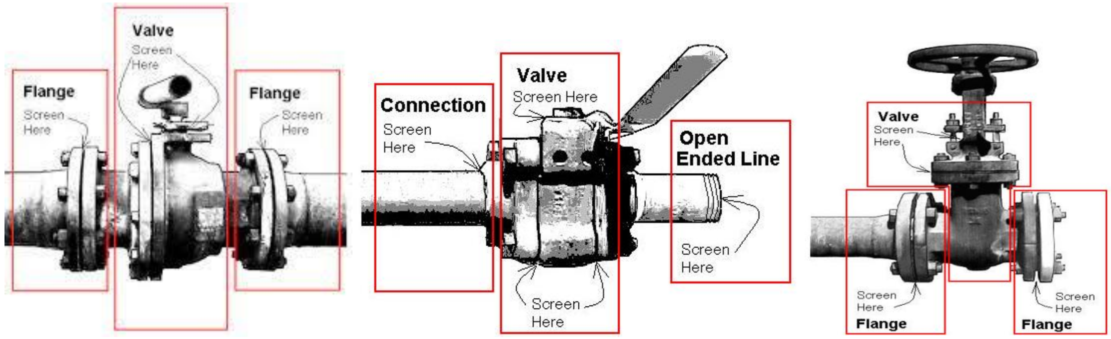

Figure 1 - U.S. EPA document EPA-453/R-95-017, issued November 1995

Issuance fee: The emission rate can be defined as the mass amount of pollutant emitted per unit of time, expressed in kilograms per hour (kg/h) or gram per second (g/s), for example (Emission rate of a block valve of gases is 0.00597 kg/hour/source) (AUSTRALIA, 2004 modified).

Estimated value: Result of an emission, using emission factor, calculations or indirect parameters. (AUSTRALIA, 2004)

Punctual issuance: Release into the atmosphere of any form of solid, liquid or gaseous matter, effected by a source provided with a device to direct or control its flow, such as ducts and chimneys. (AUSTRALIA, 2004)

Emission Factor: The representative value that relates the mass of a specific pollutant released into the atmosphere with a specific amount of material or energy processed, consumed or produced (mass/unit of production), in order to estimate the emission. (AUSTRALIA, 2004)

Calculation of Fugitive Emissions Estimate: The Australian Government Department of the Environment and Heritage has an estimation methodology for calculating fugitive emissions per Kilograms/Source/Year for total equipment types in chemical process lines. The method seeks to instruct corporate entities to help carry out the greenhouse gas inventory to comply with legislation.

Many of the engineering equations studied in the world can be applied to estimate the emissions of organic compounds, such as refrigerants. Aside from using emission factors or applying the mass balance technique to estimate emissions, little information is currently available to estimate fugitive emissions from inorganic compounds. However, in the manufacture of inorganic chemicals, it may be necessary to estimate emissions from inorganic compounds for inventory purposes.

Inorganic compound emission estimates can be obtained for inorganic chemical manufacturing processes by the following methods (in no specific order of preference):

• Develop specific correlations for specific chemical manufacturing processes;

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• Use a portable monitoring instrument to obtain actual concentrations of inorganic compounds and then apply the obtained screening values (see below) to the applicable correlation equation.

Emission Factor data is screened and collected using a portable monitoring instrument to sample air from potential leak interfaces on individual equipment. A screening value (SV) is a measure of the concentration of leaking compounds in ambient air that provides an indication of the leakage rate of a piece of equipment and is measured in units of parts per million by volume (ppmv).

In addition, surrogate measurements can be used to estimate emissions from inorganic compounds. For example, potassium iodide (KI), or a similar saline solution, is an indicator of leaks from acid process line equipment (hydrochloric acid [HCl], hydrofluoric acid [HF]) in chemical plants.

For the initial calculation, follow the definitions for support:

Ekpy,i = pollutant emission rate in a process stream determined by equipment type, kg / year / source

Emission Factor = applicable average emission factor for the type of equipment, kg / h / source

WFi = average weight fraction (Concentration % being 100% = 1 and 0.5 = 50%) of the pollutant in the flow equipment

Worked hours. = annual operating hours of equipment in the stream, hr / year

Equipment No. = number of pieces of the applicable equipment type in the process flow.

Thus, below we can define Equation 3 to estimate the fugitive emissions by equipment and later convert to CO2 equivalent.

Ekpy,i = EF x WFi x WH x Equip N (Fugitive Gas Emission/equipment) Eq.3

Ekpy,i x GWP = (Emissions in CO^2 equiv.) Eq 4

In the Table below we can see the values of fugitive emissions by equipment / GWP for some refrigerants that are estimated according to screening and data collection using a portable monitoring instrument, but the main focus in this work is R-134a.

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Table 1 - Fugitive Emission Factors by Equipment / GWP

Equipament Type	Service	Emission Factor (kg/hr/source)		Refrigerant Fluid	GWP (AR4)
Valves	Vapor	0,00597		R-22	1810
	Light Liquid	0,00403		R-32	675
	Heavy Liquid	0,00023		R-125	3500
Pump seals	Light Liquid	0,0199		R-410A	2088
	Heavy Liquid	0,00862		R-134a	1430
Compressor Seals	Vapor	0,228		HCFC-141b	725
Pressure Relief Valves	Vapor	0,104		R-404A	3922
Connectors and Flanges	All	0,00183		R-417A	2325
Open Lines	All	0,0017		R-407C	1774
Sampling Connections	All	0,015		R152a	124

Source: Eastern Research Group, 1996 e AR4 Climate Change 2007: Synthesis Report - IPCC 2077

Materials and Methods. The present work was developed by indirect deductive methodology from bibliographic and applied research. Initially, a broad sweep was made through Google Scholar in Brazilian and foreign articles, journals and scientific websites with the entry "fugitive emissions". Then, the search began in other scientific articles with the keywords: emission factor, Greenhouse Gases, Estimation, Calculation, and among other words.

The method found for calculating the estimate of fugitive emissions of Greenhouse Gases was from the Australian Government Department of the Environment and Heritage, which has an estimation methodology in Kilograms/Source/Year for the total types of equipment in process lines chemicals.

In table 2, we can see a simulated R-134a production line according to real process conditions and some equipment that is used, as well as its hours worked to quantify fugitive emissions according to the methodology of the Australian Government Department of Environment and Heritage. Data from Equation 3 and 4 will be used to simulate emissions.

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Table 2. 57 Kg Cylinder Production Line (Simulated)

Equipment	**Number of parts**	**Hours Worked/Year/Production**	**Model - Figure 3 - R-134a Returnable Cylinder 57 kg**
Pneumatic pumps	1	424	
Final Line - Open	1	424	
Blocking Valve	4	424	
Retention valve	2	424	
Compressor	1	424	
Pressure Relief Valve	1	424	
Flanges	6	424	
Safety valve	1	424	
Sample Connection	1	424	
Total	**18**	**3.816**	

Prepared by the Author, 2022

Results and Discussion. According to methodology and equation 3 and 4, fugitive emissions from the simulated process of 57kg R-134a Cylinders were calculated to quantify emissions for greenhouse gas inventory purposes. The equation is given as a basis for calculating the emission factors already determined by the methodology and the hours worked that must be filled in by the analyzer. The GWP data used were from the IPCC AR4 Report (2007). The GWP data estimate that for every 1Kg of R-134a Emission it is estimated that it is equivalent to 1430 Kgs of CO2 (Carbon Dioxide).

Table 3 presents the results of annual fugitive emissions by equipment in the simulated filling process of 57Kg R-134a Cylinders.

Table 3. Annual Fugitive Emissions Data / Calculation Representation

FREON 134A – Cylinder					Annual		
Equipment's	**Quantity of parts**	**% Worked hours**	**Emission Factor**	**WFI**	**Production/ Filling (Kg)**	**Worked hours**	**Fugitive Emission (Kg)**
Pneumatic pumps	1	100	0,0199	1	290.000,00	424	8,44
Final Line - Open	1	100	0,0017	1	290.000,00	424	0,72
Blocking Valve	4	100	0,00597	1	290.000,00	424	10,12
Retention valve	2	100	0,00597	1	290.000,00	424	5,06
Compressor	1	100	0,228	1	290.000,00	424	96,67
Pressure Relief Valve	1	100	0,104	1	290.000,00	424	44,09
Flanges	6	100	0,00183	1	290.000,00	424	4,66
Safety valve	1	100	0,00597	1	290.000,00	424	2,53
Sample Connection	1	50	0,015	1	290.000,00	424	3,18
Total	18	-	-	-	-	3.816	175,47

Prepared by the Author, 2022

In Table 4 shows the results of annual fugitive emissions of carbon dioxide (CO2) by equipment in the simulated filling process of 57Kg R-134a Cylinders. The GWP data estimates that for each 1Kg of R-134a Emission it is estimated that it is 1430 Kgs of CO2 (Carbon Dioxide) equivalent.

Table 04. Annual fugitive emissions data by GWP / Calculation Representation

Equipment's	**Fugitive Emission (Kg/Year)**	**GWP (AR4) R-134a (Kg/Year)**	**CO^2 Emission Equiv (Kg/Year) - GWP**
Pneumatic pumps	8,44	1.430,00	12.065,10
Final Line - Open	0,72	1.430,00	515,34
Blocking Valve	10,12	1.430,00	14.478,12
Retention valve	5,06	1.430,00	7.239,06
Compressor	96,67	1.430,00	138.233,33
Pressure Relief Valve	44,09	1.430,00	63.053,80
Flanges	4,66	1.430,00	6.657,03
Safety valve	2,53	1.430,00	3.619,53
Sample Connection	3,18	1.430,00	9.094,30
Total	175,47	-	250.923,81

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According to the data presented above, and through Figure 2, we can observe that the estimated fugitive emissions represent 6.05% of the total produced, considering only the emissions from the equipment mentioned in this process. From the mentioned equipment, we can see that the compressor and pressure relief valve are the biggest fugitive GHG emissions in this process.

Chart 1. - Estimated Fugitive Emission Data

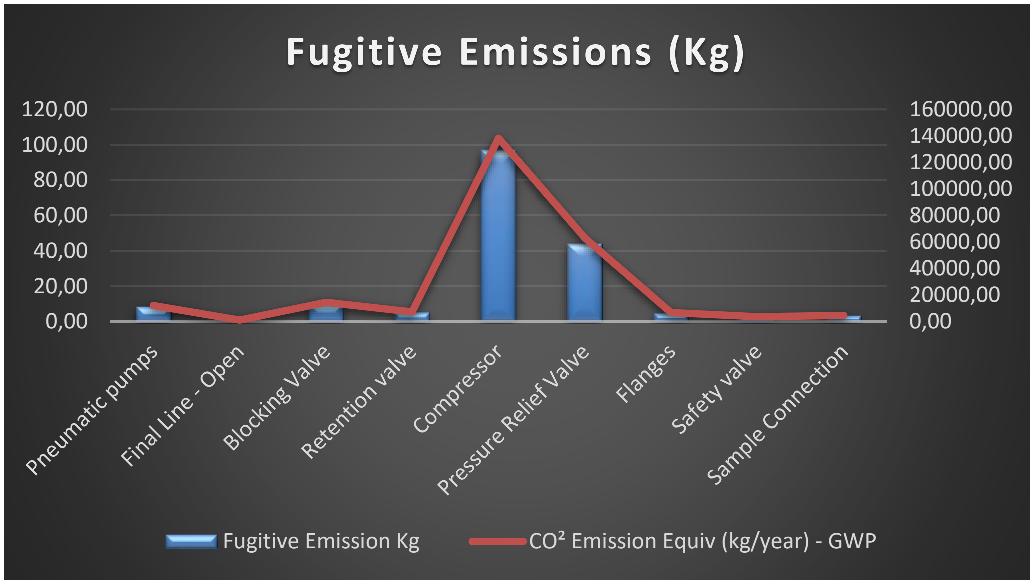

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Table 05. Data summarized in percentage of fugitive emissions

Production/ Filling (Kg/Year)	Total of Cylinder 57kgs – Filling Year	Fugitive Emission (Kg/Year)	CO^2 Emission Equiv (Kg/Year) - GWP	Total Emission %
290.000,00	5.088	175,47	250.923,81	6,05%

The atmospheric emissions of a company according to CETESB Board Decision 35/P/2021, can be represented by the quantification of Scope I Emissions (Direct GHG Emissions), Scope II (Indirect GHG Emissions) and Scope III (Indirect GHG Emissions) - the 3rd). In this work we are focusing only on fugitive GHG emissions, to quantify and estimate emissions x costs. Below we can see the description of the other issues.

<u>Scope 1 - Direct GHG emissions:</u>

• Burning of fuels to generate energy and steam;

• Other processes that emit GHGs;

• Transport of people, materials, products or waste in the enterprise's vehicles;

• Fugitive or evaporative emissions.

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Scope 2 – Indirect GHG emissions;

• Emissions from electricity purchased and consumed by the company.

Scope 3 - Indirect GHG emissions:

• Emissions from the transport of goods and services purchased or sold by an outsourced fleet equal to or greater than 300 vehicles.

In this way, by observing this calculation methodology, we can investigate and manage the emissions of a process and improve the equipment that emits the most fugitive gases, which can be quantified by specialized equipment, improving emission accuracy. This methodology only estimates the amount emitted based on an emission factor for each piece of equipment.

Another very important factor in managing emissions is when we talk about the cost lost in a fugitive process. Below we relate the average market cost per Kilogram of R-134a to the estimated issued rate.

Table 06. Annual Lost Cost by Fugitive Emissions

Average Price/kg R-134a (BRA)	Total Fugitive Emission (Kg/Year)	Annual Lost Cost
R$ 150,00	175,47	R$ 26.320,68

According to the data above, we can observe that for a simple process of filling R-134a cylinders, the lost cost of only fugitive gases in this process is R$ 26,320.68, which when added to other costs involved for other SCOPE I emissions, a chemical refrigerant filling process can become expensive if you don't have the investment in process improvements.

Conclusion. In this work, the importance of performing a calculation of estimation of fugitive emissions in a simulated filling process of 57Kgs of R-134a Cylinders was presented, as well as presenting the method to estimate these emissions and quantify the emission factor of each equipment for management and control, mainly involving operational costs and GWP when we are thinking about quantified Emissions for CO2 equivalent. Therefore, it is always important to quantify emissions to reduce the company's environmental impact and operating costs.

Disclosure. The authors report no conflicts of interest in this work.

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