

## Article

# Estimation, and Framework Proposal of Greenhouse Gas Emissions of Fluorinated Substitutes for Ozone-Depleting Substances by Application Area in the Republic of Korea

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**Abstract:** Since fluorine compounds have both high ozone depletion potential and high global warming potential, the study of hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) is crucial for climate change research. In this study, greenhouse gas (GHG) emissions from ozone-depleting industries in the Republic of Korea were estimated based on survey data on the use of fluorine compounds. This study is a response to the growing global attention to halocarbons that arose from the Kigali Amendment to the Montreal Protocol. Survey data on the consumption of fluorine compounds by application area were used to estimate emissions by applying the 2019 refinement IPCC Guidelines Tier 1a method. In addition, both the consumption ratio of fluorine compounds in the refrigeration and air conditioning application area and total fluorine compounds consumption by application area were compared with the values suggested by the UN Environment Programme to compare the current status with that in developing and developed countries. By comparing the derived GHG emissions with current emissions in the National Inventory Report, it was confirmed that 14,565 GgCO<sub>2</sub>eq of GHG emissions differed. In addition, through the replacement of fluorine compounds used as refrigerants, 14,422 GgCO<sub>2</sub>eq of the GHG emissions can be reduced.

**Keywords:** fluorine compounds; greenhouse gas emission; national inventory report; 2019 refinement IPCC Guideline

## 1. Introduction

Chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs), which are known to damage the ozone layer in the stratosphere, were widely used as refrigerants, blowing agents, electronic product cleaners, aerosols, extinguishing agents, and spray products before the environmental hazards were identified [1–4]. After the UN Environment Programme (UNEP) adopted the Montreal Protocol on the production and consumption of ozone-depleting substances (ODSs) [5–11], CFCs production and consumption were gradually reduced; CFCs' use was banned outright in 2010.

After the parties of the Montreal Protocol, which include all countries in the United Nations, signed the Kigali Amendment in 2016, the Montreal Protocol was rapidly strengthened to control additional hydrofluorocarbons (HFCs) and greenhouse gas (GHG) substances [12]. As a result, 99% of controlled ODSs were phased out in 2017, and 85% of ODS uses had been replaced [13]. The ODSs in the atmosphere are now decreasing, and the concentration of chlorine and bromine will continue to decline and return to the level of 1980 by the 2050s [14].

In response to recent amendments, including the Montreal Protocol [15–18], developed countries began to produce hydrofluorocarbons (HFCs) and perfluorocarbons (PFCs) to replace CFCs and HCFCs [19,20]. However, HFCs and PFCs are also GHGs with high global warming potential (GWP) [21,22] and atmospheric lifetimes longer than six months [23,24]. Thus, HFCs and PFCs have been listed as controlled GHGs by the Kyoto Protocol since 1997 [25,26].

Since Velders et al. [19] predicted that global fluorine compounds consumption and subsequent GHG emissions will continue to gradually increase, it is essential to accurately identify the amount of HFCs and PFCs used by each application area, and the GHG emissions from the result. The application areas that use fluorine compounds include refrigeration and air conditioning, fire suppression and explosion protection, aerosols, solvent cleaning, foam blowing, and other applications, according to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (2006 IPCC Guidelines) and the 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories (2019 refinement IPCC guideline [27,28]. In order to estimate GHG emissions in accordance with the 2019 refinement IPCC guidelines, activity data according to application area should be examined.

Accordingly, in this study, a survey of fluorine compounds distributions by companies in the Republic of Korea was conducted for five application areas including ‘refrigeration and air conditioning,’ ‘fire suppression and explosion protection,’ ‘aerosols,’ ‘solvent cleaning,’ and ‘foam blowing’ to estimate GHG emissions in each application area. To verify the survey data, we compared the results with trade statistics data managed by the Korea Customs Service and the ratio of HFCs gas usage suggested in the 2019 refinement IPCC guidelines, and we compared the difference between GHG emissions reported in the current National Inventory Report (NIR) and the emissions estimated in this study.

Furthermore, the GHG emissions that can be reduced by the distribution amount of fluorine compounds used for each application area can be determined through the survey data. Accordingly, we also estimated the amount of GHG emissions that can be reduced by the conversion of fluorine compounds to low-GWP gases, or natural substitutes in the refrigeration and air conditioning application area.

## 2. Materials and Methods

Emissions of ozone-depleting substances in the NIR are estimated using the Tier 1a methodology of the 1996 IPCC guidelines, based on HFC-152a and HFC-134a data on import and export trade statistics [29,30]. In this study, GHG emissions based on the survey data, (interpreted using the tier 1a methodology of the 2019 refinement IPCC guidelines), and emissions from the 2019 NIR were compared to improve the completeness and accuracy of the emission data in the 2019 NIR. In addition, consumption of gaseous fluorine compounds, other than HFC-152a and HFC-134a, was determined to establish a basis for calculating the future applications of the 2019 refinement IPCC guidelines, by identifying net consumption by gases and application areas in the Republic of Korea.

### 2.1. Survey Target Selection Process

Fluorine compounds distributed in Korea Republic are managed by the ten-digit Harmonized System (HS) code, which is an international nomenclature for the classification of products solely on physical characteristics [31,32]. Currently, HFC-152a and HFC-134a gases are identified using HS codes, while other gaseous fluorine compounds are labeled as ‘other’ in official documentation. Accordingly, a company list was secured by examining Korea Customs Service data on imports and exports for the three HS codes. As a result, we identified 234 companies that import or export with those 3 HS codes. The Korea Customs Service data on imports and exports companies for fluorine compounds by HS Code are shown in Table 1.

**Table 1.** Korea Customs Service data on imports and exports companies for fluorine compounds by Harmonized System (HS) Code.

| HS CODE      | Gas                                  | No. of Import Company | No. of Export Company |
|--------------|--------------------------------------|-----------------------|-----------------------|
| 2903-39-5000 | 1,1-Difluoroethane (HFC-152a)        | 9                     | 0                     |
| 2903-39-6000 | 1,1,1,2-Tetrafluoroethane (HFC-134a) | 30                    | 11                    |
| 2903-39-9000 | Other HFC Gases                      | 153                   | 31                    |
|              | Total                                | 192                   | 42                    |

We sampled companies that accounted for 97.7% of the total distribution of fluorine compounds in Korea, among a total of 234 ODS substance companies, and identified a total of 36 companies. A survey was sent to these 36 companies regarding gas type and application area.

## 2.2. Survey Method and Contents

The survey was conducted by distributing a questionnaire. The questionnaire consisted of import, export, and mixed gas production, and it was classified by the type of gases imported by the company, year, and application area. Of the 36 companies identified, 22 companies responded to the survey (95% confidence interval sampling error  $\pm 13.1\%$ ); as such, the respondents account for 98% of the total fluorine compound distribution in Korea.

## 2.3. Estimation of Emissions Based on Survey Data

To estimate GHG emissions using the Tier 1a methodology of the 2019 refinement IPCC guideline, it was necessary to take into account emission timing and the significance of banked gas reserves, and to apply the composite emission factor for each application area [28]. In the 2019 refinement IPCC guidelines, composite emission factors were presented that include all sub-uses. In refrigeration and air conditioning applications, the discharge rate of banked refrigerant is 15% per year, and the average facility lifetime is 15 years. Resin foam blowing agents are assumed to be used up at a rate of 10% for the first year, and 4.5% per year for 20 years according to the closed cell method. For fire suppression and explosion protection applications, it is assumed that 2% of gas is released annually and that the gas is discharged over 20 years, and that the remaining gas is discharged from the entire amount of waste. Aerosol and solvent cleaning gases are discharged over 2 years [28].

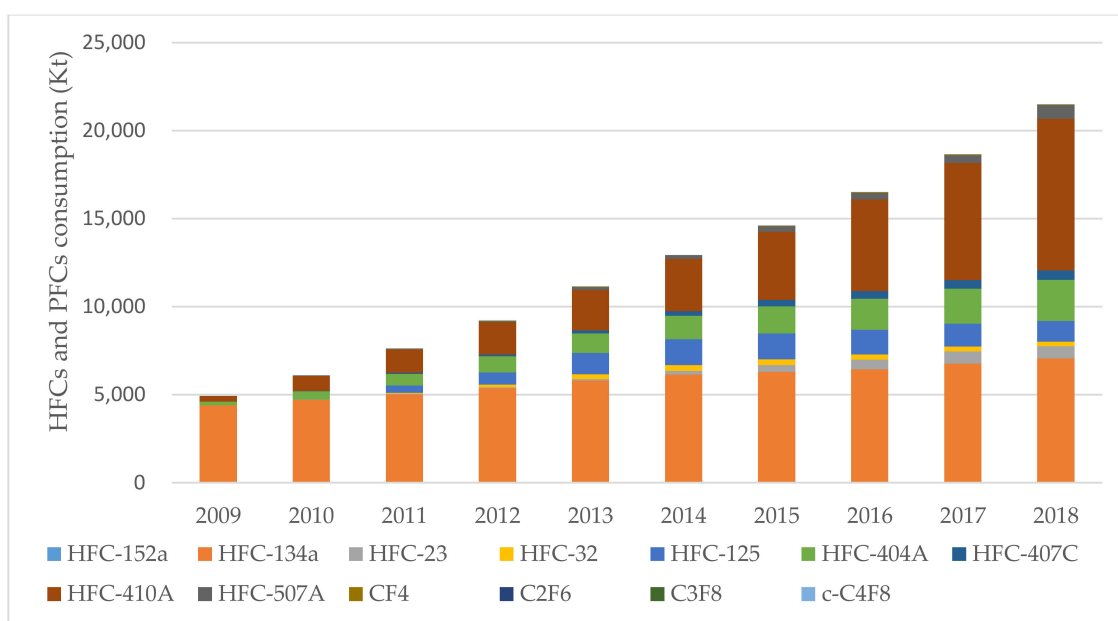
## 3. Results

### 3.1. Analysis of Consumption by Application Area

#### 3.1.1. Refrigeration and Air Conditioning Applications

The survey found that five types of HFCs, four types of mixed gas, and four types of PFCs were used for refrigeration and air conditioning applications. The total HFCs and PFCs consumption for refrigeration and air conditioning applications are shown in Figure 1. Gas analysis by year showed that use of HFC-125 and HFC-32 gases has been declining since 2013, whereas the use of the mixed gas designated HFC-410A increased at the greatest rate, with an average annual growth rate of 49.5%.

The total consumption of HFCs and mixed gas in 2018, including PFCs, was 21,491 Kt, a 15% increase from 2017 (20,233 Kt). In the Montreal Protocol, the UN Environment Programme (UNEP) put forth guidelines for fluorine compound consumption by the refrigeration and air conditioning application area [28]. Developing countries are classified as Article 5 and developed countries are non-Article 5; the Republic of Korea is currently applying the reduction programs for HCFCs and HFCs geared towards developing countries.



**Figure 1.** HFC and PFC consumption for refrigeration and air conditioning applications.

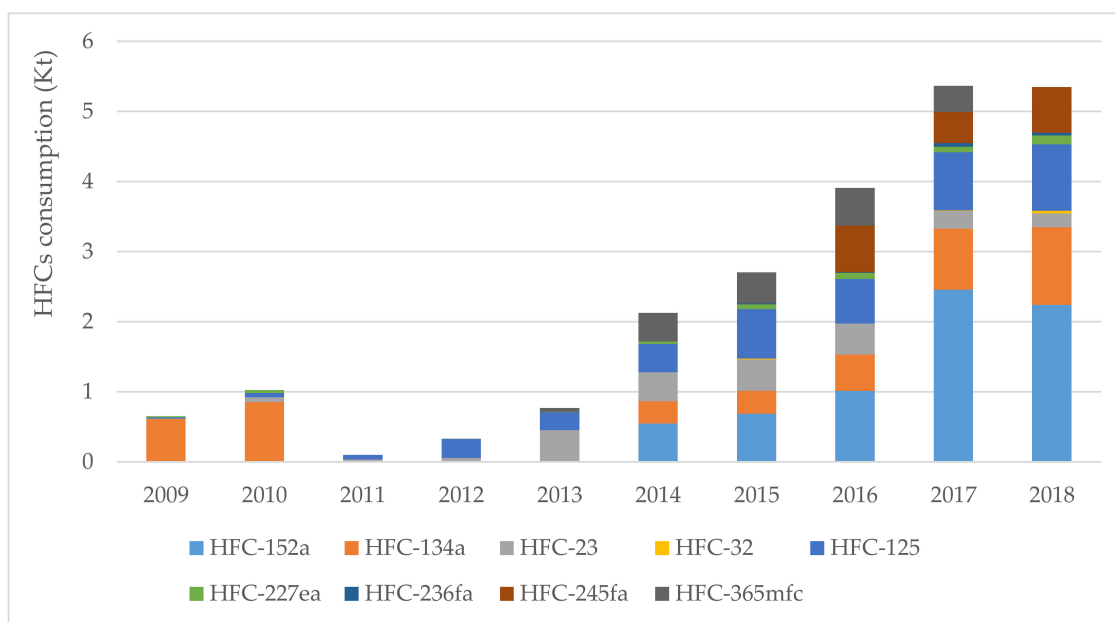
We compared the share of each HFC consumed by the refrigeration and air conditioning application area in the 2015 UNEP investigation results and in the results of this study. The comparison results of HFCs consumption in the refrigeration and air conditioning application area are shown in Table 2. The shares of HFC-134a and HFC-410A in this study are 42% and 38%, respectively, which are very similar to the shares of HFC-134a and HFC-410A in the data from non-Article 5 countries (39%). The share of HFC-407C in this study is 4%, which is significantly lower than the share in Article 5 countries (20%) and non-Article 5 countries (13%) in the UNEP investigation. This is because HFC-407C gas is mostly used in stationary air conditioners [33], but it is understood that HCFC-22 is still used in the Republic of Korea, and HFC-410A gas is mostly used in new stationary air conditioners [34–36]. HFC-404A had a 5% share in Korea, slightly lower than in Article 5 (7%) and non-Article 5 (9%) countries.

**Table 2.** HFC consumption (%) in the refrigeration and air conditioning application area.

| Kind of Data                |                              | HFC-134a | HFC-410A | HFC-407C | HFC-404A | HFC-507A |
|-----------------------------|------------------------------|----------|----------|----------|----------|----------|
| UNEP-TEAP                   | Article 5 Parties (2015)     | 27       | 39       | 20       | 7        | 7        |
|                             | Non-Article 5 Parties (2015) | 39       | 39       | 13       | 9        | -        |
| Result based on Survey Data | 2011                         | 55       | 23       | 2        | 5        | 0        |
|                             | 2012                         | 53       | 22       | 2        | 5        | 1        |
|                             | 2013                         | 48       | 20       | 3        | 5        | 2        |
|                             | 2014                         | 47       | 28       | 4        | 5        | 0        |
|                             | 2015                         | 42       | 38       | 4        | 5        | 2        |
|                             | 2016                         | 36       | 48       | 4        | 5        | 1        |
|                             | 2017                         | 40       | 46       | 3        | 5        | 1        |
|                             | 2018                         | 33       | 51       | 3        | 5        | 3        |

### 3.1.2. Foam Blowing and Fire Suppression and Explosion Application Areas

The fluorine compounds mainly used in the Republic of Korea for the foam blowing, fire suppression and explosion application areas are HFC-152a, HFC-134a, HFC-32, HFC-125, HFC-227ea, HFC-245fa, and HFC-365mfc. The total HFC consumptions for foam blowing, fire suppression and explosion application areas are shown in Figure 2.

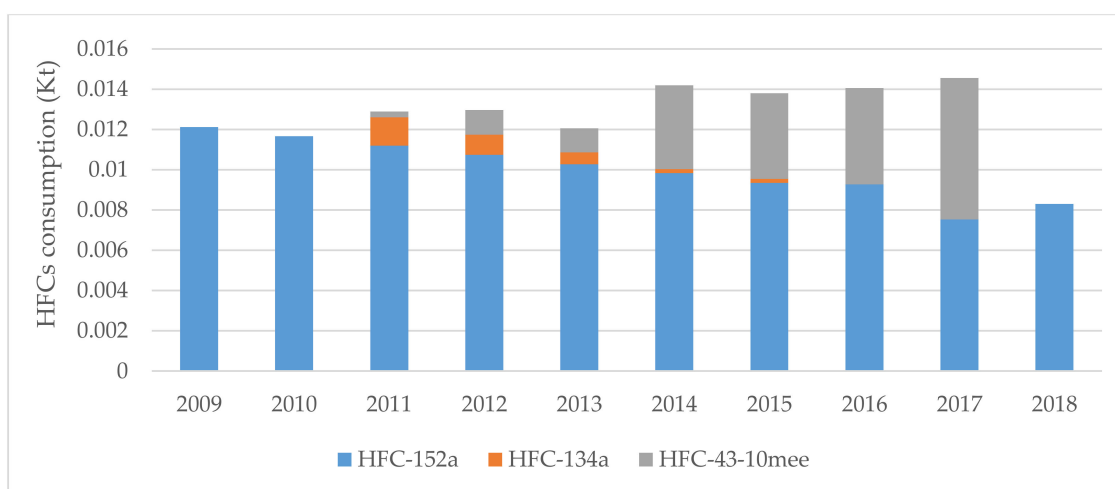


**Figure 2.** HFC consumptions for foam blowing and fire suppression and explosion application areas.

The amounts of HFC-152a and HFC-134a consumed are 2,240 tons and 1,108 tons, respectively, which together account for the largest portion of total gas consumption of 5,350 tons. This is because HFC-152a and HFC-134a are used to manufacture foam blowing, and fire suppression and explosion has increased due to the growth of the foam blowing, fire suppression and explosion industry [37–39].

### 3.1.3. Aerosol and Solvent Cleaning Application Areas

The total HFCs consumptions for aerosol and solvent cleaning application areas are shown in Figure 3. HFC-152a, HFC-134a, and HFC-43-1-mee gases have been used in aerosol and solvent cleaning applications in previous years, and only HFC-152a gas was used, at about 8.3 tons, in 2018. The reason for the relatively low usage is that low-GWP propellants (hydrocarbons, dimethylether, CO<sub>2</sub>, and nitrogen) are meant to be used in aerosol and solvent applications, according to the policy for developed countries presented in the report by the Intergovernmental Panel on Climate Change (IPCC) [40].



**Figure 3.** HFCs consumptions for aerosol and solvent cleaning application areas.

### 3.2. Estimation of HFCs Gas Consumption by Application Area

Calculation of GHG emissions using the 2019 refinement IPCC guideline Tier 1a method requires the use of composite emission factors, and information on market share by application area is required to apply the composite emission factors [28]. In this study, fluorine compound consumption by Article 5 and non-Article 5 countries in UNEP data was compared with fluorine compound consumption according to the survey, to confirm the validity of the survey data. The comparison data of fluorine compound consumption by application area are shown in Table 3.

**Table 3.** Fluorine compound consumption by application area.

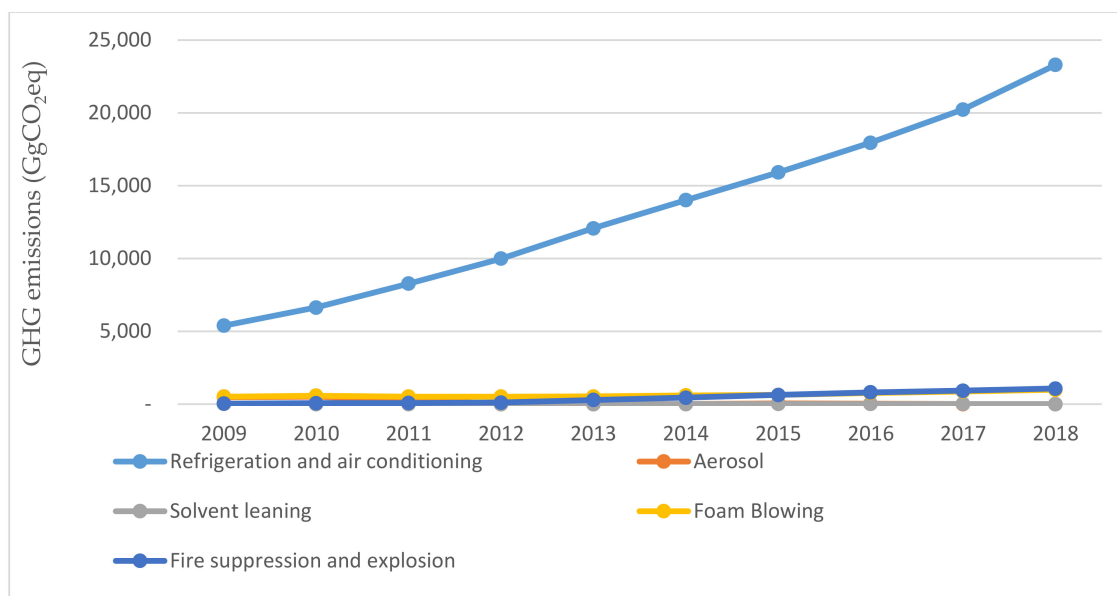
| Kind of Data                |                              | Refrigeration and Air Conditioning | Foam Blowing | Fire Suppression and Explosion | Aerosol and Solvent Cleaning |
|-----------------------------|------------------------------|------------------------------------|--------------|--------------------------------|------------------------------|
| UNEP-TEAP                   | Article 5 Parties (2015)     | 88                                 | 6            | 3                              | 3                            |
|                             | Non-Article 5 Parties (2015) | 57                                 | 22           | 19                             | 2                            |
| Result based on Survey Data | 2011                         | 98.9                               | 0.0          | 1.1                            | 0.0                          |
|                             | 2012                         | 97.1                               | 0.0          | 2.8                            | 0.0                          |
|                             | 2013                         | 94.0                               | 0.6          | 5.3                            | 0.1                          |
|                             | 2014                         | 85.0                               | 8.8          | 5.6                            | 0.6                          |
|                             | 2015                         | 82.4                               | 9.5          | 7.6                            | 0.6                          |
|                             | 2016                         | 78.0                               | 15.5         | 6.4                            | 0.1                          |
|                             | 2017                         | 75.6                               | 19.0         | 5.4                            | 0.1                          |
|                             | 2018                         | 78.9                               | 16.0         | 5.1                            | 0.0                          |

Analysis of the data from 2015 shows that the Republic of Korea is replacing HCFCs with HFCs in the foam blowing, fire suppression and explosion application areas faster than any other developing countries. As of 2018, the share used in foam blowing is 16%, which represents conversion of HCFCs to HFCs at the same speed as developed countries. However, in the case of the fire suppression and explosion and aerosols (including solvents cleaning) application areas, the level of conversion was far less than that of developed countries (19%, 2%).

### 3.3. Estimation of GHG Emissions by Application Area

GHG emissions by application area from fluorinated substitutes for ozone-depleting substances, according to the 2019 refinement IPCC Guidelines Tier 1a method [28], are shown in Figure 4.

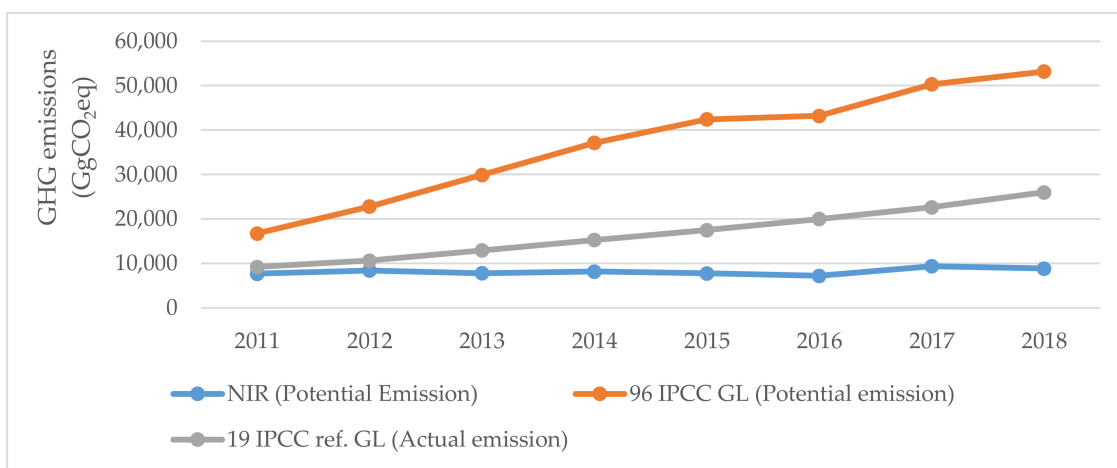
GHG emissions in 2018, calculated according to the 2019 refinement IPCC guidelines Tier 1a methodology IPCC/Fourth Assessment Report (AR4) GWPs, increased by 332% for ‘refrigeration and air conditioning,’ 112% for ‘foam blowing,’ and 3006% for ‘fire suppression and explosion’ compared to 2009. However, the GHG emissions attributable to ‘aerosols’ decreased from 473 to 1 Gg CO<sub>2</sub>eq, and the emissions from ‘solvents’ reduced from 11 to 6 Gg CO<sub>2</sub>eq.



**Figure 4.** Greenhouse gas (GHG) emissions of ozone-depleting substances by application area.

### 3.4. Comparison with Potential Emissions from the NIR and 1996 IPCC Guidelines

The GHG emissions comparison results of this study, and the potential emissions according to the NIR and 1996 IPCC guidelines, are shown in Figure 5. Potential emissions were estimated using the Tier 1a method of the 1996 IPCC Guidelines [41]. To estimate GHG emissions in the NIR (2019), data on imports and exports from the Korea Customs Service were used as the activity data. The target gases are eight types of HFCs, five types of PFCs, two types of mixed HFCs (R-406A, R-408A), and five types of mixed PFCs (R-404A, R-407C, R-410A, R-407F, R-507A).



**Figure 5.** GHG emissions compared to potential emissions according to the NIR and 1996 IPCC guidelines.

The actual emissions were estimated using the 2019 refinement IPCC Guidelines Tier 1a method by applying the composite emission factors discussed above. Emissions estimated by the NIR increased by 56%, from 5,636 GgCO<sub>2</sub>eq in 2009 to 8,822 GgCO<sub>2</sub>eq in 2018. Estimated GHG emissions according to the 2006 IPCC guidelines increased by 183%, from 9,188 GgCO<sub>2</sub>eq in 2009 to 25,989 GgCO<sub>2</sub>eq in 2018; the emissions increase was 217%, from 16,729 GgCO<sub>2</sub>eq in 2009 to 53,173 GgCO<sub>2</sub>eq in 2018, in the 1996 IPCC guidelines application.

In conclusion, we estimated actual emissions based on the 2019 refinement IPCC guidelines. Emissions of a total of eight HFCs, five PFCs, and five mixed gases were estimated. The results deviate by 2.6 times compared to the NIR result. Thus, we found that Korea's total GHG emissions by ozone-depleting substances currently reported through the NIR are lower by 14,565 GgCO<sub>2</sub>eq.

### 3.5. Evaluation of GHG Emissions Reduction through the Research Data

According to the results of this study, the amount of fluorine compounds used in refrigeration and air conditioning application areas in the Republic of Korea is overwhelming. Through these results, a reduction in GHG emissions was evaluated through the conversion of fluorine compounds used as refrigerants. R-134a and R-410a, which are the most frequently used compounds as refrigerants, are substances controlled by the Kigali Amendment to the Montreal Protocol because of their effect on climate (GWP of 1430 and 2088 respectively) [42]. After the amendment, research has been conducted in developed countries to reduce GHG emissions generated by replacing fluorine compounds used in refrigerants with low-GWP or natural refrigerants [43].

As a result of research into alternatives, R-134a can be replaced with R-1234yf gas and CO<sub>2</sub> (GWP:1), and R-32(GWP:675), DR-55(GWP:698), R-447A(GWP:583), ARM-71A(GWP:460), and HPR-2A(GWP:600) can be used as the alternatives for the R-410a [44–46]. We assumed that all of R-134a and R-410a used as refrigerants were converted into the alternatives at the same volume ratio, and the possible GHG emissions reduction was evaluated. As a result of the evaluation, a total of 14,422GgCO<sub>2</sub>eq of GHG emissions can be reduced as a replacement for refrigerants in 2018, which accounts for about 55% of total GHG emissions in 2018.

## 4. Discussion and Conclusions

Total GHG emissions of eight HFCs, five PFCs, and five mixed gases were estimated using the 2019 refinement IPCC guideline with composite emission factors, based on survey results for the verification of the impact of the 2019 refinement IPCC guideline application. The estimated potential NIR GHG emissions in 2018 were 8,822 GgCO<sub>2</sub>eq, which differs by about 14,565 GgCO<sub>2</sub>eq from the actual emissions estimated in this study. This amount of GHG emissions accounts for 2.2% of total GHG emissions in the Republic of Korea.

Provided that the GHG emissions currently proposed by the 2019 NIR and the national GHG emissions reduction goal of the Republic of Korea are underestimated, we recommend establishing an activity data tracking system under the supervision of the National Statistical Office. A statistical approach to estimating the missing data may be a good alternative.

For more efficient management of fluorine compounds, legislation should be drafted that regulates refrigerant use via the Korea Refrigeration and Air-Conditioning Industry Association, and roles and responsibilities for calculating the national inventory should be given to organizations other than the Korea Specialty Chemical Industry Association. Simultaneously, it is necessary to implement a quality assurance and quality control (QA/QC) system that can verify emissions data in the National Statistical Office, which will involve establishing a system for reporting recovery of gas wastes in the refrigerant information management system.

Reflecting all the recommendations above, to achieve the Intended National Determined Contribution (INDC) (37% reduction against BAU in 2030) in the Republic of Korea, reduction implement plans and measures under NIR emissions should be reconsidered.

The replacement of HFC-134a and HFC-410a, used in the refrigeration and air conditioning application area with low-GWP refrigerants, can reduce 14,422 GgCO<sub>2</sub>eq of GHG emissions. This amount of reduction accounts for 55% of the total emissions of fluorinated substitutes for ODS. In order to achieve INDC in the Republic of Korea, it is essential to apply the technologies that convert fluorine compounds into the low-GWP or natural substitutes.

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writing—original draft preparation, K.J. and D.R.; writing—review and editing, Y.-K.P. All authors have read and agreed to the published version of the manuscript.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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