

# 1 Estimating Perfluorocarbon Emission Factors for 2 Industrial Rare Earth Metal Electrolysis

## 4 Abstract:

5 Rare earth (RE) metals have been widely applied in new materials, leading to their drastic  
6 production increase in the last three decades. In the production process featured by the  
7 molten-fluoride electrolysis technology, perfluorocarbon (PFC) emissions are significant and  
8 therefore deserve full accounting in greenhouse gas (GHG) emission inventories. Yet, in the '2006  
9 IPCC Guidelines for National Greenhouse Gas Inventories', no method currently exists to account  
10 for PFC emissions from rare earth metal production. This research aims to determine emission  
11 factors for industrial rare earth metals production through on-site monitoring and lab analysis of  
12 PFC concentrations in the exhaust gases from rare earth metal electrolysis. Continuous FTIR  
13 measurements and time-integrated samples (analysed off-site by high-precision Medusa GC-MS)  
14 were conducted over 24-60 hour periods from three rare earth companies in China, covering  
15 production of multiple rare earth metals/alloys including Pr-Nd, La and Dy-Fe. The study  
16 confirmed that PFC emissions are generated during electrolysis, typically in the form of CF<sub>4</sub> (~90%  
17 wt of detected PFCs), C<sub>2</sub>F<sub>6</sub> (~10%) and C<sub>3</sub>F<sub>8</sub> (<1%); trace levels of *c*-C<sub>4</sub>F<sub>8</sub> and C<sub>4</sub>F<sub>10</sub> were also  
18 detected. In general, PFC emission factors vary with rare earth metal produced and from one  
19 facility to another, ranging from 26.66 to 109.43 g/t-RE for CF<sub>4</sub> emissions, 0.26 to 10.95 g/t-RE  
20 for C<sub>2</sub>F<sub>6</sub>, and 0.03 to 0.27 g/t-RE for C<sub>3</sub>F<sub>8</sub>. Converted to 211.60 to 847.41 kg CO<sub>2</sub>-e/t-RE for total  
21 PFCs, this emissions intensity for rare earths electrolysis is of lower (for most RE production) or  
22 similar (Dy-Fe production) level of magnitude to industrial aluminium electrolysis.

24 **Keywords:** PFC, greenhouse gas, emission factor, rare earth metal, electrolysis

## 26 1. Introduction

### 27 1.1. Rare Earth Metals Production & PFC Greenhouse Gas Emissions

28 'Rare earth metals' typically refer to a set of chemical elements in the periodic table, i.e. the  
29 fifteen lanthanides as well as scandium and yttrium. Rare earth (RE) metals have significant  
30 applications in new materials which are in wide demand in emerging and advanced industries such  
31 as permanent magnets and high-performance electronic devices. Therefore, in the last three  
32 decades, production of rare earth metals has soared dramatically. For instance, the global annual  
33 production of Nd<sub>2</sub>Fe<sub>14</sub>B permanent magnets have increased from around 1 tonne in the 1980s to  
34 more than 50,000 tonnes in around 2000 (Liu, 2008).

35 Since the 1990s, rare earth electrolysis using the molten *fluoride*-salt system has become  
36 the dominant production technology for rare earth metals. This replaced the molten *chloride*-salt  
37 system that was prevalent prior to the 1990s but suffered from several limitations, including: low

1 current efficiency, generation of chlorine gas as an environmental pollutant, poor metal quality /  
2 purity and other reasons.

3 The fluoride-based molten salts electrolysis process used today for rare earths production  
4 shares many similarities to the process used by the primary aluminium industry. As with  
5 aluminium electrolysis, the fluoride electrolysis route for rare earths production has the potential  
6 to form perfluorocarbon (PFC) gases, including tetrafluoromethane (CF<sub>4</sub>, PFC-14) and  
7 hexafluoroethane (C<sub>2</sub>F<sub>6</sub>, PFC-116), both of which are potent greenhouse gases; octafluoropropane  
8 (C<sub>3</sub>F<sub>8</sub>, PFC-218) is also occasionally reported in aluminium. According to the *Intergovernmental*  
9 *Panel for Climate Change (IPCC)*'s *Fifth Assessment Report* (2013), CF<sub>4</sub> has an extremely long  
10 atmospheric lifetime of 50,000 years and a global warming potential (GWP100) of 6,630  
11 compared to CO<sub>2</sub> over 100 years, C<sub>2</sub>F<sub>6</sub> has a lifetime of 10,000 years and a GWP100 of 11,100,  
12 and C<sub>3</sub>F<sub>8</sub> has a lifetime of 2,600 years and a GWP100 of 8,900.

13 While global production of rare earth metals by molten electrolysis technology is still very  
14 low compared to global aluminium production (roughly 0.1% of aluminium in 2013, based on  
15 global output of rare-earth oxides versus metallurgical-grade aluminium oxide (US Geological  
16 Survey 2014; IAI 2014)), it is possible that the resulting volume of greenhouse gas emissions can  
17 be comparatively large. Taking neodymium (Nd) metal production by Nd oxide electrolysis for  
18 example, it has been estimated by Vogel et al. (2017a) that in a *worst-case scenario*, the off-gases  
19 from the process could contain as much as 7% CF<sub>4</sub> and 0.7% C<sub>2</sub>F<sub>6</sub>. When considering the  
20 extremely large GWPs of these PFC gases and global production of roughly 30,000 t/year Nd  
21 metal, the rare earths industry could produce as much as 20 million t CO<sub>2</sub>-e/year (Vogel et al.,  
22 2017a).

23 If a significant volume of PFC generation from the rare earths industry was confirmed, this  
24 would go towards explaining the large discrepancy or 'gap' that has been found between (i) global  
25 atmospheric measurements of PFC emissions (a 'top-down' approach for accounting PFCs) and (ii)  
26 global 'bottom-up' accounting of PFC emissions from aluminium and semi-conductor industries.  
27 Both these industries are currently considered the only major anthropogenic sources of PFCs, with  
28 both employing methodologies from the IPCC to account for PFC emissions. Using atmospheric  
29 data, Kim *et al.* (2014) showed that as much as 50% of CF<sub>4</sub> and 48% of C<sub>2</sub>F<sub>6</sub> emissions over the  
30 2002–2010 period (5,200 t/year CF<sub>4</sub> and 300 t/year C<sub>2</sub>F<sub>6</sub>, equivalent to 42 million t CO<sub>2</sub>-e/year) is  
31 being under-estimated or unaccounted for from global industrial sectors.

32 The potential for large volumes of PFC gas emissions (combined with extremely high  
33 GWPs) from the rare earth metal industry implies that it should not be overlooked in terms of  
34 mitigating global warming. Therefore, evaluation and calculation of the global warming  
35 contribution from the rare earth metal industry is urgently needed. However, in the *2006 IPCC*  
36 *Guidelines for National Greenhouse Gas Inventories* (henceforth abbreviated to 2006 IPCC  
37 Guidelines), no guideline exists for the rare earth metal industry. One factor might have been the  
38 low proportion of Chinese contributors to the 2006 IPCC Guidelines (only 3.6% of authors and  
39 review editors for the entire 2006 IPCC Guidelines and only 1.1% for Volume 3 of the 2006 IPCC  
40 Guidelines: 'Industrial Processes & Product Use' (IPPU), where metal industry emissions are  
41 described) despite the fact that about 90% of rare earth metals globally are produced in China.  
42 Another more likely reason is that the global metal production of rare earths prior to 2006 was too  
43 small to consider as a significant contributor to GHG emissions, as already discussed above.

44 A further critical factor is the fact that to date there is a lack of quality academic research to

1 support a robust guideline for rare earth metal industry, if such a guideline were to be proposed.  
2 By contrast, there are numerous academic works and industrial studies on PFC emissions from  
3 primary aluminium production (Tabereaux, 1994; Rhoderick et al., 2001; Chase et al., 2005; Zhao  
4 et al., 2008; Wong et al., 2015). These have built up a robust foundation for the description of a  
5 detailed method for estimating PFC emissions from aluminium production in the 2006 IPCC  
6 Guidelines.

7 As a response to the above, recently a few studies on PFC emissions from the rare earth  
8 metals industry have been published. An example is the research conducted by Vogel et al. (2017a)  
9 who studied the electrochemistry of the neodymium oxide electrolytic system and the resulting  
10 anodic gas emissions. As the goal was to reduce PFC emissions, the paper focused on the  
11 interaction mechanism between CO/CO<sub>2</sub> and CF<sub>4</sub> emission concentrations and voltage across the  
12 electrochemical cell. With this groundwork, Vogel and Friedrich (2017b) continued the research  
13 and concluded that poor control of oxide concentrations of the electrolyte can cause higher PFC  
14 emissions. Therefore, a process control strategy similar to that in aluminium electrolysis was  
15 proposed to reduce PFCs, with continuous and precise oxide feeding being essential elements.

16 Vogel and his colleagues' research was conducted under laboratory conditions but  
17 mimicked industrial production. Given the variation of production engineering and gas scrubbing,  
18 this approach is effective in exploring the fundamental mechanisms of PFC emission but cannot  
19 be applied to estimating actual PFC emissions from the rare earth metal production industry.  
20 Therefore, Zhang et al. (2018) conducted research which measured continuous PFC emissions in  
21 an actual rare earth metal production facility. Zhang's work only focused on the PFC emission  
22 from production of Nd metal and Dy-Fe alloy at one rare earth production company. However,  
23 there are currently more than ten types of rare earth metals and alloys being produced by  
24 electrolysis every year. These include (in order of production output, from greatest to smallest):  
25 Pr-Nd, Nd, La, Dy-Fe, Ga-Fe, Ho-Fe, Pr, Ce, La-Ce and Y-Mg (ranking based on production data  
26 from major producers, covering 95% of the market). Furthermore, Zhang's data is limited in that it  
27 focused only on CF<sub>4</sub> emissions and did not measure other important PFC gases such as C<sub>2</sub>F<sub>6</sub> and  
28 C<sub>3</sub>F<sub>8</sub>. Uncertainty analyses were also not provided from the study. Since the time of Zhang's  
29 measurements in 2014, there have also been significant improvements made in rare earth  
30 electrolysis technologies and in the operations of the process, which are expected to help reduce  
31 PFC emissions. Recently, for cleaner production, gas-collection hoods for each electrolytic cell  
32 have been applied in some newly established production shops. There was no estimation of gas  
33 collection efficiencies of the hooding systems (i.e. emission factors did not take into account any  
34 fugitive emissions) in Zhang's (2018) study, which is a further significant uncertainty in this  
35 previous work.

## 36 **1.2. Aims of this Work**

37 In light of the limitations of prior studies and the new developments in rare earth metal  
38 production, this paper goes further to measure PFC emissions from the production of Pr-Nd alloy,  
39 Dy-Fe alloy and La metal from different electrolytic cell sizes and gas exhaust systems. It aims to  
40 provide greater coverage of PFC emissions in the rare earth metal industry so that PFC emission  
41 factors for different rare earth metals produced with different technologies can be proposed. The  
42 measurement of the PFC emissions was conducted in three typical rare earth companies in

1 Ganzhou, Jiangxi province. These three companies consist of diversified technologies including  
2 old and new production shop settings, small and large electrolytic cells, and low and high cell  
3 current technologies.

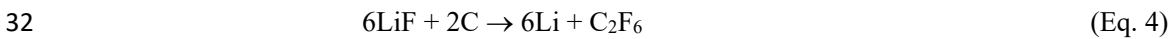
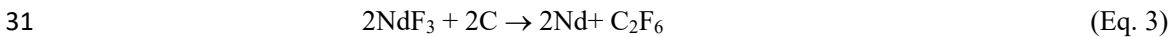
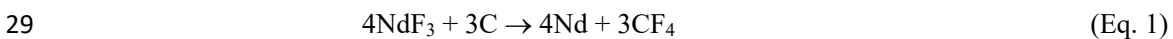
### 4 **1.3. The Fluoride Electrolysis Process for Rare Earth Metal Production**

5 The dominant technology worldwide for primary production of RE metals and alloys is using  
6 molten fluoride-salt electrolytic reduction, similar to primary aluminum's Hall-Héroult process.  
7 The raw materials for rare earths metal production are in the form of rare earth oxides (REO). In  
8 general, REOs are dissolved and electrolytically reduced in a molten salt of rare earth fluorides  
9 (REF<sub>s</sub>) and lithium fluoride (LiF), with graphite anodes and inert or consumable cathodes. The  
10 temperature in the reaction cell is high (~1000-1100 °C) and can vary depending on technology or  
11 metal produced, and be periodically interrupted or disturbed by anode replacements, cathode  
12 removal/replacement and removal of liquid RE metal/alloy.

13 Industrial fluoride-based rare earth smelters can be arbitrarily categorised into three classes  
14 by electrical current (and therefore the size of electrolysis cells), namely: low amperage (LA)  
15 technology, high amperage (HA) technology and high amperage with automatic control (HAA)  
16 technology. Low amperage technology (LA) are those with electrical current lower than 10 kA and  
17 typically employ small round-shaped cells, with only single (or several) vertical anodes and  
18 cathodes, and with very low levels of automation. For high amperage technology (HA and HAA),  
19 the current is higher than 10 kA. Typically, these use round or rectangular shaped cells, equipped  
20 with multiple vertical anodes and/or cathodes (Wen et al., 2012; Vogel & Friedrich, 2015). Some  
21 high amperage technologies may be equipped with 'Automatic Process Control' features (HAA),  
22 being able to automate rare earth oxide feeding so as to ensure continuous metal production and  
23 detection of anode effects that generate PFC.

24 PFC emissions are mainly in the form of CF<sub>4</sub> and C<sub>2</sub>F<sub>6</sub> and are often associated with 'anode  
25 effects' (AEs), resulting in passivation of graphite anodes (loss of electrical current-carrying  
26 capacity) and high cell voltage. These PFC emissions may be produced in the following manner  
27 (taking Nd metal electrolysis as an example) (equations 1-4):

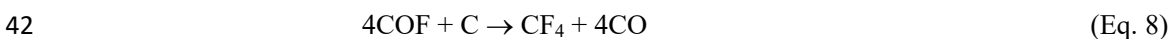
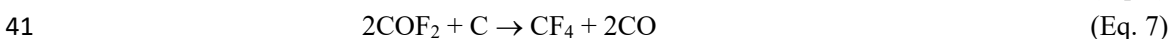
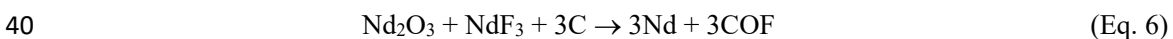
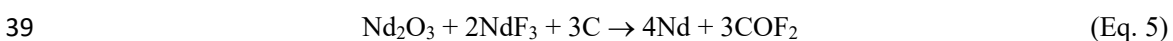
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34 In addition to being directly formed by electrolysis (equations 1-4), PFCs can also be  
35 generated by first forming COF<sub>2</sub> or COF electrochemically at lower electrode potentials  
36 (equations 5-6), which are unstable and will spontaneously react with C to form CF<sub>4</sub> and CO  
37 (equations 7-8) as follows (Vogel 2017a, Kjos 2018):

38



## 2. Methodology

Two simultaneous methods were used for sampling PFC emissions: (i) continuous onsite monitoring with an FTIR and (ii) time-integrated sampling (collection of emissions into gas canisters) with offsite lab analysis. In this study we focused on estimating emission factors for CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub>.

The most commonly applied equipment for monitoring PFC emissions is the *Fourier Transform Infrared* (FTIR) spectrometer. In theory, it is capable of capturing the emission data of all PFCs continuously in real-time. However, in RE metal production, in addition to emissions of major PFC gases (CF<sub>4</sub>), there are minor PFCs including C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> which can be more difficult to detect with the FTIR. It must be noted that when measuring these emissions with the FTIR, gases will only be detected when emission concentrations exceed certain detection limits, an issue related to the reference spectrum used in the FTIR database. Therefore during gas measurements, it is crucial to record information relating to the configuration of the production system and ensure that the on-site gas monitoring point has sufficiently high emission concentrations. In this study, CF<sub>4</sub> emissions were primarily estimated using FTIR measurements, whereas C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> emissions were estimated using the time-integrated sampling and lab analysis technique.

As emissions from a single cell can differ from other cells due to variations in structure, operating conditions and production stage, it is better to monitor emissions from a *group of cells*, e.g. 12 cells or more, even up to one ‘potroom’ or ‘potline’ of cells (a chain of cells connected in series to the same electrical circuit and same exhaust gas system) so as to reduce inherent measurement error. However, the number of cells covered must be balanced by maintaining sufficiently high concentrations of CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> that can still be accurately measured with the FTIR and time-integrated sampling. An optimum location can be in a common gas exhaust duct from a group of cells or from the gas stack before/after gas treatment for the entire potline.

Another issue that must be considered is the *gas collection efficiency*. During the monitoring process, the percentage of gases that is fully captured by the gas collection systems against all gases generated by the cells must be estimated. If gas collection efficiency is lower than 90% (i.e. more than 10% fugitive or uncontrolled emissions is not collected by the gas suction system), then there should be extra measurements of fugitive emissions at the exit point of the potroom/production shop where electrolysis cells are located, e.g. if there is a natural flow of air to the roof (Fraser et al., 2013). However, since the three rare earth companies monitored did not have any readily identifiable / accessible roof vents, gas collection efficiency was estimated at each company by releasing known quantities of SF<sub>6</sub> gas at the cell and using this as a tracer.

The amount of gas emissions in RE metal production can change over time. This may result from a variety of major process events on the cells, such as anode/cathode changes, metal removal, voltage/current change, RE oxide additions and the triggered ‘anode effect’ events. Therefore, to capture a representative period of operations/events, continuous measurements should ideally be conducted over a minimum of 24 hours (generally, the longer the time frame the better). During monitoring, all major process events were recorded so that any features in the gas concentrations can be correlated with these events – note that these features are not the focus of this paper and therefore are not elaborated further in this work. Furthermore, production data (metal and quantity produced) and technology factors (line current and cell size, which can fundamentally change the

1 emission outcomes) were also recorded.

2 For this research, three companies located in Ganzhou, Jiangxi province of China were  
3 selected as the representative cases: *Qiandong Rare Earths Group Co. Ltd* (Qiandong in  
4 abbreviation), *National Engineering Research Center for Ionic Rare Earth* (Ionic Rare Earth  
5 Center in abbreviation) and *Jiangxi South Rare Earth High Tech. Co. Ltd* (Jiangxi South in  
6 abbreviation). Qiandong specialises in three widely used RE metals including Pr-Nd alloy, La and  
7 Dy-Fe alloy. In contrast, Ionic Rare Earth Center attaches more importance on research, especially  
8 on new technologies for RE metals processing from ionic rare earths. Thus, measurement data  
9 from the Ionic Rare Earth Center can provide additional new information to the PFC emission  
10 spectrum. Jiangxi South was also included in the study as it employs some of the larger  
11 high-amperage (>10 kA) cell technologies.

12 In 2016, approximately half of China’s RE metals are produced by companies/institutions  
13 in Ganzhou, and Qiandong accounts for more than 1/3 of the total production in Ganzhou. In light  
14 of the above, the three companies participating in this study can be a valid representation of PFC  
15 emissions in the RE metal production industries in China, or even in the world.

16

## 17 2.1. Continuous On-Site FTIR Monitoring

18 For on-site monitoring of the CF<sub>4</sub> emission, an FTIR spectroscopy instrument (*MKS*  
19 *MultiGas™ 2030 or MG2030*, which is competent in capture of CF<sub>4</sub> emissions) was employed.  
20 Where possible, we measured emissions from a series/group of pots so as to ensure data was  
21 representative and to minimise the risk of major inaccuracies that are more likely to occur in  
22 measuring single pots (Table 1). Figure 1 shows the schematic diagram of the intended sampling  
23 set up and process.

24

25 **Table 1: Intended sampling arrangement for monitoring PFC emissions at the three RE**  
26 **companies**

Company	Qiandong			Ionic Rare Earth Center	Jiangxi South
Potline (metal produced)	<i>Pr-Nd</i>	<i>Dy-Fe</i>	<i>La</i>	<i>Pr-Nd</i>	<i>Pr-Nd</i>
No. of cells/pots	15	1	6	6	2
Line current (kA)	5	5	5	6	11
Time frame (hours)	60	24	36	36	24
Monitoring equipment	<i>MKS MG2030</i> FTIR for continuous onsite gas analysis, with gases simultaneously collected in sampling canisters for offsite lab analysis				

27

28 As explained previously, in addition to CF<sub>4</sub> there are minor PFC gas emissions including  
29 C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> which cannot be easily detected by *MKS MG2030*. However, it is still important to  
30 estimate emission factors for these two critical PFC gases. To circumvent this issue, we also  
31 collected ‘time-integrated’ gas emission samples (simultaneously with FTIR measurements over  
32 the same sampling durations) and stored them in canisters for subsequent offsite lab analysis  
33 (Figure 1); offline analyses provided considerably lower detection limits for other C-F gases.

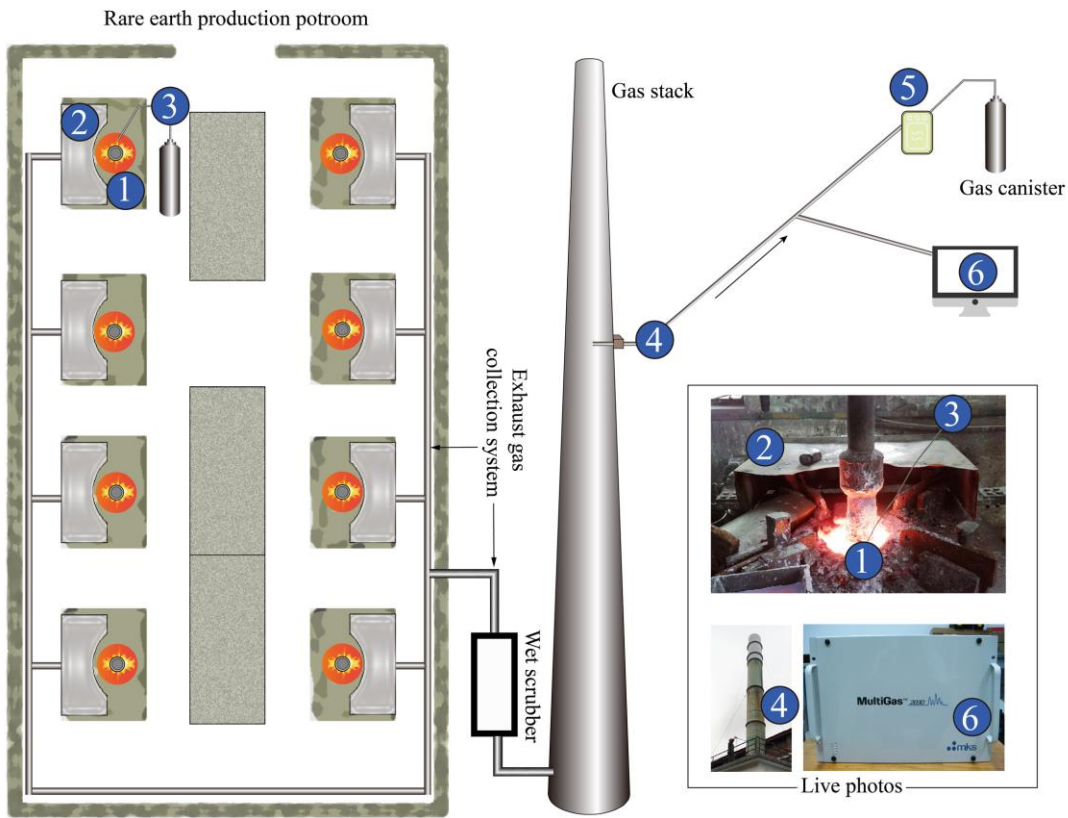
1 Through this approach, we estimate ratios of emission volumes for all other PFC components  
 2 compared to CF<sub>4</sub>, whereby emission factors for C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> can be calculated once CF<sub>4</sub>  
 3 emission factors are known/confirmed – details are explained in section 2.4.

4 In this research, the emission factor of CF<sub>4</sub> (EF<sub>CF4</sub>) is calculated directly by the equation  
 5 below:

$$6 \quad EF_{CF_4} = \frac{[CF_4] \times t \times F \times 0.088}{22.4 \times 10^6 \times Ecol \times Q} \quad (\text{Eq. 9})$$

7 Where [CF<sub>4</sub>] is volume concentration (ppmv), using data from the FTIR; *t* is total sampling  
 8 time (*h*); *F* is the average gas flow rate at the sampling point (Nm<sup>3</sup>/h); *Ecol* is gas collection  
 9 efficiency and *Q* is rare earth metal produced (tonnes) over the sampling time.

10 It was foreseeable during initial inspections that emissions from electrolysis cells could not  
 11 be completely gathered and sent by gas collection systems to the exhaust gas stack, where  
 12 sampling and measurements would be set up (Figure 1). Thus, a SF<sub>6</sub> tracer gas was employed to  
 13 estimate the gas collection efficiency at each company. Once this is known, the volumes of all gas  
 14 emissions can be calculated and accurate and reliable emission factors can then be estimated.  
 15



16 ① Rare earth electrolysis cell ② Gas collection hood ③ SF<sub>6</sub> trace gas ④ Sampling point ⑤ Time-Integrated sampler ⑥ MultiGas™ 2030

17 **Figure 1: The arrangement for on-site monitoring and sampling of PFCs from a potroom of**  
 18 **rare earth electrolysis cells.**

19 **2.2. Time-Integrated Sampling and Lab Analysis**

20 Exhaust air was drawn from the exhaust gas stack by pump (M121-FP-BA1-L) through a  
 21 sampling tube (1/4" OD Synflex tubing, Eaton, USA) at flow rate of 5 L/min to feed both the

1 in-situ FTIR system and time-integrated sampler. The sampler pressurized into pre-vacuumed 3-L  
2 stainless steel canisters (X23-2N, LabCommerce, Inc, USA) by means of a membrane pump  
3 (KNF-022, KNF Neuberger, Germany). The integrated flow was constrained by a mass flow  
4 controller (GFC17, AALBORG, USA) at a constant flow rate for each integrated sample. The  
5 flow rate ranged from 2.5 mL/min to 3.5 mL/min depending on the required integrated time of  
6 different samples.

7 Canister samples were transported to Beijing and analysed with a custom-built ‘*Medusa*’ gas  
8 chromatographic system with mass spectrometric detection (Agilent 6890/5975B, USA) for  
9 fluorinated greenhouse gases within one month. The system uses the technique developed by the  
10 *Advanced Global Atmospheric Gases Experiment* (AGAGE) and was operated in the lab of the  
11 *Meteorological Observation Centre* of the *China Meteorological Administration* (MOC/CMA).  
12 The measurements are linked to AGAGE standard scales (Prinn et al., 2018) and the precisions are  
13 <0.5% for CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>, C<sub>3</sub>F<sub>8</sub>, c-C<sub>4</sub>F<sub>8</sub> and C<sub>4</sub>F<sub>10</sub>. The detailed analysis method was described by  
14 Zhang et al. (2018). For the samples with pressure less than 10 psi, a dilution procedure was used  
15 to pressurize the sample by zero gas to ensure there is enough volume.

### 16 **2.3. Methodology for Estimating Gas Collection Efficiency using SF<sub>6</sub>**

#### 17 **Tracer**

18 As illustrated in Figure 1, high concentration SF<sub>6</sub> (0.498%, Beijing Beifenhaipu Gas  
19 Industrial, China) was released at a constant flow rate by a mass flow controller (20 to 1000  
20 mL/min depending on the releasing time period) at a chosen cell at each RE potroom. The end of  
21 the gas release tubing was placed as close to the anode as possible (generally less than 1 cm away  
22 from anodes). Actual gas release periods (i.e. monitoring period) and flow rates at each potroom  
23 are listed in Table 2.

24 SF<sub>6</sub> was diluted in the exhaust gas collection system and later collected into stainless steel  
25 canisters together with other greenhouse gases in the exhaust gas. The SF<sub>6</sub> concentration was  
26 analyzed by the Medusa-GC/MS in MOC/CMA lab (described in section 2.2) with an uncertainty  
27 of 0.5%. Thus, the gas collection efficiency was calculated as follows:

$$28 \quad E_{col} = \frac{C_{canister} \times F_{exhaust}}{C_{standard} \times F_{standard}} \quad (\text{Eq. 10});$$

29 Here  $E_{col}$  is gas exhaust collection efficiency,  $C_{canister}$  and  $C_{standard}$  are SF<sub>6</sub> concentrations of  
30 canister samples and of the SF<sub>6</sub> tracer standard, respectively, while  $F_{exhaust}$  and  $F_{standard}$  are the flow  
31 rates of the exhaust gas and the released flow rate of SF<sub>6</sub> tracer gas (high concentration standard),  
32 respectively. Here  $C_{standard}$  is 0.489%.

### 33 **2.4. Estimating Emission Factors of C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub>**

34 The *MKS MG2030* FTIR is capable of measuring the emission of CF<sub>4</sub> from production lines,  
35 yet it is unable to detect the gas emissions such as C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> directly, due to limitations of the  
36 reference spectrum used in the FTIR. To estimate emission factors for these two gases, we  
37 calculate their emission volumes by referring to the emission volume ratios (which is measured in



1 the lab from time-integrated samples) between them and CF<sub>4</sub>.

2 The emission factors (*EF*) of C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> can be calculated by the expressions as below:

3  
4 
$$EF_{C_2F_6} = EF_{CF_4} \times (138 / 88) \times (D_{C_2F_6} / D_{CF_4}) \quad (\text{Eq. 11})$$

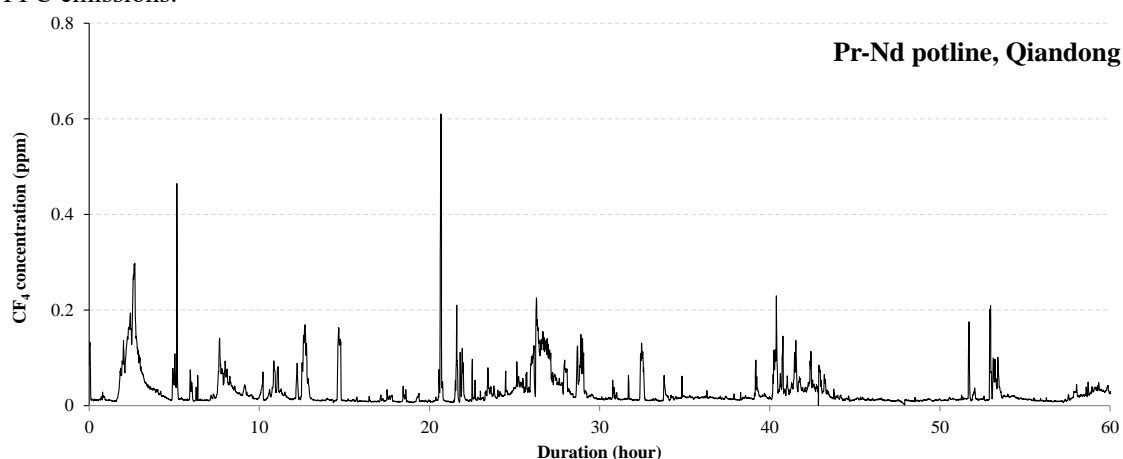
5 
$$EF_{C_3F_8} = EF_{CF_4} \times (188 / 88) \times (D_{C_3F_8} / D_{CF_4}) \quad (\text{Eq. 12})$$

6  
7 Where, *D*<sub>CF<sub>4</sub></sub>, *D*<sub>C<sub>2</sub>F<sub>6</sub></sub> and *D*<sub>C<sub>3</sub>F<sub>8</sub></sub> are the average concentration of CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub>  
8 measured in the lab from time-integrated sampling canisters are respectively, and *EF*<sub>CF<sub>4</sub></sub> is the  
9 estimated emission factor of CF<sub>4</sub> (measured simultaneously by FTIR on the same potline).  
10

### 11 3. Results and Discussion

#### 12 3.1. Estimated CF<sub>4</sub> Emission Factors from On-Site FTIR Monitoring

13 The first FTIR time series data of CF<sub>4</sub> emissions covered 15 pots from the Pr-Nd  
14 production potline in Qiandong, over 60 hours of monitoring. As shown in Figure 2, a number of  
15 anode effect events, represented by the sharp peaks in CF<sub>4</sub> emissions, were identified. The  
16 maximum observed CF<sub>4</sub> concentration was 0.610 ppm. It was estimated that 75% of CF<sub>4</sub>  
17 emissions were related to peak or anode effect (AE) emissions, with 25% attributed to ‘non-anode  
18 effect’ (NAE) emissions, as determined by integrating the emission time series for peak vs.  
19 non-peak emission periods. Based upon this data, a CF<sub>4</sub> emission factor of 26.66 g/t-RE was  
20 calculated, with a corresponding CO<sub>2</sub> equivalent emission factor of 176.76 kg CO<sub>2</sub>-e/t-RE. This is  
21 roughly 40% lower than the 307.65 kg CO<sub>2</sub>-e/t-RE emission factor measured in 2014 for Nd  
22 production (Zhang et al., 2018, with emission values converted assuming the same 57.97% gas  
23 collection efficiency). This reduction in PFC emissions coincides with the installation of an  
24 automated feeding system for the rare earth oxides into the production line over the past 3 years.  
25 Therefore, this indicates that improvements to the electrolytic process and operations can reduce  
26 PFC emissions.

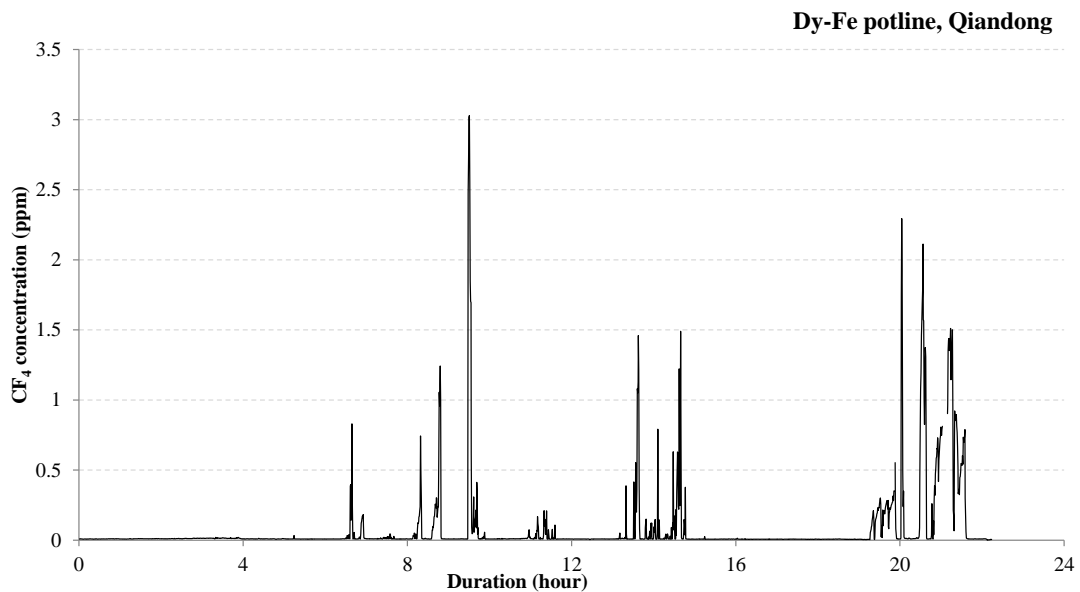


27  
28 **Figure 2: CF<sub>4</sub> concentration of Pr-Nd potline (15 cells) in Qiandong during regular**  
29 **production**

30  
31 There was only one single pot measured in the Dy-Fe potline in Qiandong. To estimate an

1 emission factor for Dy-Fe production, exhaust gases from the cell was monitored over 24 hours  
2 (Figure 3). A series of anode effects (emission peaks) were observed with 3.029 ppm as the  
3 maximum concentration of CF<sub>4</sub>. Many peak values relating to anode effects were greater than 0.5  
4 ppm; an estimated 91% of CF<sub>4</sub> emissions were related to these AEs, with 9% related to NAE  
5 emissions. The overall CF<sub>4</sub> emission factor for Dy-Fe was 109.43 g/t-RE, being equal to 725.52  
6 kg CO<sub>2</sub>-e/t-RE. In 2014, the CO<sub>2</sub>-equivalent emission factor in this same potline was 1,212.32 kg  
7 CO<sub>2</sub>-e/t-RE (Zhang et al. 2018, values converted with a 57.97% gas collection efficiency). We can  
8 see that the PFC emission had decreased significantly (~40%), which again resulted from  
9 improvements to the electrolytic process and material feeding system.

10 Compared with the other three potlines, the CF<sub>4</sub> emission factor for Dy-Fe production is the  
11 highest. One possible explanation is that the voltage for Dy-Fe electrolysis (at 12 V) is much  
12 higher than in other potlines (e.g. 4V higher than for the Pr-Nd potline). Operating at higher  
13 voltage makes it more likely to trigger anode effects which will produce more PFC gases. Another  
14 observation that supports higher PFC emissions is the fact that significantly more fluoride  
15 electrolyte is consumed in this Dy-Fe potline, with a two-fold greater fluoride consumption than  
16 Pr-Nd production. This is because more anode effects happened in this potline (Figure 3) which  
17 led to more PFC emissions. Finally, it should be stressed that there is a level of uncertainty in  
18 these results given that Dy-Fe sampling was conducted from only one cell, instead of multiple  
19 cells as discussed previously.



20

21 **Figure 3: CF<sub>4</sub> concentration of Dy-Fe potline (1 cell) in Qiandong during regular production**

22

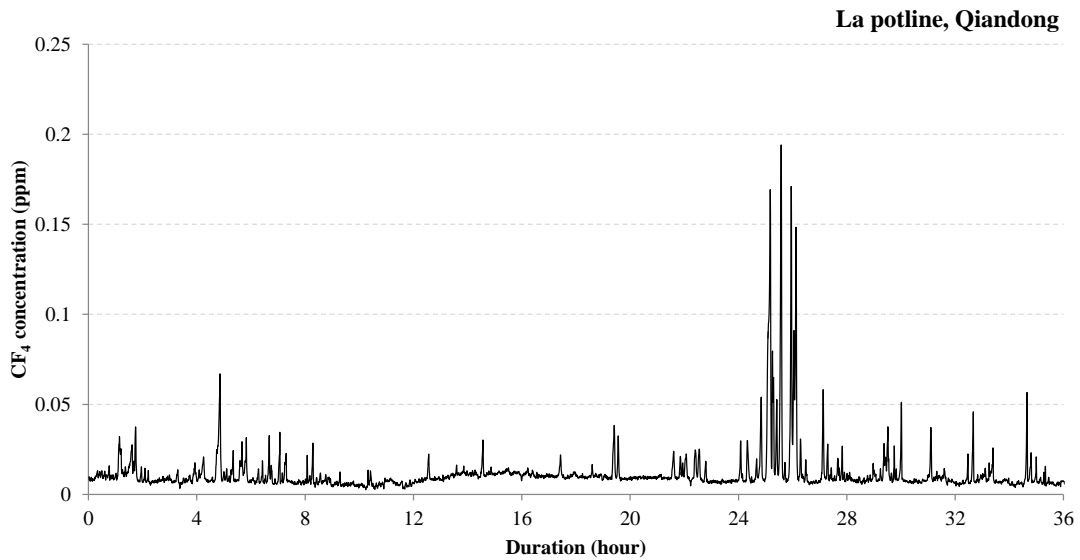
23 The La potline in Qiandong included 6 pots and monitoring was conducted over 36 hours  
24 (Figure 4). Similar to the other observations, a number of anode effect events were also captured  
25 and the maximum concentration of CF<sub>4</sub> was 0.194 ppm. An estimated 61% of emissions were  
26 attributed to AEs, with 39% attributed to NAE emissions. The overall CF<sub>4</sub> emission factor was  
27 36.16 g/t-RE and the CO<sub>2</sub>-equivalent emission factor was 239.74 kg CO<sub>2</sub>-e/t-RE.

28

29

30

1



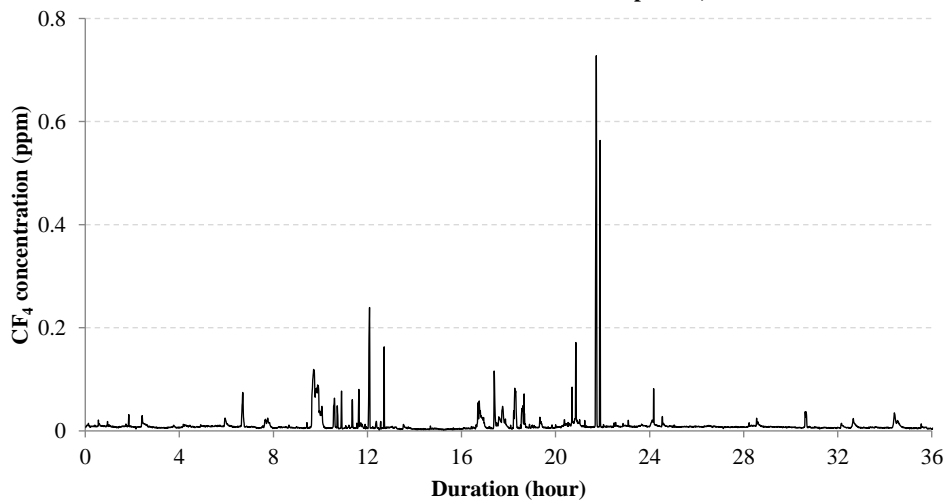
2

3 **Figure 4: CF<sub>4</sub> concentration of La potline (6 cells) in Qiandong during regular production**

4

5 At the Ionic Rare Earth Centre, the Pr-Nd potline which had 6 pots was monitored over 36  
6 hours (Figure 5). Anode effect events were not so frequent compared to other potlines, as shown  
7 by fewer peak emission events. However, when they did occur, peak emissions were greater than  
8 that in the other Pr-Nd potline at Qiandong, with a maximum CF<sub>4</sub> concentration of 0.728 ppm. In  
9 terms of total CF<sub>4</sub>, an estimated 53% were attributed to AEs, 47% to NAE emissions. A CF<sub>4</sub>  
10 emission factor of 33.96 g/t-RE was determined for this potline, equal to 225.15 kg CO<sub>2</sub>-e/t-RE.

**Pr-Nd potline, Ionic Rare Earth Centre**



11

12 **Figure 5: CF<sub>4</sub> concentration of Pr-Nd potline (8 pots) in Ionic Rare Earth Centre during**  
13 **regular production**

14

15 Originally, plans were made to examine the effect of line current and different cell  
16 technologies on PFC emissions, by measuring PFC emissions from Pr-Nd potlines at three  
17 companies for  $\geq 24$  hours, each with varying line current (Table 1). However at Jiangxi South,  
18 continuous monitoring was interrupted after only 8 hours when the company had to change the  
19 settings of the potline and the production equipment. Furthermore, no AE events were observed

1 during the 8 hours of monitoring and therefore emission factors based on this data would likely be  
2 an under-estimate of actual emission intensities. Therefore, monitoring data and a corresponding  
3 emission factor for Jiangxi South's Pr-Nd potline have not been included here. Unfortunately, this  
4 meant that emission factors for a higher-amperage cell technology (>10 kA) could not be  
5 estimated.

6 Table 2 summarizes the emission factors CF<sub>4</sub> for all four potlines (excluding Jiangxi South),  
7 alongside relevant technical process data, determined gas collection efficiencies and parameters  
8 relating to the gas sampling. As explained previously, for the same rare earth metal the magnitude  
9 of PFC emissions can vary considerably with the operating line current. Therefore, it is crucial to  
10 clearly highlight the current while calculating the emission factor. Moreover, it is crucial to take  
11 into account the gas collection efficiency at each company when determining emission factors to  
12 ensure fugitive gas emissions that escape the exhaust gas collection system are accounted for.

13 From SF<sub>6</sub> tracer studies, the gas collection efficiency at both Qiandong and Ionic Rare  
14 Earth Center was found to be approximately 57%. At Jiangxi South, this was around 53%, being  
15 slightly lower. Therefore, significant improvement in gas collection efficiency cannot be achieved  
16 by only small changes to potroom conditions with no fundamental innovation of the current  
17 electrolysis technology.

18 From Table 2, it is clear that estimated CF<sub>4</sub> emission factors vary significantly from one RE  
19 metal/alloy to another. For production of Pr-Nd alloy, CF<sub>4</sub> emission factors also varied between  
20 different companies, with CF<sub>4</sub> emissions at Ionic Rare Earth Center being roughly 80% that at  
21 Qiandong. It is possible that this is due to fluctuation of gas collection efficiency over time at each  
22 company, due to smelter workers' frequent adjustments of the gas collection hoods during  
23 monitoring (each company had differing gas collection systems). Furthermore, this might also be  
24 attributed to differences in process operating conditions (temperature, voltage, etc, as might be  
25 suggested by differences in net anode carbon consumption in Table 2).

26 Also summarized in Table 2 are the contribution of AEs (peak emission events) to total CF<sub>4</sub>,  
27 ranging from 53% to 91% over the four potlines with valid data. These highlight that PFCs can  
28 also be generated *outside* of an anode effect condition. As a point of discussion, since no anode  
29 effects were detected during Jiangxi South monitoring (data not shown here), AE-related  
30 emissions could not be estimated and therefore all measured CF<sub>4</sub> was attributed to NAE emissions.  
31 Again, the 8 hours of monitoring at this production line was deemed insufficient to provide a  
32 meaningful representation of emissions performance at this company and hence emission factors  
33 were not calculated.

34

35

36

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38

1 **Table 2: The emission factor of CF<sub>4</sub> for different production lines based on on-site**  
 2 **monitoring**

Company	Qiandong			Ionic Rare Earth Center	
	Potline	<i>Pr-Nd</i>	<i>Dy-Fe</i>	<i>La</i>	<i>Pr-Nd</i>
Gas flow rate at the sampling point (Nm <sup>3</sup> /h)	12,000	1,340	17,066	18,327	
Diameter of the gas stack (m)	1.2	0.2	0.8	0.6	
Production of RE metal (t/d)	2.173	0.150	0.930	0.965	
Net anode carbon consumption (kg-C/t-RE)	136.5	166.6	144.3	160	
Gas collection efficiency (%)	57.97	57.97	57.97	56.22	
Emission factor of CF <sub>4</sub> (g/t-RE)	26.66	109.43	36.16	33.96	
% CF <sub>4</sub> emissions related to AEs	75%	91%	61%	53%	

3

## 4 **3.2. Estimation of C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> emission factors from Time-Integrated**

### 5 **Sampling & Lab Analysis**

6 Analysis of time-integrated canister samples in the laboratory provided significantly higher  
 7 sensitivities in detecting greenhouse gases. Analysis results confirmed that addition to CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub>  
 8 and C<sub>3</sub>F<sub>8</sub>, enhanced concentrations (concentration of exhaust gas minus background samples) of  
 9 two other PFCs were observed in part of the samples, namely c-C<sub>4</sub>F<sub>8</sub> and C<sub>4</sub>F<sub>10</sub>. However, it was  
 10 found that for all four potlines (Jiangxi South excluded), the ratio of the enhanced concentrations  
 11 of c-C<sub>4</sub>F<sub>8</sub> and C<sub>4</sub>F<sub>10</sub> to CF<sub>4</sub> were <0.05%. This ratio was so low that no further emission factor  
 12 estimation was conducted for these gases.

13 Concentration ratios of C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> to CF<sub>4</sub> are presented in Table 3. It is clear that, in all  
 14 potlines regardless of line current and potroom conditions, CF<sub>4</sub> is consistently the dominant PFC  
 15 gas emitted, taking up more than 90% of total PFCs detected. Furthermore, while C<sub>3</sub>F<sub>8</sub> cannot be  
 16 ignored (compared to c-C<sub>4</sub>F<sub>8</sub> and C<sub>4</sub>F<sub>10</sub>), it contributes to less than 1% of total PFCs and is at the  
 17 lowest concentration amongst the three main PFC gases of interest.

18 By referring to the obtained CF<sub>4</sub> emission factors from FTIR measurements (Table 2) and  
 19 the corresponding concentration ratios determined from the lab analysis of time-integrated  
 20 samples (Table 3), emission factors of C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> from each potline then can be calculated by

Eq. 11 and Eq. 12. Interestingly, the mass ratio of  $EF_{C_2F_6} / EF_{CF_4}$  in two Qiandong potlines (Pr-Nd and Dy-Fe) is similar to the 10% ratio typically found in aluminium electrolysis (IAI, 2017), while the other two RE potlines differed as is the case for different technology cells in aluminium (IAI, 2017).

As presented in Table 3, Qiandong *Dy-Fe alloy* and *La* potlines had  $C_2F_6$  emission factors of 10.95 g/t-RE and 0.26 g/t-RE, respectively, and  $C_3F_8$  emission factors of 0.03 g/t-RE and 0.10 g/t-RE, respectively. Foreseeably, the emission factors of both  $C_2F_6$  and  $C_3F_8$  for Pr-Nd alloy production lines in Qiandong differ from that in the Ionic Rare Earth Center. Specifically, in Qiandong the  $C_3F_8$  emission factor is 0.19 g/t-RE, almost 30% lower than that in Ionic Rare Earth Center; this is not surprising, given that  $CF_4$  emission factors were also ~20% lower at Qiandong. Interestingly, for  $C_2F_6$ , the outcome from Qiandong is 2.98 g/t-RE, around 70% lower than that from Ionic Rare Earth Center, given the ratio of  $C_2F_6$  to  $CF_4$  at Ionic Rare Earth Centre is almost 3 times higher.

**Table 3: The estimated emission factors (EF) for  $C_2F_6$  and  $C_3F_8$**

Company		Qiandong			Ionic Rare Earth Center
Potline		<i>Pr-Nd</i>	<i>Dy-Fe</i>	<i>La</i>	<i>Pr-Nd</i>
No. of pots		15	1	6	6
Line current (kA)		5	5	5	6
Conc. Ratio	$DC_{2F_6} / DC_{CF_4}$	0.0714	0.06384	0.0046	0.2033
	$DC_{3F_8} / DC_{CF_4}$	0.00355	0.00015	0.0013	0.0037
EF Mass Ratio	$EF_{C_2F_6} / EF_{CF_4}$	0.1119	0.1001	0.0072	0.31882
	$EF_{C_3F_8} / EF_{CF_4}$	0.0076	0.00031	0.0028	0.0079
	$EF_{CF_4}$ (g/t-RE)	26.66	109.43	36.16	33.96
	$EF_{C_2F_6}$ (g/t-RE)	2.98	10.95	0.26	10.83
	$EF_{C_3F_8}$ (g/t-RE)	0.19	0.03	0.10	0.27

### 3.3. Uncertainty Analysis

Due to uncontrollable factors in monitoring, the estimated emission factors inevitably have statistically errors which should not be ignored. From Eq. 9, we can infer that the uncertainty pertaining to  $CF_4$  emission factor mainly comes from three aspects, which are the measurement of the emission concentration, of the gas flow rate and of the gas collection efficiency. Suppose the relative standard deviations in these three aspects are  $\varepsilon_{CF_4}$ ,  $\varepsilon_F$ ,  $\varepsilon_{Ecol}$ , the relative standard error ( $\varepsilon_R$ )

1 is calculated as below:

$$2 \quad \varepsilon_R = (\varepsilon_{CF_4}^2 + \varepsilon_F^2 + \varepsilon_{Ecol}^2)^{1/2} \quad (\text{Eq. 13})$$

$$3 \quad U_{CF_4} = EF_{CF_4} \times \varepsilon_R \quad (\text{Eq. 14})$$

4 and the results are listed in Table 4.

5 It is necessary to keep information in these aspects as reference for further estimation of  
6 global CO<sub>2</sub> equivalent emission from rare earth metal electrolysis.

7  
8 **Table 4: CO<sub>2</sub> equivalent emission factors and uncertainty of PFC emissions**

Company		QianDong			Ionic Rare Earth Center
Pot line		<i>Pr-Nd</i>	<i>Dy-Fe</i>	<i>La</i>	<i>Pr-Nd</i>
EF <sub>CF4</sub>	g-CF <sub>4</sub> /t-RE	26.66±2.24	109.43 ±9.63	36.16 ±3.04	33.96 ±1.56
	kg CO <sub>2</sub> -e/ t-RE	176.76±14.84	725.52 ±63.85	239.74 ±20.14	225.15 ±10.36
EF <sub>C2F6</sub>	g-C <sub>2</sub> F <sub>6</sub> /t-RE	2.98±0.28	10.95±1.08	0.26±0.02	10.83±0.71
	kg CO <sub>2</sub> -e/ t-RE	33.11±3.15	121.59±12.04	2.89±0.28	120.18±7.93
EF <sub>C3F8</sub>	g-C <sub>3</sub> F <sub>8</sub> /t-RE	0.19±0.02	0.03±0.01	0.10±0.01	0.27±0.02
	kg CO <sub>2</sub> -e/ t-RE	1.73±0.16	0.30±0.09	0.90±0.09	2.39±0.16
Total PFC EF (kg CO <sub>2</sub> -e/t-RE)		211.60±18.16	847.41±75.97	243.53±20.50	347.72±18.45

9

### 10 **3.4. Discussion on PFC Emission Factors**

11 Using GWPs for the three main PFCs of interest – CF<sub>4</sub>, C<sub>2</sub>F<sub>6</sub> and C<sub>3</sub>F<sub>8</sub> – a converted  
12 CO<sub>2</sub>-equivalent emission factor for total PFCs can be estimated for the four potlines (Table 4). All  
13 emission factors except that for *Dy-Fe* alloy production are lower than 350 kg CO<sub>2</sub>-e/t-RE, an  
14 equivalent target level for the Chinese aluminium industry by 2020. This highlights the  
15 importance of focusing on reducing PFC emissions from *Dy-Fe* alloy production, which had the  
16 highest emissions intensity from this study and the 2014 study (Zhang et al., 2018).

17 It is also pertinent to compare the rare earth PFC emission factors determined in this work  
18 with those from the aluminium industry. When compared to Chinese aluminium industry's 2020  
19 target of 350 kg CO<sub>2</sub>-e/t-Al and the global aluminium industry's 2016 overall PFC emission  
20 intensity of 640 kg CO<sub>2</sub>-e/t-Al (estimated global emission factor from the 2016 IAI Anode Effect  
21 Survey (2017), albeit using GWPs from the IPCC Fourth Assessment Report), this implies that in  
22 most situations PFC emissions intensity (on a mass basis) from rare earth metal electrolysis is on a  
23 lower or similar order of magnitude to emissions from aluminium electrolysis. Note that for  
24 fundamental understanding of PFC generation, the molar mass of rare earth metals (e.g. Nd at 144  
25 g/mol) is roughly 5 times greater than that of aluminium (27 g/mol); therefore, PFC emission

1 intensities for rare earth metals are 5 times greater on a mole-basis than for aluminium. For the  
2 practical purpose of emissions accounting from industry however, it is more appropriate to  
3 compare emission intensities by mass.

4 In a general sense, the rare earth metal production settings such as cell size, voltage and  
5 current have an evident impact upon the PFC emissions. For instance, high cell voltage operation  
6 (e.g. on Dy-Fe cells) can result in more fluoride participating in the electrolysis reaction, thus  
7 generating more anode effects which results in more frequent spikes in PFC emission. Therefore,  
8 emission factors determined for each of the four potlines should be attached with information that  
9 describes these settings.

10 Moreover, it is worth emphasising again that the high emission factor for Dy-Fe alloy has  
11 been based upon observed data from only one single pot. Inevitably it incurs the risk of  
12 uncertainty and may be less representative for *Dy-Fe* alloy production. Nonetheless, we retained  
13 this data so as to maintain coverage over production multiple rare earth metals. In any case, this  
14 salient value presents us with a necessity to explore the underlying causes that lead to greater  
15 emissions and formulate effective solutions to reduce them in *Dy-Fe* alloy production.

16 The comparison of AE-related emissions (ranging from 53% to 91%) vs. NAE-related  
17 emissions is also of particular interest since (1) it highlights that PFC emissions can be generated  
18 on industrial cells outside of an anode effect and (2) similar NAE-emissions have also been found  
19 in industrial aluminium electrolysis (Wong et al., 2015) and are now known in that industry to be  
20 difficult to detect, both by operators and control systems. In contrast, AE-related emissions are  
21 often correlated with high-voltage events on cells, which can be used to detect, mitigate and  
22 account for AE-related emissions. Applied to the rare earths industry, this would mean  
23 NAE-related PFC emissions will be difficult to detect and account for (without sophisticated  
24 instrumentation) and more importantly to mitigate.

25 Finally, the current study provides much greater coverage than previous studies in  
26 providing PFC emission factors for multiple gases ( $\text{CF}_4$ ,  $\text{C}_2\text{F}_6$ ,  $\text{C}_3\text{F}_8$ ) from three of the most  
27 common rare earth metals produced by electrolysis. However, given the lack of valid data from  
28 Jiangxi South, no conclusive observations can be made as to the impact of line-current on  
29 emissions intensity. This would be a good area of focus for further studies by the industry into  
30 PFCs from rare earths.

## 32 4. Conclusions

33 Through on-site FTIR monitoring and lab analysis of time-integrated samples, it was  
34 confirmed that in addition to  $\text{CF}_4$  there are four other PFC emissions in the exhaust gas from  
35 industrial rare earth electrolysis, namely:  $\text{C}_2\text{F}_6$ ,  $\text{C}_3\text{F}_8$ , *c*- $\text{C}_4\text{F}_8$  and  $\text{C}_4\text{F}_{10}$ . While these four  
36 components could not be measured directly by the FTIR, time-integrated samples obtained during  
37 on-site monitoring were analysed in the lab using a custom-built 'Medusa' GC-MS system. The  
38 result shows that in all the four examined potlines (Pr-Nd, Dy-Fe and La production at Qiandong  
39 and Pr-Nd production at the Ionic Rare Earth Center),  $\text{CF}_4$  is absolutely the primary PFC  
40 component, accounting for roughly 90% of PFC gases by mass. The concentration ratio of both  
41  $\text{C}_2\text{F}_6$  and  $\text{C}_3\text{F}_8$  to  $\text{CF}_4$  differs somewhat across these four pot lines – for  $\text{C}_2\text{F}_6$  it is roughly 10% wt  
42 (similar to that in industrial aluminium electrolysis) and for  $\text{C}_3\text{F}_8$  the ratio is less than 1% wt. As  
43 the ratios of the enhanced concentrations of *c*- $\text{C}_4\text{F}_8$  and  $\text{C}_4\text{F}_{10}$  to  $\text{CF}_4$  across four potlines are both



1 less than 0.05%, calculation of their emission factors was not conducted in this research.

2 Emissions vary from one rare earth metal to another and from company to another for the  
3 same metal. In general, PFC emission intensities for most rare earth metals are of a lower level of  
4 magnitude (by-mass) compared to industrial aluminium electrolysis, with the exception of Dy-Fe  
5 electrolysis. Factors behind the variation in emissions intensity include not only electrical line  
6 current, but also cell design, operating conditions and process technology employed – as shown by  
7 reductions in Pr-Nd emissions intensity at Qiandong smelter after improvements to oxide feeding  
8 technology (Zhang et al, 2018 vs. this work) – and as highlighted by differences in operating  
9 voltage, fluoride consumption and anode carbon consumption. Unfortunately, comparison of  
10 emission intensities across low vs. high-amperage cell technologies was inconclusive, given the  
11 lack of valid data at Jiangxi South smelter.

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