

Australian and global HFC, PFC, Sulfur Hexafluoride, Nitrogen Trifluoride and Sulfuryl Fluoride Emissions

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1 Introduction

Hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), sulfur hexafluoride (SF₆) and nitrogen trifluoride (NF₃) are potent greenhouse gases (GHGs), which can be collectively described as Kyoto Protocol synthetic GHGs (SGGs). HFCs are used extensively in Australia, largely in air conditioning and refrigeration, initially as 'ozone-friendly' replacements for chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs). Minor uses of HFCs in Australia are as aerosol propellants, including metered dose inhalers (MDIs), as foam blowing agents and in fire extinguishers. Perfluorocarbons are a by-product of the production of aluminium in Australia and overseas and, in addition, are used overseas in the electronics industry during the manufacture of integrated circuits and plasma screens. Refrigeration represents a minor use of PFCs in Australia and overseas. Sulfur hexafluoride is used extensively in the electricity distribution industry, both in Australia and overseas, for dielectric insulation and current interruption in circuit breakers, switchgear, and other electrical equipment, and as a cover gas in metal production, for example magnesium. There does not appear to be any significant use of nitrogen trifluoride in Australia, which is used internationally in the semi-conductor production industry, initially as a replacement for PFCs.

Sulfuryl fluoride (SO₂F₂) and trifluoromethyl sulfur pentafluoride (CF₃SF₅) are potent synthetic greenhouse gases that are not part of the Kyoto Protocol suite of SGGs. Sulfuryl fluoride use in Australia is growing rapidly, as a replacement for phosphine (PH₃) and possibly methyl bromide (CH₃Br), in grain fumigation. It is unlikely that trifluoromethyl sulfur pentafluoride is used in Australia. Its occurrence in the atmosphere is largely as a by-product of the production of perfluorooctanesulfonic acid (PFOS: CF₃(CF₂)₇SO₃H), which has never been manufactured in Australia, but is a key ingredient in fabric stain repellants (e.g. 3M's Scotchguard[®]) and fire-fighting foams. A major fire-fighting training facility in Victoria (Fiskville) was closed in early-2015 due to unacceptably-high levels of PFOS contamination. However, it is unlikely that there are significant trifluoromethyl sulphur pentafluoride emissions in Australia associated with PFOS use.

Rapidly growing HFC emissions are seen as a significant driver of climate change over the next 50 years (Velders *et al.*, 2007, 2009, 2012; Estrada *et al.*, 2013; Harris, Wuebbles *et al.*, 2014; Myhre *et al.*, 2014; Rigby *et al.*, 2014) and projections suggest that unmitigated HFC growth could result in Global Warming Potential (GWP)-weighted emissions approaching 9 Gt CO_2 -e (carbon dioxide equivalent) per year by 2050 (Daniel & Velders, 2011) and a HFC radiative forcing component as high as 0.4 W/m² by 2050 (Harris, Wuebbles *et al.*, 2014). It has been suggested that an amended Montreal Protocol could phase-down the production and consumption of HFCs (with high GWPs) sooner than possible emission mitigation under a revised Kyoto Protocol or similar agreement (Molina *et al.*, 2009).

The US Environmental Protection Agency (EPA) has proposed delisting or putting controls on a number of high GWP HFCs by end use which were previously considered acceptable alternatives to ozone depleting substances (ODSs). The European Union (EU) also has an HFC phase-down in place. There are no mandated global or Australian targets to phase-down Kyoto Protocol SGG emissions at present, except as contributors to the 'basket' of GHGs (CO_2 , methane – CH_4 , nitrous oxide – N_2O , HFCs, PFCs, sulphur hexafluoride, nitrogen trifluoride) whose total emissions from most developed countries were regulated under the Kyoto Protocol and are currently regulated under post-Kyoto agreements. In early 2010, the Australian Government made a post Kyoto Protocol committment to reduce its total GHG emissions by 5 per cent below 2000 levels by 2020. New GHG emission reduction targets for implementation post-2020 have just been announced by the Australian government: 26%-28% reduction in 2005 emissions by 2030, for submission at the UN Climate Change Conference in Paris late November 2015. This includes a commitment to look at phasing down Australian HFC emissions by 85% by 2036. There are no global or Australian targets to phase-down sulfuryl fluoride or trifluoromethyl sulfur pentafluoride emissions.

Australia reported a total of 542 million tonnes (Mt) CO_2 -e emitted from all GHG sources in 2013 (not including landuse change), which was a decrease from 2012 emissions (550 Mt) of 1.5%. HFC emissions were 10.0 Mt (9965 k tonnes) CO_2 -e in 2013, 6.5% above 2012 emissions, while the sum of HFC, PFC and sulphur hexafluoride emissions was 10.3 Mt (10285 k tonnes, Table 3) CO_2 -e, 5% above 2012 emissions, and 1.9% of total Australian greenhouse gas emissions (DoE, 2015). The total emission of Kyoto Protocol SGGs is the fastest growing emissions sector in the *Australian National GHG Inventory* (referred to subsequently as the *Inventory*).

In this Report, we estimate Australian emissions of HFCs, PFCs, sulfur hexafluoride and sulfuryl fluoride derived by inter-species correlation (ISC), inverse and forward atmospheric modelling techniques, using Cape Grim atmospheric observations. These so-called 'top-down' estimates are compared to estimates of HFCs, PFCs and sulfur hexafluoride emissions submitted by the Australian government to the United Nations Framework Convention on Climate Change (UNFCCC) (DoE 2015), based on Intergovernmental Panel on Climate Change (IPCC)-recommended 'bottom-up' methodologies for estimating national GHG emissions. Australian HFC, PFC and sulphur hexafluoride emissions are compared to global emissions estimated from AGAGE (Advanced Global Atmospheric Gases Experiment; Prinn *et al.*, 2000) atmospheric observations.

Cape Grim *in situ* measurements of nitrogen trifluoride commenced in February 2015 and trifluoromethyl sulfur pentafluoride in late 2010. Preliminary inspections of the data suggest that there are no significant Australian emissions of these species.

2 Measurements of HFCs, PFCs, sulfur hexafluoride, nitrogen trifluoride, sulfuryl fluoride and trifluromethyl sulfur pentafluoride at Cape Grim, Tasmania

Concentrations of HFCs, PFCs, sulfur hexafluoride, nitrogen trifluoride, sulfuryl fluoride and trifluromethyl sulfur pentafluoride have been measured *in situ* in the Southern Hemisphere atmosphere at Cape Grim, Tasmania, as part of the AGAGE program (Prinn *et al.*, 2000), and/or in the Cape Grim Air Archive (1978-2014) at CSIRO (Aspendale), at the Scripps Institution for Oceanography (SIO, USA), and on a sub-set of the Cape Grim Air Archive at the University of East Anglia (UEA, UK). Other flask air samples from Cape Grim have been analysed at CSIRO, at SIO, at UEA and at the University of Heidelberg (Germany). The SGGs have been measured by CSIRO *in situ* in the atmosphere (baseline and non-baseline) at Cape Grim, Tasmania, since the late-1990s (HFCs, PFC-116: CF₃CF₃) and the mid-2000s (other PFCs, sulfur hexafluoride, sulfuryl fluoride). Nitrogen trifluoride (up to 2013) and trifluromethyl sulfurpentafluoride (up to 2008) have been measured on the Cape Grim Air Archive. *In situ* measurements of nitrogen trifluoride and trifluoromethyl sulfur pentafluoride have recently become available (February 2015 for nitrogen trifluoride and recently calibrated trifluoromethyl sulfur pentafluoride going back to late 2010). These data are used, in conjunction with similar data collected from other Northern and Southern Hemispheric sites, to estimate global and regional concentration trends, atmospheric lifetimes, emissions and radiative forcings for these species:

- HFCs
 Oram et al., 1996, 1998; Oram, 1999; Prinn et al., 2000; O'Doherty et al., 2004, 2009, 2014; Greally et al., 2007; Stohl et al., 2009; Miller et al., 2010; Vollmer et al., 2011; Fortems-Cheney et al., 2013; Arnold et al., 2014; Fraser et al., 2014a; Krummel et al., 2014; Rigby et al., 2014; Lunt et al., 2015; Simmonds et al., 2015a
- PFCs
 Oram, 1999; Fraser et al., 2007, 2011, 2013; Mühle et al., 2010; Ivy, 2012; Ivy et al., 2012; Laube et al., 2012; Oram et al., 2012; Kim et al., 2014; Krummel et al., 2014; Rigby et al., 2014; Wong et al., 2015

 SF₆
 Maiss et al., 1996; Oram, 1999; Fraser et al., 2004, 2014a; Levin et al., 2010; Rigby et al., 2010, 2014;
- Ganesan et al., 2014; Krummel et al., 2014
- NF₃ Weiss *et al.*, 2008; Arnold *et al.*, 2013; Rigby *et al.*, 2014
- SO₂F₂ Muhle *et al.*, 2009; Krummel *et al.*, 2014
- CF₃SF₅ Sturges *et al.*, 2012

The abundances and trends of HFCs, PFCs, sulfur hexafluoride, nitrogen trifluoride, sulfuryl fluoride and trifluoromethyl sulfur pentafluoride in the global background atmosphere, as measured at Cape Grim, Tasmania, or in the Cape Grim air archive, are shown in Table 1 (2013-2014) and Figure 1 (1978-2014).

The major HFC in the background atmosphere at Cape Grim (and around the globe) is HFC-134a (73 ppt in 2014), followed by HFC-23 (26 ppt), HFC-125 (15 ppt), HFC-143a (15 ppt), HFC-32 (7.4 ppt) and HFC-152a (4.5 ppt). The cumulative concentration of the minor HFCs (HFC-245fa, HFC-227ea, HFC-236fa, HFC-365mfc, HFC-4310mee) is 3.9 ppt (2014). The total HFC concentration in the background atmosphere is 144 ppt (2014).

The global annual rate of increase of HFC-134a has increased from 4.2 ppt (2012-2013) to 4.9 ppt (2013-2014); the HFC-23 growth rate (0.8 ppt, 2011-2012 & 2012-2013) has increased slightly to 1.0 ppt (2013-2014); the HFC-143a annual increase (1.1 ppt, 2012-2013) has increased to 1.4 ppt (2013-2014); the HFC-125 annual increase (1.3, 2012-2013) has increased to 2.0 ppt/yr (2013-2014); the HFC-152a annual increase has remained steady (0.0 ppt, 2012-2013, 2013-2014) such that HFC-152a is no longer growing in the background atmosphere; the HFC-32 increase (0.9 ppt, 2012-2013) has increased to 1.3 ppt (2013-2014). The minor HFCs (HFC-227ea, HFC-236fa, HFC-365mfc, HFC-4310mee) are growing in the background atmosphere with a cumulative annual growth rate of 0.18 ppt (2013-2014). The annual growth in total HFCs (11 ppt, 2013-2014) has increased from 9 ppt (2012-2013), returning to the annual increase of 11 ppt seen in 2011-2012. Total HFCs in the atmosphere are growing currently (2013-2014) at 8%/yr.

The major PFC in the background atmosphere at Cape Grim (and around the globe) is PFC-14 (CF_4 : 81 ppt in 2014, about half of which is naturally-occurring), followed by PFC-116 (4.3 ppt), PFC-318 (1.4 ppt) and PFC-218 (0.59 ppt). The cumulative concentration of five minor PFCs (PFC-3110, PFC-4112, PFC-5114, PFC-6116, PFC-7118) observed at Cape Grim is 0.8 ppt (2013, 2014). The total PFC concentration in the background atmosphere is 88 ppt (2014),

currently growing at 0.84 ppt/yr (1.0 %/yr). The total anthropogenic PFC concentration in the background atmosphere is 53 ppt growing at 1.6%/yr.

Table 1. Concentrations (2013, 2014) and growth rates (2013-2014) for HFCs, PFCs, sulfur hexafluoride, nitrogen trifluoride, sulfuryl fluoride and trifluromethyl sulfurpentafluoride measured *in situ* at Cape Grim, Tasmania or on air samples collected at Cape Grim (references: see text above; CSIRO unpublished Cape Grim Air Archive data).

Species	ies Formula (Conc. (ppt)		/th	Species	Formula	Conc. (ppt)		Growth	
		2013	2014	ppt/yr	%/yr			2013	2014	ppt/yr	%/yr
HFC-134a	CH ₂ FCF ₃	67.7	72.6	4.9	7.2	PFC-14	CF_4	80.0	80.7	0.70	0.9
HFC-23	CHF ₃	25.2	26.2	1.0	4.0	PFC-14(a) ¹	CF_4	45.1	45.8	0.70	1.6
HFC-143a	CH_3CF_3	13.8	15.2	1.4	10.1	PFC-116	C_2F_6	4.26	4.34	0.08	1.9
HFC-125	CHF ₂ CF ₃	12.6	14.6	2.0	15.9	PFC-318	$c-C_4F_8$	1.37	1.41	0.04	2.9
HFC-32	CH_2F_2	6.1	7.4	1.3	21.3	PFC-218	C_3F_8	0.57	0.59	0.02	3.5
HFC-152a	CH_3CHF_2	4.5	4.5	0.0	0.0	PFC-5114 ²	$C_{6}F_{14}$	0.27	0.27	0.005	1.8
HFC-245fa	$CH_3CF_2CF_3$	1.7	1.8	0.13	7.9	PFC-3110 ²	C_4F_{10}	0.17	0.17	0.002	1.2
HFC-227ea	$CHF_2CF_2CF_3$	0.88	0.97	0.096	11.0	PFC-4112 ²	C_5F_{12}	0.12	0.12	0.001	1.1
HFC-365mfc	$CH_3CH_2CF_2CF_3$	0.67	0.73	0.059	8.8	PFC-6116 ²	$C_{7}F_{16}$	0.12	0.12	0.004	3.4
HFC-4310mee	$CF_3(CHF)_2C_2F_5$	0.22	0.23	0.014	6.4	PFC-7118 ²	C_8F_{18}	0.09	0.09	0.001	1.1
HFC-236fa	CH ₂ FCF ₂ CF ₃	0.113	0.124	0.011	9.7	total PFCs		87.0	87.8	0.84	0.97
total HFCs		133	144	10.9	8.2	total PFC(a)		52.07	52.91	0.84	1.61
HFC fluorine		493	534	41.0	8.3	PFC fluorine		372	375	3.0	0.81
sulfur hexafluoride	SF ₆	7.7	8.0	0.30	3.9	nitrogen trifluoride	NF_3	1.0	1.1	0.10^{3}	10
sulfuryl fluoride	SO_2F_2	1.78	1.90	0.12	6.5	total fluorine		918	966	48	5.2
trifluoromethyl	CF_3SF_5	0.173	0.168	-0.005	-2.9						
sulfurpentafluoride											

¹ PFC-14 (a) = CF₄ (anthro.) = total CF₄ – natural CF₄ (= 34.9 ppt, Mühle *et al.*, 2010)

² extrapolated from 2011 data (Ivy *et al.*, 2012)

³ estimated from Cape Grim & global data; assumed = 2011-2012 growth rate (Arnold *et al.*, 2013)

The annual rate of increase of PFC-14 in the atmosphere has increased from 0.56 ppt (2012-2013) to 0.70 ppt (2013-2014). The anthropogenic component (from aluminium production and the electronics industries) of the PFC-14 atmospheric abundance is growing at 1.6%/yr. The annual rate of increase of PFC-116 (CF_3CF_3) had previously remained steady at 0.06 ppt in 2011-2012 and 2012-2013, and has now increased slightly to 0.08 ppt (2013-2014); the PFC-218 annual increase (0.02 ppt, 2013-2014) is similar to the increase observed over the previous two years (2011-2013, 0.01-0.016 ppt/yr); the PFC-318 annual increase is also similar to previous years (0.04 ppt 2013-2014) compared with (0.05 ppt 2012-2013 and 0.04 ppt 2011-2012). The minor PFCs (PFC-3110, PFC-4112, PFC-5114, PFC-6116, PFC-7118) are growing in the background atmosphere with a cumulative annual growth rate likely to be of the order 0.01-0.02 ppt (2013-2014).

Annual mean sulfur hexafluoride levels reached 8.0 ppt in 2014 at Cape Grim, growing at 0.30 ppt/yr (3.9%/yr), slightly higher than the 2012-2013 growth rate (0.27 ppt, 3.6%/yr).

Annual mean sulfuryl fluoride levels reached 1.9 ppt in 2014 at Cape Grim, growing at 0.12 ppt/yr (6.5%/yr), compared to the 2012-2013 growth rate of 5.8%/yr.

Trifluoromethyl sulfur pentafluoride stopped growing in the Cape Grim atmosphere in 2007 (0.16 ppt), as seen in UEA Cape Grim Air Archive measurements, following 3M's decision to cease PFOS production (Santaro, 2000). *In situ* measurements at Cape Grim have recently been calibrated (SIO-14 scale) and show annual mean concentrations of 0.173 ppt for 2013 and 0.168 ppt for 2014, which may indicate a decrease of 0.005 ppt/yr, which is similar to the current measurement precision (± 0.004 ppt, 1 standard deviation). If there is no further production/release of trifluoromethyl sulfur pentafluoride, its concentration in the atmosphere should remain effectively constant due to its very long atmospheric lifetime (800 yr). With zero emissions, atmospheric concentrations should decline by only 0.05% (<0.001 ppt)/yr.

Nitrogen trifluoride is growing rapidly in the background atmosphere (0.10 ppt/yr, 10%/yr). In February 2015, *in situ* measurements of nitrogen trifluoride began at Cape Grim. The mean concentration of nitrogen trifluoride for February-April 2015 time period is 1.16 ppt, growing at 0.12 ppt/yr (10%/yr). Following the recent inclusion of nitrogen trifluoride into the post-Kyoto Protocol 'basket' of GHGs, it is anticipated that the current rapid growth rate will decline as alternatives are introduced into the semiconductor manufacturing industry.



Figure 1. *In situ* observations of PFCs, HFCs, sulfur hexafluoride, nitrogen trifluoride, sulfuryl fluoride and trifluoromethyl sulfur pentafluoride (1998 – 2014) showing baseline monthly mean data (dark green, Medusa; magenta, ADS) and total data (light green, Medusa; pink, ADS) obtained from the GC-MS-Medusa and GC-MS-ADS instruments at Cape Grim and from Medusa measurements at CSIRO and SIO on the Cape Grim Air Archive (orange). 2011 and 2012 nitrogentrifluoride annual means are derived from global data; 2009-2012 trifluoromethyl sulfur pentafluoride annual means are assumed = 2008; the *in situ* trifluoromethyl sulfur pentafluoride data are preliminary (references: see text above; CSIRO unpublished Cape Grim Air Archive data).

3 Global HFC, PFC, sulphur hexafluoride, nitrogen trifluoride and sulfuryl fluoride emissions

Global emissions of HFCs, PFCs, sulphur hexafluoride, nitrogen trifluoride and sulfuryl fluoride have been estimated from AGAGE global data (including Cape Grim data) by inverse modelling (Figure 2; Rigby *et al.*, 2014).



Figure 2. Global HFC, PFC, sulphur hexafluoride and nitrogen trifluoride emissions (left: k tonnes; right: M tonnes CO₂e) from global AGAGE atmospheric measurements (Rigby *et al.*, 2014).

Total global HFC emissions reached nearly 400 k tonnes per year in 2014, rising by near 20 k tonnes/yr since 1995; total global HFC emissions in 2014 were 8% higher than in 2013. Global emissions of the major HFCs (HFC-23, HFC-32, HFC-125, HFC-134a, HFC-143a) increased from 2013 to 2014: 7%, 15%, 19%, 6%, 3% respectively; global emissions of HFC-152a peaked in 2011 and are now in decline. HFC-134a emissions exhibit significant seasonality, in particular at the mid-latitudes of the northern hemisphere, with summer emissions 2-3 times winter emissions (Xiang *et al.*, 2014).

Total PFC emissions have been declining steadily (0.17 k tonnes/yr since 1995) from 19 k tonnes/yr in the late-1970s to 13 k tonnes/yr in 2014. The 2013-2014 decrease was nearly 4%. Global emissions of the major PFCs (PFC-14, PFC-216, PFC-318) decreased from 2013 to 2014: 4%, 2%, 4% respectively.

Sulfur hexafluoride emissions have been increasing (0.16 k tonnes/yr since 1995) steadily from about 3 k tonnes per year in the late-1970s to approaching 9 k tonnes per year in 2014, with a 2013-2014 increase of about 4%.

Significant nitrogen trifluoride emissions (>0.1 k tonnes per year) were first observed in the mid-1990s, growing (0.09 k tonnes/yr since 1995) to nearly 2 k tonnes per year in 2014. The 2013-2014 incease was 6%.

Total global HFC, PFC, sulphur hexafluoride, nitrogen trifluoride emissions have risen (19 k tonnes/yr, largely HFCs) from about 30 k tonnes per year in the late-1970s to over 420 k tonnes per year in 2014, with a 2013-2014 increase of 7%.

Global sulfuryl fluoride emissions (not shown in Figure 2) were 1.5 k tonnes per year in the late-1970s, rising to about 3 k tonnes/year in 2013/2014; in 2013-2014 sulfuryl fluoride emissions deceased by 13%. This may represent a turning point in global sulfuryl fluoride emissions, although the global demand for sulfuryl fluoride is presumably dependant on, in part, variable global grain production.

4 Australian HFC, PFC and sulfur hexafluoride imports, banks and emissions

HFCs and sulphur hexafluoride are not manufactured in Australia and estimates of Australian HFC and sulphur hexafluoride emissions, that are reported annually to the UNFCCC (see below), are based on import data (Table 2). HFCs and sulphur hexafluoride are imported as bulk chemicals or in pre-charged equipment (PCE), leading to estimates of 'banks' of HFCs and sulphur hexafluoride stored in equipment or products such as refrigerators, air conditioners, fire extinguishers, foams, aerosols and electrical equipment. Only small amounts of PFC are imported into Australia (Table 2) and these are not considered to be a significant source of PFC emissions; PFC emissions are assumed to originate exclusively from aluminium production (see below).

HFC imports in 2014 were 3745 tonnes, 26% in bulk, 74% in pre-charged equipment (PCE), 26% higher than HFC imports in 2013. PFC imports in 2013 and 2014 were about 0.1 tonnes. Sulfur hexafluoride imports in 2014 were 13 tonnes, 28% lower than 2013 imports.

The HFC bank in 2013 has been estimated at 58.7 M tonnes CO_2 -e (DoE, 2015) and at nearly 36,000 tonnes raw gas, 43% HFC-134a, 28% HFC-125, 24% HFC-32 and 5% HFC-143a (Brodribb & McCann, 2013, 2014) and the sulphur hexafluoride bank has been estimated at about 550 tonnes (DoE, 2015). The 2013 bank is based on a 2.1% increase in the bank from 2012.

Table 2. Australian HFC, PFC, sulfur hexafluoride imports (2013, 2014, tonnes; A. Gabriel, Department of Environment, 2014), HFC bank (2013, tonnes; Brodribb & McCann, 2013, 2014), total HFC imports in Mt CO_2 -e (DoE, 2015) and sulphur hexafluoride bank (2013, tonnes; A. Gabriel, Department of Environment, 2014); PCE = pre-charged equipment.

Species	b	ulk		PCE	То	tal	Total	Aust. bank
							Mt CO ₂ -e	
HFCs	2013	2014	2013	2014	2013	2014	2013	2013
HFC-23	0.06	0.3	0.03	0.1	0.1	0.4		
HFC-32	5.7	30	935	1059	941	1088		8738
HFC-125	16	59	883	888	899	947		10167
HFC-134, -134a	109	702	825	788	934	1490		15408
HFC-143, -143a	4.9	31	15	20	20	51		1657
HFC-152a	1.3		12	21	13	21		
HFC-227ea	0.01	14	1.2	1.7	1.2	16		
HFC-245fa	74	64	0.02	0.01	74	64		
HFC-365mfc	86	67			86	67		
total HFCs	297	967	2671	2778	2968	3745	14.78	35970
PFCs								
PFC-14	0.01	0.04	0.01	0.01	0.02	0.05		
PFC-116	0.02	0.02	0.03	0.05	0.05	0.06		
PFC-218	0.004	0.001	0.03		0.03	0.001		
PFC-318	0.03	0.01			0.03	0.01		
total PFCs	0.06	0.07	0.07	0.06	0.13	0.12		
sulphur hexafluoride	3.6	0.7	14	13	18	13		552
total HFCs, PFCs, SF ₆	301	968	2685	2791	2986	3758		

The National Greenhouse Gas Inventory (NGGI: ageis.climatechange.gov.au) published in 2015 contains estimates of Australian emissions of HFC-23, HFC-32, HFC-125, HFC-134 (CHF₂CHF₂, not measured currently at Cape Grim), HFC-134a, HFC-143a, HFC-152a, HFC-227ea, HFC-236fa, HFC-4310mee, PFC-14, PFC-116 and sulfur hexafluoride, up to 2013, and is part of the National Inventory Report 2013 (DoE, 2015), which is the Australian government submission to the United Nations Framework Convention on Climate Change and which forms a part of the Australian National Greenhouse Accounts (NGA). Note the annual emissions in the NGGI and in the National Inventory Report are for fiscal years, i.e. '2013' emissions are emissions for July 2012 to June 2013.

The HFC emissions (Table 3) are based on HFC import data (Table 2), as bulk HFCs or PCE, leading to an estimate of 'banks' of HFCs stored in equipment or products (refrigerators, air conditioners, fire extinguishers, foams, aerosols - largely metered-dose inhalers) and appropriate application-dependent emission factors from those 'banks', which allow for emissions during the lifetime of the application, as well as emissions from initial charging/re-charging of equipment and equipment disposal. The HFC emissions model contains some assumptions that simplify HFC emissions calculations. For example, the Australian HFC mix, as determined by HFC imports, is assumed to be invariant across all

HFC use categories and the application-specific emission factors are time-invariant. This means that the emissions model is likely to be more representative for total HFC emissions than for individual HFC emissions.

	HFC-134a	HFC-125	HFC-143a	HFC-32	HFC-227ea	HFC-134	HFC-43-10mee	HFC-23	total	HFCs	PFC-14	PFC-116	tota	PFCs	S	SF ₆	HFC,P	FC,SF6
					tonnes				tonnes	kt CO ₂ -e	ton	nes	tonnes	kt CO ₂ -e	tonnes	kt CO ₂ -e	tonnes	kt CO ₂ -e
1995	33	12	1.1	0.6	0.4	0.0	0.0	61	108	1004	171	22	193	1531	13	302	314	2837
1996	145	51	4.9	2.8	1.7	0.1	0.0	0.0	205	414	157	20	178	1410	12	270	395	2094
1997	247	86	8.3	4.8	2.9	0.1	0.1	0.0	349	705	137	18	155	1228	11	248	515	2182
1998	349	122	12	6.8	4.1	0.1	0.1	0.0	494	998	185	24	209	1661	9.8	223	713	2882
1999	481	168	16	9.3	5.7	0.2	0.1	0.1	681	1374	127	16	143	1139	8.6	196	833	2709
2000	565	197	19	11	6.7	0.2	0.2	0.1	799	1613	143	19	162	1287	8.4	191	969	3091
2001	807	282	27	16	9.6	0.3	0.2	0.1	1142	2306	201	26	227	1802	8.6	197	1377	4304
2002	1024	358	34	20	12	0.4	0.3	0.1	1449	2927	193	25	218	1728	8.9	202	1676	4857
2003	1252	438	42	24	15	0.5	0.4	0.2	1772	3578	188	24	212	1684	9.0	206	1993	5469
2004	1493	522	50	29	18	0.6	0.4	0.2	2113	4267	191	25	216	1714	9.1	208	2338	6189
2005	1751	612	59	34	21	0.7	0.5	0.2	2477	5002	200	26	226	1792	8.0	182	2711	6976
2006	1909	668	64	37	23	0.7	0.5	0.2	2702	5457	77	10	87	687	7.6	173	2796	6316
2007	2150	752	72	42	26	0.8	0.6	0.3	3043	6144	65	8	73	583	7.1	162	3123	6889
2008	2391	836	80	46	28	0.9	0.7	0.3	3383	6832	50	6	56	445	6.6	151	3446	7428
2009	2614	914	87	51	31	1.0	0.7	0.3	3699	7469	40	5	45	359	6.0	137	3750	7964
2010	2858	999	96	55	34	1.1	0.8	0.4	4044	8166	32	4	36	283	6.1	139	4086	8588
2011	3093	1082	104	60	37	1.2	0.9	0.4	4377	8838	35	4	38	301	5.6	128	4421	9267
2012	3273	1145	110	63	39	1.3	0.9	0.4	4632	9353	34	4	37	295	5.6	128	4675	9776
2013	3487	1219	117	67	41	1.4	1.0	0.4	4935	9965	22	2	24	192	5.6	128	4965	10285

 Table 3. Australian HFC, PFC and sulfur hexafluoride emissions (http://unfccc.int/national_reports/ annex_i_ghg_inventories/national_inventories_submissions/items/8812.php).

 HFC-23 emissions in 1995 from HCFC-22 production in Sydney. Not shown are small HFC-236fa emissions (0.004 tonnes in 2012). Note GWPs used are AR4.

The Australian HFC mix in the emissions model is 50% HFC-134a, 43% HFC-125, 5.2% HFC-143a, 0.5% HFC-32 and 1.3% HFC-227ea. The HFC mix in the 2013 import data (Table 2) is 31% HFC-134a, 30% HFC-125, 1% HFC-143a, 32% HFC-32, 2.5% HFC-245fa and 3% HFC-365mfc. The HFC mix in the Australian bank in 2013 is 43% HFC-134a, 28% HFC-125, 5% HFC-143a and 24% HFC-32 (Brodribb & McCann, 2014).

There have been a number of refinements to the HFC emissions model used for the 2014 submission:

- (i) use of country-specific (Australian) annual leakage rates for commercial refrigeration and air-conditioning, transport refrigeration and heavy vehicle air rather than default leakage rates;
- (ii) quality control checks on bulk import data showed that the previous version of the emissions model was not allocating 100% of bulk gas imports to domestic production or replenishment, leading to a minor systematic under-estimation of emissions from all equipment types; this has been corrected for the current submission to UNFCCC;
- (iii) a revision to the quantities of bulk gas allocated to foams, aerosols and fire protection equipment; and
- (iv) a minor revision to the calculation of emissions from initial charging of new equipment.

The relationship between HFC imports and emissions is not necessarily linear. For example there was a surge of HFC and sulfur hexafluoride imports into Australia in 2012 in anticipation of the imposition of the carbon tax; this presumably did not lead to a significant surge in emissions, as the HFC and sulfur hexafluoride imports, surplus to demand, presumably went into storage with minimal emissions.

In the Australian GHG emission inventory, PFC (PFC-14, PFC-116) emissions only arise from aluminium production, with total PFC emissions in 2013 of 0.19 Mt CO_2 -e (DoE, 2015). About 0.1 tonnes of PFCs (PFC-14, PFC-116) were imported into Australia in 2013 and 2014 as refrigerant blends in bulk and PCE (Table 2). It is not clear whether these PFC imports are used to calculate corresponding contributions to PFC emissions – if they are, they are very small compared to PFC emissions from the aluminium industry.

Australian sulfur hexafluoride emissions are largely from the electricity supply and distribution network. Emissions (1975-2013) are estimated as leakages from sulfur hexafluoride 'banks' and from Australian manufacture of electricity supply equipment, using a combination of default IPCC and Australian-specific emissions factors. There are three years (2012, 2013, 2014) of sulfur hexafluoride import data available, which could potentially be used to verify the calculations of sulfur hexafluoride in banks and used in manufacture. However the 2012-2013 import data suggest sulfur hexafluoride may have been stock-piled in these years in anticipation of an impending carbon tax impost on the sulfur hexafluoride price.

Australian emission factors for electrical equipment stock are global IPCC default factors: 0.05 t/t (1975-1995), 0.02 t/t (2000); an Australian-specific factor (0.0089 t/t) has been estimated for 2009 and assumed constant thereafter. For 1995-2000-2009 periods, emission factors are interpolated. The 2009-2012 emission factor is based on emission estimates from 15 utilities using their own data on sulfur hexafluoride consumption (consumption = emissions, not defaulting to the IPCC method). The emission factors assumed for Australian equipment manufacture (0.15 t/t, 1975-1995; 0.06 t/t, 1996-2012) are IPCC default factors for Europe. The original global IPCC default factor for manufacturing (0.74 t/t) was significantly higher and currently IPCC recommends 0.30-0.35 t/t (prior to 1995) and 0.12 t/t after 1995, both significantly higher than the assumed Australian emission factors are recommended for circuit breakers (0.55 t/t prior to 1995, 0.29 t/t after 1995). Circuit breakers are used extensively in the USA, GIS in Europe. The Australian inventory assumed GIS values, presumably because Australia uses largely GIS equipment in its electricity distribution networks.

Australian sulfur hexafluoride emissions from magnesium casting, tracer gas studies, eye surgery etc. have been estimated at 0.034 g/capita/yr (M. Hunstone, DoE, 2014, personal communication).

There are no import data available for sulfuryl fluoride.

The HFC, PFC and sulphur hexafluoride emissions data (1995 to 2013) are detailed in Table 3. Australian HFC emissions were about 100 tonnes in 1995, rising to 4935 tonnes (9965 CO_2 -e tonnes) in 2013. HFCs are the dominant emissions in CO_2 -e terms in this sector (97%, 2013). HFC-134a emissions increased by about 214 tonnes (6%) from 2012 to 2013, all other HFCs by about 88 tonnes (6%). Emissions of total HFCs (in CO_2 -e terms) in 2013 were 6% higher than in 2012. Australian PFC emissions were 190 tonnes in the mid-1990s, rising to 230 tonnes in the mid-2000s, before dropping to 24 tonnes in 2013. Total PFC emissions (in CO_2 -e terms) decreased by 42% from 2012 to 2013. Sulfur hexafluoride emissions are estimated to have been 13 tonnes in 1995 falling to 6 tonnes in 2013. Total HFC, PFC and sulfur hexafluoride emissions in CO_2 -e in 2013 (10285 k tonnes) were 5% higher than in 2012; this compares to a 7%/yr increase from 2007 to 2012. The overall uncertainty on the PFC/HFC/sulfur hexafluoride emissions category in the NGGI is 25-30% (DCCEE, 2011).

The HFC, PFC and sulfur hexafluoride contributions to total emissions from this sector are shown in Figure 3. The significant impact on total emissions in 2005-2006 due to reduced PFC emissions (the Kurri Kurri aluminium smelter upgrade) can be clearly seen. Prior to the significant reduction in PFC emissions at Kurri Kurri in 2005, Australian HFC/PFC/sulfur hexafluoride emissions grew at about 13%/yr (Figure 3). After the Kurri Kurri upgrade, these combined emissions grew at about 7%/yr. The Australian KP-SGG emissions are 95% HFCs, 3% PFCs and 2% sulfur hexafluoride.



Figure 3. Australian HFC, PFC and sulfur hexafluoride emissions (M tonne CO_2 -e) (DoE, 2015). Note the impact on the reduction in emissions due to the refurbishment of potline #1 at the Kurri Kurri aluminium smelter in 2005-2006. Dashed lines are exponential best fits.

5 Australian HFC, PFC, sulfur hexafluoride and sulfuryl fluoride emissions from atmospheric data

5.1 PFC-14 emissions

SE Australian emissions of PFC-14 (CF₄) are evident in the PFC-14 data collected at Cape Grim (Figure 4). Inspection of Figure 4 shows an overall decline in intensity of PFC-14 pollution episodes due to declining emissions. The year-to-year variability of the number and intensity of PFC-14 episodes seen at Cape Grim is large, so 3-yr averaging is used when deriving PFC emissions from these data (so the latest annual emission calculated from these data is for 2013). Detailed analysis of these PFC-14 pollution episodes shows clearly that the Cape Grim PFC-14 pollution originates largely from southern Victoria (from the Point Henry and Portland smelters), with some contribution from Bell Bay, Tasmania.

The Cape Grim PFC-14 pollution episode data have been used to estimate PFC-14 emissions from these SE Australian smelters. A regional transport model (TAPM – The Air Pollution Model; Hurley, 2008; Hurley *et al.*, 2008) is used to calculate emissions in which PFC-14 is released into the model atmosphere from the Point Henry, Portland and Bell Bay locations, with annual emissions that are varied, so that the resultant PFC pollution episodes seen at Cape Grim in the model are a best-fit match to observations. The emissions are constant in any one year, varied year-by-year. Smelter-specific emission factors are calculated from the emissions by dividing by the smelter-specific annual aluminium production (Fraser *et al.*, 2007, 2011).



Figure 4. Monthly-mean PFC-14 concentrations observed *in situ* at Cape Grim (dark blue), 2004-2014. PFC-14 pollution episodes (light blue), typically lasting up to 12 hours in duration, are also seen at Cape Grim (Mühle *et al.*, 2010). The NAME particle dispersion model (O'Doherty et al., 2009; Manning et al., 2003, 2011) is also used to calculate PFC-14 emissions from Victoria/Tasmania/NSW (Portland/Pt Henry/Bell Bay/Kurri Kurri/Tomago smelters). NAME (Numerical Atmospheric Dispersion Modelling Environment) is a Lagrangian particle dispersion model driven by 3dimensional wind fields from numerical weather predictions models. NAME has a horizontal resolution (grid boxes 40 km x 40 km) and a minimum boundary layer height of 100 m. NAME operates in a backward mode, so, for example, it identifies, within a 3 hr period at Cape Grim, which grid boxes in the prescribed domain impact on Cape Grim in the previous 12 days. NAME releases 33,000 particles at Cape Grim over the 3 hr period and the resultant 12-day integrated concentrations in each of the domain boxes are calculated. Operating NAME in the backward mode is numerically very efficient and is a very close approximation to the forward running mode, which is what is used to identify emission sources impacting on Cape Grim. In the inverse calculation, NAME identifies pollution episode data at Cape Grim, and starts with randomly-generated emission maps and searches for the emission map that leads to a modelled pollution time series that most accurately mimics the observations. The inversion method assumes that baseline air enters the inversion domain regardless of direction i.e. it assume that sources outside the specified domain do not impact significantly on Cape Grim. One of the major advantages of the NAME method, especially when using Cape Grim data, is that it does not require a prior estimate of emissions. Other inversion methods used to estimate regional emissions using Cape Grim data often derive emissions that are not significantly different than the prior estimates.

The TAPM and NAME estimates of regional PFC-14 emissions are scaled to derive Australian emissions on an aluminium production basis. For example, if the PFC-14 emissions derived for the SE Australian smelters, which account for about 35% of Australia's aluminium production, are assumed to be representative of all Australian aluminium production, then Australian PFC-14 emissions from aluminium production can be derived and compared to emissions in the *National Inventory Report 2013* (DoE, 2015) (Table 4). The emission factors and emissions calculated for the Point Henry, Portland and Bell Bay smelters are shown in Table 4 and Figures 5 and 6, together with national average emission factors and emissions as published in the *National Inventory Report 2013* (DoE, 2015). The Australian PFC-14 emission factors for aluminium production (Figure 5) are below the self-imposed global industry target of 80 g/tonne aluminium by 2010 and are well on the way to achieving the 2020 target of 20 g/tonne by 2020.



Figure 5. Australian PFC-14 emission factors as reported in the *National Inventory Report 2013* (NGA: DoE, 2015) and as derived from atmospheric measurements at Cape Grim using NAME/TAPM (to 2011) and TAPM (to 2013) modelling. The grey band is the average (±1 sd) emission factor derived from NAME/TAPM. The Kurri Kurri emission factor is based on direct PFC-14 measurements made at the Kurri Kurri smelter in 2009 (Fraser *et al.*, 2013).

Figure 6. Australian emissions of PFC-14 as recorded in the *National Inventory Report 2013* (NGA: DoE, 2015; uncertainty 27%: DIICCSRTE, 2013) and as obtained from TAPM (scaled from Vic/Tas emissions) and NAME-InTEM (scaled from Vic/Tas/NSW emissions) modelling. **Table 4.** Australian aluminium production (kt: 10³ tonnes), Australian PFC-14, PFC-116, PFC-218, PFC-318 emissions (t: tonnes) and PFC-14, PFC-116 emission factors (g/t aluminium, 2005-2013)derived from atmospheric observations – remote (Portland, Pt Henry, Bell Bay) (3 yr averages, e.g. 2010 = average of 2009, 2010, 2011), using the TAPM and NAME models, directly, at Kurri Kurri, and as reported in the *National Inventory Report 2013* (DoE, 2015).

	2005	2006	2007	2008	2009	2010	2011	2012	2013	Average
Alproduction										(2005-13)
	244	240	251	251	270	217	217	21E	212	220
Point Henry Vic ¹	100	100	107	108	520 185	176	177	177	176	185
Bell Bay, Tas ²	174	178	178	190	105	170	101	185	197	180
Vic/Tas	708	708	727	729	690	667	672	678	676	695
Kurri Kurri NSW ³	151	164	173	173	177	179	180	67	-	158
Tomago NSW ⁴	523	523	523	523	528	528	541	548	545	531
Vic/Tas/NSW	1382	1395	1423	1425	1395	1374	1393	1293	1220	1367
Australia ⁵	1890	1912	1954	1965	1980	1926	1943	1943	1786	1922
Inventory		-								
PFC-14 emiss.	200±60	76±22	64±19	49±15	40±12	29±9	31±9	29+9	21±6	60±18
emiss. fact.	106±32	40±12	33±10	25±8	20±6	15±5	16±5	15±5	12±4	31±9
PFC-116 emiss.	26±8	10±3	8±2	6±2	4±1	2±1	2±1	2±1	2±1	7±2
emiss. fact.	14±4	5±2	4±1	3±1	2±1	1±0.3	1±0.3	1±0.3	1±0.3	4±1
TAPM										
PFC-14 emiss.										
Portland, Vic	17±3	17±4	14±4	15±5	12±4	12±3	13±4	11±3	10±3	14±4
Point Henry, Vic	12±2	11±2	10±2	10±2	9±2	7±2	8±3	8±2	7±1	9±2
Bell Bay, Tas	5±1	5±1	6±3	4±2	3±2	3±2	3±2	3±2	4±2	4±2
Vic/Tas	34±7	34±7	30±10	29±9	25±8	23±7	25±9	22±7	21±6	27±8
Australia⁵	93±19	92±19	79±26	77±24	70±22	67±21	69±25	64±21	57±16	74±21
NAME										
PFC-14 emiss. ⁷	182±65	157±75	68±55	45±30	44±30	47±30	51±30	51 ¹³		85±50
PFC-116 emiss. ⁸	15±1	15±1	15±1	15±1	19±3	23±5	29±3	29 ¹³		20±2
PFC-218 emiss. ³	9±1	10±1	11±1	12±1	19±3	21±3	20±2	2013		15±5
TAPM/NAME avera	age								14	
PFC-14 emiss.	137±42	124±48	73±40	61±27	57±26	57±26	60±28	64±21	57±16 ¹⁴	77±30
PFC-116 emiss.°	14±4	12±5	7±4	6±3	6±3	6±3	6±3	6±2	6±2 ¹⁴	8±3
ISC										
PFC-218 emiss.	8±3	9±3	10±4	7±3	9±4	14±6	16±6	15±5	9±3	11±4
PFC-318 emiss.	27	27	27	27**	27±18	23±9	21±7	18±6	15±5	21±9
ISC/NAME average								14	14	
PFC-218 emiss.	9±2	9±2	10±3	10±2	14±3	17±4	18±4	15±5 ⁻⁺	9±3 ¹⁴	12±3
total PFC emiss.	187±45	173±50	118±40	104±30	104±30	103±30	105±30	103±25**	87±20 ^{±+}	120±33
CO ₂ -e M t	1.54±0.38	1.43±0.43	1.00±0.37	0.89±0.25	0.90±0.44	0.88±0.36	0.89±0.35	0.86±0.29	0.73±0.22	1.01±0.34
PFC-14 emission fa	ctors	52.44	40.40	12:45	07.40	20:44	10:10	24:40	22.0	
Portiand, Vic	52±10	52±11	40±12	42±15	3/±12	39±11	40±13	34±10	32±8	41±11
Point Henry, Vic	03±12 21±0	20+0	52±11	2119 22±12	48±10	40±11 10±11	45±15	43±11 10±11	42±8	49±11 22±12
Australia (TADM) ⁹	51±0 49±10	50±0 49±10	52±19 /1+1/	22±12	19±11 25±11	19±11 22±11	19±11 25±12	19±11 22+11	21+0	22±15 20+11
Australia (IAPIN)	40±10	40±10 82±40	25+20	22±14	22+15	25+15	27±15	52111	3119	J0±11 11+11
	90±34	02±40	55±20	25±14	20±12	20±12	27±15	27 ⊥11	21±0	44±11 40±11
APIVI/INAIVIE dVg	12122	05125	20171	21112	29113	29113	51 <u>1</u> 14	32 <u>1</u> 11	2113	49111
Kurri Kurri ¹¹					35±3					
¹ Alcoa Annual Sustair	nability Rep	orts, 2003,2	004, 2005, 2	006, 2009, 2	2011/2012					
² Bell Bay Aluminium	Environmer	ntal Sustaina	bility Report	2012, 2013						

⁴ Rio Tinto Annual Report 2012, 2013

⁵ DoE (2014), uncertainty 25-30% - DCCEE (2012)

⁶ Australian emissions scaled from Portland, Pt Henry, Bell Bay emissions by aluminium production

⁷ Australian emissions scaled from Portland, Pt Henry, Bell Bay, Kurri Kuri, Tomago emissions by aluminium production

 ${}^{8}C_{2}F_{6}/CF_{4} = 0.1$, Kim *et al.*, (2014)

⁹ TAPM (Portland, Pt Henry, Bell Bay average emissions)/aluminium production

¹⁰ NAME (Vic/Tas/NSW) emissions/aluminium production NAME emissions for 2011 are preliminary – based on 2010-2011 data only

¹¹ emission factor for 2009 (Fraser *et al.*, 2013)

¹² assume = 2009 emissions

¹³ assume = 2011 emissions

¹⁴ TAPM or ISC only

The Australian emissions derived from atmospheric data prior to 2006, using TAPM or NAME (Vic/Tas) are likely to be underestimated due to the very significant PFC-14 emissions from a single pot-line (#1) at Kurri Kurri, which accounted for nearly half of all Australian PFC emissions from aluminium smelting in 2005. The pot-line was upgraded in 2006 and this accounts for the dramatic drop in PFC-14 emissions from 2005 to 2006 in the *National Inventory Report*. The large PFC emissions prior to 2006 are not seen in the TAPM estimates of emissions, as they are based on Bell Bay/Pt Henry/Portland data. The NAME inversion, based on the Vic/Tas/NSW domain, uses all pollution episode data, including any Kurri Kurri- or Tomago-affected data that may have impacted on Cape Grim (the Kurri Kurri and Tomago smelters are located in the Hunter Valley, NSW). This is why the Australian PFC-14 estimates in 2005 (182 tonnes) based on the NAME (Vic/Tas/NSW) emissions are significantly greater than the TAPM estimates for 2005 (95 tonnes).

The average Australian PFC-14 emissions for 2005-2011 based on atmospheric data are similar: TAPM - 78 tonnes and NAME (Vic/Tas/NSW) - 85 tonnes, about 13% higher than reported in the *National Inventory Report 2012* (DoE, 2014; 72 tonnes). The average Australian 2008-2011 PFC-14 emissions derived from atmospheric data using NAME (Vic/Tas/NSW) are 47 tonnes, 20% higher than in the *National Inventory Report 2011* (39 tonnes). The TAPM average for the same period is 69 tonnes and 66 tonnes for 2008-2012. Arguably the NAME estimate is more accurate as it is based on 72% of Australian aluminium production (Portland, Pt Henry, Bell Bay, Kurri Kurri, Tomago), whereas the TAPM estimate is based on 35% of Australian aluminium production (Portland, Pt Henry, Bell Bay). These SE Australian smelters could have higher average PFC-14 emission factors than the Australian average (see below).

The overall agreement between the PFC emission factors in the *National Inventory Report 2013 (DE, 2015)* and as derived from atmospheric data using TAPM/NAME over the period 2005-2011 is good (Table 4, Figure 5, TAPM: 41 g CF₄/tonne aluminium; NAME: 44 g/tonne; NGA: 37 g/tonne), with uncertainties overlapping throughout the record. For the period 2005-2013 the TAPM average CF₄ emission factor is 38 g/tonne compared to the NGA: 31 g/tonne.

The atmospheric data (NAME) show a factor of 3.5 decline in the emission factors over the period 2005-2011, whereas the NGA show a factor of 6.2 decline over the same period. This decline is dominated by the influence that Kurri Kurri had on national average emissions factors between 2006 and 2007; this decline is not captured in the TAPM data which do not 'see' Kurri Kurri emissions. Given the overlapping uncertainties on both NAME and the NGA estimates of PFC-14 emissions, this difference in emissions decline over the period 2005-2011 may not be significant. However, the difference could be due, in part, to a larger than average decline in the emission factor at Boyne Island, which is not seen in the atmospheric data at Cape Grim, by either NAME or TAPM.

In 2013, Australian PFC-14 emissions (TAPM, 57 tonnes) were 0.5% of global emissions (11.3 k tonnes). The only significant PFC-14 source in Australia is aluminium production, whereas globally both aluminium production and the electronics industry are significant sources of PFC-14 emissions.

5.2 Other PFC emissions

Australian PFC emissions are dominated by the aluminium industry – only aluminium production-derived emissions of PFC-14 and PFC-116 are currently listed in the *National Inventory Report* (Table 3). PFC imports in 2013 are small (Table 2 - PFC-14: 0.02 tonne, PFC-116: 0.05 tonne, PFC-318: 0.03 tonne). These imports are presumably for minor refrigeration and fire-fighting applications, and the corresponding emissions from these non-aluminium sources are likely to be well below 1 tonne/yr.

Figure 7 shows the Australian PFC-116 emissions from the *National inventory Report*, from PFC-14 emissions (TAPM/NAME) assuming a PFC-116/PFC-14 mass emission factor of 0.10±0.01, based on atmospheric observation at Aspendale on the Pt Henry/Portland PFC plumes and on direct measurements on the Kurri Kurri exhaust stack (Fraser *et al.*, 2013; Kim *et al.*, 2014). The overall agreement is good as expected, as the emissions are from the aluminium industry only, with current emissions (since 2009) of about 6 tonnes/yr. However the PFC-116 emissions for Australia based on measurements at Cape Grim, again assuming coming from the aluminium industry only, show significant differences in emissions after 2007, with NAME PFC-116 emissions rising to approaching 30 tonnes in 2011.

Since Cape Grim largely 'sees' PFC emissions from the Victorian smelters and any other emissions from the Melbourne/Port Phillip region, the data would suggest that a significant, post -2007, non-aluminium PFC-116 source is in operation, and this PFC-116 is not captured in the import data. Australia-wide, this source could be currently approaching 20 tonnes/yr. The source of this additional PFC-116 is unknown. Further analysis of past and future Aspendale and Sydney PFC-116 data could help identify this source.

In 2011, Australian PFC-116 emissions (NAME, 29 tonnes) were 1.5% of global emissions (1.9 k tonnes).



Figure 7. Australian PFC-116, PFC-218 and PFC-318 emissions from the National Inventory Report 2013 (DoE, 2015) and from atmospheric measurements at Cape Grim using NAME, TAPM and ISC modelling; error bars are ± sd. Australian PFC-116 (NAME) are from Vic/Tas data scaled by aluminium production, PFC-116 (TAPM/NAME PFC-14) are from Australian TAPM/NAME PFC-14 emissions (Table 4) and the observed PFC-116/PFC-14 ratio (0.10±0.01) in Australian smelter plumes; PFC-218 and PFC-318 are from NAME and ISC modelling, with Australian emissions obtained from regional emissions by population scaling.

Cape Grim data also suggest that there are significant Australian PFC-218 and PFC-318 emissions, the former increasing from about 10 tonnes in the mid-2000s to nearly 20 tonnes in 2011, declining to 9 tonnes in 2013 and the latter declining from nearly 30 tonnes in 2009 to 15 tonnes in 2013. The sources of PFC-218 and PFC-318 are unknown; there are no significant PFC-218 or PFC-318 imports recorded. The PFC-218 emissions are about the same size as the non-aluminium PFC-116 emissions, with a similar growth pattern. This suggests their emissions may be related.

In 2011, Australian PFC-218 emissions (NAME, 20 tonnes) were 4% of global emissions (0.48 k tonnes).

Total PFC emissions have declined from about 190 tonnes in 2005 (80% from aluminium production) to about 100 tonnes (60% from aluminium production) in 2010, declining even further to 87 tonnes in 2013 (55% from the aluminium industry), compared to the *Inventory*, which has total PFCs (PFC-14, PFC-116) declining from 226 tonnes in 2005 to 24 tonnes in 2013 (all from the aluminium industry). In CO₂-e terms, total PFCs have declined from 1.5 Mt CO₂-e in 2005 to 0.7 Mt CO₂-e in 2013, compared to the *Inventory* figures of 1.8 Mt CO₂-e in 2005 falling to 0.19 Mt CO₂-e in 2013. This is because the *Inventory* only includes PFCs from the aluminium industry, whereas Cape Grim sees PFC emissions from all potential sources (aluminium, refrigeration, fire fighting and possibly other sources).

5.3 HFC, sulfur hexafluoride and sulfuryl fluoride emissions

CSIRO estimates emissions of a number of trace gases from the Melbourne/Port Phillip region (Dunse *et al.*, 2001, 2005; Dunse 2002; Fraser *et al.*, 2014), utilising *in situ* high frequency measurements from the Cape Grim Baseline Air Pollution Station in Tasmania and employing the interspecies correlation (ISC) technique with co-incident carbon monoxide (CO) measurements.

Port Phillip HFC, PFC-116 and sulfur hexafluoride emissions have been calculated, using ISC and Cape Grim data (2004-2013), obtained from the GC-MS-Medusa and GC-ECD instruments (Krummel *et al.*, 2011; Miller *et al.*, 2008) at Cape Grim, and presented as 3-year running averages (2005-2013; Table 5, Figure 8). The HFC, PFC-116 and sulfur hexafluoride emissions are derived from Port Phillip emissions, scaled to Australian emissions on a population basis (using a population-based scale factor of 5.4). NOAA air mass back trajectory analyses (Draxler & Hess, 1997) are used to ensure that the pollution events at Cape Grim used to derive Port Phillip emissions are imbedded in air masses that only pass over the Port Phillip region and do not include other possible high carbon monoxide source regions, in particular the Latrobe Valley, or high carbon monoxide emission events (biomass burning).

For HFC emission calculations, the NAME particle dispersion model (Manning *et al.*, 2003, 2011) 'sees' emissions from Victoria/Tasmania (the domain used in the model) and are presented as 3-yr running averages of emissions (i.e. 2008 annual emissions are derived from 2007-2009 data). The Australian emissions are calculated from Victoria/Tasmania using a population based scale factor of 3.7 and are shown in Table 5 and Figure 8.

Table 5. Australian HFC, PFC, sulfur hexafluoride and sulfuryl fluoride emissions (tonnes, 2005-2013) from atmospheric data, collected at Cape Grim, Tasmania - emissions calculated by interspecies correlation (ISC) and from inversions using the UK Met. Office NAME particle dispersion model. The emissions are 3-year running averages, i.e. '2010' = average of 2009, 2010, 2011 emissions. Australian HFC and sulfur hexafluoride emissions are scaled from regional emissions by population; GWPs (to calculate CO₂-e emissions) are from the *National Inventory Report 2013* (DoE, 2015). PFC-14 emissions are the TAPM/NAME average from Table 4. Australian PFC-116 emissions are from NAME (Vic/Tas) scaled by aluminium production; Australian PFC-218 and PFC-318 emissions are from NAME and ISC scaled by population; Australian sulfuryl fluoride emissions are from ISC and NAME estimates of SE Australian emissions scaled by grain production.

	2005		2006		2007		2008		2009		2010		2011		2012	2013
Refrigerant	ISC	NAME	ISC	NAME	ISC	NAME	ISC	NAME	ISC	NAME	ISC	NAME	ISC	NAME	ISC	ISC
	tonnes		tonnes		tonnes		ton	tonnes		tonnes		nes	tonnes		tonnes	tonnes
HFC-32	36±12	34±5	62±23	40±10	116±48	61±15	129±49	96±20	170±60	134±20	196±67	162±20	205±66	172±10	211±69	186±60
HFC-125	392±130	311±15	375±125	326±20	460±174	402±60	516±183	553±80	642±207	685±60	642±213	738±45	614±195	753±25	598±194	603±195
HFC-134a	1686±519	1613±90	1707±530	1668±110	1832±621	1830±150	1843±596	1997±165	2148±666	2279±120	2141±700	2262±120	2111±659	2169±140	2008±643	1672±526
HFC-143a	454±158	337±20	436±150	352±30	481±174	451±65	503±172	550±60	624±200	687±55	597±198	750±60	505±165	771±35	555±185	530±176
HFC-23	50	34	50	34	50±28	34	56±23	34±3	58±22	43±4	44±17	44±6	41±16	55±5	46±16	46±15
HFC-152a	32±11	31±2	38±13	35±3	51±17	41±3	49±17	43±3	52±18	68±4	59±21	69±5	56±19	72±4	77±28	69±24
HFC-227ea	15±9	15	12±6	12	17±7	17	22±8	22	36±13	36	33±12	33	26±9	26	19±6	38±14
HFC-236fa	13±10	13	5±3	5	5±3	5	4±2	4	5±2	5	6±2	6	6±2	6	5±2	4±1
HFC-245fa	27±15	27	30±13	30	32±13	32	28±11	28	32±13	32	38±14	38	49±18	49	117±43	109±39
HFC-365mfc	8±4	13	25±11	13±1	33±13	15±2	50±18	16±3	61±21	47±13	74±26	64±10	69±24	68±3	66±23	59±20
total HFCs	2714±560	2428±95	2741±565	2515±115	3078±670	2888±175	3198±650	3343±195	3828±730	4016±145	3830±760	4166±145	3682±710	4139±145	3702±700	3317±595
total HFCs NAME/ISC avg (t)	2571±	325	2628±340		2983±425		3271±420		3922±435		3998±450		3911±430		3702±700	3317±595
total HFCs (kt CO ₂ -e)	6221	650	6197±660		7057±965		7884±905		9515±950		9612±955		9393±835		8556±1435	7999±1305
SF ₆ (tonnes)	78±26	29	75±25	29±2	44±16	31±3	33±12	34±2	34±13	44±3	37±14	43±3	33±12	44±2	31±11	27±9
SF ₆ NAME/ISC avg (t)	53±	26	52±13		37±9		33±7		39±8		40	±8	38±7		31±11	27±9
SF ₆ (kt CO ₂ -e)	1179 1	298	1179±298		848±211		762±166		889±184		916±186		877±160		704±240	605±201
SO ₂ F ₂ (tonnes)		2±2		2±2		4±4		17±15	18±8	3±5	68±26	14±18	80±32	88±21	71±28	68±26
SO ₂ F ₂ NAME/ISC avg (t)	2±	2	2:	2	41	<u>-</u> 4	17±	±15	11	±6	41	22	84:	±26	71±28	68±26
SO ₂ F ₂ (kt CO ₂ -e)	9±	9	10:	±10	18:	±19	86±	±74	54:	±32	204	110	419:	±132	357±141	341±131
total PFCs (t, Table 4)	187±	45	173	±50	118	±40	104	±30	104	±30	103	±30	105	±30	103±25	87±20
total PFCs (kt CO ₂ -e)	1538	377	1430	±433	1001	±371	888±	£249	895	£438	881±	356	890	±350	862±291	725±221
total HFCs, PFCs, SF ₆ avg	2809	330	2853	±345	3138	±425	3408	±420	4065	±435	4141	±455	4054	±430	3835±700	3430±595
total HFCs, PFCs, SF ₆ avg	8938±	±780	8806	±810	8906	±1035	9535	±940	11299	±1005	11409	±1000	11160	0±880	10123±1465	9329±1330
(k tonnes CO ₂ -e)																



Figure 8. Australian HFC-32, -125, -134a, -143a, -152a, -365mfc and sulfur hexafluoride emissions (NGA: DoE 2015) compared to emissions calculated from Cape Grim data by interspecies correlation (ISC) and from the UK Met. Office NAME particle dispersion model. In the ISC calculations of HFC and sulfur hexafluoride emissions, Australian emissions are scaled from Melbourne/Port Phillip emissions on a population basis (5.4, Australia/Port Phillip); in the NAME calculations, Australian emissions are scaled from Victorian/Tasmanian emissions, also on a population basis (a scale factor of 3.7). Also shown are Australian sulfuryl fluoride emissions from ISC and NAME, scaled on a grain production basis.

Cape Grim 'sees' sulfuryl fluoride emissions from SE Australia. Cape Grim sulfuryl fluoride measurements commenced in 2004, but significant pollution episodes (as selected for ISC calculations) were not observed at Cape Grim until 2010. These pollution episodes are used to calculate SE Australian (largely Victorian) sulfuryl fluoride emissions by ISC. Grain exported from Victorian grain terminals, or produced in Victoria, accounts for about 15-20% of Australia's grain production/exports (National Transport Commission, 2008) and thus possibly 15-20% of Australia's emissions of sulfuryl fluoride. SE Australian sulfuryl fluoride emissions calculated from Cape Grim data by ISC are scaled to Australian emissions by this factor (i.e. multiplied by 6.2 ± 0.6) (Table 5, Figure 8). The NAME model domain used to derive sulfuryl fluoride emissions is Victoria/Tasmania/New South Wales, and the scale factor used to derive Australian emissions, based on grain production, is 2.1 ± 0.2 . Australian emissions of sulfuryl fluoride averaged about 74 tonnes/yr (372 k tonnes CO₂-e) over the period 2011-2013, some 2%-3% of global emissions (see above), similar to Australia's fraction of global grain production (~2%). Data on Australian sulfuryl fluoride imports are not available.

6 Comparisons of NGA, ISC & NAME emission estimates

6.1 HFC-134a

The major HFC in the *National Inventory* (ageis.climatechange.gov.au; DoE 2015) is HFC-134a, with emissions in 2013 of 3487 tonnes (Table 3). Based on Cape Grim data, Australian emissions of HFC-134a in 2013 were 1672 tonnes (ISC), ~52% lower than reported in the *Inventory* (Table 5, Figure 8). For the 9-year period 2005-2013, the average Australian emissions of HFC-134a calculated from Cape Grim data (ISC/NAME) are about 26% lower than in the *Inventory*. From 2002 to 2011, the ISC and NAME emissions estimates agree to within 2% (NAME higher). For the period 1999-2004, Australian emissions based on Cape Grim data are higher than the inventory, and, for 2006-2013, lower than the inventory. As discussed in Section 4 above, the *Inventory* best represents total HFC emissions, not individual HFC emissions.

Cape Grim data suggest that Australian emissions of HFC-134a have been in decline since 2009, whereas the *Inventory* reports growing emissions through 2013 (Figure 8), as are global emissions (see above). The Inventory assumes time-invariant emission factors, whereas the Cape Grim data may be seeing reduced emissions due to, for example, improved equipment maintenance practices, with better management of HFC emissions. Based on atmospheric data, global emissions of HFC-134a were 174 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 1.1% of global emissions based on ISC/NAME data, and 1.8% based on *Inventory* data.

USA HFC-134a emissions as reported to UNFCCC are in decline, having peaked in 2010 at 69 k tonnes; however an analysis of air masses impacted by USA emissions arriving on the west coast of Ireland (Mace Head) suggest USA HFC-134a emissions continue to increase, reaching 70 k tonnes in 2012, increasing from 62 k tonnes in 2010. The USA HFC-134a emissions exhibit seasonal behaviour, with annual emissions about 20% higher than winter emissions, attributed to seasonal automobile ac emissions (Simmonds *et al.*, 2015).

Cape Grim 'sees' largely winter emissions of HFC-134a emissions from the Melbourne/Port Phillip region; this could offer a partial explanation of the large difference between Australian emissions of HFC-134a calculated from Cape Grim data and as reported in the *Inventory*. This is being investigated further.

6.2 HFC-125

The next major HFC emitted into the Australian environment is HFC-125 with current (2013) emissions estimated to be about 1226 tonnes in the *Inventory*. Based on Cape Grim data, Australian emissions of HFC-125 in 2013 were 603 tonnes (ISC), about 50% lower than the *Inventory* data. For the period 2005-2013, the *Inventory* average emissions are 60% higher than emission estimates based on Cape Grim data (ISC/NAME). Over the period 2002-2011, NAME and ISC emission estimates agree to within 2% (NAME lower). ISC/NAME estimates of HFC-125 emissions have grown from 350 tonnes in 2005 to 600 tonnes in 2013. Like HFC-134a, the Cape Grim HFC-125 data suggest near constant or perhaps declining emissions, whereas in the *Inventory* the emissions continue to grow through 2013 (Figure 8). Based on atmospheric data, global emissions based on ISC/NAME data, and 2.6% based on *Inventory* data.

6.3 HFC-143a

According to the *Inventory* (Table 3), the next major HFC emitted into the Australian environment is HFC-143a with current (2013) emissions at about 117 tonnes. Based on Cape Grim data, Australian emissions of HFC-143a in 2013 were about 530 tonnes (ISC), almost a factor of 5 higher than *Inventory* estimates. NAME and ISC data agree to within 1% for the period 2005-2011 (NAME higher). Like HFC-134a and HFC-125, the Cape Grim HFC-143a data suggest near constant or perhaps declining emissions, whereas in the *Inventory* the emissions continue to grow through 2013 (Figure 8). Based on atmospheric data, global emissions of HFC-143a were 23 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 2.4% of global emissions based on ISC/NAME data, and 0.5% based on *Inventory* data. The Australian HFC-143a emissions based on Cape Grim data seem unusually high.

Simmonds et al. (2015b) report growing USA HFC-143a emissions through 2012 (8.5 k tonnes), with no obvious seasonality.

6.4 HFC-32

HFC-32 emission estimates are estimated in the Inventory to be 68 tonnes in 2013. The emissions based on Cape Grim data (186 tonnes in 2013) are significantly higher than in the *Inventory*. Over the period 2006-2011, ISC and NAME emission estimates agree to within 36% (NAME lower). Unlike HFC-134a, HFC-125 and HFC-143a, the Cape Grim data and the *Inventory* both suggest increasing emissions of HFC-32 through 2012, then in 2013 ISC estimates suggest a decline (Figure 8). Based on atmospheric data, global emissions of HFC-32 were 20 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 1.1% of global emissions based on ISC/NAME data, and 0.3% based on *Inventory* data.

Simmonds *et al.* (2015b) report that USA HFC-32 emissions have stabilized through 2011-2012 (5-6 k tonnes), following strong growth in emissions during 2005-2010; there is no obvious seasonality in USA HFC-32 emissions.

6.5 HFC-152a

HFC-152a emissions are not listed in the *Inventory*. The 2013 emissions have been estimated from Cape Grim data at 69 tonnes (ISC). Over the period 2002-2011, ISC and NAME estimates of Australian emissions agree to within 1% (NAME higher). Imports of HFC-152a into Australia were 13 tonnes in 2013 and 21 tonnes in 2014. Like HFC-32, the Cape Grim data show that HFC-152a emissions continue to grow through 2012, then decline in 2013.

Simmonds *et al.* (2015b) report that USA HFC-152a emissions have grown through 2012 (52-53 k tonnes); there is a suggestion of seasonality in USA HFC-152a emissions, with summer emissions lower than winter; however derivation of possible seasonality in the HFC-152a source is uncertain because HFC-152a is relatively short-lived in the atmosphere and there is a strong summer sink for HFC-152a.

Based on atmospheric data, global emissions of HFC-152a were 54 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 0.1% of global emissions based on ISC/NAME data. It would be unusual for Australian emissions of an industrial chemical to be less than 0.1% of global emissions. The possible reasons for the low Australian emission (low use in Australia compared to rest of world) are being investigated. One suggestion (M. Bennett, RRA, personal communication, 2011) is that a significant major-volume use in other parts of the world for HFC-152a is as an aerosol propellant, a use not taken up in Australia.

6.6 HFC-23

HFC-23 emission estimates are estimated in the Inventory to be 0.4 tonnes in 2012 and 2013. In the 1990s, Australian emissions were estimated as fugitive emissions from Sydney-based Australian HCFC-22 production. HCFC-22 production ceased in Australia in 1995. Imports are currently recorded at about 1 tonne in 2012, 0.1 tonnes in 2013 and 0.31 in 2014. The 2012 and 2013 emissions have been estimated from Cape Grim data at 46 tonnes (ISC). Over the period 2002-2011, ISC and NAME estimates of Australian emissions agree to within 18% (NAME lower). HFC-23 emissions have remained relatively constant since 2007. In CO₂-e terms, HFC-23 is currently the 4th largest HFC emitted in Australia (after HFC-134a, HFC-143a and HFC-125), larger than HFC-32. This is because the GWP (Global Warming Potential) of HFC-23, at 14,800, is about 3.3 times larger than the next most potent HFC (HFC-143a, GWP 4470). Based on atmospheric data, global emissions of HFC-23 were 12 k tonnes in 2012 (Rigby *et al.*, 2014). Current Australian emissions are about 0.4% of global emissions based on ISC/NAME data, 0% based on the Inventory.

The origin of these Australian HFC-23 emissions is unknown. HFC-23 is a component of the R-508 series of refrigerants, but significant imports of R-508 refrigerants into Australia have not been recorded. The other component of the R-508

refrigerants is PFC-116; current Australian emissions of PFC-116 are significantly larger than result from aluminium production and current imports. If significant amounts of R-508 refrigerants have been imported in the past into Australia, but not accounted for, then emissions of R-508 could explain these significant HFC-23 and PFC-116 emissions.

RRA (M. Bennett, private communication, 2014) is not aware of any significant historical use of R-508 refrigerants in Australia. RRA in their refrigerant reclaim operation have collected only one cylinder of R-508 from a CSIRO instrument testing laboratory in Sydney. This it is unlikely that the unaccounted for HFC-23 and PFC-116 is from past or current use of R-508 refrigerants in Australia.

Aspendale atmospheric data on HFC-23 (unpublished) and PFC-116 (Kim *et al.*, 2014) show very different concentration/wind direction (so-called concentration 'roses') at Aspendale. The PFC-116 data show a clear, strong maximum in the direction (W of Aspendale) of the Pt Henry and Portland smelters, confirming a largely aluminium-production source, and essentially noise in the other wind sectors. The HFC-23 data show a significant, but small concentration maximum in the direction NE of Aspendale (as do HFC-134a, HFC-32, HFC-125 etc). It would appear that HFC-23 is in the general mix of refrigerant emissions from Melbourne, but has not been identified, in any significant amounts, in any Australian refrigerant imports.

One possible explanation is that HFC-23 (CHF_3) is present in refrigerant blends as a contaminant – for example with blends containing HFC-32 (CH_2F_2). It is possible that overfluorination during the production of HFC-32 could produce HFC-23 and that the resultant refrigerant blends using HFC-32 could contain, likely small, amounts of HFC-23. CSIRO plan to test this hypothesis in a project designed to test the stated composition of the major refrigerants and refrigerant blends used in Australia, which are HFC-134a, R-410 (50% HFC-32, 50% HFC-125), R-404 (44% HFC-125, 4% HFC-134a, 52% HFC-143a) and R-407 (20% HFC-32, 40% HFC-125, 40% HFC-134a).

6.7 HFC-227ea

HFC-227ea emissions are now listed in the *Inventory*, with 2013 emissions estimated at 42 tonnes. The 2013 emissions have been estimated from Cape Grim data at 38 tonnes and 19 tonnes in 2012 (ISC). The *Inventory* average emissions (2005-2013) are 31 tonnes compared to 24 tonnes from Cape Grim data. There are no NAME estimates of HFC-227ea emissions from Cape Grim data. The Inventory suggests that HFC-227ea emissions continue to grow through 2013, whereas the Cape Grim data suggest that emissions peaked at 36 tonnes in 2009.

Based on atmospheric data, global emissions of HFC-227ea were 3.3 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 0.6% of global emissions based on ISC data and 1.2% based on *Inventory* data.

6.8 HFC-236fa, HFC-245fa, HFC-365mfc

The 2013 emissions for HFC-236fa, HFC-245fa and HFC-365mfc have been estimated (ISC) from Cape Grim data at about 4 (steady), 109 (growing until 2012) and 59 tonnes (peaked in 2010 at 74 tonnes) respectively. NAME estimates of HFC-365mfc emissions are available (2005-2011). From 2009-2011, the ISC and NAME estimates agree to within 15%, prior to 2009 NAME estimates of emissions are significantly higher than ISC by a factor of 2-3. There are no estimates of emissions of these HFCs in the *Inventory*.

Based on atmospheric data, global emissions of HFC-236fa, HFC-245fa and HFC-365mfc were 0.2, 8.2 and 3.3 k tonnes respectively in 2012 (Rigby *et al.*, 2014). Australian emissions are about 2.1%, 1.4% and 2.0% of global emissions based on Cape Grim data.

6.9 HFC-134 & HFC-4310mee

Emissions of HFC-134 and HFC-4310mee in the *Inventory* were about 1 tonne each in 2013. HFC-134 is not measured at Cape Grim and, although HFC-4310meee is measured, no significant Australian emissions have been detected to date. Australian emissions as low as 1 tonne/yr are unlikely to be detected in the Cape Grim data, as possible pollution episodes would be unable to be resolved from instrument noise.

6.10 Total HFCs

Total HFC emissions (Table 5), based on Cape Grim observations, have grown from over 2500 tonnes in 2005 to 4000 tonnes in 2010, declining to about 3300 tonnes in 2013. The latter estimate is from ISC data only. Total HFC emissions in 2013 are 33% lower than in the *Inventory*, due largely to the 52% lower emissions of HFC-134a emissions from atmospheric data compared to *Inventory* data. Over the period 2005-2013, total HFC emissions in the *Inventory* are in good agreement (within 8%) of total emissions based on Cape Grim data (ISC, NAME lower).

Total HFC emissions in the *Inventory* in 2013 are 9.97 Mt CO_2 -e, higher (around 25%) than emissions (7.99±1.3 Mt CO_2 -e, ISC) based on Cape Grim data (Table 3, Table 5, Figure 9). Over the period 2005-2013, the total HFC emissions in the *Inventory* are 67 Mt CO_2 -e, compared to 72 Mt CO_2 -e (8% higher) from Cape Grim data (ISC/NAME).



Figure 9. Australian emissions of HFCs - 125, -134a, -143a, -32) and other HFCs (-23, -152a, -227ea, -236fa, -365mfc) estimated from atmospheric data (ISC/NAME) measured at Cape Grim, and in the *Inventory* (DoE, 2015), expressed in units of M tonne CO₂-e.

6.11 Sulfur hexafluoride

Sulfur hexafluoride emissions in the *Inventory* are about 6 tonnes/yr in 2013 (Table 3, Figures 8, 10). Estimates using Cape Grim data (ISC/NAME: Table 5, Figure 10) are 52 tonnes in 2006, decreasing to 27 tonnes in 2013, a decrease of about 7%/yr. NAME estimates of sulfur hexafluoride emissions are about 20% lower than ISC over the period 2006-2011, but 10% higher over the period 2007-2011. In CO₂-e terms sulfur hexafluoride emissions have fallen by 0.6 Mt CO_2 -e, from 1.18 Mt CO_2 -e in 2006 to 0.61 Mt CO_2 -e in 2013 based on Cape Grim data, whereas in the *Inventory* sulfur hexafluoride emissions have fallen by only 0.04 Mt CO_2 -e, from 0.17 to 0.13 Mt CO_2 -e over the same period.

A recent inversion study, using a combined Eulerian chemical transport model and a Lagrangian particle dispersion model (CTM-LPDM) has been used to derive global and regional (including Australian) emissions (Rigby *et al.*, 2011). Using Cape Grim, the CTM-LPDM calculated emissions from Tasmania, Victoria and southern NSW (including Sydney). Scaling the results to Australian emissions, based on the fraction of the EDGAR prior estimate that is emitted into the prescribed domain (64%), results in average Australian emissions (2007-2009) of 16±10 tonnes. This study was repeated (Ganesan *et al.*, 2014), with an improved estimate of emissions and their uncertainties, resulting in Australian emissions in 2012 of 17±9 tonnes (with an expanded prior region, including Adelaide, containing 85% of Australian emissions). The combined ISC/NAME data over this period suggest emissions of 37 tonnes, whereas the *Inventory* reports 6-7 tonnes/yr over this period. In these studies, Sydney, the likely largest sulfur hexafluoride source region in Australia, is on the edge of the chosen model domain, and this may introduce potential errors into the inversion. If the inversion is not accurately capturing (underestimating) the Sydney emissions, as seen in the Cape Grim data, then the scaling factor may be too low, resulting in an underestimate of Australian emissions.

Based on atmospheric data, global emissions of sulfur hexafluoride were 8.1 k tonnes in 2012 (Rigby *et al.*, 2014). Australian emissions are about 0.4% of global emissions based on ISC/NAME data, but < 0.1% based on the *Inventory* data. It would be unusual for Australian emissions of a widely-used industrial chemical to be <0.1% of global emissions.

The Australian sulfur hexafluoride emissions from atmospheric data and in the *Inventory*, and sulfur hexafluoride import data, are shown in Figure 10. In the 2016 report, Australian sulfur hexafluoride emissions will be recalculated using IPCC default emission factors (see discussion in Section 4).

Ultimately, accurate import data will approximately match the sum of emissions and a growth in the banked sulfur hexafluoride, which have grown over the past 5 years by about 5 tonnes/yr. The average imports for 2013-2014 are 16 tonnes/yr.



Figure 10. Australian sulfur hexafluoride imports and emissions (tonnes) from Cape Grim data using ISC and NAME (Ganesan *et al.*, 2014; Rigby *et al.*, 2014) and from the *Inventory* (NGA: DoE, 2015).

6.12 Sulfuryl fluoride

Based on Cape Grim data, Australian sulfuryl fluoride emissions averaged less than 10 tonnes/yr from 2005-2007, but then increased rapidly to average about 70-80 tonnes/yr from 2011-2013 (Table 5, Figure 11). Presumably this reflects a change in grain fumigation practices away from using methyl bromide and phosphine. Global emissions were 3000 tonnes/year (2011-2012, Rigby *et al.*, 2014). Australian emissions are 2-3% of global emissions. Australia is responsible for 3% of global wheat production, but 10-15% of wheat exports. Sulfuryl fluoride is more closely related to wheat production (grain storage in Australia's interior) than to wheat export (grain storage at export ports, fumigated with methyl bromide).

In CO_2 -e terms, sulfuryl fluoride emissions averaged about 350 k tonnes CO_2 -e in 2012-2013, compared to 650 k tonnes CO_2 -e for sulfur hexafluoride and 790 k tonnes CO_2 -e for PFCs; HFCs averaged about 8300 k tonnes CO_2 -e in 2012-2013.

In climate change terms, Australian sulfuryl fluoride emissions are now about 50% of the radiative forcing of PFCs, and possibly growing quite rapidly. Clearly there needs to be a close watch on future sulfuryl fluoride emissions in relation to their contribution to radiative forcing due to SGGs.





6.13 Total HFCs, PFC and sulfur hexafluoride emissions

Total HFC, PFC and sulfur hexafluoride emissions in CO₂-e are shown in Table 5 and Figure 12.

Total Australian HFC, PFC and sulfur hexafluoride emissions in the *Inventory* are 5.0 k tonnes in 2013, higher (38%) than estimates based on Cape Grim data: 3.4±0.6 k tonnes (ISC). Over the period 2005-2013 the *Inventory* estimates are 7% higher than estimates based on Cape Grim data (ISC/NAME).

Total Australian HFC, sulfur hexafluoride and PFC emissions, expressed as CO_2 -e, in the *Inventory* are 10.3 Mt CO_2 -e in 2013, 10% higher than estimates based on Cape Grim data: 9.3±1.3 Mt CO_2 -e (ISC) (Figure 12). Over the period 2005-2013, total Australian emissions of HFCs, PFCs and sulfur hexafluoride, based on Cape Grim data (90 Mt CO_2 -e) are

about 20% higher than the same emissions in the *Inventory* (73 Mt CO_2 -e). In two categories (PFCs and sulfur hexafluoride) the atmospheric data give higher estimates of emissions than in the *Inventory*. In the HFC category the atmospheric data give lower estimates of emissions than the *Inventory*.

Australian total HFC, PFC and sulphur hexafluoride emissions in 2013 (3430 tonnes) are 0.9 % of global emissions (390 k tonnes).



Figure 12. Australian HFC, PFC, sulfur hexafluoride emissions calculated from Cape Grim observations and in the *Inventory* (NGGI/NGA, ageis.climatechange.gov.au) in M tonne CO₂-e.

Summary

- CSIRO, through involvement in the international AGAGE (Advanced Global Atmospheric Gases Experiment) program, has now measured and reported atmospheric concentrations, temporal trends and emissions for eleven HFCs (HFC-23, -32, -125, -134a, -143a, -152a, -227ea, -236fa, -245fa, -365mfc, -4310meee), nine PFCs (PFC-14, -116, -218, -318, -3110, -4112, -5114, -6116, -7118), sulfur hexafluoride, nitrogen trifluoride, trifluoromethyl sulfur pentafluoride and sulfuryl fluoride, utilizing data from the background atmosphere at Cape Grim, Tasmania. The AGAGE program involves collaborating scientists from the USA, Australia, China, Italy, Korea, Norway, Switzerland and the UK, who operate a global measurement network and, through a variety of modelling initiatives, estimate lifetimes and emissions of all the significant non-CO₂ GHGs, from both ozone depletion and climate forcing perspectives.
- All HFCs (except HFC-152a), all PFCs, sulfur hexafluoride, sulfuryl fluoride, but not trifluoromethyl sulfur pentafluoride, show increasing concentrations in the background atmosphere at Cape Grim, which accurately reflects global background atmospheric changes. Significant increases in 2013 (ppt/yr) are seen in HFC-134a (4.9), HFC-125 (2.0), HFC-143a (1.4), HFC-32 (1.3), HFC-23 (1.0), PFC-14 (0.7), sulfur hexafluoride (0.3) and sulfuryl fluoride (0.12). Total HFCs are growing at 10.9 ppt/yr or 8%/yr, total anthropogenic PFCs at 0.84 ppt/yr (1.6%/yr), sulfur hexafluoride at 3.9%/yr and sulfuryl fluoride at 6.5%/yr.
- Total global HFC, PFC, sulphur hexafluoride and nitrogen trifluoride emissions have risen from about 30 k tonnes per year in the late 1970s to over 420 k tonnes per year in 2014.
- Import data for 2014 show significant use for most of these HFCs and sulfur hexafluoride in Australia. HFC imports in 2014 are approximately 25% higher than in 2013, whereas sulfur hexafluoride imports are nearly 30% lower. Less than 1 tonne of PFCs are imported into Australia. There are no import data available for sulfuryl fluoride or nitrogen trifluoride.
- The National Greenhouse Accounts (Inventory) reports significant Australian annual (2013) emissions of HFC-134a (3487 tonne), HFC-125 (1219 tonne), HFC-143a (117 tonne), HFC-32 (67 tonne), HFC-227 ea (41 tonne), PFC-14 (22 tonne), sulfur hexafluoride (6 tonne) and PFC-116 (2 tonne). Emissions of other HFCs are about or less than 1 tonne each (HFC-23, HFC-134, HFC-4310mee). Emissions of all HFCs are increasing. The Inventory emissions calculations are constructed to best estimate total HFC emissions in CO₂-e terms, and are not necessarily representative of the emissions of individual HFCs. Emissions of PFCs (from aluminium production only) in the Inventory are declining and sulfur hexafluoride emissions have remained steady over the last 3 years. Overall the emissions of HFCs, PFCs and sulfur hexafluoride are increasing due to the dominant influence of HFC emissions.
- Regional Australian emissions have been detected in the Cape Grim data for all HFCs (except HFC-4310mee), the first four PFCs listed above, sulfur hexafluoride and sulfuryl fluoride. The emissions have been quantified using a variety of modelling techniques, including interspecies correlation (ISC), forward modelling using the CSIRO TAPM model and inverse modelling using the UK Meteorological Office model NAME. SE Australian emissions of HFC-4310mee are expected to be seen at Cape Grim over the next few years. ISC and NAME estimates for HFC, PFC and sulfur hexafluoride emissions agree, on average over the period 2005-2011, to within 2% for HFC-125, HFC-134a, HFC-143a and HFC-152a, within 15% for HFC-23, HFC-365mfc and sulfur hexafluoride, within 35% for HFC-32, within 50% for sulfuryl fluoride.
- Comparisons of emissions of individual HFCs (*Inventory* v. atmospheric data) are not likely to be meaningful, but comparisons of total HFCs could be informative. Over the period 2005-2013, total HFC emissions in the *Inventory* are about 9% higher than total HFC emissions estimated from Cape Grim data (NAME/ISC). In 2013 the *Inventory* reports 4935 tonnes emitted, compared to 3317 tonnes estimated from Cape Grim data. Unlike the *Inventory*, which shows Australian total HFC emissions continuing to grow, Cape Grim data suggest that total HFC emissions peaked in 2010 at close to 4000 tonnes, with 2013 emissions 19% below the peak.
- Over the period 2005-2013, total HFC emissions, expressed as CO₂-e emissions in the *Inventory* are about 8% lower than total HFC emissions (CO₂-e) estimated from Cape Grim data (NAME/ISC), with the *Inventory* reporting 9.97 M tonne emitted in 2013, compared to 7.99 M tonnes estimated from Cape Grim data. Total HFC emissions in CO₂-e terms from Cape Grim data peaked in 2010 at 9.6 M tonne, with 2013 emissions 19% below the peak.
- There is a small but significant source of HFC-23 emissions in Australia. HFC-23 has not been identified in past or current imports of refrigerants or refrigerant blends. It may be present as an unidentified contaminant in imported refrigerant blends.
- PFC-14 and PFC-116 emissions from the aluminium industry have been estimated using NAME and TAPM (PFC-116 indirectly via PFC-14). TAPM and NAME estimates for PFC-14 emissions agree to within 8%. Over the period

2005-2013, PFC-14 emissions in the *Inventory* are in decline and are about 25% lower than PFC-14 emissions estimated from Cape Grim data (NAME/ISC). In 2013 the *Inventory* reports 22 tonnes emitted, compared to 57 tonnes estimated from Cape Grim data. The *Inventory* data show PFC-14 emissions from the aluminium industry have declined at 10%/yr since 2006. The PFC-14 emissions from Cape Grim data have declined at 8%/yr since 2006.

- Total Australian PFC-116 emissions and PFC-116 emissions specifically from the aluminium industry have been estimated from atmospheric data using NAME and TAPM. Over the period 2005-2013, PFC-116 emissions in the *Inventory* (assumed only from the aluminium industry) and as seen at Cape Grim are in decline. The *Inventory* emissions averaged over the period 2005-2013 are equal to emissions derived from Cape Grim data (TAPM NAME PFC-14) for the aluminium industry and have declined by close to 80% since 2006.
- Total PFC-116 emissions as seen at Cape Grim (NAME) are increasing, approximately doubling since 2006, currently at 29 tonnes/yr. The data suggest that a significant, post-2007, non-aluminium PFC-116 source is in operation in Australia, and this is not captured in the import data. The source of this additional PFC-116 is unknown. Globally, the primary non-aluminium source of PFC-116 is from etching processes used in the electronics industry. Further analysis of past and future Aspendale and Sydney PFC-116 data could help identify this source.
- Cape Grim observations show that Australian emissions of PFC-218 were growing until 2011, but are now declining and PFC-318 emissions are declining, with current (2013) emissions for PFC-218 about 9 tonnes and PFC-318 about 15 tonnes. There are no significant Australian imports identified for these PFCs. Globally PFC-218 and PFC-318 are used in the electronics industry (etching), in refrigeration, in fire suppression and in medical applications. As with PFC-116, further analysis of past and future Aspendale and Sydney PFC-218 and PFC-318 data could help identify source(s) of these PFCs.
- Not surprisingly, total Australian PFCs (from Cape Grim data) declined from 2005 to 2008, largely driven by emissions from the aluminium industry. From 2008 to 2012 emissions were steady at around 100 tonnes per year and then declined further to 87 tonnes in 2013.
- Over the period 2006 2013, both the *Inventory* and Cape Grim data show that Australian sulfur hexafluoride emissions are in decline (26% decline in the Inventory, 46% seen at Cape Grim). However the *Inventory* sulfur hexafluoride emissions are significantly lower than emissions estimated from Cape Grim data; over the period 2006 to 2013 the sulfur hexafluoride emissions in the inventory average 6 tonnes/yr, about a factor of 6 lower than estimates from Cape Grim over the same period (37 tonnes/yr). This is a major discrepancy and is currently under investigation.
- Australian sulfur hexafluoride emissions in the Inventory are less <0.1% of global emissions. It would be unusual for Australian emissions of a widely-used industrial chemical like sulfur hexafluoride to be <0.1% of global emissions. A number approaching 1% would be consistent with several other widely used HFCs and HCFCs. The Cape Grim data indicate that Australian emissions of sulfur hexafluoride were 0.4% of global emissions in 2012.
- Total Australian HFC, PFC and sulfur hexafluoride emissions, expressed as CO₂-e emissions, over the period 2005-2013 estimated from atmospheric data (Cape Grim, ISC, NAME, TAPM) are about 20% higher than in the *Inventory*, although in 2013 the agreement is much closer (10%, *Inventory* lower). As indicated above, this comes about due to a reasonable agreement over the period 2005-2013 between the Inventory and atmospheric data for HFCs (8%, *Inventory* lower), whereas, for PFCs and sulfur hexafluoride, the *Inventory* emissions are substantially lower than those derived from Cape Grim data. In 2013, the combined PFC and sulfur hexafluoride emissions calculated from Cape Grim data were 114 tonnes, almost a factor of 4 higher than combined PFC and sulfur hexafluoride emissions in the *Inventory* (30 tonnes).
- Importantly, Cape Grim data suggest that the combined Australian emissions of HFCs, PFCs and sulfur hexafluoride peaked in 2010 and are now in decline, whereas the *Inventory* shows that emissions have continued to grow through 2013.

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