RRA Research Report 2011/2012

Paul Fraser, Paul Steele, Paul Krummel & Bronwyn Dunse
CSIRO Marine and Atmospheric Research
Centre for Australian Weather and Climate Research

November 2012

Australian emissions of ODSs and SGG

Methodology

Australian emissions of ozone depleting substances (ODSs – production/consumption regulated under Montreal Protocol) and synthetic greenhouse gases (SGGs – regulated under the Kyoto Protocol) have been estimated from Cape Grim data (Krummel *et al*., 2011). Emissions for 1994-2011 have been made by interspecies correlation (ISC) with carbon monoxide (CO) as the reference species (Dunse *et al*., 2005). CO data from the GC-MRD instrument at Cape Grim are available back to 1994. The Cape Grim data are used to calculate emissions from the Melbourne/Port Phillip region and Australian emissions are calculated from Port Phillip emissions scaled by population. PFC-14 (CF_4) emissions are scaled by aluminium production (Fraser *et al*., 2011).

Prior to 1994, Port Phillip CFC, methyl chloroform (CH_3CCl_3) and carbon tetrachloride (CCl_4) emissions are estimated by comparing the total pollution data for each species in 1994 to the total pollution data in previous years for each species back to 1982. The CFC, methyl chloroform and carbon tetrachloride data are from the AGAGE/GAGE GC-ECD instrument at Cape Grim.

For the other ODSs (HCFCs, halons) and the SGGs (HFCs, PFCs, SF_6) the emissions are calculated by ISC using Cape Grim GC-MS data (ADS and Medusa) for the period 2004-2011. If emission estimates are required for these species prior to 2004, the emissions are scaled to ODS import data if available. The emissions are reported as 3-year running averages, except for 2011, which is based on 2011 data only.

ODSs and SGGs

Australian ODS emissions were estimated for CFC-11 (CCl_3F), CFC-12 (CCl_2F_2), CFC-113 (CCl_2FCClF_2), methyl chloroform (CH_3CCl_3), methyl bromide (CH_3Br), HCFC-22 ($CHClF_2$), HCFC-124 ($CHClFCF_3$), HCFC-141b (CH_3CCl_2F), HCFC-142b (CH_3CClF_2), H-1211 ($CBrClF_2$) and H-1301 ($CBrF_3$).

Australian SGG emissions were estimated for HFC-32 (CH_2F_2), HFC-125 (CHF_2CF_3), HFC-134a (CH_2FCF_3), HFC-143a (CH_3CF_3), HFC-152a (CH_3CHF_2), HFC-227ea, HFC-236fa, HFC-245fa, HFC-365mfc, PFC-14 (CF_4), PFC-116 (CF_3CF_3), PFC-218 ($CF_3CF_2CF_3$) and sulphur hexafluoride (SF_6).

The Australian ODS and SGG emissions from 1990 to 2011 are shown in Figure 1 and in **Error! Reference source not found.**

Table 1Error! Reference source not found. (2005-2011).

The Australian ODS and SGG CO_2-e emissions from 1990 to 2011 are shown in Figure 2.

CFC emissions in the early 1990s were about 4000 tonnes/yr, which declined rapidly after the mid-1990s as the restrictions imposed by Montreal Protocol took effect. By 2000 emissions has declined to about 1000 tonnes/yr and have continued to decline slowly over the past decade, approaching 600 tonnes/yr in 2011, with approximately equal contributions from CFC-11 and CFC-12. These emissions over the past decade must be from CFC 'banks' – from landfills and from yet-to-be-decommissioned refrigeration/air-conditioning equipment, aerosol cans and foam plastics.

HCFC emissions totalled about 3000 tonnes/yr in 1990 and have declined slowly to about 2000 tonnes/yr currently. Emissions are dominated by HCFC-22 which is still widely used in Australia.

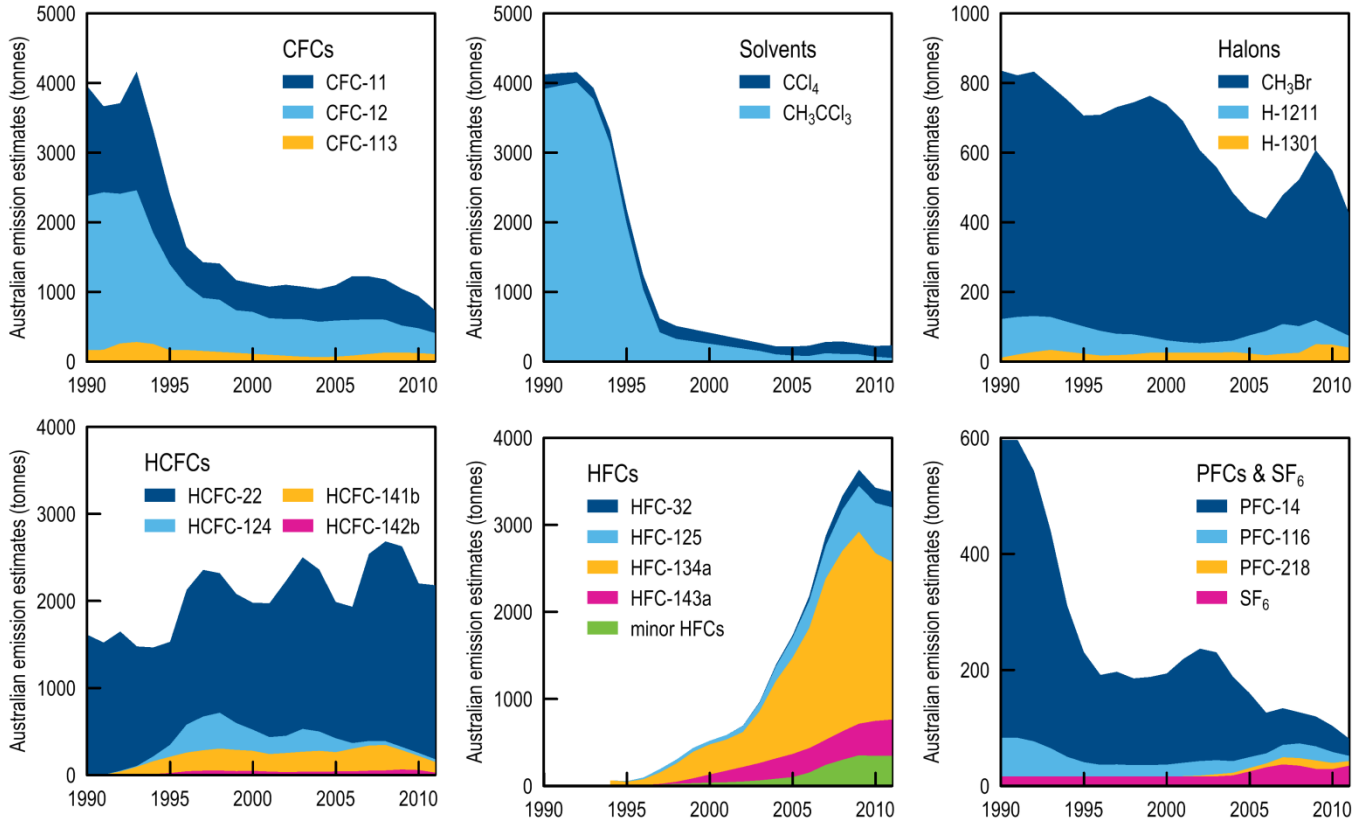

Figure 1. Australian ODS and SGG emissions from Cape Grim data based on ISC with CO as the reference species (Dunse *et al*., 2005). For the periods prior to the availability of Cape Grim data, emissions are scaled to imports (DSEWPaC, private communication). CF_4 emissions are scaled to aluminium production (Fraser *et al*., 2011).

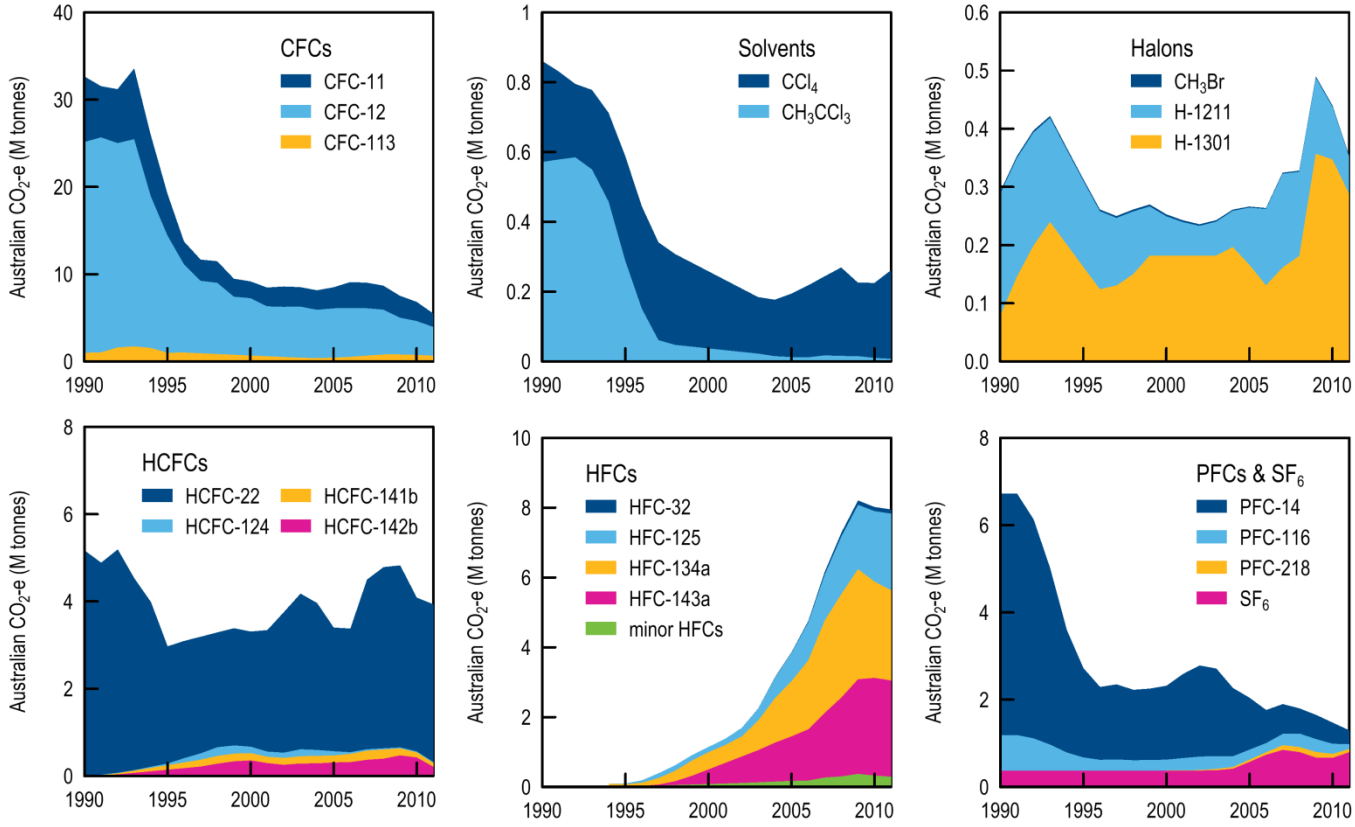

Figure 2. Australian ODS and SGG emissions, expressed as CO_2-e, from Cape Grim data based on ISC with CO as the reference species.

Table 1. Australian CFC, methyl chloroform, methyl bromide, HCFC, HFC, SF6 and PFC emissions (2005-2010) from atmospheric data, collected at Cape Grim, Tasmania on the Medusa GC-MS instrument; the carbon tetrachloride emissions are calculated from Cape Grim GC-ECD data; emissions are calculated by interspecies correlation (ISC, Dunse *et al*., 2005). The emissions are 3-year running averages, i.e. '2010' = average of 2009, 2010, 2011 emissions. Australian CFC, methyl chloroform, HFC and SF_6 emissions are scaled from regional emissions by population; the methyl bromide emissions are from Port Phillip; Australian PFC emissions are scaled by aluminium production (Fraser *et al*., 2011).

Species	2005	2006	2007	2008	2009	2010
			Tonnes			
CFC-11	508±267	626±306	617±310	578±229	578±216	462±188
CFC-12	519±195	513±182	498±185	472±180	386±153	357±139
CFC-113	70±31	87±34	109±46	131±56	131±58	122±52
methyl chloroform	87±44	81±38	120±55	109±49	105±48	71±32
carbon tetrachloride	149±58	122±42	124±63	145±67	193±99	120±55
methyl bromide	178±73	133±51	153±57	174±66	202±89	186±86
HCFC-22	1566±607	1566±584	2149±756	2295±768	2305±773	1947±669
HCFC-124	160±68	65±26	53±23	44±18	37±16	35±13
HCFC-141b	221±84	259±101	288±113	292±112	221±78	162±55
HCFC-142b	42±19	43±18	51±22	54±24	65±30	58±26
H-1211	52±25	69±31	85±36	77±31	69±30	48±22
H-1301	23±11	18±9	23±13	25±13	50±24	49±21
HFC-32	24±10	55±25	116±56	158±71	185±78	175±65
HFC-125	232±89	311±108	376±129	472±163	526±192	578±210
HFC-134a	1107±392	1379±475	1853±630	2074±693	2207±740	1926±652
HFC-143a	289±77	336±126	391±142	466±167	510±187	545±196
HFC-152a	26±10	35±12	47±18	49±19	51±21	49±18
HFC-227ea			23±12	25±11	29±13	23±9
HFC-236fa			7±3	6±2	8±4	5±2
HFC-245fa			24±12	23±11	30±14	36±14
HFC-365mfc		20±10	27±12	42±17	49±18	59±22
SF_6	25±9	32±10	37±14	35±15	29±14	29±12
PFC-14	110±87	70±46	63±46	54±37	51±39	45±31
PFC-116	19±10	17±8	21±11	25±13	24±13	19±9
PFC-218	6±3	7±3	12±6	13±7	15±7	11±5

Emissions of the major ODS solvents (CCl_4, CH_3CCl_3) were, like the CFCs, about 4000 tonnes/yr in 1990, declining rapidly in the mid-1990s, reaching a current value of about 200 tonnes/yr. These latter emissions are presumably largely from landfills (see CCl_4 discussions above).

Early (1990) halon emissions are uncertain, but likely larger than today. Based on import data, 1990 halons emissions were likely about 200 tonnes/yr declining to less than 100 tonnes/yr in 2011, with almost equal contributions from H-1211 and H-1301. Presumably these halon emissions are also from yet-to-be-decommissioned fixed and portable fire-fighting systems. The halon emissions data are shown with CH_3Br emission data, which are Australian emissions scaled from Port Phillip emissions to match a simple model of CH_3Br emissions based on QPS and non-QPS imports of CH_3Br. CH_3Br emissions were about 700 tonnes/yr in 1990, falling by about 50% to 350 tonnes/yr by 2011.

HFC emissions first appeared in the Australian environment in the mid-1990s as CFC/HCFC replacements. Their emissions grew rapidly until about 2008, reaching about 3000 tonnes/yr and have since levelled-off or even declined, despite growing imports. HFC emissions are dominated HFC-134a.

PFC emissions were about 600 tonnes/yr in the early 1990s, declining to about 200 tonnes/yr by the mid-1990s, as the aluminium industry (the major source of PFCs) adopted measures to reduce PFC emissions (Fraser *et al*., 2011). The decline in emissions has continued and today is less than about 50 tonnes. The significant PFC declines have been seen in PFC-14 (CF_4) and PFC-116 (C_2F_6), both largely produced by the aluminium industry. SF_6 emissions have grown slowly through this period reaching about 25-30 tonnes/yr in 2011.

For the ODSs, when expressed as CO_2-e emissions (Figure 2), there has been since 1990 about a 29 M tonne CO_2-e reduction in CFC emissions, a 2 M tonne reduction in HCFC emissions, 0.5 M tonnes in solvents and 0.5 M tonnes in halons, a total ODS reduction of about 32 M tonnes CO_2-e. This represents about 5% of Australia's total GHG emissions in 2010 and has had the same impact that removing approximately 300,000 cars from the Australian roads would have.

For the SGGs, HFC CO_2-e emissions have risen from approximately zero in 1995 to 7 M tonnes in 2011, while, for the PFCs and SF_6, CO_2-e emissions have fallen from about 7 M tonnes in 1990 to ~1 M tonnes in 2011. Overall the CO_2-e emissions of the SGGs fell from about 7 M tonnes in 1990 to 2 M tonnes in 1995 (driven by the fall in PFC emissions) and since 1995 risen to about 8 M tonnes in 2011, driven by HFC emissions (Figure 3).

Australia's net ODS and SGG emissions have been relatively constant since the late 1990s at about 17 m tonnes/yr, falling from about 40-50 M tonnes in the early 1990s. This has been the largest fall in Australia's GHG emissions for any sector (energy, agriculture, waste). ODS emissions are not counted as part of Australia's GHG emissions under the Kyoto Protocol.

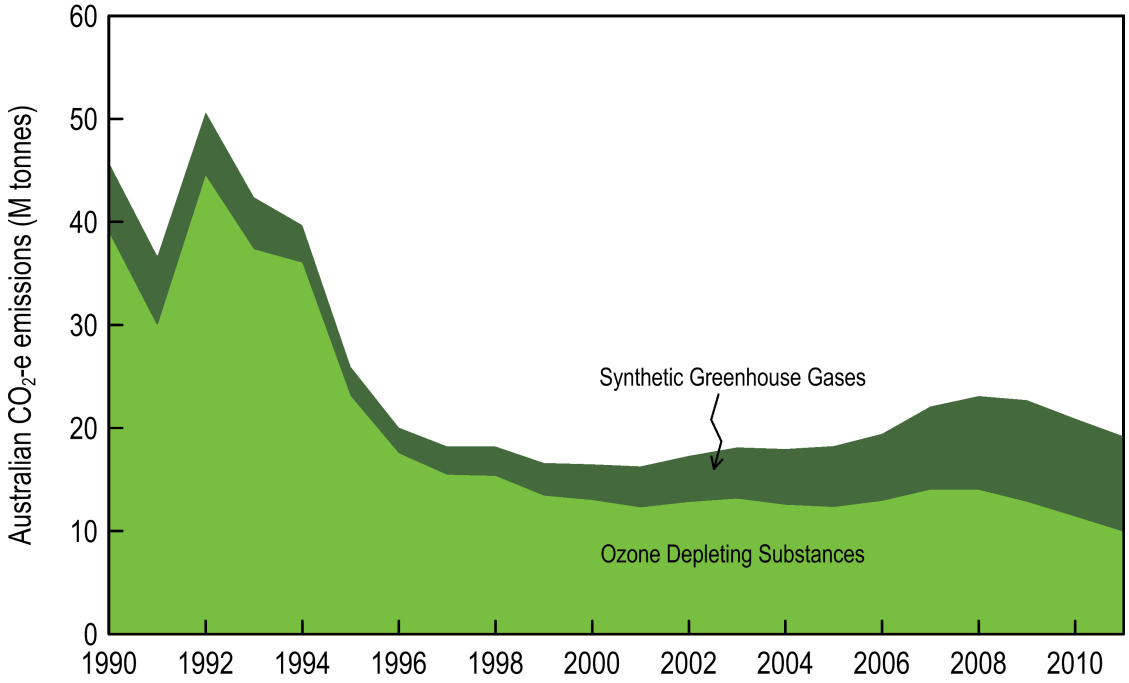

Figure 3. Total ODS and SGG CO_2-e emissions (M tonnes), based on ODS and SGG observations at Cape Grim, Tasmania, scaled to import data prior to the observation period (typically 1978-2011 ODSs, 2004-2011 SGGs).

HCFC-22 and HFC-134a

Figure 4 shows Australian HCFC-22 emissions as calculated by ISC from Cape Grim observations, HCFC-22 imports (DSEWPaC, private communication, 2012), emissions estimated by inverse modelling from Cape Grim and other global data using the MOZART model and consumption data reported to UNEP (Saikawa *et al*., 2012). The consumption data are for Australia scaled from Oceania data by population (81%).

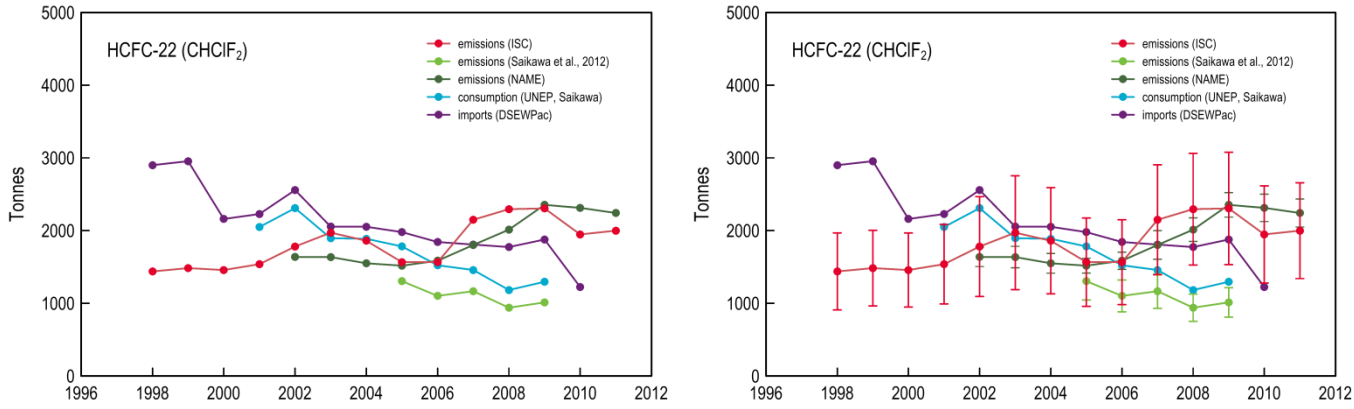

Figure 4. Australian HCFC-22 emissions (tonnes/yr), calculated from Cape Grim data by ISC (Dunse *et al*., 2005) and NAME (Manning *et al*., 2011), from Port Phillip emissions to Australian emissions on a population basis and by inverse modeling (Saikawa *et al*., 2012); Australian HCFC-22 consumption data (tonnes/yr, scaled from Oceania data on a population basis, Saikawa *et al*., 2012); Australian HCFC-22 imports (tonnes/yr, DSEWPaC, unpublished data).

There has been a steady decline in HCFC-22 imports from up to 3000 tonnes/yr (mid-to-late 1990s) to currently about 1200 tonnes/yr, a decline of about 50%. Cape Grim data suggest emissions of about 1500-2000 tonnes/yr from the late 1990s to the mid-2000s (less than imports) growing to over 2000 tonnes/yr since the mid-2000s (higher than imports). Rising emissions and declining imports can only be explained by rising emissions from the HCFC-22 'bank' – the HCFC-22 stored in existing refrigeration/air-conditioning infrastructure. Consistent with the declining imports are the declining consumption reported to UNEP. Consumption is lower than imports because some HCFC-22 is exported and it is possible that the Australia/Oceania scaling factor is too small.

The consumption data are the prior estimate of emissions used by Saikawa *et al*., and the resultant optimized annual emissions are about 1000-1300 tonnes/yr (2005-2009). It is clear that the declining trend in emissions in the prior is maintained in the optimized emissions, suggesting that the latter are not independent of the former. The Saikawa *et al.* emissions are lower than the ISC derived annual emissions over the same period (1600-2200 tonnes), but the uncertainties on the latter are large (±700 tonnes). It is also worth noting that the ISC method relies on the intensity (emissions per capita) of Port Phillip emissions being representative of Australia as a whole – if Port Phillip emissions are more intense than the national average then ISC will over-estimate Australian HCFC-22 emissions.

Figure 5 shows Australian HFC-134a emissions as calculated by ISC and NAME from Cape Grim observations, HFC-134a imports (DSEWPaC, private communication, 2012), HFC-134a emissions as reported to UNFCCC (DCCEE, 2012).

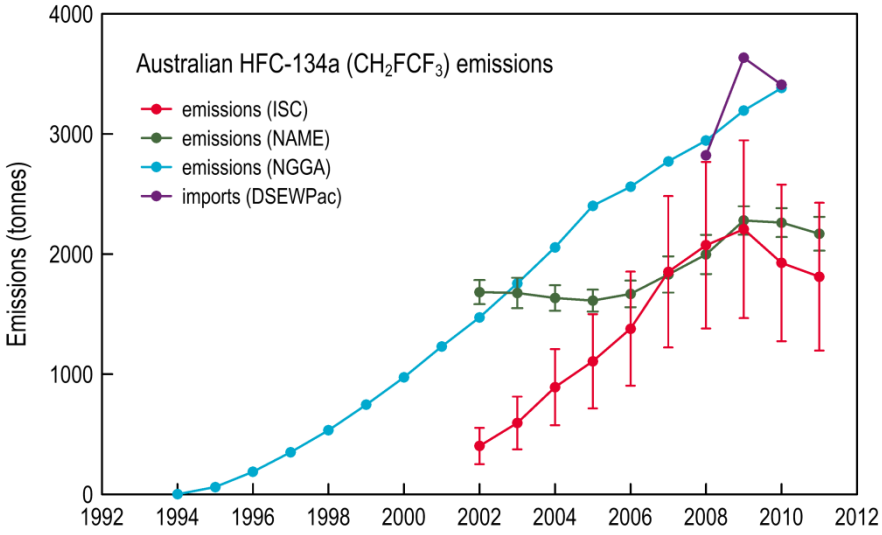

Figure 5. Australian HFC-134a emissions (tonnes/yr), calculated from Cape Grim data by ISC (Dunse *et al*., 2005) and NAME (Manning *et al*., 2011), from Port Phillip emissions to Australian emissions on a population basis; Australian HFC-134a emissions data (tonnes/yr, as published in the Australian National Greenhouse Accounts (NGGA: DCCEE, 2012); HFC-134a imports (tonnes/yr, DSEWPaC, unpublished data).

Australian emissions as reported to UNFCCC (DCCEE, 2012) have grow from near-zero tonnes in the mid-1990s to approximately 3500 tonnes currently. Imports (DSEWPaC, unpublished) approximately equal these emissions. However HFC-134a emissions estimated by ISC and NAME from Cape Grim data are smaller than reported to UNFCCC, reaching a maximum of about 2200-2300 tonnes in 2009, falling below 2000 tonnes in 2010 and 2011. It will be interesting to see if the NGGA data for 2011 (due in March 2013) show this turnaround in emissions. NAME and ISC emissions data are in general agreement from 2006 to 2011. Prior to 2006, the NAME emissions are higher than the ISC estimates – it is not obvious why this is so, but it is worth noting that there is a change in instrument at Cape Grim around 2004 (pre: GC-MS-ADS, post: GC-MS-Medusa) and the Medusa instrument is better able to identify HFC-134a pollution. The reason for this discrepancy is still being investigated.

As above for HCFC-22, the ISC method relies on the Port Phillip emission intensity of HFC-134a being similar to the national average emission intensity. Since the majority of HFC-134a emissions originate from automobile air conditioning, then emission intensity may not be nationally uniform, if, for example, it is dependent on average regional temperatures. It is generally accepted that the percentage of air conditioned vehicles is nationally uniform. Almost certainly, vehicle air conditioning is used more often in the tropics compared to mid-latitudes. If the HFC-134a leak rate when the air conditioning is in use is significantly higher than the stand-by emission rate, then tropical automobile HFC-134a emissions would be expected to be higher than mid-latitudes and then extrapolation from Port Phillip data would lead to an underestimate of national HFC-134a emissions.

References

DCCEE, *Australian National Greenhouse Accounts, National Inventory Report 2010*, Volume 1, Australian Government, Department of Climate Change and Energy Efficiency, 292 pp, 2012.

Dunse, B., P. Steele, S. Wilson, P. Fraser and P. Krummel, Trace gas emissions from Melbourne Australia, based on AGAGE observations at Cape Grim, Tasmania, 1995-2000, *Atmos. Environ.*, 39, 6334-6344, 2005.

Fraser, P., B. Dunse, P. Steele, P. Krummel & N. Derek, Perfluorocarbon (PFC) emissions from Australian aluminium smelters, 2005-2009, *Proceedings of the 10th Australasian Aluminium Smelting Technology Conference*, Launceston, Australia, 9-14 October, 2011, Welch, B., G. Stephens, J. Metson & M. Skyllos-Kazacos (eds.), Copyright School of Chemical Engineering, University of New South Wales, ISBN: 978-0-7334-3054-1, 14 pp, 2011.

Krummel, P., P. Fraser, P. Steele, L. Porter, N. Derek, C. Rickard, J. Ward, B. Dunse, R. Langenfelds, S. Baly, M. Leist & S. McEwan, The AGAGE *in situ* program for non-CO_2 greenhouse gases at Cape Grim, 2007-2008: methane, nitrous oxide, carbon monoxide, hydrogen, CFCs, HCFCs, HFCs, PFCs, halons, chlorocarbons, hydrocarbons and sulphur hexafluoride, *Baseline 2007-2008*, 67-80, 2011.

Manning, A., S. O'Doherty, A. Jones, P. Simmonds & R. Derwent, Estimating UK methane and nitrous oxide emissions from 1990 to 2007 using an inversion modelling approach, *J. Geophys. Res.,* 116, D02305, doi:10.1029/2010JD014763, 2011.

Saikawa, E., M. Rigby, R. Prinn, S. Montzka, B. Miller, L. Kuijpers, P. Fraser, M. Vollmer, T. Saito, Y. Yokouchi, C. Harth, J. Muhle, R. Weiss, P. Salameh, J. Kim, S. Li, S. Park, K.-R. Kim, D. Young, S. O'Doherty, P. Simmonds, A. McCulloch, P. Krummel, P. Steele, C. Lunder, O. Hermansen, M. Maione, J. Arduini, B Yao, L. Zhou, H. Wang, J. Elkins & B. Hall, Global and regional emissions estimates for HCFC-22, *Atmos. Chem. Phys. Discuss.*, 12, 18243-18285, 2012.