

Australian and Global Emissions of Ozone Depleting Substances

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1 Introduction

Chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), halons, carbon tetrachloride (CTC: CCl₄), methyl chloroform (MC: CH₃CCl₃) and methyl bromide (MB: CH₃Br) are all ozone depleting substances (ODSs), whose production and consumption, and resulting emissions, have been significantly reduced by national actions to comply with the Montreal Protocol. However, significant, persistent ODS emissions remain (Montzka and Reimann, 2011; Carpenter and Reimann, 2014), particularly in the developing world. If these emissions could have been reduced to zero by 2015, then ozone recovery dates at mid-latitudes and over Antarctica would have been brought forward by more than a decade (mid-latitudes: from 2047 to 2036; Antarctic: from 2073 to 2061; Bekki and Bodeker, 2011; Dameris and Godin-Beekmann, 2014; Harris and Wuebbles, 2014).

Methyl chloride (CH₃Cl), dichloromethane (CH₂Cl₂), chloroform (CHCl₃), trichloroethylene (TCE: CHClCl₂), perchloroethylene (PCE: CCl₂CCl₂), dibromomethane (CH₂Br₂), bromoform (CHBr₃) and methyl iodide (CH₃I) are not listed as ODSs in a Montreal Protocol context, because methyl chloride, chloroform, dibromomethane, bromoform and methyl iodide are predominantly natural in origin, and dichloromethane, trichloroethylene and perchloroethylene, although largely anthropogenic in origin, have relatively short atmospheric lifetimes (<0.5 yr, Montzka and Reimann, 2011) and therefore relatively low Ozone Depleting Potentials (ODPs). They are considered in this report in the context of their potential contribution to Equivalent Effective Stratospheric Chlorine (EESC), the ultimate driver of stratospheric ozone depletion (Bekki and Bodeker, 2011; Montzka and Reimann, 2011; Carpenter and Reimann, 2014; Fraser *et al.*, 2014a).

The most dramatic demonstration of the environmental impact of EESC is the annual appearance of the Antarctic ozone hole (AOH; Klekociuk *et al.*, 2015 and earlier papers). In a companion report (Krummel *et al.*, 2018 and earlier reports) we review the development and decline of the 2017 AOH, and review its metrics in light of the ongoing decline of EESC in the atmosphere. The overall ranking of the 2017 AOH is quantified in that report. In addition, ODS emissions play an important role globally in contributing to radiative forcing by greenhouse gases (GHGs), and reductions in ODSs have helped reduce the human contribution to climate change over the past 30 years (Forster and Ramaswamy, 2007; Myhre and Schindell, 2013; Carpenter and Reimann, 2014).

CFCs, halons, carbon tetrachloride and methyl chloroform are no longer imported into Australia in any significant quantities; however small amounts can still be used. Nevertheless, long-term atmospheric observations at Cape Grim, Tasmania, show that there are measurable past and current emissions of these chemicals from the Melbourne-Port Phillip-SE Australian region, and presumably from all the major Australian urban regions (Dunse *et al.*, 2005; Fraser *et al.*, 2014b). Previous research (Fraser *et al.*, 2013; 2014b) suggested that the emissions are likely from 'banks' of these species, in the form of old ODS-containing equipment and materials still in use (for example refrigerators, aerosol cans, fire extinguishers, foam plastics) or from leaking landfills, containing the aggregated emissions from buried ODS-containing equipment and materials by the Australian government to control ODS production and consumption under the Montreal Protocol – for example banning the emissions of ODSs unless permitted by regulation in the main end-use sectors: air conditioning, refrigeration and fire protection. In addition, mitigation of Australian emissions of these species is supported by government and industry initiatives in capture of used ODS substances, followed by their recycling or destruction.

HCFCs, methyl bromide, dichloromethane, trichloroethylene and perchloroethylene are imported into Australia and used in maintaining existing HCFC-containing refrigeration and air conditioning equipment, for quarantine and preshipment (QPS) uses of methyl bromide, such as grain fumigation immediately prior to international export, one exempted non-QPS use of methyl bromide (for growing strawberry runners), fire protection, and for solvent use (dichloromethane, trichloroethylene, perchloroethylene). HCFC and non-QPS methyl bromide consumptions are controlled by Australia's commitments under the Montreal Protocol and are declining, whereas QPS-uses of methyl bromide the Montreal Protocol and consequently methyl bromide emissions from QPS-use have the potential to increase. methyl bromide (SO₂F₂) are seen as suitable, but not universal, alternative or replacement chemicals. There are no controls over the use of dichloromethane, trichloroethylene, perchloroethylene from climate change or ozone depletion perspectives, but emissions of these chlorinated solvents are regulated in Australia due to their hazardous nature (toxicity) as volatile organic compounds (VOCs; EPA 1998).

2 Measurements of synthetic and natural ODSs at Cape Grim, Tasmania

The concentrations of synthetic (CFCs, HCFCs, halons, carbon tetrachloride, methyl chloroform, dichloromethane, trichloroethylene, perchloroethylene, methyl bromide-partially) and natural (methyl chloride, chloroform, methyl bromide-partially, dibromomethane, bromoform, methyl iodide) ODSs have been measured in the background (baseline) Southern Hemisphere atmosphere on air samples at or from Cape Grim, Tasmania. Measurements have been made on baseline air in the Cape Grim Air Archive (1978-2017) at CSIRO (Aspendale), at the Scripps Institution for Oceanography (SIO, USA), at the University of East Anglia (UEA, UK) and at Eidgenössische Materialprüfungs und Forschungsanstalt (Empa, Switzerland). Other flask air samples from Cape Grim and over SE Australia have been analysed at CSIRO, the Oregon Graduate Center (OGC, USA), SIO, the National Oceanographic and Atmospheric Administration (NOAA, USA), UEA, and the National Institute for Environmental Research (NIES, Japan). The synthetic and natural ODSs have been measured by CSIRO *in situ* in the atmosphere (baseline and non-baseline) at Cape Grim, Tasmania, since 1976 (CFC-11, carbon tetrachloride, methyl chloroform), since 1978 (CFC-12, chloroform), since the early 1980s (CFC-113), since the late-1990s (CFC-114, CFC-115, several HCFCs, halons), since the mid-2000s (several more HCFCs, methyl chloride, dichloromethane, trichloroethylene, perchloroethylene, methyl bromide) and more recently dibromomethane and bromoform.

CFC-114 has been measured *in situ* at Cape Grim (since 1998) and in the Cape Grim Air Archive (1978-2015) on UEA GC-MS and AGAGE ADS/Medusa GC-MS instruments (Oram, 1999; Krummel *et al.*, 2014 and earlier reports). 'CFC-114' measured on the AGAGE instruments is actually an unresolved mixture of CFC-114 (CCIF₂CCIF₂) and CFC-114a (CCl₂FCF₃), whereas the UEA GC-MS instrumentation can separate these two species, resulting in data for each species. Based on the early work of Oram (1999), in the international assessments of ozone depletion science (Carpenter and Reimann, 2014 and earlier assessments), it was assumed that AGAGE 'CFC-114' is approximately 90% CFC-114 and 10% CFC-114a, constant in time, likely reflecting a common source. Further work at UEA analysing the Archive shows that the CFC-114a: CFC-114 ratio is actually lower (currently 6%), and varies with time, suggesting, in part, separate sources for these CFCs, in particular in east Asia (Laube *et al.*, 2016).

Examples of new ODSs that have been measured recently in the Cape Grim Air Archive and/or *in situ* at Cape Grim, for which estimates of global abundances and some emissions have been made, include ³⁷Cl-CFC-11 (CCl₂³⁷ClF), ³⁷Cl-CFC-12 (CCl³⁷ClF₂), CFC-112 (CCl₂FCCl₂F), CFC-112a (CClF₂CCl₃), CFC-113a (CCl₃CF₃), ³⁷Cl-CFC-113 (CCl³⁷ClFCClF₂), CFC-216ba (CClF₂CClF₂), CFC-216ca (CClF₂CCF₂CClF₂), HCFC-31 (CH₂ClF), HCFC-133a (CH₃CClF₂) and HCFC-225ca (CHCl₂CF₂CF₃) (Kloss *et al.*, 2014; Laube *et al.*, 2014; Vollmer *et al.*, 2015; Schoenenberger *et al.*, 2015; CSIRO unpublished data). Other new ODSs recently measured at Cape Grim include HCFC-21 (CHCl₂F, commencing 2015), HCFC-132b (CHCl₂CHF₂, commencing 2015) and HCFC-1233zd (or HFO-1233zd: CHClCHCF₃, commencing 2014). Provisionally-calibrated data are available for HCFC-132b (Table 1).

These data are used, in conjunction with similar data collected from other Northern and Southern Hemispheric sites, to estimate both global and regional concentration trends, atmospheric lifetimes and emissions for these species, which have been reported in the peer-reviewed scientific literature:

- CFCs Fraser *et al.*, 1977, 1983, 1996; Fraser and Pearman, 1978a,b; Hyson *et al.*, 1980; Rasmussen and Khalil, 1979; Rasmussen *et al.*, 1982; Cunnold *et al.*, 1983; 1986, 1994, 1997; Oram, 1999; Fraser, 2000; Prinn *et al.*, 2000; Sturrock *et al.*, 2002; Dunse *et al.*, 2005; Laube *et al.*, 2013, 2014, 2016; Rigby *et al.*, 2013, 2014; Kloss *et al.*, 2014; Allin *et al.*, 2015; Meinshausen *et al.*, 2017; Montzka *et al.*, 2018, Vollmer *et al.*, 2018
- HCFCs
 Rasmussen et al., 1980, 1982; Montzka et al., 1994; Oram et al., 1995; Miller, 1998; Miller et al., 1998, 2010; Oram, 1999; Prinn et al., 2000; Sturrock et al., 2002; O'Doherty et al., 2004; Greally et al., 2007; Stohl et al., 2009; Saikawa et al., 2012; Laube et al., 2013, 2014; Kloss et al., 2014; Rigby et al., 2014, 2017; Vollmer et al., 2015; Schoenenberger et al., 2015; Chirkov et al., 2016; Liang et al., 2017; Meinshausen et al., 2017; Simmonds et al., 2017
- halons Butler *et al.*, 1994; Fraser and Prather, 1999; Fraser *et al.*, 1999; Oram, 1999; Fraser, 2000; Sturrock *et al.*, 2002; Newland *et al.*, 2013; Vollmer *et al.*, 2016; Meinshausen *et al.*, 2017
- CTC
 Fraser and Pearman, 1978a; Rasmussen et al., 1982; Simmonds et al., 1988, 1998; Prinn et al., 2000;

 Sturrock et al., 2002; Dunse et al., 2005; Xiao, 2008; Xiao et al., 2010a; Laube et al., 2013; Fraser et al., 2014b; Rigby et al., 2014; Chipperfield et al., 2016; Liang et al., 2016; Meinshausen et al., 2017
- MC
 Fraser and Pearman, 1978a; Rasmussen et al., 1982; Fraser et al., 1986; Prinn et al., 1987, 1992, 1995,

 2000, 2001, 2005; Oram, 1999; Sturrock et al., 2002; Dunse et al., 2005; Laube et al., 2013; Rigby et al.,

 2013, 2014, 2017; Patra et al., 2014; McNorton et al., 2016; Liang et al., 2017; Meinshausen et al., 2017

- CH₃CI Rasmussen et al., 1982; Prinn et al., 2000; Cox, 2001; Cox et al., 2003a, 2004; Simmonds et al., 2004; Trudinger et al., 2004; Xiao, 2008; Xiao et al., 2010b; Meinshausen et al., 2017 CH_2CI_2 Cox et al., 2000, 2003a,b; Cox, 2001; Trudinger et al., 2004; Simmonds et al., 2006; Xiao, 2008; Meinshausen et al., 2017; Hossaini et al., 2017; Oram et al., 2017 CHCl₃ Rasmussen et al., 1982; Prinn et al., 2000; O'Doherty et al., 2001; Cox, 2001; Cox et al., 2003b, 2004; Trudinger et al., 2004; Xiao, 2008; Hossaini et al., 2015; Meinshausen et al., 2017 TCE Simmonds et al., 2006 PCE Rasmussen et al., 1982; Simmonds et al., 2006 MB Cox, 2001; Sturrock et al., 2002, 2003a,b; Cox et al., 2004; Simmonds et al., 2004; Trudinger et al., 2004; Porter et al., 2006, 2009, 2010; Meinshausen et al., 2017 CH_2Br_2 Yokouchi et al., 2005 CHBr₃ Yokouchi et al., 2005
- CH₃I Cox, 2001; Cohan et al., 2003; Cox et al., 2004

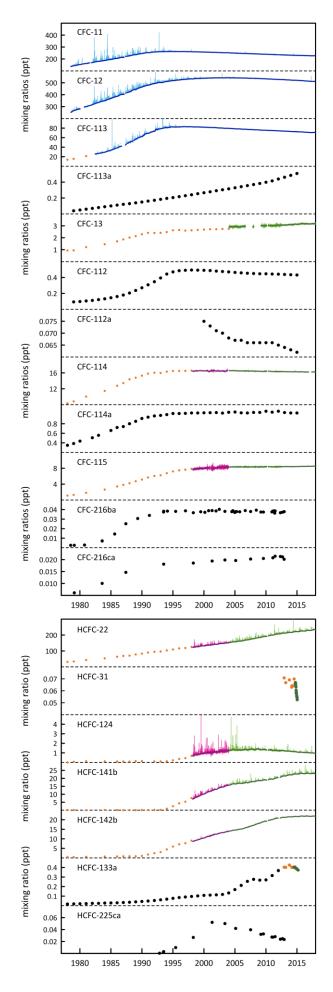
The abundances and trends of CFCs, HCFCs, halons, carbon tetrachloride, methyl chloroform, methyl bromide, methyl chloride, dichloromethane, chloroform, trichloroethylene, perchloroethylene, dibromomethane and bromoform in the global background atmosphere, as measured at Cape Grim, Tasmania, or in the Cape Grim air archive, are shown in Table 1 (2015-2017) and Figure 1 (1978-2017).

Table 1. Southern Hemisphere concentrations (2016, 2017) and growth rates (2016-2017) for CFCs, HCFCs, halons, carbon tetrachloride, methyl chloroform, methyl chloride, dichloromethane, chloroform, trichloroethylene, perchloroethylene, methyl bromide, dibromomethane and bromoform measured *in situ* at Cape Grim, Tasmania and/or in the Cape Grim Air Archive (references: see text above and CSIRO unpublished data).

Species	pecies Formula		ration	Grow	/th	Species	Formula	Concent	ration	Grow	/th
		2016	2017	ppt/yr	%/yr			2016	2017	ppt/yr	%/yr
CFCs						Halons					
CFC-11	CCl₃F	227.8	226.7	-1.2	-0.51	H-1202	CBr_2F_2	0.02 ^c	0.02 ^c	0.0	0.0
CFC-12	CCl ₂ F ₂	515.2	511.9	-3.3	-0.65	H-1211	CBrClF ₂	3.6	3.4	-0.11	-3.0
CFC-13	CCIF₃	3.2	3.2	0.00	0.09	H-1301	CBrF ₃	3.3	3.3	-0.01	-0.29
CFC-112	CCl ₂ FCCl ₂ F	0.53	0.52	-0.01	-0.72	H-2402	$CBrF_2CBrF_2$	0.4	0.4	-0.005	-1.2
CFC-112a	$CCIF_2CCI_3$	0.06	0.06	0.00	-1.7	total halons		7.3	7.2	-0.12	-1.7
CFC-113	CCl ₂ FCClF ₂	71.3	70.8	-0.49	-0.69	total halon (Cl)		3.6	3.4	-0.11	-3.0
CFC-113a	CCI_3CF_3	0.53 ^b	0.55 ^b	0.02	4.2	total halon (Br)		7.7	7.6	-0.12	-1.6
CFC-114 ^a	CCIF ₂ CCIF ₂	16.2	16.2	0.01	-0.08	Other ODSs					
CFC-114a	CCl ₂ FCF ₃					carbon tetrachloride	CCI ₄	79.1	78.0	-1.1	-1.4
CFC-115	CCIF ₂ CF ₃	8.5	8.5	0.03	0.32	methyl chloroform	CH ₃ CCl ₃	2.6	2.2	-0.4	-16.6
CFC-216ba	CF ₂ ClCFClCF ₃	0.034	0.034	0.00	-2.2	methyl chloride	CH₃CI	522.8	531.6	8.8	1.7
CFC-216ca	CF ₂ ClCF ₂ CF ₂ Cl	0.020	0.020	0.00	-1.1	dichloromethane	CH_2CI_2	12.9	15.8	3.0	20.7
total CFCs		844.5	840.5	-3.9	-0.47	chloroform	CHCl₃	6.0	5.9	-0.11	-1.8
total CFC (Cl)		1978.1	1968.6	-9.5	-0.48	TCE	CHCICCI ₂	0.021	0.021	0.001	2.4
HCFCs						PCE	CCl ₂ CCl ₂	0.35	0.36	0.01	2.9
HCFC-22	CHCIF ₂	226.9	231.3	4.4	1.9	total other CI-ODSs		623.8	633.9	10.2	1.6
HCFC-31 ^d	CH ₂ CICF ₃					total other Cl-ODSs (Cl)		892.4	901.2	8.8	1.0
HCFC-124	CHCIFCF ₃	1.0	1.0	-0.05	-5.1	methyl bromide	CH₃Br	6.7	6.1	-0.6	-9.4
HCFC-132b	$C_2H_2Cl_2F_2$	0.09	0.09	0.00	1.3	dibromomethane	CH_2Br_2	1.1	1.1	0.007	0.63
HCFC-133a	CH ₂ CICF ₃	0.40	0.42	0.02	5.5	bromoform	CHBr₃	2.0	1.8	-0.26	-13.5
HCFC-141b	CH ₃ CCl ₂ F	23.1	23.0	-0.09	-0.41	total other Br-ODSs		9.8	8.9	-0.85	-9.1
HCFC-142b	CH ₃ CCIF ₂	21.9	21.9	-0.01	-0.05	total other Br-ODSs (Br)		14.9	13.6	-1.4	-9.5
HCFC-225ca	$CF_3CF_2CHCl_2$	0.009	0.005	0.00	-56.3	total Cl		3170.6	3174.0	3.3	-0.11
total HCFCs		273.4	277.6	4.2	1.5	total Br		22.7	21.2	-1.5	-6.8
total HCFC (CI)		296.6	300.7	4.2	1.4						

^aAGAGE 'CFC-114' = CFC-114+CFC-114a ^b from 2012 concentration and growth rate ^c from 2014 concentrat

^c from 2014 concentration and growth rate ^dmeasured in 2015 only



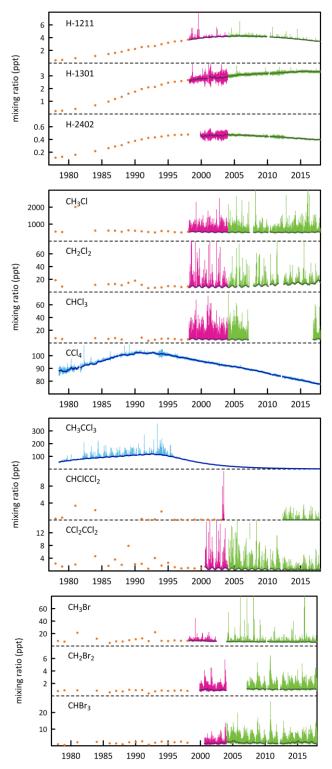


Figure 1. Cape Grim *in situ* and Air Archive observations of CFCs, HCFCs, halons, carbon tetrachloride, methyl chloroform, methyl chloride, dichloromethane, chloroform, TCE, PCE, methyl bromide, dibromomethane and bromoform (1978 – 2017) showing baseline monthly mean data (Medusa - dark green; ADS - purple; ECD – dark blue; Archive data – orange) and total data (Medusa - light green; ADS – pink; ECD – light blue) obtained from the GC-MS-Medusa, GC-MS-ADS and GC-ECD instruments at Cape Grim and Aspendale. The CFC-112, -112a, -113a, -216ba, -216ca and HCFC-133a data are from UEA and Empa measurements on the Cape Grim Air Archive (see text above and CSIRO unpublished data).

2.1 CFCs

CFC-11 and CFC-12 are the dominant CFCs in the atmosphere, constituting 88% of all CFCs in 2017. CFC-113, CFC-114 and CFC-115 constitute 11% of CFCs, the remaining minor CFCs about 1%. The CFCs account for 62% of chlorine from all ODSs in the background atmosphere.

As a result of measures undertaken within the Montreal Protocol framework, the majority of the CFCs (CFC-11, CFC-12, CFC-112, CFC-112a, CFC-113, CFC-114) measured in the atmosphere in 2017 at Cape Grim have stopped growing or are in decline, the exceptions being CFC-13, CFC-113a and CFC-115, which are growing slowly. Total CFCs are declining by 0.5% per year due to declining emissions (see below); chlorine from CFCs in the atmosphere decreased by 10 ppt (2016-2017, 0.5%) the same decline as in 2015-2016.

The summed concentration of the minor CFCs (CFC-112, CFC-112a, CFC-113a, CFC-216ba and CFC-216ca; Kloss *et al.*, 2014, Laube *et al.*, 2014) in 2017 is likely only ~1 ppt, while the other CFCs in the background atmosphere sum to 840 ppt in 2017. The summed growth of these minor CFCs in the atmosphere is 0.01 ppt/yr. These low abundance CFCs are thought to be used as chemical feedstocks, or produced as by-products, with some fugitive emissions, or used as specialised solvents (Kloss *et al.*, 2014, Laube *et al.*, 2014).

Global concentrations of the major CFCs (CFC-11, CFC-12, CFC-113) are all in decline (Figure 2). However, the rate of decline of global CFC-11 concentrations has slowed down from -1.66 ppt/yr (2012-2013) to -1.17 ppt/yr (2016-2017). The rate of decline for CFC-12 continues to increase, -2.64 ppt/yr (2012-2013) to -3.69 ppt/yr (2016-2017). The growth rate for CFC-113 has increased slightly from -0.59 ppt/yr (2012-2013) to -0.64 ppt/yr (2016-2017).

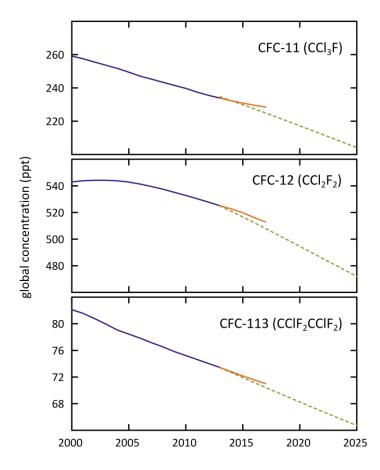


Figure 2. Global concentrations of CFC-11, CFC-12 and CFC-113 (ppt). The dashed line is the WMO 2014 A1 scenario.

2.2 HCFCs

The major HCFCs measured in the atmosphere in 2017 at Cape Grim are HCFC-22, HCFC-141b and HCFC-142b. In 2015 all three HCFCs were growing in the atmosphere, but now (2016) HCFC-22 is the only one growing (2% per year). HCFC-141b and HCFC-141b are declining by 0.41% and 0.05% per year respectively. The dominant HCFC is HCFC-22 (231 ppt in 2017), 83% of the abundance and 99% of the growth of all HCFCs. HCFC-141b and HCFC-142b constitute 16% of HCFCs, the remaining minor HCFCs close to 1%. The growth rate of total HCFCs is slowing down. Total HCFCs increased by 4.2

ppt or 1.5% per year (2016-2017) compared to 1.4% per year (2015-2016). Chlorine in the atmosphere from HCFCs (301 ppt in 2017, 9% of total chlorine from all ODSs) increased by 4.2 ppt (1.4%, 2016-2017), the only ODS sector showing an increase in chlorine (apart from methyl chloride and dichloromethane, see below).

HCFC-31 was identified in the Cape Grim Air Archive (Laube *et al.*, 2014; Schoenenberger *et al.*, 2015; Vollmer *et al.*, 2015) in 2015. The background concentration of HCFC-31 is low (0.06 ppt in 2015). HCFC-133a was also identified in the Cape Grim Air Archive and is now measured routinely at Cape Grim, with a background concentration of 0.42 ppt in 2017). There are no clearly identified sources of these HCFCs, but it is possible they are emitted to the atmosphere from an inadvertent by-product emission (Laube *et al.*, 2014; Schoenenberger *et al.*, 2015; Vollmer *et al.*, 2015). It has been suggested that there may have been a change in the industrial processes that result in their release to the atmosphere (Schoenenberger *et al.*, 2015; Vollmer *et al.*, 2015).

Measurements of HCFC-132b commenced at Cape Grim in July 2014. The 2017 annual mean HCFC-132b concentration was 0.09 ppt, increasing by 1.3%/yr (2016-2017). HCFC-123 (CHCl₂CF₃) was measured *in situ* at Cape Grim from 1998 to 2004 on the GC-MS-ADS instrument (Krummel *et al.*, 2006). The 2004 annual mean was 0.064 ppt increasing by 0.003 ppt per year. At this rate of increase, current HCFC-123 concentrations at Cape Grim would be less than 0.1 ppt. HCFC-123 is not measured on the current GC-MS-Medusa instrument at Cape Grim.

Global concentrations of the major HCFCs (HCFC-22, HCFC-141b, HCFC-142b) are growing in the atmosphere (Figure 3). The rate of increase for all 3 HCFCs has slowed since 2012. The growth rate for HCFC-22 has slowed down from 5.48 ppt/yr (2012-2013) to 3.40 ppt/yr (2016-2017). The growth rate for HCFC-141b decreased from 0.82 ppt/yr (2012-2013) to -0.06 ppt/yr (2016-2017), therefore concentrations may begin to decline. The growth rate for HCFC-142b may also declined from 0.29 ppt/yr (2012-2013) to 0.02 ppt/yr (2016-2017), therefore concentrations of HCFC-142b may also begin to decline in subsequent years.

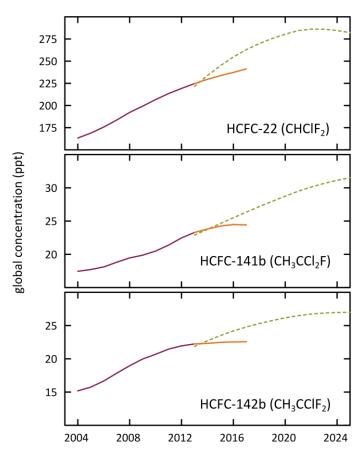


Figure 3. Global concentrations of HCFC-22, HCFC-141b and HCFC-142b (ppt). The dashed line is the WMO 2014 A1 scenario.

2.3 Other Chlorocarbons ODSs

The most abundant chlorocarbon in the background atmosphere is the largely naturally-occurring methyl chloride (CH₃Cl, 532 ppt, 2017), accounting for 84% of all chlorocarbons and 59% of chlorine from chlorocarbons. The next most abundant chlorocarbon is anthropogenic carbon tetrachloride (78.0 ppt, 2017), accounting for 35% of chlorine from chlorocarbons. The remaining minor chlorocarbons, including methyl chloroform, contribute 6% of chlorocarbon chlorocarbon account for 28% of total chlorine from all ODSs in the background atmosphere.

Carbon tetrachloride, methyl chloroform and chloroform measured at Cape Grim showed decreasing concentrations in 2016 compared to 2015, whereas methyl chloride, dichloromethane, trichloroethylene and perchloroethylene all showed increasing concentrations. The largest decrease in the chlorocarbons was 0.4 ppt (16.6%) for methyl chloroform and the largest increase in the chlorocarbons was 3.0 ppt (20.7%) for dichloromethane. Significant inter-annual variability is expected for naturally-emitted methyl chloride and chloroform, which have oceanic and biomass burning sources. Total chlorine from chlorocarbons increased by 8.8 ppt (1%), much higher than the previous years increase of 1.3 ppt (0.1%). This increase in chlorine from chlorocarbons is due predominately to dichloromethane. Two recent papers (Hossaini *et al.*, 2017 and Oram *et al.*, 2017) have shown that dichloromethane is increasing rapidly in the atmosphere. Oram et al., 2017 state that globally, dichloromethane concentrations have increased 60% over the last decade. Sustained growth in dichloromethane could offset some of the gains achieved by the Montreal Protocol. Dichloromethane will be examined more closely in a companion report (Dunse *et al.*, 2018).

The rate of decline of methyl chloroform is consistent with its relatively short atmospheric lifetime and near-zero global emissions, whereas the rate of decline of carbon tetrachloride in the atmosphere suggests there are remaining, significant carbon tetrachloride sources outside the control of the Montreal Protocol (Fraser *et al.*, 2014; Liang *et al.*, 2016).

2.4 Halons

H-1211 is the most abundant halon in the background atmosphere (3.4 ppt, 2017), followed by H-1301 (3.3 ppt) and H-2402 (0.4 ppt). H-1211, H-1301 and H-2402 are in decline in the atmosphere (-0.11 ppt/yr, -0.01 ppt/yr and -0.005 ppt/yr respectively, 2016-2017). Overall halons are in decline by 1.7% per year (compared to 2%/yr 2015-2016), a larger rate of decline (in percentage terms) compared to the CFCs. Bromine in the atmosphere from halons decreased by 0.12 ppt/yr (1.6%), which equates to about a 5 ppt/yr decline in equivalent chlorine. This is significant – chlorine from CFCs is declining currently by 10 ppt/yr. The decline in bromine from halons is a significant driver of likely ozone recovery (see Krummel *et al.*, 2016).

2.5 Other organobromine species

Methyl bromide is the most abundant (6.1 ppt) organobromine ODS in the background atmosphere in 2017, followed by H-1211 (3.4 ppt), H-1301 (3.3 ppt), bromoform (1.8 ppt) and dibromomethane (1.1 ppt).

Methyl Bromide showed a large decrease (0.6 ppt/yr 2016-2017) in the background atmosphere. There is an overall long-term decrease in methyl bromide in the atmosphere since about 2000, which briefly halted in 2012-2013 and 2014-2015. Because there are significant natural emissions of methyl bromide, year-to-year variability in the rate of decline or growth is expected.

Natural bromoform showed a decrease (0.26 ppt/yr) and natural dibromomethane showed a small increase (0.007 ppt/yr) in 2016-2017. Overall bromine from all non-halon ODSs decreased by 1.4 ppt/yr (2016-2017), a 9.5%/yr decrease, largely due to the decrease in bromine from bromoform and methyl bromide. This is the second year in a row that that background levels of bromoform have decreased. In 2014-2015, bromoform was at its highest concentration for the last decade. Significant inter-annual variability is expected for naturally emitted dibromomethane and bromoform, which have oceanic sources. Long-term trends (decadal) have not been found for these species (Carpenter and Reimann, 2014).

2.6 Total chlorine and bromine: impact on stratospheric ozone

Total chlorine from ODSs (Figure 4) increased from 3170.6 ppt in 2016 to 3174 ppt in 2017, an increase of 3.3 ppt (0.11%). This is only the second time in 20 years that there has been an increase in total chlorine from ODSs (2007-2008 increase of 2 ppt). Total chlorine from chlorocarbons increased by 8.8 ppt (1%), much higher than the previous years increase of 1.3 ppt (0.1%). This increase in chlorine from chlorocarbons is due predominately to dichloromethane. The overall decline in chlorine from CFCs was 9.5 ppt (2016-2017) and HCFC chlorine increased by 4.2 ppt.

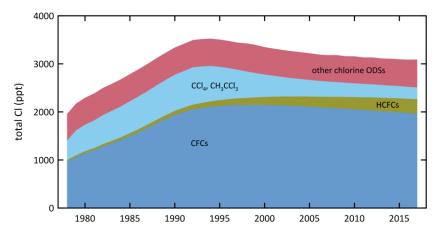


Figure 4. Total chlorine from CFCs, HCFCs, carbon tetrachloride: CCl₄, methyl chloroform: CH₃CCl₃ and other chlorine-containing ODSs (Table 1) as measured at Cape Grim.

Total bromine from organobromine ODSs (Figure 5) was 21.2 ppt (2017) – 36% from halons, 29% from methyl bromide, 25% bromoform and 10% dibromomethane. Bromine from all ODSs decreased by 1.5 ppt (2016-2017, 6.8%), due to - 0.12 ppt/yr from the halons and -1.4 ppt/yr from the non-halon ODSs. The variability in total bromine compared to total chlorine is due to the relatively larger natural (therefore variable) contributions to bromine compared to chlorine.

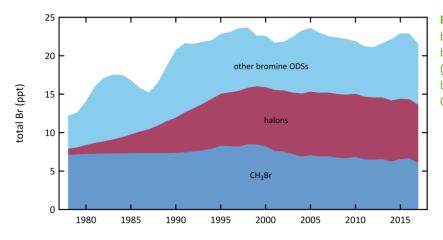


Figure 5. Total bromine from methyl bromide: CH₃Br, halons and other bromine-containing ODSs (dibromomethane - CH₂Br₂ and bromoform - CHBr₃) as measured at Cape Grim (Table 1).

The impact of total chlorine and bromine from ODSs on stratospheric ozone at polar and mid-latitudes is discussed in detail in the companion Report on the 2017 Antarctic Ozone Hole (Krummel *et al.*, 2018). Figure 6 shows the strong correlation between ozone depletion over Antarctica (Halley Station) and at mid-latitudes in the Southern Hemisphere (Melbourne). In both regions there is a strong indication of the onset of significant ozone recovery.

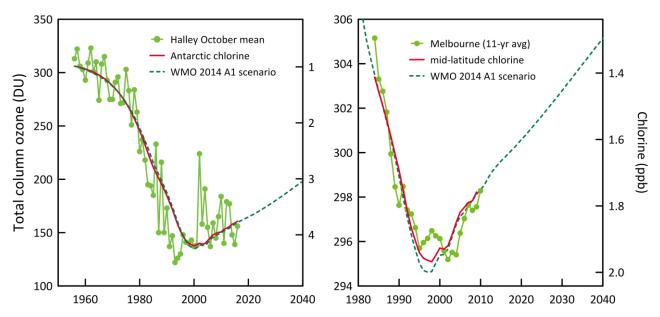


Figure 6. Total column ozone (DU) changes at Halley Station, Antarctica (76°S), and Melbourne, Australia (38°S) and Equivalent Effective Stratospheric Chlorine (EESC, ppb) changes at polar and mid-latitudes. The Melbourne ozone data are 11-year running means to minimize impacts of solar variability.

2.7 Global radiative forcing from ODSs, HFCs and other SGGs

ODSs and other synthetic greenhouse gases (SGGs, for example hydrofluorocarbons – HFCs) make a significant contribution to global radiative forcing (Figure 7). SGGs are the second most important driver of climate change after carbon dioxide since the 1950s. Radiative forcing from total SGGs almost stopped growing in the background atmosphere in the 1990s, due to the overall success of the Montreal Protocol, but recommenced growing in the 2000s due to growing global emissions of HFCs and growing emissions of HCFCs in the developing world. Montreal Protocol HCFC controls will impact these emissions from the developing world (10% reduction by 2015 and 35% reduction by 2020). Global HFC emissions should also commence to be restrained once the Kigali Amendment to the Montreal Protocol enters into force on the 1st January 2019. The Kigali Amendment mandates a phase-down schedule for HFC production and consumption. Hopefully this will lead to another 'plateau' in radiative forcing from SGGs in the next 5-10 years.

Total radiative forcing due to SGGs is increasing by 0.3%/yr (2016-2017). Radiative forcing from CFCs contributes 70% of total SGG radiative forcing but is in decline by 0.6%/yr (2016-2017). HCFCs currently contribute 16% of SGG radiative forcing and the HCFC contribution is increasing by 1.6%/yr (2016-2017). The remaining ODSs (halons, chlorocarbons) currently contribute 2.2% of SGG radiative forcing but is in decline by 3.7%/yr (2016-2017).

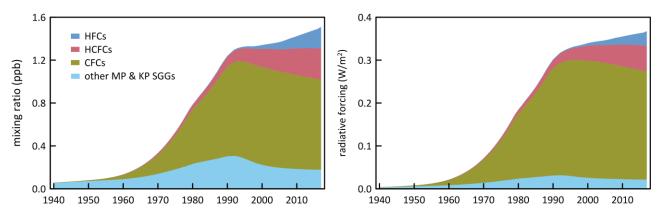


Figure 7. Global abundance (ppb, left) and radiative forcing (W/m², right) of synthetic greenhouse gases (SGGs: HFCs, HCFCs, CFCs, others).

3 Global emissions of ODSs

Background ODS observations at Cape Grim and from other AGAGE stations in the Northern and Southern Hemispheres have been used to calculate global ODS emissions up to 2016 (Figure 8; Rigby *et al.*, 2014 and updates; Vollmer *et al.*, 2015). These emissions are derived using the AGAGE 12-box global model of atmospheric chemistry and transport (Rigby *et al.*, 2013) and a Bayesian inverse method based on Rigby *et al.* (2011, 2013). ODS emissions have been calculated for recently identified ODSs in the Cape Grim Air Archive using forward modelling in a 2-D atmospheric chemistry-transport model (Laube *et al.*, 2014; Kloss *et al.*, 2014) and Bayesian inverse modelling based using the FLEXPART dispersion model (Laube *et al.*, 2014; Kloss *et al.*, 2014; Rigby *et al.*, 2014 and updates; Schoenenberger *et al.*, 2015; Vollmer *et al.*, 2015).

Total global CFC emissions are declining, dominated (91%) by emissions of CFC-11 (73 k tonnes) and CFC-12 (32 k tonnes) in 2016. Since the peak emissions of CFCs in the late 1980s (1128 k tonnes, 1987), overall CFC emissions have declined by 8%/yr, attesting to the success of the Montreal Protocol controls on CFC production and consumption. There has been no significant decline in CFC-11 emissions since 2002, whereas all other CFC emissions have shown a long-term decline. Thus CFC-11 became the dominant CFC emitted globally in 2012, taking over from CFC-12, whose emissions had previously always been larger than CFC-11 (since 1979 based on AGAGE data, and earlier based on production data). Presumably this reflects a different pattern of CFC use in developing countries compared to past use in developed countries. This is supported by a recent paper, Montzka *et al.*, 2018, that has found an unexpected increase in global emissions of CFC-11. The conclusions from this study suggest that the increase is probably not related to existing banks of CFC-11 but rather new uses. Global CFC-11 emissions from Rigby *et al.*, 2014 and updates for 2016 are 73 k tonnes and compare well with 2016 estimates from Montzka *et al.*, 2018 of 67 k tonnes.

CFC-12 emissions continue to decline from 40 k tonnes in 2015 to 32 k tonnes in 2016, continuing a remarkably monotonic decrease in emissions since the late-1980s. The 2014-2015 decline in emissions (19 k tonnes) is the largest annual decline in CFC-12 emissions since 2005-2006 (22 k tonnes).

The emissions of the other CFCs (CFC-113, -114, -115), including the minor CFCs, increased from a total of 5.3 k tonnes in 2015 to 11.1 k tonnes in 2016. This increase of 5.8 ktonnes is the largest annual increase since 2004-2005 (7.8 ktonnes). Most of the increase is due to emissions of CFC-114.

Global carbon tetrachloride emissions declined from 35 k tonnes in 2015 to 34 k tonnes in 2016. Long-term, global carbon tetrachloride emissions have declined slowly from a peak of around 120 k tonnes/yr in the late 1970s, declining to 50 k tonnes/yr by 2005, a long-term decline of about 3%/yr. Since 2005 global carbon tetrachloride emissions have declined steadily by about 1-2 k tonne per year (3%/yr) until 2016. The decline in global carbon tetrachloride emissions is not as rapid as anticipated under the Montreal Protocol (Montzka and Reimann, 2011; Carpenter and Reimann, 2014). A partial explanation may be that global emissions of carbon tetrachloride from land-fills and chlor-alkali plants may be significant, and not yet accounted for, in global budgets (Fraser *et al.*, 2014; Hu *et al.*, 2016, Liang *et al.*, 2016).

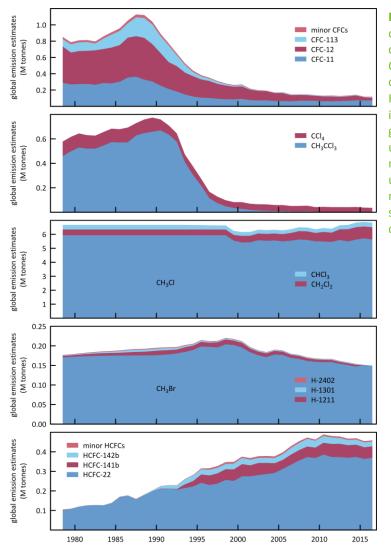


Figure 8. Global annual emissions (M tonnes) of ODSs (CFCs, chlorinated solvents (methyl chloroform: CH₃CCl₃, carbon tetrachloride: CCl₄), methyl chloride, dichloromethane, chloroform, halons, methyl bromide: CH₃Br, HCFCs) derived from global AGAGE data by inverse modelling using the 12-box AGAGE global transport model, by forward modelling using a 2-D atmospheric chemistry-transport model and by Bayesian inverse modelling using the FLEXPART dispersion model. For methyl bromide, pre-1998 emissions are scaled from global atmospheric concentrations.

Global methyl chloroform emissions peaked in 1990 at 669 k tonnes. Emissions have declined almost every year since, with a small increase in 2008 (8.1 k tonnes) then declining significantly to 1.2 k tonnes in 2016. Over the past 5 years (2012-2016) global emissions have averaged 2.2 k tonnes/yr. The long term decline in methyl chloroform emissions is in excess of 20%/yr, which is about what is expected for an ODS with low (~zero) emissions and an atmospheric lifetime of about 5 years.

Global HCFC emissions peaked in 2010 (488 k tonnes) and have since declined (6%) to 458 k tonnes in 2016, a decline of about 1%/yr. The decline in emissions since 2010 is encouraging in relation to the overall long-term decline in ODSs that will bring about, possibly hasten, ozone layer recovery. Throughout the period from the late-1970s to 2010, total HCFC emissions increased by about 4%/yr. Now the emissions of all of the major HCFCs are in decline: HCFC-22 emissions peaked in 2010 at 386 k tonnes declining (0.7%/yr) to 370 k tonnes in 2016; HCFC-141b: 68 k tonnes (2012) to 58 k tonnes (4%/yr, 2016); HCFC-142b: 39 k tonnes (2008) to 24 k tonnes (6%/yr, 2016). Declining HCFC emissions cause the observed slowing of the recent HCFC concentration growth rates in the atmosphere (see Section 2.2). If HCFC emissions continue to decline, HCFC concentrations will peak and then also start to decline over the next few years.

Data on halon emissions are now available back to the 1960s, based on AGAGE *in situ* and Cape Grim Air Archive data (Vollmer *et al.*, 2016). Total global halon emissions continue to decline (6.9 k tonnes in 2012, 4.9 k tonnes in 2016), dominated (~70%) by halon-1211 emissions, which declined by 1.3 k tonnes over the same period. Peak total halon emissions (18 k tonnes) occurred in 1990 and have declined to 4.9 k tonnes in 2016 (~5%/yr; H-1211: 4.5%/yr, H-1301: 5.5%/yr, H-2402: 6%/yr).

Global methyl bromide emissions have been estimated from AGAGE data, including Cape Grim (Rigby *et al.*, 2014) and these have been updated to 2016. The emission estimates start in 2000 (196 k tonnes), declining steadily to 149 k tonnes in 2016, a long-term decline of about 1.5% per year. methyl bromide emissions from 2013-2016 are fairly steady, ranging from 147-151 ktonnes suggesting the long-term decline in methyl bromide emissions may have levelled off in line with developing country phase-out in 2015.

The total ODS global emissions for the Montreal Protocol ODSs (CFCs, HCFCs, halons, methyl chloroform, carbon tetrachloride and methyl bromide) in ODP tonnes are shown in Figure 9, together with the global atmospheric concentration data for these species expressed as equivalent chlorine. The combined global emissions of the Montreal Protocol ODSs peaked at 1460 k tonnes (1.6 M tonnes) in the late 1980s, declining, thanks to the Montreal Protocol restrictions on ODS production and consumption, at 5%-6% per year from 1990 to 294 tonnes in 2016. Note the significant contribution of methyl bromide to global emissions in, for example, 2016 (~30%), most (96% in Australia in 2016, see below) of which is from QPS consumption (not controlled by the Montreal Protocol). At peak ODS emissions in the late 1980s, methyl bromide contributed only 7%.

The total global concentrations of ODSs, expressed as equivalent chlorine (Figure 9), peaked later in the mid-1990s at 4135 ppt (4.14 ppb) declining slowly (24 ppt per year) to 3.69 ppb by 2016, resulting largely from the long (50-100+ years) lifetimes for carbon tetrachloride and CFCs in the atmosphere.

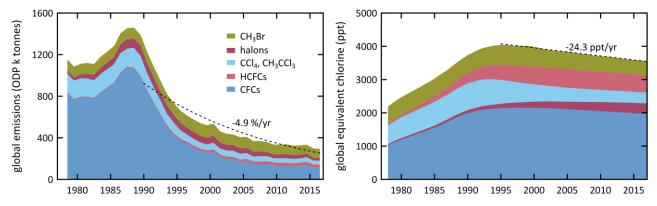


Figure 9. Global emissions (ODP tonnes) of the Montreal Protocol ODSs and global equivalent chlorine (ppt), both derived from AGAGE data (Fraser *et al.*, 2014a; Rigby *et al.*, 2014 and subsequent updates) using the 12-box AGAGE model. For methyl bromide, pre-1998 emissions are scaled to post-1998 from global atmospheric concentrations, 1978-2016.

4 Australian ODS imports and banks

Data on Australian imports of ODSs are reported to the Australian Government (Department of the Environment and Energy - DoEE) under licensing arrangements in the *Ozone Protection and Synthetic Greenhouse Gas Management Act, 1989*, with the requirement being established under the Act in 1989. Australian imports are documented in the DoEE *Ozone Licensing and Reporting System* (OLaRS: A. Gabriel, DoE, private communications, 2014-2017), which was introduced during 2011, replacing previous import data recording systems. OLaRS details imports of bulk and precharged refrigeration and air conditioning equipment containing HCFCs (individual HCFCs or HCFC blends) and other bulk ODSs (in particular carbon tetrachloride and methyl bromide).

For 2012 – 2016 (January – December) imports are reported entirely via the OLaRS protocol. Methyl bromide data were reported separately up until 2015, but are now incorporated into OLaRS. For 2008-2010, the ODS imports are reported using the pre-OLaRS protocol. Imports of ODSs are shown in Table 2 and Figure 10.

There is an overall decline in Australian ODS imports from over 15,000 tonnes in 1991 to 990 tonnes in 2017, a long-term decline of about 250 tonnes per year since 1995. ODS imports in 2017 increased by over 250 tonnes from the previous year to 990 tonnes, due to methyl bromide imports rising from 691 tonnes (2016) to 945 tonnes (2017), all for QPS applications, i.e. not subject to phase-out under the Montreal Protocol.

In 1991 CFCs were the major ODS imports (7,144 tonnes), but they declined rapidly to 371 tonnes by 1996, an overall decline of about 50% per year as a result of the phase-out by the Montreal Protocol, except for a few essential uses. By 2003 CFC imports were virtually zero. Methyl chloroform (MC) was the second largest ODS import in 1991 (4,700 tonnes) but imports ceased by 1996. HCFCs imports nearly doubled between 1991 (2,400 tonnes) and 1998 (4,200 tonnes); since 1998 there has been a long term decline in HCFC imports, falling to 43 tonnes by 2017. Australia has reached its second last step in its HCFC phase out, with an annual import limit for HCFCs of 45 tonnes from 2016 to 2029 (DoEE, 2016)

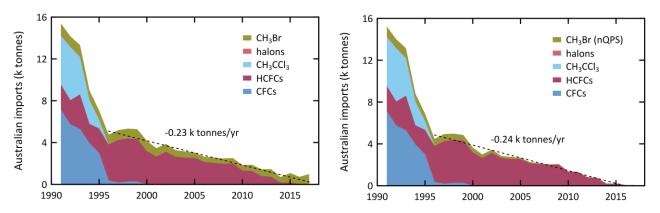


Figure 10. Australian imports (tonnes) of ODSs (CFCs, HCFCs, halons, MC: CH₃CCl₃, MB: CH₃Br – total left, MB: CH₃Br – nQPS right) (A. Gabriel, DoE, private communication, 2018). The 2012-2017 data are from the OLaRS data base, 2010 and earlier are pre-OLaRS data, and 2011 is a mixture of OLaRS and pre-OlaRS data.

Methyl bromide (MB) imports were just over 1,000 tonnes per year in the early 1990s, falling to about 350 tonnes in 2007, a long-term decline of 8% per year, driven by the reduction in non-QPS use of methyl bromide. After 2007, methyl bromide imports increased and by 2011 had reached over 730 tonnes, twice as large as the 2007 imports. Since 2011, imports have declined again to 586 tonnes in 2014, rising to 945 tonnes in 2017, the highest methyl bromide imports since 2000. The increase in methyl bromide imports since 2007 has been driven by an increase in QPS use of methyl bromide. Variations in QPS demand for methyl bromide in Australia will likely follow overall grain and wood products production trends, trading partner requirements and the use of methyl bromide alternatives (for example phosphine and sulfuryl fluoride). Grain production increased from about 20 M tonnes at the height of the recent drought (2006/2007) to 62 M tonnes in 2016/2017 (ABARES, 2018). Imports of methyl bromide for non-QPS use fell to 30-35 tonnes by the mid-2000s and have stayed at under 30 tonnes to 2017.

The Australian CFC bank (in operational refrigeration/air conditioning equipment) is now estimated to be less than 450 tonnes. The Australian HCFC-22 bank has been estimated at 13450 tonnes (2007), declining to 5265 tonnes in 2015. The only other significant ODS bank in operational equipment is HCFC-123, with a bank of 2320 tonnes in 2003 declining to 482 tonnes in 2015 (Figure 11; Brodribb and McCann, 2013, 2014, 2015a). It is estimated that about one third of all end-of-life vehicles (ELVs) have no residual air conditioning refrigerant charge when dismantled or crushed. Across the entire population of ELVs it is estimated that they contained approximately 170 tonnes of HFC-134a and 20 tonnes of CFC-12 in 2013 (Brodribb and McCann, 2015b).

Australian HCFC-22 and HCFC-123 emissions have been estimated using refrigerant bank data and refrigeration/air conditioning – specific emission factors (Figure 11; Brodribb and McCann, 2015a). HCFC-22 emissions have declined from 1320 tonnes in 2003 to 450 tonnes in 2014-2015. Comparing the estimated HCFC-22 emissions and banks indicates a HCFC-22 emission factor of 14% of the bank in 2003 declining to 6% in 2014-2015.

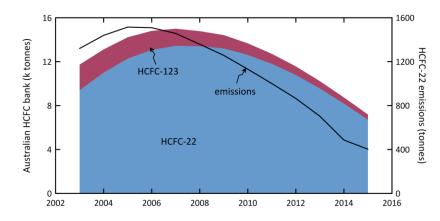




Table 2. Australian imports (bulk and pre-charged equipment, tonnes) of ODSs (CFCs, HCFCs, MC: CH₃CCl₃, halons, MB: CH₃Br) 1991-2017; 2012-2017 data are from the OLaRS data base, 2010 and earlier are pre-OLaRS data, and 2011 is a mixture of OLaRS and pre-OlaRS data (A. Gabriel, DoE, private communications, 2014-2016). The only significant carbon tetrachloride (CCl₄) imports were ~0.5 tonnes in 1995/1996 (not listed). ODS imports less than 0.1 tonne are not listed, but included in total ODS. Methyl bromide imports are listed as for QPS and non-QPS uses. Small quantities of methyl bromide imports are exported (do not influence Australian emissions); significant quantities HCFC-123 imports are exported or used as feedstock (do not influence Australian emissions) – for example 10 tonnes exported and 3 tonnes used for feedstock in 2014, ~1 tonne exported 2015).

	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017
CFC-11	1759	1786	1443	927	498	69	52	90	90				1														
CFC-12	4049	3054	3205	2784	2229	181	129	182	182	8	8	8													0.01		
CFC-113	999	808	485	168	236	118	0.1	0.1	0.1																		
CFC-114	6	19	6	11	7	3	3	3	3	1	1	1															
CFC-115	331	84	172	64	46																						
Total CFCs	7144	5751	5311	3954	3016	371	184	275	275	9	9	10	1														
HCFC-22	2402	2252	2940	1368	1663	2626	3056	2900	2955	2160	2228	2557	2054	2053	1979	1843	1808	1773	1878	1224	1288	758	714	178	181	45	43.17
HCFC-123		17	60	67	74	59	52	72	76	20	7	34	11	31	28	15	18	21	48	26	13	28	22	12	9.3		
HCFC-124			8	2	49	93	195	189	140	56	60	46	64	43	29	15	18	20	13	30	5	4	3	0.5	1.4		
HCFC-141b		30	269	411	522	579	639	827	813	892	301	446	473	396	428	241	198	155	76	49	3		1				
HCFC-142b		3	23	14	29	89	130	141	87	59	52	40	54	27	84	40	26	12	8	4	3	2		0.3			
HCFC-225ca			1.2	1.1	0.9	0.8	0.7	0.6	0.4	0.3	0.6	0.2	0.2	0.4	0.7	0.3	0.4	0.4	0.5	0.3	0.4	0.5	0.4	0.3	0.3	0.3	
Total HCFCs	2402	2302	3301	1823	2338	3448	4071	4158	4105	3187	2648	3123	2656	2551	2548	2156	2068	1981	2023	1333	1313	793	740	191	192	45.3	43.17
CH ₃ CCl ₃	4680	5086	3586	2273	846	0.1																					
H-1211	111	14																					0.1	0.4			1.26
H-1301	11	39				1																	1	2.6			0.78
Total halon	122	53				1.2																	1.3	3.0			2.04
MB-nQPS	876	799	921	704	664	631	679	570	507	451	340	323	183	207	119	55	46	41	33	34	33	33	31	30	30	28	29.75
MB-QPS	172	160	166	172	168	276	259	352	425	517	475	415	441	390	361	361	294	417	509	472	690	676	618	556	864	663	915.09
Total MB	1048	959	1087	876	832	907	1031	921	932	968	815	738	624	597	490	427	351	477	557	522	734	719	649	586	894	691	945
Total ODSs	15396	14151	13285	8926	7032	4726	5257	5354	5312	4164	3467	3870	3281	3148	3038	2583	2419	2458	2580	1859	2039	1512	1390	780	1086	736	990.21

5 Estimated Australian ODS emissions from Cape Grim data

Estimates of emissions of CFCs, HCFCs, methyl chloroform, carbon tetrachloride, halons and methyl bromide from the Melbourne/Port Phillip region (Dunse *et al.*, 2001, 2005; Dunse 2002; Greally *et al.*, 2007; Fraser *et al.*, 2012, 2013, 2014b), have been made utilising *in situ* measurements from the Cape Grim Baseline Air Pollution Station in Tasmania and an interspecies correlation (ISC) technique with co-incident carbon monoxide (CO) measurements.

The original ISC emission estimates were based on an average CO emissions from the Melbourne/Port Phillip region (600 k tonnes/yr) which were assumed to have been relatively constant during 2004-2009 (EPA, 1998). In Fraser *et al.* (2012), revised estimates of the Port Phillip region CO emissions were used (Delaney and Marshall, 2011) with 2002 emissions estimated at 605 k tonnes and 2006 emissions at 645 k tonnes. Carbon monoxide emissions were assumed constant after 2006 for the Port Phillip region. There has been a further revision of CO emissions from the Port Phillip region (S. Walsh, Victorian EPA, unpublished data, 2013). The 2006 Port Phillip CO emissions are now estimated to have been significantly higher at 796 k tonnes, with the increase in emissions compared to earlier estimates due to increased emissions from vehicles and reduced emissions from wood heaters. In this Report, the time-dependence of the Port Phillip CO emissions, Port Phillip ODS emissions have been calculated, for 1994-2016, using Cape Grim *in situ* data and ISC, obtained from the GC-ECD and GC-MS instruments at Cape Grim, and scaled to Australian emissions, where appropriate, on a population basis (using a population-based scale factor of 5.4). NOAA air mass back trajectory analyses (Draxler and 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 carbon monoxide source regions, in particular the Latrobe Valley.

SE Australian ODS emissions can be calculated from Cape Grim data using the NAME model. NAME (Numerical Atmospheric dispersion Modelling Environment) is a Lagrangian particle dispersion model (O'Doherty *et al.*, 2009; Manning *et al.*, 2003, 2011) driven by 3-dimensional 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 hour period at Cape Grim, which grid boxes in a prescribed domain impact on Cape Grim over the previous 12 days. NAME releases 33000 particles at Cape Grim over a 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 assumes 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 to the prior estimates.

The NAME model 'sees' emissions from Victoria/Tasmania or Victoria/Tasmania/New South Wales (depending on 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 NAME Victorian/Tasmanian or Victorian/Tasmanian/NSW emissions using population based scale factors of 3.7 and 1.7 respectively, when appropriate, and are shown in Figure 12.

Australian halon emissions are derived from SE Australian emissions (ISC), assuming 6-8 % of SE Australian emissions are from the National Halon Bank located in Melbourne (see below; in Fraser *et al.*, 2013, 10% emission were assumed). The SE Australian (non-Halon Bank) halon emissions are scaled to Australian emissions based on population.

Australian methyl bromide emissions (QPS) are scaled from SE Australian emissions (ISC), less non-QPS SE Australian emissions derived from non-QPS imports and a non-QPS emission factor applicable to methyl bromide use in Australia (100% of Australian non-QPS methyl bromide is assumed to originate in SE Australia); the QPS scaling factor is based on the assumption that 35% of Australian QPS methyl bromide originating from SE Australia grain export ports (35% of Australia's grain exports originate from SE Australian ports). Australian methyl bromide emissions (QPS) plus SE Australian methyl bromide emissions (non-QPS) are the sum of Australian methyl bromide emissions (QPS) plus SE Australian methyl bromide emissions (non-QPS).

Australian HCFC-22 emissions have been estimated using an emissions model based on HCFC-22 imports, and estimates of the HCFC-22 bank as refrigerants and refrigerant leakage rates from the bank (Figure 11; Brodribb and McCann, 2015a).

The Australian emissions of ODSs - CFCs, HCFCs, halons, MB, Montreal Protocol chlorocarbons (methyl chloroform, carbon tetrachloride) other chlorocarbons (dichloromethane, chloroform, trichloroethylene, perchloroethylene) - are presented as 3-year running averages (1995-2016: Table 3, Figure 12).

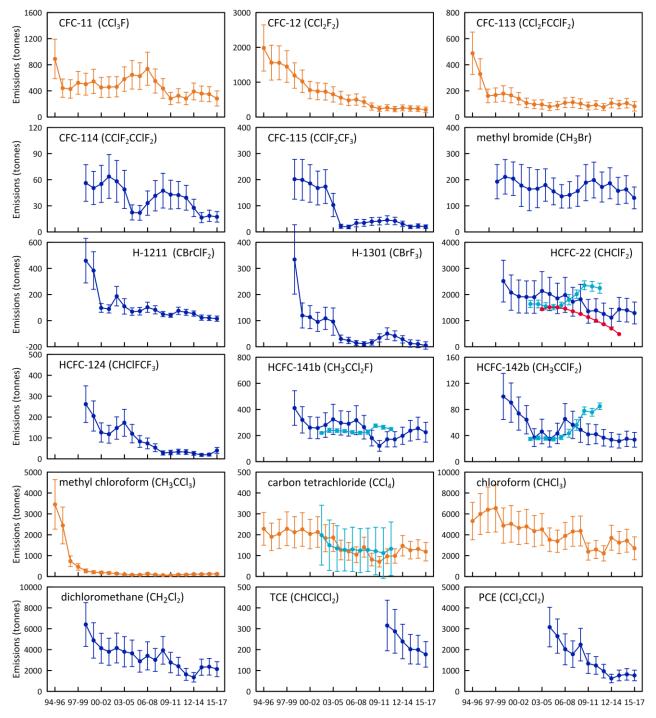


Figure 12. Annual average (3-yr running means) Australian emissions of CFCs, methyl bromide, HCFCs, halons and chlorocarbons (methyl chloroform, carbon tetrachloride, chloroform, dichloromethane, trichloroethylene, perchloroethylene) from Cape Grim AGAGE data, using ISC techniques (orange: GC-ECD data; blue: GC-MS data). methyl bromide emissions are for SE Australia. Australian emissions are scaled from SE Australian emissions on a population basis; halon emissions are adjusted to account for SE Australian Halon Bank emissions. NAME emissions are show in light blue. HCFC-22 emissions from the refrigerant bank are show in red (Brodribb and McCann, 2015a).

Table 3. Annual average (3-yr running means, i.e. 2014 = average of 2013, 2014, 2015) Australian emissions (metric tonnes unless otherwise stated) of ODSs (CFCs, HCFCs, halons, MB and chlorocarbons) from Cape Grim AGAGE data, using ISC techniques. Australian emissions are scaled from SE Australian emissions on a population basis; Australian halon emissions are from SE Australian emissions adjusted for the impact of emissions from the National Halon Bank in Melbourne; Australian methyl bromide emissions are from a DPI-modified UNEP model of methyl bromide emissions based on QPS and non-QPS methyl bromide consumption (see text). GWPs (to calculate CO₂-e emissions) are from Forster and Ramaswamy (2007); ODPs (to calculate ODP-weighted emissions) are from Montzka and Reimann (2011); assumed GWPs for trichloroethylene (0.67) and perchloroethylene (0.53). Pre-1999 emissions of CFC-114, -115, HCFCs, halons and dichloromethane (shown in red) are assumed equal to 1999 emissions; pre-2005 emissions of perchloroethylene (shown in red) are assumed equal to 2005 emissions.

Refrigerant	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016
CFCs																						
CFC-11	889	441	429	523	503	545	453	458	462	580	647	627	739	551	437	283	325	283	391	358	349	283
CFC-12	1982	1561	1555	1451	1190	1025	774	738	727	649	552	480	500	433	300	235	265	226	261	243	237	208
CFC-113	488	330	161	168	179	166	140	108	96	96	79	87	109	112	102	84	92	75	107	95	106	82
CFC-114	56	56	56	56	56	50	55	64	58	49	22	22	33	42	47	43	42	39	28	16	19	19
CFC-115	202	202	202	202	202	199	186	168	173	103	21	19	33	35	40	41	45	42	31	19	23	23
total	3359	2332	2146	2142	1872	1736	1367	1304	1286	1324	1278	1194	1348	1095	839	601	681	585	759	695	691	573
ODP tonnes	3438	2444	2291	2286	2014	1873	1506	1447	1429	1416	1298	1210	1380	1135	890	652	733	634	783	705	702	589
M tonnes CO ₂ -e	30.9	23.2	22.0	21.4	18.5	16.8	13.4	12.8	12.6	11.7	10.0	9.1	10.2	8.7	6.7	5.1	5.7	5.0	5.9	5.2	5.2	4.5
HCFCs																						
HCFC-22	2514	2514	2514	2514	2514	2073	1924	1904	1903	2135	2021	1852	1984	1722	1817	1358	1389	1258	1113	1432	1401	1293
HCFC-124	262	262	262	262	262	205	126	118	147	173	120	83	74	53	28	30	34	33	26	19	20	40
HCFC-142b	100	100	100	100	100	90	74	64	37	46	35	43	65	56	49	42	42	37	33	31	35	33
HCFC-141b	411	411	411	411	411	320	261	258	280	324	296	290	318	264	180	120	171	171	198	237	255	225
total	3286	3286	3286	3286	3286	2689	2384	2344	2367	2678	2472	2268	2441	2096	2074	1550	1636	1499	1371	1719	1711	1591
ODP tonnes	196	196	196	196	196	160	142	140	141	160	149	138	150	129	124	91	99	91	86	107	108	99
M tonnes CO ₂ -e	5.2	5.2	5.2	5.2	5.2	4.3	3.9	3.9	3.8	4.3	4.0	3.7	4.0	3.5	3.5	2.7	2.8	2.5	2.3	2.8	2.8	2.6
Halons																						
H-1211	459	459	459	459	459	384	98	90	187	109	69	72	102	83	50	41	75	64	54	23	21	15
H-1301	335	335	335	335	335	120	113	95	109	96	30	24	14	11	17	34	50	42	28	12	11	5
total	794	794	794	794	794	503	211	185	296	205	99	96	116	94	66	75	125	105	82	35	31	20
ODP tonnes	4724	4724	4724	4724	4724	2348	1426	1220	1650	1290	506	457	449	361	315	461	728	608	446	192	167	93
M tonnes CO ₂ -e	2.9	2.9	2.9	2.9	2.9	1.3	0.91	0.77	0.99	0.81	0.30	0.26	0.23	0.19	0.18	0.29	0.44	0.37	0.27	0.11	0.10	0.05
methyl bromide (MB)																						
total	542	581	623	650	662	661	627	560	512	459	410	351	352	395	447	534	587	634	583	625	638	696
ODP tonnes	325	349	374	390	397	396	376	336	307	275	246	210	211	237	268	321	352	381	350	375	383	417
k tonnes CO ₂ -e	2.7	2.9	3.1	3.2	3.3	3.3	3.1	2.8	2.6	2.3	2.1	1.8	1.8	2.0	2.2	2.7	2.9	3.2	2.9	3.1	3.2	3.5

chlorocarbons (MP)																						
carbon tetrachloride (CT)	228	190	204	229	212	225	203	213	183	187	127	120	104	140	81	71	97	98	147	126	132	119
methyl chloroform (MC)	3454	2446	731	464	270	207	190	171	146	100	85	87	137	90	66	68	96	96	115	113	127	124
total	3682	2636	935	693	482	432	393	384	329	287	212	207	241	230	147	138	193	194	262	239	259	242
ODP tonnes	596	454	297	298	260	269	243	251	216	215	148	141	128	164	96	84	116	118	174	150	158	143
M tonnes CO ₂ -e	0.91	0.70	0.47	0.48	0.42	0.44	0.39	0.41	0.35	0.35	0.24	0.23	0.21	0.27	0.16	0.14	0.19	0.19	0.28	0.24	0.26	0.23
Montreal Protocol (MP) ODSs																						
MP ODSs (k tonnes)	12	9.6	7.8	7.6	7.1	6.0	5.0	4.8	4.8	5.0	4.5	4.1	4.5	3.9	3.6	2.9	3.2	3.0	3.1	3.3	3.3	3.1
ODP (k tonnes)	9.3	8.2	7.9	7.9	7.6	5.0	3.7	3.4	3.7	3.4	2.3	2.2	2.3	2.0	1.7	1.6	2.0	1.8	1.8	1.5	1.4	1.2
M tonnes CO ₂ -e	40	32	31	30	27	23	19	18	18	17	15	13	15	13	11	8.2	9.1	8.0	8.7	8.4	8.4	7.4
other chlorocarbons																						
dichloromethane	6406	6406	6406	6406	6406	4881	4137	3798	4154	3795	3650	2880	3402	3012	3930	2764	2408	1629	1348	2315	2361	2131
chloroform	5313	5989	6400	6544	4882	5046	4652	4792	4352	4515	3525	3377	3888	4324	4361	2394	2590	2217	3714	3253	3428	2706
PCE	3080	3080	3080	3080	3080	3080	3080	3080	3080	3080	3080	2646	2015	1784	2237	1333	1243	969	617	762	814	762
TCE																	316	288	239	202	199	177
total (k tonnes)	15	15	16	16	14	13	12	12	12	11	10	8.9	9.3	9.1	11	6.5	6.6	5.1	5.9	6.5	6.8	5.8
ODP tonnes	125	132	135	137	122	109	99	97	97	95	85	74	79	78	89	55	53	41	50	55	58	49
M tonnes CO ₂ -e	0.25	0.27	0.28	0.29	0.24	0.23	0.21	0.21	0.20	0.21	0.17	0.16	0.17	0.18	0.19	0.11	0.12	0.09	0.13	0.13	0.13	0.11
all ODSs																						
all ODS (k tonnes)	26	25	24	24	21	19	17	16	16	16	15	13	14	13	14	9.4	9.8	8.1	9.0	10	10	8.9
ODP k tonnes	9.4	8.3	8.0	8.0	7.7	5.2	3.8	3.5	3.8	3.5	2.4	2.2	2.4	2.1	1.8	1.7	2.1	1.9	1.9	1.6	1.4	1.3
M tonnes CO ₂ -e	40	32	31	30	27	23	19	18	18	17	15	13	15	13	11	8.3	9.3	8.1	8.8	8.6	8.5	7.5

5.1 CFCs

CFC-11 emissions have averaged about 460 tonnes from 1996 to 2016 (Table 3, Figure 12). CFC-11 emissions increased from 2003 (462 tonnes) to 2007 (739 tonnes) - the cause of this increase is unclear. Since 2007 CFC-11 emissions have declined to about 325 tonnes per year (2010-2016), an encouraging 46% lower than average 2004-2009 emissions (600 tonnes per year). CFC-11 emissions have been steadily declining for the past 4 years and reached 283 tonnes in 2016. It appears the long-term decline in emissions has resumed after being stalled for several years (2009-2013).

CFC-12 emissions have declined steadily since 1995 (1982 tonnes) to 235 tonnes by 2010, a long-term decline of 13% per year. CFC-12 emissions were 208 tonnes in 2016 and have averaged 239 tonnes from 2010-2016, 50% lower than average 2004-2009 emissions (486 tonnes per year). As with CFC-11 above, CFC-12 emissions have also been steadily declining for the last 4 years, suggesting the long-term decline has resumed after being stalled for several years (2009-2013).

CFC-113 emissions declined rapidly from 488 tonnes in 1995 to under 100 tonnes in the early 2000s, averaging close to 100 tonnes per year from 2002 to 2016. CFC-113 emissions in 2016 were 82 tonnes. CFC-113 emissions are quite variable and don't show a clear trend.

The pattern of emissions in Australian CFC-11, CFC-12 and CFC-113 emissions has changed since the last analysis with CFC-11 and CFC-12 showing that the long-term decline in emissions has resumed. CFC-12 emissions are likely largely from old (but still functional) and discarded refrigeration and air-conditioning equipment and aerosol cans, whereas CFC-11 and CFC-113 emissions are likely from existing and discarded aerosol cans and equipment with foam insulation.

Total Australian CFC (and HCFC, halon and carbon tetrachloride) emissions are shown in Figure 13. The overall decline in CFC emissions from 1995 (nearly 3400 tonnes) to 2016 (573 tonnes) is about 7.7% per year. ODP weighted CFC emissions have fallen from 3438 tonnes in 1995 to 589 tonnes in 2016 (declining by about 8% per year). Australian ODP-weighted CFC emissions in 2016 (589 tonnes) were 0.5% of global ODP-weighted CFC emissions. Australian ODP-weighted CFC emissions (589 tonnes) are currently (2016) about 45% of Australia's Montreal Protocol ODS (ODP-weighted) emissions (1300 tonnes).

CFC emissions are presumed to be from CFC-containing appliances/materials (refrigeration/ac equipment, foams, aerosol cans), either existing or buried (land-fills). If the current total emissions (\sim 573 tonnes per year) continue to decline at \sim 7.7%/yr, it will take about 22 years for Australian CFC emissions to drop below 100 tonnes/yr.

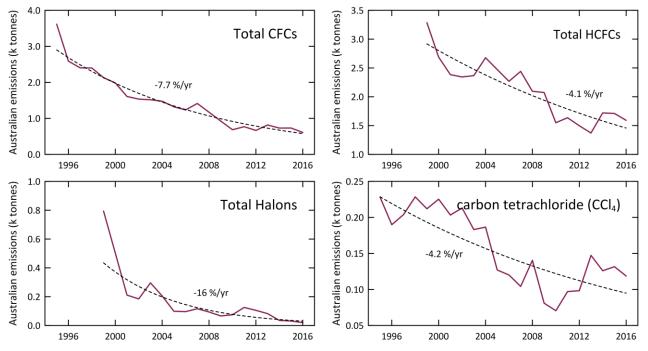


Figure 13. Declining Australian CFC, HCFC, halon and carbon tetrachloride emissions (k tonnes).

5.2 HCFCs

Total Australian HCFC-22 emissions have been estimated from Cape Grim data using ISC and NAME methods. Australian HCFC-22 emissions from operational and end-of-life refrigeration/air conditioning equipment has been estimated from an emissions model based on imports/consumption data for HCFC-22, estimates of the resultant HCFC-22 bank in the operational and retired equipment bank and assumptions about emission rates from the bank (Brodribb and McCann, 2015a).

There has been an overall decline in Australian HCFC-22 emissions estimated by ISC from about 2500 tonnes in 1999 to around 1300 tonnes in 2016 (2.5% per year over this period) (Table 3, Figure 12). HCFC-22 emissions declined rapidly from 2135 tonnes in 2004 to 1113 tonnes in 2013 (7% per year), and then increased again in 2014 and 2015 to 1432 and 1401 tonnes respectively, declining again in 2016 to 1293 tonnes.

Australian HCFC-22 emissions have been estimated by inverse modelling (NAME, Figure 12) for the period 2002-2011. The overall agreement with ISC estimates is good (better than 99%), with average NAME emissions being lower than ISC emissions pre-2007 and higher post-2007 (Figure 12).

The Australian HCFC-22 bank in operational refrigeration/air conditioning equipment peaked at ~13500 tonnes in 2007 declining by 40% to ~6700 tonnes in 2015 (Figure 11; Brodribb and McCann, 2013, 2014, 2015a). Over the same period emissions from the bank have fallen from 1300 tonnes in 2007 to 400 tonnes in 2015 (a 69% decline), an implied emission factor of 6%-10% per year. Some of the additional HCFC-22 emissions implied from the Cape Grim data could be from refrigeration/ac equipment and other HCFC-22-containing products (e.g. plastic foams, aerosol cans) in landfills.

Total HCFC-22 emissions from Cape Grim data and as projected leakage from the HCFC-22 bank are shown in Figure 14 as a function of the HCFC-22 bank. An assumed linear relationship between emissions and bank size, implies a bank emission factor from operational refrigeration equipment of 14%/yr. The difference between total HCFC-22 emissions and bank HCFC-22 emissions could be emissions from landfills (Figure 15) or other minor uses such as from foam, fire protection or aerosols. These emissions are relatively constant over the period 2002-2012 (approx. 500 tonnes) increasing to about 900 tonnes (2014-2015).

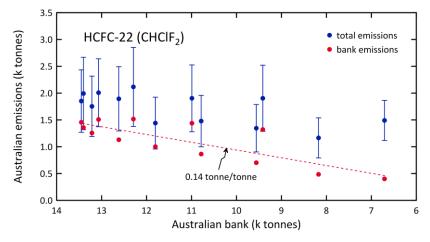


Figure 14. Total Australian HCFC-22 emissions (k tonnes) from Cape Grim data and HCFC-22 bank emissions as functions of the HCFC-22 bank (k tonnes; Brodribb and McCann, 2015a). The dashed line is a linear regression: slope = 0.14 tonne/tonne banked.

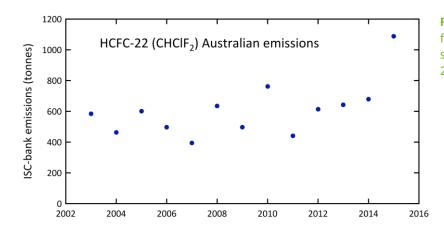


Figure 15. Possible HCFC-22 emissions from non-equipment bank sources such as landfills, increasing by about 25 tonnes per year.

HCFC-124 emissions have declined steadily from 262 tonnes in 1999 to 40 tonnes in 2016 (a decline of over 10% per year). Emissions levelled off from 2009-2013, staying steady at about 30 tonnes per year, declining to 19 tonnes in 2014 and increasing again to 20 tonnes in 2015 and 40 tonnes in 2016. It is unclear why HCFC-124 emissions doubled from 2015-2016 and will be explored in the next report.

HCFC-141b emissions have fluctuated over the period 1999-2016, falling from over 400 tonnes in 1999 to 260-320 tonnes in 2000-2008, falling again to 120 tonnes in 2010, then rising every year since to 255 tonnes in 2015, dropping again to 225 tonnes in 2016. Australian HCFC-141b emissions have been estimated by inverse modelling (NAME, Figure 12) for the period 2002-2011. The overall agreement with ISC estimates is good, with average NAME emissions being 6% lower than ISC emissions.

HCFC-142b emissions have also fluctuated over the period 1999-2016, falling from 100 tonnes in 1999 to 35 tonnes in 2005, rising to 65 tonnes in 2007, then declining each year to 31 tonnes in 2014. Emissions remain stable for the next two years at 35 tonnes (2015) and 33 tonnes (2016). Australian HCFC-141b emissions have been estimated by inverse modelling (NAME, Figure 12) for the period 2002-2011. The overall agreement with ISC estimates is good, with average NAME emissions being 4% lower than ISC emissions (similar to the HCFC-141b comparison).

Total HCFC emissions (Figure 13) have fallen by about 52% from 3,286 tonnes in 1999 to 1,591 tonnes in 2016, an overall decline of about 4% per year. ODP-weighted HCFC emissions have fallen from 196 tonnes in 1999 to about 100 tonnes per year, 2010-2016, 8% of Australia's Montreal Protocol ODS (ODP-weighted) emissions in 2016 (1200 tonnes). GWP-weighted HCFC emissions have fallen from 5.2 M tonnes CO₂-e in 1999 to 2.6 M tonnes CO₂-e in 2016, an overall decline of 50%.

Australian HCFC emissions are likely from a combination of service and malfunction leaks from existing refrigeration/ac equipment, from other minor uses and from land-fills. Australian HCFC emissions in 2016 (1591 tonnes) were 0.35% of global HCFC emissions (458 k tonnes) on a metric tonne basis.

5.3 Halons

Australian halon emissions are likely to originate from existing building fire-fighting systems (largely H-1301) still in service (but being phased-out) and existing portable fire extinguishers (largely H-1211), though now largely withdrawn from service. It may not be appropriate to directly scale the SE Australian halon emissions (as determined from Cape Grim data) to Australian emissions on a simple population basis. This is because the Melbourne/Port Philip region contains an additional likely halon source compared to other Australian regions, the National Halon Bank. The Bank collected about 5 tonnes of halon in 2014, with a collection/processing loss rate of less than 2% (0.01 tonne) and has stored a total of about 550 tonnes of halons by 2013 (140 tonnes H-1211, 410 tonnes H-1301) (E. Nigido, A-Gas (Australia) Pty. Ltd., personal communications, May 2014 and August 2015). Assuming that typical leaks from the storage of halons are less than 1% (Nigido 2015; we have assumed 0.5%), then halon emissions. The Australian halon emissions shown in Table 3 are the sum of Halon Bank emissions and Australian non-Halon Bank emissions. The SE Australian non-Halon Bank emissions are obtained from SE Australian Halon emissions (from Cape Grim data) by subtracting the Halon Bank emissions. Australian non-Halon Bank emissions equal SE Australian non-Halon Bank emissions.

It is assumed that the Halon Bank started to accumulate, process and store halons in 1993 and that the halon bank has grown linearly for both halons to reach current banked levels of 140 tonnes (H-1211) and 410 tonnes (H-1301), accumulation rates of approximately 7 and 20 tonnes per year.

Australian halon emissions fell from nearly 800 tonnes in 1999 to 200-300 tonnes in 2001-2004 and then to 66-125 tonnes in 2005-2013, before declining each following year to reach an all time low of 20 tonnes in 2016 (Figure 12, 13), an overall decline of about 16% per year. Approximately 60%-75% of Australian halon emissions over the period 2012-2016 are H-1211, 25%-40% H-1301. The increase in halon emissions seen in 2011, has not been maintained and emissions have declined each year since. SE Australian halon emissions are likely to show significant inter-annual variability.

Australian H-1211 emissions in 2016 were 15 tonnes, 0.45 % of global emissions (3300 tonnes) and Australian H-1301 emissions in 2016 were 5 tonnes, also 0.4% of global emissions (1200 tonnes). The Australian H-1211/H-1301 emissions ratio in 2016 was 3.0, compared to the global emissions ratio of 2.75.

5.4 Methyl bromide

Methyl bromide (MB) is used in Australia as a fumigant for cereals, such as wheat, and for cottonseed and timber logs prior to export and also as a soil sterilant, during the production of strawberry runners. The former uses are QPS, which are exempted from Montreal Protocol controls, and the latter use is non-QPS (n-QPS), which is restricted by the Montreal Protocol and for which Australia has to apply for a Critical Use Exemption (CUE) under the Montreal Protocol on an annual basis.

Today methyl bromide is imported into Australia predominantly for QPS use. 2/3 of QPS methyl bromide use in Australia is for timber log and wood chip fumigation (80% export, 20% import). The major timber log fumigation sites are Eden (60%, NSW), Geelong/Portland (40%, Vic) (personal communication, Mebrom Pty. Ltd., 2014). 1/3 of QPS methyl bromide use in Australia is for grain fumigation, largely export. Grain exported from SE Australian grain terminals account for about 20% of Australia's grain exports (National Transport Commission, 2008; GrainCorp, 2012). 100% of Australia's current n-QPS methyl bromide use occurs at Toolangi, NE of Melbourne, for growing strawberry runners.

A UNEP model of methyl bromide emissions suggests that 80-90 % of methyl bromide QPS use escapes to the atmosphere (UNEP, 2007). The UNEP methyl bromide emissions model may not be suitable for Australian methyl bromide consumption. It has been suggested (I. Porter, DPI Victoria, private communication) that close to 95% of QPS and about 50% of n-QPS methyl bromide are emitted to the atmosphere under Australian conditions, resulting in emissions of 600±30 tonnes in 2011-2015 (6% of global QPS plus n-QPS emissions, 9.9 k tonnes in 2012; Carpenter and Reimann, 2014). Note that both models do not take into account some recapture of methyl bromide which appears anecdotally to be increasing to meet local OH&S requirements.

The results from the modified UNEP model of Australian methyl bromide emissions are shown in Figure 16. SE Australian methyl bromide emissions from this model (assuming the speciation of SE Australian methyl bromide emissions from timber, grain exports and strawberry runner production discussed above) are compared to SE Australian emissions calculated from Cape Grim data by ISC in Figure 17 (also Figure 12). It is also worth noting that using imports in any given year as a basis for estimating emissions may bring in unrealistic year-to-year variability in calculated emissions as the amount of methyl bromide used in any year can be sourced from both imports and stock-in-hand. However, long term growth in methyl bromide stock is unlikely – in order to diminish the impact of short-term impacts of methyl bromide stock changes, 3-year average emissions from the model are also shown in Figure 15.

The emissions from the model and derived from atmospheric data for SE Australia show reasonable overall agreement (within 7%, model higher) over the period 1999-2015, and all model estimates of emissions (except for 1999) lie within the uncertainty range of the emissions estimated from Cape Grim observations. Prior to 2004, the model methyl bromide emissions were 20% higher than methyl bromide emissions derived from Cape Grim observations. The QPS and n-QPS emission factors in the model are assumed to be time-invariant. Lower emission factors before 2004 would help explain this difference; 2004 is also the time of a methyl bromide instrument change at Cape Grim. Both these factors are being investigated for possible impacts on pre-2004 emission data from Cape Grim observations. The emissions calculated from atmospheric data are consistent with the model emissions for SE Australia and it can be assumed that the model emissions for Australia are close to actual emissions.

Australian methyl bromide emissions based on the emissions model peaked at about 660 tonnes per year in 1999-2000, falling to 350 tonnes per year in 2006-2007, due to declining n-QPS methyl bromide use, increasing again to over 600 tonnes per year for 2012-2015, due to increased QPS methyl bromide use (Figure 16). The 383 ODP tonnes in 2015 are about 25% of Australia's Montreal Protocol ODS emissions in ODP tonnes.

Victorian methyl bromide consumption data for 2014 have been obtained from Mebrom Pty. Ltd. (K. Bartolo, private communication, 2014). Victorian methyl bromide consumption in 2014 was 170 tonnes, 125 tonnes in Melbourne (grain, wood products)/Toolangi (n-QPS), 35 tonnes in Portland (grain/wood) and 10 tonnes in Geelong (grain/timber). The methyl bromide consumption is similar to the emissions from Melbourne/Victoria calculated from Cape Grim data (160±60 tonnes in 2014). These data would imply that most of the methyl bromide consumed is emitted to the atmosphere.

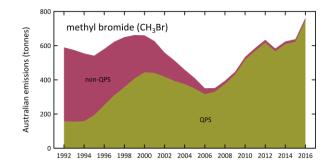


Figure 16. Australian methyl bromide emissions (3 year average) calculated from Australian methyl bromide import data and the modified UNEP emissions model (UNEP, 2007).

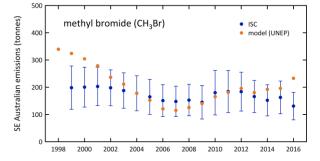


Figure 17. SE Australian methyl bromide emissions calculated (see text for details) by ISC from Cape Grim *in situ* AGAGE methyl bromide data (blue) and from a modified UNEP (2007) emissions model (orange, 3 year average) based on Australian methyl bromide imports and the SE Australian methyl bromide consumption for timber/grain exports and strawberry runner production.

5.5 Carbon tetrachloride and methyl chloroform (Montreal Protocol chlorocarbons)

Carbon tetrachloride (CTC) emissions are calculated from Cape Grim *in situ* GC-ECD data (reliable carbon tetrachloride data are not collected on the Cape Grim GC-MS Medusa instrument). The GC-ECD carbon tetrachloride data have been reprocessed for use with the recalculated Port Phillip CO emissions. The latest available estimates of Australian carbon tetrachloride emissions by ISC and NAME are shown in Figure 18.

Carbon tetrachloride emissions estimated by ISC were 228 tonnes in 1995, declining to below 100 tonnes per year for 2009-2012, before rising again to about 147 tonnes in 2013, declining again to 119 tonnes by 2016. Carbon tetrachloride emissions estimated using the NAME model were 198 tonnes in 2002, declining by 5% per year to 133 tonnes in 2011. Over the same period carbon tetrachloride emissions estimated by ISC fell by 8% per year. Overall ISC and NAME estimates of carbon tetrachloride emissions obtained from Cape Grim data agree to within 6% over this period, NAME higher.

The ISC and NAME estimates of Australian carbon tetrachloride emissions have been published in the peer-reviewed literature and used to identify possible 'missing' carbon tetrachloride sources on a global scale (Fraser *et al.*, 2014b). These findings have been incorporated into the *Scientific Assessment of Ozone Depletion: 2014* (Carpenter and Reimann, 2014) and the recent *SPARC Report on the Mystery of Carbon Tetrachloride* (Liang *et al.*, 2016). The Cape Grim data have been essential in identifying emissions of carbon tetrachloride not associated with carbon tetrachloride production and use. The carbon tetrachloride sources seen in the Cape Grim data are likely from landfills and/or chlor-alkali production. The Cape Grim data have encouraged new initiatives around the world in estimating these previously unaccounted-for emissions. The SPARC Report now estimates global carbon tetrachloride 'bottom-up' emissions of 25 k tonnes per year, whereas atmospheric data indicate emissions of 30-40 k tonnes per year. The gap between these emission estimates has closed to 5-15 k tonnes, significantly less than the 54 k tonne 'gap' reported in the last international scientific assessment of ozone depletion (Carpenter and Reimann, 2014).

Australian methyl chloroform emissions declined from about 3,500 tonnes in 1995 to 100 tonnes by 2004. Methyl chloroform emissions stabilised at about 100 tonnes per year from 2004-2012. In 2013-2016 Australian methyl chloroform emissions averaged about 120 tonnes per year, 4-8% of global emissions (1400-3100 tonnes) in 2013-2016. In 2016 Australian methyl chloroform emissions were 124 tonnes.

The combined carbon tetrachloride/methyl chloroform emissions totalled 600 ODP tonnes in 1995, declining to about 90 tonnes in 2010, an overall decline of about 12% per year. The combined carbon tetrachloride/methyl chloroform emissions rose to 174 ODP tonnes in 2013. In 2014-2016 Australian methyl chloroform/carbon tetrachloride emissions averaged around 150 ODP tonnes and were about 12% of Australian Montreal Protocol ODS emissions.

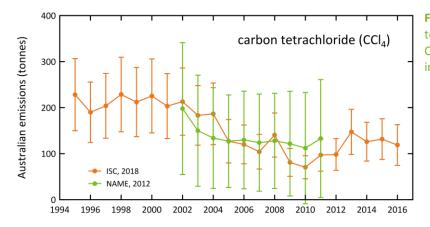


Figure 18. Australian carbon tetrachloride emissions calculated from Cape Grim *in situ* AGAGE data by ISC and inverse modelling (NAME).

5.6 Other chlorocarbons

Dichloromethane (CH_2Cl_2), chloroform ($CHcl_3$), CHClCCl_2 (trichloroethylene) and CCl_2CCl_2 (perchloroethylene) are shortlived ODSs whose production and consumption are not controlled by the Montreal Protocol. Significant emissions of all these ODSs are seen in the Cape Grim data (Figure 1). Australian dichloromethane emissions were about 6400 tonnes in 1999 declining by 10% per year to about 1400 tonnes in 2013, rising to 2361 tonnes in 2015, declining again to 2131 tonnes in 2016.

Australian chloroform emissions were over 5,300 tonnes in 1995 declining overall by 4% per year to about 2,217 tonnes in 2012, rising to over 3000 tonnes per year for the next 3 years and then declining to 2131 tonnes in 2016. There are large, natural soil emissions of chloroform and it is possible that the chloroform emissions calculated from Cape Grim data contain a large, natural, difficult to quantify, component.

Australian perchloroethylene emissions were over 3,100 tonnes in 2004 declining by about 18% per year to about 617 tonnes in 2013, increasing again 2014-2016, reaching 762 tonnes in 2016. trichloroethylene emissions were about 316 tonnes in 2011 falling by 11% per year to 177 tonnes in 2016.

Total emissions for these short-lived ODSs (not including trichloroethylene) were 11k tonnes (95 ODP tonnes) in 2004, falling by 5% per year to 5.8 k tonnes (49 ODP tonnes) in 2016. The ODP-weighted emissions of these short-lived ODSs were about 4% of total ODS emissions (ODP weighted).

5.7 GWP-weighted ODS emissions

The overall decline in GWP-weighted CFC emissions from 1995 (31 M tonnes CO₂-e) to 2016 (4.5 M tonnes CO₂-e) is 9% per year. Australian GWP-weighted CFC emissions in 2016 (4.5 M tonnes CO₂-e) were 0.9 % of Australia's total GHG emissions (525 M tonnes CO₂-e, including land use change, 2016). CFC emissions are not included in Australia's national GHG emissions, as CFCs have been phased-out under the Montreal Protocol and are therefore not subject to separate controls under the Kyoto Protocol and subsequent agreements. Nevertheless, the 26.5 M tonnes CO₂-e decline in GWP-weighted CFC emissions since 1995 is significant compared to other changes in Australian GHG emissions over the same period. Australian emissions of GHGs (CO₂, CH₄, N₂O, HFCs, PFCs, SF₆ including land use change), as reported to UNFCCC, have increased by a net 35 M tonnes CO₂-e from 1995 to 2016. The decrease in Australian CFC emissions alone over the same period (26.5 M tonnes CO₂-e) can be seen as negating 75% of this increase in Australia's reported net GHG emissions.

HCFC emissions, like CFC emission, are not included in Australia's national GHG emissions (525 M tonnes CO₂-e in 2016) as HCFCs are being phased-out under the Montreal Protocol and are therefore not subject to separate controls under the Kyoto Protocol and subsequent agreements. The overall decline in GWP-weighted HCFC emissions from 1999 (5.2 M tonnes CO₂-e) to 2016 (2.6 M tonnes CO₂-e) is 4% per year; CO₂-e weighted HCFC emissions in 2016 are 0.5% of Australia's reported net GHG emissions.

The CO_2 -e weighted emissions of other ODSs (halons, methyl bromide, chlorocarbons, other chlorocarbons) totalled 0.39 M tonnes in 2016, around 0.07% of Australia's reported net GHG emissions.

5.8 Total ODS emissions

Total Australian ODS emissions (CFCs, HCFCs, halons, methyl bromide, carbon tetrachloride, methyl chloroform, ODPand GWP-weighted), with and without 'other chlorocarbons' (see above) are shown in Figure 19. From 1995 the emissions are based on Cape Grim GC-ECD data for the major CFCs (CFC-11, -12, -113), methyl chloroform and carbon tetrachloride. From 1999 the emissions for all species are calculated from Cape Grim GC-ECD and GC-MS data, except for perchloroethylene whose GC-MS observations commenced in 2004 (1995 to 2003 perchloroethylene emissions are assumed equal to 2005 emissions) and trichloroethylene whose GC-MS observations commenced in 2011. In previous reports trichloroethylene observations were reported back to 2005, but these earlier trichloroethylene data have now been assessed as unreliable.

For the minor CFCs, HCFCs and halons it is assumed that annual emissions from 1995-1999 equal 1999 emissions. This could lead to a significant underestimate of emissions during this period (the ISC method is limited to the period when well-calibrated CO data were measured at Cape Grim - 1994 onwards). Methodologies are being investigated to extend ODS emissions estimates prior to 1994 – these will be evaluated in a future report.

The other chlorocarbons (dichloromethane, chloroform, perchloroethylene, trichloroethylene) make a very significant contribution to total Australian ODS emissions. Since 2011, 60%-70% of ODS emissions are from these other chlorocarbons. However they only make small contributions to ODP (2%-3%) or GWP (1%-2%) weighted emissions (or ozone depletion and climate change), because of their low ODPs and GWPs.

ODP-weighted ODS emissions fell by about 9% per year from 9.4 k tonnes in 1995 to 1.3 k tonnes in 2016. The largest decline is due to the halons, falling from emissions of 4.7 k tonnes (ODP) in 1999 to 0.09 k tonnes (ODP) in 2016, a fall of 4.6 k tonnes compared to a fall of 1.5 k tonnes (ODP) in CFC emissions and 0.1 k tonnes ODP in HCFC emissions over the same period.

GWP weighted ODS emissions fell by about 8% per year from 40 M tonnes CO_2 -e in 1995 to 7.5 M tonnes CO_2 -e in 2016. As discussed above the fall due to CFCs alone is 26.5 M tonnes CO_2 -e, more than 80% of the overall decline in GWP-weighted ODS emissions.

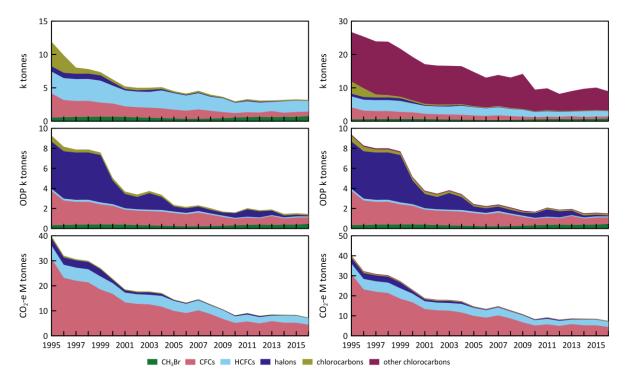


Figure 19. Total Australian ODS emissions; without other chlorocarbons (Section 5.6, left) and with other chlorocarbons (right).

The significance of the decline of GWP-weighted ODS emissions compared to GWP-weighted emissions of the GHGs reported to UNFCCC (CO₂, CH₄, N₂O, HFCs, PFCs, SF₆) are shown in Figure 20. The 32.5 M tonnes CO₂-e decline in GWP-weighted ODS emissions since 1995 is significant compared to other changes in Australian GHG emissions over the same period: as discussed above, Australian emissions of GHGs (carbon dioxide, methane, nitrous oxide, HFCs, PFCs, NF₃ and SF₆, including land use change emissions), as reported to UNFCCC, increased by a net 35 M tonnes CO₂-e from 1995 to 2016. The decrease in Australian ODS emissions negated nearly 93% of this increase and, if ODS emissions were included in Australia's GHG accounts, then Australia would record a net increase in GHG emissions of 2.5 M tonnes CO₂-e from 1995 to 2016, compared to the 35 M tonne increase as reported to UNFCCC.

Australia's GHG emissions, including land-use change, reached a peak of 617 M tonnes CO_2 -e in 2007 declining to 525 in 2016, a decline of 1.8% per year

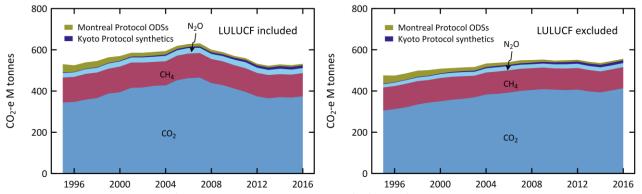


Figure 20. Australian emissions (GWP-weighted: M tonnes CO₂-e) of ODSs (Montreal Protocol species: CFCs, HCFC, halons, methyl bromide, methyl chloroform, carbon tetrachloride) and the GHGs reported to UNFCCC (carbon dioxide, methane, nitrous oxide, Kyoto Protocol synthetics: HFC, PFCs and sulfur hexafluoride), including and excluding GHG emissions due to land-use/land-use change and forestry (LULUCF).

Figure 21 shows a comparison of global and Australian CFC, HCFC and HFC emissions in M tonnes CO₂-e from atmospheric abundance data. There has been about a 72% decline in global CFC, HCFC and HFC emissions since their peak (9300 M tonnes CO₂-e) in the late 1980s and about a 57% decline since 1995. There has been a corresponding 60% decline in Australian CFC, HCFC and HFC emissions since 1995 (41 M tonnes CO₂-e). Australian ODS emissions are typically about 0.5% to 1% of global emissions. In 2016, global CFC, HCFC and HFC emissions are similar in their climate impact (32%, 32%, 36%), whereas in Australia HFC emissions have become dominant (49%) with CFCs at 32% and HCFCs at 19%. The decline in Australian CFC, HCFC and HFC emissions stalled in 2012 at about 15 M tonnes CO₂-e, declining slightly 13.9 M tonnes CO₂-e in 2016. Global CFC, HCFC and HFC emissions have been about 2400-2600 M tonnes CO₂-e since 2010.

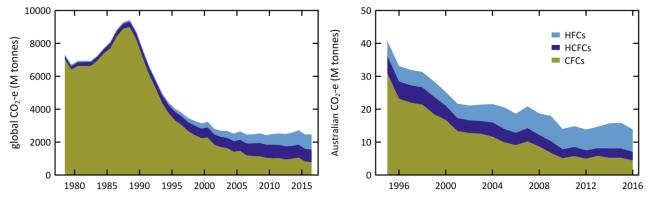


Figure 21. Global (since 1978) and Australian (since 1995) CFC, HCFC and HFC emissions (M tonne CO₂-e).

Summary

- CSIRO and collaborating laboratories measure the abundances and trends of thirty four (34) ODSs at Cape Grim, comprising twelve CFCs, eight HCFCs, four halons, seven chlorocarbons and three bromocarbons. Cape Grim has the most comprehensive list of measured ODSs anywhere in the world.
- As a result of measures undertaken within the Montreal Protocol framework, most of the CFCs measured in the
 atmosphere at Cape Grim in 2017 have stopped growing or are in decline, the exceptions being CFC-13, CFC-113a
 and CFC-115, which are growing slowly. Total CFCs in the background atmosphere declined by 0.5% (2016-2017),
 as did chlorine from CFCs.
- The atmospheric abundance of all of the major HCFCs measured in the atmosphere at Cape Grim were growing in 2015, but now (2016) HCFC-22 is the only one growing (2% per year). HCFC-141b and HCFC-141b are declining by 0.41% and 0.05% per year respectively. The growth rate of total HCFCs is slowing down. Total HCFCs increased by 4.2 ppt or 1.5% per year (2016-2017).). Chlorine in the atmosphere from HCFCs (301 ppt in 2017, 9% of total chlorine from all ODSs) increased by 4.2 ppt (1.4%, 2016-2017), the only ODS sector showing an increase in chlorine (apart from methyl chloride and dichloromethane).
- The chlorocarbons account for 28% of total chlorine from all ODSs in the background atmosphere. Carbon tetrachloride, methyl chloroform and chloroform measured at Cape Grim showed decreasing concentrations in 2016 compared to 2015, whereas methyl chloride, dichloromethane, trichloroethylene and perchloroethylene all showed increasing concentrations. The largest decrease in the chlorocarbons was 0.4 ppt (16.6%) for methyl chloride increase in the chlorocarbons was 3.0 ppt (20.7%) for dichloromethane. Total chlorine from chlorocarbons increased by 8.8 ppt (1%), much higher than the previous years increase of 1.3 ppt (0.1%). This increase in chlorine from chlorocarbons is due predominately to dichloromethane.
- H-1211, H-2404 and H-1301 continued to show declining concentrations from 2016 to 2017. Overall halons are in decline by 1.7% per year (compared to 2%/yr 2015-2016). Overall bromine from halons is in decline by 1.6% per year (0.12 ppt/yr).
- Methyl Bromide showed a large decrease (0.6 ppt/yr 2016-2017) in the background atmosphere. There is an overall long-term decrease in methyl bromide in the atmosphere since about 2000, which briefly halted in 2012-2013 and 2014-2015.
- Total chlorine from all ODSs increased by 0.11% (3 ppt, 2016-2017). This is only the second time in 20 years that there has been an increase in total chlorine from ODSs (2007-2008 increase of 2 ppt).
- Total bromine from all ODSs decreased 6.8% (1.5 ppt, 2016-2017)
- Global emissions of most of the major MP ODSs have now stopped growing or are in decline.
- Global CFC-11 emissions increased from 71 k tonnes in 2015 to 73 k tonnes in 2016. There has been no significant decline in CFC-11 emissions since 2002, whereas all other CFC emissions have shown a long-term decline
- Global CFC-12 emissions continue to decline from 44 k tonnes in 2015 to 33 k tonnes in 2016, continuing a remarkably monotonic decrease in emissions since the late-1980s. The 2014-2015 and 2015-2106 decline in emissions (11 k tonnes) is the largest annual decline in CFC-12 emissions since 2005-2006 (14 k tonnes).
- Global emissions of the other CFCs (CFC-113, -114, -115), including the minor CFCs, increased from a total of 5.3 k tonnes in 2015 to 11.1 k tonnes in 2016. This increase of 5.8 ktonnes is the largest annual increase since 2004-2005 (7.8 ktonnes). Most of the increase is due to emissions of CFC-114.
- Global carbon tetrachloride emissions declined from 35 k tonnes in 2015 to 34 k tonnes in 2016. Long-term, global carbon tetrachloride emissions have declined slowly from a peak of around 120 k tonnes/yr in the late 1970s, declining to 50 k tonnes/yr by 2005, a long-term decline of about 3%/yr. Since 2005 global carbon tetrachloride emissions have declined steadily by about 1-2 k tonne per year (3%/yr) until 2016. The decline in global carbon tetrachloride emissions is not as rapid as anticipated under the Montreal Protocol
- Global methyl bromide emissions estimates start in 2000 (196 k tonnes), declining steadily to 149 k tonnes in 2016, a long-term decline of about 1.5% per year. methyl bromide emissions from 2013-2016 are fairly steady, ranging from 147-151 ktonnes suggesting the long-term decline in methyl bromide emissions may have levelled off in line with developing country phase-out in 2015.
- Australian imports of ODSs (in tonnes) increased by 34% from 2016 to 2017, due to methyl bromide imports rising from 691 tonnes (2016) to 945 tonnes (2017), all for QPS applications.

- Australian CFC emissions, based on Cape Grim data, have decreased by 17% from 2015 (691 tonnes) to 2016 (573 tonnes), 7.7% per year since 1995. Australian ODP-weighted CFC emissions in 2016 (589 tonnes) were 0.5% of global ODP-weighted CFC emissions. Australian ODP-weighted CFC emissions (589 tonnes) are currently (2016) about 45% of Australia's Montreal Protocol ODS (ODP-weighted) emissions (1300 tonnes).
- Total HCFC emissions (Figure 13) have fallen by about 52% from 3,286 tonnes in 1999 to 1,591 tonnes in 2016, an overall decline of about 4% per year. ODP-weighted HCFC emissions have fallen from 196 tonnes in 1999 to about 100 tonnes per year, 2010-2016, 8% of Australia's Montreal Protocol ODS (ODP-weighted) emissions in 2016 (1200 tonnes). GWP-weighted HCFC emissions have fallen from 5.2 M tonnes CO₂-e in 1999 to 2.6 M tonnes CO₂-e in 2016, an overall decline of 50%.
- Australian halon emissions fell from nearly 800 tonnes in 1999 to 200-300 tonnes in 2001-2004 and then to 66-125 tonnes in 2005-2013, before declining each following year to reach an all time low of 20 tonnes in 2016, an overall decline of about 16% per year. Approximately 60%-75% of Australian halon emissions over the period 2012-2016 are H-1211, 25%-40% H-1301. The increase in halon emissions seen in 2011, has not been maintained and emissions have declined each year since
- Australian methyl bromide (MB) emissions based on imports and an emissions model peaked at about 660 tonnes per year in 1999-2000, falling to 350 tonnes per year in 2006-2007, due to declining n-QPS methyl bromide use, increasing again to over 600 tonnes per year for 2012-2015, due to increased QPS methyl bromide use. The 383 ODP tonnes in 2015 are about 25% of Australia's Montreal Protocol ODS emissions in ODP tonnes. The emissions from the model and derived from atmospheric data for SE Australia show reasonable overall agreement (within 7%, model higher) over the period 1997-2015, and all model estimates of emissions (except for 1999) lie within the uncertainty range of the emissions estimated from Cape Grim observations.
- Australian carbon tetrachloride emissions estimated from Cape Grim data were 228 tonnes in 1995, declining to below 100 tonnes per year for 2009-2010, before rising again to about 147 tonnes in 2013, declining again to 119 tonnes by 2016. The carbon tetrachloride sources seen in the Cape Grim data are likely from landfills and/or chloralkali production.
- GWP weighted ODS emissions fell by about 8% per year from 40 M tonnes CO₂-e in 1995 to 7.5 M tonnes CO₂-e in 2016. The fall due to CFCs alone is 26.5 M tonnes CO₂-e, more than 80% of the overall decline in GWP-weighted ODS emissions. The 26.5 M tonnes CO₂-e decline in GWP-weighted CFC emissions since 1995 is significant compared to other changes in Australian GHG emissions over the same period. Australian emissions of GHGs (CO₂, CH₄, N₂O, HFCs, PFCs, SF₆ including land use change), as reported to UNFCCC, have increased by a net 35 M tonnes CO₂-e from 1995 to 2016. The decrease in Australian CFC emissions alone over the same period (26.5 M tonnes CO₂-e) can be seen as negating 75% of this increase in Australia's reported net GHG emissions.
- In Australia (as elsewhere) the Montreal Protocol has been very effective in controlling the consumption, and therefore the emissions, of ODSs that cause stratospheric ozone depletion to the extent that ozone recovery is being detected at mid- and polar latitudes. In addition, the reduction in emissions of ODSs has significantly slowed the growth in overall GHG emissions that drive climate change.

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