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Australian and Global Emissions of Ozone Depleting Substances

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CSIRO Oceans and Atmosphere, Climate Science Centre

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Contents

Figures

Tables

Executive summary

- CSIRO and collaborating laboratories measure the abundances and trends of thirty-four (34) ozone depleting substances (ODSs) at Cape Grim, comprising twelve chlorofluorocarbons (CFC-11, -12, -13, -112, -112a -113, -113a, -114, -114a, -115, -216ba, -216ca), eight hydrochlorofluorocarbons (HCFC-22, -31d, -124, -132b, -133a, -141b, -142b, -225ca) four halons (-1201, -1202, -1301, -2402) , seven chlorocarbons (CH_3Cl, CH_2Cl_2, $CHCl_3$, CCl_4, CH_3CCl_3, $CHClCCl_2$, CCl_2CCl_2) and three bromocarbons (CH_3Br, $CHBr_3$, CH_2Br_2). Cape Grim has the most comprehensive list of measured ODSs anywhere in the world.
- 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 over the Antarctic.
  - Total chlorine from all ODSs decreased by 0.3% (10 ppt, 2019-2020). This total is 10% lower than its peak value in 1994.
  - Total bromine from all ODSs increased 2.5% (0.51 ppt, 2019-2020). This total is 15% lower than its peak value in 1998. The variability in total bromine compared to total chlorine is relatively large due to natural contributions.

Measurements of ozone depleting substances in the 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 and CFC-113) measured in the atmosphere in 2020 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 measured in the atmosphere are declining by 0.6% per year due to declining emissions; chlorine from CFCs in the atmosphere decreased by 13 ppt (0.7% from 2019 to 2020) slightly more than the previous annual decline.
- The atmospheric abundance of HCFC-22 measured in the atmosphere at Cape Grim is currently (2019-2020) growing at 1% per year. HCFC-141b increased by 0.3% from 2019 to 2020, after declining for two years. HCFC-142b is currently declining by 0.6% per year. The growth rate of total HCFCs is slowing down but still increased by 1.9 ppt or 0.7% from 2019 to 2020. Chlorine in the atmosphere from HCFCs (309 ppt in 2020, 10% of total chlorine from all ODSs) increased by 2 ppt (0.7% from 2019 to 2020), the only ODS sector showing an increase in chlorine.
- Carbon tetrachloride and methyl chloroform are chlorocarbons controlled by the Montreal Protocol. Carbon tetrachloride and methyl chloroform measured at Cape Grim showed decreasing concentrations in 2020 compared to 2019. The largest decrease in the chlorocarbons was 0.21 ppt (14%) for methyl chloroform.
- Methyl chloride, dichloromethane, perchloroethylene and trichloroethylene are chlorocarbons not controlled by the Montreal Protocol. Methyl chloride and dichloromethane measured at Cape Grim showed increasing concentrations in 2020 compared to 2019, whereas perchloroethylene and trichloroethylene showed decreasing concentrations.

- Overall halons are in decline by 1.4% per year (compared to 1.3%/yr 2018-2019), a larger rate of decline (in percentage terms) compared to the CFCs. Bromine in the atmosphere from halons decreased by 0.11 ppt/yr (1.4% from 2019 to 2020). H-1301 is now the most abundant halon in the background atmosphere (3.3 ppt, 2020), followed by H-1211 (3.2 ppt). Halons H-1211 and H-2402 are in decline in the atmosphere (-0.1 ppt/yr and -0.006 ppt/yr respectively, 2019-2020). H-1301 growth rates have decreased slightly from (0.009 ppt from 2018 to 2019) to 0.007 ppt from 2019 to 2020.
- Methyl Bromide showed a small increase (0.13 ppt from 2019 to 2020) in the background atmosphere however there is an overall long-term decrease in methyl bromide in the atmosphere since about 2000.

Estimated global emissions of ozone depleting substances

- Global ODS emissions have been calculated using background ODS observations at a number of AGAGE stations in the Northern and Southern Hemispheres up to 2019. 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).
- Global CFC-11 emissions averaged about 78 k tonnes/yr (2016-2018), an increase of 14 k tonne per year compared to the 2008-2012 period. Global CFC-11 emissions decreased significantly to 52 k tonnes in 2019 most likely due to the recent decline in CFC-11 emissions from eastern China (Park *et al*., 2021).
- Since the peak emissions of global CFC-12 in the late 1980s (513 k tonnes, 1988), CFC-12 emissions have declined by 96% (10%/yr). Current global CFC-12 emissions are about 22 k tonnes/yr (2019).
- Global emissions of the other CFCs (CFC-113, -114, -115), including the minor CFCs, decreased from a total of 276 k tonnes in 1989 to 12 k tonnes in 2019 (10%/yr). Most of the decrease is due to a decline in emissions of CFC-113.
- Global HCFC emissions peaked in 2010 (488 k tonnes) and have since declined (10%) to 441 k tonnes in 2019, a decline of about 1%/yr.
- Long-term, global carbon tetrachloride emissions have declined slowly from a peak of around 120 k tonnes/yr in the late 1970s to 38 k tonnes/yr in 2019. The decline in global carbon tetrachloride emissions is not as rapid as modelled under the Montreal Protocol.
- Global methyl bromide emissions decline from 182 k tonnes in 1999 to 127 k tonnes in 2019, a decline of about 1.8%/yr.

Estimated Australian emissions of ozone depleting substances

- Australian emissions of CFCs, HCFCs, methyl chloroform, carbon tetrachloride, halons and methyl bromide have been detected in the Cape Grim data. The emissions have been quantified using two techniques, interspecies correlation (ISC), and inverse modelling using the UK Meteorological Office transport model NAME (Numerical Atmospheric Dispersion Modelling Environment) coupled to the inversion model InTEM (Inversion Technique for Emission Modelling).
- The overall decline in Australian CFC emissions from 1995 (3,627 metric tonnes) to 2019 (438 metric tonnes) is about 8% per year, however Australian CFC emissions increased

from 2018 (392 metric tonnes) to 2019 (438 metric tonnes). Australian ODP-weighted CFC emissions are currently (2019) about 38% of Australia's total ODS (ODP-weighted) emissions (1100 tonnes).

- Total HCFC emissions have fallen by about 80% from 3,286 metric tonnes in 1999 to 657 metric tonnes in 2019, an overall decline of about 6.5% per year. ODP-weighted HCFC emissions have fallen from 196 tonnes in 1999 to 45 tonnes in 2019, 4% of Australia's total ODS (ODP-weighted) emissions in 2019 (1100 tonnes).
- Australian halon emissions fell from nearly 800 tonnes in 1999 to 13 tonnes in 2019, an overall decline of about 19% per year. The majority of Australian halon emissions over the period 2012-2019 are H-1211.
- Australian methyl bromide emissions estimations are based on imports and peaked at about 600 metric tonnes per year in 1999-2000, falling to 300 metric tonnes per year in 2006-2007, due in part to declining non-QPS methyl bromide use, and also a decline in QPS use (possibly due to reduced grain production as a result of the severe drought in SE Australia), increasing again to 630 metric tonnes in 2019, due to increased QPS methyl bromide use resulting from grain harvests recovering, trade patterns and trade partner requirements. The 378 ODP tonnes of methyl bromide emitted in 2019 are about 34% of Australia's total ODS emissions in ODP tonnes.
- Carbon tetrachloride emissions were estimated at 227 tonnes in 1995, declining to 93 tonnes by 2019, a decline of 4% per year. The carbon tetrachloride sources seen in the Cape Grim data are likely from landfills and/or chlor-alkali production.
- Total Australian ODP weighted emissions of ODS controlled by the Montreal Protocol fell by about 9% per year from 9.4 k tonnes in 1995 to 1.1 k tonnes in 2019. The largest decline is due to the halons, with emissions of 4.7 k tonnes (ODP) in 1999 falling to 0.07 k tonnes (ODP) in 2019, a fall of 4.63 k tonnes compared to a fall of 3 k tonnes (ODP) in CFC emissions and 0.15 k tonnes ODP in HCFC emissions over the same period.
- Total Australian GWP weighted emissions of ODS controlled by the Montreal Protocol fell by about 7% per year from 43 M tonnes CO_2-e in 1995 to 8 M tonnes CO_2-e in 2019. The fall due to CFCs alone is 28 M tonnes CO_2-e, 80% of the overall decline in GWP-weighted ODS emissions. The 28 M tonnes CO_2-e decline in GWP-weighted CFC emissions since 1995 is significant compared to other changes in Australian GHG emissions over the same period.

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Introduction

Chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), halons, carbon tetrachloride (CTC: CCl_4), methyl chloroform (MC: CH_3CCl_3) and methyl bromide (MB: CH_3Br) are all ozone depleting substances (ODSs), whose production and consumption, and resulting emissions, have been significantly reduced by national actions taken by countries to comply with the Montreal Protocol. These reductions in ODSs have led to the start of the recovery of stratospheric ozone (WMO 2018).

Methyl chloride (CH_3Cl), dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$), trichloroethylene (TCE: $CHClCl_2$), perchloroethylene (PCE: CCl_2CCl_2), dibromomethane (CH_2Br_2), bromoform ($CHBr_3$) and methyl iodide (CH_3I) are not listed as ODSs in a Montreal Protocol context. 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 & 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 (Fraser *et al*. 2014a; Engel & Rigby 2018).

The most dramatic demonstration of the environmental effect of EESC is the annual appearance of the Antarctic ozone hole (AOH; Klekociuk *et al*. 2021 and earlier papers) in the southern hemisphere spring. In a companion report (Krummel *et al*. 2021 and earlier reports) we review the development and decline of the 2020 AOH and review its metrics in light of the ongoing decline of EESC in the atmosphere. In addition, ODS emissions play an important role globally in contributing to radiative forcing as many of them are also greenhouse gases (GHGs), and reductions in ODSs have helped reduce the human contribution to climate change over the past 30 years (Carpenter & Reimann 2014; Engel & Rigby 2018).

CFCs, halons, carbon tetrachloride and methyl chloroform are no longer imported into Australia in any significant quantities; however small amounts can still be used for essential purposes. 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 by extrapolation from all 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. ODS emissions are not controlled by the Montreal Protocol. However, ODS emissions in Australia are controlled by the Australian government as it implements additional measures to reduce emissions – 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 the capture of used ODS substances, followed by their recycling or destruction.

HCFCs and methyl bromide are still imported into Australia and used in maintaining existing HCFC-containing refrigeration and air conditioning equipment, for quarantine and pre-shipment

(QPS) uses of methyl bromide, one exempted non-QPS use of methyl bromide (for growing strawberry runners) and fire protection applications. HCFC and non-QPS methyl bromide consumption is controlled by Australia's commitments under the Montreal Protocol, whereas QPS-uses of methyl bromide are not subject to phase-out under the Montreal Protocol and consequently methyl bromide emissions from QPS-use can vary from year to year.

Dichloromethane, trichloroethylene and trichloroethylene are imported into Australia and are mainly used as industrial solvents. 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).

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

The concentrations of synthetic and naturally occurring ODSs have been measured in the background (baseline) Southern Hemisphere atmosphere on air samples captured at Cape Grim, Tasmania. For more detail on the Cape Grim *in situ* and air archive measurements see Appendix A.

The synthetic and natural ODSs have been measured by CSIRO at Cape Grim since:

- 1976 (CFC-11, carbon tetrachloride, methyl chloroform)
- 1978 (CFC-12, chloroform)
- early 1980s (CFC-113)
- late-1990s (CFC-114, CFC-115, several HCFCs, halons)
- mid-2000s (several more HCFCs, methyl chloride, dichloromethane, trichloroethylene, perchloroethylene, methyl bromide, dibromomethane and bromoform).

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 and used extensively in reports to the Montreal Protocol. Key references for each species are listed in Appendix A

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 (2019-2020) and Figure 1 (1978-2020).

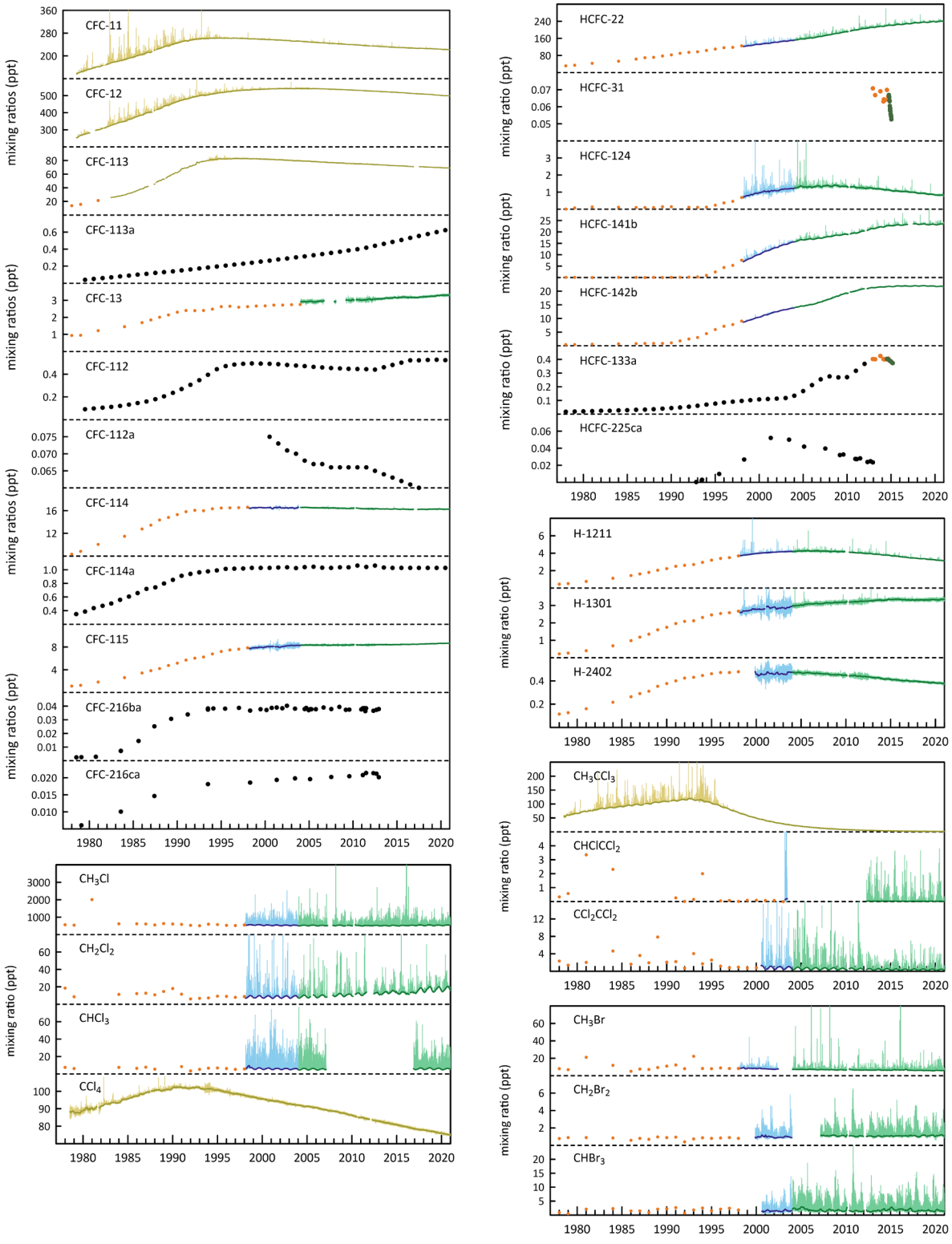
Table 1. Southern Hemisphere concentrations (2019, 2020) and growth rates (2019–2020) for CFCs, HCFCs, halons, carbon tetrachloride, methyl chloroform, methyl chloride, dichloromethane, chloroform, trichloroethylene, perchloroethylene, methyl bromide, dibromomethane and bromoform measured *in situ* at Cape Grim, Tasmania and/or in the Cape Grim Air Archive (CSIRO unpublished data).

Species	Concentration ppt				Growth ppt/yr	Growth %/yr
	2017	2018	2019	2020	2019-2020	2019-2020
CFCs						
CFC-11	226.4	225.7	224.3	222.4	-1.9	-0.84
CFC-12	510.6	507.8	503.8	499.9	-3.9	-0.78
CFC-13	3.2	3.2	3.3	3.3	0.03	1.05
CFC-112	0.52	0.53	0.53	0.52	0.00	-0.65
CFC-112a	0.06	0.06	0.06	0.06	0.00	-1.7
CFC-113	70.9	70.4	69.8	69.3	-0.52	-0.74
CFC-113a	0.55	0.58	0.60	0.62	0.02	3.8
CFC-114	16.2	16.3	16.3	16.3	-0.02	-0.10
CFC-114a	-0.9	0.1	1.1	2.1	1.00	62.5
CFC-115	8.5	8.6	8.6	8.6	0.03	0.36
CFC-216ba	0.034	0.033	0.032	0.031	0.00	-2.3
CFC-216ca	0.020	0.020	0.019	0.019	0.00	-1.1
total CFCs	**836.1**	**833.2**	**828.4**	**823.2**	**-5.3**	**-0.64**
total CFC (Cl)	**1959.6**	**1952.3**	**1940.8**	**1927.9**	**-13.0**	**-0.67**
HCFCs						
HCFC-22	231.2	234.7	237.2	239.2	2.1	0.9
HCFC-31						
HCFC-124	1.0	0.9	0.9	0.8	-0.05	-5.6
HCFC-132b	0.09	0.10	0.11	0.11	0.01	6.4
HCFC-133a	0.42	0.43	0.42	0.41	-0.01	-2.5
HCFC-141b	23.5	23.4	23.3	23.3	0.06	0.27
HCFC-142b	21.9	21.9	21.9	21.7	-0.13	-0.58
HCFC-225ca	0.005	0.001	-0.003	-0.007	0.00	81.7
total HCFCs	**278.1**	**281.5**	**283.7**	**285.7**	**1.9**	**0.7**
total HCFC (Cl)	**301.8**	**305.0**	**307.1**	**309.1**	**2.0**	**0.7**

Species	Concentration ppt				Growth ppt/yr	Growth %/yr
	2017	2018	2019	2020	2019-2020	2019-2020
Halons						
H-1202	-1.98	-0.98	0.02	0.02	0.00	0.0
H-1211	3.4	3.4	3.3	3.2	-0.10	-3.1
H-1301	3.3	3.3	3.3	3.3	0.007	0.22
H-2402	0.4	0.4	0.4	0.4	-0.006	-1.4
total halons	**5.2**	**6.1**	**7.0**	**6.9**	**-0.10**	**-1.4**
total halon (Cl)	**3.4**	**3.4**	**3.3**	**3.2**	**-0.10**	**-3.1**
total halon (Br)	**3.6**	**5.5**	**7.4**	**7.3**	**-0.11**	**-1.4**
Other ODSs						
carbon tetrachloride	78.1	77.4	76.4	75.4	-1.01	-1.3
methyl chloroform	2.2	1.9	1.6	1.4	-0.21	-14.1
methyl chloride	532.3	531.1	526.8	532.5	5.7	1.1
dichloromethane	15.8	17.1	17.1	17.4	0.4	2.2
chloroform	5.9	6.0	5.8	5.7	-0.12	-2.1
TCE	0.021	0.018	0.019	0.018	-0.001	-4.1
PCE	0.36	0.36	0.35	0.33	-0.016	-4.7
total other Cl-ODSs	**634.7**	**633.8**	**628.1**	**632.8**	**4.7**	**0.7**
total other Cl-ODSs (Cl)	**902.1**	**899.9**	**890.4**	**891.7**	**1.3**	**0.1**
methyl bromide	6.1	6.0	5.8	6.0	0.13	2.2
dibromomethane	1.1	1.1	1.1	1.1	0.029	2.60
bromoform	1.7	1.6	1.5	1.6	0.14	9.1
total other Br-ODSs	**8.9**	**8.7**	**8.4**	**8.7**	**0.30**	**3.5**
total other Br-ODSs (Br)	**13.4**	**12.9**	**12.5**	**13.1**	**0.61**	**4.8**
total Cl	**3166.9**	**3160.6**	**3141.6**	**3131.9**	**-9.74**	**-0.31**
total Br	**17.0**	**18.4**	**19.9**	**20.4**	**0.51**	**2.5**

[a] AGAGE 'CFC-114' = CFC-114+CFC-114a [b] from 2012 concentration and growth rate [c] from 2014 concentration and growth rate [d] measured 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 – 2019) 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.



1.1 CFCs

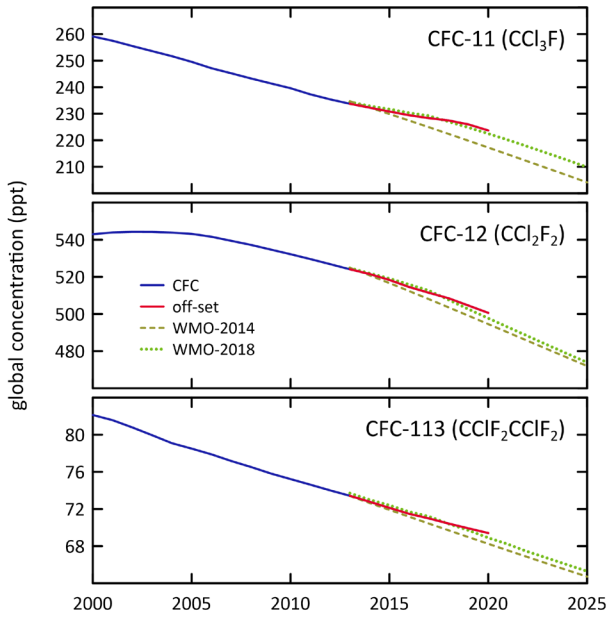
CFC-11 and CFC-12 are the dominant CFCs in the atmosphere, constituting 88% of all CFCs in 2020. CFC-113, CFC-114 and CFC-115 constitute 11% of CFCs, the remaining minor CFCs about 1%. The CFCs account for 62% of chlorine from all ODSs in the background atmosphere.

As a result of measures undertaken within the Montreal Protocol framework, the majority of the CFCs (CFC-11, CFC-12, CFC-112, CFC-112a and CFC-113) measured in the atmosphere in 2020 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 measured in the atmosphere are declining by 0.6% per year due to declining emissions; chlorine from CFCs in the atmosphere decreased by 13 ppt (2019-2020, 0.7%) slightly more than the decline in 2018-2019.

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 2020 is 1.25 ppt, while the total CFCs in the background atmosphere sum to 823 ppt in 2020. The summed growth of these minor CFCs in the atmosphere is 0.02 ppt/yr. These 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 slowed down from -1.67 ppt/yr (2012-2013) to -0.9 ppt/yr (2017-2018). This slow-down in the decline of CFC-11 has been attributed to new likely illegal production of CFC-11 and subsequent emissions from east Asia, around 50 % of which is attributed to China (Montzka *et al*. 2018; Rigby *et al*. 2019). This slow-down in the decline of CFC-11 in the atmosphere appears to be short lived with the rate of decline for 2019-2020 increasing to -2.3 ppt/yr. The rate of decline for CFC-12 has increased from, -2.79 ppt/yr (2012-2013) to -4.1 ppt/yr (2019-2020). The rate of decline for CFC-113 has decreased from -0.59 ppt/yr (2012-2013) to -0.38 ppt/yr (2019-2020).

Figure 2. Global concentrations of CFC-11, CFC-12 and CFC-113 (ppt) and WMO A1 scenarios (dashed lines; Harris & Wuebbles, 2014; Carpenter & Daniel, 2018).



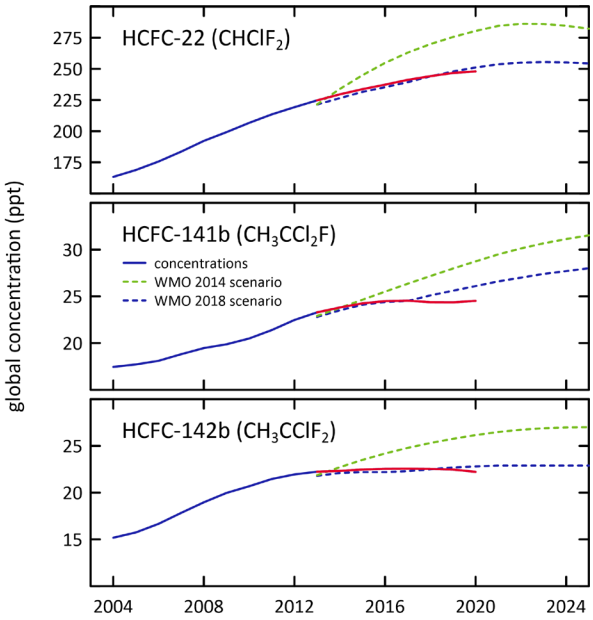
1.2 HCFCs

The major HCFCs measured in the atmosphere in 2020 at Cape Grim are HCFC-22, HCFC-141b and HCFC-142b. HCFC-22 is currently growing at 1% per year, however its growth rate has been declining since around 2010. HCFC-141b increased by 0.3% 2019 to 2020, after declining for two years. HCFC-142b is currently declining by 0.6% per year and its growth rate has also been declining since 2010. The dominant HCFC is HCFC-22 (239.2 ppt in 2020), constituting 84% 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 1.9 ppt or 0.7% per year (2019-2020) compared to 2.1 ppt per year (2018-2019). Chlorine in the atmosphere from HCFCs (309 ppt in 2020, 10% of total chlorine from all ODSs) increased by 2.0 ppt (0.7%, 2019-2020), the only ODS sector showing an increase in chlorine.

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 0.41 ppt in 2020). 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 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 2020 annual mean HCFC-132b concentration was 0.11 ppt, increasing by 6.4%/yr (2019-2020).

Global concentrations of the most abundant HCFC, HCFC-22, continue to grow in the atmosphere (Figure 3), however the rate of increase has declined almost every year since 2008. The growth rate for HCFC-22 has slowed down from 8.6 ppt/yr (2007-2008) to 1.3 ppt/yr (2019-2020). The growth rate for HCFC-141b decreased from 1.08 ppt/yr (2011-2012) to 0.16 ppt/yr (2019-2020) and concentrations increased from 2019-2020 after two years of showing a decline. The growth rate for HCFC-142b has also declined substantially from 1.17 ppt/yr (2006-2007) to -0.25 ppt/yr (2019-2020) and concentrations for HCFC-142b have declined for the third year in a row. The concentrations of all three HCFCs (Figure 3) are much lower than the WMO 2014 A1 scenario (Figure 3) from around 2013 onwards. This scenario assumed that after 2012, all Article 5 countries would continue producing HCFCs at the maximum level allowed under the Montreal Protocol (Engel & Rigby, 2018). HCFC-22 and HCFC-142b track well with the more recent WMO 2018 scenarios, and HCFC-141b is lower than the WMO 2018 scenario from around 2018 onwards (Figure 3).

Figure 3. Global concentrations of HCFC-22, HCFC-141b and HCFC-142b (ppt) and WMO A1 scenarios (dashed lines; Harris & Wuebbles, 2014; Carpenter & Daniel 2018).



1.3 Chlorocarbons

The chlorocarbons measured at Cape Grim are: Carbon tetrachloride, methyl chloroform, chloroform, trichloroethylene and perchloroethylene, methyl chloride and dichloromethane. Of these, carbon tetrachloride and methyl chloroform are controlled under the Montreal Protocol.

The chlorocarbons account for 28% of total chlorine from all ODSs (HCFCs, CFCs, halons and chlorocarbons) in the background atmosphere at Cape Grim. The most abundant chlorocarbon in the background atmosphere is the largely naturally occurring methyl chloride (CH_3Cl, 532.5 ppt, 2020), accounting for 84% of all chlorocarbons and 60% of chlorine from chlorocarbons. The next most abundant chlorocarbon is anthropogenic carbon tetrachloride (75.4 ppt, 2020), accounting for 34% of chlorine from chlorocarbons. The remaining minor chlorocarbons, including methyl chloroform, contribute 6% of chlorocarbon chlorine.

Carbon tetrachloride, methyl chloroform, chloroform, trichloroethylene and perchloroethylene measured at Cape Grim showed decreasing concentrations in 2020 compared to 2019, whereas methyl chloride and dichloromethane showed increasing concentrations. The largest decrease in the chlorocarbons was 0.21 ppt (14%) for methyl chloroform. Significant inter-annual variability is expected for naturally emitted methyl chloride and chloroform, which have oceanic and biomass burning sources.

The dichloromethane growth rate has decreased substantially in the last few years (3.0 ppt/yr, 20%/yr; 2016-2017), down to 0.4 ppt/yr, 2.2%/yr 2019-2020). Two papers (Hossaini *et al*. 2017 and Oram *et al*. 2017) have shown that globally, dichloromethane is increasing rapidly in the atmosphere. However, the recent Ozone Assessment (Engel and Rigby 2018) showed recent (2015 to 2016) global growth rates have declined and are small compared to previous years. Engel and Rigby, 2018 concluded that it can't yet be determined whether the recent decrease in growth rate reflects a stabilization of emissions or reflects the large atmospheric variability.

The rate of decline of methyl chloroform is consistent with its relatively short atmospheric lifetime and near-zero global emissions, whereas the slower than expected rate of decline of carbon tetrachloride in the atmosphere suggests there are remaining, significant carbon tetrachloride sources outside the control of the Montreal Protocol (Engel and Rigby 2018).

1.4 Halons

H-1301 is now the most abundant halon in the background atmosphere (3.3 ppt, 2020), followed by H-1211 (3.2 ppt) and H-2402 (0.4 ppt). H-1211 and H-2402 are in decline in the atmosphere whereas H-1301 concentrations continue to grow. Overall halons are in decline by 1.4% per year (compared to 1.3%/yr 2018-2019), a larger rate of decline (in percentage terms) compared to the CFCs. Bromine in the atmosphere from halons decreased by 0.11 ppt/yr (1.4%). The decline in bromine from halons is a significant driver of likely ozone recovery (see Krummel *et al*. 2021).

1.5 Other organobromine species

Methyl bromide is the most abundant (6 ppt) organobromine ODS in the background atmosphere in 2020, followed by H-1301 (3.3 ppt), H-1211 (3.2 ppt), bromoform (1.6 ppt) and dibromomethane (1.1 ppt).

Methyl bromide showed a small increase (0.13 ppt/yr 2019-2020) 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. Year-to-year variability in the rate of decline or growth is expected because there are significant natural emissions of methyl bromide.

Natural bromoform and dibromomethane both showed an increase, 0.14 ppt/yr and 0.029 ppt/yr respectively in 2019-2020. Overall bromine from all non-halon ODSs increased by 0.6 ppt/yr (2019-2020), a 4.8%/yr increase. This is the first time in several years that background levels of bromoform have increased. 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 (Engel & Rigby 2018).

1.6 Total chlorine and bromine: impact on stratospheric ozone

Total chlorine from ODSs (Figure 4) decreased from 3142 ppt in 2019 to 3132 ppt in 2020, a decrease of 10 ppt (0.31%). This total is also 10% lower than its peak value in 1994. Total chlorine from chlorocarbons increased by 1.3 ppt (0.1%) and from HCFC increased by 2 ppt, however the overall decline in chlorine from CFCs was 13 ppt (2019-2020).

Total bromine from organobromine ODSs (Figure 5) was 20.4 ppt (2020) - 36% from halons, 29% from methyl bromide, 24% bromoform and 11% dibromomethane. Bromine from all ODSs increased by 0.51 ppt (2019-2020, 2.5%), due to the increase in 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 4. Total chlorine from CFCs, HCFCs, carbon tetrachloride (CTC), methyl chloroform (MC) and other chlorine-containing ODSs (Table 1) as measured at Cape Grim.

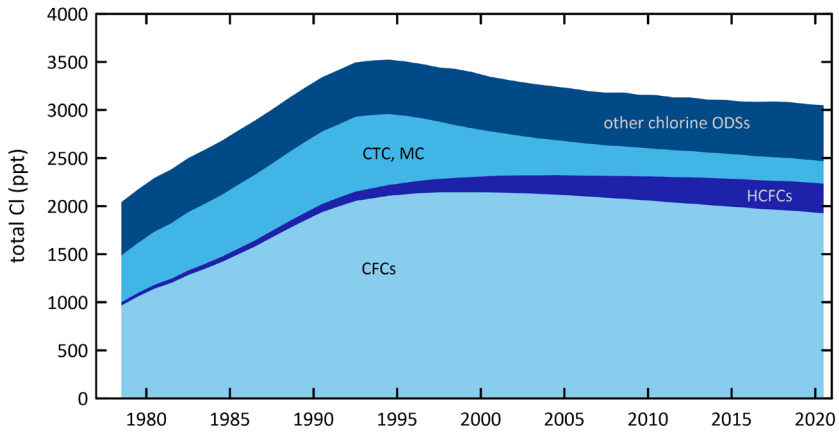
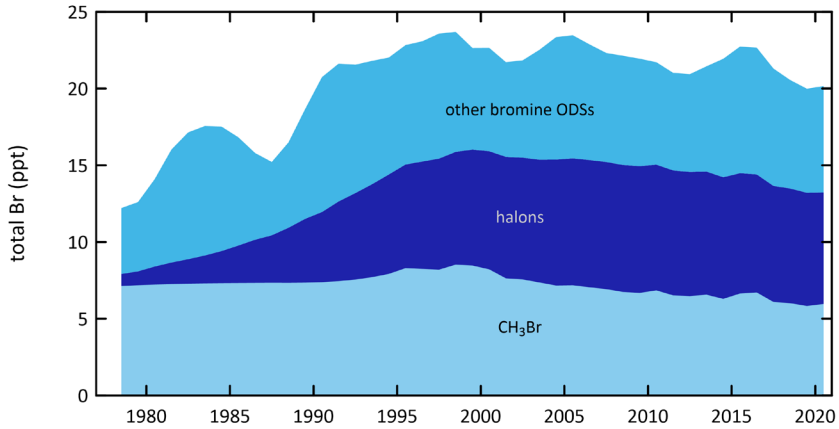


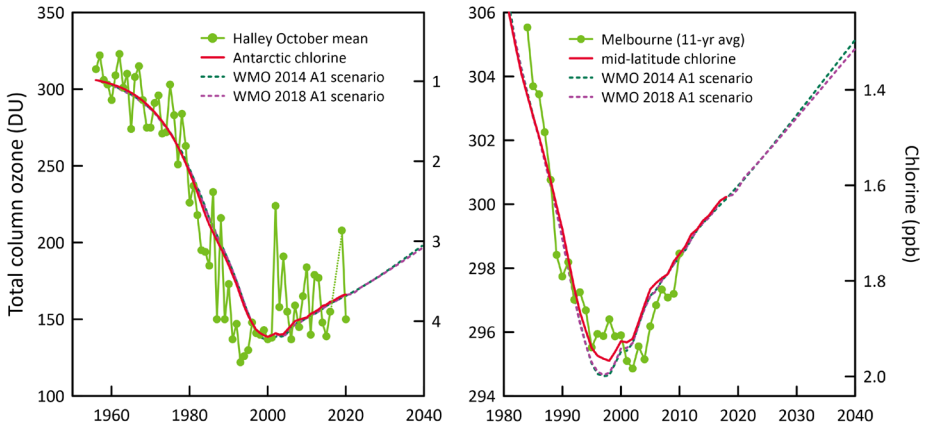
Figure 5. Total bromine from methyl bromide: CH_3Br, halons and other bromine-containing ODSs (dibromomethane: CH_2Br_2 and bromoform: $CHBr_3$) as measured at Cape Grim (Table 1).



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 2020 Antarctic Ozone Hole (Krummel *et al.*, 2021). 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; left), and Melbourne, Australia (38°S; right) and Equivalent Effective Stratospheric Chlorine (EESC,

ppb) changes at polar and mid-latitudes. The Melbourne ozone data are 11-year running means to minimise impacts of solar variability.



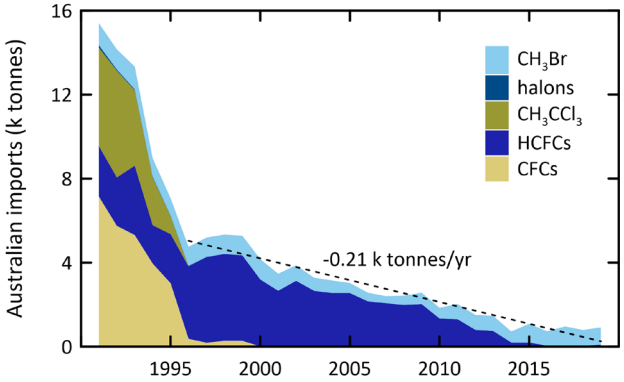
2 Australian ODS imports and banks

Data on Australian imports of ODSs are reported to the Australian Government (Department of Agriculture, Water and the Environment - DAWE) under licensing arrangements in the *Ozone Protection and Synthetic Greenhouse Gas Management Act, 1989*. Australian imports are documented in the DAWE *Ozone Licensing and Reporting System* (OLaRS: A. Gabriel, DAWE, private communications, 2014-2020). OLaRS details imports of bulk and pre-charged refrigeration and air conditioning equipment containing HCFCs (individual HCFCs or HCFC blends) and other bulk ODSs (in particular, carbon tetrachloride and methyl bromide). Imports of ODSs are shown in Table 2 and Figure 7.

There is an overall decline in Australian ODS imports from over 15,000 tonnes in 1991 to 771 tonnes in 2020, a long-term decline of about 250 tonnes per year since 1995. ODS imports in 2020 decreased by 100 tonnes from the previous year, due to methyl bromide imports decreasing from 823 tonnes (2019) to 722 tonnes (2020), for quarantine and pre-shipment applications.

In 1991, CFCs were the major ODS imports (7,144 tonnes), but they declined rapidly to 371 tonnes by 1996, and 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 import nearly doubled between 1991 (2,400 tonnes) and 1998 (4,100 tonnes); however since 1998 there has been a long-term decline in HCFC imports, falling to an average 45 tonnes per year from 2016-2020. Australia has reached its second last step in its HCFC phase out, with an annual import limit for HCFCs of around 45 tonnes of HCFC-22 from 2016 to 2029 (DoEE 2016).

Figure 7. Australian imports (k tonnes) of ODSs (CFCs, HCFCs, halons, MC: CH_3CCl_3, MB: CH_3Br).



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, drought, trade patterns and trading partner requirements (Note: MB values used here are imports minus exports). After 2007, methyl bromide imports increased and since 2011, imports have ranged from 519 to 915 tonnes per year. Methyl bromide imports in 2020 were 722 tonnes. 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). Imports of methyl bromide for non-QPS use fell to 30-35 tonnes by the mid-2000s and is now around 29 tonnes per year.

The current Australian CFC banks have been estimated at, CFC-11: 5,000-16,000 tonnes, CFC-12: 2,000-7,000 tonnes and CFC-113: 500-2,000 tonnes (Fraser *et al*. 2020). The Australian HCFC-22 bank was estimated at 4,475 tonnes in 2019 with a projected decline (Brodribb & McCann 2020) to 222 tonnes in 2030. The only other significant ODS bank in operational equipment is HCFC-123, with a bank of 190 tonnes in 2019 with a projected decline to 98 tonnes in 2030 (Brodribb & McCann 2020). Bank and bank emissions from Brodribb & McCann (2018, 2020) are included in Figure 8 (dashed lines: blue=banks, red=bank emissions).

Australian HCFC-22 and HCFC-123 emissions have been estimated using refrigerant bank data and specific emission factors for refrigeration/air conditioning equipment (Figure 8; Brodribb & McCann 2018, 2020). Estimated HCFC-22 emissions have declined from around 1,320 tonnes in 2003 to 262 tonnes in 2019. HCFC-123 emissions in 2019 were estimated at about 3.6 tonnes (Brodribb & McCann 2020).

Figure 8. Australian HCFC-22 and HCFC-123 banks and HCFC-22 bank emissions (Brodribb & McCann 2015). Dashed line is from Brodribb & McCann (2020): blue= banks, red=bank emissions.

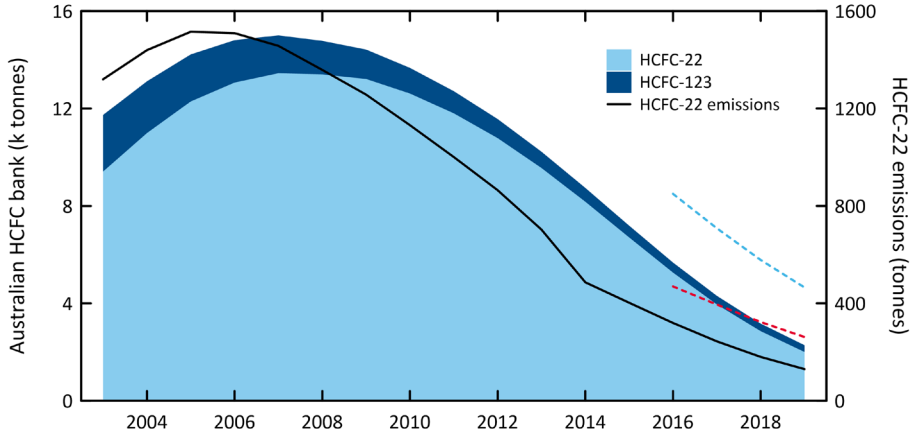


Table 2. Australian imports (bulk and pre-charged equipment, tonnes) of ODSs (CFCs, HCFCs, MC: CH_3CCl_3, halons, MB: CH_3Br) 1991-2020.

					CFCs	Total						HCFCs	Total			Halons	Total	CH3Br imports			Total
	-11	-12	-113	-114	-115	CFCs	-22	-123	-124	-141b	-142b	-225ca	HCFCs	CH_3CCl_3	-1211	-1301	Halons	n-QPS	QPS	Total	ODSs
1991	1759	4049	999	6	331	7144	2402						2402	4680	111	11	122	876	172	1048	15396
1992	1786	3054	808	19	84	5751	2252	17		30	3		2302	5086	14	39	53	799	160	959	14151
1993	1443	3205	485	6	172	5311	2940	60	8	269	23	1	3301	3586				921	166	1087	13285
1994	927	2784	168	11	64	3954	1328	67	2	411	14	1	1823	2273				704	172	876	8926
1995	498	2229	236	7	46	3016	1663	74	49	522	29	0.9	2338	846				664	168	832	7032
1996	69	181	118	3		371	2626	59	93	579	89	0.8	3448	0.1				631	276	907	4726
1997	52	129	0.1	3		184	3056	52	195	639	130	0.7	4071	0.1				660	259	919	5174
1998	90	182	0.1	3		275	2900	72	189	827	141	0.6	4129	0.1				569	352	921	5325
1999	90	182	0.1	3		275	2955	76	140	813	87	0.4	4071	0.1		1		507	425	932	5279
2000		8		1		9	2160	20	56	892	59	0.3	3187					452	516	968	4164
2001		8		1		9	2228	7	60	301	52	0.6	2648					335	475	810	3467
2002		8		1		10	2557	34	46	446	40	0.2	3123					323	415	738	3870
2003	1					1	2054	11	64	473	54	0.2	2656					183	441	624	3281
2004							2053	31	43	396	27	0.4	2551					207	390	597	3148
2005							1979	28	29	428	84	0.7	2548					119	358	477	3025
2006							1843	15	15	241	40	0.3	2156					55	355	410	2566
2007							1808	18	18	198	26	0.4	2068					46	288	334	2402
2008							1773	21	20	155	12	0.4	1981					41	401	442	2423
2009							1878	48	13	76	8	0.5	2023					33	509	543	2566
2010							1224	26	32	49	5	0.3	1336					34	472	506	1842
2011							1280	8	5	3	3	0.4	1300					33	689	722	2022
2012							758	20	4		2	0.5	784					33	676	709	1493
2013				0.0	0.0	0.0	714	14	4		2	0.4	734		0.1	1	1	32	618	650	1386
2014							181	4	0.5		0.3	0.3	186		0.4	3	3	30	489	519	708
2015		0.0				0.0	181	9	1			0.3	192					30	864	894	1086
2016							45					0.3	46					28	653	681	727
2017		0.0				0.0	42						42		1	0.8	2	30	885	915	960
2018							45						45		0.3	2	2	30	713	742	790
2019							46						46		0.5	1	2			823	870
2020							49						49		0.08	0.13	0.20			722	771

3 Estimated Australian and Global ODS emissions

CSIRO estimates of emissions of CFCs, HCFCs, methyl chloroform, carbon tetrachloride, halons and methyl bromide from the Melbourne 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 or inverse modelling (InTEM). Melbourne ODS emissions have been calculated, for 1995-2019, and scaled to Australian emissions, where appropriate, on a population basis. For details of methods and assumptions see Appendix A.

Australian ODS emissions can be calculated from Cape Grim data using the InTEM (Inversion Technique for Emission Modelling) inversion model (O'Doherty *et al*. 2009; Manning *et al*. 2003, 2011; Redington & Manning 2018). For a more detailed description of the InTEM model see Appendix A. Updates to the InTEM ODS emission estimates beyond 2018 are currently unavailable. CSIRO has recently developed in-house capability to run the InTEM model, therefore the InTEM ODS emissions estimates will be updated in the next report.

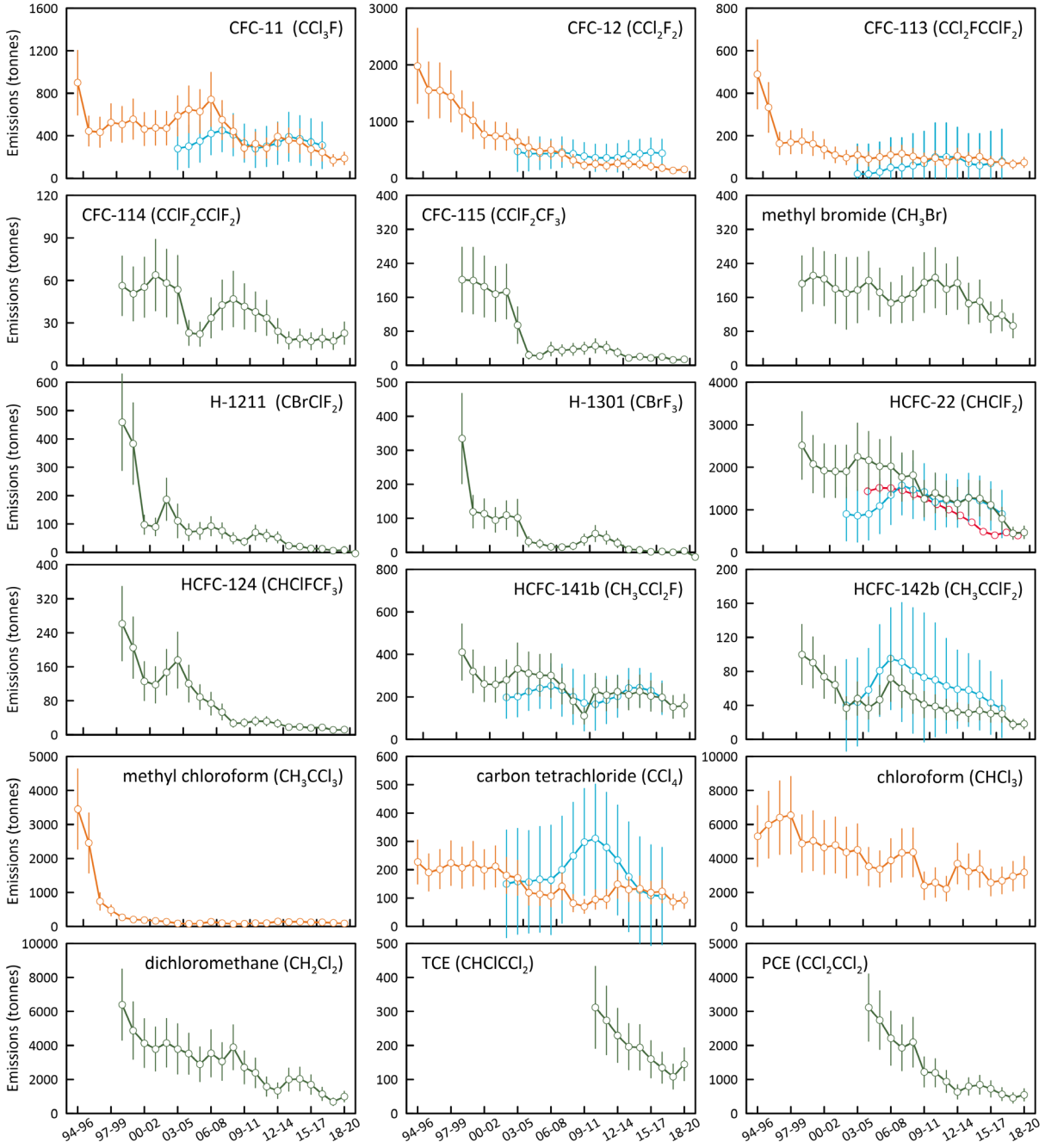
Australian halon emissions are derived from SE Australian emissions (ISC), assuming 6-8 % of SE Australian emissions are from the National Halon Bank located in Melbourne (see section 3.3). The SE Australian (non-Halon Bank) halon emissions are scaled to Australian emissions based on population.

To estimate Australian methyl bromide emissions (QPS) we begin with the SE Australian methyl bromide emissions estimated using the ISC method and then subtract the non-QPS SE Australian emissions. Australian emissions are obtained by scaling this result using a scaling factor based on the proportion of Australia's grain exports originating from SE Australian ports.

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 8; Brodribb & McCann 2015).

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 averages (1995-2019: Table 3, Figure 9)

Figure 9. Annual average (3-year 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 (orange: GC-ECD data; green: GC-MS data). InTEM emissions are show in light blue. HCFC-22 emissions from the refrigerant bank are shown in red (Brodribb & McCann 2015).



Global ODS emissions have been calculated using background ODS observations at Cape Grim and from other AGAGE stations in the Northern and Southern Hemispheres up to 2019 (Figure 10; 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).

Figure 10. Global annual emissions (M tonnes) of ODSs (CFCs, chlorinated solvents (methyl chloroform: CH_3CCl_3, carbon tetrachloride: CCl_4), methyl chloride, dichloromethane, chloroform, halons, methyl bromide: CH_3Br, HCFCs) derived from global AGAGE data.

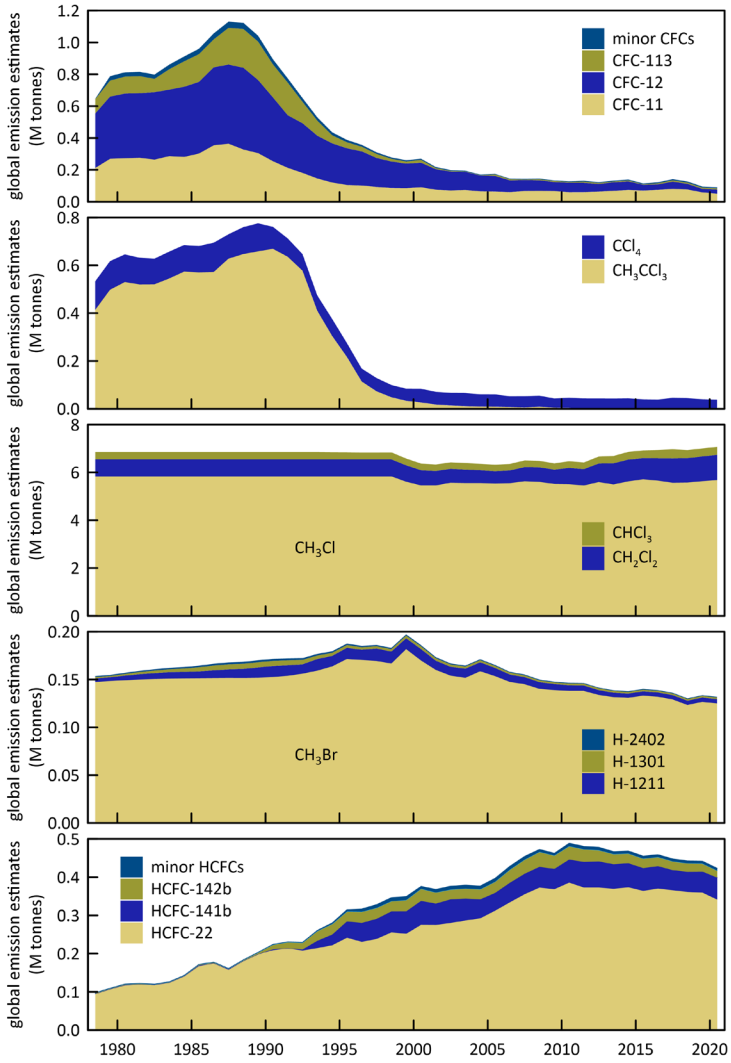


Table 3. Australian emissions (metric tonnes unless otherwise stated) of ODSs (CFCs, HCFCs, halons, MB and chlorocarbons) from Cape Grim AGAGE data. Pre-1999 emissions shown in red are assumed equal to 1999 emissions; pre-2005 emissions shown in red are assumed equal to 2005 emissions. Every second year is shown up until 2010 and then every year is shown.

ODS	1996	1998	2000	2002	2004	2006	2008	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
CFCs																	
CFC-11	445	526	556	475	587	629	548	284	326	284	392	357	348	272	243	168	188
CFC-12	1556	1445	1022	747	648	470	436	228	254	220	263	250	246	209	181	137	157
CFC-113	334	169	163	110	110	99	115	88	137	111	83	64	64	55	67	57	57
CFC-114	56	56	50	64	53	22	43	42	38	34	24	18	19	17	19	17	23
CFC-115	202	202	199	168	94	22	35	40	46	42	30	17	21	17	20	13	14
total	2592	2398	1992	1563	1492	1242	1177	682	800	690	793	706	698	570	530	392	438
ODP tonnes	2444	2283	1879	1474	1432	1214	1140	648	755	651	764	686	677	552	509	376	421
M tonnes CO_2-e	23.2	21.3	16.8	12.9	11.7	9.1	8.7	5.1	5.9	5.1	5.7	5.1	5.1	4.2	3.9	2.9	3.3
HCFCs																	
HCFC-22	2514	2514	2073	1904	2246	2025	1771	1263	1396	1257	1148	1279	1267	1119	795	447	468
HCFC-124	262	262	205	118	176	89	54	28	32	32	27	18	19	16	17	11	12
HCFC-142b	100	100	90	64	48	47	60	41	39	35	32	31	34	30	30	18	18
HCFC-141b	411	411	320	258	332	301	251	111	228	208	224	209	226	204	197	152	159
total	3286	3286	2689	2344	2802	2462	2136	1443	1695	1532	1431	1537	1546	1369	1040	628	657
ODP tonnes	196	196	160	140	167	150	130	85	105	95	91	96	97	86	68	43	45
M tonnes CO_2-e	5.2	5.2	4.3	3.9	4.5	4.0	3.6	2.5	2.8	2.5	2.3	2.5	2.5	2.3	1.7	1.0	1.0
Halons																	
H-1211	459	459	384	90	111	76	76	37	70	59	53	23	21	13	12	6	8
H-1301	335	335	120	95	101	25	15	37	55	43	27	9	7	2	2	0	5
total	794	794	503	185	212	101	92	74	125	102	80	32	28	15	15	6	13
ODP tonnes	4724	4724	2348	1220	1346	478	382	482	761	608	432	157	131	58	59	15	70
M tonnes CO_2-e	2.9	2.9	1.3	0.77	0.84	0.27	0.20	0.30	0.47	0.37	0.26	0.09	0.07	0.03	0.03	0.01	0.04
CH_3Br	523	579	608	510	416	314	351	474	521	563	509	565	576	687	645	684	630
ODP tonnes	314	348	365	306	249	188	211	284	312	338	305	339	346	412	387	411	378
k tonnes CO_2-e	2.6	2.9	3.0	2.6	2.1	1.6	1.8	2.4	2.6	2.8	2.5	2.8	2.9	3.4	3.2	3.4	3.2
Chlorocarbons (MP)																	
CCl_4	191	223	221	212	171	114	141	70	95	96	148	131	133	119	123	86	93
CH_3CCl_3	2457	475	203	165	89	87	100	76	101	98	145	133	143	120	123	103	92

ODS	1996	1998	2000	2002	2004	2006	2008	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
total	2648	699	425	377	260	201	241	147	196	194	293	263	276	239	246	189	184
ODP tonnes	455	293	264	249	197	134	165	85	114	116	178	157	161	143	148	105	111
M tonnes CO_2-e	0.70	0.47	0.43	0.40	0.32	0.22	0.27	0.14	0.19	0.19	0.29	0.25	0.26	0.23	0.24	0.17	0.18
MP ODSs (k tonnes)	9.8	7.8	6.2	5.0	5.2	4.3	4.0	2.8	3.3	3.1	3.1	3.1	3.1	2.9	2.5	1.9	1.9
ODP (k tonnes)	8.1	7.8	5.0	3.4	3.4	2.2	2.0	1.6	2.0	1.8	1.8	1.4	1.4	1.3	1.2	0.9	1.0
M tonnes CO_2-e	35	33	26	21	20	15	15	10	12	11	11	11	11	10	9.0	7.5	7.6
other chlorocarbons																	
dichloromethane	6406	6406	4881	3798	3796	2895	3055	2708	2382	1576	1339	1992	2037	1696	1149	691	991
chloroform	5990	6547	5046	4791	4516	3377	4324	2393	2590	2217	3703	3219	3385	2585	2707	2962	3178
PCE	3112	3112	3112	3112	3112	2745	1931	1220	1195	942	634	791	842	724	568	459	545
TCE									312	273	230	196	194	160	134	108	145
total (k tonnes)	16	16	13	12	11	9.0	9.3	6.3	6.5	5.0	5.9	6.2	6.5	5.2	4.6	4.2	4.9
ODP tonnes	132	137	110	98	95	74	79	54	53	40	50	52	54	43	38	36	41
M tonnes CO_2-e	0.27	0.29	0.23	0.21	0.21	0.16	0.18	0.11	0.11	0.09	0.13	0.12	0.13	0.10	0.10	0.10	0.11
all ODSs																	
all ODSs (k tonnes)	25	24	19	17	17	13	13	9	10	8	9	9	10	8	7	6	7
ODP (k tonnes)	8.3	8.0	5.1	3.5	3.5	2.2	2.1	1.6	2.1	1.8	1.8	1.5	1.5	1.3	1.2	1.0	1.1
M tonnes CO_2-e	35	33	26	21	20	15	15	10	12	11	11	11	11	10	9.1	7.6	7.7

3.1 CFCs

3.1.1 Australian emissions

Australian CFC-11 emissions have declined overall since 1995 (900 tonnes) to 188 tonnes in 2019 (Table 3, Figure 9). CFC-11 emissions increased from 2002 (475 tonnes) to 2007 (744 tonnes) - the cause of this increase is unclear. Since 2007, Australian CFC-11 emissions have been mostly in decline, declining at approximately 11% per year, reaching 188 tonnes in 2019.

Australian CFC-12 emissions have declined steadily since 1995 (1,981 tonnes) to 157 tonnes in 2019 and have averaged 215 tonnes from 2010-2019, 56% lower than average 2004-2009 emissions (490 tonnes per year). CFC-12 emissions have been steadily declining for the last 6 years after being stalled during the 2009-2013 period.

Australian CFC-113 emissions declined rapidly from 488 tonnes in 1995 to under 100 tonnes in the early 2000s, averaging close around 90 tonnes per year from 2002 to 2019. CFC-113 emissions in 2019 were 57 tonnes. Recent CFC-113 emissions are quite variable and do not show a clear trend.

Total Australian CFC (and HCFC, halon and carbon tetrachloride) emissions are shown in Figure 11. The overall decline in CFC emissions from 1995 (3,627 tonnes) to 2019 (438 tonnes) is about 8% per year. ODP weighted CFC emissions have fallen from 3449 tonnes in 1995 to 420 tonnes in 2019. Australian ODP-weighted CFC emissions in 2019 (421 tonnes) were 0.3% of global ODP-weighted CFC emissions. Australian ODP-weighted CFC emissions are currently (2019) about 38% of Australia's total ODS (ODP-weighted) emissions (1100 tonnes).

CFC emissions are presumed to be from CFC-containing appliances/materials such as refrigeration/air conditioning equipment, foams, aerosol cans, either existing or buried (landfills).

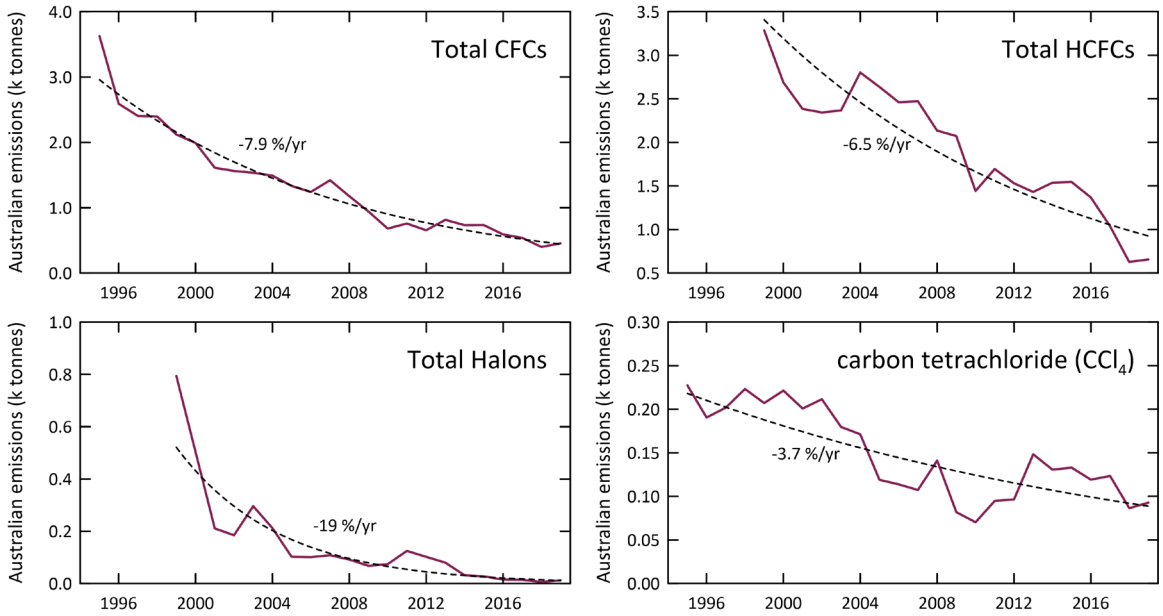
3.1.2 Global emissions

Total global CFC emissions are declining, dominated (87%) by emissions of CFC-11 and CFC-12, currently about 80 k tonnes/yr (2019) (Rigby *et al*., 2014 and updates). Since the peak emissions of CFCs in the late 1980s (1108 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. Two papers, Montzka *et al*. (2018) and Rigby *et al*. (2019), found an unexpected increase in global emissions of CFC-11 after 2012. The conclusions from both studies suggested that the increase is probably not related to existing banks of CFC-11 but rather new production and uses in east Asia, around 50% of which has been attributed to east China. The source of the remaining 50% has not been identified. Global CFC-11 emissions averaged about 78 k tonnes/yr (2016-2018), an increase of 14 k tonne per year compared to the 2008-2012 period (Rigby *et al*. 2019 and updates). Global CFC-11 emissions have decreased significantly to 52 k tonnes in 2019 (Montzka *et al*., 2021). This is likely due to the recent decline in CFC-11 emissions from eastern China (Park *et al*., 2021).

Global emissions of CFC-12 have declined substantially since its peak in the 1980s, and emissions continue to decline. Since the peak emissions of CFC-12 in the late 1980s (513 k tonnes, 1988), CFC-12 emissions have declined by 96% (10%/yr). Current global CFC-12 emissions are about 22 k tonnes/yr (2019) (Rigby *et al*. 2014 and updates).

The global emissions of the other CFCs (CFC-113, -114, -115), including the minor CFCs, decreased from a total of 276 k tonnes in 1989 to 12 k tonnes in 2019 (10%/yr). Most of the decrease is due to a decline in emissions of CFC-113.

Figure 11. Declining Australian CFC, HCFC, halon and carbon tetrachloride emissions (k tonnes).



3.2 HCFCs

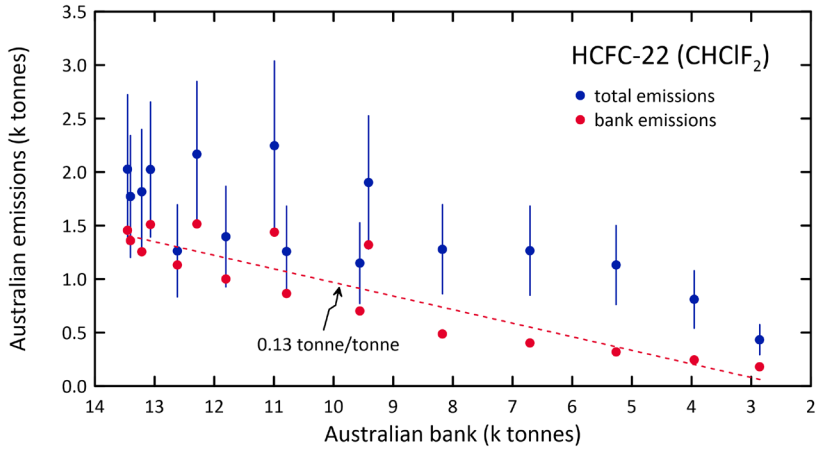
3.2.1 Australian emissions

Total Australian HCFC-22 emissions have been estimated from Cape Grim data. There has been an overall decline in Australian HCFC-22 estimated emissions from about 2,500 tonnes in 1999 to 468 tonnes in 2019 (8% per year over this period) (Table 3, Figure 9). HCFC-22 emissions declined rapidly from 2,246 tonnes in 2004 to 1,148 tonnes in 2013 (7% per year). The decline in HCFC-22 emissions briefly stalled in 2014 and 2015 but emissions have declined for the last 6 years, falling to an estimated 468 tonnes in 2019.

Australian HCFC-22 emissions have also been estimated by (Brodribb & McCann 2018, 2020) from operational and end-of-life refrigeration/air conditioning equipment using 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 (Figure 3, Figure 12). As mentioned in section 2, Brodribb & McCann (2020) estimated the Australian HCFC-22 Bank to be 4,467 tonnes in 2019 and Australian emissions of HCFC-22 in 2019 to be 262 tonnes.

Total HCFC-22 emissions from Cape Grim data and as projected leakage from the HCFC-22 bank are shown in Figure 12 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 13%/yr.

Figure 12. 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 & McCann 2020). The dashed line is a linear regression: slope = 0.13 tonne/tonne banked.



HCFC-124 emissions have declined steadily from 262 tonnes in 1999 to 12 tonnes in 2019 (a decline of around 14% per year). Emissions levelled off around 2009-2013 and then dropped again each year from 2014 to 2019.

HCFC-141b emissions have fluctuated over the period 1999-2019, falling from over 400 tonnes in 1999 to around 159 tonnes in 2019, with some falls and increases in the intervening period.

HCFC-142b emissions have also fluctuated over the period 1999-2019, falling from 100 tonnes in 1999 to 18 tonnes in 2019 with an increase estimated in 2007 before a steady decline to 2019.

Total HCFC emissions have fallen by about 80% from 3,286 tonnes in 1999 to 657 tonnes in 2019, an overall decline of about 6.5% per year. ODP-weighted HCFC emissions have fallen from 196 tonnes in 1999 to 45 tonnes in 2019, 4% of Australia's total ODS (ODP-weighted) emissions in 2019 (1100 tonnes). GWP-weighted HCFC emissions have fallen from 5.2 M tonnes CO_2-e in 1999 to 1 M tonnes CO_2-e in 2019, an overall decline of 80%.

Australian HCFC emissions are likely from a combination of service and malfunction leaks from existing refrigeration/ac equipment, from other minor uses and from landfills. Australian HCFC emissions in 2019 (657 tonnes) were 0.15% of global HCFC emissions (441 k tonnes) on a metric tonne basis.

3.2.2 Global emissions

Global HCFC emissions peaked in 2010 (488 k tonnes) and have since declined (10%) to 441 k tonnes in 2019, 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 ozone layer recovery. Throughout the period from the late-1970s to 2010, total HCFC emissions increased by about 4%/yr. Declining HCFC emissions cause the observed slowing of the recent HCFC concentration growth rates in the atmosphere (see section 1.2).

HCFC-22 emissions (the most commonly used HCFC) peaked in 2010 at 386 k tonnes declining (0.8%/yr) to 360 k tonnes in 2019; HCFC-141b: 68 k tonnes (2012) to 55 k tonnes (3%/yr, 2019); HCFC-142b: 39 k tonnes (2008) to 21 k tonnes (6%/yr, 2019).

3.3 Halons

3.3.1 Australian emissions

The origin of current Australian halon emissions is uncertain, but they could be coming from remaining building fire-fighting systems, from the Australian Halon Bank (a facility to dispose of or store and reprocess halons located in Melbourne) and exempted halon based aircraft firefighting suppression systems.

Estimated Australian halon emissions fell from nearly 800 tonnes in 1999 to 13 tonnes in 2019 (Figure 9, Figure 11), an overall decline of about 19% per year. The majority of Australian halon emissions over the period 2012-2019 are H-1211. SE Australian halon emissions are likely to show significant inter-annual variability.

Australian H-1211 emissions in 2019 were 13 tonnes, 0.4% of global emissions (3,100 tonnes) and Australian H-1301 emissions in 2019 were 5 tonnes, 0.4% of global emissions (1,200 tonnes).

3.3.2 Global emissions

Data on global halon emissions are now available back to the 1970s, based on AGAGE *in situ* and Cape Grim Air Archive data (Vollmer *et al*. 2016). Total global halon emissions continue to decline (6.8 k tonnes in 2012, 4.8 k tonnes in 2019), dominated (~70%) by halon-1211 emissions, which declined by 1.4 k tonnes over the same period. Peak total halon emissions (18 k tonnes) occurred in 1990.

3.4 Methyl bromide

3.4.1 Australian emissions

Methyl bromide (MB) is used in Australia mainly as a fumigant for cereals, such as wheat, for cottonseed and timber logs prior to export and biosecurity treatment of imported goods. These are QPS uses, which are exempted from phase-out under the Montreal Protocol. Methyl bromide is also used as a soil sterilant during the production of strawberry runners in the Toolangi district of Victoria, this being a non-QPS use 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.

For many years now, methyl bromide has been used in Australia predominantly for QPS uses and predominantly to fumigate exported commodities to meet the requirements of receiving countries. The commodities most commonly fumigated with methyl bromide are cereal grains, wood and timber (logs). It is also occasionally used in small amounts as a feedstock in chemical reactions to create other chemicals.

A UNEP model of methyl bromide emissions suggests that 80-90 % of methyl bromide QPS use escapes to the atmosphere (UNEP 2019). The results from the UNEP model of Australian methyl bromide emissions are shown in Figure 13. SE Australian methyl bromide emissions from this model (assuming speciation of SE Australian methyl bromide emissions from timber, grain exports and strawberry runner production) are compared to SE Australian emissions calculated from Cape Grim data by ISC in Figure 14 (also Figure 9). 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 14.

Figure 13. Australian methyl bromide emissions (3-year average) calculated from Australian methyl bromide import data and the modified UNEP emissions model (UNEP 2019.

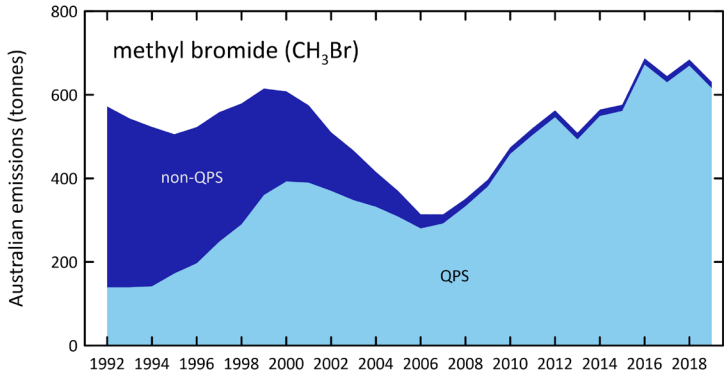
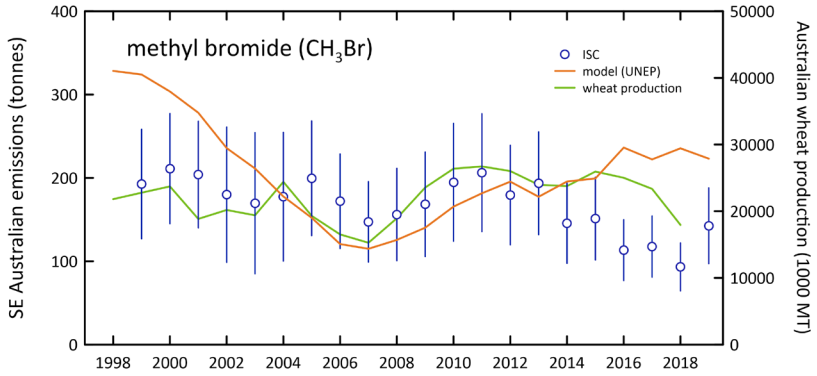


Figure 14. SE Australian methyl bromide emissions calculated by ISC from Cape Grim *in situ* AGAGE methyl bromide data (blue) and from a modified UNEP (2019) emissions model (orange, 3-year average) based on Australian methyl bromide imports and assumed SE Australian methyl bromide consumption patterns.



The emissions from the model and those derived from atmospheric data for SE Australia show reasonable overall agreement over the period 2002-2019. 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 600 tonnes per year in 1999-2000, falling to 300 tonnes per year in 2006-2007, due to declining QPS and non-QPS methyl bromide use and also a decline in QPS use (possibly due to reduced grain production as a result of the severe drought in SE Australia), increasing again to 630 tonnes in 2019, due to increased QPS methyl bromide use resulting from grain harvests recovering, trade patterns and trade partner requirements (Figure 13). The 378 ODP tonnes of methyl bromide emitted in 2019 are about 34% of Australia's total ODS emissions in ODP tonnes.

3.4.2 Global emissions

Global methyl bromide emissions have been estimated from AGAGE data, including Cape Grim (Rigby *et al*. 2014 and updates). Global methyl bromide emissions decline from 182 k tonnes in

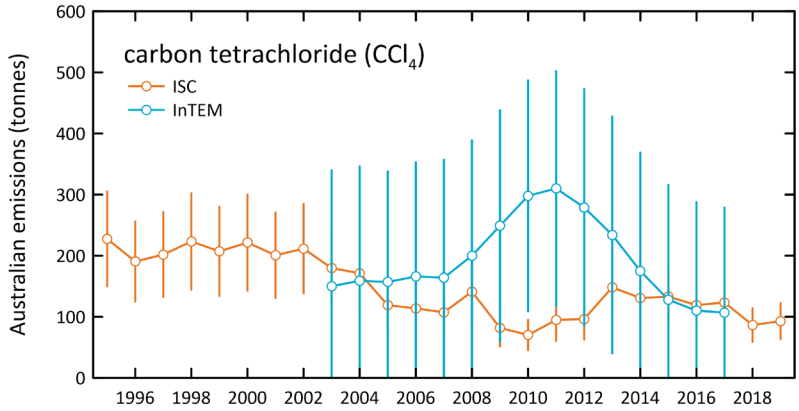
1999 to 127 k tonnes in 2019, a decline of about 1.8%/yr. Methyl bromide emissions drop from 133 k tonnes in 2015 to 127 k tonnes in 2019 reflecting in part the developing country phase-out of non-QPS uses in 2015.

3.5 Carbon tetrachloride and methyl chloroform (Montreal Protocol chlorocarbons)

3.5.1 Australian emissions

Australian carbon tetrachloride (CTC) emissions are calculated from Cape Grim *in situ* GC-ECD data. The latest available estimates of Australian carbon tetrachloride emissions by ISC and InTEM are shown in Figure 15. Carbon tetrachloride emissions estimated by ISC were 227 tonnes in 1995, declining to 93 tonnes by 2019, a decline of 4% per year.

Figure 15. Australian carbon tetrachloride emissions calculated from Cape Grim *in situ* AGAGE data by ISC and inverse modelling (InTEM).



The ISC and InTEM estimates of Australian carbon tetrachloride emissions were published in 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* and 2018 (Carpenter & Reimann 2014; Engel & Rigby 2018) and the *SPARC Report on the Mystery of Carbon Tetrachloride* (Liang *et al*. 2016).

Australian methyl chloroform emissions declined from about 3,500 tonnes in 1995 to below 100 tonnes in 2004. From 2004 to 2019 methyl chloroform emissions averaged around 106 tonnes per year. In 2019 Australian methyl chloroform emissions were estimated at 92 tonnes, 5% of global emissions (1900 tonnes).

3.5.2 Global emissions

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 1998, a long-term decline of about 4%/yr. Since 1998 global carbon tetrachloride emissions have fluctuated between increases and decreases but overall have declined steadily by about 0.5 k tonne per year (1.3%/yr) until 2019 when emissions were estimated at 38 k tonnes/yr. The decline in global carbon tetrachloride emissions is not as rapid as anticipated under the Montreal Protocol (Lunt *et al*. 2018). One explanation might be that global emissions of carbon tetrachloride from landfills and chlor-alkali plants may not be accounted for in global budgets (Fraser *et al*. 2014; Hu *et al*. 2016, Liang *et al*. 2016).

Global methyl chloroform emissions peaked in 1990 at 669 k tonnes. Emissions have declined almost every year since and are estimated at 1.9 k tonnes in 2019. Over the past 9 years (2011-2019) global emissions have averaged 2 k tonnes/yr. The long-term decline in methyl chloroform emissions is in excess of 20%/yr, which is expected for an ODS with low (~zero) emissions and an atmospheric lifetime of about 5 years.

3.6 Chlorocarbons not controlled by the Montreal Protocol

Dichloromethane (CH_2Cl_2), chloroform ($CHCl_3$), trichloroethylene ($CHClCCl_2$) and perchloroethylene (CCl_2CCl_2) are short-lived ODSs whose production and consumption are not controlled by the Montreal Protocol. Emissions of all these ODSs are seen in the Cape Grim data (Figure 1).

- Australian dichloromethane emissions were about 6400 tonnes in 1999. Since then emissions have fluctuated between increases and decreases but overall have declined steadily by about 9% per year to 991 tonnes in 2019.
- Australian chloroform emissions were over 5,300 tonnes in 1995 declining overall by 4% per year to about 2,217 tonnes in 2012, and after a rise for 3 years, is now averaging around 2,800 tonnes/yr in 2016-2019. There are natural emissions of chloroform from soil and it is possible that the chloroform emissions calculated from Cape Grim data contain natural emissions which are difficult to quantify.
- Australian perchloroethylene emissions were over 3,000 tonnes in 2004 declining to 545 tonnes in 2019 (11% per year).
- Australian trichloroethylene emissions were about 312 tonnes in 2011 falling by 9% per year to 145 tonnes in 2019.

Total Australian emissions of these short-lived ODSs (not including trichloroethylene, as reliable measurements weren't obtained until 2011) were 11k tonnes (95 ODP tonnes) in 2004, falling by 5% per year to 4.9 k tonnes (41 ODP tonnes; including trichloroethylene) in 2019. The ODP-weighted emissions of these short-lived ODSs were about 4% of total Australian ODS emissions (ODP weighted).

3.7 Australian GWP-weighted ODS emissions

The success of the Montreal Protocol in controlling the production and consumption of ODSs has protected the ozone layer. The resulting reductions in atmospheric abundances of ODSs also reduced the human influence on climate because all ODSs are greenhouse gases. In this section we will look at the GWP-weighted emissions of the ODSs as a measure of the overall threat to climate from ODSs noting that none of the ODS emissions are included in Australia's national GHG emissions inventory.

The overall decline in Australian GWP-weighted CFC emissions from 1995 (31 M tonnes CO_2-e) to 2019 (3.3 M tonnes CO_2-e) is 9% per year. 2019 emissions (3.3 M tonnes CO_2-e) were 0.6 % of Australia's total GHG emissions (519 M tonnes CO_2-e, including land use change, 2019). The 28 M tonnes CO_2-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

(CO_2, CH_4, N_2O, HFCs, PFCs, SF_6 including land use change), as reported to UNFCCC, have increased by a net 25 M tonnes CO_2-e from 1995 to 2019.

The overall decline in GWP-weighted HCFC emissions from 1999 (5.2 M tonnes CO_2-e) to 2019 (1.0 M tonnes CO_2-e) is 9% per year; CO_2-e weighted HCFC emissions in 2019 are 0.2% of Australia's reported net GHG emissions.

The CO_2-e weighted emissions of other ODSs (halons, methyl bromide, chlorocarbons, other chlorocarbons) totalled 0.33 M tonnes in 2019, around 0.06% of Australia's reported net GHG emissions.

3.8 Total ODS emissions

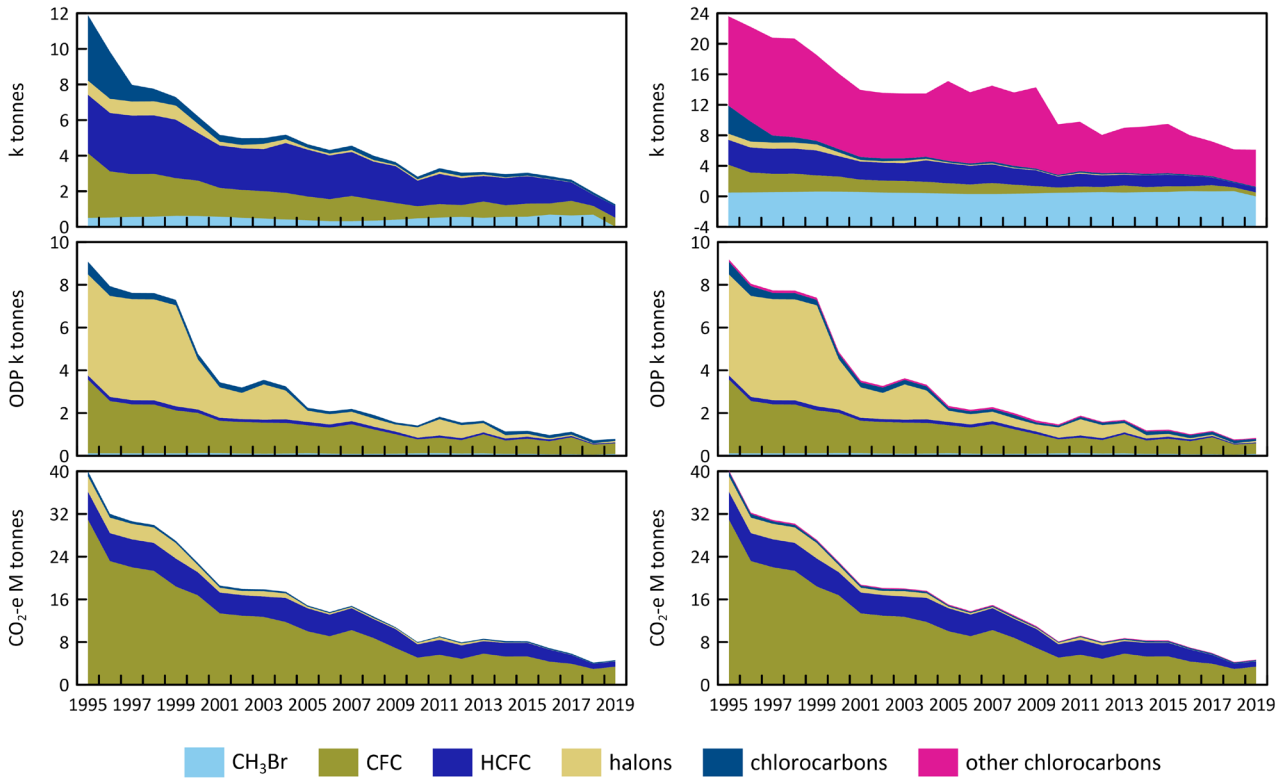
Total Australian ODS emissions (CFCs, HCFCs, halons, methyl bromide, carbon tetrachloride, methyl chloroform, both ODP- and GWP-weighted), and Montreal Protocol controlled ODS emissions are both shown in Figure 16. From 1995-1998, emissions are based on Cape Grim GC-ECD data for the major CFCs (CFC-11, -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 for which 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.

The non-Montreal Protocol chlorocarbons (dichloromethane, chloroform, perchloroethylene, trichloroethylene) make a significant contribution to total Australian ODS emissions in metric tonnes. Since 2011, 65%-70% of ODS emissions are from these other chlorocarbons, as emissions of Montreal Protocol chlorocarbons have reduced. However, they only make small contributions to ODP (2%-4%) 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 1.1 k tonnes in 2019. The largest decline is due to the halons, falling from emissions of 4.7 k tonnes (ODP) in 1999 to 0.07 k tonnes (ODP) in 2019, a fall of 4.63 k tonnes compared to a fall of 3 k tonnes (ODP) in CFC emissions and 0.15 k tonnes ODP in HCFC emissions over the same period.

GWP-weighted ODS emissions fell by about 7% per year from 43 M tonnes CO_2-e in 1995 to 8 M tonnes CO_2-e in 2019. As discussed previously the fall due to CFCs alone is 28 M tonnes CO_2-e, 80% of the overall decline in GWP-weighted ODS emissions.

Figure 16 Montreal Protocol controlled ODS emissions (Section 3.8 left); Total Australian ODS emissions (right).



The significance of the decline of GWP-weighted ODS emissions compared to GWP-weighted emissions of the GHGs reported to UNFCCC (CO_2, CH_4, N_2O, HFCs, PFCs, SF_6) is shown in Figure 17.

The 35 M tonnes CO_2-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 previously, Australian emissions of UNFCCC GHGs increased by a net 25 M tonnes CO_2-e from 1995 to 2019.

Figure 17. Australian emissions (GWP-weighted: M tonnes CO_2-e) of Montreal Protocol ODSs and the GHGs reported to UNFCCC including GHG emissions due to land-use/land-use change and forestry (LULUCF).

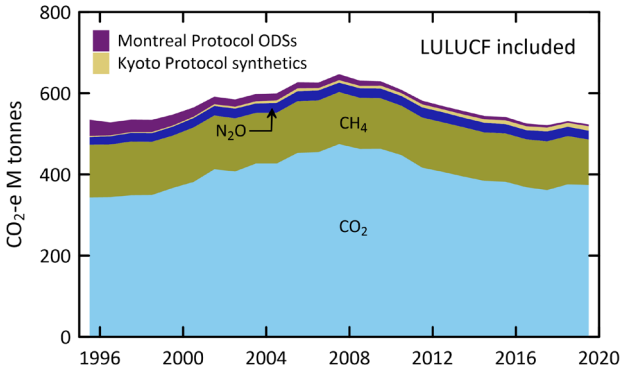
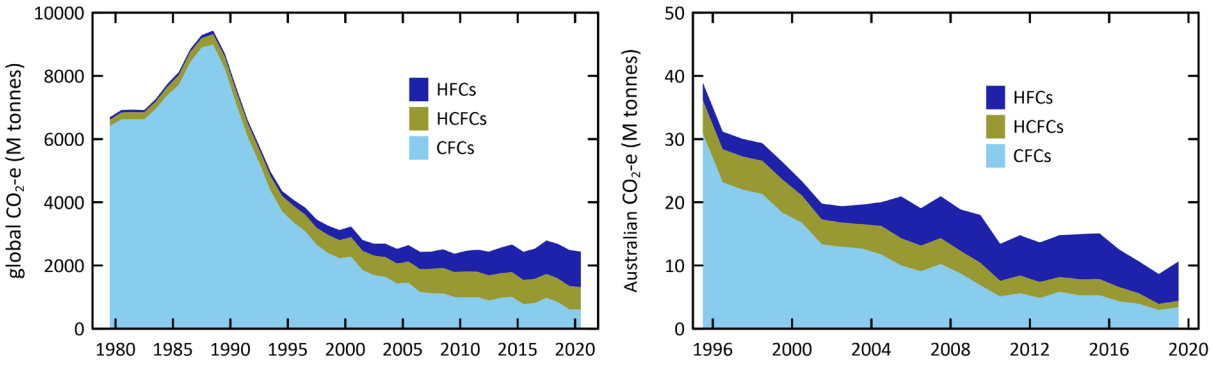


Figure 18 shows a comparison of global and Australian CFC, HCFC and HFC emissions in M tonnes CO_2-e from atmospheric abundance data. There has been about a 74% decline in global CFC, HCFC and HFC emissions since their peak (9400 M tonnes CO_2-e) in the late 1980s and about a 39% decline since 1995. There has been a corresponding 73% decline in Australian CFC, HCFC and HFC emissions since 1995 (39 M tonnes CO_2-e). Australian ODS emissions are typically about 0.5% to 1% of global emissions. In 2019, global CFC, HCFC and HFC emissions contribute 25%, 29% and 46% respectively, with HFCs clearly dominant. In Australia HFC emissions have also become dominant (59%) with CFCs at 32% and HCFCs at 9%. Global CFC, HCFC and HFC emissions have been about 2400-2700 M tonnes CO_2-e since 2004.

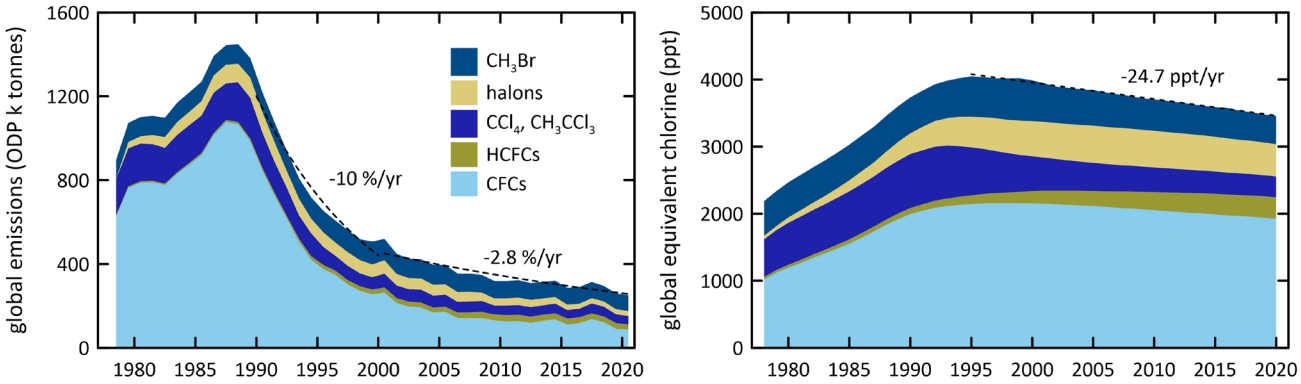
Figure 18. Global (since 1978) and Australian (since 1995) CFC, HCFC and HFC emissions (M tonne CO_2-e).



The total ODS global emissions for the Montreal Protocol ODSs in ODP tonnes are shown in Figure 19, 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 1,446 k tonnes in the late 1980s, declining at 10% per year from 1990 to 2000 and then 2.8% per year from 2000 to 2019, to reach 260 k tonnes in 2019.

The total global concentrations of ODSs, expressed as equivalent chlorine (Figure 19), peaked later in the mid-1990s at 4.05 ppb, declining slowly (24.7 ppt per year) to 3.5 ppb by 2019, this slow decline resulting largely from the long (50-100+ years) lifetimes for carbon tetrachloride and CFCs in the atmosphere.

Figure 19. Global emissions (ODP tonnes) of the Montreal Protocol ODSs and global equivalent chlorine (ppt), both derived from AGAGE data. For methyl bromide, pre-1998 emissions are scaled to post-1998 from global atmospheric concentrations..



Appendix A

A.1 Key References

CFCs Fraser *et al*. 1977, 1983, 1996, 2016, 2018; Fraser & Pearman 1978a,b; Hyson *et al*. 1980; Rasmussen & Khalil 1979; Rasmussen *et al*. 1982; Cunnold *et al*. 1983; 1986, 1994, 1997; Oram 1999; Fraser 2000; Prinn *et al*. 2000, 2018; 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; Adcock *et al*. 2018; Montzka *et al*. 2018; Vollmer *et al*. 2018; Engel & Rigby 2018; Reimann *et al*. 2018; Montzka *et al*. 2021; Park *et al*. 2021

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, 2018; 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; Fraser *et al*. 2016; Liang *et al*. 2017; Meinshausen *et al*. 2017; Simmonds *et al*. 2017, 2018; Engel & Rigby 2018; Reimann *et al*. 2018; Li *et al*. 2019

Halons Butler *et al*. 1994; Fraser & 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; Engel & Rigby 2018; Reimann *et al*. 2018

CTC Fraser & Pearman 1978a; Rasmussen *et al*. 1982; Simmonds *et al*. 1988, 1998; Prinn *et al*. 2000, 2018; Sturrock *et al*. 2002; Dunse *et al*. 2005; Xiao 2008; Xiao *et al*. 2010a; Laube *et al.* 2013; Fraser *et al*. 2014b, 2016, 2018; Rigby *et al*. 2014; Chipperfield *et al*. 2016; Liang *et al*. 2016; Meinshausen *et al*. 2017; Lunt *et al*. 2018; Engel & Rigby 2018; Reimann *et al*. 2018

MC Fraser & Pearman 1978a; Rasmussen *et al*. 1982; Fraser *et al*. 1986, 2016, 2018; Prinn *et al*. 1987, 1992, 1995, 2000, 2001, 2005, 2018; 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; Engel & Rigby 2018; Reimann *et al*. 2018

CH_3Cl 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; Engel & Rigby 2018

CH_2Cl_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

$CHCl_3$ 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; Fang *et al*. 2019

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: Engel & Rigby 2018; Reimann *et al*. 2018

CH_2Br_2 Yokouchi *et al*. 2005

$CHBr_3$ Yokouchi *et al*. 2005

CH_3I Cox 2001; Cohan *et al*. 2003; Cox *et al*. 2004

A.2 Cape Grim *in situ* and air archive measurements

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 Oceanic and Atmospheric Administration (NOAA, USA), UEA, and the National Institute for Environmental Research (NIES, Japan).

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; Vollmer *et al*. 2018). 'CFC-114' measured on the AGAGE instruments is actually an unresolved mixture of CFC-114 ($CClF_2CClF_2$) and CFC-114a (CCl_2FCF_3), 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 & 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 ^{37}Cl-CFC-11 ($CCl_2{}^{37}ClF$), ^{37}Cl-CFC-12 ($CCl^{37}ClF_2$), CFC-112 (CCl_2FCCl_2F), CFC-112a ($CClF_2CCl_3$), CFC-113a (CCl_3CF_3), ^{37}Cl-CFC-113 ($CCl^{37}ClFCClF_2$), CFC-114a (CCl_2FCF_3), CFC-133a (CCl_3CF_3), CFC-216ba ($CClF_2CClFCF_3$), CFC-216ca ($CClF_2CF_2CClF_2$), HCFC-31 (CH_2ClF), HCFC-133a (CH_3CClF_2) and HCFC-225ca ($CHCl_2CF_2CF_3$) (Kloss *et al*. 2014; Laube *et al*. 2014; Vollmer *et al*. 2015; Schoenenberger *et al*. 2015; Adcock *et al*. 2018; Engel & Rigby 2018; CSIRO unpublished data). Other new ODSs recently measured at Cape Grim include HCFC-21 ($CHCl_2F$, commencing 2015), HCFC-132b ($CHCl_2CHF_2$, commencing 2015) and HCFC-1233zd (or HFO-1233zd: $CHClCHCF_3$, commencing 2014). Provisionally calibrated data are available for HCFC-132b (Table 1).

A.3 NAME/InTEM

NAME (Numerical Atmospheric Dispersion Modelling Environment) is a Lagrangian particle dispersion model. 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-hour 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, InTEM (Inversion Technique for Emission Modelling) 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 this 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. For the current InTEM inversions (VextT), emissions were scaled from a domain that includes Victoria and Tasmania as well as southern and south western New South Wales and eastern South Australia. Emissions referred to as NAME in the report use a different domain that incorporates all of Victoria, Tasmania and New South Wales. The Australian emissions are calculated from InTEM (VextT) using a population based scale factor of 2.6, and are shown in Figure 9

A.4 ISC methods and assumptions

The original ISC emission estimates were based on 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 & 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 is estimated from the episodes of Port Phillip CO pollution observed at Cape Grim.

NOAA air mass back trajectory analyses (Draxler & Hess 1997) are used to ensure that the pollution events at Cape Grim used to derive Port Phillip emissions are imbedded in air masses that only pass over the Port Phillip region and do not include other possible carbon monoxide source regions, in particular the Latrobe Valley.

A.5 Scaling

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. GWPs (to calculate CO_2-e emissions) are from Forster & Ramaswamy (2007); ODPs (to calculate ODP-weighted emissions) are from Montzka & Reimann (2011); assumed GWPs for trichloroethylene (0.67) and perchloroethylene (0.53).

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