Supporting Information document for Manuscript:

"High-global Warming Potential F-gas Emissions in California: Comparison of Ambient-based versus Inventory-based Emission Estimates, and Implications of Refined Estimates".

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# **Supporting Information**

The supporting information contains additional details to support the primary paper, and includes the following sections:

- 1. Emission Trends for Representative F-gases CFC-12, HCFC-22, and HFC-134a.
- 2. Ambient Air Measurement Data, Mount Wilson Sampling 2007-2008.
- 3. Uncertainty Analysis, Inventory-Based Emissions Estimates.
- 4. Methodology for Inventory-Based Emission Estimates of F-Gases.

# 1. Emission Trends for Representative F-gases CFC-12, HCFC-22, and HFC-134a:

The CARB Research Division uses current data and trends to project emissions estimates of Fgases through future time-frames, such as to 2020. Using the emission refinements including improved equipment end-of-life methodology, the following projected emissions of CFC-12, HCFC-22, and HFC-134a were estimated.



Figure S1. California estimated emissions, 2000-2020, for CFC-12, HCFC-22, and HFC-134a.

The vertical axis shows both emissions by mass (in thousands of metric tonnes [MT], which equal gigagrams [Gg]), and emissions by CO<sub>2</sub>-eq GHG impact (in MMTCO<sub>2</sub>E). Dashed lines indicate emissions by mass, and solid lines indicate emissions by GHG impact. The red lines with square markers show CFC-12, purple lines with circle markers show HCFC-22, and green

lines without markers show HFC-134a. GWP values are from the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report of 2007 (AR4) 100-year GWP values.<sup>1</sup>

The emissions of these representative F-gases comprise more than 65 percent of all F-gas emissions (by CO<sub>2</sub>-eq) in the state of California (in baseline year 2008), and thus reflect aggregated emissions of CFCs, HCFCs, and HFCs very closely. As expected, emissions from CFC-12 and HCFC-22 will continue to decrease as a result of the ODS production and import phase-out. Between 2000 and 2020, CFC-12 emissions are estimated to decrease by 92 percent, and HCFC-22 emissions are estimated to decrease by 59 percent. In the same time frame, emissions of HFC-134a are expected to increase by 87 percent, as HFCs continue to replace ODS refrigerants.

The refined emission estimates conducted by the authors were also used to estimate aggregate Fgas emissions in California from the AB 32 baseline year of 1990, through 2050, a benchmark year at which time the AB 32 GHG reduction goal is that annual GHG emissions in California will be 80 percent less than baseline year 1990 emissions of 427 MMTCO<sub>2</sub>E.<sup>2, 3</sup> With increasing HFC emissions, meeting the 2050 goal of 85.5 MMTCO<sub>2</sub>E from all GHG sources will be challenging without additional reduction measures.

# 2. Ambient Air Measurement Data, Mount Wilson Sampling 2007-2008:

A "top-down" approach to calculate the total CFC-12, HCFC-22, and HFC-134a emissions inventory for LA County is based on the F-gas to CO relationship observed at Mount Wilson and the CO emissions inventory. A scaling ratio method using correlation of pollutant mixing ratios above the background to estimate trace gas emissions was utilized. This "top-down" approach relies on the fact that the selected F-gases and CO are non-reactive on the time scale of dispersion. Carbon monoxide is an excellent tracer for observing global transport in the troposphere <sup>4</sup>. Its lifetime of one to three months depending upon season is long enough for the gas to be tracked as it rises from the surface and journeys around the globe. As stated in the paper, both F-gases and CO show a high collocation of being associated with human activity in urban areas.

The following graphs and tables show the selected F-gas and CO mixing ratios correlation as measured at Mount Wilson in 2007 and 2008. Annual F-gas emissions are estimated by correlating CO emissions to measured concentrations of the F-gas, using the following formula:

F-gas emission (metric tons/year) = (CO emission, metric tons/day) \* [(correlation slope)/(1000 ppt/ppb)] \* (High-GWP molecular weight/CO molecular weight)\*(365 day/year)

# Table and Figure for CFC-12:

CFC-12	April-May, 2007	September, 2007	November, 2007	February,				
				2008				
Slope	$0.17\pm0.02$	$0.10\pm0.01$	$0.10\pm0.01$	$0.05\pm0.01$				
Intercept	$525 \pm 3.2$	$532 \pm 2.5$	$531 \pm 3.0$	$541 \pm 2.4$				
Chi Sq	4105.65	6276.92	3689.86	1209.5				
Pearson's r	0.83	0.82	0.66	0.51				
Emissions in CO	1,339	804	804	377				
(MT)/day								
Uncertainty	107	67	110	81				
(MT)								
Annual MT CFC-12 = $303 \pm 33$								

**Table S1.** CFC-12 and CO mixing ratios correlation, CFC-12 annual emissions.



**Figure S2.** Correlation of CO in ppb (X-axis) and CFC-12 in ppt (Y-axis) Measured at Mt. Wilson

Table and Figure for HCFC-22:

HCFC-22	April-May	September	November	February				
Slope	$1.5264 \pm 0.105$	$1.2808 \pm 0.0571$	$0.29544 \pm 0.129$	0.81249 ±				
				0.049				
Intercept	$-39.906 \pm 24.7$	$84.832 \pm 16.6$	$353.22 \pm 27.4$	$100.06\pm11.3$				
Chi Sq	239915	279767	308151	26832.7				
Pearson's r	0.869043	0.936874	0.263435	0.903336				
Emissions in CO	8,488	7,122	1,643	4,518				
(MT)/day								
Uncertainty	584	318	717	272				
(MT)								
Annual MT HCFC-22 = 1987 ± 173								

**Table S2.** HCFC-22 and CO mixing ratios correlation, HCFC-22 annual emissions.



**Figure S3.** Correlation of CO in ppb (X-axis) and HCFC-22 in ppt (Y-axis) Measured at Mt. Wilson

Table	and	Fig	ure	for	HF	<b>C-</b>	134a:

HFC-134a	April-May, 2007	September, 2007	November, 2007	February,				
				2008				
Slope	$0.52\pm0.02$	$0.56\pm0.02$	$0.36\pm0.02$	$0.38\pm0.03$				
Intercept	$-34 \pm 4.9$	$-8.2 \pm 4.4$	$12 \pm 5.0$	$2.5\pm6.2$				
Chi Sq	9290.87	19779.8	10198.9	7568.03				
Pearson's r	0.95	0.98	0.88	0.88				
Emissions in CO	3,430	3,699	2,393	2,500				
(MT)/day								
Uncertainty	135	100	154	176				
(MT)								
Annual MT HFC-134a = 1097 ± 52								

**Table S3.** HFC-134a and CO mixing ratios correlation; HFC-134a annual emissions.



**Figure S4.** Correlation of CO in ppb (X-axis) and HFC-134a in ppt (Y-axis) Measured at Mt. Wilson

# 3. Uncertainty Analysis, Inventory-Based Emissions Estimates:

Uncertainty estimates for F-gas emissions are upper and lower bounds (one standard deviation from the mean) based on a 95 percent confidence interval, using sampled data and Monte Carlo simulations from probability distributions. Emission sectors were aggregated into one of five categories: large refrigeration and AC (2,000-lbs. or greater charge size); medium refrigeration and AC (200 - 2,000-lb. charge size); small refrigeration and AC (< 200-lb. charge size) including residential; motor vehicle AC; and all other sectors. A weighted-value based on the portion of emissions from each emissions sector was assigned to specific F-gases (CFC-12,

HCFC-22, and HFC-134a), and to the F-gas groups of CFCs, HCFCs, HFCs, and all other F-gases (miscellaneous F-gases of SF<sub>6</sub>, PFCs, PFC/PFPEs, NF<sub>3</sub>, and SO<sub>2</sub>F<sub>2</sub>).

Uncertainties based on sampled data were assigned to each part of the basic emissions formula (number of units-equipment in use, average F-gas charge per unit, average annual loss rate, number of units-equipment reaching end-of-life, and average loss rate at end-of-life). South Coast Air Quality Management District (SCAQMD) data were used for commercial stationary refrigeration and AC sectors,<sup>5,6</sup> and residential refrigeration and AC sectors were from CARB-sponsored research.<sup>7</sup> Mobile vehicle emissions data were from several CARB-sponsored studies and CARB Research Division analysis.<sup>8, 9, 10, 11, 12, 13</sup>

The emissions estimates from several emission sectors were based on CARB-sponsored research; uncertainties as stated by the researchers were used for insulating foam,<sup>14</sup> refrigerated shipping containers,<sup>15, 16</sup> and solvents and fire suppressants.<sup>17</sup> Industry surveys and CARB analysis on usage of consumer product aerosol propellants, PFCs, and sulfur hexafluoride is believed to capture more than 80 percent of usage from these sectors.<sup>18, 19, 20, 21, 22, 23, 24</sup> F-gas emissions from transport refrigerated units and shipping are correlated with activities in these two sectors, which are reported to CARB as part of diesel emission regulations.<sup>25, 26, 27, 28</sup> We estimate that reporting to CARB captures more than 75 percent of the emissions from the sectors. Sulfuryl fluoride usage has been reported to the California Department of Pesticide Regulation (DPR) since 1990 as required by regulation. We estimate over-reporting usage is unlikely, with a greater than 90 percent compliance rate in usage reporting.<sup>29</sup> Emission estimates for two of the emission sectors, medical sterilants and metered-dose-inhalers, comprising 4 percent of F-gas emissions, relied on scaling national emissions from the U.S. EPA to the state level.<sup>30, 31</sup> The U.S. EPA GHG inventory estimates an uncertainty of ± 14 percent in their estimates.<sup>31</sup>

A preliminary (2008) uncertainty for all F-gas emissions was estimated to be  $\pm$  22 percent. The refined uncertainty using greater dis-aggregation of sectors are :  $\pm$  20 percent for CFC-12;  $\pm$  17 percent for all other CFCs and Halons;  $\pm$  25 percent for HCFC-22;  $\pm$  24 percent for all other HCFCs;  $\pm$  14 percent for HFC-134a;  $\pm$  15 percent for all other HFCs; and  $\pm$  13 percent for all other F-gases (PFCs, PFC/PFPEs, NF<sub>3</sub>, SF<sub>6</sub>, and SO<sub>2</sub>F<sub>2</sub>).

# 4. Methodology for Inventory-Based Emission Estimates of F-Gases:

The methodology used to estimate F-gas emissions in California, using an inventory-based methodology is described in this section. The methodology description is roughly divided into two major sectors: 1) Refrigeration and AC; and 2) Non-Refrigeration, Non-AC sectors.

#### **Refrigeration and Air-Conditioning emission sectors:**

The following Table S4 shows input factors used to estimate emissions from refrigeration and AC equipment (table shown on the following page to preserve table continuity).

 Table S4: Input Factors for Emission Calculations, Refrigeration and AC (stationary, transport refrigerated units, and refrigerated shipping containers), 2008

Equipment Type or Emissions sub-sector	Units in CA	Ave. Charge (amount) of F-gas in Ibs.	Ave. Annual Leak (loss) Rate	Annual Loss in Ibs. (units* charge* loss rate)	EOL units in CA	Ave. Charge (amount) in Ibs. at EOL	Ave. EOL Loss Rate	EOL Loss in Ibs. (units* charge* loss rate)	total loss in lbs. (annual + EOL)
Refrigeration Large Centralized System ≥ 907.2 kg (2,000 lbs.)	1,090	2,486	21.0%	568,914	62	2,486	20%	30,908	599,822
Refrigeration Medium Centralized System 90.7-< 907.2 kg (200-< 2,000 lbs.)	33,269	704	15.0%	3,513,168	1,924	704	20%	270,845	3,784,013
AC Large Centrifugal Chiller ≥ 907.2 kg (2,000 lbs.)	4,231	3,978	2.4%	403,915	177	3,883	20%	137,808	542,000
AC Medium Centrifugal Chiller 90.7-< 907.2 kg (200-< 2,000 lbs.)	1,330	1,007	1.4%	18,751	56	993	20%	11,108	30,000
AC Chiller - Packaged 90.7-< 907.2 kg (200-< 2,000 lbs.)	8,379	526	6.9%	304,111	352	490	20%	34,505	339,000
Refrigeration Large Cold Storage ≥ 907.2 kg (2,000 lbs.)	1,166	7,546	21.6%	1,899,770	48	5,509	16%	42,581	1,942,000
Refrigeration Medium Cold Storage 90.7-< 907.2 kg (200-< 2,000 lbs.)	4,806	565	28.8%	782,075	202	362	16%	11,660	794,000
Refrigeration Process Cooling ≥ 907.2 kg (2,000 lbs.)	395	3,640	6.8%	97,798	17	3,393	20%	11,322	109,120
Refrigerated Condensing units 22.7-≤ 90.7 kg (50-≤ 200 lbs.)	65,154	122	14.5%	1,152,583	2,738	122	20%	66,817	1,219,000
Unitary AC 22.7-≤ 90.7 kg (50-≤ 200 lbs.)	65,265	100	11.3%	737,490	3,708	77	20%	57,106	795,000
Refrigerated Condensing Units $\leq$ 22.7 kg (50-lbs. or less) <sup>(a)</sup>	262,854	31.4	15%	1,238,043	11,010	22	34%	82,351	1,320,394

Equipment Type or Emissions sub-sector	Units in CA	Ave. Charge (amount) of F-gas in Ibs.	Ave. Annual Leak (loss) Rate	Annual Loss in Ibs. (units* charge* loss rate)	EOL units in CA	Ave. Charge (amount) in Ibs. at EOL	Ave. EOL Loss Rate	EOL Loss in lbs. (units* charge* loss rate)	total loss in lbs. (annual + EOL)
Refrigerated stand-alone display cases <sup>(a)</sup>	577,457	7.1	0%	0	24,446	7	100%	173,566	173,566
Refrigerated vending machines <sup>(a)</sup>	452,086	0.66	0%	0	25,524	0.66	100%	16,846	16,846
Unitary A/C $\leq$ 22.7 kg (50-lbs. or less) (central) <sup>(b)</sup>	2,367,328	15.1	10%	3,574,665	133,608	12	56%	905,326	4,480,000
Unitary A/C $\leq$ 22.7 kg (50- lbs. or less) (window unit) <sup>(b)</sup>	639,511	1.54	2%	19,697	50,929	1.2	100%	59,587	79,000
Residential Appliance (refrigerator-freezer)	16,189,879	0.5	1%	80,949	946,725	0.4	77%	313,461	394,000
Residential A/C (central) <sup>(b)</sup>	5,994,796	7.5	10%	4,496,097	322,452	6	56%	1,083,440	5,580,000
Residential A/C (window unit)	3,558,891	1.54	2%	109,614	283,422	1.2	100%	331,604	441,000
Transport Refrigerated Units (TRUs)	57,603	20.7	18.3%	218,208	4,580	17.4	15%	11,953	230,161
Refrigerated Shipping Containers	42,941	33.1	5%	71,068	12,853	33.1	19%	80,835	151,903

Table notes:

a) Emissions were calculated separately for refrigerated condensing units  $\leq$  22.7 kg (50-lbs. or less), refrigerated stand-alone display cases, and refrigerated vending machines, and then re-aggregated in emissions summary tables for display purposes.

b) Emissions from unitary AC  $\leq$  22.7 kg (50-lbs. or less) systems, both commercial and residential, were calculated for window units and central AC units separately; then re-aggregated in emissions summary tables for display purposes.

# Commercial Refrigeration and Air-conditioning

An inventory of GHG emissions was developed for the commercial refrigeration and AC sector in the development of the CARB Stationary Refrigerant Management Program regulation adopted December 2009. The methodology used to estimate baseline emissions and future emissions is comprehensively detailed in the regulation's Initial Statement of Reasons (ISOR) Appendix B for California Facilities and Greenhouse Gas Emission Inventories.<sup>32</sup> The following is a brief summary of the methodology used.

Reports submitted by facilities to the South Coast Air Quality Management District (SCAQMD) to comply with Rule 1415, "Reduction of Refrigerant Emissions from Stationary Refrigeration and Air Conditioning Systems" formed the basis for developing "emission profiles" using the follow input data: <sup>5,6</sup>

- Numbers and types of refrigeration and AC equipment used in California,
- Refrigerant capacity in pounds,
- Annual loss (leakage) rates, and
- Types of refrigerant used and their distribution for each equipment type.

CARB Research Division staff analyzed more than 30,000 equipment records from reports submitted to SCAQMD for Rule 1415. Equipment profiles were developed for 12 sub-types of refrigeration and AC equipment, each with its own profile of refrigerant types used, average refrigerant charge size, and average annual loss (emissions). End-of-life loss rates were derived from commercial refrigeration and AC research for CARB,<sup>33</sup> U.S. EPA Vintaging Model estimates,<sup>30</sup> and UNEP reports.<sup>34, 35</sup>

Commercial refrigeration or AC units containing less than 50 pounds of refrigerant are exempt from Rule 1415 reporting, and SCAQMD records for these smaller systems do not exist. Therefore, the source of data for these units was research conducted by Saba, et al., 2009. The refrigeration sector was further broken out into the following types of equipment:

- Refrigerated condensing units (centralized or distributed systems)
- Refrigerated stand-alone display cases
- Refrigerated vending machines

To determine emissions from AC units containing less than 50 pounds of refrigerant, it was necessary to further divide this sector into central AC units used in commercial buildings, central AC units used for residential, and window AC units (commercial and residential units).

Refrigerant emissions are directly proportional to the number of equipment in use, and the number reaching end-of-life each year. As a proxy for refrigeration sales (lacking more specific sales figures), we will assume that food sales and refrigeration sales correlate well. Saba, et al. estimated that the growth in sales of refrigeration equipment was 1.7 percent annually between

1990 and 2007, and the growth in sales of AC equipment was 2.5 percent annually between 1990 and 2007.

Due to the economic downturn of the late 2000s, the refrigeration equipment sales rates were adjusted downwards beginning in 2007. Although food sales are extremely resilient to economic downturns, it was assumed that sales of new refrigeration equipment for food manufacturing, distribution, storage, and retail sales would decrease proportionally to food sales (adjusted for inflation). Food sales were estimated by the United States Department of Agriculture (USDA) to increase only 0.6 percent in 2007, and decrease one percent annually in 2008 and 2009<sup>36</sup> (although the decrease was estimated to be slightly less at 0.5 percent annually in 2009 by the Food Marketing Institute).<sup>37</sup> The USDA also estimated that food sales adjusted for inflation were estimated to increase 1.5 and 1.4 percent annually in 2010 and 2011. To simplify future growth estimates, we assume that refrigeration equipment sales (and emissions) increase proportionally to population growth in California, estimated to remain at about one percent annually for the foreseeable future, based on historical population growth.<sup>38</sup>

The AC equipment sales rates were also adjusted to the economic downturn beginning in 2007. Based on US Department of Commerce (DOC) wholesale trade surveys, we used the wholesale trade surveys for NAICS code 42 (Wholesale Trade) as the closest approximation for the more specific NAICS code that includes AC sales, NAICS 421730 "Warm air heating & air-conditioning equipment and supplies wholesale". Inflation-adjusted refinements made to the historic 2.5 percent annual increase in the AC sales growth rate were as follows: no increase or decrease in 2007, 7.2 percent decrease in 2008, 17.0 percent decrease in 2009, and a 4.3 percent annual increase in both 2010 and 2011.<sup>39, 40</sup> For 2012 and future years, we conservatively estimate half the traditional growth rate in this sector, or a 1.3 percent annual growth (to be updated and replaced periodically by actual sales data).

<u>Refined Emissions Estimates:</u> The F-gas emissions from the commercial refrigeration and AC sectors were continually refined between 2009 and 2012 as additional research and data was analyzed and used to change emission input factors where appropriate. For some sectors, the refinements were minimal and did not result in measurable changes to F-gas emissions. Other sectors underwent more substantial changes. As noted in this supporting methodology and the technical paper it supports, the greatest change in emissions estimates occurred as a result of using a more sophisticated "survival curve" or "end-of-life function curve".

Changes to the speciation of emissions by sector were largely due to the changes in the end-oflife equipment function curve, where older equipment remained in use longer than previously estimated. Additional minor changes to the speciation of emissions were due to small changes to the assigned refrigerant type used at the time of equipment manufacture.<sup>6, 30, 33</sup>

The following section describes, for each commercial refrigeration and AC sub-sector, additional changes made to the initial assumptions and emissions factors used that resulted in refined emissions estimates.

Refrigeration Large Centralized System  $\geq$  907.2 kg (2,000 lbs.): Due to a facility numbers reanalysis by CARB Research Division staff, the estimated number of facilities with centralized refrigeration systems ( $\geq$  907.2 kg) was reduced by 32% from initial unrefined estimates for baseline year 2008. CARB staff conducted four surveys between 2009 and 2012 to commercial facilities with North American Industrial Classification System (NAICS) codes of business types most likely to use refrigeration systems with more than 50 pounds of high-global warming potential refrigerant (the threshold established for registration and reporting requirements for the CARB Refrigerant Management Program rule). Initial identification NAICS codes of facilities most likely affected were based on the annual reports submitted between 2002 and 2011 to the South Coast Air Quality Management District (SCAQMD) for compliance with Rule 1415 "Reduction of Refrigerant Emissions from Stationary Refrigeration Systems". Survey results and follow-up calls to facilities were used to estimate the number of facilities within each NAICS code (or broad category of business type) that used stationary refrigeration systems with more than 50 pounds of high-GWP refrigerant.

Based on a careful re-analysis of the emissions data from the SCAQMD, the average leak rate was also very slightly revised from 21.4% annually to 21.0% annually.<sup>6</sup> Subsequently, as a result of the reduction in facility numbers, and the slight reduction in the average annual leak rate, the estimated emissions for baseline year 2008 were reduced by 36%, from 426,740 kg to 272,000 kg (940,800 lbs. to 600,000 lbs.). An ODS refrigerant blend, R-502 (comprised of CFC-115 and HCFC-22) was also added to the refined speciation, as a result of reviewing the Rule 1415 records from SCAQMD reporting beginning with year 2002.

Refrigeration Medium Centralized System 90.7-< 907.2 kg (200-< 2,000 lbs.): The number of facilities with centralized refrigeration systems between 90.7 and 907.2 kg were adjusted downwards by 3%, with the corresponding equivalent decrease in estimated F-gas emissions. The speciation changes were the same as those for the larger centralized refrigeration systems, as it is assumed that the refrigerants used are the same between large and medium sized centralized systems.

Refrigeration Large Cold Storage  $\geq$  907.2 kg (2,000 lbs.): As with the facility number re-analysis for large centralized systems, the number of facilities with large cold storage systems was also decreased by 32%. The assumed end-of-life loss rate of about 25% that had been used was simply a best estimate, and based upon additional research, was revised down to 20%.<sup>7</sup> The estimated emissions were reduced by 40%, from 1,473,600 kg to 880,900 kg (3,248,800 lbs. to 1,942,000 lbs.).

Refrigeration Medium Cold Storage 90.7-< 907.2 kg (200-< 2,000 lbs.): The number of facilities with medium-sized cold storage systems was adjusted downwards by 12%, with the corresponding equivalent decrease in estimated F-gas emissions, from 409,300 kg to 360,150 kg (902,300 lbs. to 794,000 lbs.). The speciation changes were the same as those for the larger cold storage systems, as it is assumed that the refrigerants used are the same between large and medium sized cold storage facilities.

Refrigeration Process Cooling  $\geq$  907.2 kg (2,000 lbs.): The number of process cooling (industrial cooling) facilities were adjusted downwards by 33% due to the refined facility

numbers analysis. The emissions were similarly reduced as a result of fewer facilities, from 74,300 kg to 49,500 kg (163,800 lbs. to 109,000 lbs.).

Refrigerated condensing units  $22.7 \le 90.7$  kg ( $50 \le 200$  lbs.): Refined emissions estimates were 9% less than initial estimates, with emissions reduced from 609,200 kg to 553,000 kg (1,343,000 lbs. vs. 1,219,000 lbs.), due to a slight readjustment in the number of facilities that use refrigerated condensing units. Due to the end-of-life survival curves, the distribution of refrigerant emissions increased measurably for CFC-12, and decreased for HFC-134a.

Refrigerated Condensing Units  $\leq 22.7$  kg (50 lbs.): Emission estimates from this subsector were refined when two additional emissions sources (stand-alone refrigerant cases, and refrigerated vending machines) were added and analyzed separately. All three types of equipment within this category have very different emissions profiles which did not allow for very accurate emissions estimates using average charge sizes, leak rates, and speciations, when all three sources were aggregated together. Due to disaggregated emissions calculations, the emissions increased by 7%, from 641,400 kg to 685,000 kg (1,414,000 lbs. to 1,511,000 lbs.). The condensing units contributed 87% of the emissions for this aggregated sector, the stand-alone units contributed 12% of the emissions, and the vending machines contributed only 1% of the emissions.

AC Large Centrifugal Chiller  $\geq$  907.2 kg (2,000 lbs.): Emissions estimates increased 16% when initial estimates were refined, from 212,900 kg to 246,000 kg (469,400 lbs. to 542,000 lbs.), largely as a result of refining the average annual leak rate from 2% to the more accurate 2.4%, as indicated by actual refrigerant usage records for chillers provided by SCAQMD.<sup>6</sup>

AC Medium Centrifugal Chiller 90.7-< 907.2 kg (200-< 2,000 lbs.): Emissions estimates increased 35% when the average annual leak rate of 1% was refined to the more accurate 1.4% average annual leak rate, as indicated by actual refrigerant usage records for medium-sized chillers provided by SCAQMD.<sup>6</sup> The change in leak rate inputs changed the initial estimated emissions in 2008 from 10,000 kg to 13,600 kg (22,100 lbs. to 30,000 lbs.).

AC Chiller - Packaged 90.7-< 907.2 kg (200-< 2,000 lbs.): Initial emissions estimates of 157,600 kg (347,500 lbs.) were very slightly revised downwards 2% as a result of the facility numbers refinement. Due to the changes in how equipment end-of-life survival curves were estimated, the speciation of emitted F-gases also changed slightly, with greater CFC emissions and fewer HCFC emissions.

Unitary AC 22.7- $\leq$  90.7 kg (50- $\leq$  200 lbs.): Equipment numbers were adjusted downwards 6.5% due to more recent data on the slowing growth trends of AC equipment sales,<sup>39</sup> revised end-of-life function, and the facility numbers re-assessment by CARB.<sup>41</sup> Estimated emissions were reduced by 9%, from 396,300 kg to 361,000 kg (873,800 lbs. to 795,000 lbs.).

Unitary A/C  $\leq$  22.7 kg (50-lbs. or less) (central and window units): For this smallest category of commercial AC, initial emission profiles did not break out AC systems into their two main components, central AC, and window units, which have very different average refrigerant charge sizes (6.8 kg for central AC [15.1 lbs.], and 0.7 kg for window units [1.5 lbs.]). Because the window units have small charge sizes and are hermetically sealed, they were thought to emit

minimal amounts of refrigerant, and were not included in initial estimates, but for inventory completeness, small window AC units were included in the refined emissions estimates. The window units were found to contribute approximately 2% of the F-gas emissions from this smallest commercial AC sub-sector.

Despite adding window units as a new emissions source category to this small-size commercial AC sector, the refined emissions estimates resulted in an estimated 21% decrease in emissions, from 2,611,000 kg to 2,068,000 kg (5,757,000 lbs. to 4,559,000 lbs.). The decrease in emissions can be attributed to the assumed charge size remaining at central AC end-of-life. Rather than being fully charged, it was assumed that prior to the equipment's end-of-life, it had leaked 10% of its charge for the last two years of its life, without refrigerant replacement, decreasing its average end-of-life charge amount by 20%.<sup>7</sup>

### Residential stationary refrigeration (refrigerator-freezers) and AC

Residential refrigerator-freezer emission estimates are derived from Mathis, et al., 2011,<sup>7</sup> and original CARB analysis of potential rulemaking for residential refrigerators,<sup>42</sup> with additional data on numbers of units in use and disposed of annually, average unit lifetime, and refrigerant usage data.<sup>43, 44, 45, 46</sup> The GHG contribution from refrigerator-freezer waste insulating foam is included in the separate emissions category of insulating foam.

Refrigerant emissions from refrigerant-freezers are directly proportional to the number of units used (or disposed of) on an annual basis, which is in turn directly related to the number sold for use in California in a given year. Refrigerant-freezer appliance use is estimated to have grown 2.8 percent annually between 1990 and 2007, with actual sales decreasing on average 1.5 percent per year between 2008 and 2011.<sup>7</sup> Our best estimate is that sales are expected to remain flat from 2012 through 2015, when sales will increase proportional to expected population growth of one percent annually (assumptions will be re-checked periodically for future emissions estimates).

Residential air-conditioning GHG emissions were based on equipment numbers and emission profiles as researched by Saba et al., supplemented by Vintaging Model data,<sup>30</sup> UNEP reports,<sup>34, 35, 47</sup> and applying the emissions calculation methodology used for the small AC systems in commercial facilities.<sup>32</sup> The growth in window units (and their emissions) was estimated at a slight increase of 0.1 percent annually, and the growth in central units (and their emissions) was estimated at 2.5 percent annually.<sup>33</sup> The projected growth rate was adjusted for years 2008 through 2020 due to the economic downturn, especially in housing starts which account for many of the residential AC new sales. The same downward adjustments used for refrigerator-freezers were made for years 2008 through 2014, and the same growth estimates were used for 2014 through 2020.

<u>Refined Emissions Estimates:</u> For residential refrigerator-freezers, initial estimates were based on preliminary CARB analysis of GHG emissions from this sector.<sup>42</sup> Subsequent research by Mathis, et al., 2011 was used to refine the emissions profile by clarifying the annual refrigerant leak rate (a low 1% annual leak rate on average, due to the hermetically-sealed nature of residential appliances), low refrigerant charge amount (0.23 kg, or 0.5 lbs.), and the percentage

of appliances recycled (15% or greater) with a very high confidence of refrigerant recovery.<sup>7</sup> The refinements led to an estimated 27 percent decrease in overall emissions by mass, from 243,600 kg to 179,000 kg (537,000 lbs. to 394,000 lbs.). The initial speciation profile assumed that appliances only functioned for 14 years, and therefore all pre-1995 appliances using CFC-12 refrigerant would no longer be in use after the year 2007. Based on a better end-of-life retirement function, the CFC-12 emissions from household refrigerator-freezers in 2008 were revised from 0 percent of emissions (by mass) to 32 percent of emissions, with the remaining 68 percent of emissions from HFC-134a.

For residential AC, initial emission profiles did not break out AC systems into their two main components, central AC, and window units, which have very different average refrigerant charge sizes (3.4 kg for central AC [7.5 lbs.], and 0.7 kg for window units [1.5 lbs.]). Because the window units have small charge sizes and are hermetically sealed, they were thought to emit minimal amounts of refrigerant, and were not included in initial estimates, but for inventory completeness, small window AC units were included in the refined emissions estimates. The window units were found to contribute approximately 8% of all residential AC emissions.

Despite adding window units as a new emissions source category to the residential AC sector, the refined emissions estimates resulted in an estimated 39% decrease in emissions, from 4,458,600 kg to 2,731,000 kg (9,829,600 lbs. to 6,021,000 lbs.). The decrease in emissions can be attributed to a re-assessment of the assumed growth in housing starts, which decreased in 2008 compared to historical trends, due to the economic downturn (it is assumed that all new houses in California are constructed with central AC units). Also, changes were made to the assumed charge size remaining at central AC end-of-life. Rather than being fully charged, it was assumed that prior to the equipment's end-of-life, it had leaked 10% of its charge for the last two years of its life, without refrigerant replacement, decreasing its average end-of-life charge amount by 20%. Additional refinements were made to the assumed 70% loss at end-of-life, which were decreased to a 56% loss rate.<sup>7</sup>

#### Mobile Air-conditioning (MAC) sector

Refrigerant emissions from MAC systems occur as assembly loss, regular leakage, irregular loss due to accidents, stone hits, or component failure, service loss, and end-of-life loss.

MAC emissions were based on a comprehensive emissions model developed by CARB staff and CARB-funded research and analysis in the development of potential regulations to reduce GHG emissions from MVAC sources.<sup>8, 9, 10, 11</sup> The basis of the refrigerant emissions are from vehicle data analyzed through the CARB emissions models EMissions FACtor (EMFAC) and OFFROAD. Vehicle emission profiles were developed for light duty and heavy duty vehicles, buses, and off-road heavy duty vehicles.

Table S5 is a partial summary of the types of MAC data and analysis used to determine MAC emissions, however note that the actual emissions model is more sophisticated than the summary factors shown below for reference.

Mobile AC sectors	Units in CA	Ave. Charge (amount) of F-gas in lbs.	Ave. Annual Leak (loss) Rate	Ave. EOL Loss Rate	total loss in lbs. (annual + EOL) 2011
Mobile Vehicle AC (MVAC) Light-Duty Vehicles	22,500,000	1.52 – 3.02 <sup>(a)</sup>	10.1% - 13.1% <sup>(b)</sup>	30%	4,002,497
MVAC Heavy-Duty Vehicles (non-bus) <sup>(c)</sup>	1,100,000	variable	0.79 lbs./yr	0.12 lbs./yr	1,128,473
MVAC Off-road Heavy-Duty Vehicles <sup>(c)</sup>	300,000	variable	0.79 lbs./yr	0.12 lbs./yr	173,910
MVAC Buses <sup>(d)</sup>	55,900	variable	2.55 lbs./yr	0.40 lbs./yr	276,416

 Table S5:
 Mobile AC Sectors and Emissions Data Summary

Table notes:

a) The average charge size for light-duty vehicles between model years 1965 through 2007 was 3.02 lbs. Beginning model year 2008, light-duty vehicle AC systems were manufactured with a significantly reduced average refrigerant charge of 1.52 lbs.

b) The average annual loss rate for vehicle model years 1965 through 2007 was 13.1%. Beginning model year 2008, light-duty vehicle AC systems were estimated to lose on average 10.1% of their refrigerant charge annually.

c) For heavy-duty vehicles, both on-road (non-bus) and off-road, the average emissions are based on mass-balance computations derived from CARB emissions models EMFAC and OFFROAD. The average emissions from leakage are 0.79 lbs./year and annualized end-of-life losses are 0.12 lbs./year, for a total annualized average loss 0.91 lbs./year per vehicle for heavy-duty vehicles (on-road and off-road).

d) For buses, the same note as above, except the average emissions from annual leakage are 2.55 lbs./year and annualized end-of-life losses are 0.40 lbs./year, for a total annual average loss of 2.95 lbs./year per bus.

To estimate the emissions of HFC-134a from light-duty vehicles, a model has been developed by CARB to balance the amount of refrigerant added into an MVAC system (mass-in) and the amount of refrigerant emitting from or pulled out of the system (mass-out) over the system's lifespan. The mass out of the system would become emissions to the environment unless it is recovered for recycling, reclamation, or destruction. The model parameterizes all the mass-in and mass-out terms except the number of AC service in the system's lifespan. The resulting mass balance equation is then solved for the number of AC service. The mass-out terms, together with the number of AC service, are then used to estimate refrigerant emissions.

The emissions of CFC-12 from light-duty vehicles cannot be accurately estimated directly by the lifetime mass balance model. This is mainly because CFC-12, as a controlled substance in the

US after 1996 pursuant to the Montreal Protocol, is much more expensive and less available than HFC-134a. Thus, an important model assumption that an AC system gets recharged when it needs it, does not apply to CFC-12 AC systems after 1996. In addition, the model assumes that every AC system retires at the average lifespan of all the AC systems (16 years in California as estimated previously in a CARB study).<sup>12</sup> This would dictate that every in-use vehicles equipped with CFC-12 AC systems generate no CFC-12 emissions after 16 years of age, thus artificially causing the estimate for CFC-12 emissions to abruptly decline starting from 2008 and decrease to zero in 2010 when all the CFC-12 AC systems would have retired under that assumption.

Therefore, instead of using the aforementioned model to estimate the CFC-12 emissions, those emissions are estimated based on the population estimates for in-use CFC-12 fleet and analogy to HFC-134a emission factors (per vehicle emissions), while taking into account the higher possibility that a CFC-12 AC system would not get the necessary recharge to keep it operational, as compared with an HFC-134a AC system. This method allows for a gradual retirement of the CFC-12 AC systems that emulates the reality.

The information relevant to refrigerant emissions from heavy-duty on-road (non-bus) and offroad vehicles is inadequate and cannot inform a detailed parameterization of the mass-in and mass-out terms in the lifetime mass balance model. Therefore, an average HFC-134a leak rate suggested in a CARB study for heavy-duty vehicles is scaled up to account for other types of emissions using the same ratio of leak rate to overall emission factor for light-duty vehicles estimated by the lifetime mass balance model. <sup>9</sup>

The same method is used to estimate the leakage and overall emissions of CFC-12 from the heavy-duty on-road and off-road sector. Unlike light-duty vehicles, it is assumed that heavy-duty vehicles and equipment would always receive the necessary recharge, regardless of the type of refrigerant in use. This is because heavy-duty vehicles and equipment are mainly for industrial and commercial use. Therefore, their owners are expected to be more capable of affording the cost for AC service than individuals driving their personal light-duty vehicles, and would be reluctant to compromise the productivity and safety of the drivers/workers by forgoing AC service. Such methods allows for gradual phase-out of CFC-12 emissions along with the gradual retirement of the vehicles using this refrigerant as reflected in the population projection.

Data associated with refrigerant emissions for buses are also insufficient. Therefore, an average HFC-134a emission factor suggested in a CARB study is used to estimate the overall emissions of HFC-134a for the entire in-use bus fleet.<sup>9</sup> The same emission factor is assumed to be also applicable to bus AC systems that use CFC-12 and HCFC-22. Like heavy-duty on-road vehicles and off-road vehicles and equipment, bus AC systems are also assumed to always receive necessary recharge because they are for commercial use. Similar to the cases for the heavy-duty on road fleet and off-road fleet, such a method allows for gradual phase-out of CFC-12 emissions.

<u>Refined Emissions Estimates:</u> Initial emissions estimates were simple scaled estimates from U.S. EPA national estimates for the mobile vehicle sector, where it was assumed that all HFC emissions were from HFC-134a. An in-depth analysis conducted by CARB staff to estimate

MVAC emissions led to the development of a comprehensive emissions model specific for the state of California.<sup>13</sup>

MVAC emissions were dis-aggregated into four separate sub-sectors: MVAC light-duty (LD) vehicles; MVAC heavy-duty (HD) vehicles (non-bus), MVAC bus, and MVAC off-road.

Refined emissions estimates specific to the vehicle mix and emission profiles of vehicles operating in California resulted in significantly fewer MVAC emissions than initially estimated. MVAC emissions decreased from an initial estimated 4,691,600 kg to 2,316,700 kg (10,343,300 lbs. to 5,107,500 lbs.), a decrease of 51 percent. The estimated annual leak rates as researched and analyzed were apparently much lower than historically assumed annual leak rates (nationally), although the end-of-life refrigerant recovery was still believed to be relatively low.

Initial speciation estimates assumed no CFC-12 emissions, 0.3% from HCFC-22 (bus AC is the only source of HCFC-22), and the remaining 99.7% from HFC-134a. Refined emissions speciations were as follows for the four sub-sectors:

- MVAC LD: CFC-12 (5.3%) and HFC-134a (94.7%);
- MVAC HD (non-bus): CFC-12 (19.9%), HFC-134a (80.1%);
- MVAC bus: CFC-12 (20.2%), HCFC-22 (43.2%), HFC-134a (36.6%); and
- MVAC off-road: CFC-12 (13.1%), HFC-134a (86.9%).

The changes in the proportion of CFC-12 and HFC-134a are primarily a function of the different average vehicle lifetimes.

# Refrigerated Shipping Containers

Refrigerated shipping container emissions at California ports were estimated from the methodology and refrigerant loss as outlined in CARB white paper "Defining and Determining Emissions from Refrigerated Shipping Containers and the Total Refrigeration Process in the Vicinity of Shipping Ports",<sup>15</sup> and significantly improved and refined by the additional CARB-funded research project "Evaluation of Potential for Refrigerant Recovery from Decommissioned Shipping Containers at California Ports".<sup>16</sup>

The methodology is essentially the same as that used for other refrigeration equipment, using refrigerant charge size, average leak rates, and refrigerant loss at end of life.

It was determined that refrigerated shipping containers (RSCs) are managed extremely well due to the high value of their cargo, and that the RSCs leaked very little in the first few years of their use. By the end of their useful life (10 years on average), they were leaking 10 percent of their refrigerant each year, for an average leak rate over the equipment lifetime of 5 percent annually.

The RSC end-of-life emissions were different than other refrigeration/AC equipment, in that they were determined to be of two distinct types of EOL loss: 1) Loss at the time of planned decommissioning, and 2) catastrophic loss due to accidental damage.

The refrigerant recovery at the container's planned decommissioning was estimated at 85 percent recovery, for a loss of 15 percent at EOL. However, a 100 percent catastrophic loss of refrigerant occurred when RSCs were involved in an accident that breached the refrigeration equipment. It was estimated that the accident rate (resulting in total loss of refrigerant) is about 0.5 percent of RSCs annually, with about half occurring on their way to California that would be counted as CA emissions, for a total loss accident rate of 0.25 percent (1/2 of 0.5 percent).

Based on the RSC study data, the weighted EOL loss average of 15 percent for decommissioned units and 100 percent for units reaching EOL by accident was 19 percent on average for each EOL unit.

The average refrigerant charge of 15 kg is 33.1 lbs., and due to the excellent management of the typical RSC, it is believed that they have a full refrigerant charge at their end-of-life.

Determining the number of units in California during a given year required a specific methodology, as refrigerated shipping containers are highly transient pieces of equipment. The number of units in California was estimated by first converting the 1.66 million containers that were in California (during the baseline emission year of 2010) for an average stay of 10 days, which resulted in annual "full-time equivalents". (1,666,000 containers \* 10 days/365 days/year = 45,640 "full-time equivalent" containers/year.)

The annual growth rate of emissions between 2000 and 2008 was estimated to be the same as the growth in the number of refrigerated shipping container traffic to California, at 7 percent per year.<sup>16</sup> Growth between 2008 and 2010 was estimated at 3 percent annually, and post 2010 growth was conservatively assumed at no more than 1.5 percent annually.

Based on the post-2010 growth rate in shipping container traffic, we estimate that in 2011, the number of "full-time equivalent" refrigerated shipping containers in California was 46,325 units. Note that because the baseline year for RSCs was 2010, 2008 estimates were back-calculated based on the growth/reduction change in units in use per year.

The following emissions equation was used to determine RSC emissions:

*Emissions (lbs.)* = [number of full-time-equivalent RSCs in CA \* average F-gas charge (lbs.)/unit \* average annual leak rate]

+ [number of units reaching end-of-life (EOL) from planned decommissioning \* average F-gas charge (lbs.)/unit \* average loss rate at decommissioning EOL]

+ [number of units reaching EOL through accident \* average F-gas charge (lbs.)/unit \* catastrophic loss rate from EOL as a result of an accident]

The calculated emissions are shown below:

*Emissions* (*lbs.*) = [46,325 units in CA \* 33.1 lbs./unit \* 5% annual leak rate]

+ [13,195 units reaching EOL) from planned decommissioning \* 33.1 lbs./unit \* 15% EOL loss rate]

+ [660 units reaching EOL through accident \* 33.1 lbs./unit \* 100% EOL catastrophic loss rate]

Emissions (lbs.) = 76,596 (annual loss) + 65,452 lbs. (EOL decommissioning) + 21,817 lbs. (EOL, accident)

*Emissions (lbs.)* = 163,865 *lbs. total* 

Note that the two separate steps in calculating EOL emissions can be combined into one step by using a weighted average loss at EOL. Based on the RSC study data, the weighted EOL loss average of 15 percent for decommissioned units and 100 percent for units reaching EOL by accident was 19 percent on average for each EOL unit.

<u>Refined Emissions Estimates</u>: Preliminary emissions estimates for this sector originally included it as one aggregated estimate for all (non-MVAC) transport refrigerant emissions, which have subsequently been broke out into TRUs, refrigerated shipping containers, and ships (marine vessels). Subsequent efforts were made to dis-aggregate transport refrigerant emissions, which led to a greater refinement in the emissions estimates. The first effort at refining the estimates from the refrigerated shipping container sub-sector prior to CARB-funded research, was to use the best estimate assumption that refrigerant charge.<sup>15</sup> The 2006 Report of the Refrigeration, Air Conditioning and Heat Pumps Technical Options Committee (RTOC) report from the United Nations Environment Programme (UNEP) estimated that intermodal refrigerated shipping containers had an average annual leak rate of 15 percent.<sup>35</sup> However, the CARB-funded research determined that the historically high leak rates as reported may have never been quite as significant as estimated, and at any rate, leak rates by the 2008-2010 timeframe had been reduced to just 5% percent annually.<sup>16</sup> Therefore, the annual refrigerant emissions from this sector were reduced by a factor of 5 times less than originally estimated.

# Transport Refrigerated Units (TRUs)

TRU emissions were developed from equipment numbers estimated in TRU regulation support documents,<sup>25</sup> TRU data reported to CARB by the regulated sources,<sup>26</sup> and TRU regulation staff analysis on average refrigerant charge sizes.<sup>27</sup> The TRU sector includes the following sources and emission factors:

TRU type	Units	Time	Charge	Annual	EOL	EOL	EOL
	in CA	in CA	size	Leak	units in	cnarge	loss
			(105.)	Tate	CA	(lbs.) <sup>(a)</sup>	Tate
Large refrigerated trailers over 25 feet	25,885	100%	22.0	24%	1,991	16.7	15%
Mid-size refrigerated trailers between 11 and 25 feet	6,781	100%	12.0	24%	522	9.1	15%
Mid-size refrigerated trailers between 11 and 25 feet (out-of-	106,721	12.5%	12.0	24%	8,209	9.1	15%
state)	0.1.6	1000/	1.0	2.40/	10	2.0	1.50/
than 11 feet	246	100%	4.0	24%	19	3.0	15%
Refrigerated shipping containers not in ports	7,320	100%	33.1	5%	665	31.4	15%
Refrigerated shipping containers not in ports (out-of-state)	29,124	12.5%	33.1	5%	2,648	31.4	15%
Rail cars	7,189	12.5%	33.1	24%	553	25.2	15%
Weighted average	58,110	n/a	20.7	18.3%	4,623	17.4	15%

Table S6: Transport Refrigerated Unit (TRU) Input Factors for Emission Calculations

Notes: a) For TRUs, it is assumed that during the last year of its useful life, no top-off of refrigerant occurs. The calculation for EOL charge size = Charge size - (charge size \* annual leak rate).

The number of TRUs for each type was available for years 2000 through 2010. The numbers of TRUs were further analyzed by placing them into two separate categories: 1) TRUs used in-state (completely or almost completely), and 2) TRUs registered out-of-state, but occasionally used within the state. It was assumed that TRUs registered in the state were used completely within the state. For those TRUs entering from out-of-state, it was assumed that the time they spent in state was proportional to California's share of national population, which was 12.5 percent during the baseline year. Thus, the number of out-of-state TRUs used occasionally within the state was converted to "full-time equivalents" based on the number of out-of-state TRUs registered for use within California.

The charge size and types of refrigerant used were also referenced against U.S. EPA and UNEP information.<sup>48, 49</sup> Refrigerant loss rates were from research by D. Godwin,<sup>50</sup> with additional input from Dutch transport research.<sup>51</sup> Because TRU refrigerant losses at EOL are poorly quantified, they were estimated to be the same as refrigerated shipping containers comparable in size and function to the TRU systems.<sup>16</sup>

Refrigerant emissions from TRUs are assumed to be directly proportional to TRU traffic and numbers in use, although it should be noted that even idled or mothballed TRUs can still leak refrigerant if it has not been removed from the system. Measured data from the CARB TRU program was used to estimate emissions between 2000 and 2010, which averaged a two percent annual growth rate. Due to a slowing economy, it was assumed that no growth would occur between 2010 and 2015, with an assumed growth rate of one percent annually for years 2016 through 2020. These assumptions will be checked against actual TRU numbers in use in California as reported to CARB annually.

<u>Refined Emissions Estimates:</u> As indicated for refrigerated shipping containers, all non-MVAC transport refrigerant emissions were originally aggregated, which were subsequently disaggregated into the sub-sectors TRUs, refrigerated shipping containers, and ships (marine vessels). U.S. EPA Vintaging Model scaled-down estimates formed the basis of California emissions estimates, where it was simplistically assumed that all CFC emissions were from CFC-12, all HCFC emissions were from HCFC-22, and all HFC emissions were from HFC-134a. The actual number of TRUs in operation in California and annual leak rates were not taken into consideration in the earliest estimates from the transportation sector.

# Ships (Marine Vessels)

The same basic formula used to estimate refrigerant emissions for stationary equipment was used to estimate ship refrigerant emissions.

However, there is no single "typical" ship refrigerant or AC profile; eight separate types of ship refrigeration or AC systems were identified. Before total ship refrigerant emissions could be estimated, it was first necessary to break out the different types of vessels that each had slightly different emissions profiles. Ship sectors are listed the following table.

Ship Type	Units in CA (full- time-equivalents)	Charge size (lbs.)	Annual leak rate
Merchant ships (direct refrigeration)	65	441	40%
Merchant ships (indirect refrigeration)	7	110	20%
Naval ships	14	441	40%
Large fishing vessels (25 meters or longer)	8	3,977	40%
(direct refrigeration)			
Large fishing vessels (25 meters or longer)	3.5	1,989	20%
(indirect refrigeration)			
Small fishing vessels (less than 25 meters)	513	36.5	39%
Cruise ship AC	5.4	13,228	40%
Cruise ship refrigeration	5.4	882	40%

Table S7: Ship Refrigeration and AC Sectors and Emissions Data Summary

Notes:

a) A "full-time-equivalent" approach was used to normalize the mobile and transitory nature of shipping, where the number of hours (in California waters) for each type of vessel was aggregated, then divided by 8,760 hours per year to derive the number of full-time "ship-years" in California waters.

Refrigerant emission factors from ships (marine vessels) greater than 25 meters long or more than 100 gross tonnes) were based upon UNEP data for average refrigerant charge sizes, annual leak rates, and distribution of refrigerant types.<sup>34, 35</sup> The UNEP data were used for large marine vessels including merchant ships and navy ships. Two sub-categories of merchant ships, large fishing vessels and cruise ships, had more specific emission factors assigned to them, taken from a 2007 Öko-Recherche and Ecofys study on European shipping HFC emissions.<sup>52</sup> Another sub-category not large enough to be classified as a merchant ship is the small fishing vessel category, less than 25 meters long. For this category, the Öko-Recherche and Ecofys emission factors were also used. The average annual leak rates appear to be greater than similar stationary equipment, and it is possible or even likely that annual leak rates have decreased since the UNEP and Öko-Recherche studies were published. Annual leak rates will be updated as more recent UNEP reports and other studies become available.

Unlike stationary equipment, ships are highly mobile and spend most of their time away from port. Therefore, to find the "number of refrigeration or AC units in use", it was first necessary to determine the number of vessels in California ports, harbors, and waters (within 12 miles of shore), and then determine the amount of time they spent in those waters for a given year. The number of large marine vessels and the time they spent California waters, ports, and harbors were determined from data reported to CARB as part of the California shipping emissions reduction program, and analyzed using data model "CARB Marine Model" (for fuel combustion emissions), version V2.3J.<sup>28</sup>

For each marine vessel category, the number of hours spent in California waters, harbors, and ports were aggregated. The aggregate hours were divided by 8,760 hours per year (24 hours/day \* 365 days/year) to calculate the number of "ship-year-equivalents", before average annual leak rates could be applied. Average refrigerant leak rates were expected to occur at a constant rate throughout the year.

For example, if 100 vessels each spent 72 hours in California waters, ports and harbors, the equivalent ship-year equivalents would be:

100 ships \* 72 hours/ship = 7200 ship hours 7200 ship hours/ (24 hours/day) \* (365 days/year) = 0.82 ship-year equivalents

The standard refrigerant equipment emissions formula was thus adapted for marine vessels as follows for each vessel type:

Marine vessel emissions (lbs.) = Average refrigerant charge (lbs.)/ship \* average annual leak or loss rate \* total ship-year equivalents

Additional Marine Vessel Data Used: Data on cruise ship port calls to California were included in the CARB Marine Model as part of the aggregated data that were combined with merchant ship port calls, and were not shown separately. Because the emission factors used for cruise ships are different than the average merchant ship, it was necessary to estimate the number of cruise ship port calls separately, which were taken from the U.S. Department of Transportation.<sup>53</sup> The average amount of time cruise ships spend in California waters is not known with certainty; therefore, the average time used is from the CARB Marine Model for merchant ships. To avoid double-counting, the number of cruise ships were deducted from the aggregate number of merchant ships.

Similarly, the numbers of large fishing vessels were included in the CARB Marine Model, but as the emission factors are different than those of other merchant ships, it was necessary to estimate the number of large fishing vessels separately from aggregated merchant ship data, using the 2007 analysis by Schwarz and Rhiemeier.<sup>52</sup>

The number of smaller fishing vessels (less than 25 meters in length) is not included in the CARB Marine Model; these fishing vessel numbers were estimated using a separate CARB data source, from the regulatory document "Emissions Estimation Methodology for Commercial Harbor Craft Operating in California".<sup>54</sup> Emission factors are from Schwarz and Rhiemeier, 2007.<sup>52</sup>

Additionally, the number of Navy ships is not tracked by the CARB model; data on the number of Navy ships in CA ports were derived from the U.S. Naval list of ships and their homeports.<sup>55</sup> The amount of time Navy ships spent in port or harbor was not available, likely due to security concerns; therefore, a conservative factor of 25 percent of time spent at the home port was applied. Only those ships with a home port assignment in California were included. According to UNEP analysis, Navy ships have the same average refrigerant emissions profile as merchant ships.<sup>34, 35</sup>

Due to the lack of data available, no EOL emissions from ship refrigerant and AC equipment were estimated. While likely to be greater than zero, it is also likely that with the large average refrigerant charge size of systems used, it is economically desirable to recover all refrigerant in refrigeration and AC equipment prior to disposal or recycling (as it is for large stationary refrigeration and AC equipment),<sup>7</sup> and in accordance with good refrigerant management practice at ports as found by Dwyer, 2012.<sup>16</sup> The EOL emissions loss factor will be updated should additional information becomes available, for example, when the UNEP Refrigeration Technical Options Committee (RTOC) report is updated.

Emissions were calculated for baseline years 2002, 2007, 2008, and 2010. Interpolation was used for years between 2002 and 2010 not previously estimated. Emissions are proportional to the amount of ship time spent in California waters, which is correlated with shipping traffic and trade. Therefore, growth or decrease in emission was linked to shipping traffic data as collected by the CARB Marine Model. For years previous to 2002, the annual growth rate of 2 percent between 2002 and 2008 was applied to back-cast emissions. For years 2008 through 2015, it was estimated that shipping traffic and therefore, their refrigerant emissions would continue to decline one percent per year. For years 2016 and beyond, it was assumed that shipping traffic

would increase to previous levels and continue growing at about one percent per year. These assumptions will be periodically double-checked against collected data for shipping traffic in California waters.

<u>Refined Emissions Estimates</u>: Emissions from the ships sub-sector of transportation refrigeration were originally included as part of the aggregated transport refrigerant emissions (as scaled from the U.S. EPA Vintaging Model). A careful analysis of UNEP RTOC reports,<sup>34, 35</sup> and the Schwarz and Rhiemeier 2007 study<sup>52</sup> showed that for a coastal state such as California, refrigerant emissions from ships/marine vessels were likely to be in quantities sufficient enough to warrant further research. All emissions as shown for this sub-sector can be considered "refined" compared to initial estimates. A further refinement was to add the small amount of refrigerant loss from inland fishing vessels (less than two percent of the ship subsector's F-gas emissions).

### Aircraft Air-conditioning

Based upon research conducted by W. Schwarz of Öko-Recherche and J.M. Rhiemeier of Ecofys,<sup>52</sup> the refrigerant emissions from aircraft were assumed to be negligible, and were not included in the CARB F-gas inventory. Most aircraft do have AC units, but for flight altitudes greater than 10,000 feet, an HFC-based vapor cycle is not used to cool the aircraft; it is cooled with bleed air from the jet engine. For air-conditioning purposes prior to take-off and after landing, semi-mobile units at the plane docking sites are connected to the plane's air circulation system. Annual refrigerant emissions from aircraft in the European Union 27 (EU 27) countries were estimated to be less than 400 kilograms per year.<sup>52</sup> With a population of approximately 500 million in EU 27 countries in 2010, these emissions, if scaled to California' population in 2010 (37.2 million), would be less than 30 kilograms.

# Non-Refrigeration, Non-AC Sectors

The sources of non-refrigerant F-gases are diverse and each source requires a separate methodology for estimating its F-gas emissions, as described in the following sub-sections.

# Metered Dose Inhaler (MDI) Aerosol Propellants

Estimates based on the US EPA Vintaging Model were the initial source of data for F-gas emissions from MDI aerosols.<sup>30</sup> The Vintaging Model inputs are proprietary and emission results are not speciated, they are expressed in teragrams of carbon dioxide equivalents (TgCO<sub>2</sub>e), which are equal to MMTCO<sub>2</sub>E. Scaling national Vintaging Model estimates to state population, emissions from this sector for baseline year 2010 were estimated to be 1.4 MMTCO<sub>2</sub>E CFCs, and 0.3 MMTCO<sub>2</sub>E HFCs for California (with no HCFCs used). Because the Vintaging Model aggregates emissions and groups them as total CFC, total HCFC, or total HFC, it was necessary to further speciate usage by actual F-gas used, to develop a usage "distribution profile".

The primary data source for MDI aerosol speciation was the Department of Health and Human Services rule making.<sup>56</sup> Speciation assumptions were also compared to available MDI information cited in IPCC and UNEP reports.<sup>57, 58</sup> Speciation profiles for MDI usage were developed for CFCs (equal distribution of CFC-11 and CFC-12) and HFCs (90 percent HFC-134a and 10 percent HFC-227ea). After developing the F-gas distribution profile for MDI aerosol propellants, the emission results (expressed as MMTCO<sub>2</sub>E of CFC or HFC) were back-calculated into pounds of emissions from specific propellant.

To convert a known quantity of MMTCO<sub>2</sub>E into pounds of F-gas emissions, the following formula was used:

Lbs. = MMTCO<sub>2</sub>E (known) / [[(decimal portion constituent 1) \*GWP1 \*  $4.53592 \times 10^{-10}$  MMT/lb. conversion factor] + [(decimal portion constituent 2) \*GWP2 \*  $4.53592 \times 10^{-10}$  MMT/lb. conversion factor] + (repeat for all constituents)]]

Note: The conversion factor is derived from: 0.454 kg/lb. \*1 MT/1000 kg \* 1 Million MT/1000000 MT =  $4.53592 \times 10^{-10}$  MMT/lb.

For example, the following calculation was used to estimate the pounds of HFCs estimated from MDIs in California in 2010:

Given:

0.3375 MMTCO<sub>2</sub>E of all HFCs from MDI (from scaled estimates of Vintaging Model).

90 percent of HFC aggregated emissions from HFC-134a.

10 percent of HFC aggregated emissions from HFC-227ea.<sup>57, 58</sup>

GWP Values are 1300 for HFC-134a and 2900 for HFC-227ea (IPCC Second Assessment Report [SAR] GWP values).  $^{59}$ 

Lbs. of HFCs =  $0.3375 \text{ MMTCO}_{2}\text{E}$  HFCs / (0.9 lbs. (134a) \* 1300 \* 4.53592 x 10<sup>-10</sup> MMT/lb.) + (0.1 lbs. (227ea) \* 2900 \* 4.53592 x 10<sup>-10</sup> MMT/lb.) =  $0.3375 \text{ MMTCO}_{2}\text{E}$  HFCs / (5.307 x 10<sup>-7</sup> MMTCO<sub>2</sub>E/lb.) + (1.315 x 10<sup>-7</sup> MMTCO<sub>2</sub>E/lb.) =  $0.3375 \text{ MMTCO}_{2}\text{E}$  HFCs / (6.662 x 10<sup>-7</sup> MMTCO<sub>2</sub>E /lb.) = 509,630 lbs. of HFCs HFC-134a lbs. = 0.9 \* 509,630 lbs. HFC = 458,667 lbs. of HFC-134a HFC-227ea lbs. = 0.1 \* 509,630 lbs. HFC = 50,963 lbs. of HFC-227ea

Prior to 2000, only CFC-11 and CFC-12 were used as fluorocarbon propellants in MDIs. Due to lack of specific data on distribution, it was assumed that both CFC-11 and CFC-12 propellants were used in equal amounts. Because MDI are deemed an "essential use", they are exempt from the Montreal Protocol phase-out schedule that applied to other CFC usage in North America. According to the U.S. EPA website on Essential Uses of CFCs for Metered Inhalers, "Metered Dose Inhalers (MDIs) propelled by ozone-depleting CFCs are being gradually and cautiously phased out".<sup>60</sup> Beginning in 2000, HFC propellants were also used in addition to CFCs. It was assumed that the transition rate of replacement from CFCs to HFCs between 2000 and 2010 was linear, from no HFCs before 2000, to 49 percent (by mass) in emission year 2010, and 100 percent HFC beginning in 2014. Final phase-out of CFCs used in MDI manufacturing, sales, and dispensing in the U.S. is expected to be complete by December 31, 2013.<sup>61</sup> When the phase-out is complete, the annual GHG emissions from this source beginning in 2014 will be from (relatively) lower-GWP HFC propellants and the annual emissions are expected to decrease by 50 percent from 2010 emissions.<sup>30</sup> The U.S. EPA Vintaging Model emissions estimates were used as the source of data for all years through 2020, which estimated that HFC emissions would increase by 1.5 percent annually.<sup>30</sup>

The following section of the MDI emissions methodology describes further analysis that were used to further refine F-gas emissions from MDIs.

Usage estimated by Montreal Protocol "Essential Use" Nominations for MDI: The U.S. EPA Vintaging Model estimates were further refined after reviewing the United Nations Environment Programme (UNEP) Technology and Economic Assessment Panel (TEAP) progress reports for years 1999 through 2012.<sup>62</sup> Each TEAP progress report contains a section for essential use nominations (and approvals) of CFCs used for MDI production. Additionally, many of the annual progress reports contained information on the amount of CFCs used in MDI production at country-specific levels, including the United States. National usage amounts were scaled to California's share on the national population.

Emission estimates are complicated by the lack of California-specific data or even national usage estimates, and the high sensitivity of emissions to selected average MDI unit size. For the global usage scaled to California, the average unit size of 23.9 grams/unit was selected from medical literature, although individual units may contain as little as 11 grams of propellant.<sup>63</sup> Additionally, it is not clear if HFC propellants will replace CFC propellants by a one-to-one factor, or will be replaced by not-in-kind propellants or medication delivery devices. Drypowder inhalers (DPIs) contain no propellant and their usage has been increasing globally.<sup>64</sup> Although dry powder inhalers have been commonly used in Europe for more than a decade, their acceptance in the United States has been slow.<sup>65</sup>

For this MDI sector methodology, we assume that all stockpiled CFCs are used by the end of calendar year 2013, and that beginning in 2014, the only F-gases used will be HFC-134a (90 percent of F-gases) and HFC-227ea (10 percent of F-gases). An annual growth rate of 1.5 percent is used from the Vintaging Model. The growth rate in HFCs could be reduced by increased usage of dry-powder inhalers or by some non-HFC propellant to be determined. U.S. EPA Vintaging model updates will be used for future estimates if no stand-alone methodology for California MDI usage is developed.

<u>Refined Emissions Estimates:</u> As indicated previously in the methodology section for MDI, the emissions estimates were back-calculated from scaled-down and aggregated estimates from the U.S. EPA Vintaging Model, which necessitated using best estimates for emissions input factors. The emissions estimates were refined several times as more research was conducted and the emissions profile was improved. The refined emissions by mass were approximately 5% greater than initial estimates, increasing from 433,100 kg to 456,600 kg (954,900 lbs. to 1,006,600 lbs.). Additionally, CFC-114 was added as a propellant used in 2008.

# Consumer Product (and Commercial/Industrial) Aerosol Propellants

For all non-MDI aerosol propellant emissions, four separate data sources were analyzed: 1) CARB survey data, 2) U.S. EPA Vintaging Model estimates, 3) National Aerosol Association (NAA) estimates, and 4) Alliance for Responsible Atmospheric Policy (ARAP) estimates. Estimated emissions in pounds per year of HFCs were averaged from the data sources.

A 2006 CARB industry survey of consumer product emissions was used to estimate aerosol propellant F-gas emissions,<sup>18</sup> and supplemental data was used from CARB staff research in Consumer Products Regulatory Amendments.<sup>19, 20, 21</sup> The 2006 survey was back-cast to 2000 and forecast to 2020 based on population growth for consumer product aerosols. An exception was made for the assumed growth rate of duster (pressurized gas) spray, which was assumed to grow equivalent to the rate of personal computer growth of 8.5 percent annually between 2000 and 2008, due to the prevalent use of duster spray as a keyboard cleaner.<sup>66, 67</sup> From 2008 through 2020, the duster growth rate is assumed to more closely correlate to population growth. Approximately 4.8 million pounds of emissions in 2010 were estimated based on a forecast of the 2006 survey.

CARB staff research and regulations were used to establish the rate of transition for replacing HFC-134a with HFC-152 for duster use to estimate likely distribution. The CARB industry

survey was also augmented by additional speciation and background data from UNEP and IPCC research,<sup>57, 68</sup> and compared to consumer aerosol propellant usage data sources that include the U.S. EPA Vintaging Model, Earth911, the National Aerosol Association, and the Alliance for Responsible Atmospheric Policy.<sup>30, 69, 70, 71</sup>

U.S. EPA Vintaging Model data estimated HFC emissions from consumer product aerosol propellants for estimated emissions year 2010. The CO<sub>2</sub>-equivalents were back-calculated from HFC distribution estimates to derive 9.8 million pounds of emissions in 2010. Note that when using Vintaging Model estimates expressed as MMTCO<sub>2</sub>E, the resulting calculated pounds of emissions are extremely sensitive to assumed proportions of HFC-134a and HFC-152a propellants, due to the disparity of GWP between the two (1300 for HFC-134a and 140 for HFC-152a using IPCC SAR GWPs). However, Vintaging Model results compared closely to results based on national estimated aerosol usage from the National Aerosol Association (13.8 million pounds of HFCs in 2010), and the Alliance for Responsible Atmospheric Policy (10.9 million pounds of HFCs in 2010).

The following shows emission factors used from the Alliance for Responsible Atmospheric Policy and Earth 911 data:  $^{69,70}$ 

- Three billion aerosol cans used in 2007 in the U.S.
- Annual sales growth rate of cans is 1 percent (CA population growth used).
- 3.1 billion cans used in 2010 in the U.S.
- 95 percent of cans are non-HFC.
- 156.9 HFC-containing cans used in U.S. in 2010.
- California share of national population is 12.5 percent.
- 19.6 million HFC-containing cans used in California in 2010.
- 0.56 pounds propellant per can.
- 10.9 million pounds of HFC emissions from aerosol propellants in California in 2010.

Using National Aerosol Association sales data, a slightly higher figure of 13.8 million pounds of HFC emissions from this sector was estimated based upon the same data used above, but beginning with a higher production figure of 3.7 billion aerosol cans used in the U.S. in 2005.<sup>71</sup>

For aerosol propellant emissions, we assume 100% emissivity and that all emissions occur same year of aerosol can production.

Due to the wide variety of the types of aerosol propellant cans, there is no simple data point that can be used for an average aerosol propellant can. However, oversimplifying slightly by averaging the four main sources of information, the following factors are useful guidelines:

- An aerosol propellant can contains about 0.55 lbs. of propellant.
- At least 95 percent of aerosol propellant used is non-HFC.
- Aerosol cans are assumed to be 100 percent emissive in the year they were manufactured.
- Californians used about 0.48 HFC-containing aerosol propellant cans per person per year for years 2006-2011; ( for year 2011, based on HFC emissions of 9,852,400 lbs and 0.55 lbs/can = 17,913,450 cans total, for 37,570,000 people).

For emissions beyond baseline years, we assume that the CARB regulations for high-GWP consumer products will be followed as HFC-134a aerosol propellant is replaced by HFC-152a or other lower-GWP propellants in specific applications (e.g., duster spray).<sup>19</sup>

<u>Refined Emissions Estimates:</u> Due to analyzing several data sources as previously described, several emissions inputs were refined from initial estimates. The results were that emissions by mass increased 11%, from 3,816,300 kg to 4,232,200 kg (8,413,500 lbs. to 9,330,300 lbs.). Lacking more precise data, the initial estimates had assumed that HFC emissions were distributed equally between HFC-134a (50%) and HFC-152a (50%). A careful analysis of CARB survey data also indicated that a more precise speciation for aerosol propellant emissions in 2008 was HFC-134a (23.4%), HFC-152a (76.3%), and HFC-43-10mee (0.3%). The most recent CARB industry survey on consumer aerosol product use and sales was conducted in 2006. An updated survey is needed to more accurately estimate current and future emissions from aerosol propellants.

#### Fire Protection (fire suppressants

Fire protection emissions data are from the CARB-funded research inventory project number 07-313 titled "Developing a California Inventory for Industrial Applications of Perfluorocarbons, Sulfur Hexafluoride, Hydrofluorocarbons, Nitrogen Trifluoride, Hydrofluoroethers, and Ozone Depleting Substances" and conducted by K. Wolf of the Institute for Research Technology and Assistance (IRTA).<sup>17</sup>

Fire suppressants are an insignificant source of HFC emissions, accounting for just 0.25 percent of HFC emissions. High-GWP fire suppressant emissions occur from large total flooding systems, smaller streaming (extinguisher) emissions, and emissions that occur at the time of Halon recycling. The high-GWP fire suppressant compounds used are Halon 1211, Halon 1301 (both ODSs), and the ODS replacements HFC-125 and HFC-227. A negligible amount of PFC blends were also used in the early 2000s.

Emissions were estimated for baseline year 2010 and projected through 2020. Back-casting to 1990 estimates were based on data from Wolf, 2011 that included the number and types of fire suppressant systems in the early 1990s in California.

As noted in the Wolf, 2011 report, fire suppressant systems are very leak tight, with very few accidental releases. Purposeful release of fire suppressants averages just two percent of the suppressant amount per year. Recycling fire suppressants for re-use emits another one percent per year of the recycled amount.

The high-GWP fire suppressant emissions have been decreasing since 1990, and are expected to continue to decrease through 2050. Lower-GWP suppressant replacements to Halons and HFCs such as Inergen and F-K-1-5-12 continue to increase their share of total flooding and streaming fire suppressant systems. In 2011, just 51,000 lbs. of high-GWP fire suppressants were emitted, which had a GHG impact of 0.09 MMTCO2E, or 0.2 percent of all F-gas emissions.

<u>Refined Emissions Estimates:</u> F-gas emissions from fire suppressants were not included in the very first initial emissions estimates. Based upon a review of the UNEP Halons Technical Options Committee (HTOC) 2006 Assessment,<sup>72</sup> it was assumed that fire suppressant F-gas emissions by 2008 were likely to be de minimus and did not warrant further analysis. However, through additional analysis based on the findings of Wolf, 2011, we concluded that although fire suppressant emissions were quite low in 2008, estimated to be only 25,800 kg (56,900 lbs.), which accounted for only 0.3% of all F-gas emissions in that year, the emissions were still measurable and are therefore included in the refined inventory estimates.

Fire suppressant systems tend to be very leak tight, with very few accidental releases. Purposeful release of fire suppressants averages just two percent of the suppressant amount per year. Recycling fire suppressants for re-use emits another one percent per year of the recycled amount. The high-GWP fire suppressant emissions have been decreasing since 1990, and are expected to continue to decrease through 2050. Lower-GWP suppressant replacements to Halons and HFCs such as Inergen and F-K-1-5-12 continue to increase their share of total flooding and streaming fire suppressant systems.

### Insulating Foam:

F-gas emissions from insulating foam are from the CARB-funded research on insulating foam banks and emissions inventory conducted by Ashford and Vetter of Caleb Management Services, Ltd., in the 2010 Final Report titled "Developing a California Inventory for Ozone Depleting Substances (ODS) and Hydrofluorocarbon (HFC) Foam Banks and Emissions from Foams".<sup>14</sup> The Final Report also describes the emissions estimates methodology in detail.

Foam emission sectors were identified and grouped into the following five foam GHG emission sources: 1) building insulation, 2) residential appliances (refrigerator-freezers and water heaters), 3) commercial refrigeration equipment, 4) transport refrigerated units (TRUs), and 5) marine buoyancy. Building insulation was further divided into the following insulation types: extruded polystyrene (XPS), polyiso, polyurethane panel, and polyurethane spray. Additionally, there were three distinct sub-types of buildings: commercial, single-family, and multiple-family buildings. Residential appliances were divided into water heaters, refrigerator-freezers, and freezer-only. Transport refrigerated units includes refrigerated trailers and trucks, rail refrigerated units, and refrigerated shipping containers. In all, 19 separate foam categories were researched to estimate emissions from each type of foam and application combination.

A comprehensive inventory of foam in California was developed through industry surveys and analysis of foam usage.<sup>14</sup> For the building sector, the F-gas-containing foam insulation volume was estimated for building stock beginning with 1960 through 2009, for single-family homes, multi-family homes, and commercial buildings. Appliance inventory and foam types used were developed from data supplied by the Association of Home Appliance Manufacturers<sup>73</sup> and additional water heater research by Ashford and Vetter, 2010.<sup>14</sup> Commercial refrigeration equipment and transport refrigerated unit (TRU) foam emissions were developed by applying industry standard insulating foam profiles to the California inventory of commercial refrigeration equipment and TRU equipment.

The remaining foam application are in the marine sectors buoyancy, including leisure boats, canoes, and buoyancy flotation aids. Industry surveys were used to develop foam profiles for this sector. Surfboards and windsurfers were investigated as a possible source of GHG emissions, but these foam applications have used water-based foam expansion agents since the 1980's are not a GHG emissions source. Additionally, polystyrene (often called Styrofoam<sup>®</sup> after its trademarked name from Dow Chemical) used for cups, plates, and packaging has not been a source of CFC or other F-gas emissions since the late 1970s and is no longer a source of F-gas emissions.<sup>14, 74</sup>

Because of the high degree of uncertainty regarding foam GHG emissions from landfills, we assumed that all GHGs within landfilled foam are eventually emitted to the atmosphere. However, several studies conducted by researchers at the Danish Technical University indicate that CFC foam expansion agents in landfills attenuate, or biologically degrade to lower-GWP constituents.<sup>75, 76, 77</sup> Research conducted by Environment Canada also shows that emissions of CFC and HCFC compounds in landfills are greatly reduced when collected by landfill gas systems, and combusted at high temperatures above 559 °C (1038 °F).<sup>78</sup> In March 2012, CARB approved research by California Polytechnic State University, San Luis Obispo, to determine emissions of potent greenhouse gases from waste insulating foam in landfills, although results will not be available until 2015.<sup>79</sup>

### Emissions calculations for foam

The foam emissions sector is a relatively complex emissions sector which cannot be covered with a simple formula. Extensive spreadsheet calculations were developed which included emissions from the five emissive parts (processes) of the foam lifecycle: 1) at time of manufacturing, 2) during application (relevant to spray foams), 3) during lifetime of equipment, 4) at time of building decommissioning or equipment end-of-life shredding/recycling, and 5) after disposal and landfilling. Emission profiles were developed for each sector (building, residential appliances, commercial refrigeration equipment, transport refrigeration, and marine buoyancy) and each type of foam within the sector (spray foam, polyurethane block foam, extruded polystyrene foam, etc.). Profiles were further refined by assigning foam expansion agent distribution according to year of foam manufacture. The basic emissions formula for each of the emissive processes is shown below:

Emissions (lbs.) = volume of foam  $[m^3]^*$  density of foam  $[kg/m^3]^*$ % of foam expansion agent by mass \*% of foam expansion agent loss/emitted \* 2.20462 kg/lb.

Foam GHG emissions were estimated from 1975 through 2020. To cross-check back-casting emissions estimates from baseline year 2008, internal CARB analysis also refined historical foam expansion agent profiles with foam usage information contained in IPCC reports and UNEP reports.<sup>57, 80, 81, 82, 83</sup>

For residential appliance foam insulation emissions and speciation forecasting, additional CARB-funded research conducted by ICF International was used to cross-check assumptions.<sup>7</sup> The Caleb inventory was verified as highly accurate, pending additional results from landfilled foam GHG emissions research currently being conducted by the Global Waste Research

Institute, associated with California Polytechnic State University, San Luis Obispo.<sup>79</sup> Research results are expected in 2015.

<u>Refined Emissions Estimates:</u> Due to the thorough research conducted by Ashford and Vetter, it was not necessary to refine assumptions or inputs to a significant degree for emissions year 2008. However, a careful line-by-line analysis of the research results did result in a few minor refinements to the emissions estimates. Emissions by mass decreased 3 percent. Two ODS foam expansion agents (CFC-12 and HCFC-22) were added to the speciation mix, and one (HFC-134a) was removed because its emissions were likely to be de minimus in 2008.

### Semiconductor Manufacturing

Perfluorocarbons (PFCs) are the primary high-global warming potential compounds used in the manufacture of semiconductors. PFC emission estimates from semiconductor manufacturing are not within the primary emission boundaries of this particular methodology, but are briefly described here for reference. PFC emissions in California have previously been estimated by the CARB Greenhouse Gas Inventory<sup>84</sup> and are available at: http://www.arb.ca.gov/cc/inventory/data/data.htm. Approximately 142,000 pounds of PFCs

were estimated to be emitted by the semiconductor sector in 2008.

According to the U.S. EPA Greenhouse Gas Inventory – 2000-2010, HFCs are also used in the manufacture of semiconductors.<sup>31</sup> The CARB Greenhouse Gas Inventory, estimates that a small amount of HFC-23 is used in the manufacture of semi-conductors in California, about 11,000 pounds/year statewide.<sup>84</sup> The emissions of HFC-23 from this sector were taken directly from the CARB inventory, with no additional changes in methodology or emission estimates.

Nitrogen trifluoride (NF<sub>3</sub>) is also used in semiconductor manufacturing and the manufacture of plasma screen televisions. Approximately 18,000 pounds are used per year statewide, as reported by the CARB Greenhouse Gas Inventory.<sup>84</sup>

Due to CARB regulations, PFC emissions are expected to be reduced in the future, with only 44 percent of 2008 baseline emissions being emitted by 2015. HFC-23 emissions are expected to increase by 0.5 percent annually from 2010 onward, much slower than BAU estimated rates due to CARB regulations.<sup>22</sup> NF<sub>3</sub> emissions should conceivably increase even with industry GHG reduction requirements, because substituting NF<sub>3</sub> for PFC-116 (C<sub>2</sub>F<sub>6</sub>) is an approved alternative chemistry for reduction of GHGs in chemical vapor deposition (CVD) chamber cleaning. Additionally, the semiconductor GHG emission regulations do not cover NF<sub>3</sub> used in plasma televisions. Due to increasing NF<sub>3</sub> use in semiconductor manufacturing and in plasma televisions, NF<sub>3</sub> emissions were estimated to increase 11 percent annually between 1978 and 2008.<sup>85</sup> We conservatively estimate continued NF<sub>3</sub> emissions increases at one-fourth the previous growth rate, for an annual emissions increase of 2.75 percent annually through 2020.

<u>Refined Emissions Estimates:</u> No changes were made to the original estimates for the semiconductor manufacturing sector.

### **Solvents**

F-gas emissions estimates from all non-semiconductor industry solvents are from the CARBsponsored research "Developing a California Inventory for Industrial Applications of Perfluorocarbons, Sulfur Hexafluoride, Hydrofluorocarbons, Nitrogen Trifluoride, Hydrofluoroethers and Ozone Depleting Substances" conducted by K. Wolf of the Institute for Research and Technical Assistance (IRTA).<sup>17</sup> A survey of solvent-using industries in California was undertaken, along with a review of applicable air permits, to determine emissions of specific F-gases from the solvent sector. Air permit holders for high-GWP solvents in California were contacted and surveyed for their actual solvent usage of CFCs (if any), HCFCs, HFCs, PFCs, and PFPEs. If no specific usage data could be obtained from the permit holder, it was assumed they had used the entire amount they were permitted to use during the year.

Emission estimates were made during research years 2008-2010, and were used for 2007, 2008 and 2010 emission years. The 2000 through 2006 solvent F-gas emissions estimates were not within the time-frame of the research. To form a backwards trend analysis for solvent emissions prior to 2007, historical usage and speciation trends were informed by five research reports that included solvent usage trends.<sup>57, 86, 87, 88, 89</sup>

Due to stringent VOC limitations in solvent usage in California, particularly for the South Coast Air Quality Management District, many industries have converted to low-VOC and water-based cleaners for industrial applications. There has been a concurrent decrease in the amount of HCFC and HFC solvent usage as well, with California using only 10 to 50 percent as much HCFC and HFC solvent, per capita, as average national usage.<sup>17,30</sup>

The U.S. EPA estimates that solvents account for only 1 percent of all emissions by  $CO_2$ -eq from ODS replacements.<sup>31</sup> We also estimate that for 2008 in California, solvents comprised 1 percent of all HFC emissions by  $CO_2$ -eq, and 0.5 percent of all F-gas emissions (including ODS) by  $CO_2$ -eq. By 2011, the emissions from solvents were estimated to decrease to just 0.6 percent of all HFC emissions (by  $CO_2$ -eq), and 0.4 percent (by  $CO_2$ -eq) of all F-gas emissions in California. California has a different profile than nationally for solvent usage, up to a magnitude of F-gas solvent usage lower than nationally, due to stringent VOC restrictions on solvents that incentivized transition to water-based solvents beginning in the 1990s.<sup>17</sup>

Note that for discontinued production of CFC and HCFC solvents, the IPCC 2006 GHG Inventory Guidelines were not strictly followed for solvent emissions, which states, "Historically, emissions from solvent applications generally have been considered prompt emissions because 100 percent of the chemical is typically emitted within two years of initial use."<sup>90</sup> Based on additional research, estimated stockpiled amounts of CFC and HCFC solvents accumulated prior to discontinued production dates is assumed to be used at a rate of five percent of stockpiled amount per year until the stockpiles are depleted.<sup>17, 86</sup> The actual rate of depletion may be more rapid than assumed, although usage could continue longer than estimated due to an unknown amount of ODS solvents likely to have been brought into the state illegally since their production and import have been banned. For example, the California Office of Environmental Health Hazard Assessment (OEHHA) estimates that significant, but unknown quantities of CFC-113 continue to be illegally brought into California each year, most likely for the clandestine manufacture of methamphetamine.<sup>91</sup> We assume that because HFC solvents continue to be manufactured, they are not stock-piled, and are 100 percent emissive in the year they are purchased and used.

<u>Refined Emissions Estimates</u>: Original estimates as derived from scaled-down Vintaging Model estimates calculated the fluorinated solvent emissions in 2008 at 426,380 kg (940,000 lbs.). Based upon the research specific for California solvent usage, the emissions were reduced almost by half to 218,600 kg (482,000 lbs.). The original speciation was overly simplistic and included only the following two fluorinated solvents (note that percent speciation is by mass): HFC-43-10mee (90%) and HCFC-141b (10%). The refined estimates included the additional solvents: CFC-113, HCFC-225ca/cb, HFC-245fa, HFC-365mfc, and PFC-14.

# Sterilants (Medical)

The F-gas emissions from medical sterilants were estimated using the same methodology as described for metered-dose inhalers, with U.S. EPA Vintaging Model national estimates scaled to California's population for  $CO_2E$  emissions. Pounds of emissions were then back-calculated from the MMTCO<sub>2</sub>E.

The Vintaging Model estimated that the sterilants sector emitted  $0.18 \text{ MMTCO}_2\text{E}$  of HCFCs in 2010 (scaled to CA population), with no CFC or HFC emissions.

Prior to the 1995 CFC phase-out, a blend of CFC-12 and ethylene oxide (EO) was used as a medical sterilant.<sup>68</sup> Beginning in 1995, blends of EO/HCFC-22 and EO/HCFC-124 were approved to replace the EO/CFC-12 blend, with no HFCs used as medical sterilants.<sup>92</sup> Based on a review of material safety data sheets, we will assume that of the HCFCs used in EO blends, 70 percent are HCFC-124 and 30 percent are HCFC-22.<sup>93, 94</sup>

As of 2010, no HCFC-22 was allowed for use as a sterilant. Additionally, as of 2015, HCFCs will no longer be allowed for any new production or import, unless used as refrigerants for equipment manufactured prior to 2020.<sup>95</sup> We assume that sterilants are used in the same year they are manufactured, and that they are 100 percent emissive.

It is doubtful that HCFC-124 will be stockpiled in sufficient quantities for use as sterilants past 2015, as many low-GWP, non-ODS alternatives are currently available. All sterilants approved by the U.S. EPA Significant New Alternatives Program (SNAP) are low-GWP, with none containing HFCs.<sup>92</sup> Although one of the alternatives, trifluoromethyl iodide (CF<sub>3</sub>I), contains fluorines, it has a GWP of  $1.^{59}$  Therefore, sterilants will no longer be a source of high-GWP F-gas emissions beginning 2015.

To forecast emissions, U.S. EPA Vintaging Model data was used for years 2010 through 2015. Back-casting emissions were based on the sterilant speciation transitions previously described, and an assumed growth rate of 1 percent annually, roughly equivalent to the 0.95 percent annual population growth rate in California between 2000 and 2010.

<u>Refined Emissions Estimates:</u> No California-specific data was available for fluorinated medical sterilants, therefore, refinements were made to assumptions used to interpret the scaled estimates from the U.S. EPA Vintaging Model. All Vintaging Model emissions for this sector were expressed as MMTCO<sub>2</sub>E of HCFCs. Original assumptions were that the HCFC emissions consisted of 10% HCFC-22 by mass, and 90% HCFC-124 by mass, resulting in a back-calculated estimate of 241,945 kg (533,400 lbs.). Additional research into actual medical sterilants used showed that the most likely fluorinated blends consisted of 30% HCFC-22 and 70% HCFC-124.<sup>93,94</sup> The change in assumed speciation in turn refined the emissions by mass to 368,360 kg (812,100 lbs.). Note that emissions from medical sterilants remains relatively uncertain compared to other emissions sub-sectors, in that California-specific data was not available. Medical sterilants are estimated to contribute to less than 1% of all F-gas emissions in 2008, and are therefore considered a minor source of emissions. Additionally, as this sub-sector will no longer be a source of F-gas emissions beginning in 2015, conducting additional emissions refinements for this source is not a high priority.

### Sulfur Hexafluoride

Sulfur hexafluoride (SF<sub>6</sub>) emission estimates were derived by CARB staff for the following sectors: semiconductor manufacturing,<sup>22</sup> electricity generation and transmission,<sup>23</sup> and magnesium manufacturing and other miscellaneous SF<sub>6</sub> uses.<sup>24</sup> SF<sub>6</sub> usage was estimated from industry surveys, and is described more fully in the referenced studies. The SF<sub>6</sub> inventory for California was also informed by scaling emissions from national estimates as shown in the CARB greenhouse gas emissions inventory.<sup>31, 84</sup>

CARB estimates the following SF<sub>6</sub> reductions from the three SF<sub>6</sub> sectors identified: 1) From the semiconductor manufacturing sector, both PFC and SF<sub>6</sub> emissions are expected to decrease 44 percent from 2008 baseline levels by  $2020^{22}$  2) From the electricity generation and transmission sector, the SF<sub>6</sub> emission rate from Gas Insulated Switchgear (GIS) will be reduced by one percent per year from 2010 through  $2020^{23}$  3) From the magnesium manufacturing and miscellaneous uses sector, minimum annual reductions of 0.1 MMTCO<sub>2</sub>E from a 2008 baseline of 0.15 MMTCO<sub>2</sub>E are expected by 2020, with a potential of 100 percent reduction from this sector by 2020.<sup>24</sup>

<u>Refined Emissions Estimates</u>: No changes were made to the original SF<sub>6</sub> emissions estimates.

#### Fluorinated Pesticides - Recommendation to add to GHG Inventory

Sulfuryl fluoride (SO<sub>2</sub>F<sub>2</sub>), a pesticide fumigant used primarily in structures for termite control, is a high-GWP F-gas (100-year GWP value estimated at 4780) that had not been officially included in the CARB GHG Inventory as of December 2013.<sup>96, 97, 98</sup>

Sulfuryl fluoride usage records are available for California dating back to 1990 from reported pesticide usage to the California Department of Pesticide Regulation.<sup>29</sup> We assume that sulfuryl fluoride is completely emissive when used.

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