Report on the International Symposium on the Unexpected Increase in Emissions of Ozone-Depleting CFC-11

Neil R. P. Harris, Stephen A. Montzka, Paul A. Newman, with generous contributions from the Symposium attendees

1. The state of understanding before the Symposium

Recent findings of an unexpected emission increase of chlorofluorocarbon-11 (trichlorofluoromethane, CFC-11, CCl_3F) [Montzka et al., 2018][1] has raised important issues within the atmospheric sciences and policy communities. CFC-11 is a long-lasting man-made compound (52-year lifetime in the atmosphere) that is not only a powerful ozone depleting substance (ODS) but also a powerful greenhouse gas with a 100-year global warming potential of 5,160. Growth of emissions could indicate that releases from CFC-11 banks are accelerating, that the atmospheric circulation is changing such that our estimates of emissions based on observed concentrations increase, or that unreported production is leading to increased CFC-11 emissions.

Global CFC-11 emissions in the 2014-2016 period derived by Montzka et al. [2018] were shown to be about 13 gigagrams per year (Gg/yr) higher than the 2002-2012 average. Montzka et al. further showed that emissions from eastern Asia had also increased during this period. The "Scientific Assessment of Ozone Depletion: 2018" [WMO, 2018] provided additional information to that from Montzka et al. [2018] using both NOAA and AGAGE CFC-11 observations, along with some simple model simulations of impacts of these emissions. The independent AGAGE observations also showed CFC-11 emissions increased after 2012. The combined networks indicated an annual increase of 10 Gg/yr in the 2014-2016 period over the 2002-2012 baseline, consistent with the Montzka et al. estimate. While such an emission increase was highly unexpected given the reported production phase-out, the discrepancy between expected and observationally-derived emissions may have begun as early as 2007 (Figure 1). These emission increases slowed the otherwise steady decrease in atmospheric concentrations reported in previous Assessments. The global decline in CFC-11 concentration over 2014 to 2016 was only 2/3^{rds} as fast as it was from 2002 to 2012. While the CFC-11 emissions from eastern Asia have increased since 2012, the contribution of this region to the global emission rise was not well known, and the country or countries in which emissions have increased had not been identified. Subsequent on-the-ground investigations by different organizations suggested ongoing use of CFC-11 in China for the production of foams in 2018, but it was not clear if the activities they identified could account for the observed global changes [*Perry*].

[1] Citations are indicated by square brackets []. Italicized citations were Symposium presentations with only the lead presentation author cited, while regular font citations are peer-reviewed publications.

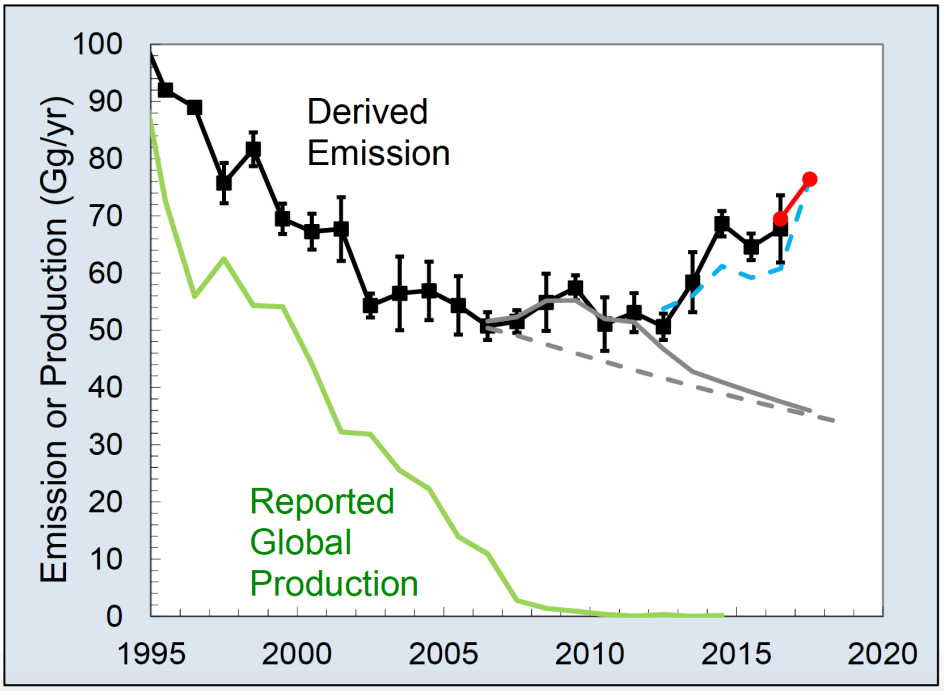

Figure 1. Global CFC-11 emissions derived from NOAA atmospheric observations with a three-box model (black squares) considering a 57.5-year lifetime[2], and production magnitudes reported to the United Nations Environment Programme (UNEP, green line). Red points represent updated values to Montzka et al. [2018] presented at the Symposium [*Montzka*], while the dashed blue line represents emissions approximately corrected for the upper end of estimated circulation changes. The dashed grey line shows expectations given measured global changes through 2005 using a data derived release fraction of 3.2% per year of CFC-11 from the bank extrapolated forward, and the solid grey line is a WMO scenario projection [WMO, 2014] that was constrained by observational data through 2012 (rescaled here to be consistent with a 57.5 yr lifetime) [as in Montzka et al., 2018].

CFC-11 production usually begins with carbon tetrachloride (CCl_4 or CTC) as feedstock (Figure 2, left) [*Sherry*]. The main commercial-scale production involves the fluorination of CTC using a liquid-phase antimony catalyst and anhydrous hydrogen fluoride (Fig. 2, left middle)[*Tope*]. In addition to CFC-11, there is typically some co-production of CFC-12 (CCl_2F_2) [*Tope*]. Before its phasedown, CFC-11 was mainly used in foams; other uses included aerosols, limited refrigeration and air conditioning units (R/AC), metered-dose inhalers, tobacco processing, and as solvents (Fig. 2, middle right). Use in aerosols and solvents resulted in the emission of the CFC-11 shortly after production; but when used to produce foams, only a small fraction of the CFC-11 escapes to the atmosphere rapidly while the majority remains in the foam cells in building insulation or in foam products. Collectively, the CFC-11 that is not immediately released is referred to as a "bank" (Fig. 2, right). While some of this "banked" CFC-11 is readily available for disposal and can be incinerated or destroyed in anaerobic processes in landfills[3], a

[2] The 57.5-year lifetime is consistent with coupled-chemistry-climate model lifetimes [Montzka et al. 2018]. The 52-year lifetime is from WMO [2018], and is a combined estimate of atmospheric observations and models.
[3] Although a few tests were completed from 2003-5 that showed some biodegradation of CFC-11 in anaerobic conditions, no large-scale or long-term in situ testing was completed to confirm that this would occur in landfill operating conditions. Additional detail can be found in the May 2005 volume 3 Technical and Economic Assessment Panel (TEAP) Report of the Task Force on Foam End-of-Life Issues. Evidence for CFC-11 loss in a limited number of landfills is also provided in Hodson et al. [2010]. CFC-11 destruction does occur in anoxic marine waters [e.g. Bullister and Lee, 1995].

substantial fraction remains and is slowly released to the atmosphere at an estimated rate of 0.5%[4] per year (e.g., the CFC-11 in foam cells of building insulation). The schematic representations of plumes in Fig. 2 collectively shows the multiple paths by which CFC-11 is emitted to the atmosphere. Once in the atmosphere, CFC-11 eventually reaches the stratosphere, where it breaks down and releases its chlorine atoms to catalytically destroy ozone. Further, the increased atmospheric CFC-11 increases the radiative climate forcing.

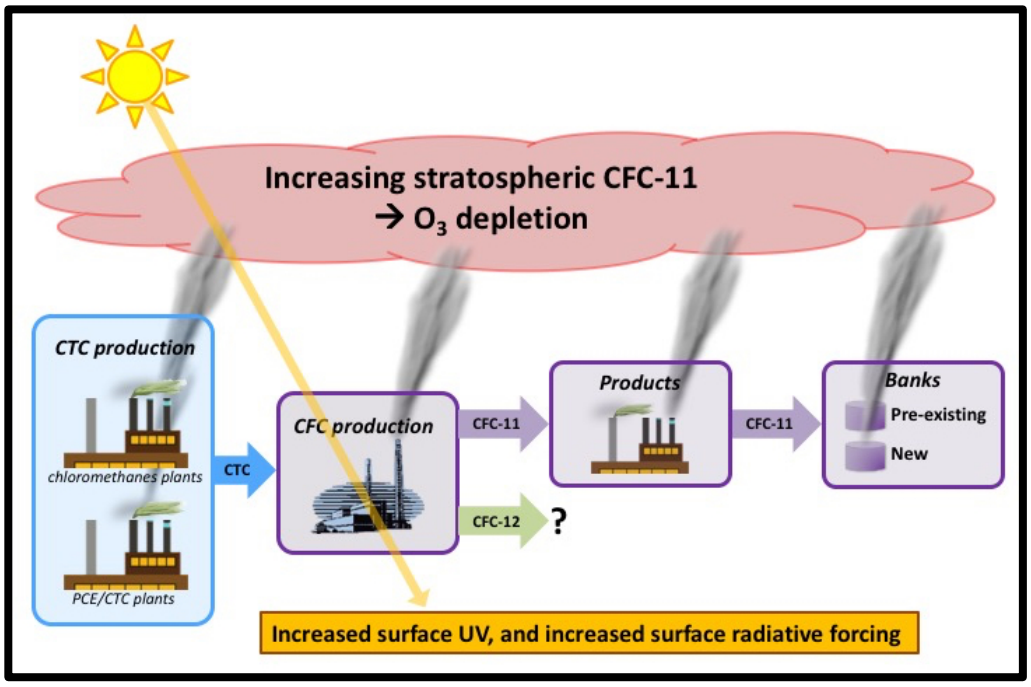

Figure 2. Schematic of CFC-11 emission routes (and also CTC). The left blue box shows production of CTC, which is used as the feedstock (blue arrow) for CFC-11 production (left middle box; PCE = perchloroethylene or C_2Cl_4). The produced CFC-11 feeds into (purple arrow) the manufacturing of products (e.g., foams), as indicated in the middle box. Some traditional CFC-11 products are directly emissive (e.g., aerosols). Foams and other less emissive products collectively form the CFC-11 bank (right hand purple box). Life-cycle emissions of CTC and CFC-11 from these sources (represented as grey cloud plumes) contribute to the atmospheric burden of chlorine that leads to ozone depletion, allowing increased penetration of ultraviolet radiation to the Earth's surface and increased surface radiative forcing.

The implications from the increased CFC-11 emissions are potentially serious. Under the Montreal Protocol on Substances that Deplete the Ozone Layer (hereafter, MP), the global consumption and production for controlled uses of CFC-11 was mandated to cease from 2010 onwards. In response to these recent observational findings concerning CFC-11, the Parties to the MP approved "Decision XXX/3: Unexpected emissions of CFC-11." This decision formally asked the Scientific Assessment Panel (SAP) to provide a summary report on this "… unexpected increase of CFC-11 emissions ..." A preliminary summary report is required for the 41st Open-ended Working Group (July 2019), a further update is requested at the 31st Meeting of the Parties (Nov. 2019), and a final report to the 32nd Meeting of the Parties (Nov. 2020). The decision also asked the Technology and Economic Assessment Panel (TEAP) to provide the parties with information on potential sources of emissions of CFC-11 in a preliminary report to the 41st Open-ended Working Group and a final report to the 31st Meeting of the Parties.

[4] The Intergovernmental Panel on Climate Change estimates that the release rate from landfills is 0.5% per year as extrapolated from foam degradation. Additional detail can be found in the May 2005 volume 3 TEAP Report of the Task Force on Foams End-of-Life Issues [TEAP, 2005].

The science community responded to this CFC-11 emissions increase by holding the "International Symposium on The Unexpected Increase in Emissions of Ozone-Depleting CFC-11" at the United Nations Office in Vienna, Austria on 25-27 March 2019. More than 70 participants from the science and technical communities from 22 different countries attended the Symposium, with 37 presentations. Representatives of the MP's SAP, TEAP and the Environmental Effects Assessment Panel (EEAP) were also present.

2. Unravelling the puzzle

Additional evidence for unreported production

Updated measurements of atmospheric CFC-11 concentrations and additional consideration of CFC-11 emission from banks that were presented at the Symposium make it more certain that the recent excess in emissions inferred for the past few years (see Figure 1) is caused by unreported CFC-11 production. The major uncertainties in quantifying the excess emissions to unreported production are (i) a possible increase in the leakage from banks; and (ii) the influence of atmospheric variability.

A preliminary estimate of CFC-11 emissions for 2018 suggests that they remained comparable with 2014-2017 rates when inferred from NOAA observations [Figure 1; update of Montzka et al., 2018]. New results presented at the Symposium based on observations conducted in eastern Asia also confirmed that emissions from this region had increased from 2012, during the 2013-2017 period [*Western; Park;* and now Rigby et al., 2019].

Other work presented at the Symposium further confirmed that enhanced atmospheric concentrations of CFC-11 have been observed recently in Asia in both focussed studies [*Benish; Simpson; Fang*] and in analyses of longer-term measurements [*Arduini; Adcock; Dang;* Lin et al., 2019]. These studies have potential for more precisely identifying the location of sources in eastern Asia and quantifying their magnitude. Observations of other ODS potentially related to CFC-11 production (e.g., CFC-12, HCFC-22) do not show comparable global emissions behaviour [*Laube; Vollmer*]. However, significant emissions of CTC (the CFC-11 feedstock) are found in eastern China in the last decade [Park et al., 2018; Lunt et al., 2018], with these CTC emissions shifting northward to Shandong province after 2012 [Lunt et al., 2018].

Considering the major uncertainties in turn: analysis of bank release rates indicates that it is unlikely that bank emissions could have increased by the amount required to explain the observations. Specifically, it seems improbable that emissions from the breakdown of insulating foams expressed as a fraction of the global CFC-11 bank could have tripled in recent years and now be as large as 10% per year. Even larger increases in the bank leakage rate (by 10 times) would be required if the increased global source was the result of enhanced releases from foams only in eastern Asia [*Western*; Rigby et al., 2019]). This is compounded by the fact that even when foams are crushed or shredded, a significant portion of the blowing agent (e.g., CFC-11) remains in the foam matrix[5]. Other traditional CFC-11 applications (e.g., aerosols, refrigeration and air-conditioning and associated banks) also appear to be unlikely sources of the increased emissions [*Walter-Terrinoni (b)*].

The main processes other than emission variations that can lead to interannual variations in CFC-11 concentrations are the quasi-biennial oscillation, El Nino-Southern Oscillation, and other large-scale dynamical changes that typically operate on timescales of a few years. Both the multi-year period during which excess CFC-11 emissions have continued (now including 2018) and especially the identification of substantial increases in CFC-11 from eastern China (see below) [*Western*; Rigby et al., 2019] put tighter bounds on the possible contribution of changing atmospheric dynamics on the observed concentration anomalies. The updated observations provide additional confidence that the influence of dynamical

[5] The 2005 TEAP report on End-of-Life of Foams estimates that 50% of the blowing agent remains in the foam matrix after crushing or shredding.

variability cannot account for more than half of the inferred emissions and further imply that the excess emissions are attributable in large part to unreported production. Several presentations examined how atmospheric variability could impact CFC-11 concentrations and global emissions when derived with simple model approaches [*Laeng; Mueller; Nuetzel; Portmann; Prather; Schuck; Sheese*].

Where do the increasing emissions come from?

Global CFC-11 emissions can be estimated using annual global concentrations and the CFC-11 lifetime (as shown in Fig. 1). The difference between the northern hemisphere (NH) and southern hemisphere (SH) average concentrations (the inter-hemispheric difference or IHD, North minus South) provides additional information on anthropogenic emission magnitudes. If there are no emissions, global mixing by atmospheric weather systems over a 1-2 year time-scale makes the IHD quite small or even slightly negative. Increasing emissions primarily in the NH would cause a temporary increase in the IHD, since the tropics inhibit rapid mixing between the hemispheres. After 2012, the IHD in both NOAA and AGAGE measurements rapidly increased [Montzka et al., 2018 and WMO 2018], and updates for 2018 presented at the Symposium show that this larger IHD persists, implying that elevated NH emissions continued through that year [*Montzka*].

Temporarily enhanced CFC-11 concentrations above background ('pollution events') can be easily identified in station observations made downwind of and relatively near sources. Owing to the long CFC-11 lifetime, pollution events are indicative of recent emissions from one or more specific upwind sources (e.g., chemical plants, production facilities, landfills, and building demolition). The Gosan station (South Korea) observations show many such events, and the concentration of CFC-11 in these events has increased since 2012 [*Park; Western;* Rigby et al., 2019]. As with smoke plumes, mixing by the atmosphere causes the CFC-11 concentrations in these plumes to eventually dilute to background levels. Hence, measurements made far downwind contain reduced quantitative information about emission magnitudes and locations in these upwind regions. Many stations outside of eastern Asia (e.g., Mace Head, Ireland) show a decreasing frequency and strength of events, suggesting fairly constant or reduced emissions in regions surrounding these sites over these same years [Rigby et al., 2019]. Station and aircraft observations in eastern Asia also suggest a continued regional source of emissions [*Adcock; Benish; Dang; Simpson;* Lin et al., 2019].

In contrast to global emission estimates, quantitative estimates of regional emissions are more difficult to obtain because they depend on a sufficient number of hourly-to-daily observations from regional stations, along with detailed knowledge of atmospheric air motions that bring air to a station. Emissions have been estimated for several areas of the world, with a recent estimate of emissions from eastern Asia, using observations from Gosan Station (S. Korea) and Hateruma (Japan) [*Western*; Rigby et al., 2019*; Park*]. These two stations are mainly sensitive to emissions only from South Korea, North Korea, southern Japan, and the eastern portion of China that accounts for about 38% of its population. Figure 3 shows the difference between emissions derived for 2014-2017 and for 2008-2012. The majority of these emission increases come from Shandong and Hebei provinces in NE China, and the integrated total increase is 7.0 ± 3.0 Gg/yr, which accounts for at least 40-60 % of the global rise in CFC-11 emission over these years [*Western*; Rigby et al., 2019].

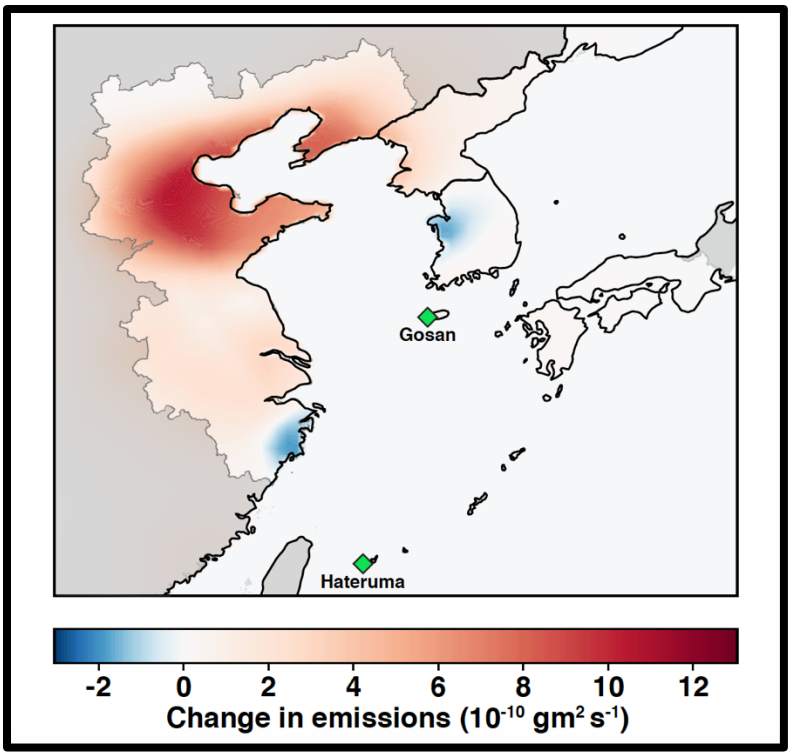

Figure 3. Spatial distribution of the changed CFC-11 emissions from 2008-2012 to 2014-2017 from an inverse analysis of the data from observations at Gosan and Hateruma (diamonds) with the NAME-HB inversion framework. The shaded areas indicate regions with low sensitivity, and therefore, are not included in the estimates of emissions and emission changes [adapted from *Western;* and Rigby et al.,2019].

While these regional inversions provide the best information about the location and strength of the emissions in this region, they require high quality measurements within or closely downwind of this region. Given that such measurements are made in only some regions, the value of this approach to sum over all potential emission regions to obtain global increases is limited (Figure 4). In addition to eastern China, estimates of CFC-11 emissions have been made for recent years for Western Europe [*Manning;* Manning et al., 2003], Australia [*Fraser;* Fraser et al., 2015], and the U.S. [Hu et al., 2017]. While these are important regions, many other important areas are not covered. Indeed, Rigby et al. [2019] point out that any unreported production and resulting emission from eastern China is unlikely to have contributed substantially to the slower than expected global decrease in atmospheric CFC-11 from 2002 to 2012. Understanding the observations during this earlier period requires further work from both a scientific and a technical perspective [*Solomon*] - one possible explanation is continued unreported CFC-11 emission from unknown locations outside of eastern China.

More geographic, though less quantitative, information is available from short-term field studies using aircraft and ground-based sampling. In India, for example, emission estimates are available from a 2-month aircraft campaign in 2016 [Say et al., 2019]. Such studies, which have been performed in China, Korea, Pakistan, Saudi Arabia and Taiwan [*Adcock; Dang; Simpson*], can (a) identify emissions hotspots, as well as areas with low emissions, and (b) provide detailed information on the gases which are co-emitted with CFC-11, and possibly also on the potential CFC-11 sources and/or co-located emissive activities. Such studies are flexible, can be quickly organized, and the information gained can be used to identify priority areas for further research.

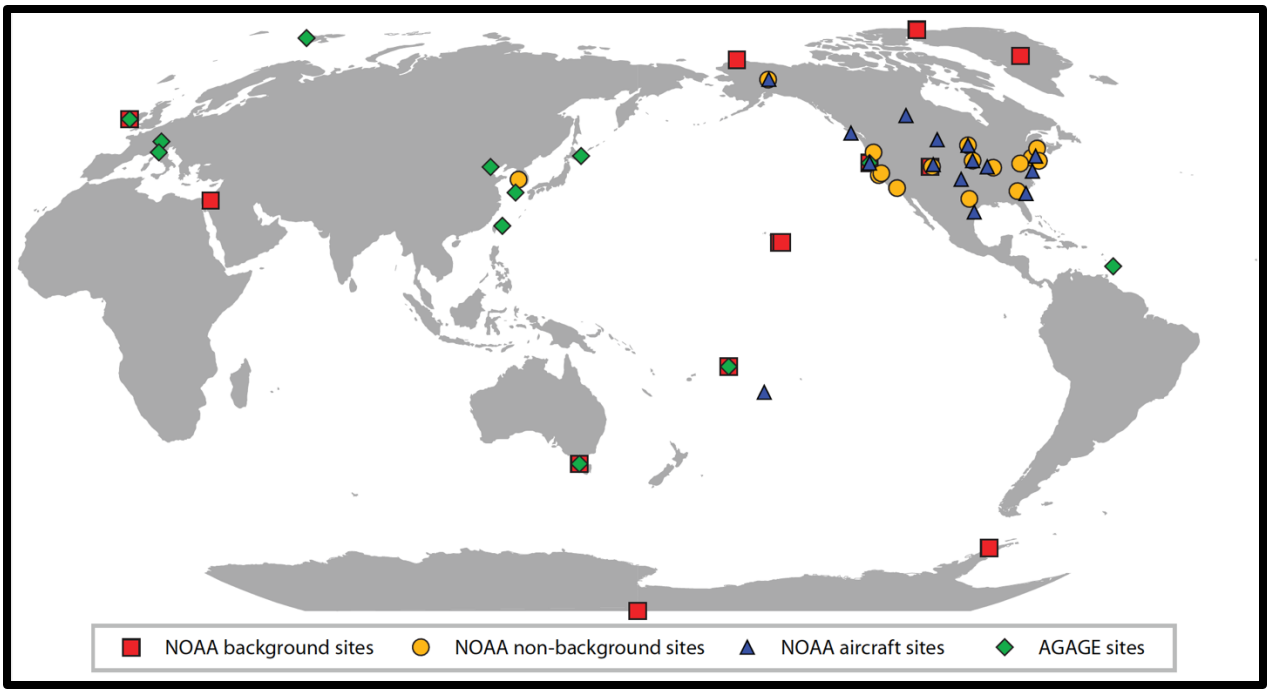

Figure 4. Global map of CFC-11 surface measurement stations. Green diamonds show AGAGE and AGAGE-affiliated site locations.

Do we understand the source(s)?

The possibility that unexpected increased emission rates from banks could lead to a rise in emissions was explored in multiple ways. A new "bottom-up" model was developed and a sensitivity analysis was used to bound the range of possible emissions related to production reported to AFEAS and the Ozone Secretariat changing the relative sizes of the market sectors and emissions rates from production, installation, and banks. The best fit corresponded to the highest emissions rates up to 2002. After that time period, none of the scenarios aligned with the derived emissions.

Emissions rates and banks were also explored for North-Western Europe to better understand bank behaviour relative to atmospheric emissions. The time period of greatest interest (1996 through the latest available data) was used to develop combinations of banks and emissions rates for this region, including an iterative analysis that was found to be consistent with a total CFC-11 bank of 100 to 120 Gg for North-Western Europe. Given this bank size and the atmosphere-derived emissions, a total emissions rate of 3-4% from the bank in 1996 is calculated, which does not align with the global emissions rate of CFC-11 during the period of the global unexpected emissions [*Walter-Terrinoni (a)*].

More extreme scenarios were developed, and scenarios related to newly produced CFC-11, were also explored. More extreme historic emissions rates did not result in scenarios that fit the currently derived emissions profile. Very high emissions rates in recent years were required (24% emissions from the global foams bank for multiple years in a row) to align with the unexpected emissions. Additional usage as a refrigerant did not produce enough emissions either. However, these are extreme scenarios related to direct emissions from production or inventory release, which seems unlikely given economic considerations [*Walter-Terrinoni (b)*].

Scenarios that include additional foam production did align with the global atmospheric top-down emissions in both closed and open cell foams. The competing low cost of dichloromethane, which is used in open-cell foams, makes those open-cell foams scenarios improbable, but the possible use of CFC-11 in closed cell foam could not be eliminated through the sensitivity analyses or due to commercial reasons or based on the analysis of replacement blowing agents [*Walter-Terrinoni (b)*].

The global top-down emissions from the foam blowing agents that replaced CFC-11 were examined and found to align with normal foam uses prior to 2002, but they did not align after 2002 without the inclusion of CFC-11. Further exploration into the potential use of CFC-11 in closed cell foams, and the economic impact of the use of CFC-11 in open cell foams, would help address remaining questions, but to date no plausible process involving emissions from banks has been found to explain the unexpected emission increase [*Walter-Terrinoni (b)*].

Finally, there was a good deal of discussion about whether the CFC-11 was being made in converted large-scale facilities [*Tope*], as would make sense based on standard economic arguments or whether it is occurring in new small-scale plants as have been suggested in China [*Perry*]. Understanding the technical as well as the economic rationale for the production is required to understand this better.

What is the expected impact?

As would be expected, additional CFC-11 emissions over our baseline expectations result in higher future levels of chlorine in the stratosphere. In turn, larger ozone depletions can be expected in the future, with delays in the recovery of ozone to pre-1980 levels. A major problem with understanding the impact of the unexpected increased emissions is to create a realistic projection of future CFC-11 levels [*Daniel*]. As noted earlier, these projections depend on CFC-11 production estimates, emissions from the manufacture of products, and emissions from CFC-11 banks [*Kuijpers; Pons; Reimann; Solomon; Walter-Terrinoni (a); Tope; Walter-Terrinoni (b)*]. These bank emissions depend on the magnitudes of the banks, and the rates of CFC-11 loss from those banks. All of these factors have significant uncertainties, and therefore CFC-11 projections are also poorly understood [*Daniel; Solomon*]. A better understanding of foam banks and emissions rates would also help to further quantify the unexpected emissions of CFC-11 [*Walter-Terrinoni (a)*].

A number of simulated CFC-11 scenarios were shown at the Symposium. The primary results were that increased CFC-11 emissions lead to increased stratospheric ozone depletion as expected, and that the additional accumulated emissions over the next few decades will control the level of additional ozone depletion [*Chipperfield; Eleftheratos; Fleming; Keeble, Liang; Nuetzel*]. At present, a temporary increase of 10-13 Gg/yr will not have a detectable impact on ozone levels, particularly if the emissions quickly decrease [*Chipperfield*]. Ground-based and satellite ozone measurements do not provide evidence yet as to an observational effect of increased CFC-11 emissions to ozone profile trends in middle latitudes [*Eleftheratos*]. However, sustained emissions over decades, e.g., of a total at the 60-80 Gg/yr level, would have a large impact.

A major uncertainty was identified with respect to the potential influence on ozone of co-production of CFC-12 with CFC-11, and the use of CCl_4 as a feedstock for CFC-11 [*Sherry; Tope*]. The consequent growth of emissions from these additional chemicals, including from a possible delayed release from an increased bank of CFC-12, would add to the impact on the ozone layer.

Model simulations of enhanced CFC-11 emissions provide a variety of estimates on the impact to stratospheric ozone to 2100. All models found that recovery was delayed depending on the emission levels [*Chipperfield; Fleming; Keeble, Liang; Nuetzel*]. For various scenarios of continuing high emissions of CFC-11 (e.g. continuing current values of 72.5 Gg/yr until 2100), the ozone hole persists until 2100 [*Keeble*; *Liang*]. There is a linear dependence of ozone depletion on the cumulative CFC-11 emissions to the year 2100, with a global ozone change of -0.29 DU (-0.1%) per 1000 Gg of emitted CFC-11, and an Antarctic spring depletion of -2.4 DU (-0.9%) per 1000 Gg of CFC-11 [*Fleming*].

Future global ozone depletion is also sensitive to the climate scenario, the depletion per 1000 Gg emission was less (-0.25 DU) for the high-GHG RCP8.5[6] than for the lower-GHG RCP2.5 (-0.3 DU) [*Fleming*]. In contrast to global ozone, the Antarctic ozone hole was minimally affected by GHG scenarios, with no perceptible difference in ozone depletion sensitivity to CFC-11 between RCP8.5 and RCP2.5. However, the ozone depletion caused by sustained CFC emissions into the future had significant impacts on the Antarctic stratospheric temperature, the Southern Hemisphere jet strength, and the Brewer-Dobson circulation [*Liang*].

Until the source and, ultimately, the likely future trajectory of the unexpected increased CFC-11 emissions becomes clearer, it will not be possible to provide more constrained emission scenarios for the atmospheric models to explore expected future impacts on stratospheric ozone depletion and climate forcing.

Suggested research directions

Two categories for future research emerged during the Symposium: (1) short-term activities that will inform the various reports to the Parties in the next two years; and (2) longer-term requirements to ensure that the community can respond to this and similar situations in the future. The latter could be related to the controlled HCFCs and HFCs in the Montreal Protocol or to other gases controlled for other purposes (e.g., greenhouse gases).

In the short term (1), the main aims identified are to better bound the problem based on analyses of existing information and to prioritize and perform any quick-response activities that can reduce the major uncertainties. These are scientific and technical in nature and include the following:

a) *Updates based on recent observations.* It is quite possible that this issue's significance will lead to actions that will then bring about rapid emissions reductions. Providing timely feedback to the Parties on any changes in emission rates is highly relevant to their decision making.

b) *Identify other possible areas of CFC-11 emissions.* While the unreported emissions were initially detected at the global scale, the sparseness of existing measurement sites means that little is known about possible emissions from many parts of the world. Analysis of existing measurements and organising focussed, internationally recognised measurement campaigns in priority areas (e.g. those with the capacity for CFC production) could rule in and/or rule out locations reasonably quickly.

c) *Providing improved knowledge on the time history of emissions*. There is clearly more that can be derived from atmospheric observations that have already been made, from new analyses of CFC-11 emissions from banks (e.g. emission rates from the disposal of foams, including breakdown rates in different types of landfill) and from foam production, etc. Further modeling work to better understand the influence of atmospheric dynamics in interannual changes in CFC-11 concentrations could better constrain global emission estimates.

d) *Review known plant capacities and other pathways for CFC-11 production.* Some plants can in principle be switched to produce CFC-11. Identifying these and their potential capacities could provide insights into how CFC-11 could be produced using established techniques in the quantities required to supply potential new foam production. This would complement analyses of potential new or less common CFC-11 production pathways.

e) *Improved assessment of unreported CFC-12 co-production.* Given current understanding of the production of CFC-11, there is a good chance that additional CFC-12 is being co-produced. If so,

[6] RCP (Representative Concentration Pathway) is a future greenhouse gas concentration trajectory adopted by the IPCC for its 5th Assessment Report [AR5] in 2014. The number (e.g., 8.5) is the expected radiative forcing in Watts per meter squared expected for those concentrations.

it has not been detected in atmospheric measurements as yet, so either it is not being released or any amount produced and subsequently released cannot currently be detected by the observational system. Addressing this requires a mixture of analyses of near-source atmospheric observations and new assessments of possible CFC-12 uses and banks.

f) Improved understanding of the relationship between the locations of production and emissions. CFC-11 containing products, such as foams, can be found in dispersed locations, including those well away from production plants. Better knowledge of the geographic distribution of foams and other products would further constrain our understanding of where emissions may occur.

Addressing these short-term research goals has the potential to put bounds on the nature and scale of the overall CFC-11 emissions problem and provide a more rounded scientific and technical basis for possible policy responses.

The emergence of the unexpected increased CFC-11 emissions raises questions about the completeness of our understanding of the ODS banks and new production and provides valuable lessons that can be taken into account when considering how to monitor emissions of other gases with restricted production. This applies to substances controlled under the Montreal Protocol and other greenhouse gases.

In the longer term (2), the main goals identified were to ensure on-going effective responses to this and similar future situations. These include the following:

a) Improve the current monitoring system to provide quantitative estimates of regional emissions throughout the world. Unmonitored regions include the Western and central China, India, Russia, Eastern Europe, Southeast Asia, South America, Africa and some of North America. Maintaining a robust system with better coverage, scientific expertise, and long-term continuity is critical.

b) Pursue a mix of atmospheric monitoring approaches. Increasing the coverage of the monitoring network requires developing and maintaining international capabilities for short-term, intensive field studies. These include static, spot or mobile measurements near source locations that ideally are coordinated with available observational systems (e.g. local government air quality networks) which can provide ancillary information. It will be important to identify priority areas for such intensive studies.

c) Reduce uncertainties in emission estimates from global and regional inversion studies. This topic is already a priority due to its importance for other greenhouse gases; nonetheless, it is important to recognise its broad importance.

d) Develop a set of more plausible CFC-11 and other ODS emission scenarios. These emission scenarios are highly dependent on production estimates, total amounts in banks, and emission rates from those banks. Constraining these scenarios would enable more realistic estimates of the possible impact on O3, UV radiation, and radiative forcing.

e) Assess similar issues that could result from the implementation of the Montreal Protocol. The production and consumption of CFCs, HCFCs and HFCs are controlled under the Montreal Protocol. It will be helpful to continue to identify and analyse potential 'pinch-points' where unreported production might be more likely to occur, e.g. when the cost of producing the replacement is more than the cost of producing the banned substance. For CFC-11, with major replacements HCFC-141b, HFC-365mfc and HFC-245fa, this could involve modelling emissions rates (to cross check with other foam blowing agent types) and to reverse engineer banks and examine transitions and compare them to production and consumption.

f) Improve our understanding of emissions from banks. Updated emissions rates from foams and further exploration of the timing of emissions from banks by region to further quantify the unexpected emissions geographically (see (d) above).

3. Summary

The Symposium brought together an international community of experts with technical and scientific expertise to update the understanding about the unexpected emissions of CFC-11. A wide range of results were presented including a review of previous work, new or updated estimates of emissions based on atmospheric measurements, field studies of the mixture of gases emitted by a number of sources, technical assessment of possible new sources of CFC-11 emissions, and atmospheric modelling studies of the impact of continuing new emissions on stratospheric ozone.

One major conclusion is that the evidence that a substantial fraction of the unreported emissions are from eastern China is now stronger. This evidence is consistent with the increase in the IHD. Conversely there is no evidence for unreported emissions from other regions, though that can certainly not be ruled out as the coverage of other regions by existing observational studies is incomplete. Sustained emissions at current rates would lead to a delay in ozone recovery.

The most likely use of the newly produced CFC-11 is thought to be in manufacturing foams as there is large global demand for insulating materials. So, a consistent picture is emerging. However, it is hard from a technical perspective to explain how the CFC-11 is being produced, whether in re-conversion of large production plants or the use of new micro-scale plants. Questions underlying this aspect are the subject of the TEAP report being produced for the 31st Meeting of the Parties in November 2019.

4. References

Papers:

Fraser P. J., B. L. Dunse, P. B. Krummel, L. P. Steele and N. Derek, Australian and Global Emissions of Ozone Depleting Substances, Report prepared for Department of the Environment, CSIRO Oceans and Atmosphere Flagship, Collaboration for Australian Weather and Climate Research, Aspendale, Australia, iii, 29 pp., (2015).

Hu, L., et al., Considerable contribution of the Montreal Protocol to declining greenhouse gas emissions from the United States, *Geophys. Res. Lett., 44*, 8075–8083, doi:10.1002/2017GL074388 (2017).

Lin, Y., et al., Observations of High Levels of Ozone-Depleting CFC-11 at a Remote Mountain-Top Site in Southern China, *Environ. Sci. Technol. Lett., 6,* 3, 114-118, doi: 10.1021/acs.estlett.9b00022 (2019).

Lunt, M. F., et al., Continued emissions of the ozone-depleting substance carbon tetrachloride from eastern Asia, *Geophys. Res. Lett., 45*, 11,423–11,430, doi: 10.1029/2018GL079500 (2018).

Manning, A. J., D. B. Ryall, R. G. Derwent, P. G. Simmonds, and S. O'Doherty, Estimating European emissions of ozone-depleting and greenhouse gases using observations and a modeling back-attribution technique, *J. Geophys. Res.*, 108 (D14), 4405, doi:10.1029/2002JD002312 (2003).

Montzka, S. A. et al. An unexpected and persistent increase in global emissions of ozone-depleting CFC-11. *Nature, 557*, 413–417, doi: 10.1038/s41586-018-0106-2 (2018).

Park, S., et al., Toward resolving the budget discrepancy of ozone-depleting carbon tetrachloride (CCl_4): an analysis of top-down emissions from China, *Atmos. Chem. Phys., 18*, 11729-11738, doi: 10.5194/acp-18-11729-2018 (2018).

Rigby, M., et al., Increase in CFC-11 emissions from eastern China based on atmospheric observations, *Nature,* 569, 546–550, doi: 10.1038/s41586-019-1193-4 (2019).

Say, D., Ganesan, A. L., Lunt, M. F., Rigby, M., O'Doherty, S., Harth, C., Manning, A. J., Krummel, P. B., and Bauguitte, S.: Emissions of CFCs, HCFCs and HFCs from India, *Atmos. Chem. Phys. Discuss.,* doi: 10.5194/acp-2018-1146, in review (2019).

TEAP, May 2005 Report of the Task Force on Foam End-of-Life Issues, Montreal Protocol On Substances that Deplete the Ozone Layer, UNEP Technology and Economic Assessment Panel, United Nations Environment Programme, Nairobi, Kenya (2005).

WMO (World Meteorological Organization), Scientific Assessment of Ozone Depletion: 2018, *Global Ozone Research and Monitoring Project* – Report No. 58, 588 pp., Geneva, Switzerland (2018).

Symposium Presentations:

Adcock, K. E. et al., 'The origin of high concentrations of CFC-11 observed in Taiwan'. (03/25/19, 1200).

Arduini, J. et al., 'CFC measurements from Nepal Climate Observatory - Pyramid (Nepal), GAW global station'. (03/25/19 1220).

Benish, S. E. et al., 'Aircraft Measurements of Elevated CFC-11 Concentrations over the North China Plain in Spring 2016'. (03/25/19 1400).

Chipperfield, M. et al., 'Determination of the Sources and Implications of Increased CFC-11 Emissions using Inverse and Forward 3-D Modelling'. (03/27/19 1400).

Dang, G. et al., 'Long-term spatiotemporal variations and source changes of halocarbons in the greater Pearl River Delta region, China'. (03/25/19 1420).

Daniel, J., Carpenter, L., 'A path towards more useful future CFC scenarios'. (03/26/19 1120).

Eleftheratos, K. et al., 'A note on possible effects of the unexpected increase in global CFC-11 to ozone profiles and erythemal doses at ground level'. (03/27/19 1420).

Fang, X. et al., 'Rapid increase in ozone-depleting CHCl3 emissions from eastern China'. (03/25/19 1440).

Fleming, E. et al., 'The impact of continuing emissions of CFC-11 on the stratospheric ozone layer'. (03/27/19 1140).

Fraser, P. et al., 'Chlorofluorocarbon (CFC-11, CFC-12, CFC-113) emissions in Australia: 1962-2017'. (03/26/19 1340).

Pons, J., 'Aerosol, solvent and other misc. uses of CFC-11 and CFC-12'. (03/25/19 1650).

Keeble, J. et al., 'The Impact of Recent East Asian Emissions of CFC-11 on Ozone Recovery'. (03/27/19 1120).

Kuijpers, L. et al., 'Considering total bottom-up emissions and comparisons with top-down numbers'. (03/26/19 1200).

Kuijpers, L. et al., 'CFC refrigerant banks and emissions 1990-2010-2018 '. (03/25/19 1640).

Laeng, A. et al., 'On natural atmospheric variability of CFC-11 '. (03/27/19 1000).

Laube, J. et al., 'Distributions and correlations of CFC-11 and other trace gases in the upper troposphere and stratosphere'. (03/26/19 1620).

Liang, Q. et al., 'GEOSCCM simulations of the Antarctic ozone hole changes due to continuing CFC-11 emissions'. (03/27/19 1340).

Manning, A. et al., 'Estimating CFC-11 emissions over Western Europe from atmospheric observations'. (03/26/19 1400).

Montzka, S. et al., 'Atmospheric measurements of CFC-11 through 2018: Are global CFC-11 emissions back on the decline?'. (03/25/19 1020).

Mueller, R. et al., 'Stratospheric loss of Stratospheric loss of CFC-11 and age-of-air in the Chemical Lagrangian model of the Stratosphere (CLaMS)'. (03/27/19 0920).

Nützel, M. et al., 'Implications of constant CFC-11 concentrations for the future ozone layer'. (03/27/19 1200).

Park, S. et al., 'Identifying potential CFC-11 emission sources in China based on atmospheric observations from 2008 to 2016 '. (03/25/19 1040).

Perry, C., 'Understanding and Mitigating the Impacts of Illegal CFC-11 Use in the Production of Polyurethane Foams'. (03/26/19 0940).

Portmann, R. et al., 'Interannual Stratospheric Transport Variability Impacts on Surface Trace Gas Concentrations'. (03/26/19 1640).

Prather, M., Ruiz, D., 'Understanding the role of the stratosphere on the lifetime and surface variability of CFC-11'. (03/26/19 1500).

Reimann, S., 'Additional CFC-11 emissions: Foam is the only answer, is it?'. (03/26/19 1000).

Schuck, T. et al., 'New Long-term Measurements of CFC-11 and other halocarbons at Taunus Observatory'. (03/26/19 1420).

Sheese, P. et al., 'Recent changes in CFCs in the upper troposphere – lower stratosphere from the ACE-FTS satellite instrument'. (03/27/19 0940).

Sherry, D., 'Production pathways and usages of CFC-11 from carbon tetrachloride'. (03/26/19 0920).

Simpson, I. et al., 'Recent CFC-11 enhancements in China, Korea, Saudi Arabia and Pakistan'. (03/25/19 1500).

Solomon, S. et al., 'Evaluation of Chlorofluorocarbon Banks, Uncertainties, and Implications for Emissions'. (03/26/19 1100).

Tope, H., et al. 'Understanding the production of CFC-11, associated chemicals, and related emissions '. (03/25/19 1600).

Vollmer, M. et al., 'Increasing Emissions of Montreal Protocol Substances Other than CFC-11'. (03/26/19 1440).

Walter-Terrinoni, H. et al., (a) 'Foam Uses '. (03/25/19 1620).

Walter-Terrinoni, H. et al., (b) 'Emissions & Hypothetical Release Scenarios'. (03/26/19 1140).

Western, L. et al., 'Estimates of CFC-11 emissions from eastern Asia based on atmospheric measurements and inverse modeling'. (03/25/19 1140).

5. Acknowledgements

The organizers wish to thank the Parties to the MP for their financial support along with thanks to the Ozone Secretariat of the MP, NASA, and the WCRP/SPARC program for their logistical support. Financial support for attendee travel was also provided by the MP, NASA, WCRP/SPARC, NOAA, USGCRP, and WMO. We also acknowledge the Symposium participants for their active contributions at the Symposium and assistance with the preparation and review of this article, in particular Helen Tope and Helen Walter-Terrinoni.