A COST-EFFECTIVE TRACE GAS MEASUREMENT PROGRAM FOR LONG-TERM MONITORING OF THE STRATOSPHERIC CIRCULATION

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A low-cost, long-term trace gas measurement program having the capability to uniquely monitor key aspects of the stratospheric circulation is proposed.

The stratospheric mean meridional circulation, commonly referred to as the Brewer–Dobson circulation (BDC), consists of upwelling in the tropics, poleward motion away from the tropics, and downwelling at mid- to high latitudes. State-of-the-art coupled chemistry–climate models predict an increase in the strength of the BDC with increasing atmospheric concentrations of greenhouse gases (e.g., Garcia and Randel 2008; McLandress and Shepherd 2009). Changes in the BDC will likely impact both the recovery of the stratospheric ozone layer (Butchart and Scaife 2001; Butchart et al. 2010) and the concentration of stratospheric water vapor entering the stratosphere (Randel et al. 2006), with subsequent radiative impacts (Forster and Shine 2002). In recent years, evidence has been rapidly accumulating that there are stratospheric climate change circulation feedbacks on tropospheric climate (e.g., Gerber et al. 2012; Karpechko and Manzini 2012; Scaife et al. 2012; Solomon et al. 2010; Dall'Amico et al. 2010). Gerber et al. (2012) sums up many of these feedbacks and highlights recent climate modeling efforts that include a realistic stratosphere [i.e., the Coupled Model Intercomparison Project phase 5 (CMIP5)] showing that the stratosphere has an impact on tropospheric climate. It has been shown that recent decadal changes in lower-stratospheric water vapor enhanced (from 1980 to 2000) or slowed (following 2000) the global surface temperature changes expected solely from increases in greenhouse gases by as much as 30% (Solomon et al. 2010). Changes in stratospheric water vapor can be directly caused by changes in the Brewer–Dobson circulation through modification of tropical cold-point temperatures. In a seasonal sense, a stronger circulation in boreal winter results in lower tropical cold-point temperatures and a smaller amount of water vapor entering the stratosphere than in boreal summer (Mote et al. 1996). Changes in the stratospheric zonal winds have been shown to induce

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a change in baroclinic eddy growth rates that shift storm tracks toward the equator, increase "storminess," and carry with them corresponding changes in precipitation distributions (Scaife et al. 2012). Such changes in turn modify the generation of wave activity, the major driver of the stratospheric circulation (e.g., Haynes et al. 1991; Plumb 2002). There is a growing understanding that climate models will be severely limited if they do not incorporate a realistic and accurate stratospheric circulation.

To help improve climate model simulations of the stratospheric circulation, climate-monitoring programs should have a stratospheric-circulation monitoring component. The original discovery of the BDC was based on measurements of lower-stratospheric water vapor (Brewer 1949) and ozone (Dobson 1956). Because the BDC describes the circulation responsible for the distribution of trace gases in the stratosphere, it has both a mean and distributed mass flux component and a quasi-horizontal mixing, or redistribution component (Fig. 1). We now have the capability to measure a variety of trace gases in addition to water vapor and ozone that are uniquely influenced by both the mean and distributed mass flux of the stratosphere and the redistribution due to mixing. It is important to have this suite of measurements in order to better understand both components of the BDC, since especially the redistribution due to mixing has not been thoroughly investigated in models primarily because of a lack of sufficient observations.

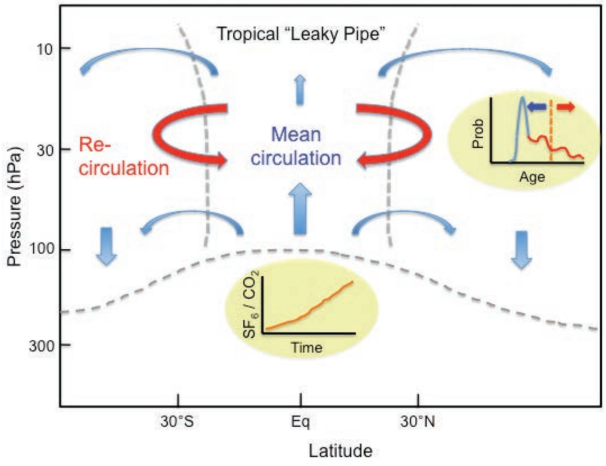

Fig. 1. A schematic with the main components of the stratospheric circulation. The mean meridional circulation (blue arrows) consists of upward motion in the tropics and downward motion in the extratropics. Recirculation (red arrows) is the mixing of air from the extratropics back into the tropics. The dashed gray lines denote the altitude of tropopause and the edges of the tropics in the stratosphere. The plot in the tropical upper troposphere represents the time series of trace gases in near-linear growth, such as SF_6 and CO_2, used to estimate the mean age of air in the stratosphere. The plot in the NH stratosphere represents an age distribution—i.e., the time that individual particles within an air parcel have resided in the stratosphere. The blue line in this plot represents the portion of the age distribution solely due to the mean circulation. The red line represents the long age tail of the distribution due to mixing between the extratropics and tropics. The orange dashed line is the mean age of this distribution.

At the present time, the cost of collecting these measurements on a routine basis using in situ techniques is prohibitive; these measurements have only been taken during occasional field campaigns. Accurately monitoring the stratospheric circulation requires high-quality, long-term measurements of a variety of long-lived trace gases. Recent measurements of SF_6 from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument on the *Environmental Satellite* (*Envisat*) show the most promise of any satellite measurements to reveal stratospheric circulation variability (Stiller et al. 2012). SF_6 can be used to estimate the mean age of air, an important indicator of the strength of the stratospheric circulation, as described in section 2. But a satellite instrument such as MIPAS was not designed for long-term monitoring and, in fact, communication with *Envisat* was lost in early 2012, ending the mission. Discontinuous satellite records make trend analysis problematic. Additionally, current satellite measurements of gases used to infer stratospheric age of air do not have the precision and vertical resolution required to quantify small and more complex changes in the components of the BDC.

To generate a long-term, cost-effective series of trace gas measurements to monitor the stratospheric circulation, we propose using a novel measurement technique called "AirCore" (Karion et al. 2010) combined with ongoing trace gas sonde measurements. The AirCore technique has been shown to be a viable means of collecting high-vertical-resolution profiles at altitudes from the surface to nearly 30 km (or ~10 hPa). For the circulation monitoring we propose here, we only require the stratospheric portion of the profile. The measurement payload is extremely light and low cost, comparable to ozone sondes, water vapor sondes, and radiosondes. This allows establishing measurement sites at a number of locations around the world with measurements of a high temporal frequency and vertical resolution, allowing monitoring of seasonal to interannual variability in the stratospheric circulation. As with other National Oceanic and Atmospheric Administration (NOAA) Global Monitoring Division long-term trace gas monitoring programs, air samples would be analyzed using a single well-calibrated instrument. In this article we briefly describe the main techniques we propose to use for quantifying the stratospheric circulation from trace gas measurements and outline the potential measurement program.

QUANTIFYING STRATOSPHERIC CIRCULATION FROM TRACE GAS MEASUREMENTS.

Photolytic trace gases: Unique indicators of stratospheric transport. Trace gases that have stratospheric loss determined almost entirely by photolysis include N_2O, CFC-12, CFC-113, CFC-11, and halon-1211. These trace gases not only have a very specific loss process, but the photolytic loss rates increase exponentially with increasing height and are unique for each gas. Figure 2 shows local photolytic lifetime profiles in the tropics for the trace gases mentioned above. Each of these trace gases has negligible loss at levels below the altitude where the local photolytic lifetime is roughly the same as the transport time scale in the stratosphere. We refer to the altitude range where the local photolytic lifetime of a particular trace gas is roughly equal to the transport time scale as the "cutoff level." The cutoff level for each trace gas is denoted in Fig. 2 by the altitude of the intersection of the vertical blue bar (a 1–2-month local time scale) with the local photolytic lifetime profile. Above its cutoff level, a trace gas is rapidly and completely destroyed. This means that the difference from the tropospheric average mixing ratio of any of these photolytic trace gases at altitudes below its cutoff level is due to transport of air from above the cutoff level. These tracers are therefore a sensitive measure of the mass flux and transport from above each respective cutoff level.

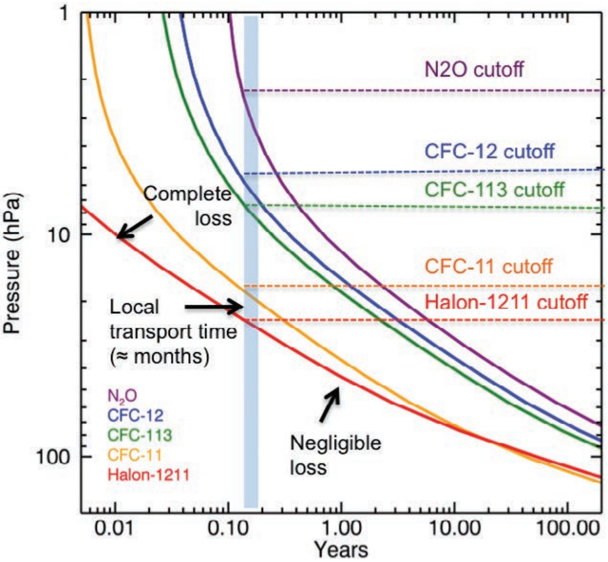

Fig. 2. Tropical average local photolytic lifetime profiles for our targeted trace gases. The values shown are from a 2D model of the stratosphere and are averaged from 20°S to 20°N (Portmann et al. 1999). Note the exponential decrease in lifetime with altitude. The vertical light-blue-shaded region represents the transport time scales in the stratosphere. The level where the local photolytic lifetime of a particular trace gas is equivalent to the transport time scale is referred to as the cutoff level for that tracer. The colored dashed lines represent the cutoff levels for each of the trace gases. For altitudes above a cutoff level, the trace gas is rapidly destroyed compared to the local time scale for transport into and out of that region (near 100% loss is expected). For altitudes below the cutoff level, there is negligible loss compared to the time scales for transport into and out of those regions.

This unique loss of the photolytic trace gases has been utilized in a number of studies to investigate stratospheric transport in both models and measurements (e.g., Plumb and Ko 1992; Volk et al. 1996; Plumb et al. 2000; Hall 2000). We intend to make use of techniques described in Volk et al. (1996) to estimate the vertical profile of advection in the tropics, which quantifies mass flux, and the entrainment of midlatitude air into the tropics, referred to as recirculation. This analysis requires measured photolytic trace gas profiles. These techniques take advantage of the compact correlations between photolytic trace gases within the tropics and extratropics. We can then use these estimates of tropical mass flux and recirculation as inputs into a simplified model of the stratosphere, the "tropical leaky pipe" (TLP) model (Neu and Plumb 1999). The TLP model will be constrained further by simultaneous SF_6 profiles that define mean age throughout the stratosphere. This model has been shown to effectively simulate average stratospheric trace gas profiles (e.g., Ray et al. 2010). With the TLP model we can refine the mean mass flux and recirculation estimates to best fit the measured trace gas profiles and correlations. The TLP model resolves time-scale variability from subseasonal to multidecadal and can be run for many different scenarios with minimal computational cost.

As an example of the utility of the TLP model for our purpose, we performed a number of simulations to quantify the sensitivity of measured trace gas profiles to changes in the stratospheric mean circulation, recirculation, and photolysis rates. These photolysis rates are anticipated to change because of changes in stratospheric ozone. Figure 3 shows modeled changes in the photolytic tracers in the tropics in response to a 5% increase in the strength of the mean meridional circulation throughout the depth of the stratosphere. All other inputs were held fixed in this simulation and the results shown are annually averaged. The shaded regions in the figure represent the amount of change we would be sensitive to with the expected measurement precision of 1% or less. The model results indicate that we would be able to detect mean circulation changes of at least 3%–5%. Current chemistry–climate model (CCM) simulations suggest a 2%–3% decade^{-1} increase in the strength of the mean meridional circulation throughout the twenty-first century (Butchart et al. 2010). Thus, our measurements and analysis technique would be able to detect a trend of this size in 10–25 years. Simulations with recirculation and photolysis rate changes showed the tracers were sensitive at the 1% level to 4%–5% changes in those inputs (not shown).

There are important caveats to the above-mentioned calculations and our ability to detect changes in the stratospheric circulation with photolytic trace gas measurements. One is that variability in tracer profiles due to the seasonal cycle and interannual forcing, such as the quasi-biennial oscillation (QBO), needs to be understood. Not only do we need to understand these relatively short-term modes of variability to help detect a trend, but they are also important and interesting to understand in their own right. These modes of variability in the trace gases are quite large, on the order of 5%–10%, so they are easily detected as long as measurements are taken with enough frequency. This is an essential reason to take enough air samples to obtain representative profiles at each location with a frequency of at least four times per year. Investigating this variability is also aided by having a set of trace gas measurements with unique sensitivity to different regions of the stratosphere and different types of transport changes.

A second caveat is our ability to obtain a representative profile at each location and season due to the zonal and meridional gradients in long-lived trace gases, primarily in the extratropics. Our previous measurement campaigns have indicated extensive filamentary structure in tracer profiles and in most cases it is clear that the filaments deviate from a background profile. This filamentary structure in trace gas mixing ratios is a result of an individual profile sampling a range of equivalent latitudes (Butchart and Remsberg 1986) different from the actual latitude where the flight took place. To obtain a representative profile from a particular location and season, we intend to take a number of samples within a several-week period during that season. The goal of this strategy is to make sure that the uncertainty of the representative profile associated with sampling at each location is small enough so that it will not impact our ability to detect relevant variability in the stratospheric circulation. We performed an analysis using Microwave Limb Sounder (MLS) N_2O measurements and Modern-Era Retrospective Analysis for Research and Applications (MERRA) reanalysis output to try to quantify the uncertainty on a representative profile due to equivalent latitude variability. For this exercise we chose Northern Hemisphere midlatitude locations, since this region is expected to have a relatively large sampling uncertainty. We obtained statistics on a range of sampling strategies, varying both the number of individual profiles taken each season and the gap in days between profiles. The results of this analysis indicate that sampling 3–6 times in a 2–3-week period and using equivalent latitude to filter the measurements can result in a sampling uncertainty of 1% or less.

A third caveat is that several mechanisms can interact to change tracer profiles in conflicting ways. Simultaneous changes in both the mean meridional circulation and recirculation will complicate the interpretation of changes in tracer profiles. However, the photolytic tracers have unique sensitivities to each forcing and with our simple model we can investigate changes due to a combination of factors.

Time scales of stratospheric transport. Time scales for transport in the stratosphere can be quantified using the concept of "age" of air (Waugh and Hall, 2002). Age is defined by the time an air parcel has resided in the stratosphere after entering through the tropical tropopause. Trace gas species for estimating stratospheric age of air should have negligible stratospheric production or loss and near-linear tropospheric growth; SF_6 and CO_2 are two such examples (e.g., Hall et al. 2011). The stratospheric profiles of these trace gases are consequently defined solely by their growth at the tropical tropopause (the gateway to the stratosphere) and the average length of time it takes for air to reach a given location in the profile. The average length of time at a given location is not just the result of direct transport through the stratospheric mean meridional circulation, but it is also substantially modified by recirculation, whereby entrainment of midlatitude air is mixed back into the tropics, resulting in longer average transit times (Fig. 1). Thus, the age of air is not solely a function of the strength of the mean meridional circulation but is also impacted by the amount of mixing, and thereby reveals information about both processes. This also means that changes in the age of air cannot uniquely deconvolve changes because of the mean meridional circulation and changes in recirculation because of tropical entrainment of midlatitude air.

Typically what is calculated in models and derived from observations is the mean age of air (e.g., Harnisch et al. 1996; Andrews et al. 2001; Engel et al.

2009). An individual air parcel is actually composed of particles with a range of transit times. The probability distribution of these times is the age spectrum (graph in the top right of Fig. 1), and the first moment of the distribution is the mean age. The strength of the mean meridional circulation defines the leading peak of the age spectra, and the mixing from midlatitudes to tropics defines the old age tail (blue and red peaks, respectively) of the age spectra in Fig. 1. Nearly all CCMs simulate a strengthening mean meridional circulation and decreasing mean age throughout the stratosphere over the past few decades with increasing greenhouse gas simulations (Butchart et al. 2010). In fact, it has been shown that different CCMs display a nearly linear inverse relationship between the strength of the mean meridional circulation and the mean age of air (Austin and Li 2006; Garcia and Randel 2008). However, it is not clear that this relationship holds in the real stratosphere. Any increase in the wave activity from the troposphere that drives the Brewer–Dobson circulation (Shepherd and McLandress 2011) may also enhance mixing between the midlatitudes and tropics. This will have the competing effect of increasing mean age by adding weight to the tail of the age spectrum.

The possibility that changes in mixing are not well represented in global models can help explain the apparent discrepancy between model predictions of younger age trends due to climate change and ages derived from measurements. A recent study by Engel et al. (2009) and extended by Ray et al. (2010) pieced together available balloon-based SF_6 and CO_2 measurements over the past three decades to show that the mean age of stratospheric air in the Northern Hemisphere midlatitudes had increased, in apparent opposition to the decreased mean age predicted by CCMs (Fig. 4). However, it should be noted that uncertainties due to limited sampling may be large. Variability in mean age due to the seasonal cycle is quite large, as seen by the modeled mean ages (green lines in Fig. 4). The model/measurement discrepancies and the large seasonal cycle in mean age highlight the need for a regular stratospheric circulation-monitoring program that can resolve the seasonal cycle to reduce trend calculation uncertainties and improve understanding of the relevant processes.

In our simple model simulation described in the previous section and shown in Fig. 3, the mean age changes are indicated by the light blue lines. The shaded regions on the plot represent mean age changes of more than 0.25 years, since this is the lowest detectable change based on the precision of the SF_6 measurement and the uncertainty in the calculation of mean age. It is apparent from this result that mean age changes because a 5% mean circulation change is not sufficiently large to be detected, whereas the 5% mean circulation change is detectable in the photolytic tracer profiles given the caveats mentioned above.

Benefits of a range of trace gas measurements. The combined set of tracers described here (SF_6, N_2O, CFC-12, CFC-113, CFC-11, and halon-1211) is sensitive to both the strength and the depth of the BDC in unique ways, and it represents a near-complete dataset for monitoring changes in the mean mass flux as well as recirculation due to mixing between the tropics and extratropics. It is important to note that information from the photolytic tracers is confined to altitudes above the lowest cutoff level (in this case for halon-1211).

Additional information on the stratospheric mass flux below the halon-1211 cutoff level can and should be obtained by including CO_2, water vapor, and ozone measurements. Water vapor contains a propagating signal in the lower stratosphere because of the variation of temperature, and subsequent dehydration, at

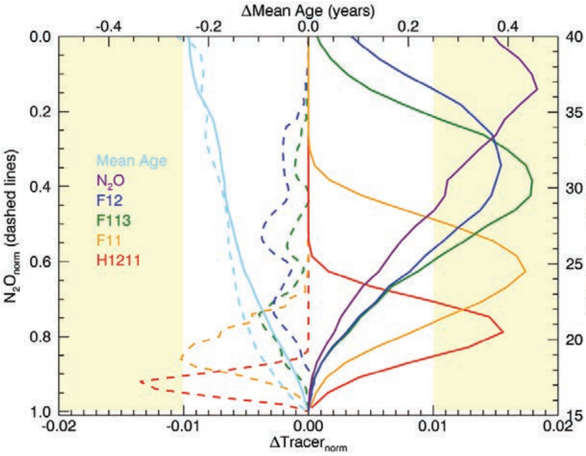

Fig. 3. Modeled tropical sensitivity profiles for photolytic trace gases and mean age in response to a 5% increase in the strength of the mean meridional circulation throughout the stratosphere. The solid lines represent the tracer (bottom axis) and mean age (top axis) changes as a function of altitude (left axis), and the dashed lines represent tracer and mean age changes as a function of normalized N_2O (right axis). Each of the tracers was normalized to the tropical tropopause value before differences were taken. So tracer differences are expressed as a fraction of the tropical tropopause value. The precision and accuracy of the measurements is 1% or better, so the detection limit of a change in the tracers is shaded at values greater than 0.01 to indicate where the changes in each profile are large enough to be detected. The calculation of mean age from SF_6 has an uncertainty of 0.25 year or less, so the shaded area represents detectable mean age changes of 0.25 year or larger.

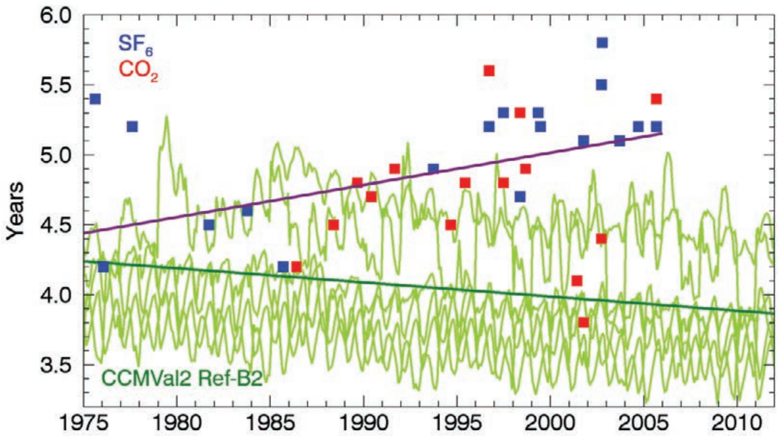

Fig. 4. Mean age of air averaged from 24 to 35 km based on SF_6 measurements in blue and CO_2 measurements in red. Mean age output averaged from 30° to 50°N and over the same altitude range for six different CCMs that were part of the CCM Validation phase 2 (CCMVal2) activity. Solid lines are linear fits through the observations (purple) and the average of all the model ages (green). The observations show an ageing trend in apparent opposition to the CCMVal2 model output.

the tropical tropopause (e.g., Mote et al. 1996). CO_2, which is currently measured as part of the AirCore program, can also be used to infer information about the mass flux in the lower stratosphere from the propagation and damping of the seasonally varying entry values at the tropical tropopause (e.g., Sawa et al. 2008). Tracking the propagating signals of these tracers upward within the tropics and meridionally out to the extratropics is an indicator of lower-stratospheric mean flow and mixing. Ozone is relatively long lived in the lower stratosphere and so it can also give information about transport in this region (e.g., Ploeger et al. 2012). Water vapor and ozone are currently measured on other relatively low-cost balloonborne packages, and lower-stratospheric CO_2 measurements by the AirCore technique are becoming more prevalent. The measurements of these complementary trace gases would not need to be simultaneous in time but would need to sample the seasonality and latitudinal coverage of the age and photolytic tracers. Additional information could also be obtained from satellite measurements in the lower stratosphere. These satellite measurements will help to connect the vertical profiles of our program to the global stratosphere.

In the winter polar vortices, measurements of several additional tracers with production or loss in the mesosphere would allow information about mesospheric influence and mass flux into the stratosphere. Trace gases such as CO and H_2, which could potentially be measured from air samples taken as part of this program, have production in the mesosphere. Combining these tracers with SF_6 and CO_2 would be useful for tracking mesospheric influence in the polar vortex (e.g., Ray et al. 2002). The mesospheric mass flux into the stratosphere is important for models to capture correctly, since the mesospheric circulation is primarily driven by gravity wave breaking (e.g., Alexander et al. 2010). Since gravity wave fluxes are not well constrained observationally, the variability of momentum deposition by gravity waves in the mesosphere as well as the variability of the mesospheric circulation is still somewhat uncertain. Near-simultaneous launching of different balloonborne sampling devices in the winter polar vortices could provide valuable information about the middle atmospheric circulation in the upper stratosphere and mesosphere.

STRATOSPHERIC MEASUREMENT PROGRAM. This stratospheric measurement program takes its inspiration from the AirCore measurement technique. In 2009 the NOAA Global Monitoring Division (GMD) Carbon Cycle and Greenhouse Gases Group (CCGG) began preliminary studies (Karion et al. 2010) with the AirCore (Tans 2006). The AirCore consists of a long tube filled with a known standard reference atmospheric gas mixture, and is open on one end. The tube is lofted into the air with a weather balloon. As it gains altitude, the tube depressurizes. When dropped from altitude, the tube fills back up with ambient air. Mixing of newly entered air with air that entered earlier, which gets compressed toward the back of the tube, is minimal. The tube is recovered after flight and the air is chemically analyzed in the laboratory. By keeping track of time, pressure, the temperatures of outside air and the tube itself, along with GPS altitude, the analyzed air can be accurately registered back to the sampled vertical levels in the atmosphere. Using a Picarro cavity ring-down instrument, CO_2 and CH_4 are being successfully measured using the AirCore technology (Karion et al. 2010).

In late 2010 the NOAA GMD Halocarbons and other Trace Species (HATS) group realized that stratospheric measurements based on an AirCore could be coupled to laboratory gas chromatographs (GC). GC sampling techniques have been developed from many previous airborne campaigns and incorporate small sample volumes of one standard cubic centimeter (scc) (Moore et al. 2003). A 1-scc GC sample volume corresponds to a high vertical resolution of 5 hPa. The AirCore technique coupled to a GC will bring the cost of a stratospheric profile of the targeted long-lived trace gases down to that comparable with ozone and water vapor sonde measurements [~$3,000 (U.S. dollars)]. Presently, NOAA GMD uses the Lightweight Airborne Chromatograph Experiment (LACE), a balloonborne in situ GC to measure the relevant age of air and photolytic tracers needed to monitor stratospheric circulation changes. Three LACE profiles were used in the Ray et al. (2010) and Engel et al. (2009) studies of age trends, with each of these profiles costing well in excess of $300,000 to obtain. Balloonborne AirCore-based GC measurements will form the basis of a financially feasible and sustainable stratospheric circulation-monitoring program for climate change.

A key aspect of the measurement program is that all the air samples would be analyzed by one laboratory GC in Boulder, Colorado, with calibrations tied directly to the NOAA surface network measurements. This should eliminate some of the discrepancies that can result from different measurement campaigns performed by different laboratories. After each tube is flown and recovered, the sample will be partitioned into individual sealed mini containers with a clean push gas. Each mini container is registered to a sample altitude through pressure and temperature measurements made during flight (Karion et al. 2010). The containers with discrete samples will then be shipped back to Boulder to be analyzed on a GC. The use of mini containers eliminates the need for a separate GC at each measurement location and will allow for measurement consistency at all locations. In this proposed program, the above-mentioned techniques are an extrapolation of tested flask, existing AirCore measurements, and instrument analysis done in the laboratory. These results indicate that the described technique is feasible. A complete integrated working version has not been constructed. If the mini containers compromise the measurements, then the program would require analysis of the samples at each site. This would require multiple instruments and a larger investment, primarily in personnel. Even with this modest increase in cost of the program, it would still be cost effective and technically feasible.

To obtain the circulation diagnostics described above, the primary trace gases targeted by this program will be SF_6, N_2O, CFC-12, CFC-113, CFC-11, and halon-1211, supplemented by existing programs measuring H_2O and CO_2. Additional trace gases specific to polar vortex profiles would be CO and H_2. Profiles would ideally be taken within five different latitudinal regions, the southern and northern mid- and high latitudes and the tropics (Fig. 5). The locations of these regions are defined by the latitudinal barriers that exist in the stratosphere in the subtropics and at the edge of the polar vortices (e.g., Haynes and Shuckburgh 2000). The program would start with samples taken from Boulder, moving to the tropics and Southern Hemisphere after technical and logistic issues have been resolved, verified, and optimized.

Within each region, the stratosphere is relatively well mixed, so that a representative profile at an equivalent latitude should represent the region as a whole for a given season. Past midlatitude profiles in the stratosphere show structure attributed to tropical or high-latitude influences. This complicates obtaining regional representativeness from a single vertical profile. To obtain better sampling statistics and to average out zonal asymmetries, we will fly and analyze 3–6 samples within a given 2–3-week period in each region. These individual profiles are filtered to define a representative profile for each season and location. To resolve the seasonal cycle, these measurements will be repeated at least four times each year.

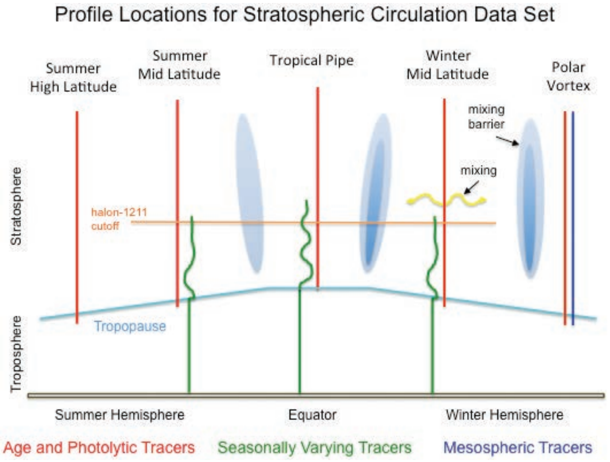

Fig. 5. Shown are proposed measurement locations with seasonal coverage from regular balloon flights in the tropics, and mid- and high latitudes from both hemispheres. The red lines at each location represent the mean age and photolytic tracers (SF_6, N_2O, CFC-12, CFC-113, CFC-11, and halon-1211), the green lines represent tracers that have seasonal variability in the lower stratosphere (ozone, water vapor, and CO_2), and the blue line represents mesospheric tracers (H_2 and CO). The blue-shaded regions represent latitudinal transport barriers that divide the sampling regions. The yellow wavy arrow represents the latitudinal mixing that takes place mostly within the winter midlatitudes.

Additional measurements of complementary trace gases such as ozone and water vapor from sondes as well as AirCore CO_2 and CH_4 would contribute substantially to the program. Ideally, this measurement program would also be complementary to and help supplement existing measurement programs such as the Network for the Detection of Atmospheric Composition Change.

SUMMARY. The program described here would provide unprecedented low-cost, long-term seasonal profile measurements of trace gases that can be used to monitor transport characteristics of the stratosphere. A dataset of this type would put invaluable constraints on climate models. These constraints would be through direct comparison via predicted tracer values because these tracers are sensitive to both path and time scales of transport in unique and quantifiable ways. In addition, the calculation of transport variables will also be quite beneficial. This dataset would also be valuable for the satellite community, both for individual calibration and validation, and for bridging the gap between different satellite datasets as they go on- and off-line.

The stratosphere is increasingly being recognized as an important piece of the climate system and necessary for a complete understanding of climate change. The stratosphere is also a difficult and expensive place to make measurements, particularly profile measurements of long-lived trace gases. The AirCore technique provides the cost savings to make a long-term measurement program feasible. This program would provide a substantial and critical improvement in the capability to evaluate climate model performance in the stratosphere and potentially help improve model prediction of the entire climate system.

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REFERENCES

Alexander, M. J., and Coauthors, 2010: Recent developments in gravity-wave effects in climate models and the global distribution of gravity-wave momentum flux from observations and models. *Quart. J. Roy. Meteor. Soc.,* **136,** 1103–1124, doi:10.1002/qj.637.

Andrews, A. E., and Coauthors, 2001: Mean ages of stratospheric air derived from in situ observations of CO_2, CH_4, and N_2O. *J. Geophys. Res.,* **106** (D23), 32 295–32 314.

Austin, J., and F. Li, 2006: On the relationship between the strength of the Brewer-Dobson circulation and the age of stratospheric air. *Geophys. Res. Lett.,* **33,** L17807, doi:10.1029/2006GL026867.

Brewer, A. W., 1949: Evidence for a world circulation provided by the measurements of helium and water vapour distribution in the stratosphere, *Quart. J. Roy. Meteor. Soc.,* **75,** 351–363.

Butchart, N., and E. E. Remsberg, 1986: The area of the stratospheric polar vortex as a diagnostic for tracer transport on an isentropic surface. *J. Atmos. Sci.,* **43,** 1319–1339.

——, and A. A. Scaife, 2001: Removal of chlorofluorocarbons by increased mass exchange between the stratosphere and troposphere in a changing climate. *Nature,* **410,** 799–802.

——, and Coauthors, 2010: Chemistry–climate model simulations of twenty-first century stratospheric climate and circulation changes. *J. Climate,* **23,** 5349–5374.

Dall'Amico, M., P. A. Stott, A. A. Scaife, L. J. Gray, K. H. Rosenlof, and A. Yu. Karpechko, 2010: Impact of stratospheric variability on tropospheric climate change. *Climate Dyn.,* **34,** 399–417.

Dobson, G., 1956: Origin and distribution of the polyatomic molecules in the atmosphere. *Proc. Roy. Soc. London,* **236,** 187–193.

Engel, A., and Coauthors, 2009: Age of stratospheric air unchanged within uncertainties over the past 30 years. *Nat. Geosci.,* **2,** 28–31.

Forster, P. M. de F., and K. P. Shine, 2002: Assessing the climate impact of trends in stratospheric water vapor. *Geophys. Res. Lett.,* **29,** 1086, doi:10.1029/2001GL013909.

Garcia, R. R., and W. J. Randel, 2008: Acceleration of the Brewer–Dobson circulation due to increases in greenhouse gases. *J. Atmos. Sci.,* **65,** 2731–2739, doi:10.1175/2008JAS2712.1.

——, ——, and D. E. Kinnison, 2011: On the determination of age of air trends from atmospheric trace species. *J. Atmos. Sci.,* **68,** 139–154.

Gerber, E. P., and Coauthors, 2012: Assessing and understanding the impact of stratospheric dynamics and variability on the Earth system. *Bull. Amer. Meteor. Soc.,* **93,** 845–859.

Hall, B. D., and Coauthors, 2011: Improving measurements of SF_6 for the study of atmospheric transport and emissions. *Atmos. Meas. Tech.,* **4,** 2441–2451, doi:10.5194/amt-4-2441-2011.

Hall, T. M., 2000: Path histories and timescales in stratospheric transport: Analysis of an idealized model. *J. Geophys. Res.,* **105** (D18), 22 811–22 823.

Harnisch, J., R. Borchers, P. Fabian, and M. Maiss, 1996: Tropospheric trends for CF_4 and C_2F_6 since 1982 derived from SF_6 dated stratospheric air. *Geophys. Res. Lett.,* **23,** 1099–1102.

Haynes, P. H., and E. Shuckburgh, 2000: Effective diffusivity as a diagnostic of atmospheric transport: 1. Stratosphere. *J. Geophys. Res.,* **105** (D18), 22 777–22 794.

——, C. J. Marks, M. E. McIntyre, T. G. Shepherd, and K. P. Shine, 1991: On the "downward control" of extratropical diabatic circulations by eddy-induced mean zonal forces. *J. Atmos. Sci.,* **48,** 651–678.

Karion, A., C. Sweeney, P. Tans, and T. Newberger, 2010: AirCore: An innovative atmospheric sampling system. *J. Atmos. Oceanic Technol.,* **27,** 1839–1853.

Karpechko, A. Y., and E. Manzini, 2012: Stratospheric influence on tropospheric climate change in the Northern Hemisphere. *J. Geophys. Res.,* **117,** D05133, doi:10.1029/2011JD017036.

McLandress, C., and T. J. Shepherd, 2009: Simulated anthropogenic changes in the Brewer–Dobson circulation, including its extension to high latitudes. *J. Climate,* **22,** 1516–1540, doi:10.1175/2008JCLI2679.1.

Moore, F. L., and Coauthors, 2003: Balloonborne in situ gas chromatograph for measurements in the troposphere and stratosphere. *J. Geophys. Res.,* **108,** 8330, doi:10.1029/2001JD000891.

Mote, P. W., and Coauthors, 1996: An atmospheric tape recorder: The imprint of tropical tropopause temperatures on stratospheric water vapor. *J. Geophys. Res.,* **101** (D2), 3989–4006.

Neu, J. L., and R. A. Plumb, 1999: Age of air in a "leaky pipe" model of stratospheric transport. *J. Geophys. Res.,* **104** (D16), 19 243–19 255.

Ploeger, F., and Coauthors, 2012: Horizontal transport affecting trace gas seasonality in the tropical tropopause layer (TTL). *J. Geophys. Res.,* **117,** D09303, doi:10.1029/2011JD017267.

Plumb, R. A., 2002: Stratospheric transport. *J. Meteor. Soc. Japan,* **80,** 793–809.

——, and M. K. W. Ko, 1992: Interrelationships between mixing ratios of long-lived stratospheric constituents. *J. Geophys. Res.,* **97** (D9), 10 145–10 156.

——, D. W. Waugh, and M. P. Chipperfield, 2000: The effects of mixing on tracer relationships in the polar vortex. *J. Geophys. Res.,* **105** (D8), 10 047–10 062.

Portmann, R. W., S. Brown, T. Gierczak, R. Talukdar, J. B. Burkholder, and A. R. Ravishankara, 1999: Role of nitrogen oxides in the stratosphere: A reevaluation based on laboratory studies. *Geophys. Res. Lett.,* **26,** 2387–2390.

Randel, W. J., F. Wu, H. Vömel, G. E. Nedoluha, and P. Forster, 2006: Decreases in stratospheric water vapor after 2001: Links to changes in the tropical tropopause and the Brewer-Dobson circulation. *J. Geophys. Res.,* **111,** D12312, doi:10.1029/2005JD006744.

Ray, E. A., F. L. Moore, J. W. Elkins, D. F. Hurst, P. A. Romashkin, G. S. Dutton, and D. W. Fahey, 2002: Descent and mixing in the 1999–2000 northern polar vortex inferred from in situ tracer measurements. *J. Geophys. Res.,* **107**, 8285, doi:10.1029/2001JD000961.

——, and Coauthors, 2010: Evidence for changes in stratospheric transport and mixing over the past three decades based on multiple data sets and tropical leaky pipe analysis. *J. Geophys. Res.,* **115,** D21304, doi:10.1029/2010JD014206.

Sawa, Y., T. Machida, and H. Matsueda, 2008: Seasonal variations of CO_2 near the tropopause observed by commercial aircraft. *J. Geophys. Res.,* **113,** D23301, doi:10.1029/2008JD010568.

Scaife, A. A., and Coauthors, 2012: Climate change and stratosphere–troposphere interaction. *Climate Dyn.,* **38,** 2089–2097, doi:10.1007/s00382-011-1080-7.

Shepherd, T., and C. McLandress, 2011: A robust mechanism for strengthening of the Brewer–Dobson circulation in response to climate change: Critical-layer control of subtropical wave breaking. *J. Atmos. Sci.,* **68,** 784–797.

Solomon, S., K. H. Rosenlof, R. W. Portmann, J. S. Daniel, S. M. Davis, T. J. Sanford, and G.-K. Plattner, 2010: Contributions of stratospheric water vapor to decadal changes in the rate of global warming. *Science,* **327,** 1219–1223.

Stiller, G. P., and Coauthors, 2012: Observed temporal evolution of global mean age of stratospheric air for the 2002 to 2010 period. *Atmos. Chem. Phys.,* **12,** 3311–3331, doi:10.5194/acp-12-3311-2012.

Tans, P. P., 2006: System and method for providing vertical profile measurements of atmospheric gases. U.S. Patent 7,597,014, filed 15 August 2006, and issued 6 October 2009.

Volk, C. M., and Coauthors, 1996: Quantifying transport between the tropical and mid-latitude lower stratosphere. *Science,* **272,** 1763–1768, doi:10.1126/science.272.5269.1763.

Waugh, D. W., and T. M. Hall, 2002: Age of stratospheric air: Theory, observations, and models. *Rev. Geophys.,* **40**, 1010, doi:10.1029/2000RG000101.