Final Report

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Scaling from Flux Towers to Ecosystem Models: Regional Constraints on Carbon Cycle Processes from Atmospheric Carbonyl Sulfide

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Abstract

DOE supported research suggests that gross primary productivity (GPP) is largely underestimated by global earth system models [Welp et al., 2011], reflecting the persistent challenge in extrapolating from local-scale GPP observations to global-scale earth system models. This poor understanding of GPP at large spatial scales is of particular concern in tropical forests. In tropical forests, some earth systems models forecast a powerful feedback between a warming climate and a decline in GPP resulting in forest dieback. While this simulated feedback is intensely debated, we lack robust large-scale constraints on GPP that are needed to resolve this debate. In particular, carbon dioxide measurements provide valuable information on net carbon flux, but not on the gross flux associated with GPP. Here we conducted a study of regional-to-global scale GPP using atmospheric carbonyl sulfide to provide a new constraint on GPP mechanisms in earth system models. Our project activities integrated modeling, in situ measurement, and remote sensing techniques to resolve GPP for the Amazon as well as global scale trends. The results of this work included initiating airborne carbonyl sulfide monitoring in the Amazon, training for postdocs and graduate students at a Hispanic Serving Institution, fundamental advances in carbonyl sulfide budgets [e.g. Hilton et al., *Nature Climate Change*, 2017], and high-profile publications that focused on GPP trends for the Amazon [Stinecipher et al., *GRL*, 2022] and global historical GPP trends [Campbell, et al., *Nature*, 2017]. Based on the suggestion of our DOE program manager, we published a state-of-the-science commentary to the scientific community on GPP monitoring with COS [Campbell et al., *EOS*, 2017] which was selected as the cover story. DOE support was acknowledged in all reports. The importance of this research to understanding climate change was communicated to the general public through community seminars (Rotary, Public Libraries, State Parks), an op-ed (SF Chronicle), and interviews in the mass media including two stories in the New York Times (4/5/17; 7/30/18), one of which was especially widely read after it was featured in the New York Time's Quote of the Day.

1 Introduction

A central barrier preventing the scientific community from developing predictive capabilities for carbon-climate feedbacks is methodological; it is challenging to bridge the gap in spatial scales from the detailed process-level understanding obtained from local field measurements and the large-scale earth system models where the understanding of these processes is implemented. This methodological challenge has resulted in large uncertainties in understanding the two primary processes that underlie net ecosystem carbon fluxes: gross primary production (GPP) and respiration [*Beer et al.*, 2010; *Welp et al.*, 2011]. An important finding from the North American Carbon Program (NACP) interim synthesis effort is that ecosystem models disagree strongly about the magnitude of regional GPP fluxes [*Huntzinger et al.*, 2012]. Figure 1 shows GPP from six terrestrial biosphere models integrated across the temperate North American domain. Summertime GPP differs among these estimates by a factor of two. These same models largely agree on the magnitude of net ecosystem exchange (NEE, not shown). Since NEE is the imbalance between GPP and respiration fluxes, the models must also have large disagreements about ecosystem respiration. Similar discrepancies are found in the Amazon [*Gloor et al.*, 2012]. Given this poor diagnostic capability it is not surprising that predictive models of GPP out to the year 2100 vary widely with respect to magnitude, inter-annual variability, and long-term trends [*Friedlingstein et al.*, 2006].

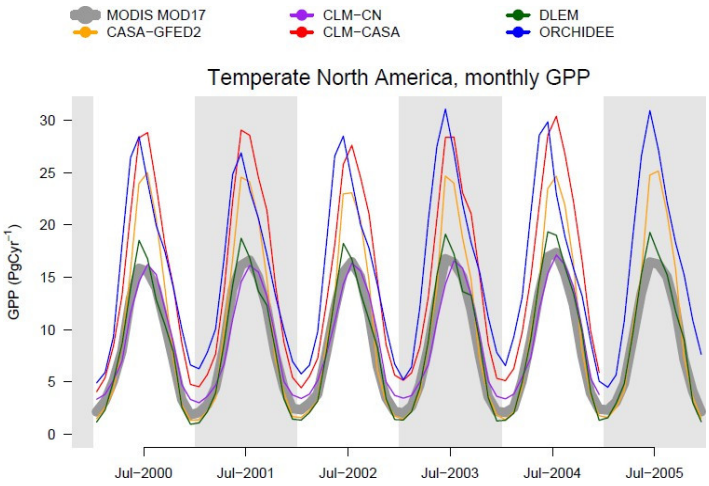

Figure 1. Gross primary production (GPP) estimates from NACP interim synthesis terrestrial biosphere models for temperate North America over the period 2000-2005 (Courtesy of Andy Jacobson at NOAA ESRL).

In order to develop predictive capacity for ecosystem responses to global change, the process fluxes underlying NEE must be better understood. Thus, observational methodologies for evaluating these process fluxes are needed. GPP and respiration flux estimates have generally been made at small spatial scales using eddy flux techniques [e.g. *Ciais et al.*, 2005; *Falge et al.*, 2002]. Robust techniques are urgently needed for extending these local eddy flux measurements to spatial scales that are commensurate with the large scales (>10^4 km^2) of global earth systems models used to forecast climate-carbon feedbacks.

Atmospheric analysis suggests the potential of carbonyl sulfide (COS) measurements to be used as a large-scale tracer of GPP and simultaneous COS and CO_2 measurements to be used to obtain

additional information on carbon cycle processes [*Campbell et al.*, 2008; *Montzka et al.*, 2007]. The relationship between COS and CO_2 fluxes across different plant types and environmental conditions is due to the simultaneous uptake of COS and CO_2 by photosynthesis enzymes in terrestrial plants [*Billesbach et al.*, In Press; *Sandoval-Soto et al.*, 2005; *Stimler et al.*, 2011]. While the gross plant uptakes of these two gases are related, the net ecosystem fluxes are distinct. The net CO_2 flux is the result of the sum of off-setting component fluxes - the GPP sink and the respiration source, while the net COS surface flux is dominated by the plant uptake sink. While potentially confounding continental fluxes require continued investigation (e.g. soils, biomass burning, anthropogenic activities, nocturnal conductance), field observations suggest that they are small relative to COS plant uptake in most upland terrestrial ecosystems and the primary source is physically remote, located over the oceans. One field site exhibited significant soil fluxes at times [*Seibt et al.*, In Preparation] but field investigations and atmospheric studies are broadly consistent with a small soil flux [*Asaf et al.*, 2013; *Campbell et al.*, 2008; *Kettle et al.*, 2002; *Montzka et al.*, 2007; *Steinbacher et al.*, 2004; *White et al.*, 2010]. It follows that COS measurements could be used as a tracer of GPP and that simultaneous COS and CO_2 measurements could be used to deconvolve the GPP and respiration contributions to net CO_2 exchange.

We have integrated modeling and measurements in recent work to develop the COS tracer approach within a 3-D atmospheric framework, finding this GPP tracer particularly suitable for applications to large-scale analysis [*Berry et al.*, 2013; *Billesbach et al.*, In Press; *Campbell et al.*, 2008]. The atmospheric COS observations from 1741 NASA INTEX-NA samples in the North American growing season (July/August 2004) showed considerable drawdown in the boundary layer (difference between 6-8 km and 0-2 km altitudes) of 59.9 ± 8.9 ppt (mean ± 95% c.i., n = 50 vertical profiles) which was consistent with the NOAA/ESRL drawdown of 55.9 ± 19.0 ppt (mean ± 95% c.i., n = 12 airborne sites, July and August of 2005 through 2007). This is a 13% drawdown relative to the background (CO_2 had a 2% drawdown). Furthermore, the regional model results show that this drawdown was driven by the photosynthesis-linked uptake rather than other continental and oceanic fluxes or boundary condition variability (Figure 2). Anthropogenic COS emissions included in the model (direct and indirect) were concentrated in the eastern U.S. but resulted in a boundary layer enhancement that was less than one third of the vegetative drawdown. While the COS ocean flux is a large source globally, simulations showed only a small marine influence on the vertical profile over the continent during the growing season. The COS soil uptake during the growing season was also small relative to the plant uptake which was consistent with field observations [*Asaf et al.*, 2013; *Steinbacher et al.*, 2004; *White et al.*, 2010]. These results provide quantitative evidence that COS measurements may have broad use as a measurement-based, large-scale GPP tracer.

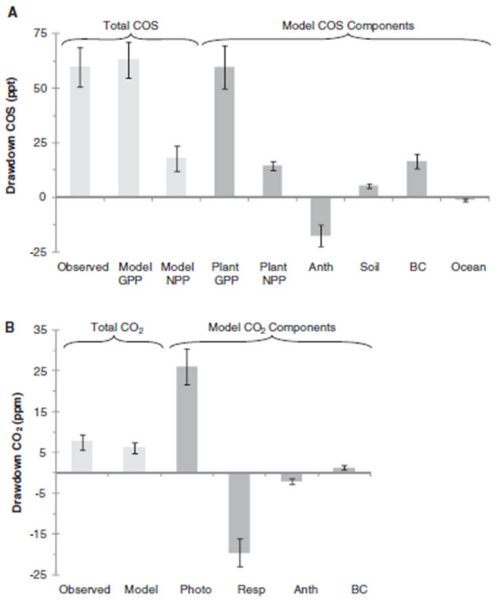

Figure 2. *Tropospheric drawdown for observed and modeled concentrations along continental NASA INTEX-NA DC8 flight paths (July/August 2004). Tropospheric drawdown of COS (A) and CO_2 (B), for total (left brace) and modeled components (right brace) as the difference between mean 6-8 km and 0-2 km altitude asl concentrations for all continental INTEX-NA data (error bars ± 95% c.i., n = 50 vertical profiles, Anth = anthropogenic, Photo = photosynthesis, Resp = ecosystem respiration, BC = boundary condition). Positive drawdown is removal from the atmosphere and negative drawdown is a source to the atmosphere.*

While recent atmospheric COS analysis is focused on North America, a process-level understanding of the carbon cycle is of particular concern for the tropics. There is a scientific debate concerning whether a warming climate and the resulting moisture stress could result in a decline in tropical rainforest GPP and a subsequent dieback of rainforests releasing massive quantities of carbon as predicted by some earth systems models [*Cox et al.*, 2013; *Malhi et al.*, 2009]. Furthermore, changes in GPP in the Amazon are also thought to be driven by the increase in atmospheric CO_2 over pre-industrial concentrations [*Gedney et al.*, 2006; *Hietz et al.*, 2005; *Lloyd and Farquhar*, 1996]. Not only do these GPP processes need to be developed in earth systems models, but we also need to be able to validate these models. Current attempts to validate GPP process models are hampered by the fact that large-scale CO_2 measurements are directly useful for inferring information about NEE but are far more uncertain in quantifying the underlying GPP and respiration processes, particularly at the regional scale.

GPP investigations focused on temperature and hydrologic variability may be particularly well suited for a COS analysis. Variation in the rates of GPP and respiration between ecosystems and within an ecosystem to environmental stress tend to have offsetting effects on net CO2 exchange. For example, drought stress inhibits both GPP and respiration resulting in non-linear responses of net CO2 exchange to drought stress. In contrast, drought would be expected to inhibit leaf COS uptake (because of reduced stomatal conductance), which is the dominant terrestrial flux, resulting in a more or less linear response of COS exchange to drought stress. Similarly, it is difficult to resolve differences between ecosystems in their overall productivity based on net CO2 exchange alone because the differences in GPP and ecosystem respiration tend to cancel. In contrast COS exchange should scale linearly with productivity.

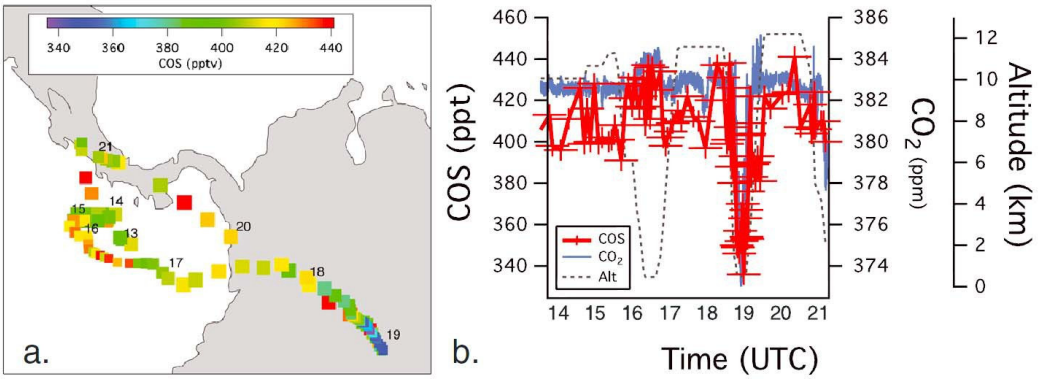

Figure 3. *CO_2 and COS concentration measured in one flight leg (8 August 2007) of the TC^4 campaign. (a) COS concentration along the flight path into the Columbian Amazon. COS is depicted by marker color (warmer color is higher concentration) and flight altitude is depicted by marker size (larger size is lower altitude). (b) Time series of CO_2, COS and altitude for the flight depicted in panel a. Low altitude passes include collection in the marine boundary layer (UTC 16 h) and the continental boundary layer (UTC 19 h).*

While a COS tracer analysis has not previously been undertaken for the tropics, preliminary results suggest promise for such an application [*Berry et al.*, 2013]. Airborne measurements from the NASA TC^4 campaign measured a simultaneous depletion of COS and CO_2 in the boundary layer over the Columbian Amazon that is consistent with simulations of COS and CO_2 (Figure 3). Furthermore, an atmospheric-ecosystem simulation experiment was used to show that atmospheric COS data could provide valuable information on the separate responses of photosynthesis and respiration to environmental forcing. Two global simulations were conducted, each with a different implementation of soil hydrology and water stress (Figure 4). The original simulation was known from comparisons with ecosystem scale measurements at an eddy correlation tower in the Eastern Amazon near Santarem to over-estimate drought stress. In the original simulation, the canopy developed severe water stress near the end of the dry season, yet this was not seen in the eddy correlation studies conducted in the forest. To correct this problem, the soil was made deeper, and root-mediated redistribution of soil water was implemented in a second simulation [*Baker et al.*, 2008]. This modification reduced the simulated inhibition of photosynthesis and enhanced COS uptake by eliminating soil water stress. The two different model implementations were run globally, including atmospheric transport. These simulations provide examples of the differential responses of photosynthesis and respiration that have interpretable manifestations in the comparative drawdown of CO_2 and COS. Clearly, COS data could provide process level insights on GPP, additional to those we could distinguish from only looking at the CO_2 alone.

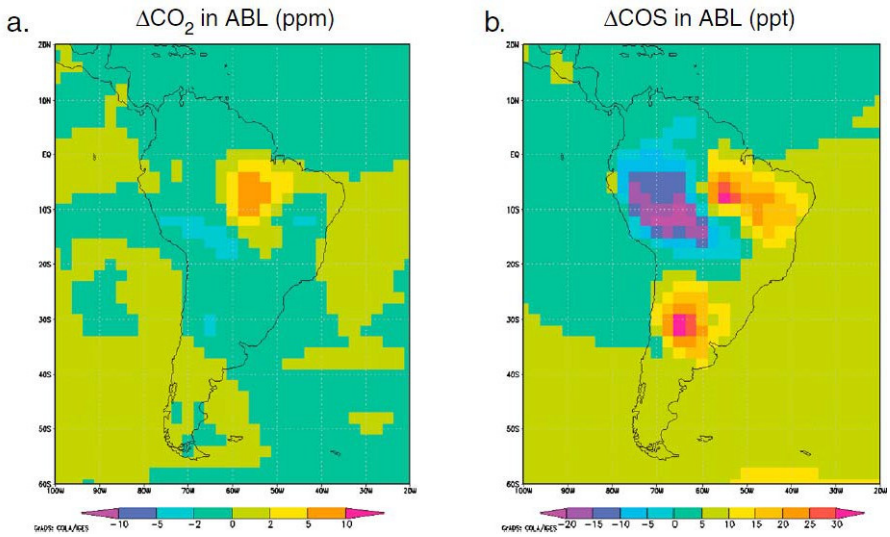

Figure 4. Maps illustrating the use of COS as a diagnostic between different simulated soil hydrology implementations. Maps show the difference in ABL concentration for CO_2 and COS for different parameterizations (new-old) over South America in January 2005. Positive value indicates increased drawdown indicative of increased net uptake in the new parameterization. An area of enhanced CO_2 drawdown (5–10 ppm) in the ABL is seen over the Eastern Amazon, with corresponding enhanced COS drawdown (20–40 ppt) over the same area. Another area of COS drawdown is seen over an area south of the Amazon, but with no corresponding area of enhanced CO_2 drawdown.

2. Results

Given the critical need for large-scale, process-level information in the tropics and the preliminary evidence for a strong COS-GPP relationship in this region, we propose the first integrated measurement and modeling study of GPP. This project included activities between terrestrial ecology, atmospheric science, and oceanography to assess GPP in the Amazon to global scales using in situ and remote sensing observations. The following sections describe the core results of this work. The PhD students and postdocs who were trained on this projected are noted through the underlined authors in the references.

2.1 Carbonyl Sulfide Budgets

In order to apply atmospheric COS to estimate GPP, the large-scale sources and sinks of COS must be well quantified. We advanced the understanding of these sources and sinks with a focus on anthropogenic and biomass burning inventories as well as the relationship between plant uptake of CO_2 and plant uptake of COS.

2.1.1 Anthropogenic

We developed a global COS anthropogenic inventory for the years 1850 to 2013 based on new emission measurements and material-specific data [*Campbell et al., 2015*]. Previous results from a recent regional inventory suggest that the anthropogenic source is similar in magnitude to the plant sink, confounding carbon cycle applications. However, our material-specific approach revealed that the current anthropogenic source is only one third of plant uptake and is concentrated in Asia, supporting GPP-estimation applications of COS.

We extended our global anthropogenic estimates to a gridded emissions inventory with a monthly, 1-degree resolution for the years 1980 through 2012 [*Zumkehr et al., 2018*]. This inventory accounted for recent emissions factor reports of previously overlooked emissions sectors including domestic coal burning. We estimate a global source in year 2012 of 406 Gg S y^{-1} (range of 223–586 Gg S y^{-1}), which is highly concentrated in China and is twice as large as the previous gridded inventory. Our large upward revision in the bottom-up estimate of the source is consistent with a recent top-down estimate based on air-monitoring and Antarctic firn data *[Campbell et al. 2017a]*. Furthermore, our inventory time trends, including a decline in the 1990's and growth after the year 2000, are qualitatively consistent with trends in atmospheric data. Finally, similarities between the spatial distribution in this inventory and remote sensing data suggest that the anthropogenic source could potentially play a role in explaining a missing source in the global COS budget. These emissions estimates are now widely applied in other atmospheric studies [e.g. *Hannigan et al., 2022; Ma et al., 2021*].

Given the availability of higher-resolution driver data over the U.S., we refined the gridded inventory [*Zumkehr et al., 2017*]. We updated emission factor data and industry activity data to develop a gridded inventory with a 0.1° spatial resolution for the U.S. domain. The inventory includes the primary anthropogenic COS sources including direct emissions from the coal and aluminum industries as well as indirect sources from industrial carbon disulfide emissions. Compared to the previously published inventory estimates, we found that the total anthropogenic source (direct and indirect) is 47% smaller. Using this new gridded inventory to drive the Sulfur Transport and Deposition Model/Weather Research and Forecasting chemical atmospheric transport model, we found that the anthropogenic contribution to COS variation in the troposphere is small relative to the biosphere influence, which is encouraging for carbon cycle applications in this region.

2.1.2 Biomass Burning

We developed a global gridded inventory of COS biomass burning and evaluated this inventory using atmospheric modeling constrained by air-monitoring data [*Stinecipher et al., 2019*]. COS emissions due to biomass burning are a variable and substantial (over 10%) part of the current COS budget. COS emission ratios from open burning fires, coupled with 1997–2016 data from the Global Fire Emissions Database (GFED4), yield COS biomass burning emissions with a global average annual flux of 60 ± 37 Gg(S) $year^{-1}$. A global box model suggests these emissions are more consistent with observations from global atmospheric composition monitoring networks than fluxes derived from previous synthesis papers. Even after considering the uncertainty in emission factor observations for each category of emissions and the interannual variation in total burned dry matter, the total COS emissions from open burning are insufficient to account for the large imbalance between current estimates of global COS sources and sinks.

2.1.3 Plant Flux

Another important prerequisite for applying COS as a GPP tracer is to understand the relationship between GPP and the plant uptake of COS. We completed a field experiment to provide a proof-of-concept of the measurement apparatus that we exported to Brazil for this DOE project which included an automated flask sampling system and interface for the quantum cascade laser spectrometer system [*Campbell et al., 2015*]. Flask samples were collected in a redwood forest at ground level as well as within and above the canopy. These results provided strong evidence of a close relationship between

COS plant uptake, CO_2 uptake for photosynthesis, and stomatal conductance. The first line of evidence is that the daytime COS to CO_2 ratio in the redwoods is also observed in continental and hemispheric observations. The second line of evidence was the longitudinal drawdown of these gases as marine air moved across the forest was in close agreement with high-resolution atmospheric transport simulations that were driven by ecosystem models that incorporated COS-to-CO_2 flux relationships.

2.2 Remote Sensing

We applied the source and sink project results described above to examine global trends in atmospheric COS from remote sensing. MIPAS satellite retrievals of the upper troposphere revealed remarkable drawdown of COS over the Amazon which is consistent with a large plant sink and vigorous convection (Glatthor et al., 2015). Mid-troposphere retrievals from TES reveal evidence of outflow of the Asian anthropogenic source (Kuai et al., 2014), which is consistent with revised inventories (Campbell et al., 2015).

2.3 Amazon GPP Estimation

Given the large COS depression over the Amazon revealed in MIPAS data (Figure 5), we applied these data as a benchmark for Amazon GPP estimates from global ecosystem models [Stinecipher et al., 2022]. Understanding the magnitude of tropical gross primary production (GPP) is critical for carbon cycle modeling and climate projections, but this quantity is poorly constrained for the Amazon. We simulated COS concentrations driven by surface flux scenarios encompassing a wide range of GPP estimates for the Amazon basin. We compared the model output to the satellite retrievals and found a regional GPP estimate of 2375 ± 914 g(C) m^{-2} yr^{-1}, consistent with previous estimates, including the TRENDY model ensemble range of 1431–3812 g(C) m^{-2} yr^{-1}.

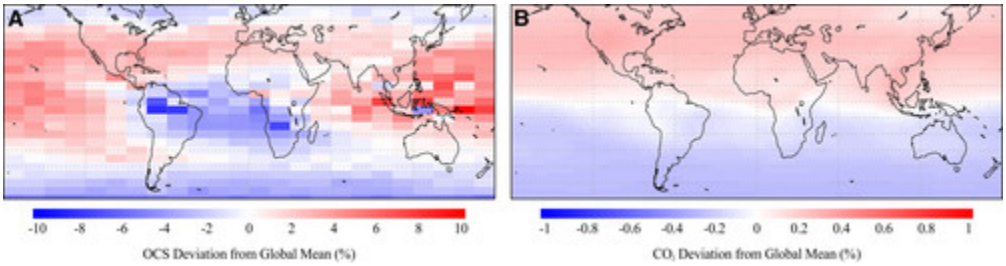

Figure 5. Upper troposphere deviations in (a) COS concentration from MIPAS (average binned observations, 2002–2012) and (b) CO_2 concentration from GOSAT's Level 4B product (averaged 2010–2013). Both plots express changes in concentration as percent deviation from the global annual mean at 250 hPa. Note that the CO_2 signal is an order of magnitude less than that of COS.

2.3 Global GPP Trends Estimation

We extended the Amazon work to the global scale using long-term COS air-monitoring data from atmospheric observatories and firn data [Campbell et al., 2017c]. We interpreted these records using a model that simulates changes in COS concentration according to changes in its sources and sinks—including a large sink that is related to GPP. We find that the observation-based COS record is most

consistent with simulations of climate and the carbon cycle that assume large GPP growth during the twentieth century (31% ± 5% growth; mean ± 95% confidence interval). This GPP growth is at the high end of the range reported from different carbon/climate models, ranging from 5% to 34% over the past century.

2.4 State-of-the-Science Report

Based on the suggestion of our DOE program manager, we published a state-of-the-science report on the COS approach to studying GPP [Campbell et al., 2017b]. The report identified the unique role that COS plays with respect to temporal and spatial constraints in relation to other diagnostics including atmospheric CO_2 measurements and SIF observations. This report was selected as the cover story for a widely read scientific newsletter (*EOS*) and helped to disseminate information on this emerging approach to understanding terrestrial ecosystems.

3.0 Science Communication

We shared the results of this work with the general public through community seminars hosted by local institutions (Santa Cruz Rotary, Santa Cruz Public Library, Point Lobos State Park) while also reading out to a larger audience through an op-ed in the San Francisco Chronicle (7/10/18), and interviews in the mass media including two profiles of this research covered in articles in the New York Times (4/5/17; 7/30/18). One of our quotations was selected by the editor for the New York Times Quote of the Day which significantly enhanced the reach of this science communication piece.

4.0 Future Work

A key prediction is that the COS seasonal amplitude in the northern hemisphere should provide a signature of photosynthesis. In future work we will examine long-term trends in the COS seasonal amplitude across multiple observatories (continental, marine, boundary layer, free troposphere) and atmospheric sensing platforms (flask, FTIR, and satellite). Our preliminary results show that the seasonal amplitude trends coincide with long-term trends in mean annual concentrations, but change at five times the rate. The observations predict a compensation point of over 300 ppm at which the amplitude extrapolates to zero. We will use global chemical transport simulations to interpret these trends and their relationship to plant uptake and other sources and sinks.

We will also continue to engage with our Brazilian collaborators from this project who are using our quantum cascade laser spectrometer to analyze flask samples collected from their airborne observing network. While our MIPAS remote sensing analysis provided a top-down constraint on Amazon GPP, we expect the near-surface airborne data to open a new window into GPP analysis by revealing detailed seasonal and regional pattern in GPP that were not preserved in the high-altitude MIPAS data.

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