Understanding the Co-Evolution of Air Quality and Climate Forcing in a Rapidly Changing World

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What are the key challenges or questions for Earth System Science across the spectrum of basic research, applied research, applications, and/or operations in the coming decade?

One of the greatest challenges facing society is the need to develop targeted, optimal strategies for both improving air quality and reducing anthropogenic global warming. The well-mixed greenhouse gases CO_2 and CH_4 and short-lived air quality species ozone and aerosols together constitute the largest components of radiative forcing, and they are coupled through common anthropogenic, biogenic, and chemical processes [*Arneth et al*, 2010]. Development of such mitigation strategies therefore requires an integrated approach to accurately quantifying emissions and relating them to atmospheric concentrations and radiative forcing.

Quantifying CO_2 and CH_4 emissions is a national [*Pacala et al.,* 2010] and international [*IPCC*, 2007] priority. Fossil fuel emissions of CO_2 ($FFCO_2$), 45% of which are produced by the US and China, are the largest net flux in the current global carbon cycle and their uncertainty is projected to surpass the magnitude of land emissions (11.0 PgC^{-1}) by 2020 [*World Energy Outlook*, 2010]. Additional large uncertainties in carbon storage and exchange between the atmosphere and terrestrial biosphere prevent closure of the carbon budget at all scales. CH_4 emissions are even less well-known, with the best national inventories reporting uncertainties from individual sectors of ± (20–50) % [*US EPA*, 2012]. CH_4 plays a critical role in both short and long-term climate warming [*Shindell et al*, 2012], and changes in its atmospheric burden over the past decade, which appear to be driven by changes in tropical emissions [*Kirschke et al.,* 2013], are not well-understood.

The emissions and atmospheric burdens of long- and short-lived climate forcers are inextricably linked. In the US, a dramatic shift in energy dependence from coal (with large CO_2, NO_x, and SO_2 emissions) to natural gas over the past decade-and-a-half has improved air quality [*de Gouw et al.,* 2014] but driven increases in CH_4 [e.g. *Lu et al,* 2012] as well as in volatile organic compounds (VOCs) released during natural gas extraction. Air quality regulations have reduced CO and NO_x emissions, abating tropospheric ozone pollution and attenuating its climate forcing, but driving increases in CH_4 lifetime and radiative forcing. At the same time, background ozone has increased due to transport of ozone and its precursors from Eastern China [*Verstraeten et al.*, 2015; *Huang et al.,* 2015].

Rapid population growth and industrialization in China combined with dependence on coal as an energy source has driven rapid increases not only in CO_2, but also NO_x and SO_2, with disastrous consequences for air quality. More than 4000 people die every day in China from particulate matter (PM) exposure [*Rohde and Muller*, 2015]. These rapid changes in Chinese emissions have had complex and uncertain implications for climate forcing from ozone, CH_4, and aerosols due to the dependence of ozone radiative effect on the location of emissions [*Bowman and Henze*, 2012], the opposing responses of CH_4 lifetime

to tropospheric ozone precursors NO_x and CO, and large uncertainties in aerosol direct and indirect effects.

Addressing the challenge of developing strategies for optimizing the co-evolution of air quality and climate forcing therefore requires:

1. Accurate knowledge of emissions of $FFCO_2$, CH_4, and precursors of tropospheric ozone and aerosols
2. Reduced uncertainties in the magnitude and variability of CO_2 land fluxes to allow better closure of the carbon budget
3. Quantification of how changes in precursor emissions affect tropospheric ozone and aerosols and their radiative forcing locally and globally
4. Understanding of how emissions controls targeting ozone affect CH_4 through better understanding of, and constraints on, OH.

Why are these challenges/questions timely to address now especially with respect to readiness?

Regulation of air quality and climate gases over next several decades will be critical in terms of protecting human and ecosystem health and limiting anthropogenic global warming over the next century. In the US, we are nearing the point at which further air quality gains require reductions in energy production and consumption. The Chinese response to air quality concerns may commit the country to either a high or low carbon pathway, depending on whether they continue to invest in reducing pollutant emissions from coal-based power plants [*Karplus*, 2015] or shift toward natural gas and renewable energy [*Sheehan et al.,* 2014].

From a technological point of view, the upcoming geostationary Earth orbit (GEO) missions TEMPO (NASA), GEMS (KARI), and Sentinel 4 (ESA) will provide unprecedented new capabilities to assess the evolution of air quality from space [*Bowman*, 2013] and will feed continued advances in inverse modeling and data assimilation that permit attribution of composition changes to spatially explicit emissions [*Streets et al*, 2013] and multi-species assimilation that can provide, for example, constraints on OH (which is critical to understanding and predicting changes in CH_4 lifetime and burden) [*Miyazaki et al.,* 2012]. However, as discussed in the next section, these observations were not designed for quantifying climate forcing, nor do they make key carbon measurements. The GEO-CAPE Atmospheric Science Working Group, a broad cross-section of ~35 scientists from universities and government laboratories participating in pre-formulation studies for the GEO-CAPE air quality mission recommended by the 2007 Decadal Survey [http://geo-cape.larc.nasa.gov/], has endorsed this white paper in recognition of the need for and value of additional measurements to tackle this challenge.

Why are space-based observations fundamental to addressing these challenges/questions?

Climate forcing occurs at the global scale but is initiated from fluxes and emissions at local scales, which are also the main drivers of air quality. Measurements from ground-based networks provide accurate samples of concentrations and fluxes, with limited geographic coverage. Airborne platforms provide frequent sampling that is limited to selected regions or intensive campaigns. Only satellite measurements can capture the daily, seasonal and interannual variability of key emissions and processes at the spatiotemporal scales needed to inform optimized air quality and climate mitigation strategies.

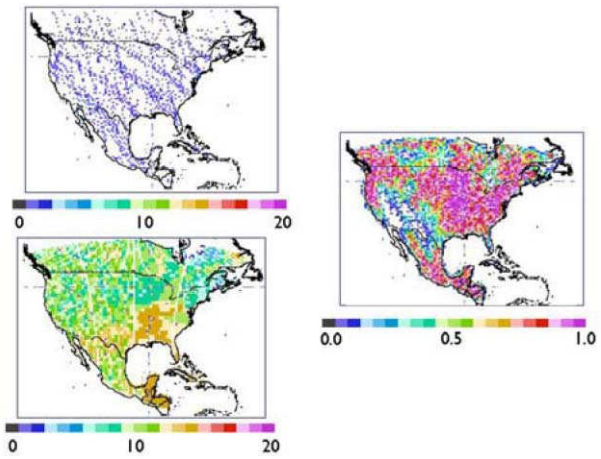

Figure 1. OSSE simulations of weekly clear-sky CO_2 sampling frequency for a combined GOSAT+OCO-2+OCO-3 constellation (top left) and for nominal geostationary measurements (bottom left). The 10x more frequent measurements from GEO provide a 5- to 10-fold reduction in flux errors (1-posterior error/prior error)(right).

The GEO-CAPE pre-formulation studies have shown that monitoring and understanding air quality requires multiple observations per day, at horizontal resolutions of ~10 km or finer and with as much near-surface sensitivity as possible, in order to separate emissions sources at the urban scale, understand diurnal and day-to-day variability in emissions and chemical processing, and provide sufficient measurement statistics to enable quantitative studies in cloudy regions [e.g. *Fishman et al.*, 2012]. Likewise, multiple measurements of CO_2 and CH_4 per day at high spatial resolution are needed to constrain fluxes (Figure 1 and Bousserez et al. [2015]) and map emissions, including those with large diurnal variability such as CH_4 "super-emitters", at policy-relevant scales and uncertainty levels. For this reason, a recent European Commission report on monitoring of $FFCO_2$ emissions recommends geostationary CO_2 measurements [*EC Report*, 2015].

Sentinel 4, TEMPO, and GEMS will measure multiple air quality species at <8km x 8km spatial resolution multiple times per day over Europe, the US, and part of East Asia. Sentinel 4, however, is the only mission that will have concurrent measurements of CO, a key ozone precursor and tracer of anthropogenic activity that provides constraints on OH (which couples CH_4 and O_3), and thermal infrared (TIR) ozone, which can quantify free tropospheric ozone and its radiative effect. None of the geostationary missions will measure the key climate forcers CH_4 and CO_2.

JPSS, the MetOP series, and Sentinel 5/5p will provide global LEO coverage of many of the relevant air quality species. However, these measurements lack the spatiotemporal resolution needed to sufficiently understand and quantify the link between emissions and atmospheric concentrations at the scales needed to optimize emissions regulations. Furthermore, many of these missions are funded to provide a routine set of data to weather forecasting communities and the simplified, fast retrieval approaches needed for that purpose are not well-suited to the quantitative analysis needed for climate forcing. There is no current pathway to producing optimally-retrieved products operationally. Other ongoing and planned missions will provide CO_2 columns from LEO orbit. These measurements are well-suited for studying the global carbon cycle in an aggregated sense (near-continental spatial scales and monthly periods), but again lack the spatiotemporal resolution and sampling density needed to constrain fluxes and reduce emissions uncertainties at the required level.

To support national priorities, there is therefore a need for a TEMPO follow-on mission to provide geostationary measurements of not only air quality species, but also TIR ozone, CO, CH_4, and CO_2 (see also white paper by D. Jacob et al. on "Air Quality and Climate Forcing over North America in the Next Decade"). Measuring these species concurrently provides several advantages. For ozone, combining TIR measurements, which have enabled constraints on ozone radiative forcing [*Bowman et al.,* 2013] and are being used to benchmark model radiative transfer codes, with those in the UV/VIS provides

robust sensitivity to near-surface ozone [*Natraj et al.,* 2011; *Zoogman et al.*, 2011], which is critical for understanding effects on human health and ecosystems. Concurrent measurement of CO_2 and CH_4 significantly improves detection of anthropogenically-enhanced CH_4 concentrations [*Gerilowski et al.*, 2011] and provides a greater reduction in uncertainty in CH_4 emissions than CH_4 measurements alone [*Wong et al.,* 2015]. Given that both CO_2 and CH_4 have significant natural sources, measuring them in combination with CO and NO_2 provides much-improved ability to identify fossil fuel-based emissions [*Newman et al.,* 2013; *Lindenmaier et al.,* 2014].

These measurements would benefit the air quality and carbon cycle communities and would support the Congressionally-mandated Carbon Monitoring System goals of informing carbon policy and management. They would provide a foundation for decision-making aimed at optimizing emissions controls to improve air quality and prevent dangerous warming of the climate system. Extension of a US-based mission's spatial coverage to include South America would allow much-improved constraints on tropical wetland CH_4 emissions and climate forcing associated with biomass burning, while additional GEO or LEO satellites providing the required spatiotemporal resolution over the rest of the globe would serve as an integrated, global air quality / climate forcing monitoring system covering regions with the most rapidly growing emissions (see also companion white paper on "The Future of Air Quality in the Developing World").

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