Quantifying sources and sinks of trace gases using space-borne measurements: current and future science

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We have been observing the Earth’s upper atmosphere from space for several decades, but only over the past decade has the necessary technology begun to match our desire to observe surface air pollutants and climate-relevant trace gases in the lower troposphere, where we live and breathe. A new generation of Earth-observing satellites, capable of probing the lower troposphere, are already orbiting hundreds of kilometres above the Earth’s surface with several more ready for launch or in the planning stages. Consequently, this is one of the most exciting times for the Earth system scientists who study the countless current-day physical, chemical and biological interactions between the Earth’s land, ocean and atmosphere. First, I briefly review the theory behind measuring the atmosphere from space, and how these data can be used to infer surface sources and sinks of trace gases. I then present some of the science highlights associated with these data and how they can be used to improve fundamental understanding of the Earth’s climate system. I conclude the paper by discussing the future role of satellite measurements of tropospheric trace gases in mitigating surface air pollution and carbon trading.

Keywords: global troposphere; air pollution; greenhouse gases; satellite remote sensing; source-sink estimation

1. Introduction

Quantitatively understanding the physical, chemical and biological processes that determine contemporary climate is a prerequisite for developing confident projections of how the Earth’s climate system will evolve on decadal to centennial time scales (IPCC 2007). The climate community thus far has relied largely on (i) detailed, small-scale in situ measurements (e.g. small land plots in the Amazon that monitor carbon uptake from vegetation), (ii) large-scale space-borne measurements that provide information on variables that embody a changing climate state (e.g. atmospheric temperature, ice sheet thickness) and (iii) mathematical models of the Earth’s climate, which attempt to link different components of the Earth (e.g. land, ocean, atmosphere). Despite many successes in understanding the broad-scale nature of the Earth’s climate using the
aforementioned data and mathematical models, the tropospheric chemistry community has, until recently, been starved of satellite measurements, instead relying on sparse ground-based and aircraft measurements of trace gases. There has been a reversal of fortunes, and here I argue that the wide range of current measurements of tropospheric trace gases and surface properties from space-borne instrumentation, and their associated temporal and spatial distributions, represent a rich resource for testing understanding of couplings between land, ocean and atmospheres, and for guiding subsequent in situ measurement campaigns. Relating these satellite observations to specific climate processes is challenging, often demanding an appreciation of physics, chemistry and biology, disciplines that have traditionally been taught separately. Satellite observations of trace gases provide an ability to sample large spatial scales on hourly time scales, as discussed below, which complement the high temporal or spatial resolution distributions provided by in situ instruments. Consequently, measurement campaigns are already integrating space-based and in situ datastreams in an attempt to relate trace gas distribution over different spatial and temporal scales (Jacob et al. 2003). I shall focus my research on satellite observations of trace gases in the lower troposphere (less than 6 km).

The general problem of relating trace gas concentration measurements to surface processes is straightforward, and in this paper, I shall consider the mass balance of a generic trace gas \( C \) at time \( t \) in a frame of reference fixed on the Earth,

\[
\frac{dC(t)}{dt} = P(t) - L(t) + T(t) + X(t). \tag{1.1}
\]

The term on the left-hand side describes how \( C \) varies with time at a particular point in the reference frame. The terms on the right-hand side describe surface production \( (P) \) and loss \( (L) \), atmospheric transport \( (T) \) and additional processes \( (X) \), e.g. atmospheric chemistry, that determine the temporal changes in measurements of \( C \), all of which are likely to be functions of time. I will refer back to this mass balance equation throughout the paper. To predict how \( C \) will change with time, we have to know the functional forms of \( P, L, T \) and \( X \); better quantitative understanding of these functions represents one of the key challenges in predicting future changes in climate. For example, we have only a limited biological understanding of \( P \) and \( L \) for CO\(_2\); without better quantitative understanding of the underlying processes, we cannot confidently predict how the terrestrial biosphere will respond to future warming.

I provide a brief overview of satellite remote sensing of lower tropospheric trace gases in §2 and the estimation of surface sources and sinks of trace gases using satellite measurements in §3. Section 4 summarizes the current science headlines associated with satellite observations of trace gases. I conclude the paper in §5 by exploring the future of atmosphere remote sensing of trace gases.

2. Remote sensing of tropospheric trace gases

Earth observation (EO) satellite instruments that measure atmospheric trace gases generally exploit the spectroscopic properties of atmospheric trace gases and the underlying surface, which are sensitive to atmospheric pressure and the abundance of absorbing gas in the line of sight. Most current EO satellites
Table 1. Nadir sensors that measure trace gases in the lower troposphere (less than 6 km), which can be used for estimating surface sources and sinks. The local equatorial crossing time (LECT) denotes the sunlit portion of the complete orbit.

<table>
<thead>
<tr>
<th>Nadir sensors (launch year)</th>
<th>Orbit (LECT)</th>
<th>Horizontal resolution (vertical degrees of freedom)</th>
<th>Approximate repeat time</th>
<th>Gases measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aqua satellite (2002)</td>
<td>LEO (1330)</td>
<td>13.5 km diameter circle (1 d.f.)</td>
<td>Daily</td>
<td>CO, CO₂, O₃, CH₄</td>
</tr>
<tr>
<td>AIRS</td>
<td></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Aura satellite (2003)</td>
<td>LEO (1338)</td>
<td>13×24 km² (column)</td>
<td>Daily</td>
<td>O₃, HCHO, NO₂, SO₂</td>
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<tr>
<td>OMI</td>
<td></td>
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<tr>
<td>TES</td>
<td></td>
<td>5×8 km² (1–2 d.f.)</td>
<td>6 days</td>
<td>O₃, CH₄</td>
</tr>
<tr>
<td>Envisat satellite (2002)</td>
<td>LEO (1000)</td>
<td>30×60 km² (column)</td>
<td>6 days</td>
<td>O₃, HCHO, NO₂, SO₂</td>
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<tr>
<td>SCIAMACHY</td>
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<tr>
<td>MetOp satellite (2007)</td>
<td>LEO (0930)</td>
<td>12 km diameter circle (1–3 d.f.)</td>
<td>12 hours</td>
<td>O₃, CO, CH₄, BrO, SO₂</td>
</tr>
<tr>
<td>IASI</td>
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<tr>
<td>GOME-2</td>
<td></td>
<td>40×80 km² (column)</td>
<td>Daily</td>
<td>O₃, HCHO, NO₂, BrO, SO₂</td>
</tr>
<tr>
<td>Terra satellite (1999)</td>
<td>LEO (1030)</td>
<td>22×22 km² (1–2 d.f.)</td>
<td>3 days</td>
<td>CO</td>
</tr>
<tr>
<td>MOPITT</td>
<td></td>
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<tr>
<td>ERS-2 satellite (1995)</td>
<td>LEO (1030)</td>
<td>40×320 km² (column)</td>
<td>3 days</td>
<td>O₃, HCHO, NO₂, BrO, SO₂</td>
</tr>
<tr>
<td>GOME</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>to be launched</td>
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<td></td>
</tr>
<tr>
<td>OCO (2008)</td>
<td>LEO (c1315)</td>
<td>1.3×2.3 km² (column)</td>
<td>16 days</td>
<td>CO₂</td>
</tr>
<tr>
<td>GOSAT (2008)</td>
<td>LEO (c1300)</td>
<td>10 km diameter circle (1–2 levels)</td>
<td>3 days</td>
<td>CO₂, CH₄</td>
</tr>
<tr>
<td>Sentinel 4/5 (c2018)</td>
<td>GEO (all)</td>
<td>to be determined</td>
<td>n.a.</td>
<td>TBD</td>
</tr>
</tbody>
</table>

Measuring tropospheric composition are in a Sun-synchronous low-Earth orbit (LEO, defined here as between 200 and 1000 km; table 1), in which an observation at any given point on the Earth is taken at the same local solar time over a particular latitude zone. As I discuss in §5, alternative orbits can improve spatial and temporal coverage of the Earth.

The majority of current EO instruments that measure trace gases in the lower troposphere use nadir geometry (table 1), where the line of sight intersects the Earth’s surface. Other measurement approaches (e.g. measuring Rayleigh scattered radiation along the atmosphere limb) have difficulty in measuring...
below the upper troposphere owing to optically thick clouds along the line of sight; nadir measurements also suffer from this problem but their horizontal resolutions are typically small enough that they have a higher probability of measuring (partial) cloud-free scenes. Limb-viewing instruments, such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS; Fischer et al. 2008), the Microwave Limb Sounder (MLS; Waters et al. 2006) and the Atmospheric Chemistry Experiment (ACE; Bernath et al. 2005), occasionally provide valuable information on the distribution of trace gases in the free troposphere, but they do not have sensitivity to the boundary layer. In this paper, I focus on nadir sounders.

Nadir measurements use backscattered solar radiation at ultraviolet/visible (UV/Vis) and short-wave infrared (SWIR) wavelengths, which are sensitive to cloud, aerosol and Rayleigh scattering. The thermal IR (TIR) region is also measured in this geometry using atmospheric absorption and emission phenomena. In general, observations of TIR wavelengths will be most sensitive to the middle and upper troposphere, but will have some sensitivity to the lower troposphere when there is significant thermal contrast between the lower and middle troposphere (Deeter et al. 2007). Shorter wavelengths such as SWIR/Vis/UV will be sensitive to lower altitudes and these are the wavelengths used by most tropospheric sensors. Other factors, such as spectral resolution, play a key role in the ability of an instrument to accurately measure variations in lower tropospheric trace gases that are (i) orders of magnitude optically thinner than gases in the free troposphere (e.g. formaldehyde, HCHO) or (ii) well mixed owing to long atmospheric lifetimes (e.g. CO₂).

There has been a rapid change in the emphasis of atmospheric trace gas measurements from those associated primarily with stratosphere O₃ depletion to those in the troposphere, associated with climate and surface air quality (AQ). This has been driven by a combination of a growing desire to quantitatively understand surface sources and sinks of trace gases and the technological advances that are beginning to match this desire. Difficulties in measuring tropospheric concentrations arise from clouds and aerosols, a large number of ‘interfering’ trace gases with spectral lines that sometimes are close or overlap target gases, weak absorption of target gases (e.g. HCHO), large stratospheric contributions of target gases (e.g. nitrogen dioxide, NO₂) and surface source–sink variations that represent only a small percentage of tropospheric columns of target gases (e.g. CO₂). Improvements in spectroscopy (e.g. Gratien et al. 2007; Toth et al. 2007), radiative transfer modelling (e.g. Natraj & Spurr 2007) and detector technology (increased sensitivity and reliability) have all led to our ability to make more precise and accurate measurements of tropospheric trace gases. These improvements are particularly important for short-lived (lifetimes of less than hours to days) and long-lived (lifetimes of several months to years) trace gases in the troposphere. Short-lived trace gases emitted (in)directly from surface sources typically have low concentrations (ppt to low ppb) and reside in the lower most atmosphere, so observing them from space involves particularly sensitive analysis of the observed spectra. Long-lived trace gases have large background values, and fresh sources and sinks represent only a few per cent of this background so, as I discuss in §4, this has led to precision requirements unprecedented in atmospheric remote sensing.

Carbon monoxide (CO) has an atmospheric lifetime ranging between weeks and months, depending on the abundance of its OH sink, which is long enough that it can be used effectively to track transport of atmospheric pollution.
but short enough that pollution plumes can be distinguished from the global background. It is therefore no coincidence that CO was one of the first molecules to be measured in the troposphere at TIR wavelengths back in 1984 from the Measurement of Air Pollution from Space (MAPS) instrument aboard the NASA Shuttle (Connors et al. 1999). The resulting free tropospheric distributions of CO, although sparse, were tantalizing. MAPS was a forerunner of the Measurement Of Pollution In The Troposphere (MOPITT), discussed in §4.

3. Estimating surface sources and sinks of trace gases from satellite data

Combining measurements and models, accounting for their respective errors, generally provides a better estimate of surface sources and sinks than using either measurements or models alone. The underlying mathematics originates from engineering and is largely developed for studying the Earth’s climate by the physical oceanography and numerical weather prediction (NWP) communities (Kalnay 2006). The application of this mathematics to atmospheric chemistry is relatively recent.

Relating observed variations in trace gas concentrations to the underlying processes that determine the variability generally relies on using a model to describe the processes. I define a forward model $M$, which includes all known processes that determine variability in the observed tracer $C$,

$$ C(t) = M(P_a, L_a, T_a, X_a, ...) + \varepsilon, $$

where $P$, $L$, $T$ and $X$ are defined as before, with subscript ‘a’ denoting our best a priori understanding of the real process. Model error, $\varepsilon$, represents the sum of errors from the a priori, $\varepsilon_a$, and the model physics and chemistry, $\varepsilon_m$. For the purpose of this discussion, I will estimate only surface production $P$. The Jacobian matrix $K$, containing the sensitivity of $C(t)$ to changes in $P$ (i.e. $\partial C/\partial P_a$), is used to relate differences between model and observed concentrations of trace gas $C$ to $P$.

The observation and its associated error $\varepsilon_o$, and the a priori and its error $\varepsilon_a$, represent two pieces of information. I assume that $\varepsilon_o$ and $\varepsilon_a$ are unbiased and that the expected values (the average obtained after making many similar measurements) of the variances of the observation and model error are $\sigma_o^2$ and $\sigma_a^2$. The inverse model $I$ uses the measurements and a priori information to determine the optimal estimate of production, $\hat{P}$: $\hat{P} = I(M(P_a), \varepsilon_a, C, \varepsilon_o)$. There are a number of ways to reach the optimal solution, but I use the maximum a posteriori solution to show that

$$ \hat{P} = \frac{\sigma_a^2}{K\sigma_o^2 + \sigma_a^2} C + \frac{K\sigma_o^2}{K\sigma_o^2 + \sigma_a^2} P_a, $$

which is essentially a linear least-squares fit of model production rate $P_a$ to observed trace gas measurements $C$, weighted by their respective uncertainties. For example, as the relative error on the a priori $\sigma_a^2$ increases (i.e. we have less confidence in the a priori compared with the measurements), $\hat{P}$ is increasingly determined by the first right-hand side term that contains information from the measurements. Of course, this is a simple example, in which I have considered a linear problem with only one measurement and one variable to estimate, but it
captures the essence of an active research problem. It is also worth noting that measurements from satellite instruments generally represent a weighted mean of the real vertical atmospheric profile. The vertical weighting profile, determined by the instrument type, wavelength studied and the geophysical scenario (e.g. surface albedo), should be applied to the forward model $M$ in the inverse model calculation.

As the precision and spatial and temporal resolutions of satellite data improve, and, more importantly, confidence about these data grows in the community, a more fundamental science objective is the estimation of the smaller scale processes that determine the large-scale source and sink distributions. This involves solving for model parameters, such as the temperature sensitivity of production. The method outlined above remains the same, but $M$ is more complex (now explicitly describing small-scale processes that are sometimes parametrized in large-scale models) and $K$ now describes the sensitivity of $C(t)$ to changes in the model parameters.

4. Current science highlights

Here, I outline a few of the many science highlights associated with understanding atmospheric and surface sources and sinks of trace gases using satellite measurements. As the reader will appreciate, the work shown here is the subject of ongoing research.

(a) Fossil fuel emissions

A number of trace gases are associated with fossil fuel emissions, and here I discuss NO$_2$ and CO$_2$. Fossil fuel emissions represent approximately half the global budget of NO$_x$ (NO + NO$_2$), originating from oxygen thermolysis at high temperatures (approximately equal to 2000 K) and subsequent reactions with N$_2$. Space-borne NO$_2$ measurements have been available for more than a decade (table 1). Tropospheric NO$_2$ columns are inferred from the total columns by subtracting the stratospheric contribution, assuming zonal invariance of the stratospheric contribution. Emissions of NO$_x$ can then be estimated from these data by assuming knowledge of the lifetime of NO$_x$ and of the ratio NO$_2$–NO$_x$ that can be taken from a model of atmospheric chemistry (Martin et al. 2003).

Figure 1 shows tropospheric NO$_2$ columns from the SCanning ImAging spectroMeter for Atmospheric CHartographY (SCIAMACHY) between May 2004 and April 2005 (Martin et al. 2006). The largest columns are generally associated with major industrial and metropolitan areas. There are also enhancements over central Africa, reflecting seasonal biomass burning; over northern equatorial Africa, attributed to rain-induced soil NO$_x$ emissions (discussed below); and near Kuala Lumpur along ship tracks. Figure 1 also shows NO$_x$ emissions determined by combining a priori NO$_x$ emissions and the SCIAMACHY NO$_x$ emissions following equation (3.2). The resulting a posteriori global emissions are more than 20 per cent higher than prior estimates, with the associated uncertainty reduced by half. The biggest discrepancies between the a priori and the a posteriori are over major industrial areas, including Beijing, Tokyo, Buenos Aires and New York City; the ability to relate these discrepancies to particular cities is a reflection of the spatial resolution of the data and the
annual mean quantities that reduce the random noise on these data. Some studies have adopted a first-order approximation of this approach to directly use variations in tropospheric NO$_2$ as a proxy for local emissions (e.g. Richter et al. 2005). Interpretation of these NO$_2$ columns over multiple years allows NO$_x$ emission trends to be studied. Data from the Global Ozone Monitoring Experiment (GOME) and SCIAMACHY instruments between 1996 and 2004 showed a larger than expected accelerating trend of emissions over the industrial regions within China (50% increase over the 9 year period), due to energy growth and technology renewal (Richter et al. 2005). This study also showed a substantial decrease over Europe and the USA, which the authors relate to cleaner car exhausts and changing economical circumstances. The SCIAMACHY has also been used to revise emissions of NO$_x$ from international shipping, a large source of many trace gases that is currently not included in emission treaties.

*Figure 1. (a) Tropospheric NO$_2$ columns (10$^{15}$ molecules cm$^{-2}$) retrieved from the SCIAMACHY for May 2004–April 2005 and (b) associated NO$_x$ emissions (10$^{11}$ atoms N cm$^{-2}$ s$^{-1}$). NO$_2$ columns, filtered for cloud radiance less than 0.5, are averaged on a 0.4°×0.4° grid. The NO$_x$ emissions, averaged on a 2°×2.5° grid, are determined through inverse modelling of the NO$_2$ columns using the GEOS-Chem chemistry transport model. Figure courtesy of Randall Martin, Dalhousie University.*

*Phil. Trans. R. Soc. A (2008)*
The spatial distribution of observed and model NO\textsubscript{x} emissions from shipping was consistent, but SCIAMACHY observations were typically lower, although they had large uncertainties (Richter et al. 2004).

CO\textsubscript{2} observations are currently available from three satellite sensors: the SCIAMACHY; the Atmospheric InfraRed Sounder (AIRS); and the Television Infrared Observation Satellite Operational Vertical Sounder (TOVS), and will be available from two instruments due for launch in early 2009, the Orbiting Carbon Observatory (OCO) and the Greenhouse Observing SATellite (GOSAT; table 1). Synthetic studies have shown that for these CO\textsubscript{2} data to be useful for regional-scale source–sink estimates, they have to have precisions of 1–2 ppm (Rayner & O’Brien 2001); even precisions as poor as 5 ppm on regional scales would improve upon our current understanding of the carbon cycle (CC; Miller et al. 2007), given the spatial coverage provided by these data. Biases on spatial scales shorter than 100×100 km\textsuperscript{2} can be discounted as random noise. Similarly, a global offset in the observations will not affect the inverse modelling since they introduce no error in the observed gradients. Biases on regional to continental scales (i.e. greater than 10\textsuperscript{4} km\textsuperscript{2}) have the largest impact on inferred CO\textsubscript{2} surface fluxes (Miller et al. 2007). Studies to infer CO\textsubscript{2} sources and sinks from the AIRS and TOVS data, using TIR wavelengths, have concluded that the regional biases in the observations, and low sensitivity to the lower troposphere, compromise the estimates (Chevallier et al. 2005a,b). The SCIAMACHY is the only instrument currently in orbit which measures CO\textsubscript{2} columns that are sensitive to the lower troposphere by measuring SWIR wavelengths. Work has shown that the data are broadly consistent with the observed seasonal variations of vegetation patterns over North America and agree with column variations observed by high-precision ground-based instrument and AIRS data (Barkley et al. 2006a,b; Bösch et al. 2006). The year to year trend is also remarkably consistent with surface observations that show an annual cycle, driven by photosynthesis and respiration, on an upward trend (Buchwitz et al. 2007). However, the SCIAMACHY suffers from an unexplained bias that is the subject of ongoing work. The OCO and GOSAT instruments will improve on the SCIAMACHY by having an order of magnitude better resolution that permits improved fitting on CO\textsubscript{2} absorption features at SWIR wavelengths and improved spatial resolution.

(b) Natural sources and sinks of trace gases

Terrestrial vegetation, soils, lightning (Martin et al. 2007) and wetlands are examples of natural sources and sinks of a range of trace gases, which we are beginning to quantify at regional scales through the use of satellite measurements.

Soils represent approximately 15 per cent of global NO\textsubscript{x} emissions, of which 70 per cent are estimated to originate from the tropics. Long, dry periods in tropical ecosystems allow soils to accumulate inorganic nitrogen. Through field measurements, we have known for some time that the onset of the wet season activates water-stressed nitrifying bacteria, leading to large pulses of NO as a by-product of consuming the inorganic nitrogen, but the spatial and temporal extent of this source was not understood until recently. Analysis of satellite observations of NO\textsubscript{2} from the GOME showed that, following the onset of the wet season over the Sahel, large pulses of NO lasting one to three weeks affect 3 million km\textsuperscript{2} of semiarid sub-Saharan savannah (Jaegle et al. 2004).
estimated an African soil source of 3.3 ± 1.8 Tg N yr⁻¹, comparable in size to the biomass burning source (3.8 ± 2.1 Tg N yr⁻¹), and contributing 40 per cent to nitrogen emissions over the Africa continent. Extrapolating these values over the tropics leads to an emission estimate that is a factor of two larger than model-based estimates but consistent with the estimates inferred from other measurements, with implications for modelling tropical tropospheric chemistry (Jaegle et al. 2004). Figure 1 shows that even the annual mean of NOₓ emissions shows some hint of these emissions over northern equatorial Africa.

Figure 2a shows the global distribution of HCHO measured by the Ozone Monitoring Instrument (OMI) during August 2006. Formaldehyde is a high yield product of hydrocarbon oxidation with losses from the oxidation by OH and photolysis, leading to a lifetime of a few hours. Variations in HCHO columns therefore provide constraints on the emissions of short-lived hydrocarbons (Palmer et al. 2003). In the remote atmosphere, the oxidation of CH₄ provides a relatively uniform concentration of HCHO (approximately equal to 4×10¹⁵ molecules cm⁻²), which is significantly elevated by the oxidation of short-lived hydrocarbons in the continental boundary layer. The observed distribution in August shows continental enhancements, an order of magnitude higher than concentrations over the oceans determined largely by CH₄, that reflect biomass burning (South America, south equatorial Africa, Malaysia, Indonesia and parts of China). Elevated concentrations over the southeastern USA, peaking over the Ozarks Plateau (figure 2b), are due to emissions of isoprene (C₅H₈) emitted from a high density of oak trees (Wiedinmyer et al. 2005). Isoprene is a natural hydrocarbon precursor of surface O₃ pollution and the formation of organic aerosols, with an atmospheric lifetime of approximately an hour and a high oxidation yield of HCHO. The southeastern USA, with high surface temperatures and high levels of photosynthetic active radiation, is ideal for isoprene-emitting vegetation. The temporal and seasonal variations of these HCHO column data are remarkably consistent with the ground-based data and model predictions, determined largely by variations in surface temperature (figure 2c). However, the magnitude and spatial distribution of emissions are very different from model predictions with more pronounced year to year variability (Palmer et al. 2006).

(c) Biomass burning

The two main sources of CO are (incomplete) combustion and the oxidation of CH₄ and other hydrocarbons; the chemical source is diffuse and does not contribute significantly to the observed variability in CO. CO has been measured successfully for a number of years by the MAPS (§2), MOPITT, SCIAMACHY, Infrared Atmospheric Sounding Interferometer (IASI), AIRS and Tropospheric Emission Spectrometer (TES). Figure 3 shows the global CO distribution measured at 700 hPa by the MOPITT for July 2004 (Pfister et al. 2005). These data show a large hemispheric gradient with values typically 30–40% lower in the Southern Hemisphere, consistent with ground-based data. The two prominent features are over North America and equatorial Africa, both of which are related to biomass burning. The transport of plumes from Africa over the equatorial Atlantic is evident, despite showing a monthly mean, a reflection of sustained African burning activity. Over North America during this month, elevated CO concentrations originate from large forest fires that raged for most of that
summer. An inverse model analysis of these North American data concluded that 30 Tg of CO was emitted by these fires over the summer, a value comparable to annual anthropogenic CO emissions over North America (Pfister et al. 2005).

Similar studies over eastern Asia (Heald et al. 2005) and the Southern
Hemisphere (Edwards et al. 2006) have drawn attention to the widespread nature of biomass burning emissions and their subsequent long-range transport of pollutants.

Rapid vertical mixing of trace gases associated with intense surface burning can reach the upper troposphere (Jost et al. 2004), but the frequency and nature of these events is currently unknown. Large-scale transport models generally do not capture this intense, vertical transport, often occurring on horizontal scales of less than a few kilometres. Current satellite observations offer, at best, one to two degrees of freedom within the troposphere (table 1), so without careful attention, estimating biomass burning emissions using these data will probably have larger-than-predicted errors. These rapid convective processes are the subject of ongoing research in the field.

5. Concluding remarks: a view of the future

We are towards the end of the preliminary proof of concept stage for several instrument types, such as the GOME, TES, MOPITT and SCIAMACHY, which have shown that it is possible to observe tropospheric O₃, HCHO, CO, NO₂, CH₄ and CO₂ from space. These pioneering instruments have tended to be part of larger interdisciplinary missions that require more than a decade of preparation. As instruments are designed and refined to improve instrument sensitivity and spatial and temporal coverage, and in the light of better scientific knowledge, the user community is looking forward to how the next-generation instruments will improve knowledge of the Earth system in which trace gases and their sources and sinks play a starring role. Here, I outline the requirements for dedicated
atmospheric chemistry and CC missions and critically assess the options that are currently being discussed in the international community. I conclude the paper by discussing two future applications of EO data: (i) surface AQ mitigation and (ii) carbon trading.

(a) Orbital configuration: horses for courses

The orbit in which a satellite instrument resides partly determines the science objectives it can achieve. I shall consider three orbits for future tropospheric trace gas measurements: low-Earth orbit (Sun-synchronous and precessing); geostationary orbit (GEO); and a Lagrange-1 (L1) orbit. Here, I outline the orbit requirements for AQ and CC missions and discuss them in the context of LEO, GEO and L1, based on recommendations put forward by the AQ community to the US National Research Council decadal survey. Many, if not all, of these requirements would also be desirable for a CC mission. I have not considered a mid-Earth orbit because, at these altitudes (1500–15 000 km), incoming radiation would damage current-generation UV/Vis technology, seriously limiting its science deliverables.

Fine horizontal sampling in satellite measurements is obvious for almost any EO sensor. A horizontal resolution of 1 km or better over land, particularly over source regions, would be required for an AQ mission, but an instrument should also provide regional-scale information. A broad swath capability with overlapping measurements would effectively relax the horizontal resolution restriction to 10 km.

Figure 4 illustrates the large diurnal variation of O₃ and NO₂ in an urban atmosphere. Current Sun-synchronous LEO instruments observe this diurnal cycle once in a day (table 1), sometimes early in the morning when it is difficult to relate these data to peak mid-day values. Better temporal resolution enables the characterization of (i) synoptic-scale development of air pollution episodes (as discussed below), (ii) the diurnal variation of emissions, (iii) stochastic processes (e.g. wildfire emission injected into the free atmosphere), and (iv) AQ forecasting. An hourly resolution is desirable over urban areas, with less frequent observations (e.g. every 3–4 hours) suitable for studying long-range transport.

Vertical resolution is a major priority for AQ applications, particularly sensitivity to the continental boundary layer. As discussed in §2, this is a difficult problem owing to interferences due to aerosols and clouds. Combining UV/Vis, SWIR and TIR wavelengths can provide vertical resolution throughout the troposphere, but is unlikely to provide resolution within the boundary layer. Vertical resolution in the free troposphere is also required where long-range transport of pollutants often involves coherent plumes with an approximate thickness of 1 km, resulting from emissions uplifted by fronts, convection and rapid vertical mixing due to surface fires. Vertical resolution is of less importance to a CC mission because lower free tropospheric distributions of long-lived trace gases still bear the signature of surface processes.

Table 2 matches the desired observation requirements, as discussed above, with orbital characteristics. LEO is the least expensive orbit. A Sun-synchronous orbit provides no diurnal information on trace gases, limiting some of the key science objectives of AQ and CC missions. Precessing an instrument in LEO would provide diurnal information, but the repeat time over a specific location.
Figure 4. Example of diurnal variation in surface concentrations of $O_3$ and NO$_2$ in an urban atmosphere at 40°N, and the sampling provided by instruments in a Sun-synchronous LEO (S), a precessing LEO (P), GEO and an L1 orbit. Light and dark regions denote day- and night-times. For UV/Vis sensors, the Sun-synchronous orbit will sample the diurnal cycle once in a day during daylight; the three rows of ‘P’ represent the coverage provided by the precessing LEO for 3 days separated by 15 days; GEO will sample all day but will require three satellites for global coverage and the latitude coverage will be limited beyond ±60°; L1 will continually sample the sunlit disc. Figure courtesy of Nathaniel Livesey, Jet Propulsion Laboratory.

Table 2. Candidate orbits for dedicated air quality and carbon cycle missions and the mission requirements.

<table>
<thead>
<tr>
<th></th>
<th>LEO (Sun-synchronous)</th>
<th>LEO (precessed)</th>
<th>GEO</th>
<th>L1</th>
</tr>
</thead>
<tbody>
<tr>
<td>minimum horizontal</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
<td>yes</td>
</tr>
<tr>
<td>sampling (10 km)</td>
<td></td>
<td></td>
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<tr>
<td>minimum temporal</td>
<td>no, once or twice</td>
<td>no, close with</td>
<td>yes</td>
<td>yes</td>
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<tr>
<td>sampling (1 hour)</td>
<td>and only during day</td>
<td>2-hour repeat</td>
<td></td>
<td></td>
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<tr>
<td>minimum vertical</td>
<td>yes</td>
<td>yes, with caveats</td>
<td>yes, but</td>
<td>limited and</td>
</tr>
<tr>
<td>sampling in troposphere (4–5 levels)</td>
<td></td>
<td></td>
<td>expensive</td>
<td>expensive</td>
</tr>
<tr>
<td>global coverage with</td>
<td>yes</td>
<td>yes</td>
<td>no</td>
<td>yes, sunlit disc</td>
</tr>
<tr>
<td>one sensor</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>expense</td>
<td>cheap</td>
<td>cheap</td>
<td>expensive</td>
<td>most expensive</td>
</tr>
</tbody>
</table>

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will be longer and interpretation of these data would be difficult without a model. Temporal resolution could also be improved by (i) broad across-track swath so that 2 hours temporal resolution can be achieved by overlapping measurements in successive orbits, but this is eventually limited by viewing angle and also leads to a larger probability of cloudy scenes, (ii) using an inclined orbit but this sacrifices high latitude measurements, and (iii) using a constellation of satellites. At the moment, the GOME-2 aboard MetOp and the OMI aboard Aura sample the diurnal cycle twice (table 1). Constellation costs (each platform with a different local equatorial crossing time) could be reduced by exploiting commercial piggybacking opportunities, e.g. the Iridium NEXT constellation representing 66 LEO payloads that will be launched in 2013.

A GEO places a satellite at approximately 36 000 km above the Earth’s equator and has an orbital period equal to the Earth period of rotation so that the satellite observes the same view all day. Such an orbit is often used for meteorological or communication satellites. Measurements can made at UV to TIR wavelengths and fulfils all the resolution requirements for an AQ mission (table 1), with the caveat that achieving a vertical resolution similar to the next generation of LEO will be expensive. Continuous temporal coverage of the sunlit disc effectively removes the problem of cloud obscuration. Three satellites are required to globally observe low to mid-latitudes, with resolution and signal-to-noise decreasing at higher latitudes and associated viewing zenith angles. Geostationary measurements of aerosol optical properties, available from the Meteosat Second Generation Spinning Enhanced Visible and InfraRed Imager (SEVIRI), show great promise in helping to unravel the role of aerosols in modifying cloud radiative properties. The European Space Agency have nominally assigned Sentinel 4 as a geostationary platform for atmospheric chemistry studies to be launched in ca 2018.

The Lagrangian (L1) orbit places an instrument at approximately 1.5 million km away from the Earth, a special point between the Earth and Sun, where the instrument will always view the complete sunlit disc. This combines the advantage of global coverage provided by LEO/MEO and the temporal coverage of GEO, and fulfils all the air quality mission requirements (table 1). This orbit suffers from a lack of vertical resolution, which, as I discussed above, is an important dimension to consider for trace gases with short lifetimes and for studies over geographical regions with rapid vertical transport. This is the most technically challenging and the most costly of all the orbits considered here, owing to the larger telescope required to observe the Earth from such a distance, but it does have a precedent. The SOlar and Heliospheric Observatory (SOHO) is the only instrument that has resided in the L1 orbit at an approximate cost of 1000 M euros.

(b) Adopting an Earth system approach

Partly due to the success of past and existing instruments, the number of new questions we have about the Earth system is larger than the original set of questions we had a decade ago. This is also a reflection of our increasing awareness that the Earth system is connected together via a complex web of interactions that act on a wide spectrum of spatial and temporal scales. As a community, we are also becoming aware that some climate feedbacks from radiative forcing can be sudden, responding to warming within a decade, e.g. melting of arctic permafrost that could release uncertain amounts of CH₄ into the

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atmosphere. So how do we proceed? Do we move towards a framework of rapid response instruments that can be proposed, built and deployed within a few years, which can monitor rapid climate change? Do we focus on comprehensive monitoring of key trace gases in the atmosphere? Or do we move towards a more integrated approach of measuring the Earth by designing Earth system monitoring platforms that measure a wide suite of variables that help interpret trace gas distributions in the troposphere?

The most scientifically fruitful approach is, perhaps, to develop intermediate size EO satellite platforms that comprehensively sample a particular aspect of the Earth system and can be developed and flown within 4–5 years of being proposed. Measuring trace gases such as CH₄ and CO₂ to a precision required to observe surface fluxes represents an engineering feat, but there is no unique information within the measurement itself to improve understanding about source attribution, i.e. whether elevated CO₂ is from biomass burning, fossil fuel combustion or respiration. Using correlations between different trace gases (e.g. using CO to inform about the combustion source) will help with this issue but they will not provide useful information with which to better understand the underlying processes. Coincident measurements of terrestrial photosynthesis (using ratios of spectral bands), fire radiative power (Wooster et al. 2005) and soil moisture, for instance, would disproportionately increase the science return of satellite observations of trace gases. Such a synergistic approach to measuring the CC would facilitate, for example, the study of the daily interplay between leaf phenology, hydrology, biology and atmospheric chemistry and transport.

(c) A grand challenge: integrating EO data and climate policy

The science and policy of climate is becoming progressively interlinked as the economic and humanitarian impacts of projected climate scenarios are being realized. Here, I discuss two examples in which I believe EO measurements of tropospheric trace gases can play a prominent role: (i) mitigating chronic surface air pollution and (ii) reducing global anthropogenic carbon emissions through carbon trading schemes.

(i) Surface air pollution forecasting and mitigation

The chronic surface AQ event over Europe during the August 2003 heatwave serves as an example of the human impacts of surface air pollution, with many hundreds of deaths linked with elevated concentrations of surface ozone (greater than 100 ppb; Stedman 2004). The build up on pollution over the UK during that period was due to a stable high-pressure system that brought in pollution from mainland Europe and prevented dilution of boundary-layer pollution to the free troposphere. The increased temperatures associated with this meteorology led to rapid production of pollution exacerbated by temperature-dependent emissions of isoprene, which effectively increase surface O₃ concentrations (Lee et al. 2006). The frequency of heatwaves as hot as 2003 has been projected to increase 100-fold over the next 40 years (Stott et al. 2004). If these projections are realized, current European surface air pollution episodes are likely to become more extreme with associated impacts on human health and agriculture.

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So how can satellite measurements help? We already have NWP models, one of the principal outputs from which is the weather forecasts shown in the media. NWP centres are already moving towards using satellite observations of trace gases to provide regional AQ forecasts. At some stage, we should expect forecast maps of surface O₃ and other pollutants that will guide our day-to-day lives, e.g. not to exercise vigorously when surface O₃ is above a critical threshold. What this approach does not consider is using these forecasts to develop mitigation strategies. As discussed above, distributions of NO₂ and HCHO provide information on their sources and sinks, which can be used to improve the predictive capabilities of emission and chemistry models. If we know ahead of time, even a few days, that the meteorological and AQ conditions over London during mid-summer 2012 will be similar to those experienced during the 2003 heatwave, we would have the ability to minimize or even avert a chronic AQ episode. In this case, instead of solving just for physical and chemical parameters, we can broaden equation (3.2) to simultaneously minimize the economic ‘costs’ of air pollution episodes on regional human health, industry and food supply. Estimating the costs and risks of surface air pollution required extensive economic and epidemiological analyses. However, armed with such a predictive mitigation system, national governments will be able to help control the health and economic implications of surface air pollution.

(ii) Carbon trading

International trading of carbon emissions on the open market is an approach that is being implemented to incrementally reduce anthropogenic CO₂ emissions. The premise is that emissions from individual countries will be capped at a figure determined by emissions from prior years. If a country emits more than the capped amount, they must buy permits to cover the excess carbon from another country’s allocation that is surplus to requirement. The global cap will be reduced over a number of years and the associated price of carbon emission will increase, if we assume no rapid increase in carbon capture technology. This cap and trade approach to carbon trading is a new and rapidly growing financial market that is likely to be worth trillions of sterling in coming years.

To ensure the successful operation of carbon trading schemes, it is important to ensure subsequent adherence to nationwide emission commitments. A major criticism of carbon trading as an effective method of reducing emissions of CO₂ is that there is currently no objective measure of adherence that has realistic confidence intervals. Carbon emission permit and trading schemes currently rely on self-reporting by firms, based on (essentially) fixed ratios of emissions to output, with monitoring by local or national environmental agencies, with powers to penalize offending firms. The quality of regulation is likely to vary greatly between industries, regions and countries, which is clearly not satisfactory for mitigating the anthropogenic contribution to climate change. Reporting carbon emissions is less time sensitive than air quality mitigation strategies, but rapid dissemination of results is still useful. It is unlikely that, in the near future (within 10 years), satellite observations of CO₂ will be sufficiently accurate to provide reliable independent flux estimates on spatial scales less than 100 km, but subcontinent regional assessments should be possible with upcoming satellite instruments. The first step to integrate atmospheric measurements of CO₂ in carbon trading schemes...
is to determine the measurement network necessary to provide robust CO₂ flux estimates and associated uncertainties on spatial and temporal scales of interest. The extent to which satellites play a role in such a network will partly rely on the success of the upcoming OCO and GOSAT instruments.

I thank Nathaniel Livesey for a critical review of an earlier draft and much advice; Randall Martin, Thomas Kurosu, Louisa Emmons and Christian Frankenberg for providing figures and general support; and John Burrows, Cathy Clerbaux, Folkert Boersma and Paul Monks for advice on the work shown. I also thank Hartmut Bösch and an anonymous reviewer for providing their useful input on the submitted paper.

References


Observing trace gases from space


Paul I. Palmer obtained his BSc in physics at the University of Bristol in 1995, and his DPhil in physics at the University of Oxford in 1999, specializing in satellite remote sensing of the Earth’s atmosphere. He spent the following six years working at Harvard University employing new satellite observations of tropospheric trace gases to improve quantitative understanding of tropospheric chemistry. He returned to the UK in late 2005 as a University Research Fellow at the University of Leeds and took up his present appointment as a Lecturer at the University of Edinburgh in 2006. Knowledge of atmospheric remote sensing and tropospheric chemistry have underpinned much of his subsequent research, and have allowed him to explore topics in climate science, which have until recently suffered from lack of data. His current research interests focus on using space-borne observations to improve estimates of the magnitude and distribution of natural sources and sinks of trace gases from tropical terrestrial vegetation, and estimates of the magnitude, distribution and vertical transport of biomass burning emissions.