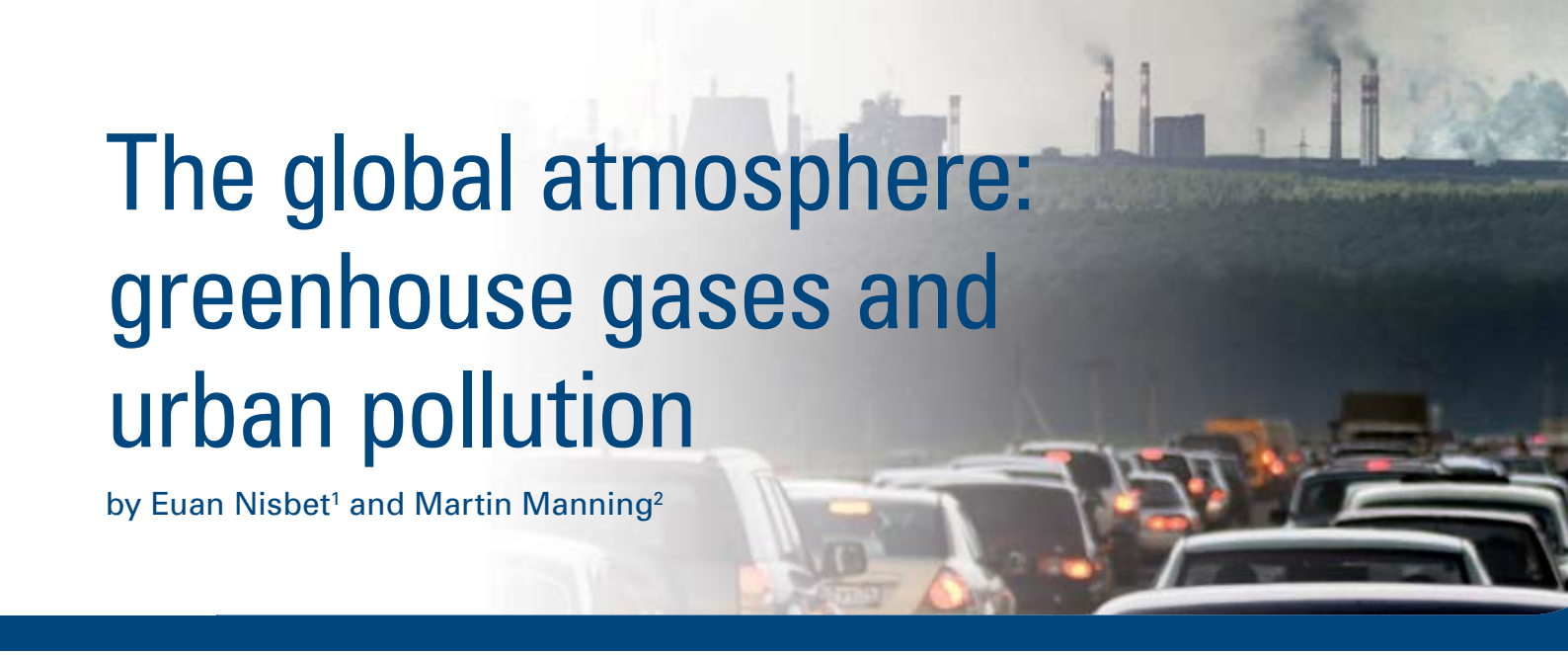

The global atmosphere: greenhouse gases and urban pollution

by Euan Nisbet[1] and Martin Manning[2]

Introduction

For 50 years, since Dave Keeling started monitoring carbon dioxide at Mauna Loa, Hawaii, and the South Pole, scientists have been tracking greenhouse and other trace gases in the global atmosphere. The results have revolutionized our understanding of biogeochemistry and demonstrated that human activities affect climate change and air quality. Precise measurements of trace-gas concentrations, pioneered by Dave Keeling, have led from exciting scientific inquiry to arguably the most serious socio-economic and political challenges that humankind has ever faced.

It has only been through precise and calibrated measurements of trace gases at multiple locations that we have been able to construct reliable budgets for the sources and sinks of both greenhouse gases and the pollutants that control air quality. Anthropogenic effects on the atmosphere have become clearly identified. At the same time, we have developed some understanding of the scale of damage that can be caused by unfettered atmospheric changes. This is now causing a global rethink of the direction and design of economic activity.

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The technological advances necessary to avoid dangerous atmospheric changes come at a cost but, increasingly, the evidence is that the cost of doing nothing would be much larger. Air-quality impacts and those of climate change run potentially into trillions of dollars (Burtraw et al., 2003; Metz et al., 2007; Sitch et al., 2007). Ironically, despite the international attention given to global change, the precise and strategic monitoring of atmospheric trace gases—which identified the problem in the first place—remains markedly underfunded (Nisbet, 2007). Yet this Cinderella science still provides the only way of telling whether mitigation is working.

If we could take atmospheric change for granted, a minimalist approach to monitoring might be justified. The ozone hole, however, showed that atmospheric chemistry is capable of producing surprises. What is more: atmospheric monitoring provides us with the most comprehensive overview that we have of biogeochemical changes in a rapidly warming world. This is no time to take atmospheric monitoring for granted.

Which greenhouse gases are monitored and where?

Greenhouse gases need to be monitored for many reasons. First, this is fundamental science: the breathing of the Earth. Keeling (1960), in his initial report, documented the planetary biosphere's seasonal cycle of growth and decay and showed the dominant effect of the northern hemisphere land masses over the south. Secondly, and more worryingly, monitoring has tracked the steady rise in carbon dioxide. Measurement began in Hawaii in March 1958, recording 316 parts per million (ppm) of carbon dioxide. By March 2007, the comparable value was 384 ppm. The Mauna Loa curve, simple and unambiguous, changed the way we look at the world and at our own actions.

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Greenhouse emissions are now self-declared by many emitting nations. They are estimated from economic and statistical data, such as tonnes of fossil fuel burned, landfill leaks or estimates of methane outbreathings by cows (which can vary significantly across national borders). There are paper audits of the data but, despite the potential for errors in the data-gathering process, there is as yet no independent and comprehensive verification of emissions. This is a major flaw in the Kyoto process, as emissions become associated with actual financial costs or benefits.

Today, global monitoring of greenhouse gases and related species, done for the public good by many nations, is beginning to provide an independent, scientific approach to estimate greenhouse-gas budgets. At present, the data are only adequate to provide global and very broad regional insights or to quantify plumes from large, localized sources. In future, under a Kyoto follow-up treaty, we should be doing much more to audit compliance directly at all levels—local (e.g. factory), regional and national and continental.

The main atmospheric trace constituents monitored at global scales are:

- The core Kyoto gases carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), hydrofluorocarbons (HFCs), perfluorocarbons (PFCs) and sulphur hexafluoride (SF_6);
- Ozone-depleting substances such as chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) controlled under the Montreal Protocol;
- The indirect greenhouse gases hydrogen (H_2) (likely to grow sharply in a hydrogen economy) and carbon monoxide (CO) (involved in methane chemistry and air quality);
- Isotopic carbon dioxide ($^{13}CO_2$), istopic methane ($^{13}CH_4$) and ozone (O_2) (to constrain emission sources and budgets of CO_2).

At local and regional scales many short-lived pollutants, such as volatile organic compounds (VOCs), nitrogen oxide (NO_x) and particulates, are monitored along with species resulting from their chemical reactions, such as ozone.

Not all these gases are monitored equally and the spatial and temporal coverage of available data varies enormously. But, in all cases, restricted coverage or the sporadic nature of measurement programmes that are not funded continuously is limiting our ability to identify changes of either natural or human origin. Verification of the effects of policy on emissions control has been identified as a potential goal of national and international atmospheric science programmes for many years, but has yet to be achieved in a meaningful way.

Monitoring is carried out by national and multinational groups, some governmental, others linked to universities. The most extensive global monitoring programme is run by the US National Oceanic and Atmospheric Administration (NOAA), which also provides most calibration standards. NOAA's carbon dioxide work is closely coordinated with national programmes in Australia, Canada, China, Japan, New Zealand, South Africa and many other nations. The notable developed-nation exception is the United Kingdom.

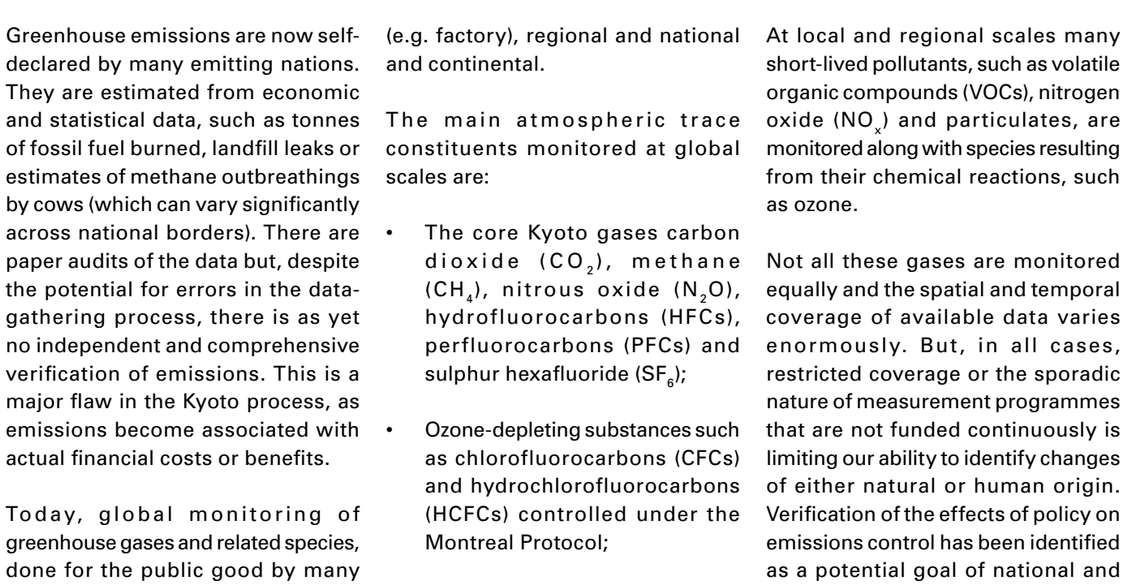

Figure 1—Global carbon dioxide monitoring sites in November 2008 (courtesy A.C. Manning). Stations shown are from programmes run by NOAA (USA), Scripps (USA), Princeton (USA), Commonwealth Scientific and Research Organization (Australia), National Institute of Water & Atmospheric Research (New Zealand), National Institute for Environmental Studies(Japan), the South Africa Weather Service and CarboEurope-IP (EU, including French RAMCES programme).

European Union programmes such as Carbo-Europe and GEOmon make major contributions, coordinating national efforts such as the French RAMCES (Réseau Atmosphérique de Mesure des Composés à Effet de Serre) network, and helping with measurements in India and Africa. However, many monitoring programmes are poorly funded or liable to severe cut-back (as has happened to world-class work in Australia and Canada and to European Union methane monitoring). The major gaps in the monitoring network are in the tropics, especially in India (where there is some French monitoring), Arabia, tropical Africa and Brazil (from where shipment of flask samples is difficult).

WMO coordinates global measurements and analysis, for instance by supporting (since 1975) an international biennial meeting of a carbon dioxide and trace-gas measurement experts panel. Through its Global Atmosphere Watch (GAW) programme, WMO is responsible for supporting international partners that maintain the key components of the GAW global carbon dioxide and methane monitoring network that is part of the Global Climate Observing System. A high level of engagement of the international carbon cycle research community has enabled agreement on analytical standards and methodologies. This community assists WMO in issuing an annual greenhouse-gas bulletin describing consensus on global greenhouse- gas composition and trends. Importantly, GAW measurement expert groups initiate vital round-robin intercomparison studies, unloved by funding agencies but without which the global collaboration and most budget modelling would be almost worthless. Approximately 25 national programmes participate in the work (the number is growing). The work has global scope, with NOAA and RAMCES, in particular, making strategic use of oceanic islands. However, there are major gaps in the coverage (Figure 1), especially in the tropics.

Satellite monitoring of trace gases is still at a preliminary stage but should provide a valuable broader picture. Satellite systems, such as the European Union's Envisat's Sciamachy (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography) instrument use the near infrared to measure the total amount of carbon dioxide and methane in the atmospheric column. Soon, the US National Aeronautics and Space Administration's Orbiting Carbon Observatory and the greenhouse- gases observing satellite IBUKI (GOSAT) of the Japan Aerospace Exploration Agency will provide additional coverage over much of the world. In principle, the near-global coverage provided by remote-sensing techniques may lead to a significant improvement in our ability to relate the fingerprints in trace-gas concentrations to the distribution of the sources and sinks that cause them (Rayner and O'Brien, 2001). However, this approach has yet to be proven. In the near future, we are clearly reliant on the ground-based network. Moreover, in the longer term, we will continue to need the ground truth supplied by surface and upward-looking total column measurements.

Carbon dioxide varies across the planet. Keeling showed it is well mixed on a multi-year timescale, but there are major seasonal and latitudinal variations. Figure 2 shows the "carbon rug" or "flying carpet"—carbon dioxide in the marine boundary layer by time and latitude. This is a marvellous record of the breathing of our planetary biosphere and the increment of human activity. The fine topography of global carbon dioxide variation bears comparison with the global variation in absolute temperature. At the same moment in a spring anticyclone, ground-level carbon dioxide in a large industrialized region may exceed 450 ppm, the value in a budding forest one thousand kilometres away may be 100 ppm lower. Contrast this with a July temperature in the Sahara of 330K and 230K at the South Pole. There

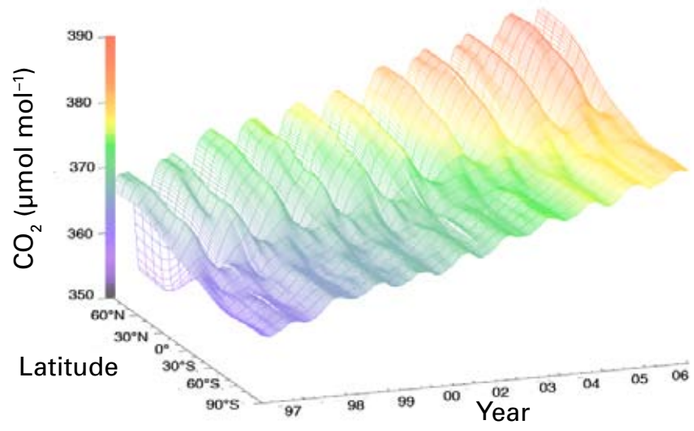

Figure 2—Global average distribution of atmospheric carbon dioxide in the marine background, by time and latitude (from NOAA Earth System Research Laboratory cooperative air sampling network (www.esrl.noaa.gov))

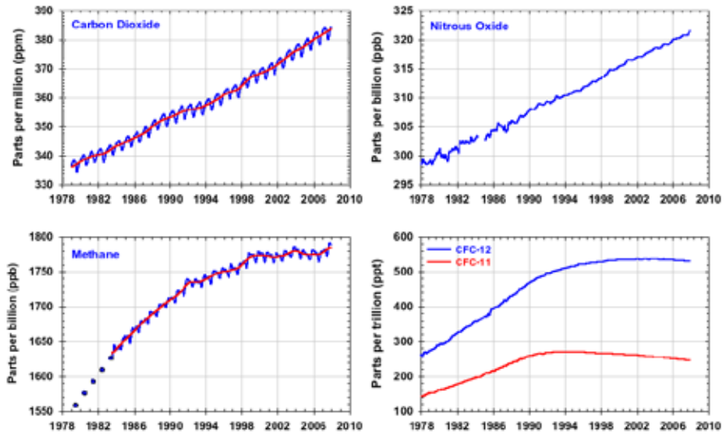

Figure 3—Global averages of the concentrations of the major, well-mixed, long-lived greenhouse gases carbon dioxide, methane, nitrous oxide, CFC-12 and CFC-11 from the NOAA Earth System Research Laboratory cooperative air sampling network since 1978 (www.esrl.noaa.gov/gmd/aggi/)

are large seasonal cycles, and a major hemispheric gradient.

The global greenhouse

Figure 3 shows the evolution of the global averages of the major greenhouse gases since 1978, as measured by the US NOAA Earth System Research Laboratory cooperative air-sampling network. For carbon dioxide, the rise seems inexorable, particularly in the past decade. Note the change in slope in the time of slower growth and El Niño in the early 1990s. Nitrous oxide is also increasing steadily: this gas, released in nylon manufacture and by agriculture, may be a very cost-effective target for reduction measures.

Methane, even more than nitrous oxide, is an attractive first target for reduction efforts as many of its emissions, such as those from landfills and gas pipes, are economically wasteful as well as environmentally damaging. The methane budget reached near-equilibrium in the early 1990s but methane may now be increasing again, especially in the Arctic (unpublished NOAA results and Rigby and Prinn, 2008). The causes are not yet known and, as long as this remains the case, that is a telling indictment of our ability to diagnose atmospheric change. Formerly, the European Union supported methane isotopic monitoring of Arctic methane, which could, in principle, distinguish source inputs. This was terminated and isotopic work in the Arctic now largely depends on US and national programmes. Finally, in Figure 3, the chlorofluorocarbons show encouraging declines, reflecting the success of the Montreal Protocol process. This outcome is a hopeful pointer for follow-up to the Kyoto Protocol.

What is the point of measuring greenhouse gases?

Regional studies

Studying the detailed distribution of carbon dioxide as revealed by the monitoring network addresses a wide variety of scientific questions. Examples include quantifying sinks of carbon dioxide on land versus ocean; assessing the impacts of the 2003 European heatwave on atmospheric carbon dioxide; studying the impact of the El Niño/Southern Oscillation cycle; or watching the wider impacts of volcanic events.

In the USA, NOAA monitors carbon dioxide using continuous observations from tall towers and sampling by small aircraft. The data give regional gradients in space and time, fed into a data-assimilation carbon cycle modelling system called Carbon Tracker. From this, Peters et al. (2007) estimated the net carbon dioxide exchanges between the ground and the atmosphere from 2000 to 2005. They found that the North American land biosphere is a major carbon sink, absorbing some 0.65 x 1 015 grams of carbon per year (note: the variability is large, from 0.4 to 1.01 x 1 015 g/yr). This partly offsets the fossil-fuel emissions, estimated as 1.8 x 1 015 g/yr. In Europe, similar tall tower monitoring is under way as part of Carbo-Europe's Chiotto programme and an emerging Integrated Carbon Observing System.

Stephens et al. (2007) used vertical profiles from aircraft measurements to infer that northern uptakes were around 1.5 x 1 015 g/yr, less than previously estimated, and that net tropical emissions were small (0.1 x 1 015 g/yr), implying that strong tropical uptakes largely balanced the major emissions from tropical forest clearing and grass fires. Piao et al. (2008) also sounded a warning note, using both NOAA observational records and modelling to find that autumn warming may have a significant impact on carbon dioxide budgets.

Isotopic monitoring is particularly powerful. Because emissions from different sources typically have different isotopic ratios, source strengths can be estimated. For example, if winds blowing from a coalfield are enriched in $^{13}CH_4$, then that increment can be related to the total methane they release. Using wind back-trajectory analysis, such source inferences can be made at great distances: methane from Canadian fires can be "smelled" in Ireland; African methane reaches New Zealand. The isotopes eventually mix almost as coloured streamers do in smoke trails.

Levin et al. (2007) used radiogenic ^{14}C observations at regional stations in Germany, compared with measurement in the free troposphere at Jungfraujoch, Swiss Alps, to estimate regional carbon dioxide surpluses over background.

More generally, Bakwin et al. (2004) showed that, by using carbon dioxide measurements, it is possible to assess carbon dioxide emissions on a regional scale (of the order of one million square kilometres, provided the monitoring network is adequate). With relatively inexpensive modern instruments and some supporting aircraft sampling, it should be possible to quantify emissions by quantity and by isotopic source. The increase in effort required is not large, compared to existing programmes. In principle, it should be possible directly to audit compliance with any future Kyoto-like agreement, at least in major industrial regions such as China, Europe, India and the USA.

Urban air impacts

In the developed world, urban air quality has improved markedly over the past decade. Figure 4 shows mean monthly carbon monoxide, a good proxy for overall air quality, for Egham, south-west of London, United Kingdom. Days with very high carbon monoxide are now rare. This improvement has occurred since 1997 as a consequence of reductions in car emissions brought about by catalytic exhaust converters, a tax regime that made unleaded fuel cheaper than leaded (thereby reducing catalyst poisoning) and a more rigorous annual inspection regime. London's air is now frequently sparkling. London is not alone: most European and US cities have similar improvements. Though very significant health risks remain, the overall picture is much brighter than a decade ago.

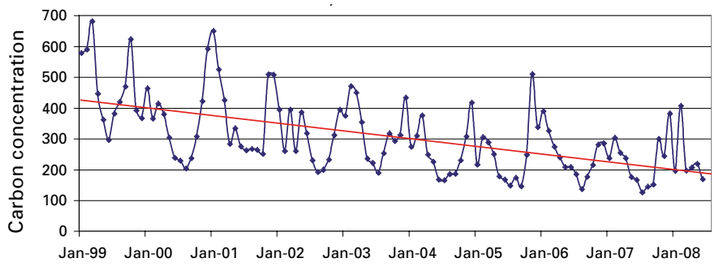

Figure 4—10-year record for carbon monoxide, Egham, west of London. Mean monthly carbon mixing ratios are shown. The detailed record for methane also shows a sharp decrease in severe pollution events since 1997. From 2006 to 2008, many days with westerly winds had carbon not far above contemporary (seasonal) Atlantic background levels and methane close to simultaneous measurements at Mace Head, western Ireland (unpublished preliminary data, Royal Holloway Group: note that the line is indicative only).

Urban air quality in many cities in newly industrializing countries, especially China and India, remains extremely poor. Here, too, however, improvements are beginning to take place. Beijing's air quality is illustrative: the heroic efforts to improve conditions for the 2008 Olympics led to public desire for better air nationally. In India, too, public pressure may bring about improvements. North American and European experience, both recently with air quality and in the decade previously with acid rain, show that, with determined effort, significant betterment of air quality can take place within a decade.

While local air quality around major urban centres is reasonably well understood, the expected growth of megacities is likely to raise new issues, particularly as we release new synthetic gas species to the atmosphere at a challenging rate and before we understand the full environmental consequences of doing so. What is perhaps of even more concern is that we still know little about the broad background of atmospheric chemistry changes that may be occurring on larger scales. For example, there is evidence that fluctuations in the concentration of the dominant oxidizing species, the hidroxil (OH) radical, can be quite large (Manning et al., 2005) and it is revealing that much of what we infer about hidroxil comes from atmospheric chemistry models rather than from observations (Jöckel et al., 2003; Spivakovsky et al., 2000).

Ironically, Chinese and Indian aerosol pollution acts on a global scale as a negative greenhouse forcing. Rightly, these nations will cleanse their air. In so doing, they ameliorate local problems but accelerate global warming. Moreover, catalytic exhaust converters add weight to a car, and inefficiency. The overall fuel consumption of vehicles in western nations is thus higher than it would be if air were dirtier. Local environmental improvement can mean a transient increase in global warming.

Conclusion

With the international support of WMO, atmospheric greenhouse-gas monitoring and analysis have been developed in a global integrated system reaching 50 years back to the pioneering measurements of Dave Keeling on Mauna Loa Hawaii and the South Pole. The enormous scientific and socio-economic implications of this work have too often been taken for granted. The global increase in greenhouse warming and the hot spots of degraded air quality are reasonably well understood. Now, the need is for more comprehensive and more detailed (and relatively inexpensive) monitoring designed

to support relevant modelling in order to determine trace-gas budgets regionally and locally. This will enable an independent audit of emissions, by source, by location and by time. An effective carbon tracking tool is now possible.

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