Chapter 2. Black carbon and tropospheric ozone precursors: drivers, emissions and trends

Coordinating lead author: David Streets (Argonne National Laboratory, USA).

Lead authors: Hajime Akimoto (Asia Center for Air Pollution Research, Japan), Paulo Artaxo (University of Sao Paulo, Brazil), Zbigniew Klimont (International Institute for Applied Systems Analysis, Austria), Kaarle Kupiainen (International Institute for Applied Systems Analysis, Austria), Greet Janssens-Maenhout (Joint Research Centre, European Commission, Italy), Harry Vallack (Stockholm Environment Institute, Environment Department, University of York, UK), Elisabetta Vignati (Joint Research Centre, European Commission, Italy).

Contributing authors: Gufran Beig (Indian Institute of Tropical Meteorology, India), Christopher Heyes (International Institute for Applied Systems Analysis, Austria), Lena Höglund Isaksson (International Institute for Applied Systems Analysis, Austria), Jean-François Lamarque (National Center for Atmospheric Research, USA), Zifeng Lu (Argonne National Laboratory, USA), N.T. Kim Oanh (Asian Institute of Technology, Thailand), Yuxuan Wang (Tsinghua University, China).

Key findings

Air pollution control policies will be effective. Up to the present time, emission reduction policies in North America and Europe have reduced emissions of sulphur dioxide (SO_2), nitrogen oxides (NO_X) and particulate matter (PM), including black carbon (BC), from many sectors. Current and planned regulations are expected to continue to drive down emissions in these two regions. In others, similar policies are expected to slow down their rate of growth in the future and, in the case of Northeast Asia, to bring about reduced emissions of BC, organic carbon (OC) and SO_2. Ensuring effective implementation of these policies is necessary to achieve the reductions. Sources emit a range of pollutants at the same time, and it is essential to consider co-emitted ones when designing abatement measures.

Without implementation of measures beyond current and planned regulations, methane (CH_4) emissions are expected to increase in the future. Increased coal mining and oil and gas production, coupled with growth in agricultural activities and municipal waste generation, are likely to lead to more than 25 per cent higher global anthropogenic CH_4 emissions by 2030 relative to 2005. The projected increase in fossil fuel production is the main driving force behind this growth.

Without implementation of measures beyond current and planned regulations, emissions of BC, OC and carbon monoxide (CO) are projected to remain relatively constant to 2030, as planned improvement in combustion technology will be offset by continued economic growth. The regional trends vary, however, with decreases in North America, Europe and Northeast Asia being balanced by increases in Africa and South Asia. Emission reductions in the transport sector will be the main cause of improvement. As the role of traditional biomass in residential combustion declines and improved technology comes into play, it is generally expected that primary OC/BC ratios will fall.

Emissions from residential biomass combustion are expected to become even more important in the future than they are today. Because efforts to curtail BC and OC emissions from transport and industrial sources are projected to be increasingly successful, the residential use of traditional biofuels in developing countries of Asia and Africa will dominate global emissions of anthropogenic carbonaceous aerosols in the future, contributing nearly half of BC emissions and about two-thirds of OC emissions by 2030.

Of those pollutants with a long history of control policies, NO_X emissions are expected to remain relatively constant in the future, while SO_2 emissions will decline between 2005 and 2030. Modest reductions in NO_X emissions in North America and Europe will be offset by increases everywhere else. Despite a strong increase, estimated at nearly 50 per cent, in South, West and Central Asia, global SO_2 emissions are projected to decline because of emissions reduction elsewhere, particularly in North America and Europe.

Open biomass burning is a large contributor to global emissions of carbonaceous aerosols and ozone (O_3) precursors. Recent estimates suggest that forest fires, savannah and grassland burning, etc., excluding agricultural burning, contribute about half of global BC and CO emissions and two-thirds of OC ones. It is not known how these emissions will change in the future and for this study they are assumed to remain constant at today's levels. Open biomass burning can be a key source contributor if it occurs close to a sensitive ecosystem such as the Arctic or the Himalayas, so the regional emissions distribution is important.

Confidence in emission estimates of primary aerosols and O_3 precursors varies by pollutant and world region. For NO_X, SO_2 and carbon dioxide (CO_2), knowledge of emission levels is relatively good. This derives from a history of emission measurements and consistent emissions from a given source type. However, despite an influx of new observational and measurement data in recent years, emissions of BC, OC, CO, CH_4 and non-methane volatile organic compounds (NMVOCs) remain quite uncertain for many source types. For CH_4 and NMVOCs, confidence is within a factor of three; and for the main products of incomplete combustion, BC, OC and CO, confidence at present is still only within a factor of roughly 2–5. Confidence is greater for the developed world than the developing regions because of a longer history of emission estimation, a wider dataset of source measurements, and more reliable statistical information on the number of sources and activity levels. As more sources are measured and more survey data gathered, confidence in emission estimates is expected to grow.

2.1 Introduction

This chapter discusses the current state of knowledge of primary emissions of carbonaceous aerosols, BC and OC, other fine particles and their precursors, SO_2 and ammonia (NH_3) and the precursors of tropospheric O_3, NO_X, CO, NMVOCs and CH_4. Emissions of CO_2 are also included in some presentations for comparison purposes. The chapter presents anthropogenic emissions of these species, as generated by the International Institute for Applied Systems Analysis Greenhouse Gas and Air Pollution Interactions and Synergies (IIASA GAINS) model for 1990, 2005 and 2030, the future reference scenario, as well as information on emissions from natural sources that are part of global climate modelling. It also shows results from some other global emission inventories by way of comparison and confirmation. Emissions of each species are disaggregated by major world region and emitting source category in order to highlight the most important contributors to global change and to direct the reader towards opportunities for mitigation action, which are discussed in detail in Chapter 5. To place present-day emissions in context, historical trends and likely future directions of emissions by considering socioeconomic and other drivers are also examined. The degree of confidence in these emission estimates is discussed, with reference to available quantitative estimates of uncertainty. For some species the emission estimates are quite uncertain, and this carries over into uncertainty about their impacts and mitigation.

2.2 Socioeconomic and other drivers of emissions

Emissions of BC and tropospheric O_3 precursors are driven by basic societal forces that govern the production and consumption of energy, food and materials:

1. Population growth;

2. Economic development that drives, for example, the transition away from residential burning of solid fuels, the extent of private vehicle ownership, and dietary habits such as increased meat consumption that affects CH_4 emissions;

3. Consumption patterns, driven by population pressure, that include land-use changes involving deforestation and wetlands removal;

4. Technological change that can help in faster emission reductions through the spread of new, high-efficiency and low-emission technologies;

5. Environmental regulations driven by regional health and ecosystem concerns, resulting in, for example, reductions in PM, SO_2 and NO_X emissions; and

6. Other policy drivers such as global climate change mitigation agreements.

All these drivers interact with different sectors of human activity and have distinct timeframes for implementation and the achievement of objectives.

There are other driving forces that affect emissions but are not so directly linked to human activities. These include future climatological changes that may affect, for example, water availability and drought, which would in turn have an influence on food production and wildfires, and consequently on emissions. Other examples might relate to the frequency and intensity of lightning and associated NO_X, soil NO_X and CH_4 released from warming tundra, increased tropospheric O_3 formation due to higher temperatures, etc. At the same time, previously unavailable areas may open up to enable some economic activities to shift or expand, such as drilling for oil and opening new shipping routes in the Arctic.

Before the start of the industrial revolution, emissions of BC and tropospheric O_3 precursors were essentially driven by natural episodic events – predominantly wildfires, volcanoes and other geogenic releases – and by the same kinds of continuous natural releases that are seen today, such as CH_4 emissions from wetlands, biogenic NMVOCs, and NO_X from lightning. Anthropogenic emissions at that time were limited to biomass burning for cooking and heating and represented only a small fraction of total emissions: for O_3 precursors and SO_2 less than 10 per cent, while for BC about a third of the total (Lamarque *et al.*, 2010). Some episodic releases, such as the large volcanic eruptions of El Chichon (1982) and Mount Pinatubo (1991), injected enormous amounts of ash, dust and gases into the atmosphere and had worldwide effects lasting two or three years (McCormick *et al.*, 1995).

The same is true of the large wildfires that periodically break out in Siberia and are often initiated by lightning strikes. Large boreal wildfires may also periodically inject smoke into the lower stratosphere (Fromm *et al.*, 2005). These episodic natural emissions can be of similar magnitude to anthropogenic emissions. Even some of the continuous natural emissions are on the same scale as present-day human-made emissions, examples of which include CH_4 from wetlands, bogs, swamps and tundra (200–400 Mt/yr, from Fung *et al.*, 1991); biogenic NMVOCs (500–750 Mt/yr, from Guenther *et al.*, 2006); volcanic SO_2 (13–15 Mt/yr, from Andres and Kasgnoc, 1998); lightning NO_X (about 5 Mt Nitrogen/year, from Schumann and Huntrieser, 2007) and NMVOCs from biomass burning (about 80 Mt/yr, from Lamarque *et al.*, 2010).

Though emissions from natural sources can show large inter-annual variability, they have not shown significant long-term trends (except for open biomass burning which has been growing strongly since the 1950s, largely from deforestation and land clearance, and hence is no longer exclusively natural). Anthropogenic emissions, on the other hand, have increased dramatically since pre-industrial times: for example, SO_2 and NO_X emissions had increased nearly 50-fold and emissions of BC and CO by factors of 5 and 10 respectively by the year 2000 (Lamarque *et al.*, 2010).

In the early to mid-19th century, widespread industrialization saw the advent of the use of fossil fuels, initially coal, to provide energy to power the production of goods and services. The development of the Haber-Bosch process for producing nitrogen fertilizer at the beginning of the 20th century led to an unprecedented growth in food production that supported strong population growth but at the same time caused a cascade of environmental changes, including increased air pollution and the perturbation of greenhouse-gas levels (Erisman *et al.*, 2008). At the same time, biofuels were being consumed in large amounts wherever human settlement occurred in timber-rich parts of the world (Fernandes *et al.*, 2007). At the beginning, the burning of wood and coal in factories and homes and on railways was an inefficient process that paid no regard to smoke emissions; large quantities of PM, rich in the carbonaceous products BC and OC, were therefore generated. Gaseous species including NO_X, CO, NMVOCs and SO_2 were produced at the same time in varying quantities.

Anthropogenic emissions began to grow rapidly, and it was not until the 1920s that attention began to be paid to limiting the emissions of PM through various add-on removal devices. The economic depression of the 1930s and installation of rudimentary emission controls led to a modest decline in most emissions. In many parts of the world, however, wartime production reversed this trend in the 1940s, and emissions began to increase rapidly in the post-war economic revival, beginning in about 1950. This trend spread quickly to most parts of the world, especially in the northern hemisphere, which until now has been responsible for the majority of human-caused pollution. As emissions grew and associated environmental and health impacts – acid rain, respiratory diseases, and heavy metals poisoning – became more evident, serious attention started to be paid to preventing atmospheric emissions in the early 1970s, at least in the developed world. The last few decades have seen a strong decline in emissions of PM and acidifying pollutants from industrial and transport sources in the developed world. However, since the late 1990s, a rapid increase in emissions in Asia has offset these reductions, often leading to continued growth at global level. Emissions of CO_2 have kept growing, and only the most recent economic crisis resulted in their temporary stabilization (Friedlingstein *et al.*, 2010).

For the future, it is generally expected that growth in emissions of air pollutants will be tempered by continuing pressure to achieve ever more ambitious environmental targets. In some cases emissions will be ameliorated simply by economic development, which will, for example, gradually replace residential solid fuel combustion with cleaner fuels and remove older vehicles from the road. This will go some way towards reducing emissions of PM, including BC and OC, as well as hydrocarbons and CO. For other species, primarily SO_2 and NO_X, where there is now a fairly long history of pollution control, the experience of developed countries can be expected to spread gradually throughout the world and hold down future emissions; recent emission inventories and assessments in Asia seem to confirm this assumption (for example, Zhang *et al.*, 2009a; Klimont *et al.*, 2009; Xu *et al.*, 2009). For some sources of all species, however, mitigation is either difficult or expensive and, without new policies, emissions will persist for the foreseeable future. This may lead to growth in total global emissions if the spread of cleaner technology cannot keep pace with economic and industrial growth. Emission projections are sensitive to the socioeconomic attributes of the scenarios, however, and it is not always clear which pathways will generate the highest emissions. For example, while high economic growth is likely to result in high levels of emissions, it may also promote the transfer of high-performing technologies to less developed countries.

2.3 Global BC, OC and O_3 precursor emissions

This section reviews estimates of global emissions of BC, OC and tropospheric O_3 precursors, highlighting the important source types, economic sectors and geographical regions. It begins by describing the IIASA GAINS model used in this Assessment for emissions calculations, together with its assumptions about emission factors (Section 2.3.1). Section 2.3.2 presents emissions for the year 2005 from the IIASA GAINS model that were used to anchor the future reference scenario emissions to 2030. Though emphasis is on anthropogenic emissions, the major natural sources of relevant species are also introduced. Biomass-burning emissions are discussed in section 2.3.3, as they represent a large source of all relevant species. Uncertainties in present-day emissions are discussed in section 2.3.4, and key observational support for current emission trends is provided in section 2.3.5. How present-day emission levels were established is discussed in section 2.3.6, through examples of historical emission estimates since the beginning of industrialization. Section 2.4 describes the assumptions that were made for the construction of the reference scenario, and section 2.5 contains a detailed description of recent past, present and future emission estimates from the IIASA GAINS model used as the basis for the analysis of mitigation options (Chapter 5).

2.3.1 The IIASA GAINS model

Estimation of anthropogenic emissions in this chapter and the analysis of mitigation opportunities (Chapter 5) have been performed with the IIASA GAINS model (http://gains.iiasa.ac.at), an integrated assessment model that explores cost-effective emission control strategies to reduce greenhouse gases and/or improve local and regional air quality. The model has been applied to analyse strategies in a number of regions and countries and, recently, for the whole world. The emission calculation in IIASA GAINS draws on the available measurement literature and has been reviewed by experts from academia, government and industry who work on source-specific issues and national aspects of emissions. The model applies emission factors that reflect country-specific conditions such as fuel quality, combustion technologies, fleet composition, maintenance levels and the application of control technologies.

Emission factors for BC and OC in the GAINS framework have been developed in parallel with the assessment of PM emissions to assure overall consistency (Kupiainen and Klimont, 2007). Detailed discussion of the calculation of BC and OC emissions in GAINS, including emission factors for the developed countries, is given in Kupiainen and Klimont (2007) and for Asia in Klimont *et al.* (2009). Emission factors for other world regions (see Cofala *et al.*, 2007) are based on data from Bond *et al.* (2004, 2007). Methane emission factors have been derived following the guidelines (1997 and 2006 versions) of the Intergovernmental Panel on Climate Change (IPCC) as closely as data availability allows. Country-specific information has been included from the United Nations Framework Convention on Climate Change (UNFCCC) and other external sources. For some source categories (e.g., livestock and coal mines), country-specific implied emission factors have been used directly as reported to the UNFCCC, while for others (e.g., oil and gas production and transportation) emission factors have been derived from underlying data (e.g., fractions of heavy/conventional oil, current utilization of associated gas, pipeline lengths and amounts transported). For countries not reporting to the UNFCCC, emission factors have been derived based on underlying factors available from international statistics and national sources. For NO_X, the emission factors originate from the Regional Air Pollution Information and Simulation (RAINS) databases for Europe (Cofala and Syri, 1998a) and the GAINS model implementation for Asia (Klimont *et al.*, 2009). For other regions, the default factors that distinguish the effects of applied emission control measures were used as a starting point, then adjusted to the values implied by available national or international emission inventories (Cofala *et al.*, 2007). Similarly, for SO_2, information originating from Cofala and Syri (1998b), Cofala *et al.* (2007) and Klimont *et al.* (2009) was modified for other countries with coal quality parameters provided in the IEA Coal Research database (IEA, 1997). Emission factors for CO are based on the IPCC guidelines (Houghton *et al.*, 1997) and modified for country-specific conditions (e.g., for wood burning in the domestic sector and for transport) to the extent that appropriate information was available. For NMVOCs, sources include Klimont *et al.* (2000, 2002), Wei *et al.* (2008), and the EDGAR v4 database (EC-JRC/PBL, 2010). The CO_2 emission factors rely on IPCC recommended values and for NH_3 on Klimont and Brink (2004), Bouwman *et al.* (2002), Fischer *et al.* (2010) and EDGAR v4 (EC-JRC/PBL, 2010).

During 2010, for the purpose of this Assessment, emission factors for several priority sectors were revisited to include results from the most recent scientific literature. Emission factors for cook stoves and power generation now take into account measurements by Roden *et al.* (2006, 2009), Christian *et al.* (2010), and recent studies by Chen *et al.* (2009), Zhi *et al.* (2009) and Zhao *et al.* (2010). In the transport sector a major new feature is the estimation of emissions from high-emitting vehicles based on studies by Durbin *et al.* (1999), Yanowitz *et al.* (1999), Hsu and Mullen (2007), Ban-Weiss *et al.* (2009) and Subramanian *et al.* (2009). The update of input data to GAINS also included a revision of emission factors for diesel cars and trucks, for which emissions in real operating conditions differ from emission factors derived from test cycles. This revision was based on the results of research projects like ARTEMIS (http://ec.europa.eu/transport/road safety/projects/doc/artemis.pdf) and HBEFA (http://www.hbefa.net/e/index.html). Data used by IIASA were provided by the COPERT 4 (v7.1) model (http://lat.eng.auth.gr/copert/files/COPERT4_v7_1.pdf). Emission factors from brick manufacturing, coke production and garbage burning in the developing world were revised to take into account the recent source measurements conducted by Christian *et al.* (2010) and Bellprat (2009).

2.3.2 Emissions for the year 2005

Table 2.1 presents anthropogenic and natural emissions of each species for the year 2005, as used by the atmospheric models in this Assessment. The anthropogenic totals are taken from the GAINS model and are shown in Figure 2.1; other estimates of present-day emissions are presented in Appendix A.2. Anthropogenic contributions are discussed in detail in section 2.5. The transport sector in Table 2.1 includes emissions from international shipping and aviation that come from Representative Concentration Pathway 8.5 (Lamarque *et al.*, 2010). Aviation is a minor contributor to global anthropogenic emissions of these species, but international shipping contributes about 14 per cent and 23 per cent of global SO_2 and NO_X, respectively. As discussed in section 2.1, emissions from natural sources are significant for all pollutants and are essential inputs to the atmospheric and climate models. Table 2.1 includes the values of natural emissions that are used in the GISS and ECHAM models. With the exception of agricultural burning, open biomass burning is categorized as a natural source in Table 2.1. While it is clear that wildfires are under notable human influence (Achard *et al.*, 2008), it is not possible to quantify the human causation globally. Wildfires are also affected by local climatic factors (Bowman *et al.*, 2009) that may change over time. Policy measures to reduce wildfires and their emissions can only be of limited value, therefore, and are not a focus of this mitigation-oriented Assessment. Nevertheless, it must be remembered that wildfires in forested land and savannah are important sources of global carbonaceous aerosols and contribute substantially to O_3 precursors. Section 2.3.3 discusses biomass burning in greater detail.

2.3.3 Emissions from biomass burning

Emissions from fire are an important component of the Earth's system, controlling atmospheric composition at a global level (Bowman *et al.*, 2009). Biomass burning is the major source of BC, OC and O_3 precursors in the southern hemisphere and is an important contributor in the northern hemisphere.
It is seasonal, with significant variability in

Table 2.1 Anthropogenic and natural emissions for the year 2005 used in this assessment (Mt/yr)

	BC	OC	$PM_{2.5}$	SO_2	NO_x[a]	CH_4	NMVOC	CO	NH_3[b]
Anthropogenic									
Large-scale combustion	0.10	0.15	8.1	71.6	34.1	0.36	1.2	29.9	0.07
Industrial processes	0.43	0.66	4.5	12.7	2.4	0	8.0	74.2	0.11
Residential-commercial combustion	2.7	9.6	17.8	5.8	5.0	8.8	37.9	195	0.34
Transport	1.6	1.4	3.4	15.9	71.5	2.3	38.5	266	0.36
Fossil-fuel extraction and distribution	0.28	0.06	0.51	2.4	1.4	101	36.4	2.0	0
Solvents	N/A	N/A	N/A	N/A	N/A	N/A	23.4	N/A	N/A
Waste/landfill	0.1	0.75	1.3	0.06	0.12	49.8	1.1	6.2	0.02
Agriculture[c]	0.31	1.2	3.4	0.16	0.26	126	4.0	25.5	39.5
Total anthropogenic	5.5	13.8	39.0	109	115	288	150	599	40.4
Natural[d]	3-3.7	33-38	6000[e]	28-31	54-60	210	470-549[f]	46-63	21
Global total	8.5-9.2	47-52	6039	137-140	169-175	498	620-699	645-662	61

a Reported as NO_2.

b NH_3 emissions are from EDGAR v4.1.

c Includes the burning of agricultural residues.

d Includes the open burning of all biomass other than agricultural residues.

e Includes total fluxes of sea salt and dust. According to IPCC (2007), the submicron shares of sea salt and dust are 15 per cent and 7–20 per cent, respectively.

f Isoprene.

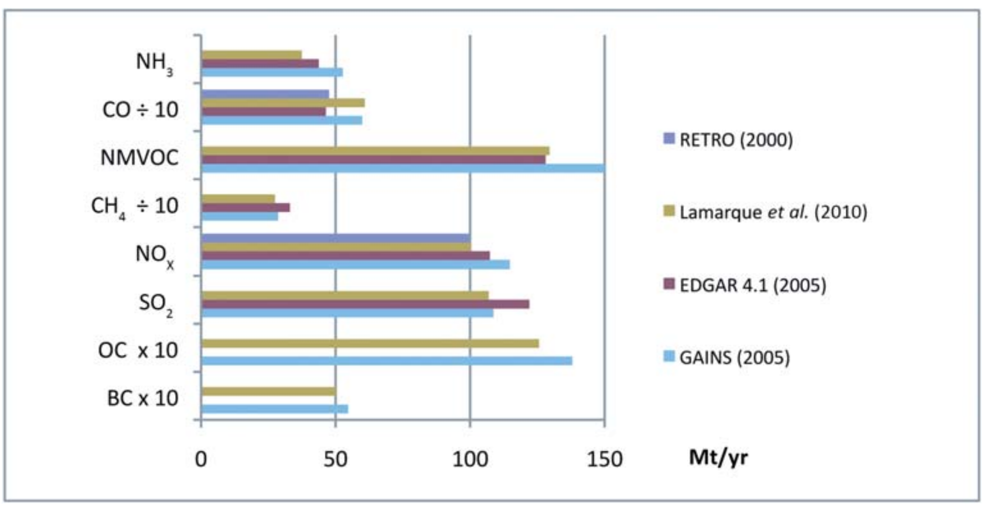

Figure 2.1. Comparison of GAINS 2005 emissions with other global inventories (not including open biomass burning).

emissions both temporally and geographically (Andreae and Merlet, 2001; Streets *et al.*, 2003b). Particles from biomass burning are the major anthropogenic aerosol component in South America, Africa and Southeast Asia during their respective dry seasons. Such emissions have been shown to significantly alter the radiation balance and cloud properties over large regions of the southern hemisphere (Reid *et al.*, 2005a, b; Andreae *et al.*, 2004; Artaxo *et al.*, 2002). Different approaches have been used to obtain emission factors for biomass-burning, such as direct measurements over fires in field experiments (Yamasoe *et al.*, 2000) and aircraft experiments (Yokelson *et al.*, 2007), as well as laboratory measurements (Christian *et al.*, 2003). Andreae and Merlet (2001) compiled emission factors for over 100 trace-gas species and aerosol components for tropical forests, extratropical forests and savannah and grassland emissions.

There are two main components to human-caused biomass burning: deforestation fires and management fires, for example of agricultural residue or as pasture maintenance. Although deforestation rates have been reduced since 2005, especially in the tropics, neither the incidence of fire nor the quantity of smoke in tropical regions has fallen commensurately (Artaxo and Andreae, 2007). Recent estimates of carbon emissions from open biomass burning range from 1 500 to 2 800 Mt C/yr (van der Werf *et al.*, 2010) over the period 1997–2009. In its Fourth Assessment Report (AR4) the IPCC estimated emissions from deforestation to be 1 600 Mt C/yr (IPCC, 2007). Efforts to provide online fire data worldwide through the Global Fire Monitoring Center show a very dynamic system that responds quickly to precipitation changes as well as policy measures.

Several detailed global inventories of biomass-burning emissions are available. These include EDGAR (Olivier *et al.*, 1996; EC-JRC/PBL, 2010), Bond *et al.* (2007) and Junker and Liousse (2008). A gridded historical (1850–2000) biomass-burning emissions product was recently made available in support of the IPCC's Fifth Assessment Report (AR5) (Lamarque *et al.*, 2010). However, only a few inventories include biomass-burning emissions for past decades (Ito and Penner, 2004; Schultz *et al.*, 2008). For the principal categories of biomass burning the main emission datasets are:

1. The RETRO inventory (Schultz *et al.*, 2008), which provides emissions from wildfires for each year during the 1960–2000 period, on a monthly basis;

2. The GICC inventory (Mieville *et al.*, 2010), which gives emissions from open biomass burning for the 20th century (1900–2000) on a decadal basis, based on Mouillot and Field (2005); and

3. The Global Fire Emissions Database (GFED) inventory (van der Werf *et al.*, 2006, 2010), which now covers emissions for the 1997–2009 period in GFED3.

Due to the efforts of several countries to reduce deforestation to avoid carbon emissions, several regions are observing important changes in deforestation rates and biomass-burning emissions (Artaxo, 2010). These efforts have been encouraged by the Reducing Emissions from Deforestation and Degradation (REDD) scheme, in which carbon credits can allow funding from developed nations to be used to protect forests in developing nations. Another important reason to reduce biomass burning is to lessen the effect of the smoke in suppressing cloud formation and precipitation, resulting in a slow-down of the hydrological cycle (Andreae *et al.*, 2004). Figure 2.2 gives deforestation rates in the Brazilian Amazonia from 1980 to 2009, showing a strong reduction in the last five years – from 27 000 km^2 deforested in 2004 to 7 000 km^2 in 2009 – through Brazil's implementation of national policies. Similar reductions are happening in other tropical areas including Indonesia, and if global policies to reduce deforestation come into place, these reductions can be even more significant and may change the future picture of emissions quite rapidly. On the other hand, a future climate with higher temperatures and reduced precipitation and soil moisture may increase the incidence of forest fires.

Even though progress is being made in reducing biomass burning in many parts of the world, it can still have an important influence on sensitive ecosystems nearby, for example, springtime fires in the boreal forests of Europe, Central Asia and Siberia have been shown to contribute significantly to pollution in the Arctic (Quinn *et al.*, 2008). Similarly, the Himalayas are vulnerable to biomass burning in nearby regions of South and Southeast Asia (Ramanathan and Carmichael, 2008). In both cases, enhanced environmental damage can arise from BC deposition on snow and ice and associated influences on the local albedo.

Emissions from biomass burning, which can be substantial percentages of total emissions (e.g., 34 per cent of total global BC, 65 per cent of OC and 43 per cent of CO), are included in the natural emissions category presented in Table 2.1. As mentioned earlier, the amount of biomass burning likely to occur in future is sensitive not only to socioeconomic forces but also to future climatological changes that may affect water availability and soil moisture, which will in turn influence the amount of open biomass burning. Emissions from burning of agricultural residues may increase in the developing countries of Asia, for example, as the living conditions of farmers improve and pressure increases to have more crop cycles per year. For the purposes of this study, however, open biomass-burning emissions are assumed to remain constant between 2005 and 2030.

2.3.4 Uncertainties and issues in quantifying emission estimates

The degree of confidence in emission estimates varies widely depending on species, sector and region of the world. For some sources in some countries very little is known about activity levels and real-world emission factors, and choices for parameter values in such cases rely heavily on inferences of activity levels from quite limited and uncertain statistical information and extrapolations of emission factors from sources that have been measured in other parts of the world. Often, these are underestimated: many sources are uncounted in statistics and unmeasured sources tend to be poorer performers than tested ones. Additional factors influencing uncertainty in the estimation of emissions include fuel quality and the penetration and efficiency of abatement technology.

For global emissions of BC and OC, Bond *et al.* (2004) presented a formal uncertainty analysis with 95 per cent confidence intervals

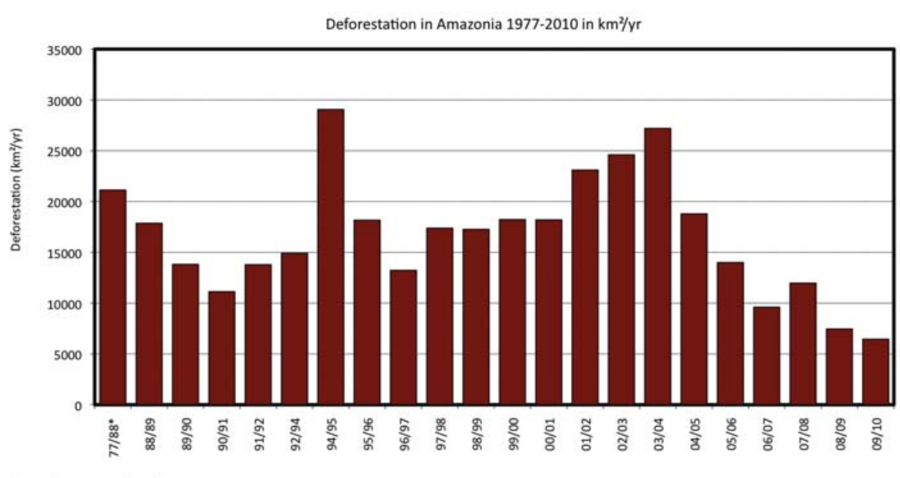

Figure 2.2. Annual deforestation rates in Brazil for the last 30 years, showing a strong reduction after 2004 (Artaxo, 2010).

for total global emissions. For BC emissions from contained combustion (essentially anthropogenic burning), the range was -30 per cent to +120 per cent. For BC from open biomass burning the range was -50 per cent to +200 per cent. For OC, the corresponding ranges were - 40 per cent to +100 per cent and -50 per cent to +130 per cent. The major source types contributing to uncertainty in anthropogenic BC emissions were identified as coke making, residential wood combustion, industrial coal combustion and on-road diesel vehicles. For anthropogenic OC, the largest contributors were residential wood combustion (by far), gasoline vehicles, and agricultural waste burning. Bond *et al.* (2004) showed that emissions are not predictable from overall stoichiometry (the amounts of fuel and air used in combustion). Rather they are determined by combustion kinetics and mixing. Moreover, carbonaceous aerosols are usually formed under poor combustion conditions, when insufficient oxygen is available for complete combustion. These conditions are generally associated with small, inefficient devices using poor-quality fuels, which are mostly, but by no means exclusively, to be found in developing countries. Residential cook stoves are a typical example of a combustion device that yields high emissions of carbonaceous aerosols; and if the extremely wide variation in stoves and their usage (stove type, fuel type and quality, fuel moisture content, oxygen availability, draft and ventilation conditions, temperature requirements, etc.) is taken into account, it should not be surprising that the uncertainty in emissions is high. The wide variation among emission factors reported from field and laboratory tests confirms this (e.g., Roden *et al.*, 2006, 2009).

Uncertainty in emissions of other species, including the precursors of tropospheric O_3 is generally not as high as for carbonaceous aerosols. As far as can be ascertained, there have been no formal estimates of uncertainty in global emissions of these species, such as are contained in the EDGAR inventory. Estimates for Asian gaseous emissions conducted for the NASA TRACE-P mission (Table 2.2) provide useful illustrations of the relative uncertainties in emissions by species and by level of economic development (Streets *et al.*, 2003a). The least uncertainty is associated with SO_2 (9–44 per cent) and CO_2 (7–91 per cent), both of which are to some extent constrained by the elemental concentrations in the fuels (sulphur and carbon). CO_2 can actually be an indicator of how well activity levels are known. Uncertainty in emissions of BC and OC were the highest by far (160–500 per cent for developing countries and 80–180 per cent for developed countries in Asia), mostly driven by high uncertainty in the emission factors of small sources like cook stoves, kilns and ovens, and some kinds of vehicles. NO_X, CO, CH_4 and NMVOCs are all subject to considerable uncertainty because of their sensitivity to combustion conditions, just like BC and OC; in addition, there are other, non-combustion contributions to CH_4 and NMVOC emissions that are hard to characterize. Emissions of NH_3 are largely associated with agricultural emissions, the sources of which are not easily quantified. Table 2.2 also shows the lower levels of uncertainty for developed countries like Japan (e.g., ±19 per cent for NO_X emissions), compared with the developing regions of South Asia (±63 per cent) and Southeast Asia (±92 per cent). This difference relates to variations in the availability and reliability of detailed activity statistics and the availability of measured emission factors (or lack of them). Discussion of uncertainties in Asian emissions was further developed by Zhang *et al.* (2009a) in the context of the updated TRACE-P emission inventory for the INTEX-B mission. They showed that incorporation of local knowledge for developed countries such as Japan and the Republic of Korea could reduce the uncertainties.

Uncertainties in the GAINS estimates of base-year emissions are similar to those reported in Table 2.2. Schöpp *et al.* (2005) discussed the uncertainties of emission estimates in the integrated assessment model RAINS, considering the uncertainties in the model parameters themselves. Overall, it was found that a typical range of uncertainties for modelled national emissions of SO_2, NO_X, and NH_3 in Europe lies between 10 and 30 per cent, which is consistent with the results presented in Table 2.2 for developed countries. In general, the uncertainties are strongly dependent on the potential for error compensation. This compensation potential is larger (and uncertainties are smaller) if calculated emissions are composed of a larger number of equal-sized source categories, where the errors in input parameters are not correlated with each other. Thus, estimates of the national total emissions are generally more certain than estimates of sectoral emissions. While the analysis of Schöpp *et al.* (2005) showed that the actual uncertainties are critically influenced by the specific situation – including the pollutant, year and region – the emission factor is an important contributor to uncertainty in estimates of historical emissions, while uncertainty in the activity data dominates the uncertainty in future emission estimates.

A quantitative assessment of uncertainty for future-year emissions was not performed for this Assessment. As discussed in Nakicenovic *et al.* (2000) and Streets *et al.* (2004), uncertainties associated with long-term global projections are best treated within a scenario context. This helps in the assessment of future developments in complex systems that are either inherently unpredictable or that have high scientific uncertainties. In all stages of the scenario-building process, uncertainties of different kinds are encountered. A large uncertainty surrounds future emissions and the possible evolution of their underlying driving forces, as reflected in a wide range of future emission paths in the literature. The uncertainty is further compounded in going from emission paths to formulating adaptation and mitigation measures and policies. In this Assessment a set of alternative scenarios was devised drawing on the two different developments of the energy system (reference and CO_2 measures scenarios from IEA, 2009) for which a number of control scenarios were prepared (Chapter 5). Hence, the alternative scenarios describe the range of possible future emissions.

2.3.5 Observational support for emission estimates

Present-day emission inventories of O_3 precursors can be verified by satellite and airborne and ground-based observational data. The estimation of emissions constrained by satellite observations is often called a top-down approach, as compared with the conventional bottom-up inventories based on activity data and emission factors. Further, *a priori* bottom-up emission estimates can now be assimilated with satellite data using inversion techniques to yield *a posteriori* estimates that are more consistent with observational data.

Table 2.2. TRACE-P uncertainty estimates (per cent) for Asian emissions (upper bound, +95 per cent confidence interval).

	SO_2	NO_X	BC	OC	CO_2	CO	CH_4	NMVOCs	NH_3
China	13	23	484	495	16	156	71	59	53
Japan	9	19	83	181	7	34	52	35	29
Other East Asia	12	24	160	233	13	84	101	49	31
Southeast Asia	27	92	257	345	91	214	95	218	87
India	26	48	359	544	33	238	67	149	101
Other South Asia	35	63	379	531	44	291	109	148	101
International shipping	44	56	402	402	40	72	72	204	–
All Asia	16	37	364	450	31	185	65	130	72

NO_X emissions

Much of the interest in verification of regional emissions of NO_X has focused on East China, where a rapid increase in emissions in recent decades is thought to have been a driver of the increase of O_3 in the northern hemisphere. From SCIAMACHY and OMI satellite data studies such as Ma *et al.* (2006), Akimoto *et al.* (2006), Zhang *et al.* (2007) and Lin *et al.* (2010), it can be concluded that, although emission inventories of fossil-fuel NO_X in China in the late 1990s to early 2000s could have been underestimated by more than 30 per cent due to errors in coal consumption statistics and other uncertainties, more recent bottom-up emission inventories of NO_X agree within 15 per cent with satellite-constrained estimates (Zhang *et al.*, 2009a). However, satellite data are able to provide more accurate spatial distributions of power plants and other anthropogenic and natural activities and therefore improve emission inventory mapping. For example, the OMI satellite products have been used to identify newly added NO_X emissions from new power plants in Inner Mongolia, China (Zhang *et al.*, 2009b).

CO emissions

Top-down approaches to constraining global CO emissions using satellite remote sensing data and surface observational data have long been pursued to validate inventory data and estimate their uncertainties. Several studies using inverse modelling techniques found that the earlier EDGAR inventory (Olivier *et al.*, 1996) appears to have underestimated sources for various regions (Kasibhatla *et al.*, 2002; Pétron *et al.*, 2004; Mueller and Stavrakou, 2005; Arellano *et al.*, 2006). Particularly, global inversion analyses using MOPITT satellite data found that sources in Asia and Africa are significantly underestimated by the EDGAR inventory. The inter-comparison study of Shindell *et al.* (2006) showed that forward models underestimate CO in the northern extratropics. More specifically, analyses of aircraft data downwind of Asia obtained in the TRACE-P campaign imply that emissions of CO from China were too low in the original TRACE-P emission inventory (Carmichael *et al.*, 2003; Allen *et al.*, 2004). Motivated by these studies, Streets *et al.* (2006) developed a new bottom-up inventory of CO emissions for China that are 36 per cent higher than the earlier work and give much better agreement between modelled and observed CO concentrations.

NMVOC emissions

Among the key precursors of tropospheric O_3, NMVOC emissions are difficult to validate by observational data, since continuous measurement of each NMVOC component on the ground is very limited and satellite data are not available for non-methane hydrocarbons (NMHC). Recently, however, satellite data using GOME, SCIAMACHY and OMI sensors for column distribution of formaldehyde (HCHO) (e.g., De Smedt *et al.*, 2008) have been utilized to verify NMVOCs from biomass burning and to estimate emissions of isoprene, the most significant precursor of HCHO from the biosphere. For example, Fu *et al.* (2007) found that the biomass-burning source for East and South Asia is almost five times the TRACE-P inventory estimate, while an estimate of anthropogenic reactive NMVOC emissions from China is only 25 per cent higher based on wintertime GOME observations. Generally, NMVOC emissions from biomass burning in Asia are greatly underestimated in emission inventories. Dufour *et al.* (2009) studied HCHO tropospheric columns from SCIAMACHY with the CHIMERE chemical transport model for Europe, and reported that the agreement between measurements and model was within 20 per cent on average.

Decadal ozone trends

Consistency between trends of observed tropospheric O_3 abundance and precursor emissions in the last couple of decades is very important for assessing the relative importance of reducing O_3 compared to long-lived greenhouse gases for climate-change mitigation up to 2030. Emissions of O_3 precursors generally increased until the end of the 1980s in the northern hemisphere. After the 1990s, emissions in Europe and North America started to decrease due to successful control measures – including stringent regulations in the USA (Monks *et al.*, 2009) – and remained stable or slightly decreased through the 2000s, consistent with long-term baseline O_3 measurements at Mace Head on the Atlantic coast of Ireland (Figure 2.3).

In contrast, emissions in Asia have been increasing rapidly since the 1980s, becoming comparable to emissions in Europe and North America in the 1990s, and have continued to increase in the 2000s, exceeding emissions from Europe and North America in the northern hemisphere (Akimoto, 2003). Figure 2.4, and similar data from other stations, confirms the increase in O_3 on the Asian continent.

Substantial amounts of surface O_3 trend data have been gathered in Europe, North America and Northeast Asia in the last couple of decades, and increasing trends are observed at most of the baseline sites in the northern hemisphere. Baseline observation sites in both Europe and North America typically show an increase of 0.3–0.4 parts per billion (ppb) per year of surface O_3 during the 1990s to early 2000s (Monks *et al.*, 2009). However, this rate of increase, which is more than 50 per cent of the rate observed in Northeast Asia, is too high to be ascribed to the precursor emission increase in Northeast Asia resulting from intercontinental transport. There is still significant uncertainty about the exact cause of these increases, which deserves further research.

2.3.6 Historical trends in emissions

BC emissions

There have been four studies to date that have attempted to reconstruct historical emission

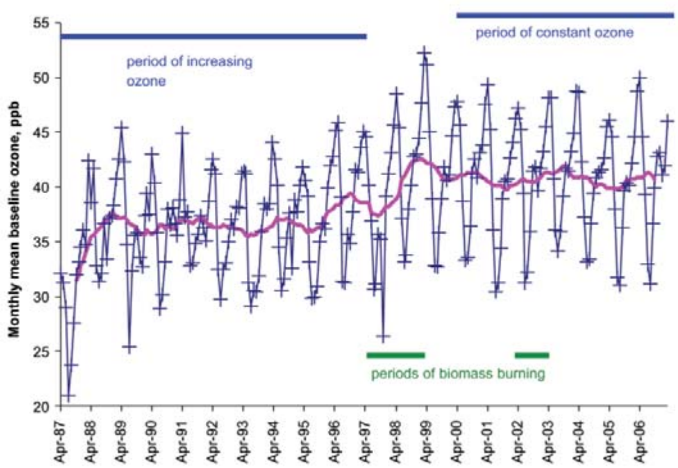

Figure 2.3. Time development of the monthly mean (+) and 12-month running mean (solid line) baseline O_3 mixing ratios from April 1987 at Mace Head (from Derwent *et al*., 2007).

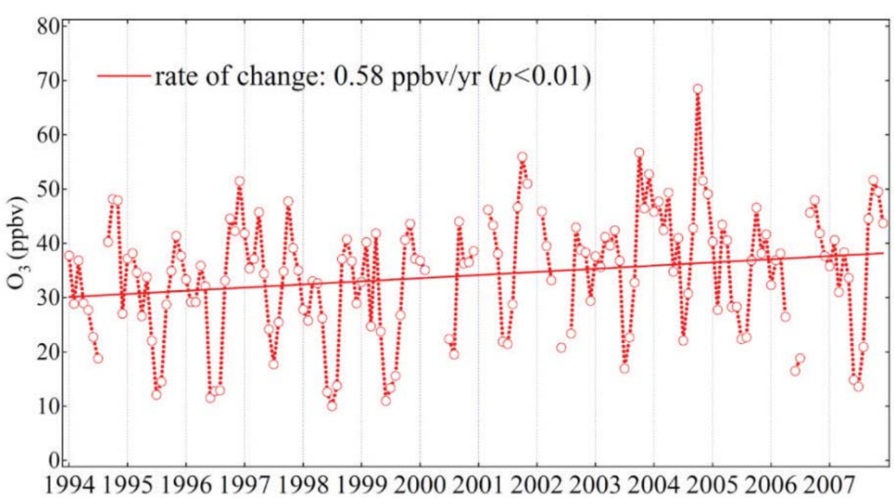

Figure 2.4. Monthly mean O_3 mixing ratios and the linear fit line at Hok Tsui, Hong Kong, during 1994–2007 (from Wang *et al.*, 2009).

inventories of carbonaceous aerosols. Bond *et al.* (2007) extended the earlier Bond *et al.* (2004) inventory for the year 1996 to the period 1850–2000 for biofuel and fossil-fuel combustion, building on a companion trend of global biofuel use for 1850–2000 (Fernandes *et al.*, 2007). Figure 2.5 compares the historical BC emission trends of Bond *et al.* (2007), Ito and Penner (2004) and Novakov *et al.* (2003). Qualitatively the trends of all three studies are similar. The major features are growth from the 19th century until about 1920, a period of little change from 1920 to about 1950, rapid increase from 1950 to about 1990, and then attenuation of the growth and perhaps a levelling off after 1990.

Quantitatively, however, the three studies show significant differences. The fossil-fuel trend of Novakov *et al.* tracks Bond *et al.* until 1950, after which it increases much more rapidly, ending up in 2000 at roughly double (~6 Mt/yr versus ~3 Mt/yr). This is attributed to the use by Novakov *et al.* of the emission factors of Cooke *et al.* (1999), which are now thought to be too high. On the other hand, the fossil-fuel BC trend of Ito and Penner agrees much better with Bond *et al.* in modern times, but is significantly lower in the early part of the 20th century. This might be due to the fact that Ito and Penner, for the most part, did not take into consideration the relatively poor technologies in the early time period, with much higher emission factors than today and no emission controls. The estimates of BC emissions from biofuel combustion of Bond *et al.* and Ito and Penner are quite similar. It should also be noted that Novakov *et al.* did not consider biofuel combustion.

Ozone precursor emissions

The radiative forcing of tropospheric O_3 from the pre-industrial era to the present is very much dependent on the pre-industrial concentration of O_3. Estimates of surface O_3 concentrations at the end of the 19th century are of the order of 10 ppb, but are based on extremely limited quantitative data or more widespread data that are only qualitatively reliable (Pavelin *et al.*, 1999). Such low values cannot be explained by current models which, when anthropogenic emissions of NO_X, CO, NMVOCs and CH_4 are excluded, give around 20 ppb or more (Gauss *et al.*, 2006). A large reduction in natural-source emissions in the current inventories is required in order to reproduce the observed O_3 abundance (Mickley *et al.*, 2001). Although there remains uncertainty in the difference in tropospheric O_3 abundance between the pre-industrial and present day, current estimates of radiative forcing, which are comparable with that of methane, are generally based on the converging estimates of multi-model studies overestimating the pre-industrial level (IPCC, 2007). This apparent overestimation does not, however, imply a similar underestimation of the response to modern emission changes, as the uncertainty stems from poor knowledge of 19th century emissions rather than poor understanding of the atmospheric processes governing O_3. Consistency between trends of observed tropospheric O_3 abundance and precursor emissions in the last couple of decades is very important for assessing the relative importance of reducing O_3 emissions alongside other greenhouse gases for climate-change mitigation over the next few decades.

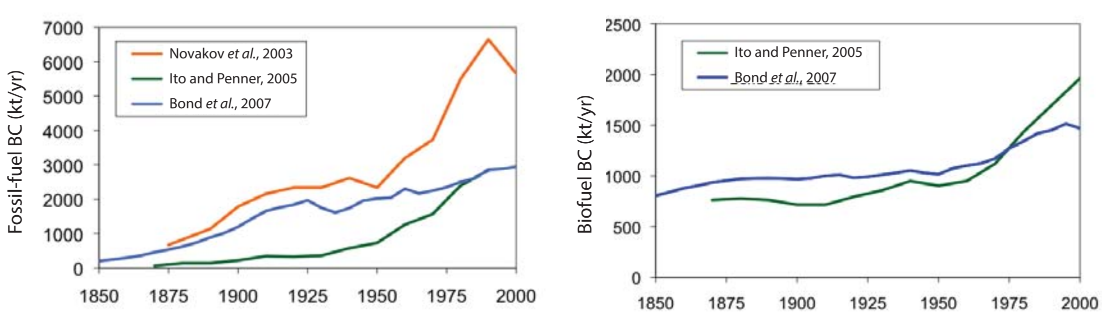

Figure 2.5. Historical BC emission trends from fossil-fuel (left) and biofuel (right) combustion from Novakov *et al.* (2003), Ito and Penner (2004) and Bond *et al.* (2007).

The most recent compilation of historical estimates of emissions of tropospheric O_3 precursors is the one developed for use in the chemistry model simulations that form part of the basis of the IPCC's fifth assessment report, as documented in Lamarque *et al.* (2010). This dataset is an historical reconstruction of global emissions back to 1850, normalized to year-2000 estimates. It does not develop new emission estimates from original data, but rather tries to take previously compiled trends and harmonize them. For BC and OC, the inventory of Bond *et al.* (2007) is used, and results are therefore identical to what was presented in the previous section. The historical trends for NO_X, CO, NMVOCs and CH_4 are developed from a combination of primary sources including the EDGAR-HYDE dataset (Olivier and Berdowski, 2001; van Aardenne *et al.*, 2001) and the RETRO dataset (Schultz *et al.*, 2008). Figure 2.6 shows the resulting trends for global anthropogenic emissions. Open biomass burning is not included. (The trends are rather smooth and do not contain the same level of temporal resolution as the Bond *et al.* inventory for BC). They all indicate growth in emissions from pre-industrial times until about 1990, when emission control measures in developed countries began to reverse the trend.

2.4 Development of emissions under the reference scenario

The GAINS reference scenario developed for this Assessment begins with the key macroeconomic assumptions and energy demand presented in the *World Energy Outlook 2009* reference scenario (IEA, 2009), taking into account existing air quality and climate-related policies that imply implementation of emission control measures. Projections cover the period to 2030.

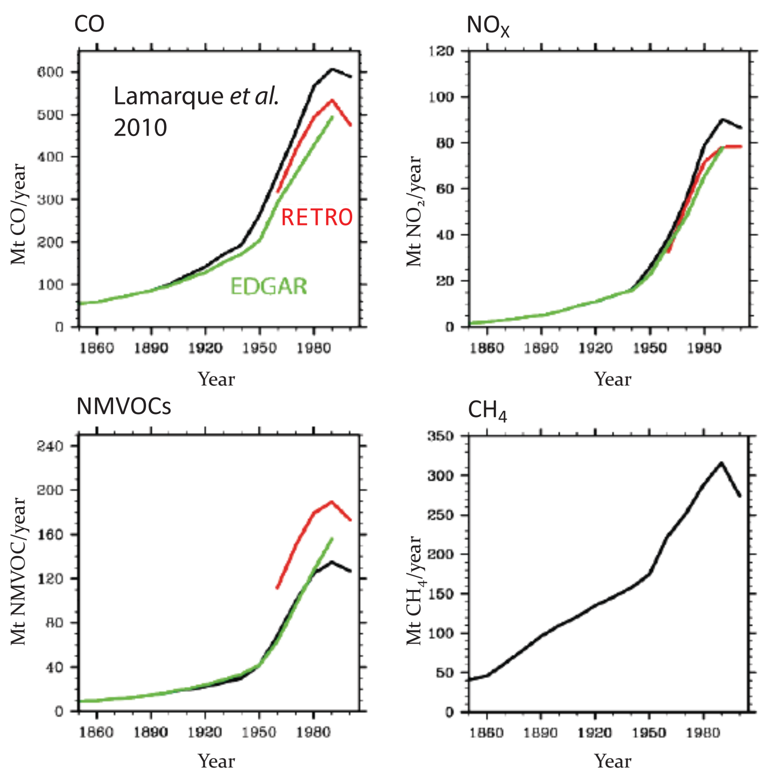

Figure 2.6. Historical trends in human-driven emissions of O_3 precursors from Lamarque *et al.* (2010), in black; RETRO (Schultz *et al.*, 2008; in red) and EDGAR-HYDE (Olivier and Berdowski, 2001; van Aardenne *et al.*, 2001) in green. Open biomass burning is not included.

2.4.1 Macroeconomic trends and energy demand

Figure 2.7 shows global development trends, differentiating regions that come under the Organisation for Economic Co-operation and Development (OECD) from those that do not. Global population is projected to grow by 1 per cent per year, from 6.6 billion in 2007 to 8.2 billion in 2030. Most of the increase will take place in Asia and Africa and will largely be in urban areas. Major drivers behind population projections are success in controlling the further spread of HIV, greater availability of treatment for AIDS and population aging.

Gross domestic product (GDP) projections consider the anticipated impacts of the global economic crisis of 2008–09. It is assumed that the rate of GDP growth recovers from its fall of 1.4 per cent in 2009 to reach 4.1 per cent in 2015, with an average of 3 per cent per year for the remaining period. The GDP of non-OECD countries grows significantly faster and is estimated to exceed that of the OECD region before 2020. Primary energy demand increases by 1.5 per cent per year on average between 2007 and 2030, with an overall increase of 40 per cent. The increase is driven mainly by development in China, India and the Middle East; overall, about 90 per cent of the increase occurs in non-OECD countries. However, their per capita energy consumption remains much lower than in the rest of the world. Fossil fuels continue to be the dominant source of primary energy supply to the world economy, representing 80 per cent in 2030, with oil and coal shares of about 30 per cent each. Coal is one of the most important fuels in industry, although the picture is different across regions. Power plants and industrial boilers in Northeast and South Asia are major coal consumers and coal use is expected to grow in the future. Consequently, production, conversion and transportation of fossil fuels will remain a significant activity with associated fugitive emissions of CH_4.

Transport and industry are the sectors with the highest fossil-fuel consumption, representing 62 per cent of global demand in 2030. Figure 2.8 shows regional development in the road transport sector. (See section 2.5 for a full description of these regions.) Global fuel consumption is shown to rise by 46 per cent between 2005 and 2030, driven by growth in the developing world; however, fuel use continues to be high in North America and Europe, and is approximately equal to that of Asia in 2030. Growth in fuel use in the OECD region is expected to be marginal due to enforcement of new fuel economy standards, increased penetration of smaller vehicles, and new engine technologies such as hybrid cars, which are projected to account for about 6 per cent of the global car fleet. It is important to note that the share of diesel fuel (an important source of BC emissions) grows from 39 per cent in 2005 to 46 per cent in 2030.

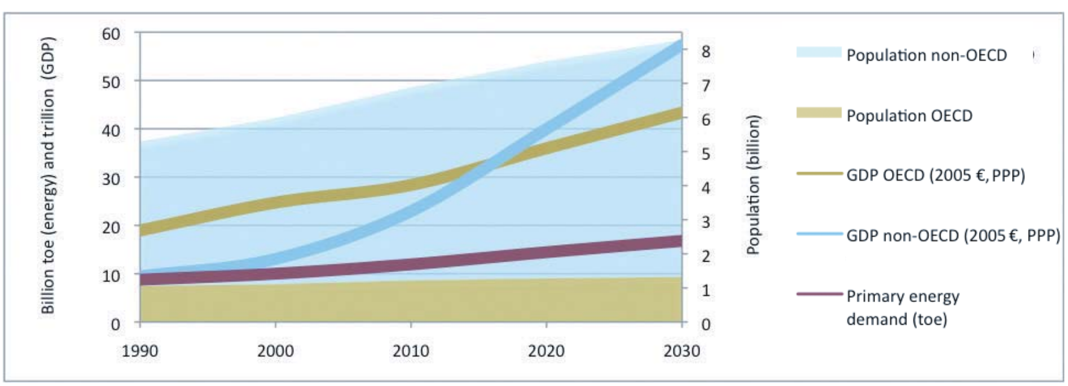

Figure 2.7. Key macroeconomic assumptions for the OECD and non-OECD regions and world primary energy demand (IEA, 2009).
Note: *PPP* = purchasing power parity; *toe* = tonnes of oil equivalent.

Solid fuel use (i.e., biofuel and coal) in the residential sector remains fairly constant during the period 2005–30, although there are differences between regions (Figure 2.9). Biomass, mostly used in the developing world in cooking and heating stoves, represents nearly 90 per cent of total domestic use of solid fuel, and is expected to grow slightly in all regions except Northeast Asia. Coal use in the residential sector is projected to decline by about 4 per cent, mostly in Northeast Asia and Europe.

Livestock breeding and crop production are associated with nearly half of CH_4 and over 80 per cent of NH_3 emissions. Growth in agricultural production is strongly linked with population and economic growth, the latter affecting lifestyles and leading to, for example, increased meat consumption. Most recently, greater demand for biofuels has resulted in additional growth in this sector. Agricultural data used in this study originate from global (Bruinsma, 2003; Alexandratos *et al.*, 2006; FAO, 2010; IFA, 2003; Döring *et al.*, 2010), regional (EFMA, 2010; Klimont, 2005) and national (Fischer *et al.*, 2010; Amann *et al.*, 2008) statistics, outlooks and projects. Figure 2.10 shows changes in selected key indicators in the reference scenario. All the activity growth is expected in non-OECD countries, as demonstrated in the value added (VA) forecast. While growth is moderate for dairy cattle, demand for meat leads to strong growth in beef production.

2.4.2 Air pollution legislation in the reference scenario

The GAINS reference scenario assumes successful implementation of current air pollution control policies (i.e., policies that were in force or in the final stage of the legislative process as of mid-2010 in each country). The most influenced sectors are transport, power generation and industrial production. The degree and stringency of regulation varies across regions, however; strongly controlled sectors tend to converge in the longer time horizon to a comparable implied emission rate largely independent of the region. At the same time, it is assumed that there will be changes that are autonomous from legislation, such as the transition from traditional heating and cooking stoves to improved and clean stoves through turnover of the equipment, and the introduction of new vehicles with better fuel efficiency. However, the effects of such improvements are often only seen towards the end of the modelling horizon owing to the long lifetime of specific installations – such as heating stoves or off-road machinery – or other con-

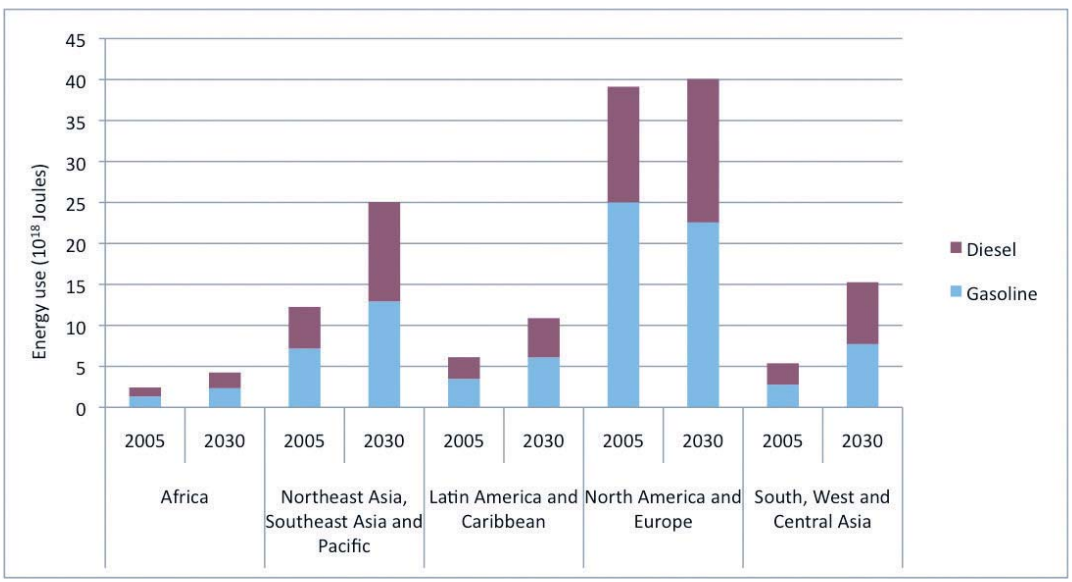

Figure 2.8. Use of diesel and gasoline fuel in the road transport sector in 2005 and 2030 in the reference scenario.

straints including a lack of infrastructure or economic stimulus in specific regions.

Implementation of national air pollution legislation, fuel efficiency standards, installation-specific emission limit values, and international laws and agreements such as the Convention on Long-range Transboundary Air Pollution (CLRTAP), implies changes in emission factors over time. These changes will vary across regions and sectors, reflecting the stringency of legislation. One additional aspect of environmental legislation is the actual level of compliance, both in terms of the timely introduction of the laws (emission standards) and the actual performance of the control equipment. For projections in this study, it has been assumed that the technical abatement measures will be installed in a timely manner and will achieve the emission levels required to comply with the law. The only exception is the transport sector, where high-emitting vehicles (or super-emitters) are explicitly considered and assumptions are made about their region-specific shares and technology-specific deterioration factors. Figure 2.11 shows examples

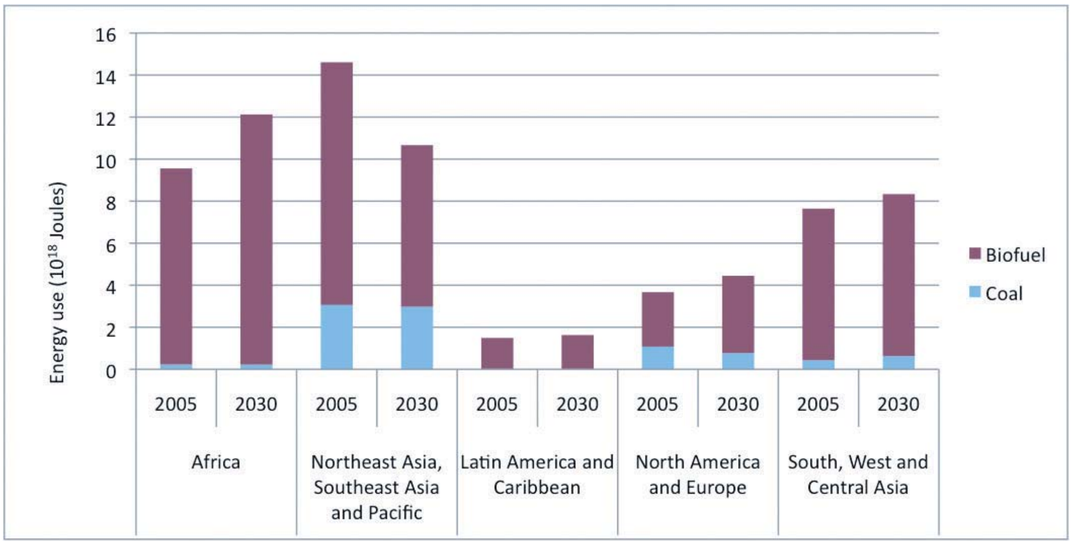

Figure 2.9. Use of solid fuels in the residential sector in 2005 and 2030 in the reference scenario.

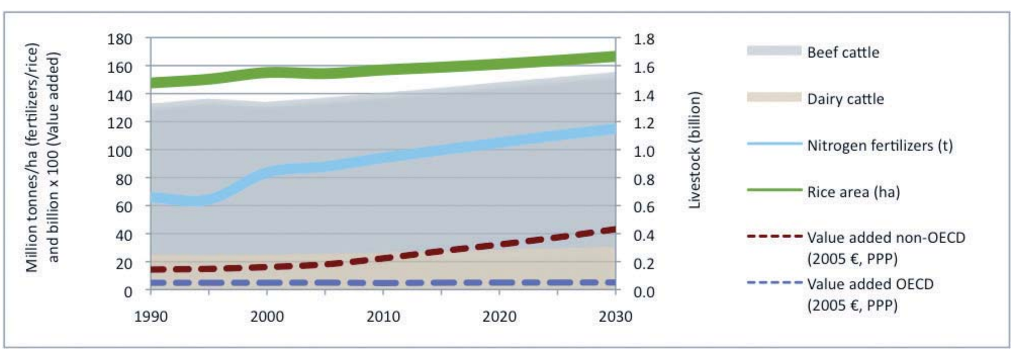

Figure 2.10. Key reference scenario developments for the agriculture sector.
Note: *PPP* = purchasing power parity.

of how implied emission factors change over time in China and India in selected sectors. The emission factors presented in this figure do not explicitly consider super-emitters.

For transport sources and non-road machinery, the reference scenario includes implementation of the Euro-standards, the US federal emission standards, the Japanese emission standards, and their fuel quality requirements. Other countries have adopted one or more of the above standards, mostly the European ones, with a somewhat delayed implementation stage (e.g., DieselNet, 2010; CAI, 2008). Assumptions about current and future emission controls in the power-plant sector have been cross-checked with detailed information from the database on world coal-fired power plants (IEA CCC, 2010) as well as collaboration with national expert teams, including from Asia (Klimont *et al.*, 2009), and various literature sources (e.g., Xu *et al.*, 2009; Zhang *et al.*, 2009a; Smith et al., 2011). Although several countries have emission standards for cooking and heating stoves and boilers, most of the solid fuels used in this sector are in the developing world where such legislation is lacking, and many of these traditional appliances are expected to remain in use until 2030.

2.5 Reference scenario emissions of BC, OC and O_3 precursors from key sectors and regions

The emissions described here are mainly derived from the GAINS model for the years 1990, 2005 and 2030 (see Sections 2.3.1 and 2.4). Global NH_3 and NMVOC emissions in developing countries originate from the CIRCE project, based on EDGAR v4.1, as documented in Döring et al. (2010). These global emissions are disaggregated into five regional groupings based on UNEP's sub-regional geographical breakdown (available at http://geodata.grid.unep.ch/extras/geosub-regions.php.) (Figure 2.12) plus an additional category concerning emissions from international shipping and aviation. Global emissions are also disaggregated into nine key anthropogenic emission source sectors:

1. Large-scale combustion (including power plants and industrial boilers);

2. Industrial processes (including cement, lime and brick kilns);

3. Residential-commercial combustion;

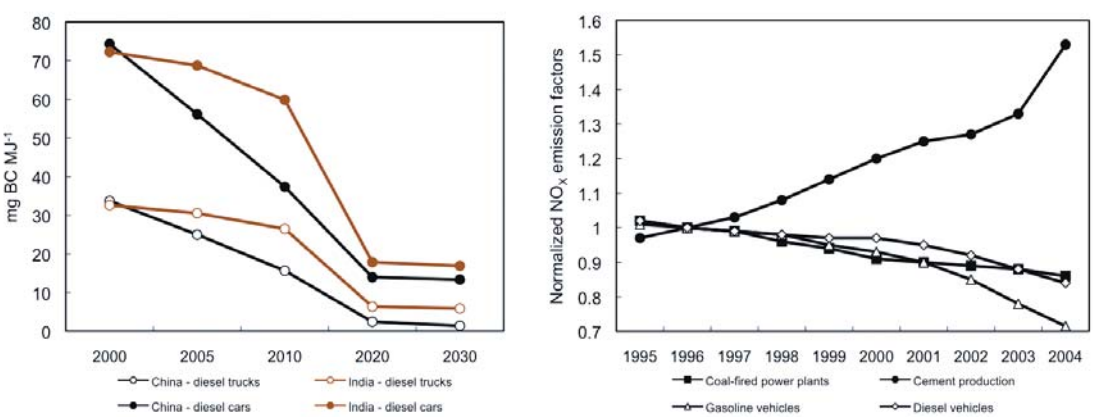

Figure 2.11. Examples of changes in emission factors over time. Left: transport-related BC in China and India (Klimont *et al.*, 2009). Right: NO_X (relative change to 1996) in several sectors in China (Zhang *et al.*, 2007).

4. Transport (excluding international shipping and aviation);

5. Fossil fuel extraction and distribution (including coal, oil, gas production, storage and distribution);

6. Waste/landfill (including open garbage burning);

7. Agriculture (including open burning of crop residues);

8. Solvent use; and

9. International shipping and aviation.

Emission densities vary regionally, depending on many factors such us structure of fuel use, population density, economic development and air quality legislation. Consequently, they are expected to change over the modelling time horizon. Figure 2.13 illustrates the spatial distributions of BC and NO_X emissions in 2005 and in the 2030 reference scenario, accordingly. In general, the regional trends for both pollutants are quite similar, with declining emissions in North America and Europe and increases projected in Asia. However, while BC emissions in Northeast Asia show a decline, this region becomes the largest emitter of NO_X. A more detailed analysis of regional and sectoral trends is included later in this chapter. Shifts in the spatial distribution of emissions over time are important, because emissions from different locations cause different amounts of radiative forcing (Chapter 3). Figure 2.13 also includes international shipping emissions, which are not significant for BC emissions but represent a sizable part of NO_X and are expected to increase by 2030.

Emissions of NMVOCs, NH_3 and total $PM_{2.5}$ are discussed only in section 2.5.1. NMVOCs and NH_3 are presented in Figures 2.14 and 2.15, but because they are of lesser importance in the modelling and mitigation sections of this Assessment, they are not discussed thereafter. Total $PM_{2.5}$, which includes all primary emitted particles (BC, OC, sulphates, metals and minerals) but no secondary particles, is very relevant to human health and is further discussed in Chapter 4, while in this chapter emissions of the radiatively active components of primary particulate matter, BC and OC, are stressed. All 10 of the species presented in the following section are intimately connected and have their own unique suite of environmental effects. In addition, each source category emits a variety of species, and therefore a mitigation measure directed at a single one will simultaneously affect co-emissions. This may alter the benefit balance, as discussed at length in Chapter 5. Emissions from vegetation fires (including for-

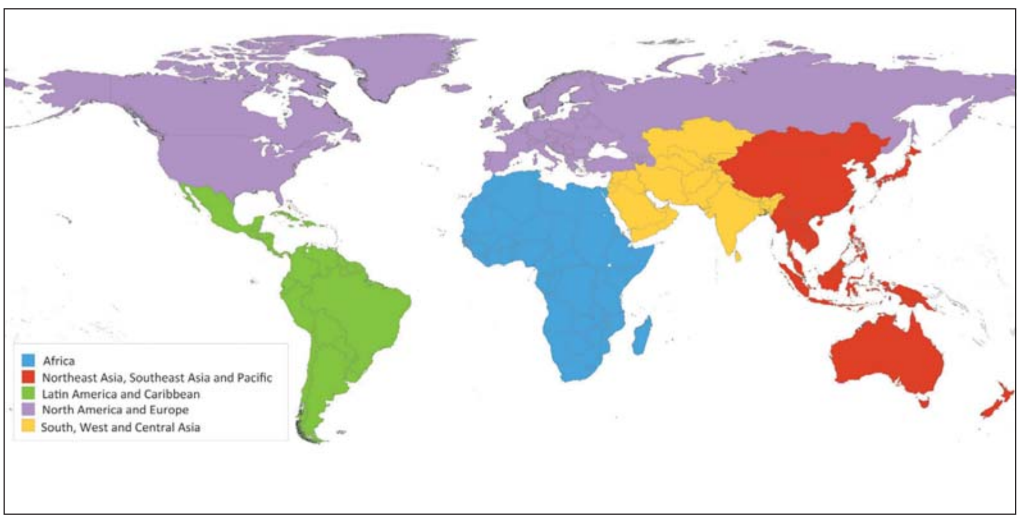

Figure 2.12. The regional groupings used in this analysis.

est and savannah burning) and natural sources are not covered in this section.

2.5.1 Sectoral/regional shares

The regional shares of global anthropogenic emissions for 2005 are shown in Figure 2.14. The Northeast Asia, Southeast Asia and Pacific region represents the largest source of emissions, the majority of which (60–80 per cent) originate in China (data not shown), except for NO_X and CO_2, where North America and Europe are responsible for the greatest share of global emissions. For BC, the next most important regions are North America and Europe, and South, West and Central Asia (of which 63 per cent is from India), each emitting almost a fifth of the global total. The second largest contribution for SO_2, CO and CH_4 comes from North America and Europe, while for OC it is Africa and for total $PM_{2.5}$ it is South, West and Central Asia (almost 70 per cent of both pollutants coming from India). Africa contributes only a small

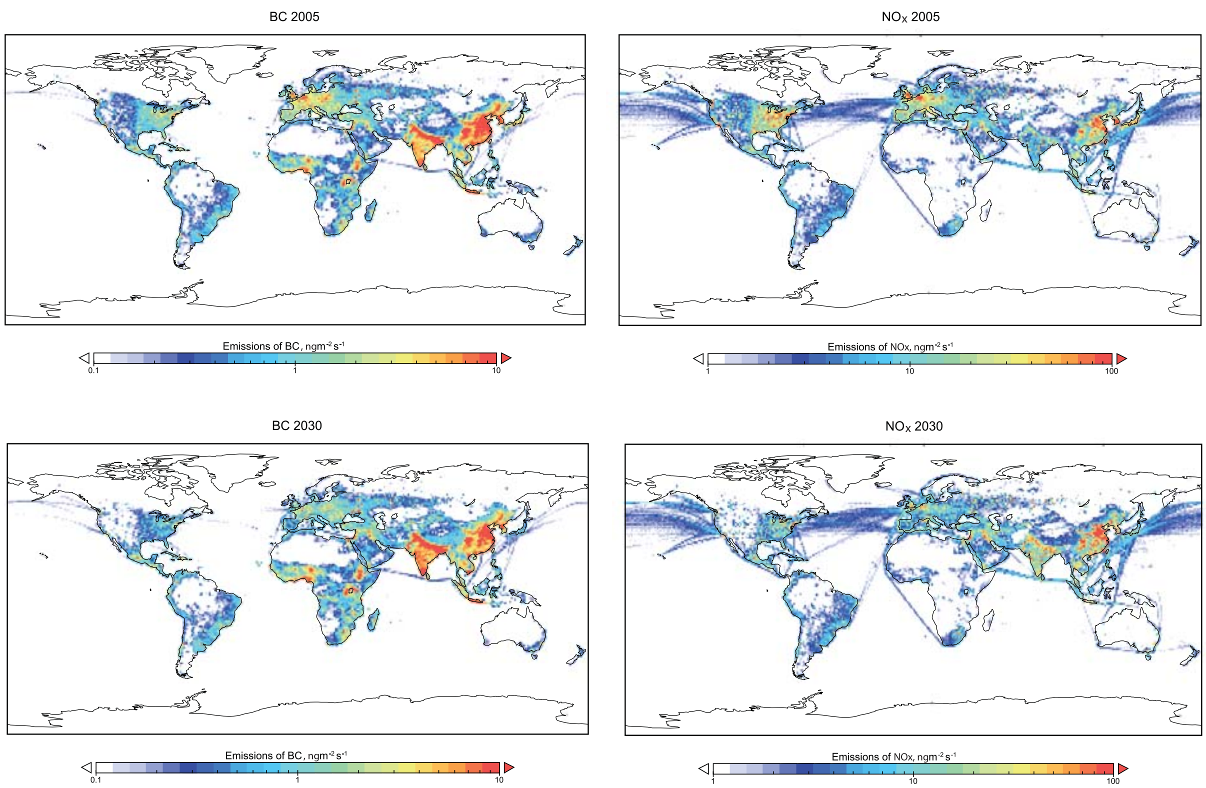

Figure 2.13. Spatial distribution of BC and NO_X (as N) emissions in 2005 and the 2030 reference scenario. Note the difference of scale between BC and NO_X emissions. The GAINS emissions were gridded on a 0.1° × 0.1° grid by the EDGAR team at the European Commission's Joint Research Centre, Ispra, Italy.

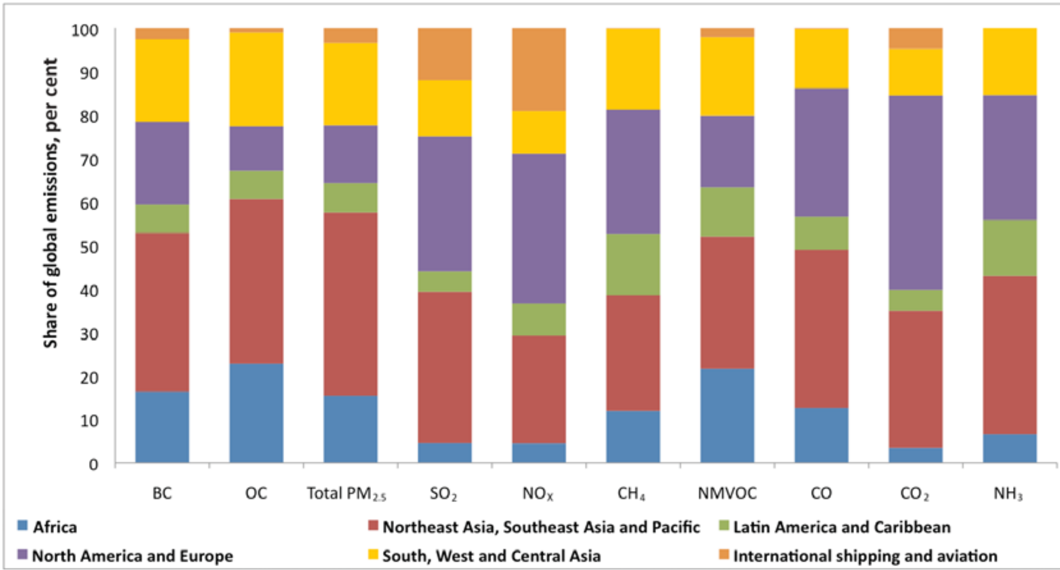

Figure 2.14. Regional shares of global anthropogenic emissions for 2005.

percentage of emissions associated with transportation and industrial activities, but ranks high for pollutants originating from incomplete combustion (traditional cook stoves), i.e., BC, OC, NMVOC, CO and total $PM_{2.5}$.

For each pollutant, the percentage shares of global emissions for 2005 represented by each of the nine key sectors are shown in Figure 2.15; in this figure transport does not include emissions from international shipping and aviation, which are in a separate category. Primary carbonaceous aerosols (BC and OC) and total $PM_{2.5}$ emissions originate predominantly in the residential-commercial sector, which also contributes significant shares of NMVOCs and CO, consistent with the emission profiles of incomplete

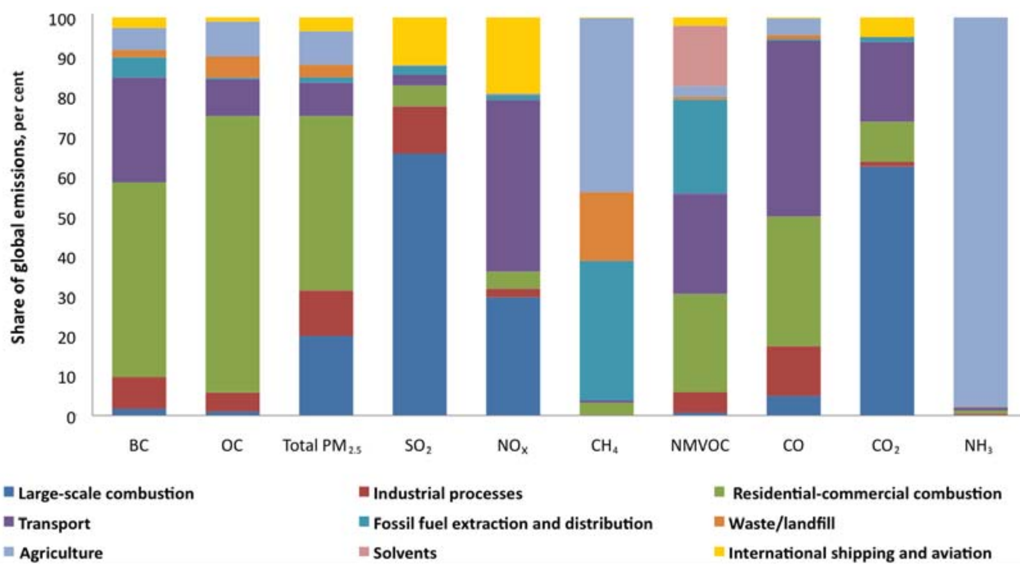

Figure 2.15. Sectoral shares of global anthropogenic emissions for 2005.

combustion sources. The large-scale combustion sector is responsible for the majority of CO_2 and SO_2, as well as contributing nearly 30 per cent of NO_X and 20 per cent of total $PM_{2.5}$, while for other pollutants it does not play a significant role. Emissions from the transport and international shipping and aviation sectors taken together are key for NO_X and CO (45–60 per cent), and contribute about 25–30 per cent of CO_2, NMVOCs and BC. The agriculture sector accounts for the vast majority of NH_3 emissions and is the main source of CH_4; for other pollutants its contribution is linked with open burning of agricultural residues, which makes up almost 10 per cent of total OC. Fossil fuel extraction and distribution is a significant source of CH_4 and NMVOC emissions, also contributing about 5 per cent of BC, which is associated with oil and gas flaring. The waste/landfill sector represents about 15 per cent of CH_4 emissions, while solvents account for a similar share of NMVOCs.

2.5.2 Global trends

Figure 2.16 presents global emission trends by world region for the seven major pollutants that are of special relevance to modelling and mitigation analysis. Emissions of BC, OC, CO and NO_X declined in North America and Europe due to implementation of strict air pollution control policies, especially in the transport, industry and power plant sectors, as well as the economic restructuring of Eastern Europe at the beginning of the 1990s. Emissions rose, however, between 1990 and 2005 in the other regions due to increasing activities and less ambitious implementation of emission controls. This resulted in slight net increases in global emissions of these four pollutants. Conversely, net global emissions of SO_2 declined between 1990 and 2005, mainly due to the large reduction in North America and Europe caused by end-of-pipe emission abatement and reductions in the sulphur content of fuels. These measures offset increased emissions from South, West and Central Asia. From 1990 to 2005, CO_2 emissions increased in all regions apart from North America and Europe where they remained relatively unchanged; this trend is projected to continue up to 2030. Emissions of BC, OC and CO, however, are projected to decrease between 2005 and 2030 in both Northeast Asia, Southeast Asia and Pacific and North America and Europe due to further air pollution legislation, resulting in slight net global decreases despite increases in most other regions. Emissions of CH_4 and NO_X are expected to grow between 2005 and 2030 in all regions apart from North America and Europe, resulting in a net

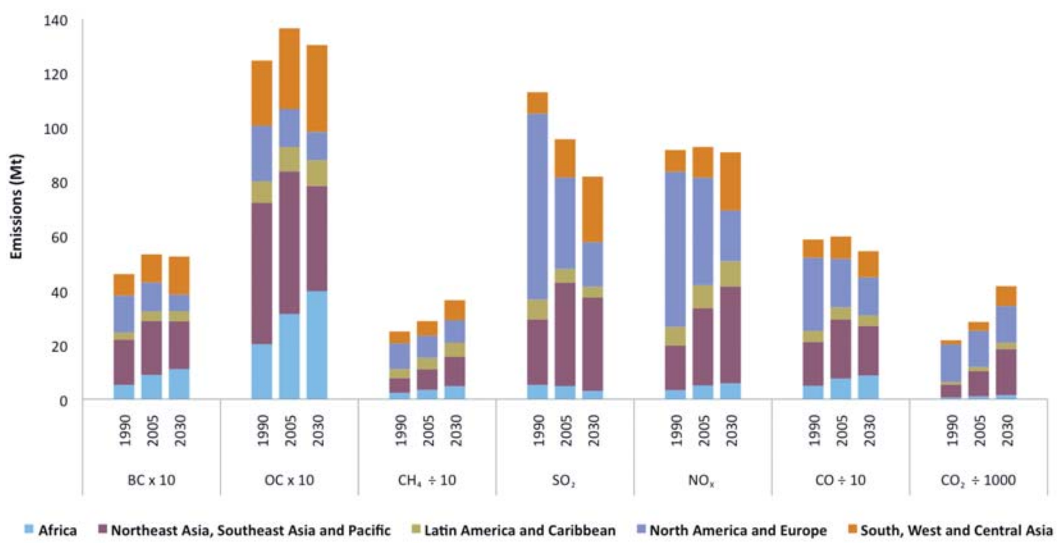

Figure 2.16. Regional splits of global emission trends for the major emitted pollutants.

global increase in the case of CH_4 and a slight decrease for NO_X.

For BC, OC and NO_X, Figure 2.17 does not show much change in the sectoral shares over time. For CO, however, there was a declining contribution from transport between 1990 and 2005, with this trend set to continue up to 2030. The transport sector has experienced a step-wise introduction of emission abatement technologies which will continue to reduce emissions as a result of equipment turnover. In some regions the emission benefits of further controls are offset by increases in activity levels. Over the same period, CO emissions from the residential-commercial sector remain static, whilst those from the other three sectors show a rising trend. For CH_4, the increased global emissions between 1990 and 2005 are largely due to fossil fuel extraction and distribution, and this trend is projected to continue up to 2030. The decline in total SO_2 emissions from 1990 to 2005 was mainly caused by reduced emissions from large-scale combustion and the residential-commercial sector, with a further decrease in emissions from large-scale combustion driving the ongoing reduction in global SO_2 emissions projected to 2030. The decline is a result of implementation of end-of-pipe control technologies, as well as fuel-related changes – mainly switches to lower sulphur content and away from solid fuels. Large-scale combustion was also largely responsible for the increase in CO_2 emissions between 1990 and 2005, with the further increase projected to 2030 also driven by emissions from this sector.

2.5.3 Sectoral trends

Large-scale combustion (power plants and industrial boilers)

Carbon dioxide emissions from this sector (Figure 2.18) are a good illustration of expected growth in fossil-fuel use, especially in Asia where they increase nearly fourfold. At the same time, the trends for key air pollutants (SO_2 and NO_X) indicate increasing implementation of abatement technologies and fuel quality improvement. This figure also shows the relatively small emissions of BC, OC and CH_4 in comparison with other pollutants. SO_2 emissions from large-scale combustion declined from 1990 to 2005 due to large reductions in emissions from North America and Europe outweighing rising trends in Northeast Asia, Southeast Asia and Pacific (mainly China) and, to a lesser extent, in South, West and Central Asia (mainly India). The major drivers for SO_2 emission reductions in North America and Europe have been

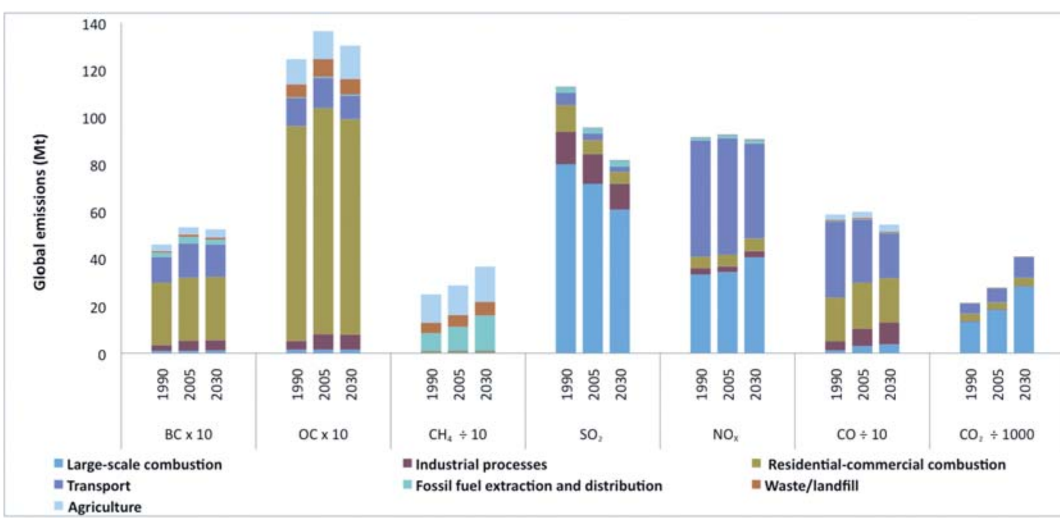

Figure 2.17. Sectoral splits of global emission trends for the major pollutants.

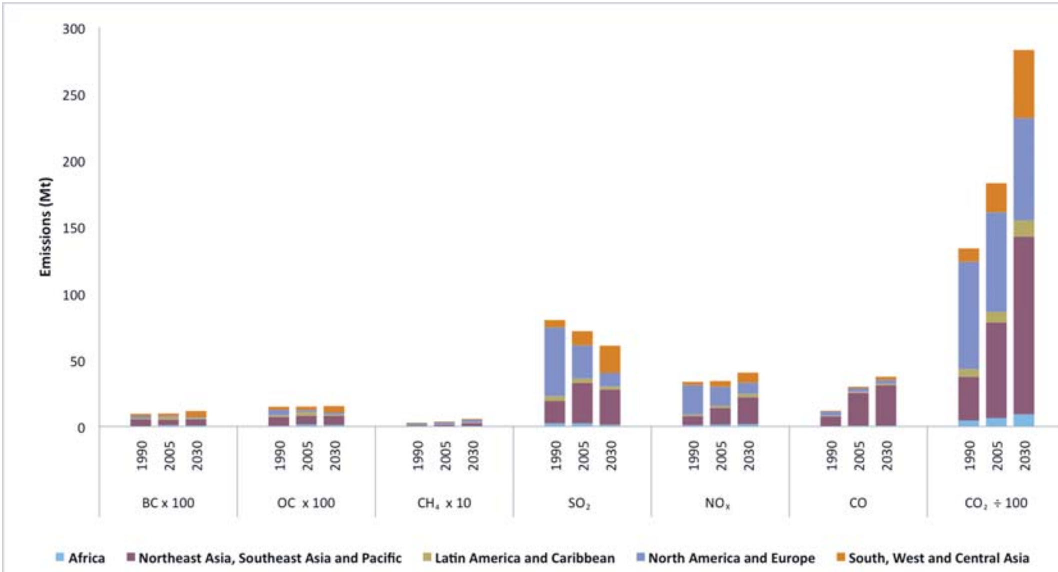

Figure 2.18. Trends in emissions from large-scale combustion.

efficient end-of-pipe measures accompanied by fuel switching and a reduced sulphur content in fuels. This overall declining trend in SO_2 emissions is set to continue to 2030 owing to sustained reductions in North America and Europe combined with more modest reductions in all other regions apart from South, West and Central Asia, where SO_2 emissions are actually projected to increase significantly due to expanded coal use without effective emission controls. For NO_X, overall emissions in 2005 were similar to those in 1990, with a marked decrease for North America and Europe being balanced by increases for all other regions. By 2030, the continued reduction in NO_X emissions projected for North America and Europe is more than offset by increases in all other regions, resulting in a net global increase for this sector. The increase is not, however, as large as in the case of CO_2, indicating that effective implementation of emission controls decouples emission rates from growth in fuel use. Global emissions of CO increased sharply between 1990 and 2005, largely due to rising emissions from coal combustion in Northeast Asia, Southeast Asia and Pacific. Emissions are projected to increase until 2030, but at a lower rate owing to better efficiency of new power plants.

Industrial processes

Globally, emissions from industrial processes are of relatively minor importance for the pollutants under discussion. Emissions of BC and OC in the study period are dominated by Northeast Asia, Southeast Asia and Pacific, and South, West and Central Asia; for CO, North America and Europe follows Northeast Asia, Southeast Asia and Pacific (Figure 2.19). Global emissions of these three pollutants almost doubled from 1990 to 2005. Major sources in these areas are brick- and coke-making (especially for BC and OC), which use rather primitive and low-cost production technologies without emission controls. Whereas BC and OC emissions are projected to stabilize (the reference scenario assumes partial replacement of the most inefficient technologies), the rising trend continues for CO to 2030 with increased emissions in all regions. Global SO_2 emissions declined slightly from 1990 to 2005 due to decreased emissions in regions other than Northeast Asia, Southeast Asia and Pacific (China represented 67 per cent of the regional total in 2005 and was responsible for most of the growth) and South, West and Central Asia. The gradual decline in SO_2 emissions globally is projected to continue to 2030, even though

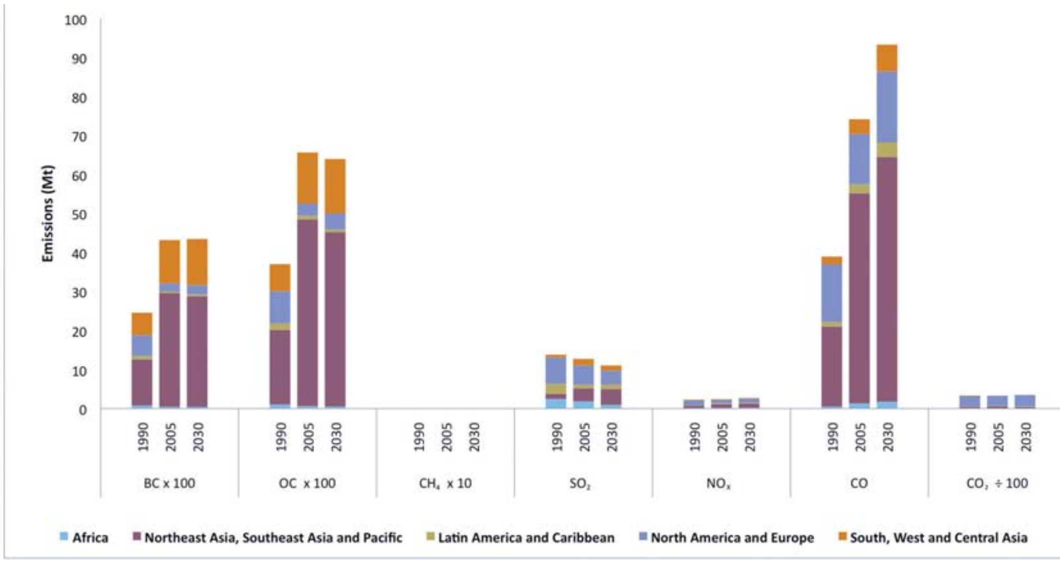

Figure 2.19. Trends in emissions from industrial processes.

emissions from Northeast Asia, Southeast Asia and Pacific (again, mostly from China) continue to increase due to high growth in activity. Emissions of CH_4, NO_X and CO_2 from this sector are relatively insignificant.

Residential-commercial combustion

This sector is the most important contributor to BC, OC and CO emissions. Emissions of all pollutants remain fairly constant over the whole modelling horizon (Figure 2.20), although regional shares change over time. The stability in emissions in this sector is explained by the rather constant global fuel use and, in the absence of regulation, only small switches towards improved and cleaner stoves. Patterns of variation in regional emissions of all species are strikingly similar owing to similarities in the emission profiles of key polluting installations, and consequently reflect fuel-use changes. Three regions – Africa, Northeast Asia, Southeast Asia and Pacific, and South, West and Central Asia – tend to dominate emissions of BC, OC, CO and CH_4, with decreases in Northeast Asia, Southeast Asia and Pacific and increases in Africa tending to cancel each other out, reflecting changing fuel-use patterns. Emissions of SO_2, NO_X and CO_2 from this sector are relatively insignificant. In the case of CO_2 this is because emissions from biomass combustion are regarded as carbon neutral, that is to say, do not lead to a net increase in atmospheric CO_2 concentrations; it is acknowledged that this may not be true for every situation.

Transport

The study period covers a timeframe of strict air pollution control policies for on-road transport and to some extent also for off-road transport. This can be seen clearly in the development of emissions, especially in North America and Europe, where transport volumes remained rather constant or increased only slightly, while emissions of all pollutants, except BC, declined by 2005 (Figure 2.21). For BC, the increasing shares of diesel vehicles offset improvements in engine technology and reductions achieved in gasoline vehicles, as shown in Figure 2.8 and the accompanying text. Existing legislation for new vehicles results in a significant reduction in emissions from diesel vehicles, including BC and NO_X, and is expected to bring a substantial reduction in emissions by 2030, especially in North America and Europe. The other regions are adopting similar legislation to that of North America and Europe, but with delayed implementation timetables. This also applies to off-road transport, for which standards are not as strict as for the on-road sector. Increased emissions in other regions are mainly a result of dramatic increases in traffic volumes, particularly in Asia. Despite

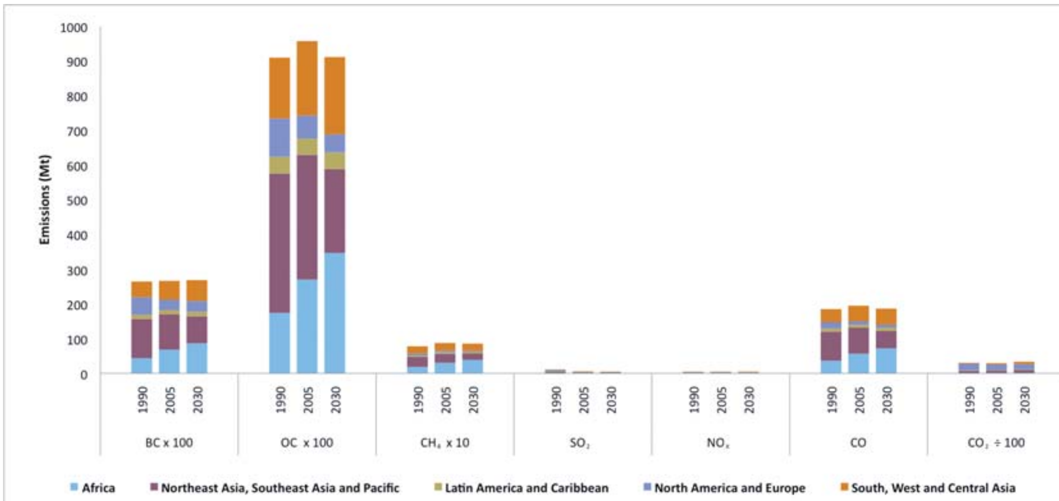

Figure 2.20. Trends in emissions from residential-commercial combustion.

some improvements in fuel economy, trends in transport activities can be seen in global emissions of CO_2, which increased from 1990 to 2005 and are projected to continue rising up to 2030. With the exception of international shipping, SO_2 is a relatively minor component of emissions from the transport sector because of fuel standards requiring the use of low-sulphur fuels.

International shipping emissions represent a relatively high share of global NO_X and SO_2 emissions, comprising 23 and 14 per cent of the anthropogenic and 15 and 11 per cent of total global emissions, respectively. The corresponding shares for BC are lower, at 3 per cent and 2 per cent respectively, and for the other pollutants are less than 1 per cent. Within the transport sector, international shipping contributed over 80 per cent of SO_2, 30 per cent of NO_X, 10 per cent of BC and 9 per cent of OC emissions in 2005. For CO the contribution is less than 1 per cent. About 80 per cent of the international shipping traffic occurs in the northern

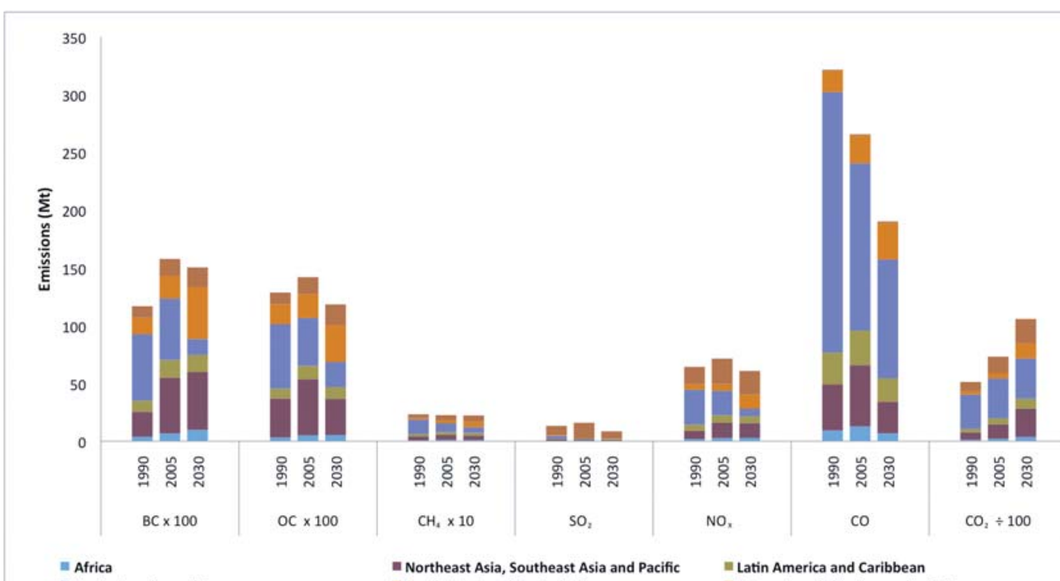

Figure 2.21. Trends in emissions from the transport sector.

hemisphere, with 32 per cent in the Atlantic, 29 per cent in the Pacific, 14 per cent in the Indian Ocean and 5 per cent in the Mediterranean (Eyring *et al.*, 2010). However, increasing Asian shipping activity may change the distribution in the future (Eyring *et al.*, 2010). At present, Arctic shipping contributes 1–3 per cent to global shipping emissions, depending on the pollutant, but its proximity to sensitive ecosystems might make it a much more important source locally in the future, especially when an extended period of ice-free Arctic waters is considered in future scenarios (Corbett *et al.*, 2010). Global annual growth of international ship traffic is estimated to be 2–3 per cent, depending on the scenario (Buhaug *et al.*, 2009), and annual growth in fuel use could be about 2 per cent (Eyring *et al.*, 2010). However, recent regulations of the International Maritime Organization (IMO) address emissions of SO_2 and NO_X, and some changes are expected for other pollutants as well. Since part of the fuel sulphur is emitted as particulate sulphate, the reductions may also affect primary PM emissions, including BC and OC (Buhaug *et al.*, 2009; Lack *et al.*, 2009). In the RCP 8.5 scenario, the average compound annual growth rates (CAGR) for global shipping emissions between 2005 and 2030 are 0.83–0.87 per cent for BC, OC, CH_4, CO and NMVOC. The NO_X annual growth rate is smaller at 0.44 per cent, and annual SO_2 emissions decline by 2.5 per cent on average; however, a significant decline is expected in the period between 2010 and 2020, following implementation of the IMO regulation.

Fossil fuel extraction and distribution

In 1990, North America and Europe accounted for more CH_4 emissions than any of the other four regions (Figure 2.22). By 2005, a fall in emissions from North America and Europe was more than offset by a rise in emissions from the other regions, producing an overall net increase; CH_4 emissions from Northeast Asia, Southeast Asia and Pacific (of which China accounted for over 80 per cent, mainly from coal mining) increased to equal those from North America and Europe. The projected global increase of almost 40 per cent by 2030 is mainly due to rapidly growing emissions from Northeast Asia, Southeast Asia and Pacific, driven strongly by growth of the Chinese coal-mining industry (see Figure 2.28), although increases also occur in the other four regions.

Waste/landfill

Emissions from this sector are dominated by CH_4. Global CH_4 emissions from waste/landfill increased by 10 per cent between 1990 and 2005 and are projected to increase by 20 per cent by 2030 (Figure 2.23). North America and Europe is the largest emitting region in all three years, although their CH_4 emissions actually decline over the period, the overall rising trend being driven by growth in emissions from the other four regions. For OC, global emissions increased from 1990 to 2005 and are projected to decrease slightly by 2030, although not back down to 1990 levels. For this

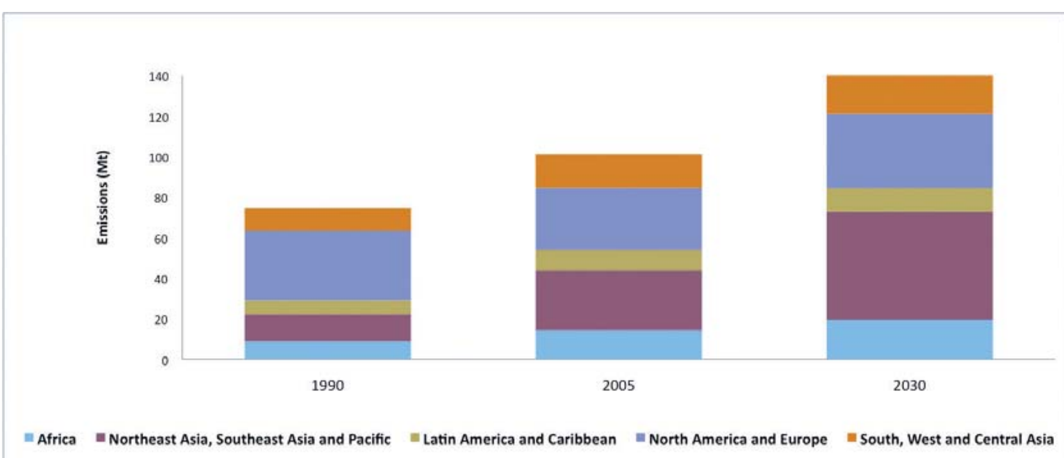

Figure 2.22. Trends in CH_4 emissions from fossil fuel extraction and distribution.

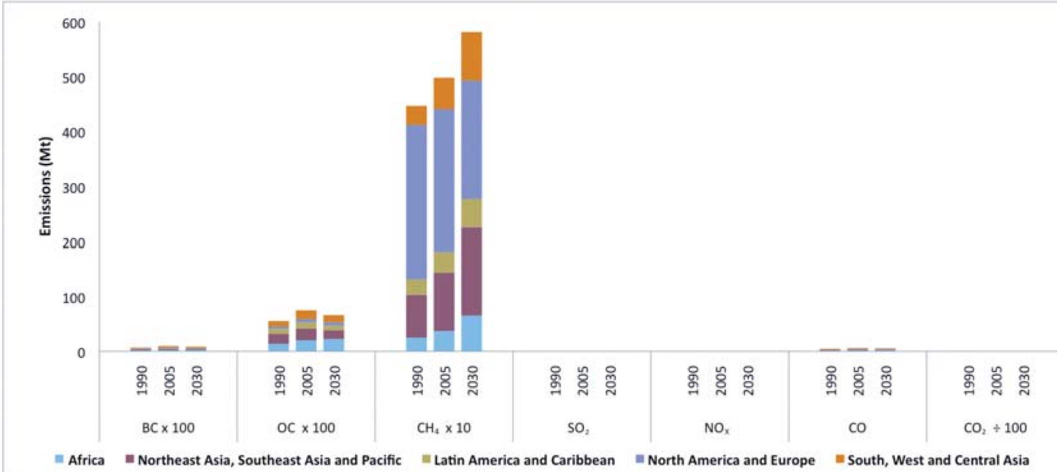

Figure 2.23. Trends in emissions from waste/landfill.

sector, emissions of BC, SO_2, NO_X, CO and CO_2 are relatively insignificant. In developing countries, where municipal waste collection and handling are not adequate, burning of solid waste (backyard and landfill fires) may contribute significantly to emissions of CO, NMVOCs, BC and OC. However, estimates and projections of emissions from such activities are almost completely lacking and further research is needed.

Agriculture

Global emissions of BC, OC, CH_4 and CO all increased between 1990 and 2005 and are projected to rise still further by 2030 (Figure 2.24). The BC, OC and CO emissions in this sector come mostly from open burning of crop residues, whilst CH_4 emissions are mainly due to enteric fermentation in livestock. Population and economic growth lead to changes in lifestyles and typically increased demand for meat and dairy products; consequently, agricultural activities increase sharply, leading to higher emissions of CH_4 and NH_3 (not shown). In all three years, the two regions with the greatest emissions of BC, OC and CO are Northeast Asia, Southeast Asia and Pacific (mainly China) and South, West and Central Asia (mainly India). Emissions of CH_4 tend to be more evenly distributed between regions. By 2030, Northeast Asia, Southeast Asia and Pacific, Latin America and Caribbean and South, West and Central Asia become the three regions with the largest CH_4 emissions.

Emissions of SO_2, NO_X and CO_2 from agriculture are relatively insignificant.

2.5.4 Sub-sector analysis for carbonaceous aerosols

Figure 2.15 showed that the residential-commercial sector is the predominant source of primary $PM_{2.5}$, this being particularly true for BC and OC. The transport sector is also an important emitter of BC and, to a lesser extent, OC. In developing countries, certain industrial processes, such as brick kilns and coke ovens, and the burning of crop residues within the agriculture sector are also notable contributors to primary aerosol emissions. In this section, therefore, these sectors are analysed at a greater level of disaggregation, focusing on those sub-sectors that are of most relevance to primary BC and OC emissions and where control measures (Chapter 5) have the greatest potential impact. Figures 2.25 and 2.26 (note the difference of scale between BC and OC emissions) show that for BC and OC emissions, residential biofuel use was a dominant and growing global source from 1990 to 2005, with Northeast Asia, Southeast Asia and Pacific being responsible for the greatest share (of which approximately two-thirds came from China) followed by Africa and then South, West and Central Asia. Despite a decline in solid fuel use and a resulting reduction in emissions from Northeast Asia, Southeast Asia and Pacific

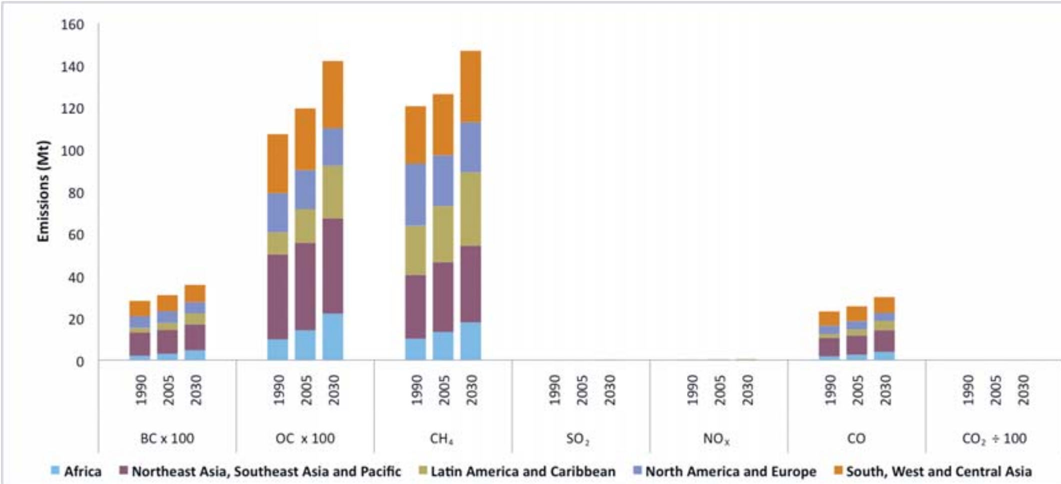

Figure 2.24. Trends in emissions from agriculture.

(almost entirely from China), global emissions from this sub-sector are projected to increase further by 2030. The development is largely driven by increases in biomass fuel use and consequent emissions from Africa. The main difference between BC and OC is that, while the BC trend is towards increasing emissions, global OC emissions are projected to decrease slightly by 2030, because improved combustion technologies are expected to reduce relatively more OC than BC.

Global emissions of BC and OC from residential coal use decreased between 1990 and 2005 and are projected to continue to decline up to 2030 as result of reduced emissions from North America and Europe and from Northeast Asia, Southeast Asia and Pacific (again almost entirely reductions in China due to improvements in combustion technologies). In 1990 and 2005, BC and OC emissions from brick and coke kilns were dominated by Northeast Asia, Southeast Asia and Pacific (China accounting for 85 per cent and 95 per cent of BC and OC, respectively) and South, West and Central Asia (India accounting for 50 per cent and 76 per cent, respectively). There was an increase in global emissions for this sector between 1990 and 2005, due mainly to an increase in Northeast Asia, Southeast Asia and Pacific (mainly China), with little subsequent change projected for 2030.

Road transport diesel combustion is also an important global source of BC and OC, with emissions having increased between 1990 and 2005, largely due to increases from Northeast Asia, Southeast Asia and Pacific, as well as South, West and Central Asia and Latin America and Caribbean, due to growth in transport activities. Reduced global BC and OC emissions from road transport diesel are projected by 2030. This is mainly a result of large reductions in emissions from North America and Europe resulting from strict emission controls and rather stable activity. Continued increases in emissions are projected for Northeast Asia, Southeast Asia and Pacific, and South, West and Central Asia, where the implementation of stricter air pollution policies is offset by growth in activity. The temporal trends and regional shares for off-road diesel closely follow those for road transport diesel, the main difference being a projected decrease for Northeast Asia, Southeast Asia and Pacific, and a projected increase for Latin America and Caribbean between 2005 and 2030. It is also worth noting that abatement technologies are penetrating the off-road transport sector more slowly than the on-road one. BC emissions from road transport powered by fuels other than diesel rose between 1990 and 2005, and this trend is projected to continue to 2030 as a result of a large increase in Northeast Asia, Southeast Asia and Pacific (entirely due to China). BC and OC emissions from agriculture, consist-

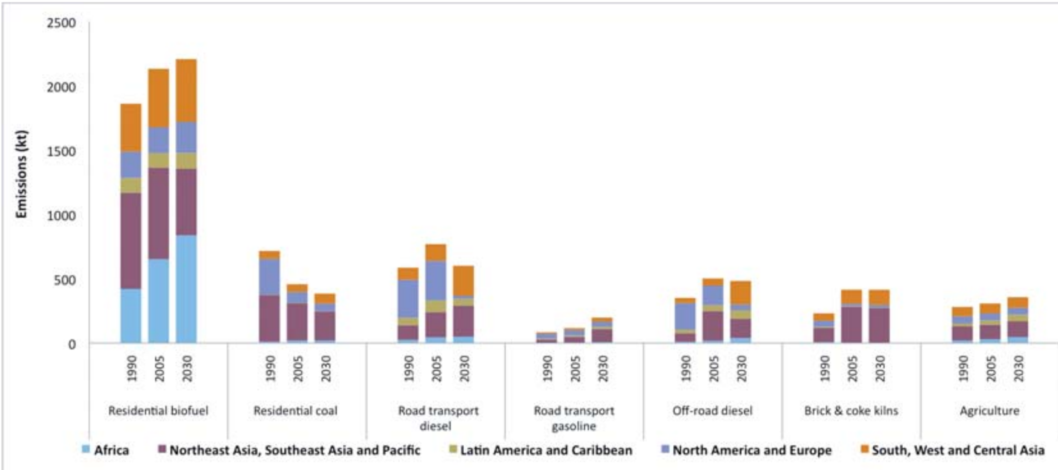

Figure 2.25. Trends in sub-sector BC emissions by region.

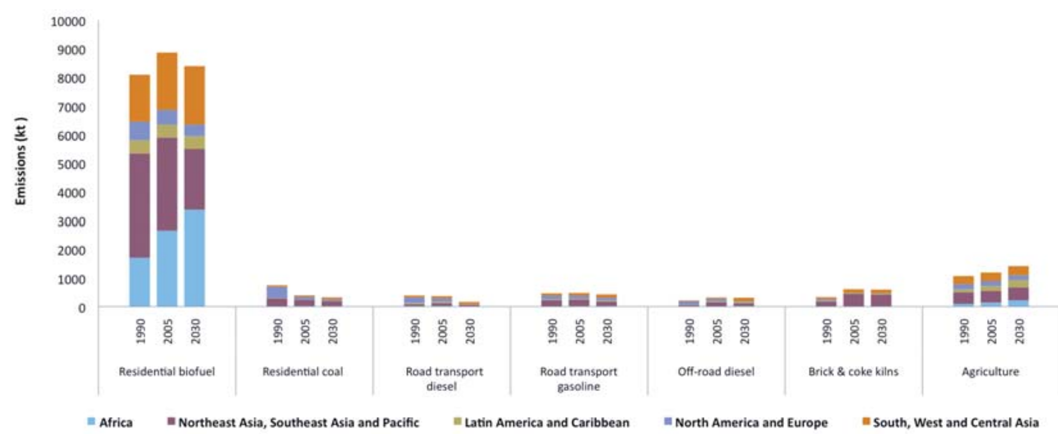

Figure 2.26. Trends in sub-sector OC emissions by region.

ing mainly of the burning of crop residues in the field, in 1990 and 2005 were also dominated by Northeast Asia, Southeast Asia and Pacific (China accounting for about 70 per cent) and South, West and Central Asia (India accounting for about 60 per cent). The climbing trend, which is projected to continue to 2030, is mainly due to increases in Africa and in Latin America and Caribbean. North America and Europe is the only region where a decline in emissions is expected.

OC/BC ratios

One significant aspect of carbonaceous aerosol emissions is the relative proportions of BC and OC. Because they have contrasting effects on radiative forcing, it is essential to know whether a particular type of source produces more BC than OC or vice versa. The OC/BC ratio is not the only factor determining sectoral contributions to a change in radiative forcing: other co-emitted aerosols and gases like SO_2 (especially from combustion of coal and high-sulphur oil), CO, NMVOCs, and NO_X, play a role too. However, for several sources, BC and OC are estimated to be the key determinants. Work assessing the magnitudes of direct and indirect effects, or even their sign, for BC and OC emissions and other aerosols is underway (Chapter 3).

It should be stressed that this section only addresses *primary* emissions of BC and OC and does not include secondary OC formed in the atmosphere after emission; this may appear to conflict with OC/BC ratios from modelling studies that include both primary and secondary OC combined.

Figure 2.27 shows the OC/BC mass ratios of the seven major source categories discussed above. The ratio of 1 (equal emissions) is highlighted as a reference level, but this does not imply equal effects on radiative forcing, because the forcing of BC per unit mass is greater than that of OC. In the case of emissions of a pure carbonaceous aerosol, an OC/BC mass ratio of more than about 10 would lead to cooling, since the global warming potential of OC and BC have been estimated to be about -70 and 700, respectively (Chapter 4). Because these source categories are relatively broad, they sometimes reflect mixtures of particular technologies and fuels, and they also reflect changing technologies and fuels into the future. This is an advantage, however, because it gives information on the overall proportions of BC and OC emitted from the sub-sector and how they are changing over time, and this gives insight into trends and opportunities for climate change mitigation. The different ratios in each of the five world regions are shown. A high OC/BC ratio represents a relatively unfavourable mitigation opportunity, because it reduces the cooling component more than the warming component. The three major source categories leading to high OC/BC ratios in 2005 are: residential biofuel use (OC/BC = 1.5–4.5), gasoline road transport (OC/BC = 3–6) and agriculture (OC/BC = 3–5). Residential coal use and brick and coke kilns have relatively similar emissions (OC/BC ≈ 1). Diesel road transport and off-road diesel have the lowest ratios (0.2–0.8) and are therefore good candidates for reducing net forcing.

Some ratios are likely to stay effectively constant until 2030, but others, mainly residential biofuel, diesel road transport and gasoline road transport, show a significant decline in their OC/BC ratio. In principle, the reductions in the OC/BC ratios reflect improvements in combustion or the effects of exhaust after-treatment technologies. In the cases of residential biofuel and coal, the development of the ratio reflects the transition from traditional heating and cooking stoves to improved and cleaner stoves through turnover of the equipment. The improved stoves often reduce OC more than BC, which sometimes may remain at the level of traditional stoves (Roden *et al.*, 2006, 2009). The technologies in use are often different in different regions, which is reflected in the ratios. The development of the OC/BC ratio in the transport sector is the result of a mix of different stages of emission abatement technologies and the pace at which they are implemented in a given region. For both diesel and gasoline, the later stages tend to reduce more OC than BC, which is why the later year ratios are lower. However, these lower ratios are associated with small overall emissions and so there is very little reduction potential left.

For agriculture, the ratio does not change over time, as the only option included in this study is a ban that affects both compounds in the same way. Furthermore, we do not assume any changes over time in the shares of various crop residues burned. The regional differences visible in Figure 2.27 stem from different assumptions about emission factors for key crop residues, the shares of which vary from region to region.

2.5.5 Sub-sector analysis for methane

Figure 2.28 shows that enteric fermentation in domestic livestock was the predominant source of global anthropogenic CH_4 emissions between 1990 and 2005 and is projected to remain so to 2030. A slight decreasing trend in North America and Europe from 1990 to 2005 is more than offset by increasing emissions from all other regions, particularly Latin America and Caribbean (43 per cent from Brazil in both years), and this trend is set to continue to 2030. The next most important CH_4 source sector is oil and gas production and distribution, with a rising trend between 1990 and 2005 projected to continue to 2030. This growth is driven mainly by increasing emissions from South, West and Central Asia (almost half from the Middle East), although there are also greater contributions projected for Africa and North America and Europe. In 1990, CH_4 emissions from landfill (dominated by North America and Europe) and rice production (dominated by Northeast Asia, Southeast Asia and Pacific, and South, West and Central Asia) were the next two most important source sectors. However, between 1990 and 2005, emissions from coal mining grew more rapidly than those from either rice production or landfill, due to a large increase in Northeast Asia, Southeast Asia and Pacific, and this trend is set to continue to 2030. Thus coal mining is projected to become the third most important CH_4 source sector by 2030, with Northeast Asia, Southeast Asia and Pacific the predominant emitting region (with 85 per cent from China). Combined, the two fossil-fuel related sectors would be the largest single source category. The treatment of waste water (municipal and industrial) is a minor contributor to global anthropogenic CH_4 production, although emissions increased in all regions between 1990 and 2005 and this trend is set to continue up to 2030.

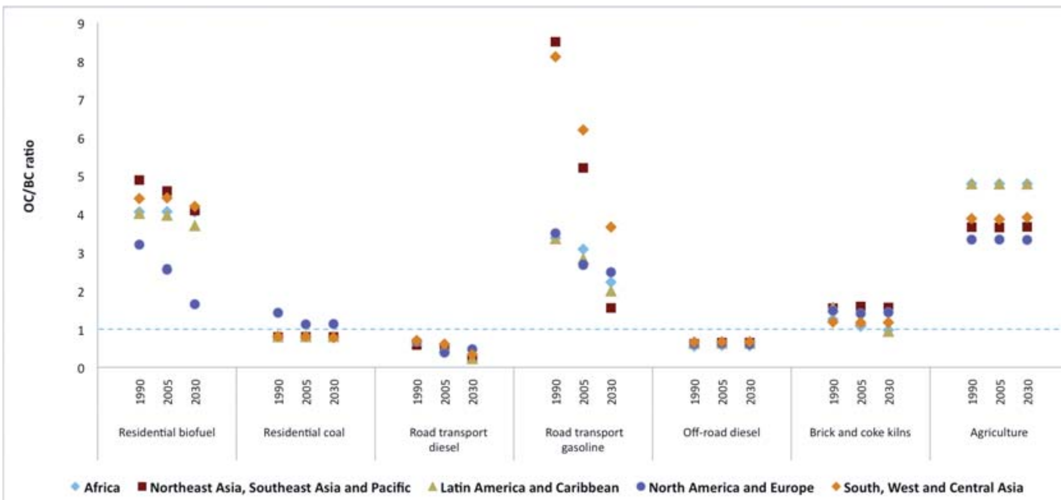

Figure 2.27. Trends in OC/BC mass ratios for important sub-sectors in each region.

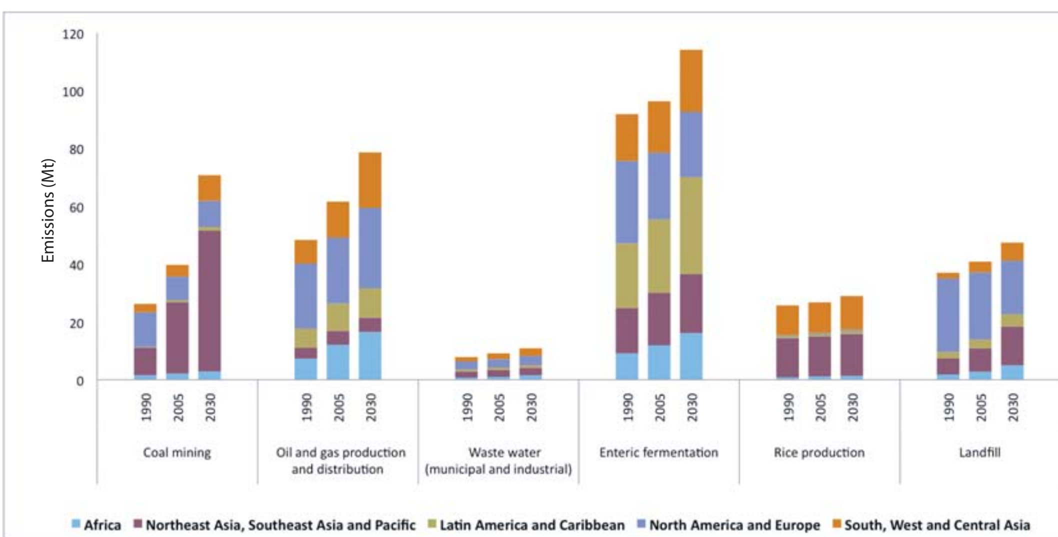

Figure 2.28. Trends in sub-sector CH_4 emissions by region.

Appendix A.2

There have been a number of attempts to compile estimates of present-day global emissions of pollutants relevant to this Assessment. These are not always consistent in their approaches to the three main categories of emission sources:

1. Fossil-fuel combustion, for which international statistics on fuel use are available;

2. Biofuel combustion, which is a large source in the developing world and for which international statistics are poor or non-existent; and

3. Open biomass burning, for which a variety of techniques, including national and regional surveys of burnt area and satellite observations, can be applied.

This section presents estimates of current emissions of BC, OC and tropospheric O_3 precursors developed in other works, against which the GAINS values can be compared. Although there are several of these inventories available, we present results from the two best known and most widely used inventories: Bond et al. (2004) for BC and OC and the EDGAR inventory for tropospheric O_3 precursors.

A.2.1 Present-day BC and OC emissions

A concise overview of global BC emissions can be obtained from Figures A.2.1 and A.2.2, based on data in Bond *et al.* (2004) for the year 1996. Although these data are somewhat old now, the distribution of emissions between regions and source types has not changed dramatically since then, and they are still informative. Total global BC emissions in 1996 were estimated at 8.0 Mt: 4.6 Mt from contained combustion – the burning of fossil fuels and biofuels for energy and other anthropogenic releases – and 3.3 Mt from open biomass burning. For contained combustion, Asia is the largest contributor, producing 2.5 Mt/yr or 55 per cent of the global total. China and India are the two largest emitting countries. With respect to open biomass burning – the accidental or intentional burning of forests, savannah/grassland and agricultural waste in fields – Africa produces the most BC: 1.9 Mt/yr or 44 per cent of the global total. Central/South America produces 0.91 Mt/yr or 27 per cent. The developed world (Europe, Former USSR, North America, Japan and the Pacific) is a relatively minor producer of BC of either kind. The global distributions of BC and OC are quite different, with over 70 per cent of BC emitted in the northern hemisphere and over 60 per cent of OC emitted in the southern hemisphere.

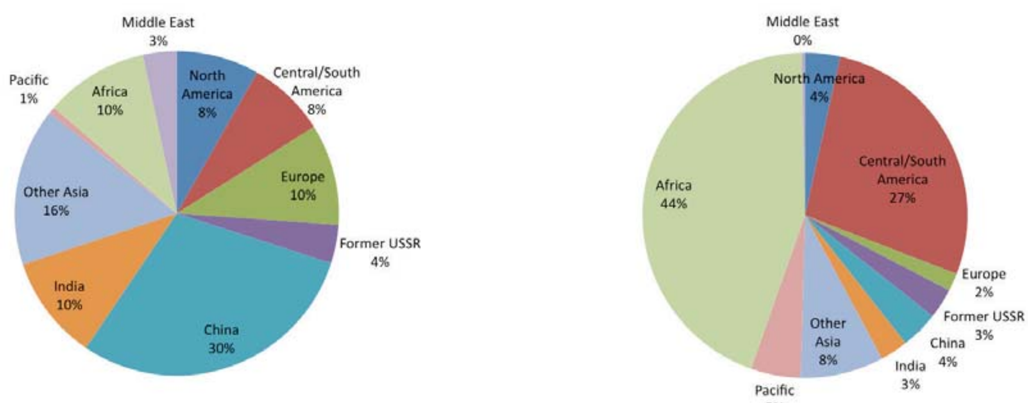

Figure A.2.1. Distribution of global BC emissions by world region for contained combustion (left) and open biomass burning (right) from Bond *et al.,* 2004.

Figure A.2.2 shows the relative contributions of all source types (both contained and open burning) to total global BC emissions. Open biomass burning is the largest individual contributor, with residential biofuel combustion the single largest anthropogenic source type at 1.5 Mt/yr or 19 per cent of the global total. On-road and off-road diesels produce a combined 1.4 Mt/yr or 17 per cent, while coal combustion in industry and the residential sector produces another 1.1 Mt/yr or 14 per cent. It should be noted that electric power generation, a major contributor to CO_2 emissions, does not appear on this chart because its contribution is so small. This is a powerful illustration of the very different nature of the sources of CO_2 and BC, and hence the different nature of the targets for mitigation. Although the spatial, temporal, sectoral and technological resolution of the inventory of Bond *et al.* and the GAINS model are different, the development of both has been closely coordinated between the groups, specifically with regard to the use of emission factors and principal assumptions on combustion technology in use. Therefore, the results are consistent between the two studies.

Global emissions of primary OC are about four times as large as global emissions of BC; note that secondary organic aerosols (SOA) that are formed in the atmosphere through chemical reactions of gaseous species are not included. The total primary OC emissions reported by Bond *et al.* (2004) are 33.9 Mt for 1996. The underlying reason for differences in total BC and OC emissions lies in the fact that the OC/BC emission ratio for the burning of vegetation is much higher than the OC/BC ratio for fossil-fuel combustion. The regional distributions of global OC emissions from contained and open combustion are quite similar to those of BC, with the shares of regions that burn excessive amounts of biofuel and biomass being somewhat enhanced (e.g., India, Africa). China's share is lower because of its greater reliance on coal than biofuels. As regards the distribution of OC emissions by source type, we see a major difference from BC. Open biomass burning is by far the dominant source type (74 per cent versus 41 per cent for BC). Residential biofuel combustion comprises 17 per cent of the total, about the same as for BC. However, all other contributing source types are smaller for OC than for BC, showing the dominant role of vegetation burning in OC emissions.

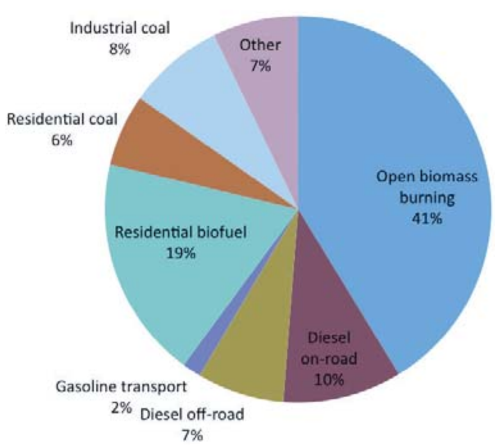

Figure A.2.2. Distribution of global BC emissions by major contributing source type from Bond *et al.*, 2004.

A.2.2 Present-day emissions of tropospheric O_3 precursors

Levels of tropospheric O_3 at regional scale (as distinct from peak O_3 levels observed in urban areas) are determined primarily by the emissions of four major pollutants: NO_X, CO, NMVOCs and CH_4. Together, these four control the formation and destruction of tropospheric O_3 under prevailing atmospheric conditions of temperature, insolation, humidity, etc. NMVOCs encompass a wide range of compounds of very different reactivities and ozone formation potentials. In order to determine the role that NMVOCs plays in O_3 formation, the speciation profiles of the emissions in different sectors and regions of the world need to be known. And though we discuss below the anthropogenic emissions of NMVOCs, it should be remembered that emissions of biogenic NMVOCs are larger by a factor of 5–7 and highly relevant.

There are relatively few global emission inventories that are comprehensive in nature, including all major species, source types and regions of the world. Perhaps the most well-known of them is the EDGAR (Emission Database for Global Atmospheric Research) (Olivier et al., 1996), presently hosted by the Joint Research Centre of the European Commission (EC-JRC/PBL, 2010). Results for the year 2005 from the recently released EDGAR v4.1 are shown below to illustrate the major sources of emissions of the four primary anthropogenic precursors of tropospheric O_3. Note that EDGAR only includes anthropogenic sources of emissions, following the UNFCCC convention. Open biomass burning is included but does not differentiate between wildfires and anthropogenic forest fires. The burning of forests and grassland for agriculture or habitation are included. As the EDGAR inventory is the most widely accepted global inventory of present-day emissions, it is desirable to compare its results with those that form the base case of the GAINS results for this study. Table A.2.1 summarizes the EDGAR v4.1 global emissions by species and major emitting sector.

NO_X emissions

In a similar way to the analysis followed above for BC, we can trace the major contributors to global NO_X emissions by world region, shown below in Figure A.2.3 (left) and by major emitting source type (right).

China is the largest contributor to global NO_X emissions, with 22 per cent of the total emission of 118.7 Mt. This reflects the large-scale use of fossil fuels, mainly coal, to power industrial and energy generation activities. In addition, China's NO_X emissions have grown very rapidly since 2000. North America (16 per cent), Europe (11 per cent), Africa (11 per cent) and Other Asia (11 per cent) are the other major emitting regions. Whereas industry and transport dominate the sources of NO_X in North America and Europe, it is biomass burning that largely accounts for the contributions of Africa and Other Asia. Sectoral emissions are dominated by industrial fuel combustion (including power generation) (43 per cent). On-road (22 per cent) and off-road (16 per cent) transport are also significant.

Table A.2.1. Global emissions of tropospheric O_3 precursors in 2005 by main source category from EDGAR v4.1 (Mt/yr).

	CO	NO_X	CH_4	NMVOCs
Industrial combustion	37.9	51.4	0.8	1.7
Fuel fugitive emissions	10.0	0.1	115.1	54.4
Domestic combustion	157.3	5.0	11.6	16.4
Transport on-road	147.0	25.9	0.7	21.9
Transport off-road	6.6	18.8	<0.1	1.4
Industrial processes	52.0	0.4	0.2	4.1
Solvent and other product use	0	0	0	21.9
Agriculture	248.4	12.9	150.5	14.5
Land-use change and forestry	188.3	3.9	11.7	14.2
Waste	<0.1	0.2	58.1	2.3
Other	2.5	<0.1	0.2	0.1
Global total	**850.2**	**118.7**	**349.0**	**152.9**

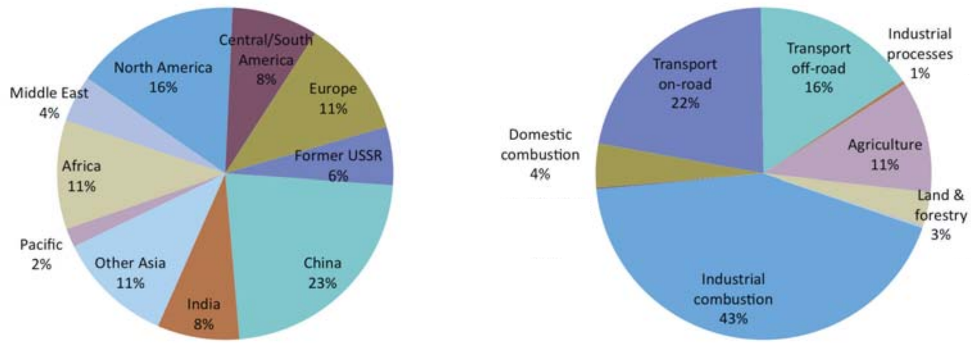

Figure A.2.3. Distribution of global NO_X emissions in 2005 by world region (left) and by major source type (right), from EDGAR v4.1.

CO emissions

Because CO emissions (Figure A.2.4) are primarily caused by poor combustion, global emissions are dominated by the developing regions of the world, where biofuels and fossil fuels continue to be burned in small, inefficient devices for residential cooking and heating and small-scale industrial operations. The EDGAR v4.1 inventory shows that the total global CO emissions of 850 Mt in 2005 are dominated by Asia (34 per cent) and Africa (30 per cent), with relatively small contributions from North America (8 per cent) and Europe (4 per cent). The largest source categories are agriculture (29 per cent) and land use and forestry (22 per cent), mostly consisting of various forms of vegetation burning for farming and human habitation. Residential combustion is also a large source of CO in the developing world (19 per cent) and transport (18 per cent) in all parts of the world.

NMVOC emissions

NMVOC emissions are more evenly distributed between the regions of the world (Figure A.2.5 (left)). The developed regions contribute significantly because of their large industrial sectors, handling substantial volumes of industrial chemicals, as well as their extensive transportation systems, processing and handling large quantities of oil products. On the other hand, NMVOC emissions are also

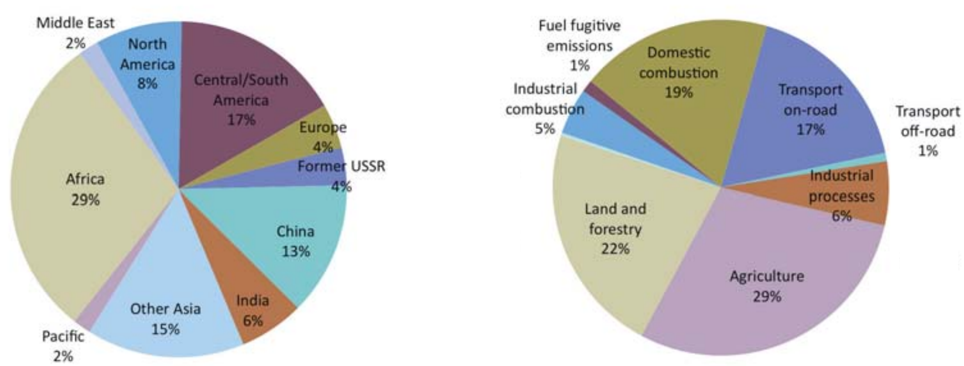

Figure A.2.4. Distribution of global CO emissions in 2005 by world region (left) and by major source type (right), from EDGAR v4.1.

a significant product of incomplete combustion, typically from traditional cooking and heating stoves, and therefore developing nations also contribute. On balance, Africa (20 per cent) and Other Asia (17 per cent) are the two largest emitting regions. North America (10 per cent) and Europe (10 per cent) contribute somewhat less. Fugitive emissions from oil, natural gas and solid fuels (36 per cent) is the largest contributing source category to global NMVOC emissions (Figure A.2.5 (right)), with transport (14 per cent) and solvent use (14 per cent) following. Several other source categories (residential fuel combustion, agriculture, and land use/ forestry) are significant contributors to NMVOC emissions.

CH_4 emissions

Methane has rather a different sectoral profile to the other pollutants (Figure A.2.6 (right)). Distribution around world regions (Figure A.2.6 - left) is perhaps the most even of all, with the largest contributors being China (18 per cent), Central/South America (14 per cent), and Other Asia (13 per cent). Because of the contribution from coal mining, those regions that produce large quantities of coal are preferentially higher (like the former USSR). By source category (Figure A.2.6 - right), agriculture is the largest (43 per cent), reflecting the close association of CH_4 with rice growing and wetlands. Fugitive emissions from fuels generate 33 per cent of global CH_4, and waste disposal through landfills (17 per cent) is also a large source. All other sources of CH_4 are small.

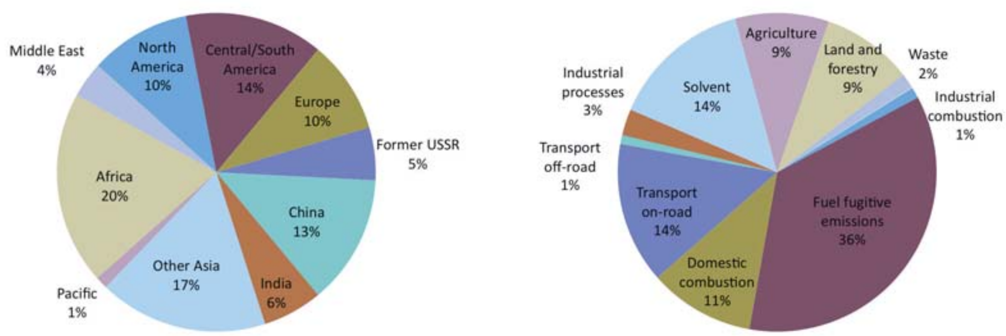

Figure A.2.5. Distribution of global NMVOC emissions in 2005 by world region (left) and by major source type (right), from EDGAR v4.1.

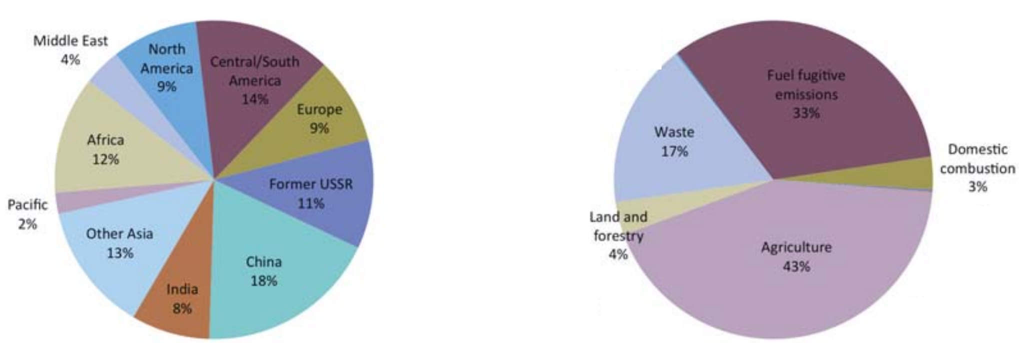

Figure A.2.6. Distribution of global CH_4 emissions in 2005 by world region (left) and by major source type (right), from EDGAR v4.1.

References

Achard, F., Eva, H. D., Mollicone, D. and Beuchle, R. (2008). The effect of climate anomalies and human ignition factor on wildfires in Russian boreal forests. *Philos. T. Roy. Soc. B*, 363, 2329–2337.

Akimoto, H. (2003). Global air quality and pollution. *Science*, 302, 1716–1719.

Akimoto, H., Ohara, T., Kurokawa, J. and Horii, N. (2006). Verification of energy consumption in China during 1996–2003 by using satellite observational data. *Atmos. Environ.*, 40, 7663–7667.

Alexandratos, N., Bruinsma, J., Boedeker, G., Schmidhuber, J., Broca, S., Shetty, P., and Ottaviani, M. G. (2006). *World agriculture: towards 2030/2050. Interim report. Prospects for food, nutrition, agriculture and major commodity groups.* FAO, Rome. http://www.fao.org/es/esd/AT2050web.pdf.

Allen, D., Pickering, K. and Fox-Rabinovitz, M. (2004). Evaluation of pollutant outflow and CO sources during TRACE-P using model-calculated, aircraft-based, and Measurements of Pollution in the Troposphere (MOPITT)-derived CO concentrations. *J. Geophys. Res.*, 109, D15S03, doi:10.1029/2003JD004250.

Amann, M., Kejun, J., Jiming, H., Wang, S., Xing, Z., Wei, W., Yi Xiang, D., Hong, L., Jia, X., Chuying, Z., Bertok, I., Borken, J., Cofala, J., Heyes, C., Höglund, L., Klimont, Z., Purohit, P., Rafaj, P., Schöpp, W., Toth, G., Wagner, F. and Winiwarter, W. (2008). *GAINS ASIA: Scenarios for Cost-effective Control of Air Pollution and Greenhouse Gases in China*. IIASA Policy Report. IIASA, Laxenburg, Austria.

Andreae, M. O. and Merlet, P. (2001). Emission of trace gases and aerosols from biomass burning. *Global Biogeochem. Cycles*, 15, 955–966.

Andreae, M. O., Rosenfeld, D., Artaxo, P., Costa, A. A., Frank, G. P., Longo, K. M. and Silva-Dias, M. A. F. (2004). Smoking rain clouds over the Amazon. *Science*, 303, 1337–1342.

Andres, R. J. and Kasgnoc, A. D. (1998). A time-averaged inventory of subaerial volcanic sulfur emissions. *J. Geophys. Res.,* 103, 25,251–25,261.

Arellano, A. F., Kasibhatla, P. S., Giglio, L., van der Werf, G. R., Randerson, J. T. and Collatz, G. J. (2006). Time-dependent inversion estimates of global biomass-burning CO emissions using Measurement of Pollution in the Troposphere (MOPITT) measurements. *J. Geophys. Res.*, 111, D09303, doi:10.1029/2005JD006613.

Artaxo, P., Martins, J. V., Yamasoe, M. A., Procopio, A. S., Pauliquevis, T. M., Andreae, M. O., Guyon, P., Gatti, L. V. and Leal, A. M. C. (2002). Physical and chemical properties of aerosols in the wet and dry seasons in Rondonia, Amazonia. *J. Geophys. Res.,* 107, 8081, doi:10.1029/2001JD000666.

Artaxo, P. and Andreae, M. O. (2007). Biomass burning as a driver for atmospheric composition and ecosystem changes. *iLEAPS Newsletter*, 4, 12–14.

Artaxo, P. (2010). Reductions in deforestation rates in Amazonia. *Global Atmospheric Pollution Forum Newsletter*, 8, 2–3.

Ban-Weiss, G. A., Lunden, M. M., Kirchstetter, T. W. and Harley, R.A. (2009). Measurement of black carbon and particle number emissions factors from individual heavy duty trucks. *Environ. Sci. Technol.*, 43, 1419–1424.

Bellprat, O. (2009). *Brick Kiln Evaluation Study in the Bajío Region GTO, México*. Instituto Nacional de Ecologia Mexico and Swiss Federal Institute of Technology, Zürich. September 2009.

Bond, T. C., Streets, D. G., Yarber, K. F., Nelson, S. M., Woo, J.-H. and Klimont, Z. (2004). A technology-based global inventory of black and organic carbon emissions from combustion. *J. Geophys. Res.,* 109, D14203, doi:10.1029/2003JD003697.

Bond, T. C., Bhardwaj, E., Dong, R., Jogani, R., Jung, S. K., Roden, C., Streets, D. G. and Trautmann, N. M. (2007). Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850–2000. *Global Biogeochem. Cycles,* 21, GB2018, doi:10.1029/2006GB002840.

Bouwman, A. F., Boumans, L. J. M. and Batjes, N. H. (2002). Estimation of global NH_3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands. *Global Biogeochem. Cycles*, 16, 1024, doi:10.1029/2000GB001389.

Bowman, D. M. J. S., Balch, J. K., Artaxo, P., Bond, W. J., Carlson, J. M., Cochrane, M. A., D'Antonio, C. M., DeFries, R. S., Doyle, J. C., Harrison, S. P., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Marston, J. B., Moritz, M. A., Prentice, I. C., Roos, C. I., Scott, A. C., Swetnam, T. W., van der Werf, G. R. and Pyne, S. J. (2009). Fire in the Earth system. *Science*, 324, 481–484.

Bruinsma, J. (ed.) (2003). *World Agriculture: Towards 2015/2030, An FAO Perspective.* Earthscan, London and FAO, Rome. http://www.fao.org/DOCREP/005/Y4252E/Y4252E00.htm.

Buhaug, Ø., Corbett, J.J., Endresen, Ø., Eyring, V., Faber, J., Hanayama, S., Lee, D.S., Lee, D., Lindstad, H., Markowska, A.Z., Mjelde, A., Nelissen, D., Nilsen, J., Pålsson, C., Winebrake, J.J., Wu, W. and Yoshida, K. (2009). *Second IMO GHG Study 2009*. International Maritime Organization, London, UK.

CAI-Asia (2008). Emission standards for new vehicles (light duty). Clean Air Initiative for Asian Cities (CAI-Asia). http://www.cleanairnet.org/caiasia/1412/articles-58969_new.pdf.

Carmichael, G. R., Tang, Y., Kurata, G., Uno, I., Streets, D. G., Thongboonchoo, N., Woo, J. H., Guttikunda, S., White, A., Wang, T., Blake, D. R., Atlas, E., Fried, A., Potter, B., Avery, M. A., Sachse, G. W., Sandholm, S. T., Kondo, Y., Talbot, R. W., Bandy, A., Thornton, D. and Clarke, A. D. (2003). Evaluating regional emission estimates using the TRACE-P observations. *J. Geophys. Res.*, 108, 8810, doi:10.1029/2002JD003116.

Chen, Y., Zhi, G., Feng, Y., Liu, D., Zhang, G., Li, J., Sheng, G. and Fu, J. (2009). Measurements of black and organic carbon emission factors for household coal combustion in China: Implication for emission reduction. *Environ. Sci. Technol*., 43, 9495–9500.

Christian, T. J., Kleiss, B., Yokelson, R. J., Holzinger, R., Crutzen, P. J., Hao, W. M., Saharjo, B. H. and Ward, D. E. (2003). Comprehensive laboratory measurements of biomass-burning emissions: 1. Emissions from Indonesian, African, and other fuels. *J. Geophys. Res*., 108, 4719, doi:10.1029/2003JD003704.

Christian, T. J., Yokelson, R. J., Cárdenas, B., Molina, L. T., Engling, G. and Hsu, S.-C. (2010). Trace gas and particle emissions from domestic and industrial biofuel use and garbage burning in central Mexico. *Atmos. Chem. Phys*., 10, 565–584.

Cofala, J. and Syri, S. (1998a). *Sulfur Emissions, Abatement Technologies and Related Costs for Europe in the RAINS Model Database.* International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Cofala, J. and Syri, S., (1998b). *Nitrogen Oxides Emissions, Abatement Technologies and Related Costs for Europe in the RAINS Model Database.* International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria.

Cofala, J., Amann, M., Klimont, Z., Kupiainen, K. and Höglund-Isaksson, L. (2007). Scenarios of global anthropogenic emissions of air pollutants and methane until 2030. *Atmos. Environ*., 41, 8486–8499.

Cooke, W., Liousse, C., Cachier, H. and Feichter, J. (1999). Construction of a 1° × 1° fossil fuel emission data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model. *J. Geophys. Res.,* 104, 22,137–162.

Corbett, J. J., Lack, D. A., Winebrake, J. J., Harder, S., Silberman, J. A. and Gold, M. (2010). Arctic shipping emissions inventories and future scenarios. *Atmos. Chem. Phys*., 10, 9689–9704.

De Smedt, I., Müller, J.-F., Stavrakou, T., van der A, R., Eskes, H. and Van Roozendael, M. (2008). Twelve years of global observations of formaldehyde in the troposphere using GOME and SCIAMACHY sensors. *Atmos. Chem. Phys*., 8, 4947–4963.

Derwent, R. G., Jenkin, M. E., Passant, N. R. and Pilling, M. J. (2007). Reactivity-based strategies for photochemical ozone control in Europe. *Environ. Sci. Policy,* 10, 445–453.

DieselNet (2010). Summary of worldwide diesel emission standards, http://www.dieselnet.com/standards.

Döring, U., Janssens-Maenhout, G., van Aardenne, J. and Pagliari, V. (2010). *Scenarios of Future Climate Change.* FP6 IP CIRCE del. 3.3.1, JRC technical note, Joint Research Centre (JRC).

Dufour, G., Wittrock, F., Camredon, M., Beekmann, M., Richter, A., Aumont, B. and Burrows, J. P. (2009). SCIAMACHY formaldehyde observations: constraint for isoprene emission estimates over Europe? *Atmos. Chem. Phys.,* 9, 1647–1664.

Durbin, T. D., Norbeck, J. M., Smith, M. R. and Truex, T. J. (1999). Particulate emission rates from light duty vehicles in the South Coast Air Quality Management District. *Environ. Sci. Technol*., 33, 4401–4406.

EC-JRC/PBL (European Commission Joint Research Centre/Netherlands Environmental Assessment Agency) (2010). Emission Database for Global Atmospheric Research (EDGAR), http://edgar.jrc.ec.europa.eu.

EFMA (European Fertilizer Manufacturers Association) (2010). *Forecast of Food, Farming and Fertilizer Use in the European Union - 2009 to 2019*. Brussels, http://www.fertilizerseurope.com.

Erisman, J. W., Sutton, M. A., Galloway, J., Klimont, Z. and Winiwarter, W. (2008). How a century of ammonia synthesis changed the world. *Nat. Geosci.*, 1, 636-639.

Eyring, V., Isaksen, I. S. A., Berntsen, T., Collins, W. J., Corbett, J. J., Endresen, O., Grainger, R. G., Moldanova, J., Schlager, H. and Stevenson, D. S. (2010). Assessment of transport impacts on climate and ozone: Shipping. *Atmos. Environ*., 44, 4735–4771.

FAO (2010). FAOSTAT: United Nations Food and Agriculture Organization (FAO) statistical databases, available at http://faostat.fao.org/site/339/default.aspx.

Fernandes, S. D., Trautmann, N. M., Streets, D. G., Roden, C. A. and Bond, T. C. (2007). Global biofuel use, 1850–2000. *Global Biogeochem. Cycles*, 21, GB2019, doi:10.1029/2006GB002836.

Fischer, G., Winiwarter, W., Ermolieva, T., Cao, G.-Y., Qui, H., Klimont, Z., Wiberg, D. and Wagner, F. (2010). Integrated modeling framework for assessment and mitigation of nitrogen pollution from agriculture: Concept and case study for China. *Agr., Ecosys. and Env.*, 136, 116–124.

Friedlingstein, P., Houghton, R. A., Marland, G., Hackler, J., Boden, T. A., Conway, T. J., Canadell, J. G., Raupach, M. R., Ciais, P. and Le Quéré, C. (2010). Update on CO_2 emissions. *Nat. Geosci*., 3, 811–812.

Fromm, M., Bevilacqua, R., Servranckx, R., Rosen, J., Thayer, J. P., Herman, J. and Larko, D. (2005). Pyro-cumulonimbus injection of smoke to the stratosphere: Observations and impact of a super blowup in northwestern Canada on 3–4 August 1998. *J. Geophys. Res*., 110, D08205, doi:10.1029/2004JD005350.

Fu, T.-M., Jacob, D. J., Palmer, P. I., Chance, K., Wang, Y. X., Barletta, B., Blake, D. R., Stanton, J. C. and Pilling, M. J. (2007). Space-based formaldehyde measurements as constraints on volatile organic compound emissions in east and south Asia and implications for ozone. *J. Geophys. Res.*, 112, D06312, doi:10.1029/2006JD007853.

Fung, I., John, J., Lerner, J., Matthews, E., Prather, M., Steele, L. P. and Fraser, P. J. (1991). Three-dimensional model synthesis of the global methane cycle. *J. Geophys. Res.*, 96, 13,033–13,065.

Gauss, M., Myhre, G., Isaksen, I. S. A., Grewe, V., Pitari, G., Wild, O., Collins, W. J., Dentener, F. J., Ellingsen, K., Gohar, L. K., Hauglustaine, D. A., Iachetti, D., Lamarque, J.-F., Mancini, E., Mickley, L. J., Prather, M. J., Pyle, J. A., Sanderson, M. G., Shine, K. P., Stevenson, D. S., Sudo, K., Szopa, S. and Zeng, G. (2006). Radiative forcing since preindustrial times due to ozone change in the troposphere and the lower stratosphere. *Atmos. Chem. Phys.*, 6, 575–599.

Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., van der Gon, H. D., Frost, G. J., Heil, A., Kaiser, J. W., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F., Liousse, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M. G., Smith, S. J., Thompson, A., van Aardenne, J., van der Werf, G. R. and van Vuuren, D. P. (2011). Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period. *Climatic Change* 109, 163-190. DOI 10.1007/s10584-011-0154-1

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I. and Geron, C. (2006). Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmos. Chem. Phys.*, 6, 3181–3210.

Houghton, J.T., Meira Filho, L.G., Lim, B., Tre'anton, K., Mamaty, I., Bonduki, Y., Griggs, D.J., and Callander, B.A., (eds.). (1997). *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories*. Vol.1–3. Hadley Centre Meteorological Office, Bracknell, United Kingdom.

Hsu, Y. and Mullen, M. (2007). *Compilation of Diesel Emissions Speciation Data. Final Report.* CRC contract No. E-75.

IEA (1997). *Coal Power 2: World Coal-Fired Power Stations and Their Pollution Control Systems*. International Energy Agency, London, U.K.

IEA (2009). *World Energy Outlook 2009*. OECD/IEA, Paris, France.

IEA CCC (International Energy Agency Clean Coal Centre) (2010). Coal Power Database. IEA, available at http://www.iea-coal.org.uk.

IFA (International Fertilizer Industry Association) (2003). World Fertilizer Consumption Statistics, available from http://www.fertilizer.org/ifa/Home-Page/STATISTICS.

IPCC (2006). *2006 IPCC Guidelines for National Greenhouse Gas Inventories*. Report prepared by the National Greenhouse Gas Inventories Programme. Eggleston, H. S., Buendia, L., Miwa, K., Ngara, T. and Tanabe, K. (eds). IGES, Japan.

IPCC (2007). *Climate Change 2007: Synthesis Report. Contribution of Working Groups I, II and III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change.* Geneva, Switzerland.

Ito, A. and Penner, J. E. (2004). Global estimates of biomass burning emissions based on satellite imagery for the year 2000. *J. Geophys. Res.,* 109, D14S05, doi:10.1029/2003JD004423.

Junker, C. and Liousse, C. (2008). A global emission inventory of carbonaceous aerosol from historic records of fossil fuel and biofuel consumption for the period 1860–1997. *Atmos. Chem. Phys.*, 8, 1195–1207.

Kasibhatla, P., Arellano, A., Logan, J. A., Palmer, P. I. and Novelli, P. (2002). Top-down estimate of a large source of atmospheric carbon monoxide associated with fuel combustion in Asia. *Geophys. Res. Lett.,* 29, 1900, doi:10.1029/2002GL015581.

Klimont, Z., Amann, M. and Cofala, J. (2000). *Estimating Costs for Controlling Emissions of Volatile Organic Compounds from Stationary Sources in Europe.* Interim Report IR-00-51, International Institute for Applied Systems Analysis, Laxenburg, Austria.

Klimont, Z., Streets, D., Gupta, S., Cofala, J., Lixin, F. and Ichikawa, Y. (2002). Anthropogenic emissions of non-methane volatile organic compounds (NMVOC) in China. *Atmos. Environ*., 36, 1309–1322.

Klimont, Z. and Brink, C. (2004). *Modeling of Emissions of Air Pollutants and Greenhouse Gases from Agricultural Sources in Europe.* Interim Report IR-04-048, International Institute for Applied Systems Analysis, Laxenburg, Austria.

Klimont, Z. (2005). Projections of agricultural emissions of ammonia in the European Union. In: Kuczynski, T. *et al.* (eds.) *Emissions from European Agriculture*. Wageningen Academic Publishers, Wageningen, Netherlands, pp. 227–246.

Klimont, Z., Cofala, J., Xing, J., Wei, W., Zhang, C., Wang, S., Kejun, J., Bhandari, P., Mathur, R., Purohit, P., Rafaj, P., Chambers, A. and Amann, M. (2009). Projections of SO_2, NO_X and carbonaceous aerosols emissions in Asia. *Tellus B,* 61, 602–617.

Kupiainen, K. and Klimont, Z. (2007). Primary emissions of fine carbonaceous particles in Europe. *Atmos. Environ*., 41, 2156–2170.

Lack, D. A. *et al.* (2009). Particulate emissions from commercial shipping: Chemical, physical, and optical properties. *J. Geophys. Res*., 114, D00F04, doi:10.1029/2008JD011300.

Lamarque, J.-F., Bond, T. C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K. and van Vuuren, D. P. (2010). Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmos. Chem. Phys.,* 10, 7017–7039.

Lin, J. T., McElroy, M. B. and Boersma, K. F. (2010). Constraint of anthropogenic NO_X emissions in China from different sectors: a new methodology using multiple satellite retrievals. *Atmos. Chem. Phys.,* 10, 63–78.

Ma, J., Richter, A., Burrows, J. P., Nüss, H. and van Aardenne, J. A. (2006). Comparison of model-simulated tropospheric NO_2 over China with GOME-satellite data. *Atmos. Environ.,* 40, 593–604.

McCormick, M. P., Thomason, L. W. and Trepte, C. R. (1995). Atmospheric effects of the Mt Pinatubo eruption. *Nature*, 373, 399–404.

Mickley, L. J., Jacob, D. J. and Rind, D. (2001). Uncertainty in preindustrial abundance of tropospheric ozone: Implications for radiative forcing calculations. *J. Geophys. Res.,* 106, 3389–3399.

Mieville, A., Granier, C., Liousse, C., Guillaume, B., Mouillot, F., Lamarque, J.-F., Grégoire, J.-M. and Pétron, G. (2010). Emissions of gases and particles from biomass burning during the 20th century using satellite data and an historical reconstruction. *Atmos. Environ.,* 44, 1469–1477.

Monks, P.S., Granier, C., Fuzzi, S., Stohl, A., Williams, M.L., Akimoto, H., Amman, M., Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R. S., Carslaw, K., Cooper, O. R., Dentener, F., Fowler, D., Fragkou, E., Frost, G. J., Generoso, S., Ginoux, P., Grewe, V., Guenther, A., Hansson, H.C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I.S.A., Jenkin, M.E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M.G., Lee, J.D., Liousse, C., Maione, M., McFiggans, G., Metzger, A., Mieville, A., Moussiopoulos, N., Orlando, J.J., O'Dowd, C.D., Palmer, P.I., Parrish, D. D., Petzold, A., Platt, U., Pöschl, U., Prévôt, A.S.H., Reeves, C.E., Reimann, S., Rudich, Y., Sellegri, K., Steinbrecher, R., Simpson, D., t. Brink, H., Theloke, J., v. d. Werf, G., Vautard, R., Vestreng, V., Vlachokostas, C. and v. Glasow, R. (2009). Atmospheric composition change – global and regional air quality. *Atmos. Environ.*, 43, 5268–5350.

Mouillot, F. and Field, C. B. (2005). Fire history and the global carbon budget: a 1° × 1° fire history reconstruction for the 20th century. *Glob. Change Biol.*, 11, 398–420.

Mueller, J. F. and Stavrakou, T. (2005). Inversion of CO and NO_X emissions using the adjoint of the IMAGES model. *Atmos. Chem. Phys.*, 5, 1157–1186.

Nakicenovic, N. et al. (2000). Emissions Scenarios: A Special Report of Working Group III of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, UK.

Novakov, T., Ramanathan, V., Hansen, J. E., Kirchstetter, T. W., Sato, M., Sinton, J. E. and Sathaye, J. A. (2003). Large historical changes of fossil-fuel black carbon aerosols. *Geophys. Res. Lett.,* 30, 1324, doi:10.1029/2002GL016345.

Olivier, J. G. J., Bouwman, A. F., Van der Maas, C. W. M., Berdowski, J. J. M., Veldt, C., Bloos, J. P. J., Visschedijk, A. J. H., Zandveld, P. Y. J. and Haverlag, J. L. (1996). Description of EDGAR Version 2.0, a set of global emission inventories of greenhouse gases and ozone-depleting substances for all anthropogenic and most natural sources on a per country basis and on 1×1 degree grid, RIVM/TNO report, Bilthoven, The Netherlands.

Olivier, J. G. J. and Berdowski, J. J. M. (2001). Global emissions sources and sinks. In: Berdowski, J. J. M., Guicherit, R. and Heij, B. J. (eds.), *The Climate System.* A. A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands.

Pavelin, E. G., Johnson, C. E., Rughooputh, S. and Toumi, R. (1999). Evaluation of pre-industrial surface ozone measurements made using Schönbein's method. *Atmos. Environ.,* 33, 919–929.

Pétron, G., Granier, C., Khattatov, B., Yudin, V., Lamarque, J.-F., Emmons, L., Gille, J. and Edwards, D. P. (2004). Monthly CO surface sources inventory based on the 2000–2001 MOPITT satellite data. *Geophys. Res. Lett.,* 31, L21107, doi:10.1029/2004GL020560.

Quinn, P. K., Bates, T. S., Baum, E., Doubleday, N., Fiore, A. M., Flanner, M., Fridland, A., Garrett, T. J., Koch, D., Menon, S., Shindell, D., Stohl, A. and Warren, S. G. (2008). Short-lived pollutants in the Arctic: their climate impact and possible mitigation strategies. *Atmos. Chem. Phys*., 8, 1723–1735.

Ramanathan, V. and Carmichael, G. (2008). Global and regional climate changes due to black carbon. *Nat. Geosci.*, 1, 221–227.

Reid, J. S., Koppmann, R., Eck, T. F. and Eleuterio, D. P. (2005a). A review of biomass burning emissions part II: intensive physical properties of biomass burning particles. *Atmos. Chem. Phys*., 5, 799–825.

Reid, J. S., Eck, T. F., Christopher, S. A., Koppmann, R., Dubovik, O., Eleuterio, D. P., Holben, B. N., Reid, E. A. and Zhang, J. (2005b). A review of biomass burning emissions part III: intensive optical properties of biomass burning particles. *Atmos. Chem. Phys.*, 5, 827–849.

Roden, C. A., Bond, T. C., Conway, S., Benjamin, A. and Pinel, O. (2006). Emission factors and real-time optical properties of particles emitted from traditional wood burning cookstoves. *Environ. Sci. Technol.,* 40, 6750–6757.

Roden, C. A., Bond, T. C., Conway, S., Pinel, A. B. S., MacCarty, N. and Still, D. (2009). Laboratory and field investigations of particulate and carbon monoxide emissions from traditional and improved cookstoves. *Atmos. Environ.,* 43, 1170–1181.

Schöpp, W., Klimont, Z., Suutari, R. and Cofala, J. (2005). Uncertainty analysis of emission estimates in the RAINS integrated assessment model. *Environ. Sci. Policy*, 8, 601–613.

Schultz, M. G., Heil, A., Hoelzemann, J. J., Spessa, A., Thonicke, K., Goldammer, J. G., Held, A. C., Pereira, J. M. C. and van het Bolscher, M. (2008). Global wildland fire emissions from 1960 to 2000. *Global Biogeochem. Cycles*, 22, GB2002, doi:10.1029/2007GB003031.

Schumann, U. and Huntrieser, H. (2007). The global lightning-induced nitrogen oxides source. *Atmos. Chem. Phys.*, 7, 3823–3907.

Shindell, D. T. *et al.* (2006). Multimodel simulations of carbon monoxide: Comparison with observations and projected near-future changes. *J. Geophys. Res.,* 111, D19306, doi:10.1029/2006JD007100.

Smith, S. J., van Aardenne, J., Klimont, Z., Andres, R. J., Volke, A. and Delgado Arias, S. (2011). Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmos. Chem Phys., 11, 1101-1116.*

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H. and Yarber, K. F. (2003a). An inventory of gaseous and primary aerosol emissions in Asia in the year 2000., *J. Geophys. Res*., 108, 8809, doi:10.1029/2002JD003093.

Streets, D. G., Yarber, K. F., Woo, J.-H. and Carmichael, G. R. (2003b). Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions. *Global Biogeochem. Cycles,* 17, 1099, doi:10.1029/2003GB002040.

Streets, D. G., Bond, T. C., Lee, T. and Jang, C. (2004). On the future of carbonaceous aerosol emissions, *J. Geophys. Res*., 109, D24212, doi:10.1029/2004JD004902.

Streets, D. G., Zhang, Q., Wang, L. T., He, K. B., Hao, J. M., Wu, Y., Tang, Y. H. and Carmichael, G. R. (2006). Revisiting China's CO emissions after the Transport and Chemical Evolution over the Pacific (TRACE-P) mission: Synthesis of inventories, atmospheric modeling, and observations. *J. Geophys. Res.,* 111, D14306, doi:10.1029/2006JD007118.

Subramanian, R., Winijkul, E., Bond, T. C., Thiansathit, W., Oanh, N. T. K., Paw-Armart, I. and Duleep, K. G. (2009). Climate-relevant properties of diesel particulate emissions: results from a piggyback study in Bangkok, Thailand. *Environ. Sci. Technol*., 43, 4213–4218.

van Aardenne, J. A., Dentener, F. J., Olivier, J. G. J., Goldewijk, C. G. M. K. and Lelieveld, J. (2001). A 1º × 1º resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990. *Global Biogeochem. Cycles*, 15, 909–928.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Kasibhatla, P. S. and Arellano, A. F. (2006). Interannual variability in global biomass burning emissions from 1997 to 2004. *Atmos. Chem. Phys.,* 6, 3423–3441.

van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y. and van Leeuwen, T. T. (2010). Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009). *Atmos. Chem. Phys*., 10, 11707–11735.

Wang, T., Wei, X. L., Ding, A. J., Poon, C. N., Lam, K. S., Li, Y. S., Chan, L. Y. and Anson, M. (2009). Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994–2007. *Atmos. Chem. Phys.,* 9, 6216–6226.

Wei, W., Wang, S., Chatani, S., Klimont, Z., Cofala, J. and Hao, J. (2008). Emission and speciation of non-methane volatile organic compounds from anthropogenic sources in China. *Atmos. Environ*., 42, 4976–4988.

Xu, Y., Williams, R. H. and Socolow, R. H. (2009). China's rapid deployment of SO_2 scrubbers. *Energy Environ. Sci.,* 2, 459–465.

Yamasoe, M. A., Artaxo, P., Miguel, A. H. and Allen, A. G. (2000). Chemical composition of aerosol particles from direct emissions of vegetation fires in the Amazon Basin: water-soluble species and trace elements. *Atmos. Environ.,* 34, 1641–1653.

Yanowitz, J., McCormick, R. L. and Graboski, M. S. (1999). In-use emissions from heavy-duty diesel vehicles. *Environ. Sci. Technol*., 34, 729–740.

Yokelson, R. J., Karl, T., Artaxo, P., Blake, D. R., Christian, T. J., Griffith, D. W. T., Guenther, A. and Hao, W. M. (2007). The Tropical Forest and Fire Emissions Experiment: overview and airborne fire emission factor measurements. *Atmos. Chem. Phys.,* 7, 5175–5196.

Zhang, Q., Streets, D. G., He, K., Wang, Y., Richter, A., Burrows, J. P., Uno, I., Jang, C. J., Chen, D., Yao, Z. and Lei, Y. (2007). NO_X emission trends for China, 1995–2004: The view from the ground and the view from space. *J. Geophys. Res.,* 112, D22306, doi:10.1029/2007JD008684.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, I. T. and Yao, Z. L. (2009a). Asian emissions in 2006 for the NASA INTEX-B mission. *Atmos. Chem. Phys.,* 9, 5131–5153.

Zhang, Q., Streets, D. G. and He, K. B. (2009b). Satellite observations of recent power plant construction in Inner Mongolia, China. *Geophys. Res. Lett.,* 36, L15809, doi:10.1029/2009GL038984.

Zhao, Y., Wang, S., Nielsen, C. P., Li, X. and Hao, J. (2010). Establishment of a database of emissions factors for atmospheric pollutants from Chinese coal-fired power plants. *Atmos. Environ*., 44, 1515–1523.

Zhi, G., Peng, C., Chen, Y., Liu, D., Sheng, G. and Fu, J. (2009). Deployment of coal briquettes and improved stoves: possibly an option for both environment and climate. *Environ. Sci. Technol*., 43, 5586–5591.