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PART A: OZONE AND PARTICULATE MATTER

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Chapter 5
Impacts on Health, Ecosystems, and Climate

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Given the LRT of ozone (O_3) and particulate matter (PM) discussed in previous chapters, this chapter reviews the literature on the impacts of that LRT on human health, ecosystems, and climate. Concerns over human health and ecosystems have been the major drivers for air pollution regulations. In addition, O_3 and PM are important for climate, so actions to reduce emissions relevant for O_3 and PM will also influence climate. Other impacts of the LRT of O_3 and PM that are not addressed in this chapter include the degradation of visibility (mainly due to PM) and damage to materials [*U.S. EPA*, 2006; 2009b].

Figures 5.1 and 5.2 show selected Air Quality Standards (AQS) and guidelines for O_3 and PM from around the world, relative to baseline levels. These standards and guidelines have been motivated to protect health, ecosystems, and visibility, although some are explicitly only motivated for health purposes. Nonetheless, research has not identified clear "threshold" concentrations of O_3 or PM below which there are no adverse health effects [*Krzyzanowski and Cohen*, 2008; *WHO*, 2006] and therefore current standards do not necessarily fully protect human health. For ozone, present-day baseline concentrations approach the lowest of the air quality standards, and as shown in chapters A2 and A4, measured baseline levels for O_3 have sometimes exceeded these standards.

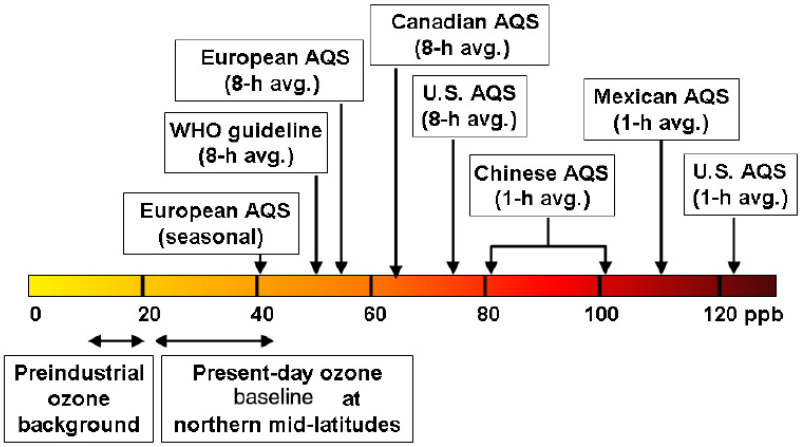


Figure 5.1. Ozone air quality standards in ppb for different nations, as well as international guidelines and estimates of the preindustrial background (i.e., with no anthropogenic emissions) and present-day baseline (i.e., not affected by local pollution). Note that the different standards use different averaging times and the determination of violations also differ among nations. [Reprinted with permission from Figure 2.1 in *Global Sources of Local Pollution: an Assessment of Long-Range Transport of Key Air Pollutants to and from the United States* (2010), by the National Academy of Sciences, Courtesy of the National Academies Press, Washington, D.C.]

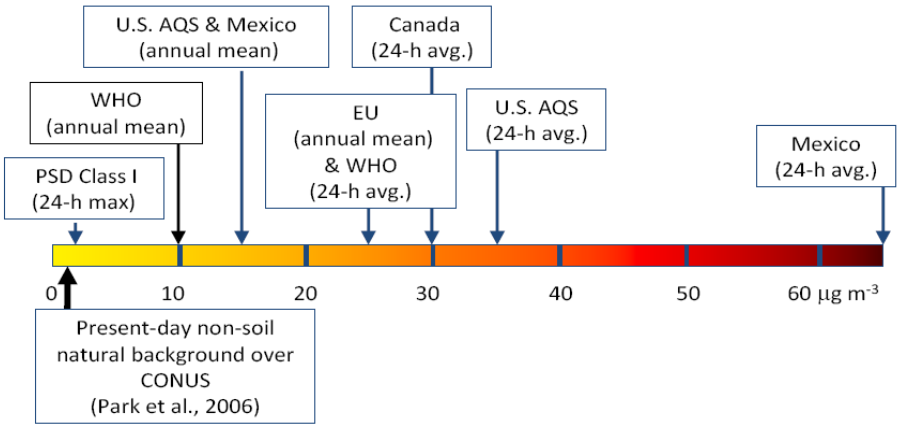


Figure 5.2. Current 24-hr. and annual mean $PM_{2.5}$ standards for selected countries, and U.S. allowable 24-hr emissions increment for Class I areas under the Prevention of Significant Deterioration rule (CONUS is the continental United States). [Reprinted with permission from Figure 3.4 in *Global Sources of Local Pollution: an Assessment of Long-Range Transport of Key Air Pollutants to and from the United States* (2010), by the National Academy of Sciences, Courtesy of the National Academies Press, Washington, D.C.]

5.1 Impacts of Long-range Transport on Human Health

5.1.1. Evidence for effects of ozone and PM on human health

Over the past 20 years, evidence showing the health effects of air pollution has grown exponentially. It is now widely accepted that exposure to outdoor air pollution, a complex mixture of particles and gases, contributes to a broad range of acute and chronic health effects, ranging from minor physiological disturbances to death from respiratory and cardiovascular disease [*ATS*, 2000; *Bascom et al.*, 1996a; b; *Brook et al.*, 2010; *Royal Society*, 2008]. Evidence for these health effects comes in several forms. Epidemiologic studies that find statistical relationships between air pollutant concentrations and health outcomes, looking at effects over large populations, provide the strongest quantitative basis for understanding health impacts of air pollution. These studies also offer the strongest evidence that pollutants have widespread effects in a population, and the large body of epidemiologic studies has provided the foundation for establishing ambient air quality standards and policy guidelines, nationally and internationally. Based on this evidence, WHO estimated that urban outdoor $PM_{2.5}$ caused 800,000 deaths (1.2% of all deaths) and 6.4 million years of lost healthy life globally in the year 2000. Thus, outdoor air quality ranks as the third most important environmental risk factor for mortality in the WHO Global Burden of Disease study, behind unsafe water, sanitation and hygiene, and indoor air pollution from household use of solid fuels [*Cohen et al.*, 2004; *Ezzati et al.*, 2004].

In addition to epidemiologic studies, other types of studies provide strong supporting evidence linking air pollutants and human health: direct human exposures under controlled conditions; laboratory exposures of rodents and other animals; and *in vitro* studies of exposure of cells and tissues to air pollutants [*WHO*, 2006]. In this section, we summarize the epidemiologic research on the health effects of PM and O_3, two components of air pollution mixtures with well-established adverse health effects.

Epidemiologic research plays a key role in air pollution health effects science and in policy, providing estimates of the health effects of both short- and long-term exposure to air pollution in many parts of the world. The epidemiology of air pollution takes advantage of the fact that concentrations of outdoor air pollution, and thus human exposure, vary in both time and space. Short-term temporal variation in concentrations of air pollution over days or weeks has been used to estimate effects on daily mortality and morbidity in time-series studies. They provide estimates of the proportional increase in the number of deaths brought forward in time by recent exposure. Spatial variation in long-term average concentrations of air pollution has provided the basis for cohort studies of long-term exposure. Cohort studies include not only those whose deaths were brought forward by

recent exposure to air pollution, but also those who died from chronic disease caused by long-term exposure [*Künzli et al.*, 2000]; thus, they provide a more complete estimate of the effects and, for this reason, are used to estimate public health impacts in terms of annual attributable deaths and changes in duration of survival [*Brunekreef et al.*, 2007; *Pope III et al.*, 2009]. Estimates of effect are often expressed in relative terms, either as a relative risk or as a percent increase in incidence of mortality in a study population under different exposure conditions, with the precision of the relative risk expressed as a confidence interval.

Epidemiologic studies of the health effects of outdoor air pollution are generally based on measurements of air pollution at fixed monitoring sites that have been established to ensure compliance with air quality standards. Most human populations are exposed to a complex mixture of air pollutants in a variety of indoor and outdoor environments, but studies have found that there are strong correlations between personal exposure to pollutants from outdoor sources and the concentrations measured at fixed monitoring sites [Janssen and Mehta in *WHO*, 2006]. Air pollutant concentrations have been measured routinely in North America and Western Europe since the 1970s; currently, these networks are extensive. Networks are more limited but expanding in Latin America and South and East Asia, but are virtually non-existent in Africa, Central Asia and the Middle East. Consequently, epidemiologic evidence for the health effects of air pollutants is most extensive in North America and Western Europe [*HEI*, 2004; 2010]. In epidemiologic studies the monitored levels are used either alone or as part of more complex geo-spatial models to estimate the individual exposure of study participants [*Jerrett et al.*, 2005]. Neither approach, however, is entirely accurate, and measurement error in exposure remains a concern in all epidemiologic research. Generally, such errors will be smaller for pollutants which tend to be uniformly distributed over large urban areas, and which penetrate efficiently indoors, such as fine PM produced by combustion. If the errors in the estimates of exposure are uncorrelated with the risk of the health outcome, then the estimates of relative risk attributable to air pollution will, in most cases, be too low, but the precision of relative risk estimates will be reduced [*Navidi and Lurmann*, 1995; *Szpiro et al.*, 2008; *Zeger et al.*, 2000].

Epidemiologic studies of the adverse health effects of outdoor air pollution are almost always observational studies. Therefore, the validity of their results requires that they account for other factors related to human health that may be correlated in space or time with exposure to air pollution and whose effects could potentially be confused with those of air pollution. Such factors, termed confounders, include season, meteorology, influenza, tobacco smoking, indoor burning of solid fuels, and access to medical care. In addition, because outdoor air pollution comprises a mixture of pollutants whose concentrations are often highly correlated, estimates of the effects of single pollutants must be interpreted with caution. Epidemiologists address these issues in either the design or the statistical analysis of study data, for example by restricting study populations to those who are unexposed to potential confounders (e.g., non-smokers) or through multivariable regression techniques [*Rothman et al.*, 2008].

FINDING: There is broad consensus that exposure to ambient PM and ozone causes adverse health effects that range from minor sensory irritation to death.

Health effects of particulate air pollution (PM)

Particulate air pollution is a complex mixture whose chemical and physical characteristics vary over space and time due to different emission sources and atmospheric conditions. Small particles emitted directly into the ambient environment or formed in the atmosphere as a result of combustion or mechanical processes may be inhaled into the respiratory tract (inhalable PM or PM_{10}). Smaller particles (respirable PM or $PM_{2.5}$) may be deposited in the deep lung, potentially causing effects on the cardiovascular and other organ systems. There are a variety of potential mechanisms through which inhalable and respirable PM could cause adverse effects on the respiratory and cardiovascular systems, including via effects on neurologic, immunologic and inflammatory responses. Inflammation induced by oxidative stress has recently emerged as a possible common underlying mechanism for a variety of adverse effects of PM [*WHO*, 2006]. Definitive knowledge of which specific characteristics of PM_{10} or $PM_{2.5}$ are responsible for effects is currently lacking, though current epidemiologic and toxicologic findings suggest that certain physical characteristics (e.g., particle size, number and surface area) and

chemical characteristics (e.g., transition metal content) may play a role [*U.S. EPA*, 2009b; *WHO*, 2006]. Because current evidence does not support species-specific PM risk characterization, air quality guidelines from WHO and national regulatory standards specify limits for undifferentiated mass concentrations of PM_{10} or $PM_{2.5}$ (Figure 5.2). There is limited information on the extent to which the LRT of PM alters its toxicologic properties. One recent laboratory study reported that the toxicity of diesel emissions in mice was enhanced by exposure to sunlight and the addition of VOCs. Although aging of diesel PM appears to increase toxicity, it is currently unknown which of the reaction products are responsible, as O_3 also increased [*Zielinska et al.*, 2010].

Hundreds of studies of short-term exposure to air pollution and adverse health effects have been conducted in many regions of the world [*WHO*, 2006]. PM, measured as Total Suspended Particles (TSP), PM_{10}, $PM_{2.5}$, and Black Smoke, has been associated with increases in daily mortality rates from all natural causes, and specifically from respiratory and cardiovascular causes, hospital admissions for respiratory diseases (from all causes, chronic obstructive pulmonary disease, asthma, or pneumonia), and hospital admissions for cardiovascular diseases (from acute myocardial infarction or congestive cardiac failure [*Pope III and Dockery*, 2006]. Exposure to particulate air pollution has also been associated with adverse reproductive outcomes, including low birth weight and pre-term birth [*Glinianaia et al.*, 2004], and acute lower respiratory tract infections in young children [*Romieu et al.*, 2002].

Meta-analytic summaries of the results of individual studies, and coordinated multi-city studies, which apply a common analytic approach to data from multiple cities, now provide the most robust and precise estimates of the effects of short-term exposure to PM_{10} on daily mortality (Figure 5.3). The evidence is consistent among different regions of the world with small relative increases in daily mortality rates, on the order of 0.05 and 0.1 percent increase in daily mortality per 1 $\mu g\ m^{-3}$ PM_{10}. There are fewer reports for $PM_{2.5}$ associations. However, the available evidence from meta-analysis and multi city studies around the world are generally consistent, suggesting associations of around 0.1% per 1 $\mu g\ m^{-3}$ $PM_{2.5}$ (Table 5.1). A recent systematic review and meta-analysis by Smith et al [2009] assessed current evidence for health effects associated with short-term exposure to both sulphate and back carbon (indicated by black smoke) and concluded that short-term exposure time-series studies of black smoke and sulphate PM was associated with increased daily mortality (Table 5.1). For PM associations, larger relative effects tend to be seen in older people and for mortality from cardiovascular and respiratory causes.

PM transported between continents has also been associated with increases in daily mortality. A recent study in Barcelona observed increased daily mortality from all-natural causes associated with both the coarse (PM_{10} minus $PM_{2.5}$) and fine ($PM_{2.5}$) PM components during Saharan dust episodes in 2003-2004 [*Perez et al.*, 2008]. Larger relative effects were reported for exposure to both fine and coarse PM on Saharan dust days versus non-Saharan dust days, but the effects were particularly increased for the coarse fraction. This was not explained by differences in chemical composition, and the authors hypothesized that unmeasured biological agents ("irritants and allergens") might be responsible [e.g., *Griffin et al.*, 2001] .

The literature on exposure to particulate air pollution and mortality in adults from chronic disease has been recently and extensively reviewed [e.g., *U.K. COMEAP*, 2009; *U.S. EPA*, 2009b; *WHO*, 2006]. These reviews, and other efforts specifically designed to elicit the views of the expert community on the strength of the existing scientific evidence [*Cooke et al.*, 2007; *Roman et al.*, 2008; *U.K. COMEAP*, 2009] support the conclusion that exposure to particulate air pollution is causally related to mortality from chronic cardiovascular and respiratory disease and lung cancer.

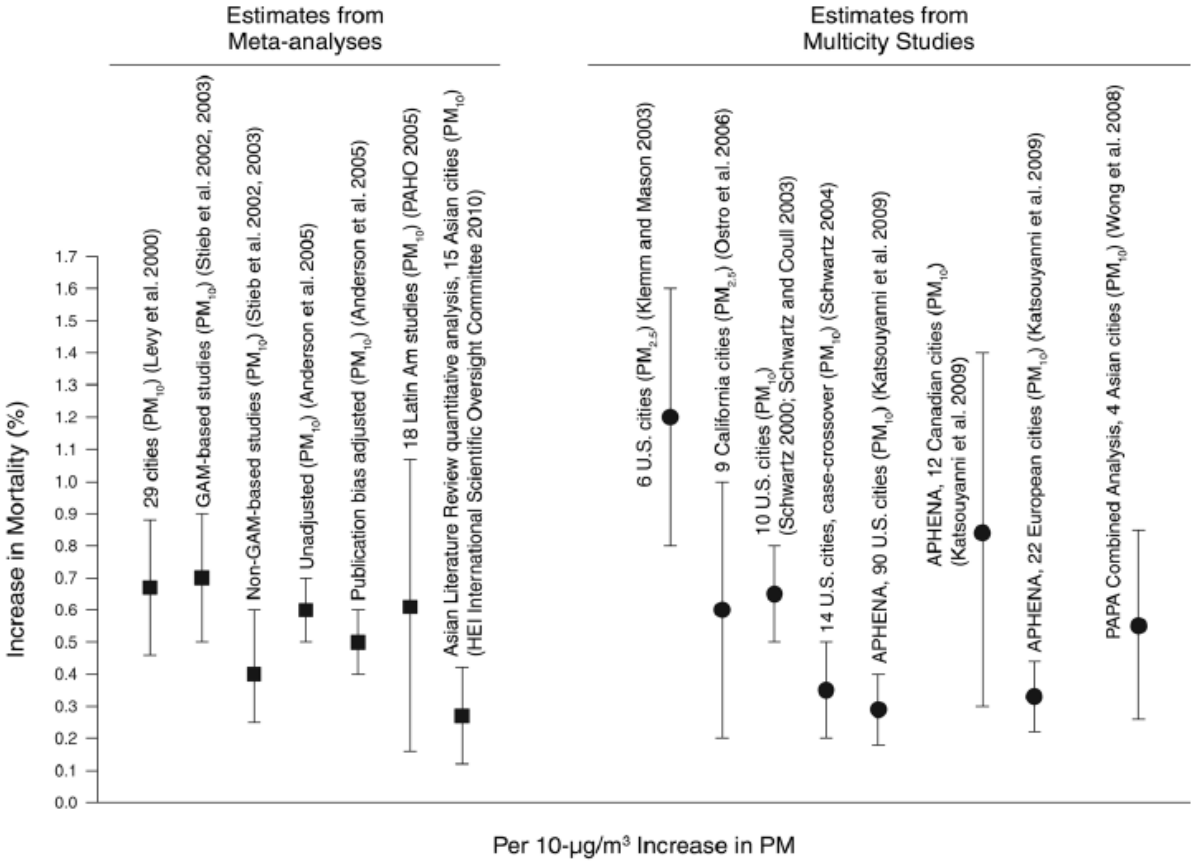
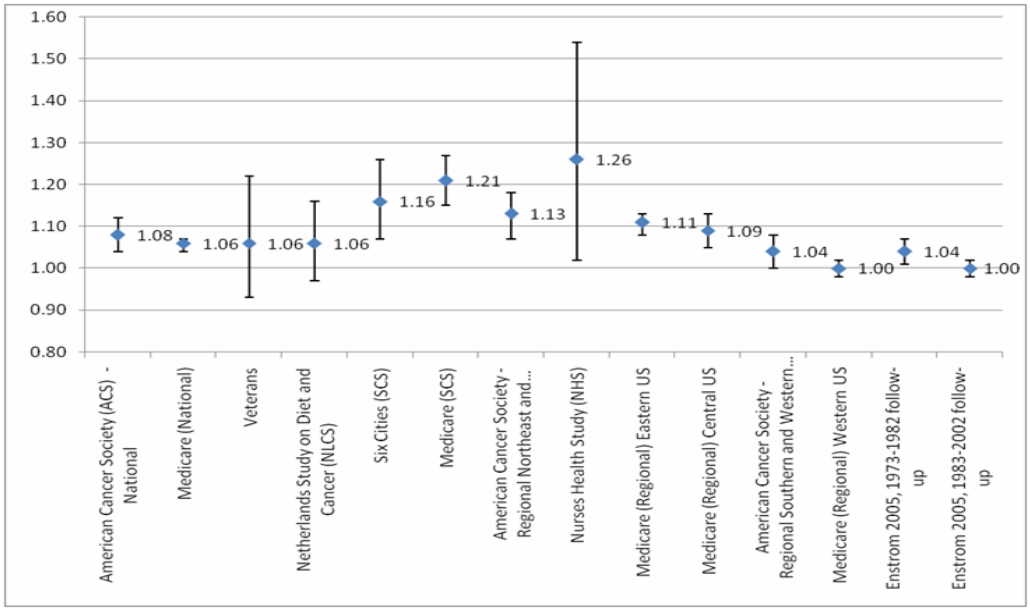


Figure 5.3. Estimates of the effect on all natural mortality per 10 µg m-3 increase in PM reported in several recent meta-analyses and multicity studies. (APHENA = Air Pollution and Health: A European and North American Approach; GAM = generalized additive model; PAPA = Public Health and Air Pollution in Asia program). [Based in part on Katsouyanni, K., et al. (2009), *Air Pollution and Health: A Combined European and North American Approach (APHENA)*, Research Report 142, Health Effects Institute, Boston, MA.]

Although time-series studies of the effects of short-term exposure are numerous and have been conducted in most regions of the world, only nine cohort studies in the US and Europe have estimated the relationship between long-term exposure to $PM_{2.5}$ and mortality (Figure 5.4). No cohort studies of long-term exposure to $PM_{2.5}$ and mortality outside of North America and Europe have been published to-date, although, as noted above, evidence from time-series studies in other regions, such as Asia, find results that are similar to those reported in North America and European studies, as do Asian studies of chronic effects of long-term exposure on respiratory symptoms, asthma, lung cancer and adverse birth outcomes [*HEI*, 2010].

The largest cohort study, and the one that has been used most extensively for quantitative risk assessment, is the American Cancer Society study that relates $PM_{2.5}$ with risks of mortality for adults in 50 US cities. Since the original American Cancer Society study, subsequent studies have extended and reanalyzed the results [*Krewski et al.*, 2009; *Pope III et al.*, 1995; 2002]. Krewski et al. [2009] find results that are similar to, though generally higher than earlier analyses, reporting an increased relative risk of all cause mortality of 0.3% (95% CI of 0.1-0.5%) per 1 µg m^{-3} $PM_{2.5}$, and increases in relative risk for ischemic heart disease (1.5%, 95% CI of 1.1-2%) (Table 5.1). Smith et al. [2009] recently reviewed the evidence regarding the relative toxicity of short-lived greenhouse pollutants, black carbon and sulphate components of $PM_{2.5}$, and ozone. This review included new analyses of the American Cancer Society cohort and concluded that although it was difficult to draw firm conclusions regarding the relative toxicity of the three pollutants, sulphate particles "seem[ed] to have the most robust effects in multi-pollutant models" (Table 5.1).

A



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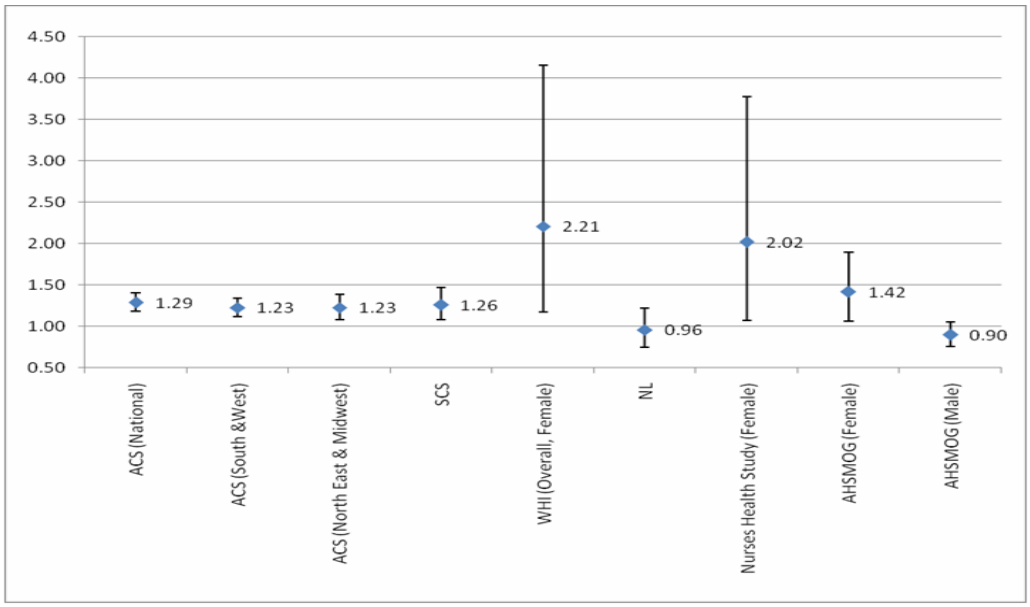


Figure 5.4. Relative risk (95% CIs) of mortality from (A) all-natural causes and (B) ischemic heart disease per 10 µg m-3 PM2.5 from current U.S. and European cohort studies. Based on on-going work of the Global Burden of Disease, Outdoor Air Pollution Expert Group, http://www.globalburden.org/.

Long-term reductions in ambient levels of PM have been associated with reduced mortality rates and improved life-expectancy in the US [*Laden et al.*, 2006; *Pope III et al.*, 2009]. More rapid changes in air pollutant concentrations, including the closing of industrial facilities [*Pope III et al.*, 1992], reductions in residential coal burning [*Clancy et al.*, 2002], and restrictions on the sulphur content of fuel [*Hedley et al.*, 2002] have also been associated with reduced mortality from cardiovascular and respiratory disease.

Table 5.1. Results from selected cohort and time series studies of PM and human mortality

Study	*Outcome*	*Risk estimate*[1]	*95% Confidence Interval*
$PM_{2.5}$			
Cohort studies			
American Cancer Society; Krewski et al. [2009]	All causes	0.3	(0.1 to 0.5)
American Cancer Society; Krewski et al. [2009]	Ischaemic Heart Disease	1.5	(1.1 to 2.0)
Time series studies			
Single-city estimates (n=42); Smith et al. [2009]	All causes	0.10	(0.08 to 0.13)
Multi-city studies			
10 Canadian cities; Brook et al. [2007]	All causes	0.11	(0.01 to 0.21)
25 US Communities; Franklin et al. [2008]	All causes	0.07	(0.04 to 0.11)
5 Australian cities; Simpson et al. [2005]	All causes	0.90	(-0.70 to 2.53)
9 French cities; Blanchard et al. [2008]	All causes	0.16	(0.08 to 0.24)
Sulfate particles			
Time series studies			
Single city estimates (10 studies); Smith et al. [2009]	All causes	0.21	(0.11 to 0.30)
Cohort studies			
American Cancer Society; Smith et al. [2009]	All causes	1.07	(0.73 to 1.40)
American Cancer Society; Smith et al. [2009]	Cardiopulmonary	1.51	1.01 to 2.01)
Carbon			
Time series studies (Black Smoke)			
Single city estimates (25 studies) Smith et al. [2009]	All causes	0.05	(0.03 to 0.07)
Cohort studies(Elemental Carbon)			
American Cancer Society; Smith et al. [2009]	All causes	2.11	(-2.44 to 6.89)
American Cancer Society; Smith et al. [2009]	Cardiopulmonary	2.09	(-4.53 to 9.18)

1 Percent per increment of 1 μg m^{-3} in pollutant

As noted above, short-term exposure to coarse PM has been associated with increased daily mortality. However, there is currently little evidence for a relationship between *long-term* exposure to coarse PM and annual average mortality. Two large cohort studies of women reported no evidence of an effect [*Miller et al.*, 2007; *Puett et al.*, 2008]. A smaller cohort study reported increased mortality in women (but not men), but the results were inconsistent across a range of statistical models [*Chen et al.*, 2005]. The effects of long-term exposure to PM on mortality from chronic cardiovascular and respiratory disease have only been studied over the range of ambient concentrations that exist in the US and Western Europe: approximately 5–30 μg m^{-3}. Over that range the concentration-response relationship appears to be both linear and monotonic [*Krewski et al.*, 2009]. Because no cohort studies of mortality have been conducted in Asia or other regions with higher annual average PM concentrations, the concentration-response function at those higher levels is currently unknown. This contributes considerable uncertainty to global quantitative risk assessments of air pollution [*Anenberg et al.*, 2010; *Cohen et al.*, 2004], including the assessments of impacts of LRT in Section 5.1.2. Additional sources of uncertainty in such risk assessments include uncertainties in the baseline rates of mortality and levels of exposure to PM in many low- and middle- income countries (see Section 5.1.4).

FINDING: Short-term exposure to PM is associated with increased daily mortality and morbidity in hundreds of studies worldwide; long-term exposure to $PM_{2.5}$ is associated with

increased mortality from chronic cardiovascular and respiratory disease in multiple studies. Adverse effects appear to extend linearly to levels below current air quality standards with no evidence of a threshold. Although ambient air pollution is a complex mixture, most assessments of the public health impacts of ambient air pollution have focused on PM, because of the strength and consistency of the evidence for PM. Current impact estimates are subject to considerable uncertainty due to lack of information on the concentration-response function for PM and mortality in the most highly polluted regions.

Health effects of ozone

The adverse effects of both short- and long-term exposure to O_3 are well-documented and have been recently reviewed [*U.S. EPA*, 2006; *WHO*, 2006]. Experimental evidence from studies in humans and animals indicates that short-term exposure causes the exacerbation of existing lung diseases, and toxicologic and epidemiologic studies indicate that long-term exposure can produce irreversible changes in the lung structure and function.

Ground-level O_3 is formed by the chemical reaction between NO_x and VOCs in the presence of sunlight; so ambient concentrations and human exposure, are typically higher in warm seasons. Experimental studies in animals designed to simulate these conditions appear to indicate cumulative impacts of seasonal exposure, and long-term exposure to high seasonal levels of O_3 have been associated with reduced lung function growth in children [*Gauderman et al.*, 2000; 2002; *Rojas-Martinez et al.*, 2007]**.**

Daily time series studies of O_3 and mortality have shown consistent adverse health effects. These include many single-city studies which have then been analyzed in meta-analyses [*Anderson et al.*, 2004; *Bell et al.*, 2005; *Ito et al.*, 2005; *Levy et al.*, 2005; *Thurston and Ito*, 2001] and large multi-city studies [*Bell et al.*, 2004; *Gryparis et al.*, 2004; *Katsouyanni et al.*, 2009] (Table 5.2). These results also illustrate the variability in the size and precision of effect estimates observed in different studies. The recent APHENA study, a multi-city study of over 100 cities in North America and Europe [*Katsouyanni et al.*, 2009], estimated that all-cause mortality increased by 0.52% for a 10 ppb increase in O_3 across the two continents, with considerably larger values and imprecision for Canadian studies. Adverse health effects of O_3 on mortality have also been observed at concentrations below current air quality guidelines. If a threshold exists, a concentration below which ozone has no measurable effects, then it is likely at concentrations below the current air quality standards [*Bell and Dominici*, 2006; *Gryparis et al.*, 2004; *Katsouyanni et al.*, 2009; *U.S. EPA*, 2006].

There is currently limited evidence that long-term exposure to O_3 causes increased mortality from chronic cardiovascular or respiratory disease. Recently, however, Jerrett et al. [2009] reported an association between long-term exposure to O_3 and respiratory mortality in the American Cancer Society cohort (Table 5.2). A 10 ppb increase in O_3 is associated with a 4% (95% confidence interval, 1.3-6.7%) increase in relative risk of mortality from non-malignant respiratory disease. This study, however, did not detect an increase in cardiovascular mortality that could be attributed to O_3 independent of the influence of $PM_{2.5}$. An earlier study had reported increased respiratory mortality associated with long-term exposure to O_3 with more pronounced increases in men than women [*Abbey et al.*, 1999]. Given well-documented acute and chronic effects of exposure to O_3 on the respiratory system, these associations seem plausible.

FINDING: Short-term exposure to ozone is associated with increased daily mortality and morbidity in many studies worldwide. However, the current evidence linking long-term exposure to ozone with mortality from cardiovascular and respiratory disease is limited. Adverse effects appear to extend linearly to levels below current air quality standards with no evidence of a threshold.

Table 5.2. Results from selected cohort and time series studies of ozone and human mortality

Study	*Outcome*	*Risk estimate*[1]	*95% Confidence Interval*
Cohort studies			
American Cancer Society; Jerrett et al. [2009]	All causes	0.1	(-0.4 to 0.7)
	Cardiovascular	1.1	(0.3 to 2.3)
	Respiratory	2.9	(1.0 to 4.8)
Time series studies[2]			
Multi-city studies			
95 US urban communities; Bell [2004]	All causes	0.52	(0.27 to 0.77)
23 European cities; Gryparis et al. [2004]	All causes	0.06	(-0.36 to 0.42)
APHENA (Canada, US, Europe); Katsouyanni et al. [2009]	All causes	0.52	(0.30 to 0.74)
Meta-analysis (24-hr)			
Thurston & Ito [2001]	All causes	0.89	(0.56 to 1.22)
Stieb et al [2003]	All causes	1.12	(0.32 to 1.92)
Levy et al [2001]	All causes	0.98	(0.59 to 1.58)
Anderson et al [2004]	All causes	1.11	(0.55 to 1.67)
Bell et al [2005]	All causes	0.87	(0.55 to 1.18)

1 Percent per increment of 10 ppb in ozone

2 Results are for daily mean ozone concentration except Gryparis et al. [2004] where the daily 8-hr maximum is used

Evidence for interaction between PM and ozone

Although air pollution is always present as a complex mixture of particles and gases, the overwhelming majority of studies have estimated the effects of single pollutants, sometimes controlling analytically for the effects of other pollutants. Few studies have explored whether the effects of specific components of the mixture enhance or suppress the effects of others, and as a result, there is relatively little information about whether the health effects of exposure to PM or O_3 are dependent on the co-existing levels of the other pollutants in space or time. The APHENA study [*Katsouyanni et al.*, 2009], which analyzed data from over 120 major cities in Europe and North America, observed that in the US cities with higher levels of O_3, the effects of short-term exposure to PM_{10} on daily mortality among the elderly (>=75 yr.) were smaller. However, a study in Shanghai reported that the percent increase in daily mortality associated with PM_{10} was increased at higher levels of O_3 and that the effect of O_3 was increased at higher levels of PM_{10} [*Chen et al.*, 2005]. Katsouyanni et al. [2009] also reported that the effects on daily mortality of short-term exposure to PM_{10} were greater at higher levels of NO_2 (an ozone precursor) and when the NO_2 /PM_{10} ratio was higher, a pattern more pronounced in Europe than the US.

5.1.2. Quantified influences of long-range transport on human health

Although the influence of particular emission sources on atmospheric concentration is generally greatest near those sources, LRT influences pollutants far downwind. As pollutants move to downwind regions and continents, much larger populations may be exposed to this change in concentration. The total risks for human health in downwind continents may be comparable to the risks within the source continent, even if the effects on the concentrations that people breathe are much smaller [e.g., *Anenberg et al.*, 2009].

Several studies have specifically modelled the LRT of O_3 and PM, and have estimated the effects of this LRT on premature mortality. These several studies have all used similar methods, and uncertainties in these methods should be addressed in future research. First, all studies used global

CTMs to represent LRT. While these coarse resolution CTMs (typically 200 km in horizontal resolution) are necessary to represent LRT, their use for health impact analysis may cause uncertainties in concentrations and exposures near urban regions, where there are strong gradients in population density and pollutant concentrations and urban chemistry may modify the effects of LRT (Chapter A4). Second, all studies apply concentration-response relationships from epidemiological studies in industrialized nations to the rest of the world. While some studies in developing nations suggest that the same relationships may hold, the body of evidence is weaker. These uncertainties can be reduced when considering cause-specific mortalities and when using appropriate baseline mortality rates based on observed disease incidence in different world regions. Therefore, while epidemiological studies provide strong evidence for quantifying health impacts, and while it is common to use regional CTMs to drive health impact analyses within North America or Europe, the use of global CTMs for global health impact analyses should be a focus of future research to improve methods [*NRC*, 2010]. Recommendations to resolve these uncertainties are in Section 5.1.4.

FINDING: LRT of ozone and PM can have widespread effects on human health in other continents, as the combined populations of affected regions are likely greater than that of the source region.

Impacts of the long-range transport of ozone on premature mortality

Modelled contributions of LRT to O_3 concentrations have been shown to be significant causes of premature mortality. Anenberg et al. [2009] modelled the effects of intercontinental O_3 from the HTAP SR6 model simulations, in which anthropogenic emissions of NO_x, non-methane volatile organic compounds (NMVOCs), and carbon monoxide (CO) were reduced by 20% from each of the four HTAP source regions (see Chapter A4). Avoided premature cardiopulmonary mortalities were calculated for the multi-model mean change in O_3, as shown in Table 5.3 and Figure 5.5. Changes in emissions from any of the four regions have widespread effects throughout the Northern Hemisphere, influencing mortality in population centres on other continents.

When the influences of all four source regions are combined, foreign emissions contribute roughly 30%, 30%, 20% and >50% to the avoided mortalities in the receptor regions NA, EA, SA, and EU (Table 5.3). In particular, EU is shown to be influenced strongly by emissions from NA. The reductions in emissions in NA and EU are estimated to cause more avoided mortalities outside of these source regions than within, due to the large populations affected in downwind regions. In addition, many CTMs show small decreases in local O_3 within parts of the NA and EU source regions, due to nonlinear chemical processes, which contributes to the small numbers of avoided mortalities in these regions relative to the whole hemisphere.

Anenberg et al. [2009] also found that for some source-receptor pairs, the uncertainty in the avoided premature mortalities associated with the modelled O_3 responses is greater than the uncertainty caused by the health impact function parameters (Figure 5.6), although neither the range of CTM responses nor the confidence intervals on relative risk fully reflect the total uncertainty. In particular, the several models analyzed showed a large discrepancy in the response of O_3 to changes in precursor emissions within the source region. The uncertainties in source-receptor responses were large enough to change the conclusions above about the relative numbers of avoided mortalities within versus outside of the source regions.

FINDING: For ozone, one study based on the HTAP multi-model comparison found that ozone resulting from emissions from foreign regions is estimated to contribute 20% to >50% of ozone mortalities, subject to large uncertainty.

The finding that O_3 precursor reductions in NA and EU cause more avoided mortalities outside of the source regions than within is supported by Duncan et al. [2008] and West et al. [2009]. Duncan et al. [2008] modelled the full contribution of European emissions on nearby regions by removing anthropogenic emissions from Europe. They found that Europe has a large impact on surface O_3 concentrations in Northern Africa, the Middle East, and the Former Soviet Union, and smaller influences elsewhere in the Northern Hemisphere. While O_3 caused by European emissions

causes ~18,000 annual premature mortalities within Europe, Duncan et al. [2008] find a total of ~51,000 mortalities in the Northern Hemisphere.

Table 5.3. Annual avoided premature cardiopulmonary mortalities (hundreds) following 20% NO_x, NMVOC, and CO emission reductions in each region, for the HTAP SR6 multi-model mean (relative to SR1), assuming no concentration threshold (italics) and assuming a concentration threshold of 35 ppb (normal font). Confidence intervals (95%) reflect uncertainty in the CRF only. [Reprinted with permission from Table 2 in Anenberg, S., et al. (2009), Intercontinental impacts of ozone pollution on human mortality, *Environmental Science and Technology*, *43*(17): 6482-6487. Copyright 2009 American Chemical Society.]

Source Region	Receptor Region				
	NA	EA	SA	EU	NH
NA	*9 (4 - 13)*	*7 (3 - 10)*	*6 (3 -9)*	*11 (5 – 17)*	*36 (18 – 55)*
	9 (4 - 14)	4 (2 - 6)	5 (3 – 8)	6 (3 – 9)	27 (13 – 41)
EA	*2 (1 - 3)*	*43 (21 – 66)*	*6 (3 – 9)*	*5 (3 – 8)*	*59 (29 – 91)*
	1 (1 - 2)	40 (19 – 61)	5 (2 – 8)	3 (1 – 4)	49 (24 – 76)
SA	*1 (0 - 1)*	*4 (2 – 6)*	*76 (37 – 117)*	*2 (1 – 3)*	*85 (41 – 130)*
	0 (0 - 1)	3 (1 – 4)	66 (32 – 101)	1 (0 – 2)	71 (34 – 108)
EU	*2 (1 - 3)*	*8 (4 – 12)*	*6 (3 – 10)*	*17 (8 – 26)*	*38 (18 – 58)*
	1 (0 - 1)	6 (3 – 8)	6 (3 – 9)	25 (12 – 38)	40 (19 – 61)

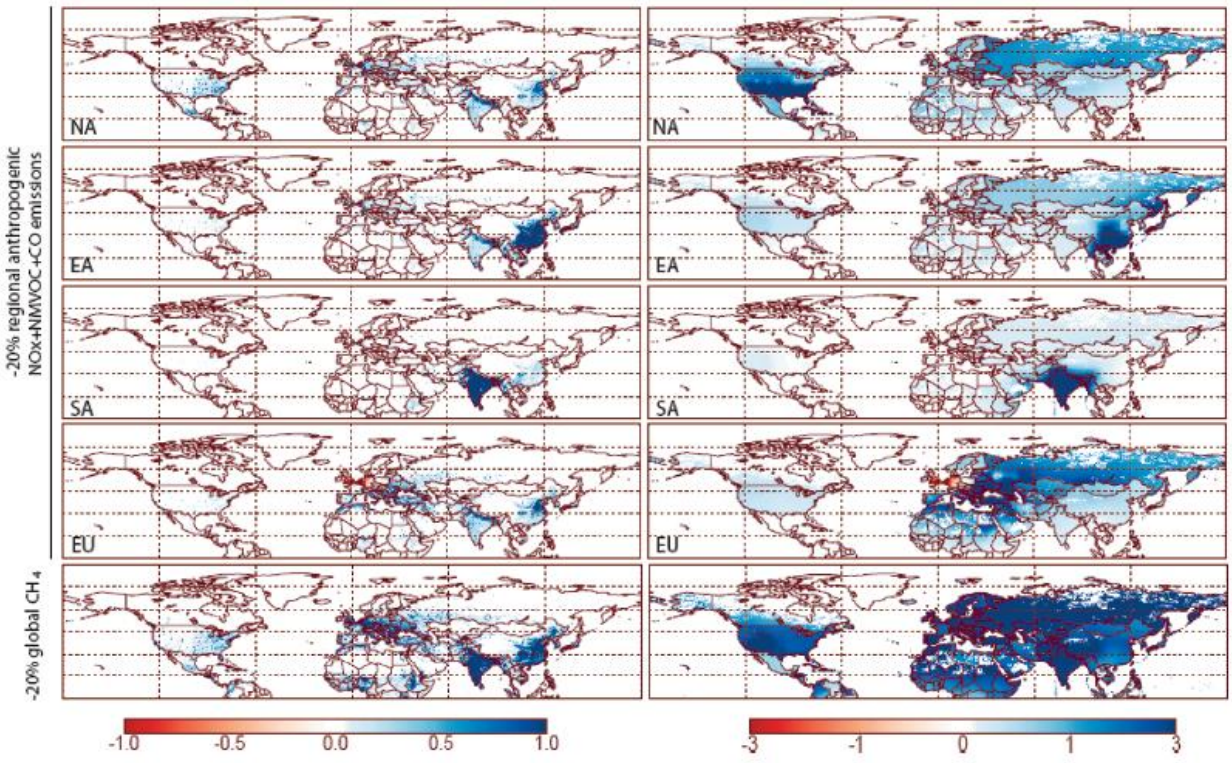


Figure 5.5. Annual avoided premature cardiopulmonary mortalities per 1,000 km^2 (left) and per million people (right) resulting from 20% NO_x, NMVOC, and CO emission reduction in the region shown and a 20% global methane mixing ratio reduction, assuming no low-concentration threshold. [Reprinted with permission from Figure 1 in Anenberg, S., et al. (2009), Intercontinental impacts of ozone pollution on human mortality, *Environmental Science and Technology*, *43*(17): 6482-6487. Copyright 2009 American Chemical Society.]

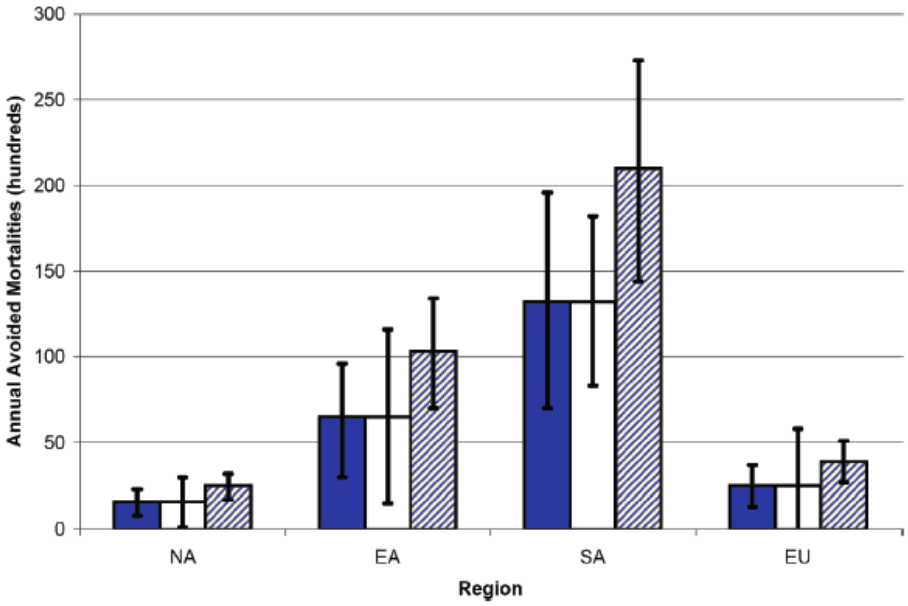


Figure 5.6. Annual avoided premature nonaccidental mortalities (hundreds) in each region from 20% NO_x, NMVOC, and CO emission reductions in the same region using the concentration-response function (CRF) and confidence interval (95%) from Bell et al. [2004] (solid bars), using the CRF from Bell et al. [2004] and confidence intervals (68%) from ±1 standard deviation of the model ensemble O_3 perturbation in each grid cell [*Fiore et al.*, 2009] (white bars), and using the mean and confidence intervals (95%) of the CRFs from three meta-analyses of O_3 mortality [*Bell et al.*, 2005; *Ito et al.*, 2005; *Levy et al.*, 2005] (striped bars). [Reprinted with permission from Figure 2 in Anenberg, S., et al. (2009), Intercontinental impacts of ozone pollution on human mortality, *Environmental Science and Technology, 43*(17): 6482-6487. Copyright 2009 American Chemical Society.]

West et al. [2009] analyzed 10% reductions in anthropogenic NO_x emissions from each of nine world regions, and 10% combined reductions of O_3 precursors from three world regions. Reductions in emissions from North America and Europe each caused more avoided mortalities outside of these source regions than within, for the NO_x reductions, combined O_3 precursor reductions, and for short-term and steady-state results. The same was also true for NO_x reductions from the Former Soviet Union. In analyzing the changes in long-term O_3 via methane, NO_x reductions in one hemisphere were estimated to increase O_3 and premature mortalities in the opposite hemisphere, as NO_x reductions increase the lifetime of methane (see Chapter A4). In fact, the NO_x reduction in Australia was found to cause a net increase in mortality, as the long-term increase in O_3 in the Northern Hemisphere was shown to dominate (Figure 5.7).

Reductions in global methane emissions and impacts on O_3 and health have also been estimated by West et al. [2006] and Anenberg et al. [2009]. For the HTAP SR2 simulations, a 20% decrease in the global methane mixing ratio was shown to result in 16,000 avoided premature cardiopulmonary mortalities annually in the Northern Hemisphere, with the greatest numbers in highly populated regions [*Anenberg et al.*, 2009]. These results agree roughly with those of West et al. [2006], after accounting for differences in study design, who found that a 20% decrease in global anthropogenic methane emissions (in 2010) would result in ~30,000 avoided premature mortalities in 2030 globally, and ~370,000 accumulated over the period 2010-2030. These benefits of avoided mortalities exceed the marginal costs of methane reduction using known technologies, depending on the valuation of mortalities, and can justify the 20% methane reduction without considering non-health benefits of reduced O_3 and decreased climate change (see Section 5.3) [*West et al.*, 2006].

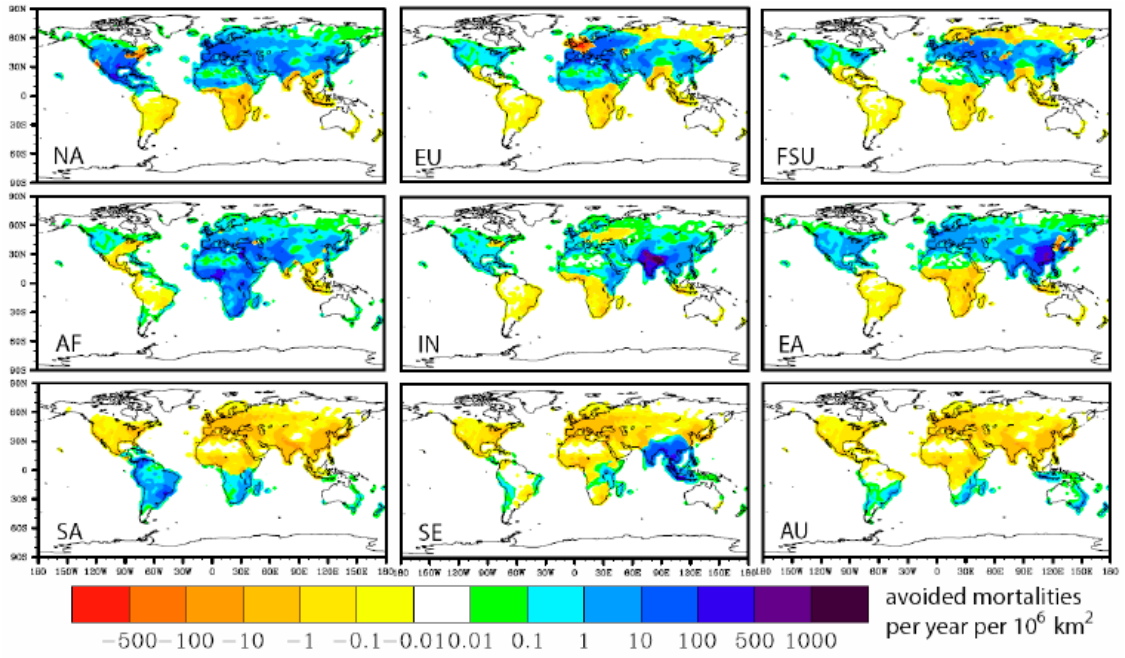


Figure 5.7. Annual avoided premature moralities from 10% anthropogenic NO_x reductions in each of nine world regions, at steady state and using a low-concentration threshold of 25 ppb. [Reprinted from Figure 3 in West, J. J., et al. (2009), Effect of regional precursor emission controls on long-range ozone transport - Part 2: Steady-state changes in ozone air quality and impacts on human mortality, *Atmospheric Chemistry and Physics*, *9*(16): 6095-6107.]

All of the studies of O_3 mortality described here use the same daily time series study [*Bell et al.*, 2004] to give the concentration-response function, and comparable but not identical population and baseline mortality rates, but make different assumptions about a low-concentration threshold, below which changes in O_3 are assumed to have no effect on premature mortality, and may report results for total (non-accidental) mortality or for cardiovascular and respiratory mortality. More recent findings suggest that O_3 may influence chronic mortality to a greater extent than is represented by the daily time series studies [*Jerrett et al.*, 2009]; were these studies to be reproduced with new concentration-response functions, the total avoided mortalities would likely be greater, but the relative importance of source-receptor pairs would not be likely to change.

FINDING: Three studies have estimated that reductions in ozone precursor emissions may avoid more premature mortalities outside of some source regions than within, mainly because of larger populations outside of those source regions.

Impacts of the long-range transport of PM on premature mortality

LRT of PM can adversely impact public health in downwind regions. In Chapter A4, source-receptor relationships for aerosols were presented, based on the HTAP SR6 multi-model experiments. A new analysis conducted for this report examined the impacts of these source-receptor relationships on premature mortality. Mortalities were calculated using the median annual average surface $PM_{2.5}$ (taken as the sum of black carbon (BC), polycyclic/particulate organic matter (POM), and sulphate (SO_4)) response from the multi-model SR6 simulations of regional 20% primary PM and PM precursor emissions reductions, and using the long-term concentration-response factor from Krewski et al. [2009] for adults age 30+. The same concentration-response factor is assumed to apply to all chemical components of $PM_{2.5}$. Methods for estimating mortalities and baseline mortality rates are comparable to those used previously for ozone [*Anenberg et al.*, 2009].

The results show that every region is influenced most by its own emissions (Table 5.4). In contrast to O_3, the large majority of mortalities in each receptor region is due to emissions from within that region, with only 3-5% resulting from emissions in the three other regions. This finding for

mortality is consistent with the findings in Chapter A4 that the import sensitivities for PM are less than that for O_3.

Table 5.4. Annual avoided premature cardiopulmonary and lung cancer mortalities (in hundreds) due to 20% reductions in primary $PM_{2.5}$ and PM precursor emissions in each region, assuming no concentration threshold and assuming a low-concentration threshold of 5.8 μg m^{-3} (italics). Confidence intervals (95%) reflect uncertainty in the CRF only.

Source Region	Receptor Region				
	NA	EA	SA	EU	World
NA	103 (76-130) *71 (52-89)*	4 (3-5) *4 (3-5)*	2 (1-2) *2 (1-2)*	10 (7-12) *8 (6-10)*	121 (90-152) *84 (62-105)*
EA	2 (1-2) *1 (1-1)*	941 (715-1161) *926 (703-1142)*	5 (4-6) *5 (4-6)*	4 (3-4) *3 (2-4)*	958 (727-1182) *935 (710-1154)*
SA	0 (0-0) *0 (0-0)*	8 (6-10) *8 (6-10)*	442 (342-541) *428 (330-523)*	1 (1-1) *1 (1-1)*	455 (351-556) *437 (337-534)*
EU	2 (1-2) *1 (1-1)*	16 (12-20) *16 (12-20)*	14 (11-17) *11 (9-14)*	364 (256-451) *293 (221-362)*	413 (313-511) *321 (243-398)*

While PM has lower import sensitivities than O_3, its concentration-response factor for human mortality is also stronger. Consequently, the total mortalities avoided by 20% emission reductions are much greater for PM, with most of those benefits realized within the source regions. We find that the inter-continental influences on mortality are comparable for PM and O_3; inter-continental O_3 mortalities in Table 5.3 are greater for O_3 for most source-receptor pairs, but are greater for PM for pairs not separated by an ocean – EU on EA, EU on SA, and SA on EA. Because of the large uncertainties in atmospheric modelling (including the fact that we do not account for all components of PM) and in applying concentration-response functions globally, and because of new findings suggesting chronic mortality effects of O_3 [*Jerrett et al.*, 2009], we conclude that the influences of LRT on human mortality are comparable for O_3 and PM.

Among the source regions, the 20% reduction in EU has the largest impact on the other three regions (~3,200 premature mortalities annually), while the reduction in EA causes the greatest overall decrease in mortality (~96,000) due to the large population density and high PM concentrations. Of the total avoided mortalities from the emissions in EA and SA, only 2% and 3% result outside of those source regions, while 15% and 12% occur outside of NA and EU due mainly to smaller populations within those source regions. In total, transport of PM from the four HTAP regions is estimated to result in ~9,600 avoided mortalities annually (for the 20% reductions).

Since the $PM_{2.5}$ concentration-response factor from Krewski et al. [2009] was based on concentrations ranging from 5.8-30 μg m^{-3}, effects of $PM_{2.5}$ on mortality outside of that range are unknown. Applying a high-concentration threshold of 30 μg m^{-3}, above which additional $PM_{2.5}$ is assumed to have no additional impact on health, has no impact on these results, since modelled annual average concentrations for all scenarios are below the threshold. However, applying a low-concentration threshold of 5.8 μg m^{-3} causes the estimated avoided mortalities to decrease. The low-concentration is much more important for the receptor regions NA and EU (32% and 20% reductions) than for EA and SA, since PM concentrations are generally lower and therefore more frequently below the threshold. Here, the definition of $PM_{2.5}$ includes only BC, POM, and SO_4, and inclusion of

dust, nitrates, and other $PM_{2.5}$ components would likely raise concentrations in many cases to above the low-concentration threshold, reducing the impact of the threshold on estimated mortalities.

Related studies by Liu et al. [2009a; 2009b] tracked the intercontinental transport of $PM_{2.5}$ by tagging aerosols (including sulphate, black carbon, organic carbon, and mineral dust) originating from 10 continental regions in year 2000, and quantified the global impact of this transport on premature mortality. They found that the simulated annual mean population-weighted concentrations of total $PM_{2.5}$ were highest in East Asia (EA, 30 $\mu g\ m^{-3}$) and lowest in Australia (3.6 $\mu g \cdot m^{-3}$). The global annual premature mortalities (for adults over age 30) due to inter-continental transport of $PM_{2.5}$ were nearly 380,000 in 2000 (95% confidence interval of 68,000-910,000). Figure 5.8a shows that approximately half of these deaths occurred in the Indian subcontinent (IN), mostly due to dust aerosols transported from Africa and the Middle East (ME). Dust is the dominant component of $PM_{2.5}$ transported between continents and approximately 290,000 deaths globally were associated with exposure to foreign (i.e., originating outside a receptor region) dust $PM_{2.5}$. Intercontinental transport of non-dust aerosols accounted for nearly 90,000 deaths in 2000. As shown in Figure 5.8b, more than half of the premature mortalities associated with foreign non-dust aerosols are due to aerosols originating from Europe, ME and EA; and nearly 60% of the 90,000 deaths occur in EA, IN, and Southeast Asia. These estimates are substantially greater than those in Table 5.4 (after multiplying by five because of the 20% reduction) mainly because of the inclusion of natural dust emissions.

Influence potentials (IPs), which quantify the human exposure that occurs in a receptor region as a result of a unit of emissions from a source region, were developed in Liu and Mauzerall [2007]. Inter-continental IPs are usually less than domestic IPs by 1–3 orders of magnitude. They found high influence potentials connecting Eurasian regions with Africa (Figure 5.9), and found that NA is impacted less by foreign $PM_{2.5}$ than are receptors in Eurasia. However, in 2000 nearly 6,600 premature deaths in North America (NA) were associated with exposure to foreign $PM_{2.5}$ (5,500 from dust $PM_{2.5}$) [*Liu et al.*, 2009b].

The conclusion that PM causes most mortalities domestically was supported by Saikawa et al. [2009], who analyzed the effect of China's emissions of SO_2, SO_4, organic carbon (OC) and BC on PM concentrations and mortality in 2000 and for three emission scenarios in 2030. In 2000, aerosols from Chinese emissions were estimated to have caused approximately 470,000 premature deaths in China and an additional 30,000 deaths globally. In 2030, aggressive emission controls were found to reduce premature deaths in China by half (relative to 2000) and to 10,000 elsewhere, while under a high emissions scenario premature deaths increased 50% in China and to 40,000 elsewhere [*Saikawa et al.*, 2009].

Finally, Corbett et al. [2007] used similar methods to model the contribution of ship emissions to global $PM_{2.5}$ concentrations and related premature mortalities. They estimated that ~60,000 cardiovascular and lung cancer mortalities can be attributed to ship emissions annually, with most of these mortalities near the coastlines of Europe, East Asia, and South Asia.

FINDING: Intercontinental transport of PM is estimated to cause human mortalities that are comparable to those estimated for ozone, due to the stronger relationships between PM and mortality, and despite the smaller fraction of PM being transported between continents. A large majority (>80%) of the health benefits of reductions in PM are expected to be realized within the source continent.

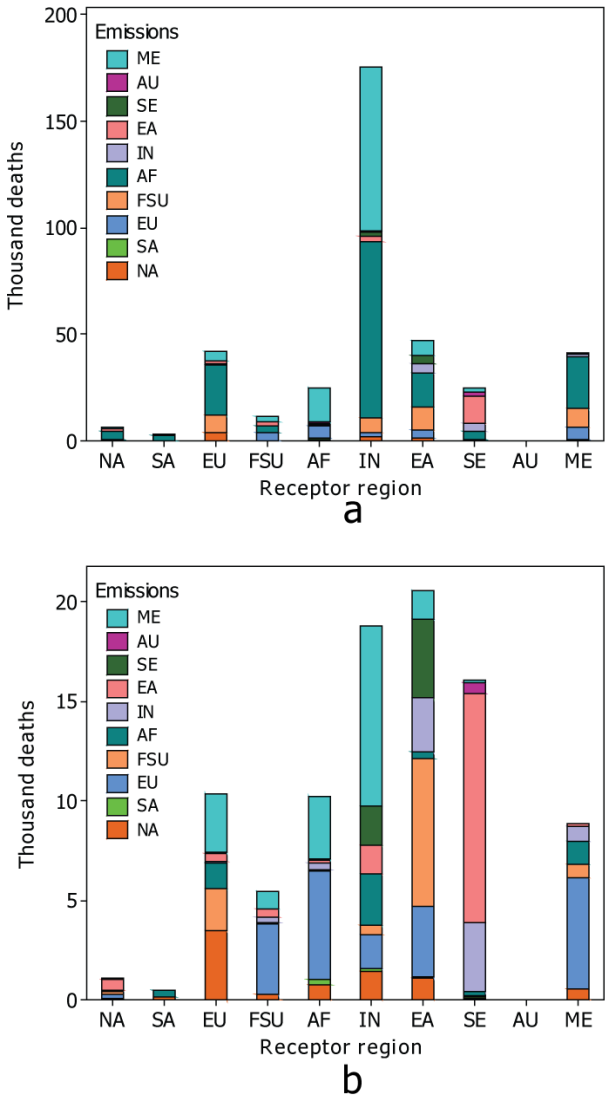


Figure 5.8. Annual adult (age 30 and over) premature mortalities in each receptor region associated with inter-continental transport of (a) fine aerosols ($PM_{2.5}$) (b) non-dust aerosols from the nine other source regions. [Reprinted from Figure 2 in Liu, J., et al. (2009a), Evaluating inter-continental transport of fine aerosols: (2) Global Health Impacts, *Atmospheric Environment, 43*(28): 4339-4347.]

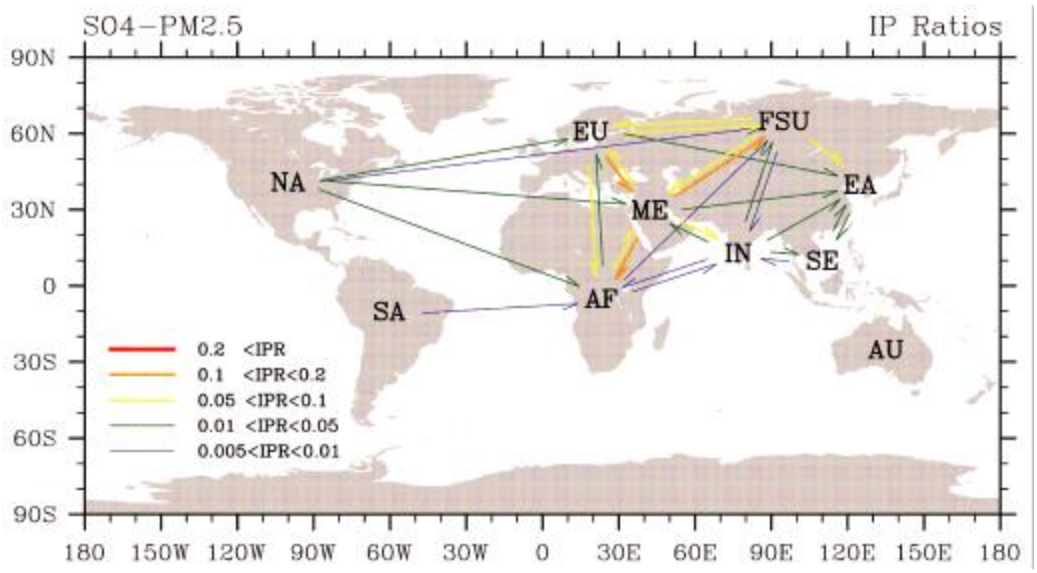


Figure 5.9. Influence potential ratios (IPR) of inter-continental transport of fine ($PM_{2.5}$) sulphate aerosols. Arrows indicate the influence direction from a source to a receptor region. Colours indicate the magnitudes of IPR ranging from red (strong influence) to blue (weak influence). Arrows with IPR less than 0.005 are not shown. [Reprinted from Figure 5 in Liu, J., and D. L. Mauzerall (2007), Evaluating the potential influence of inter-continental transport of sulphate aerosols on air quality, *Environmental Research Letters, 2*(4): 045029.]

5.1.3. Future changes in human health impacts due to LRT

In Chapter A3, the future emissions of air pollutants from different regions of the world are presented for the "Representative Concentration Pathway" (RCP) scenarios. These scenarios show that emissions of ozone precursors, and PM and its precursors are generally expected to decline in NA and EU to 2050, but depending on the pollutant and emissions scenario, will increase in SA and EA before declining. CTM simulations with future emission scenarios (Chapter A4) suggest that for ozone the importance of foreign emission changes compared to domestic changes is likely to increase substantially for NA and EU, as domestic emissions fall and emissions from EA and SA continue to grow. The ozone-related health impacts due to LRT of EA and SA emissions are therefore likely to be enhanced in the future. Since, for most future RCP emission scenarios, ozone concentrations in NA and EU decline in 2050 (Figure 4.45; section 4.6.1), ozone-related human health impacts due to LRT from these two source regions would be expected to lessen. While PM in these scenarios is not modelled in Chapter A4, the emissions scenarios project similar relationships, suggesting that the relative importance of foreign emissions for PM concentrations in NA and EU may also grow.

The impact of climate change is such that when applying the same emission reductions of ozone precursors to the same baseline emissions for a present-day (2000s) and for a future (2090s) climate, the impact on ozone concentrations is slightly greater in the future as compared to present-day (Section 4.6.2). In terms of health impacts it is likely that the downwind ozone-related effects will be reduced due to climate change. The impact of climate change on future LRT of PM has not been modelled.

While ozone LRT may decrease in the future as emissions decline and because of climate change, populations are also likely to increase, particularly in less industrialized nations, leading to greater exposure to LRT and pollutants in general, and the total incidence of mortality caused by LRT may increase [e.g., *West et al.*, 2007b]. Future estimates of health effects should account for changing populations and health characteristics. Improved health care globally may increase life expectancy, possibly making populations more susceptible to cardiopulmonary disease and therefore to health effects associated with air pollutants.

5.1.4. Major uncertainties and research needs

Quantitative estimates of the health impacts of air pollutants have large uncertainties, and assessments of the health effects associated with LRT compound several uncertainties. As discussed

earlier, the existing studies of LRT influences on mortality apply concentration-response functions from the US and Europe globally, assume that all components of PM are equally toxic, and ignore possible interactions between pollutants for human health. There are also important uncertainties related to the use of coarse resolution global models, which are not designed to represent urban concentrations well [Chapter A4, *Lin et al.*, 2010]. The overall uncertainties are likely greater than represented in the individual studies in Section 5.1.2, because the uncertainty in concentration-response functions from a single epidemiological study does not reflect the full scientific uncertainty, and because of the combination of factors discussed here. The approaches used in Section 5.1.2 reflect the current state of knowledge and analytical tools, and there are clear opportunities to reduce these uncertainties through future research.

RECOMMENDATION: Estimates of health impacts of air pollutants and long-range transport are uncertain and can be improved through research on: concentration-response relationships in less industrialized nations and over a range of concentrations, including short-term and long-term effects and possible low- or high-concentration thresholds; improving the resolution of global atmospheric models and improving nested models that encompass the global, regional, and urban scales, to better represent concentration gradients in and near urban areas; the possible differential toxicity of different PM components and particle sizes; possible changes in PM and pollutant mixtures as they are transported and age, and the effects of such changes on toxicity; and possible interactive effects of PM, ozone, and other pollutants on human health.

5.2. Impact of Long-range Transport on Ecosystems

The doubling of tropospheric ozone (O_3) in the Northern Hemisphere since the Industrial Revolution [Chapter 1, 2] [*Vingarzan*, 2004] has had significant, negative impacts on crop production, forest productivity and has been shown to cause changes in the species composition of semi-natural systems [*Ashmore*, 2005]. Rapid industrialization in the Northern Hemisphere has also resulted in increases in emissions of pollutants such as SO_2, NO_x and BC that enhance atmospheric PM and can impact ecosystems through acidification, eutrophication and perturbations to the quality of photosynthetically active radiation. Here we make a first attempt to investigate the implications of intercontinental long-range transport (ICT) of these pollutants for ecosystems. The focus of this section is on terrestrial ecosystems, however impacts on oceans are also briefly covered.

5.2.1. Evidence for effects of ozone and PM on ecosystems

The regional distribution of experimental evidence for impacts on ecosystems is predominantly driven by the historical identification of impacts related to the regional occurrence of elevated pollutant concentrations and associated deposition. Hence, most evidence has been collected from North America and Europe over the past 30 years. The relatively recent advent of rapid industrialization and associated pollutant emissions in Asia [*Ohara et al.*, 2007] has led to a disconnect between the level of experimental evidence available and the scale of the pollutant problem by world region.

Impacts on ecosystems caused by O_3

The majority of the existing experimental evidence comes from bio-monitoring and Open Top Chamber studies that have been conducted first in North America [under the NCLAN Programme, *Heck et al.*, 1988] and later in Europe under the European Open Top Chamber [EOTC, see review by *Jager et al.*, 1992] and UNECE ICP Vegetation Programmes [*Hayes et al.*, 2007; *Mills et al.*, 2000]; these have mainly focussed on arable crops. Over the last decade similar studies are now increasingly being conducted in Asia [*Emberson et al.*, 2009]. The Free Air Concentration Enrichment approach has recently been used as an experimental method to assess impacts on crops (soybean in the US; *[Morgan et al., 2006]*; rice in China: *[Shi et al., 2009]*) forest trees (Aspen, Maple and birch) in Wisconsin, US: *[Karnosky et al., 2003]*; mature beech stands in Bavaria, Germany: *[Matyssek et al., 2010]*) and grasslands (alpine semi-natural grassland in the Swiss Alps *[Volk et al., 2006]*). These methods have advantages of being closer to field conditions but are limited in their ability to define impacts at or below ambient pollutant concentrations and in defining dose-response relationships which are necessary for regional scale risk assessments.

In addition to experimental studies, epidemiologic methods have also been used, initially driven by a need to overcome difficulties in extrapolating experimental studies conducted on young forest tree species in Open Top Chambers to understand effects on mature trees growing under forest stand conditions [*Bussotti and Ferretti*, 2009]. Such studies have consistently demonstrated that O_3 can negatively influence a variety of forest responses from crown condition to radial growth [*Braun S et al.*, 2010]. More recently, similar spatially relevant studies have been extended to crop loss assessments [*Fishman et al.*, 2010; *Kaliakatsou et al.*, 2010]. These studies have found that the influence of O_3 can be detected in regional level production statistics and field trial data although damage estimates have been found to differ from those obtained from risk assessments performed using empirically derived dose-response relationships [*Kaliakatsou et al.*, 2010]. This may be due to these methods being most effective in those regions characterized by higher average O_3 concentrations [*Fishman et al.*, 2010] where the O_3 signal is strong enough to overcome the influence of confounding variables affecting yield.

Rising background concentrations experienced over the last few decades can particularly enhance spring and autumn O_3 levels [see *Fiore et al.*, 2008], in effect lengthening the period of elevated O_3 concentrations from the existing summer peak O_3 exposures; the influence of these new seasonal profiles on ecosystems needs to be understood, this can only be achieved with new experimental investigations. To date, such new investigations have used diurnal fumigation patterns that emphasise chronic rather than peak O_3 exposures and investigation of species with growth periods that extend into those times of the year when ICT is a more substantial fraction of the total pollution load i.e. the spring and autumn periods [*RoTAP*, 2010].

Range of response parameters

Key impacts for agriculture include visible injury to leafy crop species [*Emberson et al.*, 2003; *Mills et al.*, 2010, in press]; declines in arable yields [*Mills et al.*, 2007] and effects on crop quality (e.g. nitrogen content of grains, tubers etc. and nutritive quality of forage crops) [*Pleijel et al.*, 1999]. Importantly, impacts have been found to vary substantially according to crop species and cultivars [*Betzelberger et al.*, 2010]. Prevailing climatic and meteorological conditions and agricultural management practices (e.g. irrigation) will also affect response to O_3 [*Fuhrer and Booker*, 2003].

For forest trees, O_3 has been shown to impact visible foliar injury, accelerate leaf senescence, reduce photosynthesis, alter carbon allocation, and reduce growth and productivity [*Karnosky et al.*, 2007; *Skarby et al.*, 1998]; again, these effects vary by forest tree species and genotype [*Karnosky and Steiner*, 1981]. O_3 also appears to weaken tree resilience to a range of biotic (e.g. pest and pathogen attack) and abiotic (e.g. drought, frost hardiness) stresses. The extent to which results obtained from tree seedlings/saplings can be extrapolated to mature trees under real forest condition has been severely challenged [*Kolb and Matyssek*, 2001] and resulted in a study conducted at Kranzberg Forest, Germany on naturally growing and late-successional, adult forest trees [*Matyssek and Sandermann*, 2003; *Matyssek et al.*, 2010]. This study found reductions in annual whole-stem volume increments for beech which, when scaled to stand level, supported modelling predictions that claim elevated O_3 to cause substantial reduction of C sink strength in trees [*Sitch et al.*, 2007].

Semi-natural grasslands are genetically highly diverse multi-species communities ranging from low to high productivity depending on site conditions and management. Component species differ strongly in their sensitivity to O_3 [*Bungener et al.*, 1999; *Hayes et al.*, 2007; *Timonen et al.*, 2004] and thus community response to O_3 is likely to be species-driven [*Jones et al.*, 2007]. However, changes in productivity and species composition in established temperate [*Volk et al.*, 2006], calcareous [*Thwaites et al.*, 2006] or alpine grassland [*Bassin et al.*, 2007] are difficult to detect against a background of considerable natural spatial and temporal variability. Subtle changes in C-assimilation and water economy in selected component species, as inferred from shifts in stable C and O isotopic signatures [*Bassin et al.*, 2008; *Jaggi and Fuhrer*, 2007], reduced leaf longevity [*Bassin et al.*, 2007], and altered biomass partitioning suggest that in the longer run, productivity may decline and species dominance may change in response to ICT. This is in contrast to observations in Mediterranean therophytic grasslands, where short-term effects on reproductive traits of annuals have

been observed [*Gimeno et al.*, 2004]. Experimental studies in the US and Europe have also highlighted O_3 impacts on nutritive quality of forage crops [*Krupa et al.*, 2004].

FINDING: There is evidence that O_3 can cause a variety of damage responses to crops, forests and grasslands. The strength of this evidence varies with receptor type and location, with more evidence on crops than forest trees, more on trees than grasslands, and equal evidence in North America and Europe, but less in Asia.

Experimental derivation of dose-response indices

The experimental campaigns conducted in North America [NCLAN: *Heck et al.*, 1988] and Europe [EOTC: *Jager et al.*, 1992] were instrumental in providing experimental data describing yield and growth responses for a range of crop species (and a far more limited number of forest and grassland species) that could be used to define O_3 metrics and dose-response relationships (see Box 5.1). It is important to note that in Europe, the selection of the AOT (accumulated ozone concentration over a threshold over a growing season) cut-off concentration of 40 ppb was actually driven by consideration of the level of background O_3 concentrations; AOT30 was as statistically robust in terms of defining crop damage but was considered to have implications for control strategies outside of Europe and hence the 40 ppb cut-off concentration was retained [*Karenlampi and Skarby*, 1996]. Since the background O_3 level can vary considerably across different regions, and most importantly with altitude, a single cut-off value may not be suitable for risk assessments in every geographical region with important implications for use of such threshold indices for assessment of ICT.

In the development of these indices it was recognised that high O_3 concentrations tended to co-occur with environmental conditions that restrict uptake (e.g. hot, dry sunny conditions). In Europe this has led to the development of the O_3 flux metric, PODy (Phytotoxic O_3 dose above a stomatal flux threshold y; formerly known as the accumulated stomatal O_3 flux, AFstY) [*LRTAP Convention*, 2010]. Re-analysis of existing Open Top Chamber experimental data for European wheat and potato [*Pleijel et al.*, 2007] and for a number of forest trees [*Karlsson et al.*, 2007] showed PODy to more accurately predict yield or biomass loss as compare to the AOT40 index. Although the flux metrics still have a threshold, which is assumed to act as a surrogate for the internal detoxification capacity of the plant [*Pleijel et al.*, 2007], the ambient O_3 concentrations that can contribute to accumulated flux will be substantially lower than the 40 ppb cut-off concentration used in the AOT40 index under optimum environmental conditions for plant gas exchange. Hence, the flux metric is likely to be better suited to assess the implications of rising background O_3 concentrations. It should also be noted that the increasing levels of background O_3 concentration translate into proportionally higher risk estimates, since both AOT40 and PODy involve a threshold value. This is due to a fundamental property of any similar threshold index and effectively means that with an increasing threshold these metrics become increasingly sensitive to the exceedance of the threshold value [*Tuovinen et al.*, 2007]. The sensitivity also depends on the characteristics of the frequency distribution of the data. Owing to the relatively wider distribution of stomatal fluxes, the PODy index for crops has been shown to be less sensitive to such perturbations than the corresponding AOT index [*Tuovinen et al.*, 2007].

Box 5.1 Explanation of different ozone metrics used to derive dose-response relationships.

Table 5.5 describes the formulation of different metrics that have been developed to assess O_3 damage to crops, forests and grasslands in both North America and Europe. The profile of the dose-response relationship relating the characterization of O_3 exposure to the response is also described along with the international organisations that have adopted the metrics. These metrics have evolved from average mean growing season indices (e.g. M7 and M12 (here referred to jointly as Mx indices) to indices that give greater weight to higher or peak O_3 concentrations deemed more biologically relevant to the induction of damage (e.g. AOT40 in Europe, SUM06 and W126 in North America) and most recently to flux based metrics (PODy). The profile of the dose-response relationship should also be considered; for example the curvilinearity of the M7/M12 indices essentially results in lower average O_3 concentrations contributing less to damage thereby having a similar effect in terms of weighting the relative importance of higher end O_3 concentrations albeit as growing season values rather than hourly averages; the importance of this effect varies according to the species-specific relationship. It should also be noted that dose-response relationships have yet to be derived for Asian conditions.

Table 5.5. O_3 metrics used to derive dose-response relationships for ecosystem protection in air quality management.

Metric	**Definition**	**Formulation**	**Dose-response relationship**	**Application**
M7 or M12	Daylight (7 or 12 hours) growing season average	M7 = mean 7 hour daylight [O3] M12 = mean 12 hour daylight [O3]	Weibull	-
AOT40	Accumulated concentration over a threshold of 40 ppb over a growing season	AOT40 = $\sum_{i=1}^{n}[O3-40]_i$ for [O3] ≥ 40 ppb during daylight hours [AOT40 units: ppb.hrs]	Linear	UNECE, EU
W126	Weighted accumulation over a growing season	W126 = $\sum_{i=1}^{n}[O3]_i * Wi$ where wi =(1/ (1+4403 *exp(-0.126*[O_3]))) over 24 hours [W126 units: ppb.hrs]	Curvilinear	USEPA (under consideration)
SUM06	The sum of all hourly ozone concentrations greater than 0.06 ppm, accumulated over a growing season	SUM06 = $\sum_{i=1}^{n}[O3]_i$ for [O3] > 60 ppb over 24 hours [SUM06 units: ppb.hrs]	Curvilinear	USEPA (under consideration)
PODy	Phytotoxic ozone dose expressed as the accumulated stomatal ozone flux over a threshold of y nmol m^{-2} s^{-1} over a growing season	PODy = $\sum_{i=1}^{n}[FO3-y]_i$ for [FO3] ≥ *y* nmol m^{-2} s^{-1} [PODy units: mmol m^{-2}]	Linear	UNECE

N.B. *[O3]* is the hourly mean O_3concentration, *i* is the index, *wi* is a weighting scheme that relates ambient O_3 concentrations to flux into the plant, *FO3* is calculated stomatal flux into the plant, *y* is a threshold above which stomatal O_3 flux causes damage and *n* is the number of hours over which O_3 concentrations are summed.

A recent study [*Mills et al.*, 2010] collating evidence from across Europe for ~30 species of agricultural and horticultural crops and almost 150 (semi)natural vegetation species (trees were excluded) between 1990 to 2006 showed that risk maps based on stomatal O_3 flux were better predictors of the areas where O_3 damage occurred than those based on AOT40. Under Mediterranean conditions, such improved performance of flux based risk assessments have been attributed to dry summer conditions that dramatically limit the accumulated stomatal flux through the occurrence of drought-limited gas exchange, despite the occurrence of higher O_3 concentrations that result in enhanced seasonal AOT40 values [*Gerosa et al.*, 2008]. Similar results were found by [*Matyssek et al.*, 2007] for mature beech trees growing under drought conditions in Bavaria, Germany.

Regional scale impact assessment studies that have used different metrics and appropriate dose-response relationships have illustrated considerable differences in resulting yield loss predictions. For example, a global study by van Dingenen et al. [2009] showed differences in crop yield losses of up to 74% using M7 and AOT40 indices. The cause of such inconsistencies are unclear and could be related to variability in the experiments used to derive dose-response indices (e.g. cultivar sensitivity, experimental O_3 concentration profiles) or application of the risk assessment methods (e.g. uncertainties in modelling O_3 concentrations, local O_3 concentration profiles, local conditions affecting the connection between O_3 concentration and absorbed O_3 dose).

Mechanisms of adverse effects of O_3 relevant for ICT

Phenology will play an important role in determining the importance of ICT damage to ecosystems. The timing and duration of growth periods will determine species exposure to ICT. For example, early or late season crops such as oil seed rape and maize may be more at risk, similarly forests and grasslands which have appreciably longer growth periods will be more likely to experience ICT. Within life-cycle and annual growing season variability in sensitivity may also be important. For example, Wieser et al. [2003] found that age dependent changes in leaf morphology were related to changes in the defence capacity against oxidative stress, with concentrations of antioxidants increasing with tree age; therefore, early season ICT O_3 exposures may occur when anti-oxidative defence capacities are still establishing. In more remote regions with early springtime peaks in O_3, together with conditions favouring high stomatal conductance, the situation may be worse as the advancement of plant development may lead to more frequent co-occurrence of sensitive stages and early-season O_3 stress [*Karlsson et al.*, 2007].

Species distribution will also affect vulnerability to ICT. For example, many forests and grassland communities extend to high altitudes where the planetary boundary is more likely to be coupled to the lower layers of the free troposphere and hence more prone to ICT influence. Forests also host understory species, which often have ephemeral growth periods in the spring before closure of the forest canopy restricts growth.

FINDING: Concentration-based indices to assess the importance of O_3 damage, especially those which use thresholds (e.g. AOT40) may not be appropriate for assessment of damage resulting from ICT. O_3 flux metrics (e.g. PODy) that incorporate the effects of differences in phenology and environmental conditions in estimates of O_3 damage are more suitable for assessments of the potential impact of ICT.

Impacts on ecosystems caused by PM

PM can affect ecosystems in a number of ways depending on its characteristics, atmospheric concentration and deposition mechanisms; here we identify acidification, eutrophication and changes in radiation quality as the most important of these in terms of ICT.

Sulphur and nitrogen (N) deposition can cause soil and water acidification through long-term deposition of nitrate, ammonium and sulphate, which combine to add to the acidifying load that soils and aquatic ecosystems receive [*Fowler et al.*, 1991]. Such acidification potential can be counteracted by atmospheric deposition of base cations (calcium, potassium and magnesium) that contribute to the buffering capacity of sensitive soils and can negate the effects of acidic deposition [e.g., *Johnson et*

al., 1994]. Acidification has been linked to adverse effects on forest growth and vitality, to loss of sensitive fish populations and to changes in soil and aquatic invertebrate communities.

In ecosystems for which N is the limiting nutrient, moderate rates of N deposition can also result in stimulation of plant growth; increasing forest and grassland productivity. Likewise, moderate rates of sulphur deposition can enhance growth in sulphur deficient soils [*McGrath and Zhao*, 1995]. However, excess rates of N deposition can cause the loss of characteristic plant species of N limited systems and reductions in species diversity in terrestrial systems, it can also cause oxygen deficiency in fresh surface waters that are N, rather than P limited, with an associated loss of biodiversity and habitat degradation.

Since the process by which PM impacts ecosystems through eutrophication and acidification is reasonably well understood and is linked only to the total annual deposition load (i.e. will not be affected by within year variation in deposition loads resulting from ICT), we discuss these mechanisms of PM impacts only in relation to current knowledge based on ICT modelling studies in section 2.3.1.

While little is known about the broad scale impact on marine ecosystems, there are several recent papers [*Duce et al.*, 2008; *Krishnamurthy et al.*, 2009] that have investigated the impact of the deposition of anthropogenic N species on the biological productivity of the open ocean, and the impacts are not insignificant. For example, Duce et al. [2008] suggest that currently the quantity of atmospheric anthropogenic fixed N entering the open ocean (i.e. ocean fertilization) could account for up to a third of the ocean's external (non-recycled) N supply and up to ~3% of the annual new marine biological production, ~0.3 petagram of carbon per year. The resulting drawdown of CO_2 from the atmosphere would represent ~10% of the ocean's entire drawdown of atmospheric anthropogenic CO_2 each year, leading to a decrease in radiative forcing. However, the atmospheric deposition of anthropogenic N could also lead to increased production of nitrous oxide in seawater, and its subsequent emission to the atmosphere could offset some of the radiative forcing effects from atmospheric CO_2 reduction.

Atmospheric PM (including dust, sulphates, nitrates, secondary organics, OC, BC, and directly emitted PM) can also alter radiation quality by scattering and reflecting incoming solar radiation leading to an increase in the diffuse component. Up to a certain point the resulting increase in the diffuse component can enhance plant growth as this type of radiation is used more efficiently in photosynthesis with implications for the terrestrial carbon sink [*Mercado et al.*, 2009]. The response to PM changes in radiation quality vary by vegetation type [*Niyogi et al.*, 2004] and has been explained as an effect of canopy architecture and influence on light distribution through the canopy. *Niyogi et al.* [2004] found forest and croplands to be more sensitive to changes in diffuse radiation than grasslands [*Niyogi et al.*, 2004]. Unfortunately, only very few experimental studies have been conducted investigating PM influence on radiation quality.

FINDING: There is evidence that PM can affect crop, forest and grassland production through the processes of acidification, eutrophication and alteration of radiation quality. Acidification and eutrophication are associated with the loss of characteristic species and a reduction of species diversity in sensitive terrestrial and aquatic ecosystems. Limited evidence also suggests that biological productivity of the ocean is affected through fertilization by N deposition with implications for atmospheric CO_2 and radiative forcing. Alteration of radiation quality can enhance crop and forest photosynthesis, up to a threshold value of PM above which photosynthesis can decrease.

Critical thresholds for adverse effects of PM

The long-term nature of effects of acidification and eutrophication (which have cumulative impacts over decades) coupled with limited experimental data, mean that dose-response relationships cannot be identified to assess the impacts of ICT of PM. Instead, critical thresholds for adverse effects are used to assess impacts.

The effects based approach, developed by the UNECE CLRTAP for use in Europe [*LRTAP Convention*, 2010] employs the concept of critical loads, the maximum deposition below which significant harmful effects on specified sensitive elements of the ecosystem will not occur. These critical loads refer to long-term effects of acidification and eutrophication. For acidification and nutrient mass balance, steady-state biogeochemical models are used to determine the deposition rate that results in the exceedance of a pre-defined chemical threshold value in environmental media (e.g. soil, surface waters) for effects in different ecosystems [*Spranger et al.*, 2008]. The calculation of these critical loads depends on local soil and water characteristics, climate and land use. For eutrophication, empirical critical N loads have also been established based on field experiments and gradient studies; these focus on impacts on biodiversity in natural and semi-natural systems [*Bobbink et al.*, 2003]. A recent synthesis of results of European N addition experiments in grasslands, (sub)Arctic and alpine vegetation, and temperate forests showed clear relationships between exceedance of empirical N critical loads and either plant species richness or similarity to reference plant species composition [*Bobbink*, 2008] thus supporting the use of this method to identify the harmful effects of eutrophication across Europe [*Bobbink et al.*, 2010].

Empirical critical loads are primarily defined for temperate and arctic-alpine ecosystems of Europe, although values have also been identified from field experiments in North America. A recent overview of the evidence of effects of N deposition on terrestrial plant diversity [*Bobbink et al.*, 2010] identifies that the critical load for many sensitive ecosystems lies in the range 10-15 kilograms per hectare per year (kg ha^{-1} y^{-1}), but may fall to 5-10 kg ha^{-1} y^{-1} for particular sensitive ecosystems or groups of organisms. Bobbink et al. [2010] identify Mediterranean ecosystems of southern Europe and California, and sensitive ecosystems in southern and eastern Asia as systems where there may already be impacts of N deposition but critical loads have not been defined.

Discussion of metrics to quantify effects of PM enhancing diffuse radiation on ecosystems photosynthesis has been limited to interpretation of modelling studies that attempt to calculate the diffuse fraction value (threshold) at which photosynthesis starts decreasing. A study by *Knohl et al*. [2008] for a broad leaf forest site in Germany estimates that above a diffuse fraction of 0.4, forest photosynthesis will decrease. A similar value for the same location was found in the modelling performed by *Mercado et al*. [2009]. A modelling study by *Alton* [2008] claimed that diffuse radiation under cloudy skies makes plants photosynthesise less; however, these global fields of diffuse radiation were not extensively validated leading to uncertainties in the results and interpretation of this work. As such, a suitable metric may be one that is able to capture the effect of diffuse radiation fraction on photosynthesis with a threshold defined as that diffuse radiation fraction at which photosynthesis starts decreasing. However, this is difficult to estimate using experimental or modelling studies in isolation. Once models are better calibrated and evaluated using data for single sites, it might be possible, on consideration of the sensitivities of the model, to estimate such thresholds.

FINDING: In contrast to O_3, PM dose-response relationships are not available to estimate the effects of intercontinental transported PM. Critical thresholds to prevent long-term adverse effects can be identified or modelled, but much of the empirical evidence from which these thresholds have been identified comes from Europe and North America limiting applicability to other parts of the world.

5.2.2. Quantified influences of ICT pollution on ecosystems

Role of ecosystems in determining long-range transport

Ecosystems play an important role in determining the global mass balance of atmospheric gases and aerosols by providing an effective sink for the removal of substantial fractions of atmospheric O_3 and PM by dry deposition. For O_3, this surface removal has been found to exceed the net stratospheric input term [*Stevenson et al.*, 2006]; these same deposition processes induce a potentially harmful load to ecosystems. This load depends not only on the atmospheric abundance of pollutants but also on the efficiency of dry deposition. Therefore an improved mechanistic understanding of the atmosphere-biosphere exchange processes would contribute to our understanding of both ICT and ecological impacts of pollutants. For O_3, in particular, it is necessary to differentiate

between the stomatal and non-stomatal deposition pathways, as it is the former that provide the primary potential for plant injury [*Emberson et al.*, 2001]. Modelling studies [*Solberg et al.*, 2008] have shown that reduced stomatal uptake, and consequently slower depletion by dry deposition, significantly enhances ground-level O_3concentrations; such conditions tend to occur during hot and dry periods in which stomates close to limit plant water loss. Given these criteria it is likely that climate change will play a significant role in determining future dry deposition removal processes by ecosystems with implications for atmospheric concentrations and human health [see section 5.2.4.4 and *Klingberg et al.*, 2011]

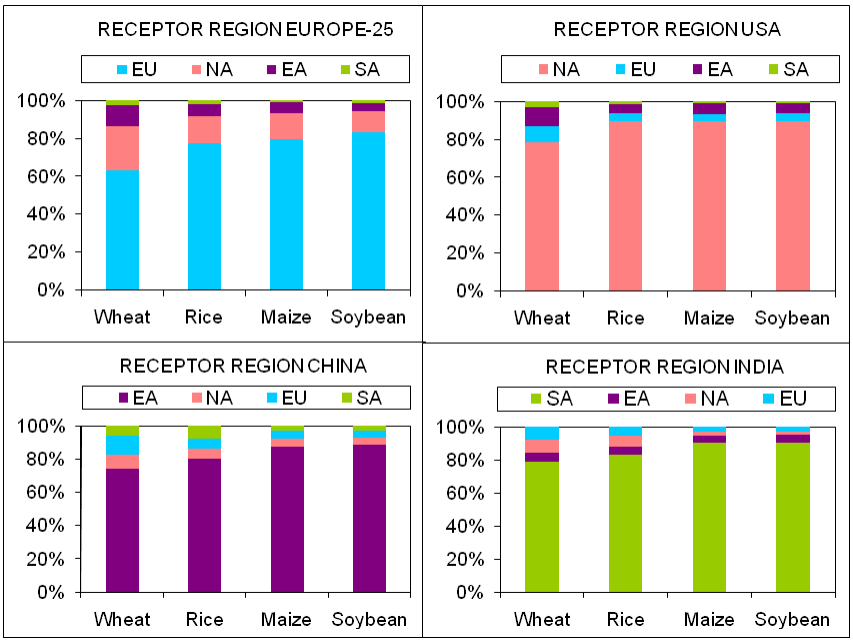
FINDING: Deposition of pollutants to vegetated land surfaces plays an important role in determining atmospheric pollution mass balance and hence ICT. These same deposition processes also represent the pathways by which ecosystems are impacted by air pollution. As such, an integrated approach to improve our understanding of these deposition processes may benefit our understanding of ICT as well as impacts.

Impacts of long-range transport of ozone on crop yield

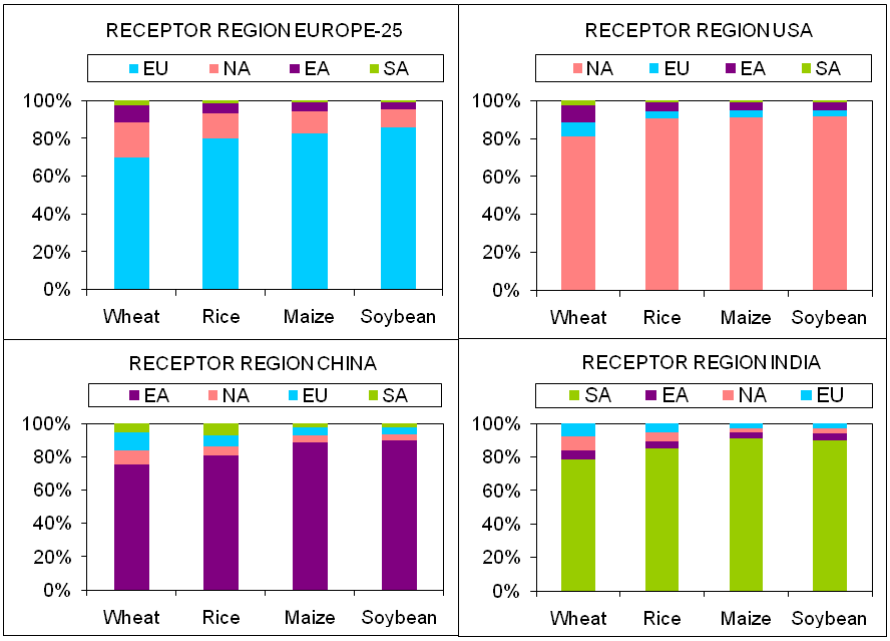
The impact of ICT of tropospheric O_3 on ecosystems has not previously been investigated in published studies. Specifically for this report, crop losses were estimated from the HTAP model ensemble of O_3 concentration based metrics (flux based metrics could not be used since this methodology has not yet been parameterized for application at the global scale). Using hourly O_3 concentration output of a subset of models, AOT40 and Mx metrics for four staple crops (wheat, rice, maize and soybean) were estimated for crop specific growing seasons according to climatically determined growth periods *[as described in Van Dingenen et al., 2009]*. For each individual model, the difference in AOT40 and Mx between the base run and each of the four perturbation simulations (20% emission reductions in each of the four regions) was calculated on the model's own grid resolution and then interpolated to a 1°x1° grid to provide the ensemble average. The resulting ensemble AOT40 and Mx 1°x1° fields were then analyzed as in *Van Dingenen* et al. [2009], leading to gridded fields of crop relative yield loss, which were then aggregated to regional averages. The regional averages were used to calculate Relative Annual Intercontinental Response (RAIR) values in terms of relative crop yield loss. As in Chapter 4, the RAIR values are calculated for each region as the sum of the changes due to emission changes in the three other regions divided by the sum of the changes due to emission changes in all four regions combined.

Figure 5.10 shows the Relative Annual Intercontinental Response (RAIR) of each HTAP region to changes in ozone crop damage (expressed as relative yield loss) in each of four receptor regions for simulation set SR6 (20% emission reductions of NO_x, VOC, CO, SO_2, and PM). These results clearly show that emissions from one region can cause yield losses in another region. The contribution of ICT from the three other source regions to yield losses in each receptor region varies between ~5 and 35% of the estimated losses. However, this range is very dependent upon the response function used and should be considered with caution, especially given the disadvantages in using the AOT40 index for estimating the influence of ICT. The largest influence of ICT is found from North American emissions impacting on European crop yields. The difference in the influence of ICT on damage to each crop for a given region depends upon differences in the contribution of domestic emissions to O_3 concentration, crop growing season and geographical location and the crop-specific response function. It should also be noted that emission reductions outside the indicated HTAP areas have not been included and could further impact background O_3,;Also, the indicators have not been corrected for the crop canopy height. As such, limiting analyses of these results to relative rather than absolute contributions to crop damage makes the results more robust (as also appears from the very similar outcome for both indicators) since inter-model differences of absolute yield losses are rather large, and more difficult to interpret.

a). The contribution of source regions to relative yield losses estimated using growing season Mx concentration response functions specific to each of four staple crops.



b). The contribution of source regions to relative yield losses estimated using AOT40 concentration response functions specific to each of four staple crops.



HTAP region key: EU = Europe, NA = North America, EA = East Asia, SA = South Asia

Figure 5.10. The relative contribution of each HTAP region to crop damage (expressed as relative yield loss) in each of 4 regions for run SR6 (20% decrease of O_3 and PM precursor emissions) using Mx and AOT40 indices.

In a separate, and as yet unpublished study, the TOMCAT global chemical transport model [Arnold et al., 2005 ; Chipperfield, 2006] has been used to investigate the impact of hemispheric-scale transport of O3 and its precursors on crops using the AOT40 O3 index to assess crops yield loss. In this study three perturbation simulations were carried out where NOx emissions over three major

global regions were cut by 90%. Similar to the HTAP study described above, these results also suggested that of all global regions, it was emissions from North American impacting on European yields that gave the largest transboundary effect. The use of 90% NOx reductions in this study is likely to provide a stronger indication of the actual contributions from the different emission sources. This is due to the threshold nature of the AOT40 index, which means that contributions to AOT40 exposure can only be defined against a given scenario of local and non-local O3 precursor emissions. The contribution of a non-local NOx source to AOT40 is dependent on the extent to which that source enhances surface O3 concentrations above 40ppb, which will change depending on the strength of local and other non-local sources. In this sense, the AOT40 contributions from ICT diagnosed over each region are particular to the current configuration of local emissions in each region, whereas non-local contributions to the mean O3 (represented by the Mx indices) over each region are largely independent of local emissions. This further emphasizes the weakness in the use of threshold indices such as AOT40 in attributing plant O3 damage to different emission sources.

A further key limitation of the model based risk assessment studies discussed above is associated with uncertainties in the simulated O_3 concentrations. While there are dense networks of near-surface O_3 measurements in North America, Europe and Japan [*EEA*, 2009; *EMEP*, 2010; *Oltmans et al.*, 2006; *U.S. EPA*, 2009a], the availability of measurement data for model validation is more limited for Africa, Latin America and China [*Ellingsen et al.*, 2008], and often non-existent in rural parts of these regions due to the focus on monitoring to assess risks to human health. It has also been found that models tend to be less accurate in their predictions of higher O_3 concentrations; this coupled with the increased mathematical sensitivity of the threshold-based risk indices makes the quantification of such indices more uncertain than that of mean concentrations [*Tuovinen et al.*, 2007].

FINDING: There is considerable uncertainty as to how much of the damage to ecosystems caused by O_3 is attributable to hemispheric ICT. Provisional results from HTAP model-based risk assessments using AOT40 and Mx indices indicate that emissions from one continent have the potential to influence crop productivity on other continents by affecting O_3 concentrations, with the largest influence of ICT found from North American emissions impacting on European crop yields. Based on the HTAP multi-model experiments, ICT may be responsible for about 5 to 35% of the estimated crop yield losses depending on the location, crop, and response function used, subject to large uncertainties.

Impacts and potential vulnerability of ecosystems to long-range transport of PM

The observations of forest decline and lake acidification in Europe and North America during the 1970s and 1980s [*Fowler et al.*, 1999] prompted research which clearly identified the adverse effects of deposition of S, and to a lesser extent, N. Emission control programmes have helped to improve the situation in these regions, and attention is now focused on the rate and nature of recovery of soils and waters from long-term acidification [e.g. *Ormerod and Durance*, 2009]. The region of greatest concern is now Asia, where acidification problems are expected to increase in the future [*Fowler et al.*, 1999]. There are now indications that acidification may have already occurred in parts of China related to air pollution [*Hicks et al.*, 2008]. Modelling by Hicks et al. [2008] has described the variability in susceptibility to acid rain across south and east Asia, and suggests that soil acidification effects are unlikely to be widespread due to insensitivity of soils and high concentrations of alkaline dust in the atmosphere, and the timescale for significant acidification is likely to be long. Areas at risk of acidification over the next 50 years are mainly restricted to southern East and Southeast Asia (Figure 5.12) but the characteristics and severity of these problems vary with biotic and abiotic factors [e.g. *Hicks et al.*, 2008].

HTAP model ensemble results presented in *Sanderson et al*. [2006] examined acidification risk of soils in the present day and 2090s using a single chemistry-transport model coupled to a global climate model, and examined the impact of climate change. The areas most at risk from acidification were projected to be the eastern parts of the U.S., central Europe, and particularly south and east Asia for both time periods. Future deposition fluxes of N and sulphur compounds were mostly controlled by changes in emissions. Climate change and atmospheric composition (which will affect the oxidising capacity of the atmosphere) had only a small impact on pollutant transport and deposition.

In terms of observed effects in the region, it is also important to note that the forest decline that has occurred in some areas of China may be due to direct effects of gaseous SO_2, and to extremely acidic mist or rain events, which are due to local emissions, rather than the long-term effects of soil acidification caused by regional and ICT sulphur and N deposition. Hence, although parts of east and south east Asia can be identified as the third largest acid rain prone region in the world, more information on rates of change and the regional sensitivity to acidification, and the importance of pollutant emissions from local sources, are needed before accurate assessment of the impact of ICT can be undertaken.

In relation to eutrophication, recent global modelling studies suggest that N deposition related to NO_x and NH_3 emissions are now as high in some parts of south, south-east and southern east Asia as they are in Europe and North America (Figure 5.11). There is growing scientific consensus that impacts will also be considerable in sensitive ecosystems of Asia, although local evidence is only just starting to emerge. For example, in China, manipulation experiments suggest that N deposition has the potential to influence the species richness of the under-storey of temperate and tropical forests [*Bobbink et al.*, 2010]. The WWF terrestrial Global 200 eco-regions were ranked by conservation status identifying those that were critical, endangered, or vulnerable as a result of direct human impacts [*Olson and Dinerstein*, 2002]. Figure 5.12 shows that many of the WWF G200 eco-regions of global conservation significance in Asia already had deposition rates in 2000 that are well above the critical load for sensitive ecosystems that have been established in Europe, while the threshold range for sensitive ecosystems of 10-15 kg ha^{-1} y^{-1} is reached in eco-regions in several other parts of the world.

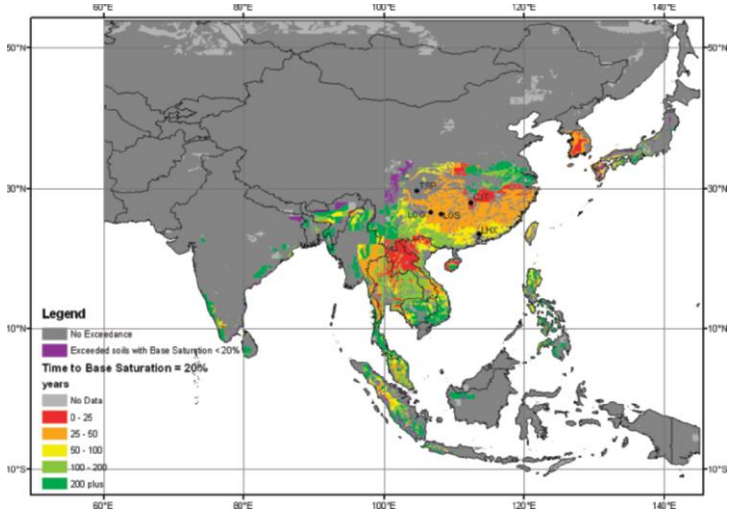


Figure 5.11. Time development of soil acidification damage according to a modelling study for Asia using best available data for soil and deposition parameters and deposition estimates obtained using a rather pessimistic emission scenario (IPCC SRES A2) for 2030. The model calculates the time it takes for the neutralizing capacity of the soil (expressed as base saturation) to be reduced to a level where acidification effects are observed (i.e., approx. 20% base saturation). [Reprinted from Figure 3 of Hicks, W. K., et al. (2008), Soil sensitivity to acidification in Asia: Status and prospects, *Ambio, 37*, 295-303. Copyright: the Royal Swedish Academy of Sciences.]

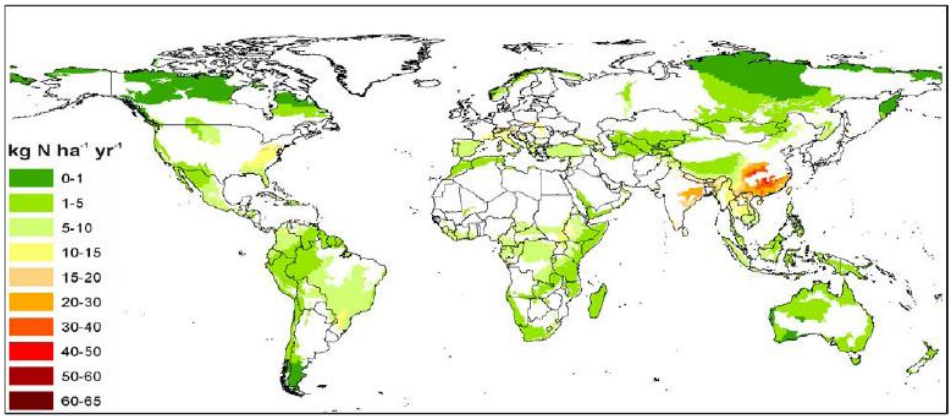


Figure 5.12. The WWF G200 regions [*Olson and Dinerstein*, 2002], showing total N deposition rates for the year 2000. N deposition to areas outside the G200 regions is not shown. [Reprinted from Figure 5(a) of Bobbink, R., et al. (2010), Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis, *Ecological Applications, 20*: 30-59.]

The importance of ICT to these exceedances of critical loads in regions with globally significant eco-regions cannot be quantified with large certainty. Reduced N (NH_x) is important in determining local eutrophication but due to its lower average atmospheric lifetime (e.g. the mean residence time of NH_x is 5 hours, compared to 30 hours for NO_y); mean travel distances of NH_x are only 150 kilometres (km) compared with 1,000 km for NO_y [*NEGTAP*, 2001]. Hence NH_x is not often considered in the context of ICT although ammonium sulphate has a longer lifetime than ammonia (approximately 5 days) and can be transported larger distances. A recent study by Sanderson et al. [2008] attempted to quantify the ICT of oxidised N (NO_y) from one source region to another in the Northern Hemisphere. They used results from 15 chemistry-transport models to quantify the transport of NO_y across four major source regions in the Northern Hemisphere. They found that 8-15% of NO_x emitted from each region is transported over 1,000 km and that between 3-10% of the NO_x emitted from each region is deposited as NO_y in the other three regions. However, a fixed reduction in emissions in one source region had little influence in other regions, with the percentage change in deposition in any receptor region being typically only 1-2% of the change in emissions in the source region; the one exception was east Asia, for which there was a 6.4% reduction from emission reductions in South Asia. We note here however, that these numbers mask large geographical variability, and that in sub-regions closer to the source the impacts can be larger.

On this basis, it is unlikely that ICT makes substantial contributions to large scale adverse effects of N deposition in other regions. However, it is important to emphasise that the situation may be different for specific remote and sensitive regions. For example, Arctic systems are very sensitive to eutrophication and may have a critical load close to 5 kg ha^{-1} y^{-1} [*Bobbink et al.*, 2010]. These areas have very little influence from local sources, but *Wolfe et al*. [2006] have suggested that N deposition from distant sources can contribute to changes in the biogeochemistry and ecology of two remote lakes on Baffin Island in the eastern Canadian Arctic**.** In addition, deposition of transported PM to snow and ice surfaces in the Arctic has been linked to accelerated melting [*Shindell*, 2007] with implications for ecosystems. More analysis of the significance of ICT in specific remote and sensitive parts of each region would be valuable since these are the locations where it may be most significant.

FINDING: There is considerable uncertainty over the importance of the contribution of ICT of PM to regional acidification and eutrophication. However, on a regional basis, the contribution of ICT of NO_x to adverse effects on sensitive ecosystems is likely to be small. Focus is needed on specific remote and sensitive ecosystems where the contribution of ICT may be more significant.

5.2.3. The potential vulnerability of ecosystems to long-range transport of O_3

This section investigates the vulnerability of ecosystems to ICT, through identification of ecosystems of particular global importance in terms of agriculture, forest cover and biodiversity. This type of analysis equates with findings from the health section (see section 5.1.2) that identified the

added importance of foreign sourced emissions to pollution in regions of high population density. Here we replace high population density with ecosystem importance to identify equivalent regions where ICT may be of particular relevance.

Agriculture and Food security

O_3 crop yield response functions have been used with global chemistry transport models to estimate current and future relative yield losses due to O_3 [e.g., *Tong et al.*, 2007; *Van Dingenen et al.*, 2009; *Wang and Mauzerall*, 2004]. Although each of these modelling studies uses different O_3 models and crop production and distribution data they all make use of the range of dose-response response functions described in Box 5.1, and hence provide a reasonably standardized indication of production and economic losses associated with O_3 exposures. Currently, global yield losses are predicted to range between 3% and 5% for maize, 7% and 12% for wheat, 6% and 16% for soybean, and 3% and 4% for rice, which represents an annual economic loss of $14-$26 billion [see Figure 5.2.3 for wheat production losses due to O3, *Van Dingenen et al.*, 2009]. Agricultural regions in North America, Europe and Asia are identified as particularly vulnerable to O_3damage. About 40% of this damage was found to occur in parts of China and India. The substantial impacts found in the Asian region may be particularly relevant given the importance of agriculture within these country economies; e.g. losses were estimated to offset a significant portion (between 20 to 80%) of the increase in GDP in the year 2000 in such economies.

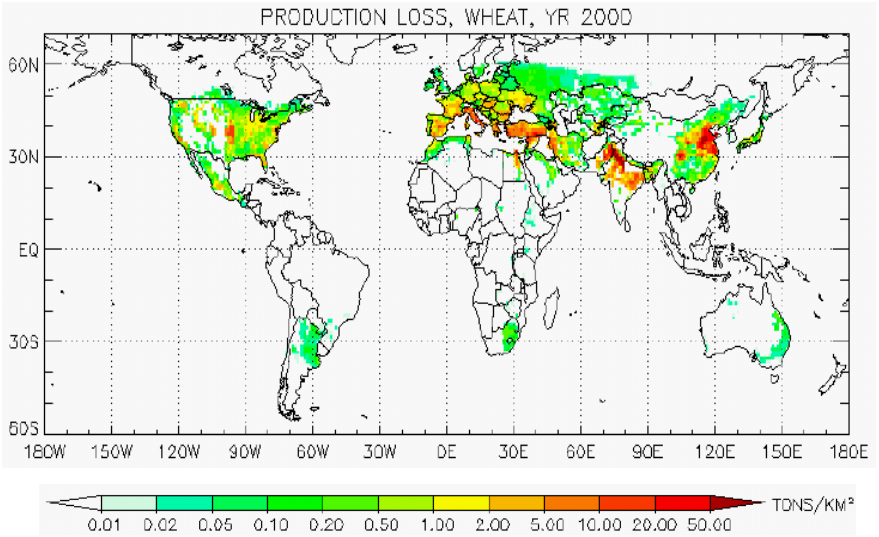


Figure 5.13. Average wheat crop production losses due to O_3 estimated for the year 2000 using European and North American concentration based exposure-response relationships. [Adapted from Figure 10 of Van Dingenen, R., et al. (2009), The global impact of ozone on agricultural crop yields under current and future air quality legislation, *Atmospheric Environment, 43*(3): 604-618, with permission from Elsevier.]

Importantly, these modelling studies have relied on North American or European dose-response relationships to assess the yield losses caused by O_3; a recent synthesis of data [*Emberson et al.*, 2009] strongly suggests that key Asian crops and cultivars may well be more sensitive to O_3 concentrations when growing under Asian conditions suggesting that the production and subsequent economic loss estimates for this region may be underestimated.

Globally, there are a number of agricultural production areas that are vulnerable to increasing O_3 pollution. The "Cornbelt" in the United States produces 40% of the world's corn and soybean crops, and this region is already potentially losing 10% of its soybean production to O_3 [*Tong et al.*, 2007]. In the U.S. as a whole, agronomic crop loss to O_3 is estimated to range from 5 – 15%, with an approximate cost of $3-$5 billion annually [*Fiscus et al.*, 2005; *U.S. EPA*, 2006]. In Europe, similar studies have identified substantial economic losses due to O_3, for example, Holland et al. [2007] estimated losses for 23 crops in 47 countries in Europe of €4.4 to 9.3 billion/year, around a best estimate of €6.7 billion/year for year 2000 emissions. Despite the overwhelming evidence that current

O_3 concentrations are causing yield losses, new O_3 tolerant crop cultivars are not being developed for a future higher-O_3 world [*Ainsworth et al.*, 2008; *Booker et al.*, 2009]. Recent successes in identifying quality trait loci associated with O_3 tolerance in rice indicate the breeding for O_3 tolerance in food crops is possible [*Frei et al.*, 2008; *Frei et al.*, 2010], yet currently, there is little if any industrial effort in this direction.

In the case of PM, eutrophying and acidifying deposition is not likely to pose a particular threat to agriculture; in the first instance, agricultural systems are often N limited and require applications of N fertilizer to enhance productivity. The adverse effects caused by acidifying substances can be negated through the application of lime in heavily managed systems although such management comes with an associated economic cost and hence may be a less favourable option in developing countries. The role that atmospheric PM may play on agricultural productivity has not been specifically assessed.

Forest health, grasslands and biodiversity

In contrast to agriculture, little work has been done to assess the impacts of ground level O_3 on important global forest biomes. Work that has been conducted has been confined to studies in Europe and North America. A Scandinavian study estimated timber yield losses due to O_3 at 2.2% in Sweden over the period 1993-2003 [*Karlsson et al.*, 2005]. The resulting economic loss was estimated to be a 2.6% decline, which is equivalent to €56 million based on 2004 prices for timber and pulpwood. Additionally, *Muller and Mendelsohn* [2009] estimated annual timber yield losses equivalent to $80 million in the U.S.

Little is therefore known of the response to O_3 by forest ecosystems that cover vast swathes of the Northern Hemisphere, even though heavily forested areas coincide with regionally high O_3 concentrations in east and southeast Asia, northern Asia, and boreal North America. Recent studies in Scandinavia have identified certain aspects of O_3 exposure of the more northerly, boreal forest ecosystems that could particularly enhance vulnerability to ICT. These include the earlier onset of the growing season as climate changes, the extensive periods of 24 hour daylight that allow more or less continuous gas exchange leading to a large cumulative O_3 dose, and lack of recovery from oxidative stress during darkness [*Karlsson et al.*, 2007]. In terms of tropical forests, *Fowler et al*. [1999] estimated that O_3 concentrations in excess of 60 ppb were experienced by an area of 3 million km^2 in 1990 (almost 20 % of total tropical forest cover) with the increase particularly great in southeast Asia. In spite of this rapid increase in exposure, our knowledge of O_3 impacts on forest ecosystems of tropical biomes is extremely limited [*Emberson et al.*, 2003].

A similar situation exists for grasslands. Projections of O_3 effects on semi-natural grasslands in different regions with widely varying climatic conditions are difficult because of the diversity of this ecosystem type, substantial intra-specific differences in O_3 sensitivity among populations [*Bassin et al.*, 2004; *Yoshida et al.*, 2001], and a lack of experimental data for most systems. In temperate latitudes, such as northwestern Europe, grasslands are dominated by perennial C3 species, whereas in warmer climates annual species form a greater component. These latter systems may be more sensitive to O_3 due to their dependence on reproductive output, which was found to respond most sensitively to elevated O_3 [*Gimeno et al.*, 2004]. Highly diverse communities with an important conservation value in regions with a warmer climate may be more vulnerable than perennial grasslands in temperate and montane habitats. This may concern regions where C3 species predominate and where typically high O_3 levels are observed, such as the Mediterranean basin in Europe or in coastal parts of southern California. Grasslands dominated by C4 grasses in warmer regions such as India, southeast Asia, southern China and in much of the Southern Hemisphere may be less sensitive to O_3 as the C4 photosynthetic pathway (which is capable of providing a near constant and optimum supply of CO_2 for photosynthesis with relatively low stomatal conductance) will confer some protection against O_3. Similarly, therophytic grasslands in arid and semi-arid regions such as northern China may be less affected with only a few percent simulated reductions in net primary production due to O_3 alone [*Ren et al.*, 2007].

FINDING: There is evidence of impacts of O_3 on vulnerable and important agricultural, forest and grassland ecosystems across the Northern Hemisphere; over such ecosystems the enhancement of locally derived O_3 concentrations by ICT may be particularly important.

5.2.4. Interactions with climate change

Effect of atmospheric composition on plant physiology

Vegetation plays an important role in determining surface O_3 levels, via dry deposition of O_3 to the interior of leaves through stomata. As atmospheric CO_2 levels rise, the stomata will not need to open as widely to allow sufficient CO_2 to enter for photosynthesis. This may reduce O_3 uptake, decreasing the sensitivity of the plants to O_3 [*Sitch et al.*, 2007]. Such reductions in stomatal conductance of plants would result in both lower uptake rates and increased O_3 concentrations in the boundary layer. *Sanderson et al*. [2007] found that surface O_3 levels over parts of Europe, Asia and the Americas were 4-8 ppb larger under doubled CO_2 conditions during April, May and June (the approximate growing season for crops in northern Europe). Similarly, *Klingberg et al.*[2011] found that despite substantially increased modelled future O_3 concentrations in central and southern Europe, the flux-based risk for O_3 damage to vegetation is predicted to remain unchanged or decrease at most sites, mainly as a result of projected reductions in stomatal conductance under rising CO_2 concentrations although soil moisture and temperature were also found to play an important role in determining stomatal O_3 flux. However, the relationship between stomatal conductance and CO_2 concentration may prove to be more complex and depend on O_3-CO_2 interactions [*Uddling et al.*, 2010]. In addition, recent research has found that effective regulation of stomatal conductance under drought conditions was disrupted by increasing background O_3 concentration [*Mills et al.*, 2010; *Wilkinson and Davies*, 2009; 2010]. For a further discussion of potential climate change effects on O_3 fluxes, see *Fowler et al*. [2009] and *Fuhrer* [2009].

Impact of future atmospheric conditions on ecosystems

Models simulate that global mean precipitation increases with global warming [*Meehl et al.*, 2007]. However, there are substantial spatial and seasonal variations. Increases in the amount of precipitation are *very likely* in high latitudes, while decreases are *likely* in most subtropical land regions (section 1.5.2), continuing observed patterns in recent trends in observations. Precipitation is projected to decrease in many areas already suffering from water shortages [e.g., the Mediterranean and parts of Africa, *IPCC*, 2007a], which together with rising temperatures, will increase stress among plants. Reduced stomatal conductance that may occur in response to elevated CO_2 may enhance water use efficiency of plants, which may help to partly alleviate the effects of reduced rainfall. The projected increase in temperatures in many parts of the world mean that yields from crops may also be reduced [*Lobell and Field*, 2007]. Increased water stress in a warmer climate may be expected to decrease sensitivity to O_3 via reduced uptake; however O_3 induced damage to stomatal functioning (*Mills et al*, 2010, *Wilkinson and Davies*, 2009, 2010) might confound this effect. The exact impacts of pollutants on vegetation in the future will be complicated by the differential response of plants to climate change and rising CO_2 levels, whereby the latter will increase growth and might offset some of the projected yield losses from crops by the former.

A mechanistic model of plant-O_3 interactions was implemented into the Hadley Centre land surface model and run with O_3 scenarios from the STOCHEM chemistry transport model driven with SRES A2 emission scenario [*Sanderson et al.*, 2003; *Sitch et al.*, 2007]. Results suggest a large negative impact of near-surface O_3 on plant productivity. These model results have been used to identify eco-regions where a significant effect on global primary productivity might be expected to occur [*Royal Society*, 2008]. The results in Figure 5.14 are based on the 'low sensitivity' simulation of *Sitch et al*. [2007] which is overlain with the G200 regions and used to assess threats of O_3 deposition to biodiversity. This identifies eco-regions of south and east Asia, central Africa and Latin America as being at risk from elevated O_3 levels during this century, in addition to areas of North America and Europe where the effects of O_3 are better documented. However, there is almost no information available on whether the plant communities in these other regions of the world are as sensitive to O_3

as those that have been used to define critical levels, and hence the real significance of these areas of potential risk to biodiversity is completely unknown.

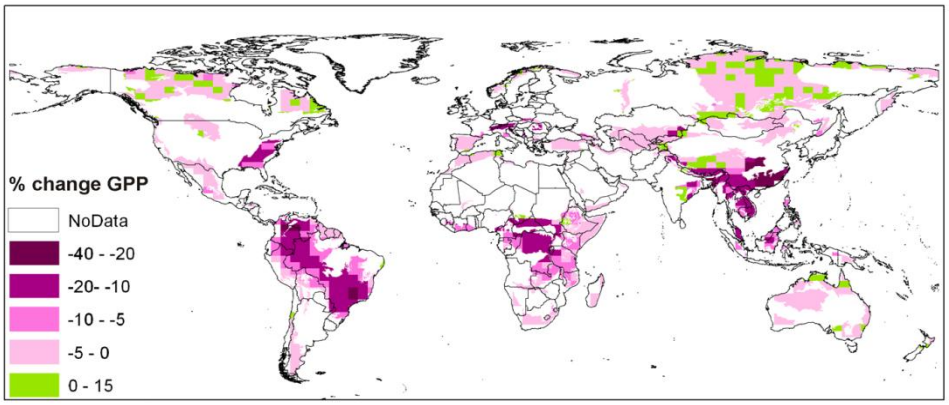


Figure 5.14. Global assessment of the key biodiversity areas at high risk from O_3 impacts; the figure shows the projected percent decrease in gross primary productivity due to O_3 within the Global 200 priority conservation areas. [Reprinted from Figure 8.8 in *Ground-level ozone in the 21st century: future trends, impacts and policy implications* (2008), RS Policy document 15/08, The Royal Society, London, United Kingdom.]

Impact of air pollution on climate change

The direct radiative impact of pollutants and their ICT is covered in detail in section 5.3. Here we cover indirect climate impacts of pollutants mediated through their effects on ecosystems.

A recent study by *Sitch et al*. [2007] found a significant suppression of the global land carbon sink due to O_3 induced damage to vegetation leading to reduced net primary productivity; with estimates of the reduction in land carbon sequestration being up to 260 PgC by 2100 based on SRES A2 emission scenarios. This reduced carbon sequestration leads to a higher atmospheric CO_2 concentration which was estimated to constitute an indirect radiative forcing that could exceed warming due to the direct radiative effect of tropospheric O_3 increases. However, elevated CO_2 concentrations may provide a degree of protection against O_3 damage, though there is uncertainty as to the magnitude and temporal longevity of this protection as well as which species groups would be affected (see Section 5.2.4.1).

The O_3 changes from the HTAP O_3 precursor experiments from the STOCHEM-HadGEM model have been applied to the offline dynamic vegetation model of *Sitch et al*. [2007] to quantify their impact on the carbon cycle [*Collins et al.*, 2010, see Figure 5.2.6]. The impacts of O_3 on vegetation and hence the land carbon storage are closely tied to the O_3 footprints of the precursor changes themselves. For NO_x emission changes, the impact of ICT on vegetation is small, with the possible exception of south Asian emissions impacting on east Asian vegetation. For emissions of longer-lived species such as CO, the resulting O_3 causing vegetation damage has a small component from ICT, but the dominant impact comes from emissions within the source region. On a 20 year timescale, O_3 precursor emissions cause a larger temperature change through indirectly increasing CO_2 than from the changes in O_3 or methane themselves.

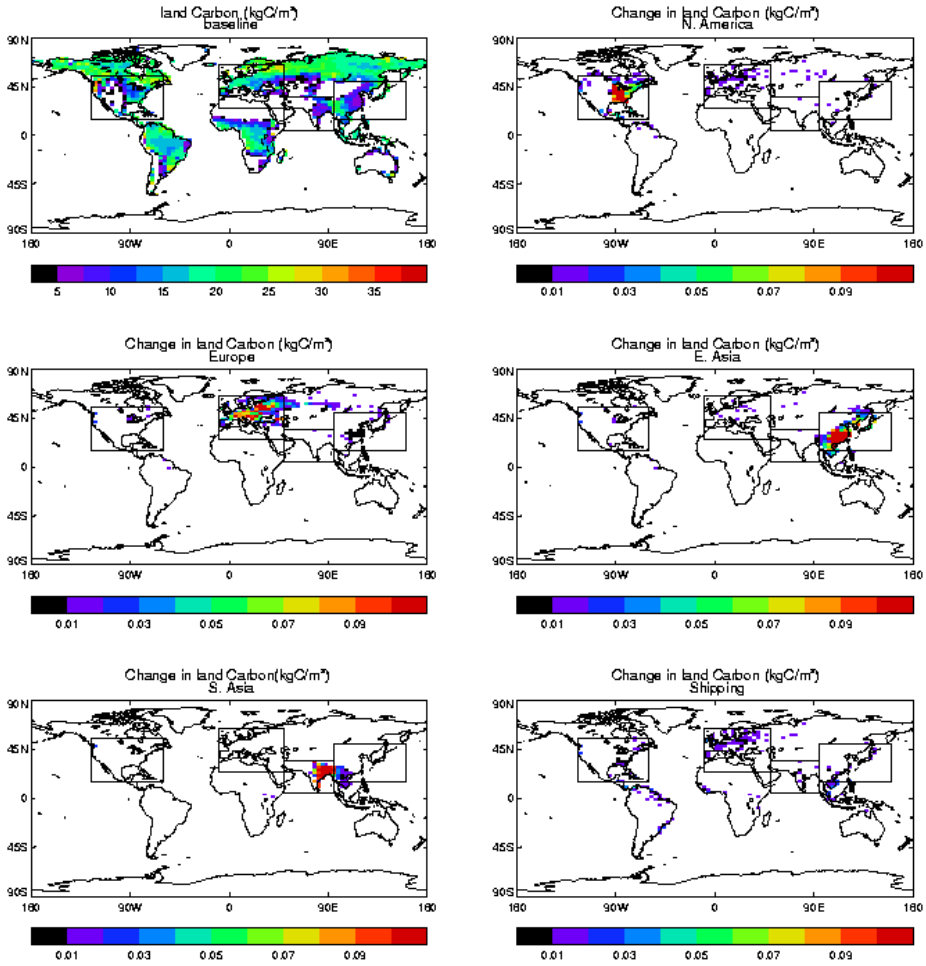


Figure 5.15. Change in land carbon at the end of the year following the HTAP 20% NO_x emission reductions. Boxes show the extent of the HTAP regions.

Global models that are now able to separate direct and diffuse radiation to assess the contribution of sunlit and shaded leaves to canopy photosynthesis [*Mercado et al.*, 2009] have recently been incorporated into fully coupled land-ocean atmosphere simulations and are hence capable of accounting for effects of short time-scale variability of PM loading. A first attempt to quantify the effects of PM and clouds on the regional and global C sinks [*Mercado et al.*, 2009] has estimated changes in diffuse fraction of -5 % to 30% during the global dimming period (1950-1980) which correspond to a contribution to the regional C sink of up to 30 gC m^{-2} yr^{-1} across Europe, eastern U.S., east Asia and some tropical regions in Asia (Figure 5.16). Conversely, during the brightening period (1980-2000), a reduction in diffuse fraction over Europe, eastern USA, western Australia, and some regions of Russia and China, led to a lower regional contribution to the land C sink from diffuse radiation. Globally, over the 1960-2000 period, diffuse radiation effects associated with changes in PM and clouds in the atmosphere enhanced the land C sink by about 25%. This more than offsets the negative effect of reduced surface radiation on the land carbon sink, giving a net effect of changes in radiation on the land carbon sink of 10% [*Mercado et al.*, 2009].'

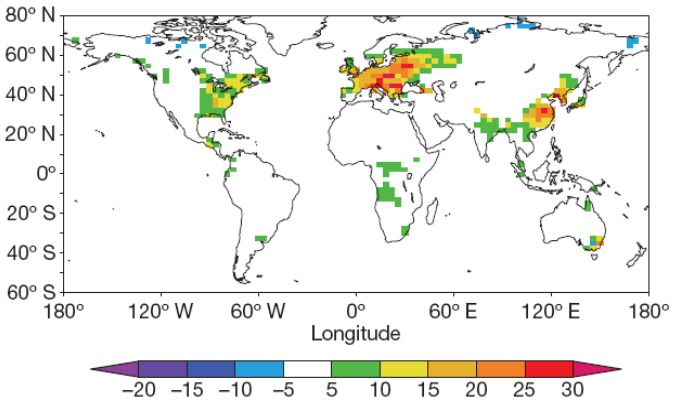


Figure 5.16. Simulated change (colour scale, grams carbon per square metre per year) in diffuse fraction contribution to land carbon accumulation between 1950 and 1980. [Reprinted from Figure 3(d) in Mercado, L. M., et al. (2009), Impact of changes in diffuse radiation on the global land carbon sink, *Nature, 458*: 1014-1017.]

5.2.5. Interactions with other pollutants

Damage to plants by a single pollutant can be altered by the presence of other pollutants, frequently with synergistic effects (e.g. between NO_2 and SO_2), but calculating response functions for each pollutant in the presence of others is difficult [*Bell*, 1985; *Bender and Weigel*, 1993].

There are also indirect feedback effects on climate of N and O_3 deposition to ecosystems. In particular, the enhanced atmospheric levels of N species and ground-level O_3 in Asia mean that this region will play an important role in determining the magnitude of these potential feedbacks with important implications for the climate system.

N deposition can lead to increased emissions of the potent GHG nitrous oxide (N_2O) from soils. It can also increase soil emissions of nitric oxide (NO), one of the important chemical precursors for O_3 formation. N deposition can also enhance the growth-rates of N-limited forests [*Hungate et al.*, 2009] resulting in enhanced uptake/sequestration of C in terrestrial ecosystems where N is the limiting nutrient; under enhanced CO_2 conditions, N limitation may become more common. However, where N deposition exceeds critical loads, adverse effects on growth and carbon sequestration can occur.

There is some evidence that a long-term trend of increased productivity in European forests is associated with environmental factors, including N deposition, as well as increased CO_2, and climate change [*Nabuurs et al.*, 2003], and is not simply due to improved management. These factors ameliorate the resilience of trees against O_3, but, on the other hand, O_3 itself is considered a factor potentially capable of reducing the ‘‘benefits’’ of CO_2 and N fertilization [*King et al.*, 2005; *Magnani et al.*, 2007]. Semi-natural grasslands are often limited by nutrients such as N or phosphorous. Alleviating such constraints for instance by the addition of N could decrease the sensitivity of the plant community to O_3through increasing biochemical detoxification capacity, or increase the sensitivity though increased stomatal conductance. However, to date evidence of these effects is limited and contradictory. Research into the N cycle is relatively less well developed than for the carbon cycle. Incorporation of the N cycle into earth system models is urgently needed to make further progress in assessing the interactions between air quality pollutants and climate.

FINDING: There are important interactions between O_3 and N deposition, and between both pollutants and climate and CO_2 concentrations, which have implications for future crop and forest production and global GHG budgets. However, these interactive effects are currently not well quantified.

RECOMMENDATIONS: Conduct experimental studies using O_3 profiles that simulate enhanced background O_3 concentrations to develop an improved understanding of the processes by which seasonality in O_3 exposure influence damage to ecosystems, perhaps with a focus on crops, forests and grasslands that have growth periods extending into those seasonal periods when the relative ICT contribution to pollution is greatest. Conduct a pan-Asian Open Top Chamber/Free Air Concentration Enrichment field campaign to establish dose-response relationships specific for Asian species (crops, forests and grasslands) growing under Asian climatic and management conditions. Assess the suitability of the flux-based O_3 index to identify ICT effects on ecosystems. Develop flux networks so that monitoring of O_3 fluxes in addition to other biogeochemical species (such as N, C and water vapour) is performed as standard. Conduct global modelling experiments that use flux-based indices to estimate the influence of ICT on ecosystem damage. Improve critical loads for tropical and Asian ecosystems. Improve understanding of how ICT may influence recovery or time-development of damage of ecosystems to acidification and eutrophication. Improve understanding of the aerosol-loading limit up to which ecosystems respond positively to an increase in diffuse fraction, the global and regional effect of diffuse radiation changes on the hydrological cycle, the implications for aerosol-based geo-engineering schemes and, the effect of diffuse radiation on croplands. Improve understanding of the interactions between acidification, eutrophication, ground level O_3, atmospheric aerosol loading and climate change. Research could be led by modelling efforts which identify knowledge gaps and hence drive experimental design to further scientific understanding. Improved modelling of ICT feedbacks to global climate taking into account the effects of ICT pollutants on GHG absorption by vegetation.

5.3 Impact of Long-range Transport on Climate

Ozone (O_3) and particulate matter (PM) have mainly been addressed by policy because of their impacts on air quality, but these air pollutants also have important influences on the Earth's climate. This section emphasizes the effects of anthropogenic O_3 and PM on regional and global climate, at present and in the future, and the effects of actions to reduce emissions to address these pollutants. In doing so, we mainly consider effects on radiative climate forcing, as well as on climate parameters. We do not estimate the ultimate impacts of climate change, including those to human health, ecosystems, agriculture and food supply, and coastal infrastructure. These impacts of climate change are detailed elsewhere [*IPCC*, 2007b].

The previous sections on the health and ecosystem impacts associated with LRT emphasize the effects of emissions from a source region on impacts in a receptor region, determined by pollutant concentrations or deposition. For climate, however, the effect of emissions from a source region on the climate of a receptor region is not determined entirely by pollutant concentrations over the receptor, and the source-receptor relationships in Chapter A4. Rather, the effects of emissions on regional forcings and atmospheric circulation become important, as do the effects on global average forcings and climate. Nonetheless, since O_3 and PM are short-lived climate forcing agents, the effect of a unit change in emissions on global and regional climate will vary with the location of those emissions.

We begin by analyzing the radiative forcing associated with the emissions relevant for O_3 and PM, and the effects of changes in emissions from different world regions. We then consider the importance of regional forcings for their ultimate influences on regional and global climate, and highlight the Arctic as a unique region that is clearly influenced by LRT. We conclude by considering future changes in the forcings of O_3 and PM and provide recommendations for future research.

5.3.1. Effects of ozone and particulate matter on global average radiative forcing

Radiative forcing and its relation to global climate change

Climate change can be driven by changes in atmospheric composition (e.g., carbon dioxide, methane, O_3, and PM), surface albedo (e.g., through changes in land use), and solar irradiance. Radiative forcing (RF) has been designated and extensively used to measure and compare, to a first order, the global climate impacts of different climate change drivers. RF is defined as the difference in net (down-welling minus upwelling) irradiance (solar plus thermal infrared radiation, in Watts per meter squared, Wm^{-2}) between an initial and a perturbed state of forcing agents, usually at the tropopause or the top of the atmosphere (TOA), with surface and tropospheric temperatures and state held fixed [*Forster et al.*, 2007]. To assess anthropogenic RF, the initial state is usually set in 1750, the approximate start of the industrial era, and the perturbed state in a recent year. By definition, a negative (positive) RF implies that the change in a climate driver exerts a cooling (warming) influence on the Earth-atmosphere system. Figure 5.17 summarizes the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4) estimates of major components of global average RF between 1750 and 2005 [*Forster et al.*, 2007]. While Figure 5.17 shows global mean annual average RF, the RF of particular forcing agents varies strongly with location and season.

For a sustained forcing, RF can be linearly related to the global mean equilibrium surface temperature response (ΔT_s),

$$\Delta T_s = \lambda \, RF$$

where λ is the climate sensitivity. For forcing agents like long-lived greenhouse gases (LLGHGs), approximately the same value of λ applies. However, for O_3 or aerosols, λ can be substantially different from that of LLGHGs, and more uncertain, and the estimated ratio of λ for a given forcing to that of CO_2 is defined as "climate efficacy" (Figure 5.17). Although RF does not reflect the overall climate response, providing only a limited measure of climate change, it is the most simple and commonly accepted indicator for quantitatively assessing the drivers of climate change [*NRC*, 2005].

Mechanisms and magnitudes of anthropogenic radiative forcing

Ozone

Anthropogenic emissions of O_3 precursors – NO_x, CO, NMVOCs, and methane – react chemically in the troposphere to produce O_3. Since O_3 is a radiatively active gas, absorbing in both the short-wave and long-wave, changes in O_3 precursors that increase O_3 will lead to a positive RF from O_3. However, these chemical reactions also affect methane and aerosol concentrations, leading to forcings that may affect the net RF [*Shindell et al.*, 2009], as discussed in section 5.3.2.1.

The AR4 estimate for the forcing from tropospheric O_3 changes from 1750 to 2000, due to anthropogenic emissions of precursors, is 0.25-0.65 Wm^{-2} (5-95%) with a best estimate of 0.35 Wm^{-2}. These estimates of O_3 forcing are largely based on chemistry model results, since few measurements are available of pre-industrial O_3. Gauss et al. [2006] showed that uncertainty in the O_3 forcing is largely driven by the variation in the O_3 changes simulated by the chemistry models, reflecting differing sensitivities of model chemistry schemes to changes in emissions of O_3 precursors.

O_3 also has an indirect warming effect on climate by inhibiting the natural uptake of CO_2 [*Sitch et al.*, 2007], as discussed in section 5.2.2.5. This may have led to a doubling of the climate forcing attributable to O_3 since pre-industrial times. The estimates are very uncertain as studies of O_3 impacts have focused on crop species, with fewer studies of natural forests (especially in the tropics).

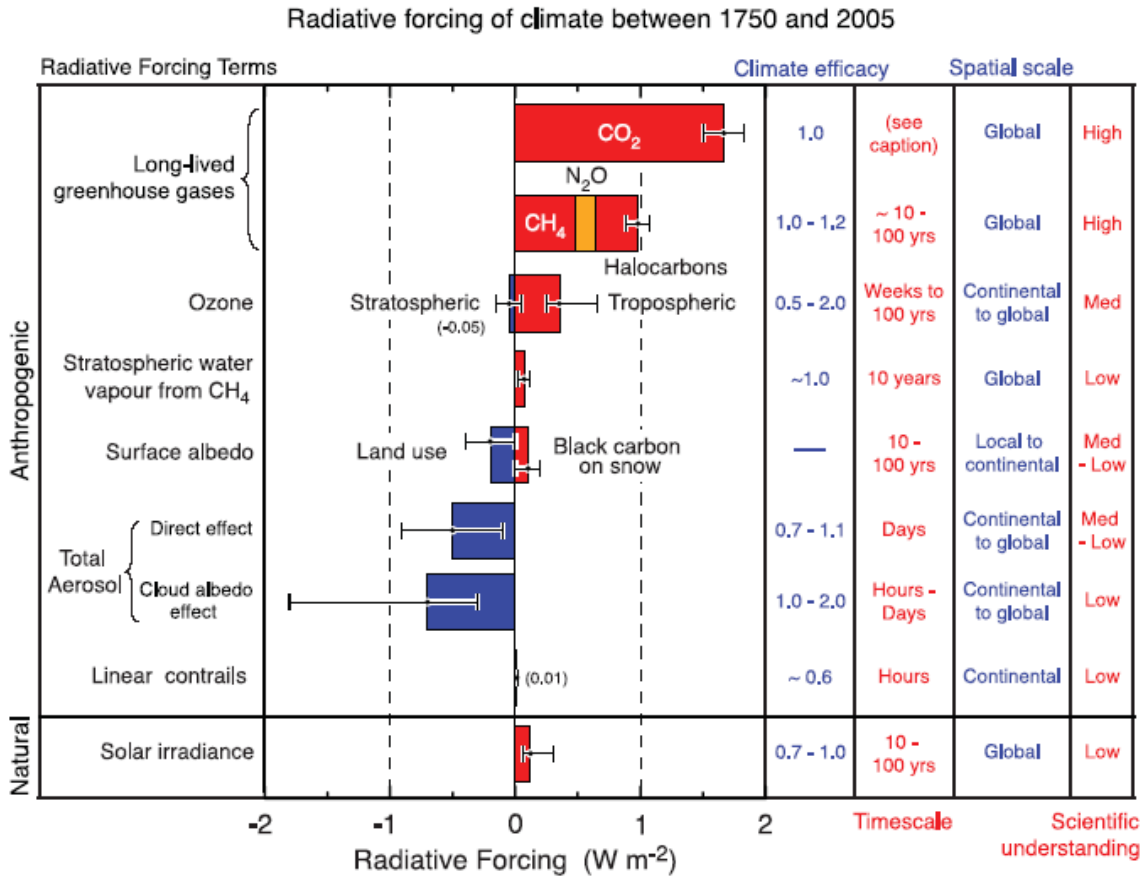


Figure 5.17 IPCC AR4 summary of averages and ranges of global mean radiative forcing (RF) in units of Watts per square meter (Wm^{-2}) between 1750 and 2005 for major forcing agents, including carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), ozone, land use, aerosols, and solar irradiance. Typical climate efficacy, timescale, geographical extent of forcing, and the assessed level of scientific understanding are also given. No CO_2 time scale is given, as its removal from the atmosphere involves a range of processes that can span long time scales, and thus cannot be expressed accurately with a narrow range of lifetime values. [Adapted from Forster, P. et al. (2007), Changes in atmospheric constituents and radiative forcing, *Climate Change 2007: The Physical Scientific Basis*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.]

PM

Aerosols perturb the Earth's energy budget *directly* by scattering and absorbing radiation and *indirectly* by acting as cloud condensation nuclei and in doing so changing cloud properties. Box 5.2 describes a suite of mechanisms by which aerosols interact with radiation and affect climate. The IPCC estimates that anthropogenic aerosol direct RF is -0.5 [±0.4] Wm^{-2} and indirect RF through the cloud albedo effect on liquid water clouds is -0.7 [-1.8, -0.3] Wm^{-2}. Globally-averaged, the sum of aerosol direct and indirect RF of -1.2 Wm^{-2} [-2.4 to -0.6 Wm^{-2}] (cooling) is significant compared to the positive (warming) forcing of 2.63 [±0.26] Wm^{-2} by anthropogenic LLGHGs [*Forster et al.*, 2007]. However, the uncertainty in aerosol RF is much greater than that of LLGHGs, and is among the most important uncertainties in understanding anthropogenic climate change [*Andreae et al.*, 2005]. These uncertainties result from the strong spatial heterogeneity in concentrations, uncertainties in emissions, chemical processing and lifetimes, and uncertainties in the effects of aerosols on cloud processes. Different aerosol components may provide warming or cooling based on the properties of the aerosol, atmosphere, and underlying surface. BC is estimated to cause a total globally-averaged positive forcing of 0.34 (0.09 to 0.59) Wm^{-2} (Figure 5.17). This includes a forcing from deposition on snow estimated as +0.1 [±0.1] Wm^{-2}, or +0.05 Wm^{-2} [0.007, 0.13 Wm^{-2}] in a recent study [*Flanner et al.*, 2007].

Box 5.2. Mechanisms of Aerosol Radiative Forcing

Aerosols interact strongly with solar and terrestrial radiation in several ways, as illustrated in Figure 5.18. They scatter and absorb radiation, thereby perturbing the radiative balance of the Earth-atmosphere system [*McCormick and Ludwig*, 1967]; these are described as "aerosol direct effects". Aerosol direct effects in the solar spectrum generally dominate over that in the thermal infrared, except for large particles such as mineral dust. Purely scattering aerosols (including sulphates, nitrates, and sea salt) exert a negative RF by reflecting solar radiation back to the space, while partly absorbing aerosols (including dust and organics) may exert a negative RF over dark oceans and vegetated land but a positive RF over bright surfaces such as deserts, snow, ice, and clouds. Black carbon is a strongly absorbing aerosol, expected to give a positive global average RF. As particles age, their chemical composition can change to more of a mixture, affecting their optical properties, lifetimes, and uptake of water.

Aerosols also interact indirectly with radiation through acting as cloud condensation nuclei and/or ice nuclei. Increases in aerosol particle concentrations may increase the ambient concentration of cloud condensation and ice nuclei, causing smaller but more numerous cloud droplets for fixed cloud water content [*Twomey*, 1977]. This makes clouds reflect more solar radiation, causing a negative RF (the "indirect effect" or "cloud albedo effect").

Other effects of aerosols are also important for climate change. Both aerosol scattering and absorption decrease the solar radiation reaching the surface, causing the negative RF at the surface to be stronger than at the TOA. Aerosol absorption can also increase atmospheric radiative heating. The simultaneous surface cooling and atmospheric heating alter convection and cloud formation, thus changing cloud extent and optical properties as well as circulation and precipitation patterns [*Hansen et al.*, 1997]. The changes in cloud microphysics due to increases in aerosols may also alter the likelihood and intensity with which a cloud eventually precipitates [e.g., *Albrecht*, 1989; *Gunn and Phillips*, 1957], but these effects, particularly on mixed-phase or ice clouds, are not well understood. It has proven difficult to establish climatically meaningful relationships between aerosols, clouds, and precipitation [*Stevens and Feingold*, 2009].

Finally, the deposition of absorbing aerosols such as black carbon and dust in snow can reduce the albedo of snow, mainly in the visible, because of enhanced absorption by multiple scattering in the snowpack. This causes a positive RF and promotes melting of snow and ice [*Warren and Wiscombe*, 1980].

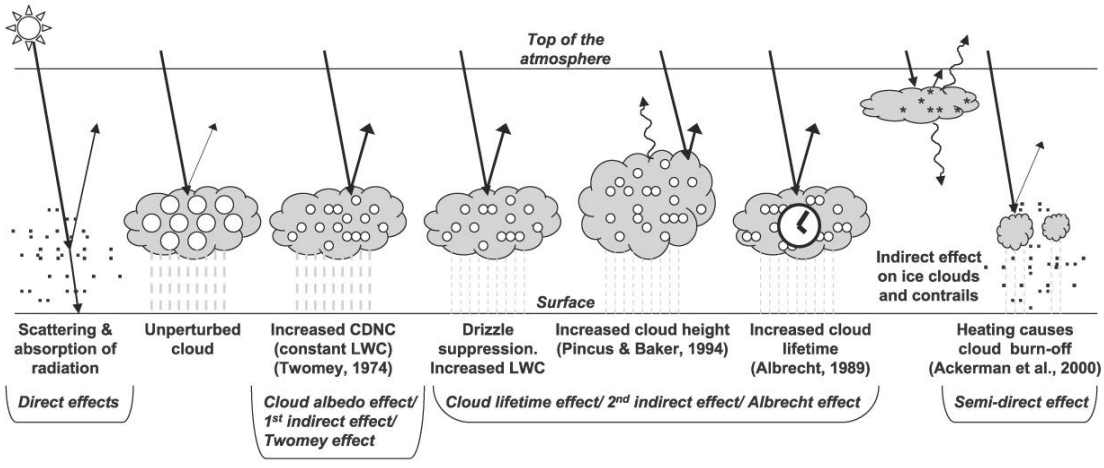


Figure 5.18. Schematic diagram showing the major radiative forcing mechanisms related to aerosols. The small black dots represent aerosol particles; the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The filled white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger cloud drops as only natural aerosols are available as cloud condensation nuclei, while the perturbed cloud contains a greater number of smaller cloud drops as both natural and anthropogenic aerosols are available as cloud condensation nuclei (CCN). The vertical grey dashes represent rainfall, and LWC refers to the liquid water content. [Adapted from Forster, P., et al. (2007), Changes in atmospheric constituents and radiative forcing, *Climate Change 2007: The Physical Scientific Basis*, Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.]

These aerosol forcing assessments, based largely on model calculations, are also constrained as much as possible by observations. For example, the IPCC estimate of aerosol RF based on forward calculations has recently been corroborated by an inverse calculation of -1.1 Wm^{-2} [-1.9 to -0.3 Wm^{-2}] [*Murphy et al.*, 2009]. Similarly, updated estimates of the aerosol direct RF, based on satellite observations, include -0.65 Wm^{-2} [*Bellouin et al.*, 2008] and –0.9 [±0.4] Wm^{-2} [*Quaas et al.*, 2008], which is stronger than the IPCC AR4 estimate and model-based estimates generally, and Myhre [2009] estimates a weaker direct RF of -0.3 [±0.2] Wm^{-2}.

FINDING: Increases in ozone and PM since preindustrial times have caused significant climate forcing. Ozone is a greenhouse gas that causes warming; in addition, damage to plants by ozone inhibits the natural uptake of CO_2. PM is a mixture containing components that mainly cool, including sulphate and organic aerosols, and black carbon that warms. PM also influences climate by changing clouds.

Short-lived vs. Long-lived Forcings and Relations to Air Pollution

Because O_3 and PM affect climate, actions to reduce air pollutant concentrations also affect climate. Reductions in O_3 would both improve air quality and slow global climate change. However, as discussed in section 5.3.2, reductions in emissions of O_3 precursors also influence methane, and can influence aerosols and the global carbon cycle. Therefore, the net effects of changes in emissions of each O_3 precursor need to be evaluated for both air quality and net radiative climate forcing. Among the components of PM, BC aerosols are identified as the only form that causes warming on a global scale, and reductions in BC emissions would likely improve PM air quality and slow climate change. Other components of PM have a net cooling influence, and so reducing these emissions likely exacerbate warming. However, analysts and governments have been reluctant to treat emissions of PM and their precursors as a benefit to climate, for two main reasons: (i.) their significant effects on air quality, health, and ecosystems, and (ii.) the possible effects of their regionally-specific forcings on climate (Section 5.3.3).

FINDING: Any change in emissions that influences ozone and/or PM will generate a climate forcing.

O_3 and PM differ from the major greenhouse gases in that perturbations to them produce relatively short-lived forcings (Figure 5.17). Whereas CO_2 and N_2O perturbations persist in the atmosphere for over a century, and those of methane decay on decadal timescales, the lifetime of O_3 is on the order of weeks, while those of PM and its precursors are on the order of days. Despite their short life, emissions of PM and its precursors significantly counteract the RF due to the accumulation of years of emissions of the LLGHGs. As emissions continue in the future, the LLGHGs will continue to accumulate in the atmosphere, while the short-lived forcers will not. Over the long term, LLGHGs will very likely dominate. However, because of the short lifetimes of O_3, BC, and also methane, actions to reduce these emissions would be very effective at slowing the short-term rate of global warming over the coming decades [*Hansen et al.*, 2000], leading some to define Short-Lived Climate Forcers (SLCFs) as a problem separate from, but related to the warming of LLGHGs [*Jackson*, 2009; *U.S. CCSP*, 2008]. Despite the clear benefits of reducing these emissions, BC and O_3 precursors (with the exception of methane) have not been included in international actions to address climate change. Similarly, reductions in PM components other than BC will exacerbate short-term warming, and may cause the rate of climate change to be greatest before the 2030s [*Raes and Seinfeld*, 2009].

FINDING: Emissions relevant for ozone and PM have short atmospheric lifetimes, compared to the long-lived greenhouse gases, and will strongly influence the rate of climate change in the coming decades. There are opportunities to improve air quality while slowing climate change, but many actions to reduce PM would exacerbate warming.

For LLGHGs, it is common to compare the climate forcings of different changes in emissions by using the Global Warming Potential (GWP), which integrates forcing over a selected time horizon, typically 20 or 100 years. Because O_3 and PM have lifetimes much shorter than that of LLGHGs, the choice of time horizon has a large influence on the estimated GWP.

Because short-lived species are distributed inhomogeneously, the resulting forcing will depend on where the species are emitted and where they or their products are transported. Section 5.3.2 addresses the spatial extent of RF caused by emissions from a source region, as well as the nonlinear dependencies of forcing on precursor emissions. The impacts of spatially inhomogeneous forcings on regional and global climate are then addressed in Section 5.3.3.

5.3.2. Radiative Forcing of Ozone and PM: Regional Extent and Effects of Varying Precursor Emissions

Because O_3 and PM have short lifetimes, their forcings are largely exerted on regional to intercontinental scales. In this section, we address the spatial extents of forcings from regional emissions and the importance of LRT for climate. In addition, for both O_3 and PM, the effects of changes in emissions depend strongly on chemistry, gas-aerosol interactions, and other factors. Consequently, we explore the importance of these many influences in determining the RF of changes in emissions of different precursors from different world regions.

Ozone

RF generated by increases in tropospheric O_3 since pre-industrial times [e.g., *Berntsen and Isaksen*, 1997; *Gauss et al.*, 2006; *Shindell et al.*, 2003], and also projected for the future [e.g., *Gauss et al.*, 2003; *Stevenson et al.*, 2006], has a distinct regional structure. This is the result of spatial variations in several contributing factors: (i) horizontal and vertical distributions of changes in O_3; (ii) various physical aspects of the climate system that determine the passage of radiation through the atmosphere (e.g., surface and cloud albedos, for short wave radiation; and surface and vertical profiles of temperature and clouds, for long wave radiation); and (iii) how (i) and (ii) overlap. While local changes in O_3 at the surface are very important for air quality, changes in O_3 over a larger area, and in the middle to upper troposphere (where O_3 exerts a larger RF per unit change in concentration) become particularly important for climate. While O_3 at a given location has a complex dependency on precursor emissions from local and distant sources (Chapter A4), the above factors make this picture even more complicated for RF.

Some studies have calculated RFs due to emissions from specific regions [e.g., *Derwent et al.*, 2008; *Naik et al.*, 2005; *Naik et al.*, 2007; *Stevenson and Derwent*, 2009]. Figure 5.19 shows O_3 RFs resulting from 10% NO_x emissions reductions from specific regions, including only RFs from the initial, short-term increase in O_3, and not the long-term impacts on O_3 associated with changes in CH_4. The O_3 RF is partly determined by the extent of transport of O_3 and its precursors away from source regions. The extent of transport depends upon the wind field and the lifetimes of the species, both of which may vary seasonally and inter-annually, and may also change as climate changes. Naik et al. [2005] found that the short-term O_3 RF from NO_x emissions varies with season of emission, with typically the largest O_3 RF occurring in the summer. The strong influences of emissions from North America (NA), Europe (EU) and East Asia (EA) result from the large emissions in these regions. Figure 5.20 shows that when RF is normalized by NO_x emissions, SE Asia (SE), S America (SA) and India (IN) produce the largest O_3 RF per unit NO_x emission, illustrating greater sensitivity in the tropics than mid- and high-latitudes. In a similar study of aircraft NO_x emissions, Stevenson and Derwent [2009] found that regions with low background NO_x generated the most O_3 RF per unit NO_x emission, whereas latitude was a less important influence.

Changes in emissions of O_3 precursors (NO_x, CH_4, CO and VOCs) have additional impacts on RF, as they alter hydroxyl radical (OH) concentrations, affecting the removal rate and, therefore, the concentration of CH_4. NO_x emissions tend to increase OH (which reduces CH_4, Figure 5.20); whereas CH_4, CO and VOC emissions tend to consume OH (which increases CH_4). In addition, oxidation of these C-containing species produces CO_2. The effects of decreasing emissions of NO_x have been shown to cause a positive net RF, as the RF of the CH_4 increase is greater than that of the ozone decrease [e.g., *Wild et al.*, 2001], and this has been shown to be the case for NO_x emissions from all world regions (Fig. 5.3.4). Further forcings arise because CH_4 oxidation is a significant source of tropospheric O_3, in addition to stratospheric H_2O, and NO_x produces nitrate aerosol. Any change in oxidants will also affect formation rates of secondary aerosols.

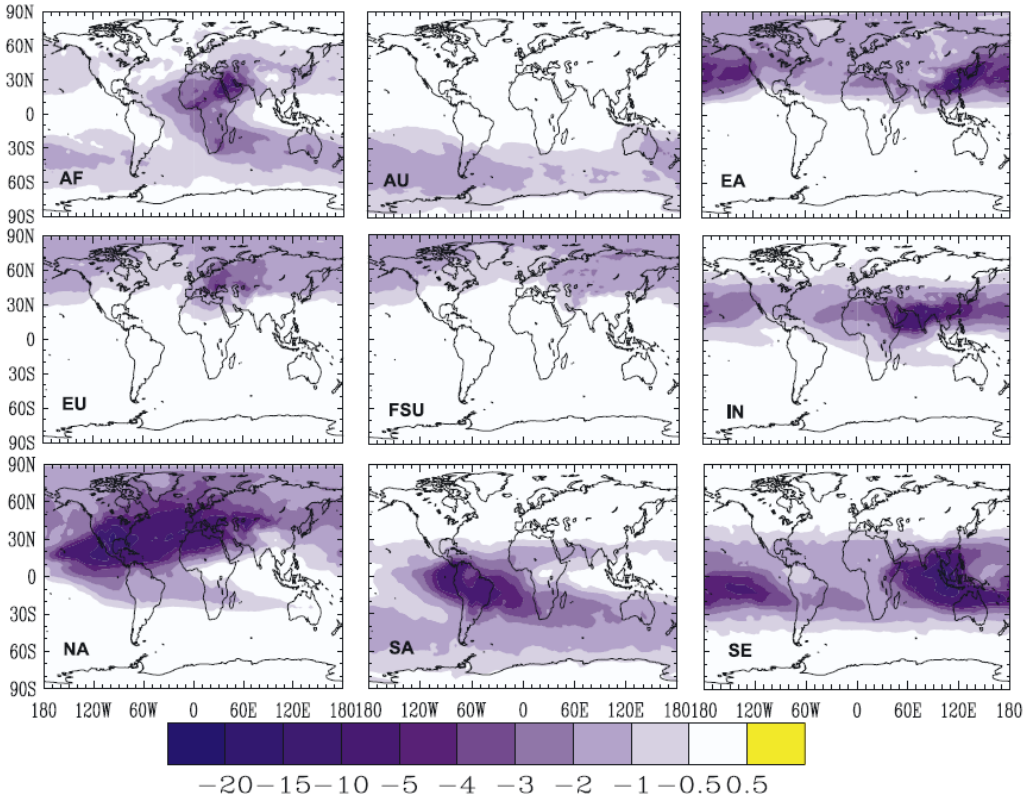


Figure 5.19. Annual total-sky instantaneous RF (mWm^{-2}) due to short-term changes in O_3 from a 10% reduction in surface anthropogenic NO_x emissions from each of nine world regions. [Reprinted from Figure 7 in Naik, V., et al. (2005), Net radiative forcing due to changes in regional emissions of tropospheric ozone precursors, *Journal of Geophysical Research, 110* (D24306).]

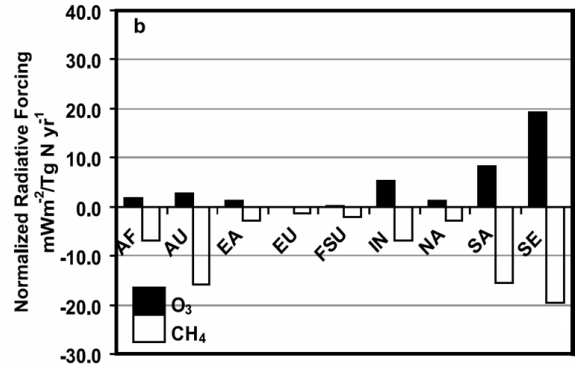


Figure 5.20. The change in annual average RF per unit change in NO_x emissions for 10% regional NO_x reductions in Figure 5.19 [adapted from Figure 9b in Naik, V., et al. (2005), Net radiative forcing due to changes in regional emissions of tropospheric ozone precursors, *Journal of Geophysical Research, 110* (D24306)]. The O_3 RFs include an estimate of the long-term contribution due to changes in methane. Associated estimates of methane RF are also shown; these have the opposite sign as the O_3 RFs, and are larger in magnitude.

By modelling the effects of emissions of each precursor, the IPCC AR4 Assessment [*Forster et al.*, 2007] attributed the global mean annual average RF (in Wm^{-2}) to anthropogenic changes in emissions: 0.86 from CH_4, 0.27 from CO and VOCs, and -0.21 from NO_x. These emissions-based estimates of forcing differ from the abundance-based estimates in Figure 5.17, and include the effects of

methane on CO_2, O_3, and stratospheric water vapour, the effects of NO_x on O_3, methane and nitrate aerosols, and the effects of CO and VOCs on CO_2, methane and O_3. A recent study [*Shindell et al.*, 2009] found somewhat different RF values (Wm^{-2}) for these O_3 precursor species (Figure 5.21): 0.99 from CH_4, 0.25 from CO+VOCs (0.19 from CO and 0.06 from VOCs), and -0.29 from NO_x. Differences relative to AR4 resulted largely from the inclusion of additional gas-aerosol interactions, so that these calculations included the effect of all emissions on sulphate and nitrate aerosols. For changes in methane emissions, the temperature change 20 years after a methane emission pulse is ~57 times that of CO_2 on a kg/kg basis. This factor of 57 can be broken down into 41 from the direct effect, 10 via tropospheric ozone production and 6 via stratospheric water vapour [*Boucher et al.*, 2009].

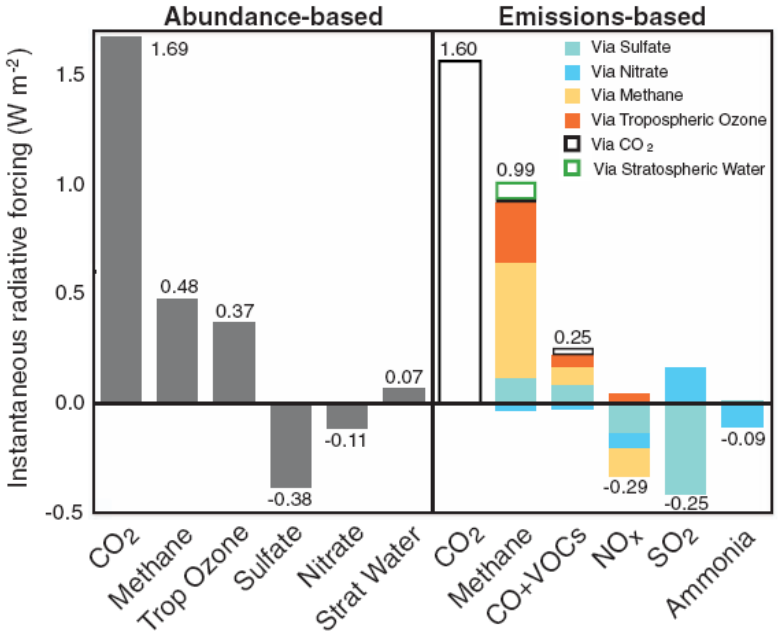


Figure 5.21. Radiative forcing from 1750 to 2000. Numerical values within the figure give the net forcing (instantaneous at the tropopause). Uncertainties in the abundance-based values are 0.16 for CO_2, 0.05 for methane, +0.15 to -0.10 for ozone, 0.20 for sulphate, 0.10 for nitrate, and 0.05 for stratospheric water. For emissions-based values, uncertainties are estimated as 0.14 for methane, 0.04 for CO+VOCs, 0.09 for NO_x, 0.23 for sulphate, and 0.10 for ammonia. The aerosol indirect effect is not included. [From Figure 1 from Shindell, D., et al. (2009), Improved attribution of climate forcing to emissions, *Science, 326(*5953): 716-718. Reprinted with permission from AAAS.]

The estimates of anthropogenic RF in Figure 5.21 also have bearing on the RF from changes in emissions. At a global scale, reductions in emissions of O_3 precursors have been studied for their effects on both surface O_3 air quality and radiative climate forcing. Among O_3 precursors, global reductions in methane emissions have been shown to best reduce RF per unit improvement in O_3 air quality metrics, mainly because of the reduction in methane itself, followed by CO and NMVOCs, while NO_x emissions are expected to lead to a net positive RF [*West et al.*, 2007a]. These conclusions are supported by Fig 5.3.5, where effects on aerosol species are included. Note, however, that the above calculations do not include the effect of O_3 on carbon uptake (section 5.2.2.5).

Except for methane, the effects of O_3 precursor emissions on global mean RF differ with the location of those emissions, depending upon factors such as the local chemical regime, lifetime, and photolysis rates of key species. Several modelling studies have attempted to quantify the RF of the geographic distribution, altitude and season of O_3 precursor emissions [e.g., *Berntsen et al.*, 2005; *Derwent et al.*, 2008; *Fuglestvedt et al.*, 1999; 2010; *Naik et al.*, 2005]. In Figure 5.20, reductions in surface NO_x emissions from all world regions cause a positive RF, for sustained emissions changes at steady state, although the sensitivity of O_3 and CH_4 per unit change in emissions varies considerably

among regions. Most studies suggest high altitude aircraft NO_x emissions produce a net warming [e.g., *Köhler et al.*, 2008; *Lee et al.*, 2009; *Wild et al.*, 2001], whilst others suggest a net cooling [*Stevenson et al.*, 2004; *Stevenson and Derwent*, 2009]. Background levels of NO_x were the primary determinant of the net climate forcing from aircraft NO_x, with larger net forcings produced by emissions into cleaner regions; latitude of emission was a lesser influence [*Stevenson et al.*, 2004; *Stevenson and Derwent*, 2009]. Analyses of the dependence of RF on the location of CO and VOC emissions have not been as detailed as for NO_x, nor have seasonal dependencies been quantified, and the available studies of regional emissions do not include the effects on aerosols [*Shindell et al.*, 2009], nor the effect of O_3 on plants and the carbon cycle [*Sitch et al.*, 2007].

The time dependence of the climate impact has been studied by Collins et al. [2010], who analysed year-long pulse emission reductions from the HTAP regions, accounting for changes in both O_3 and methane. In Figure 5.22, the HTAP NO_x reductions initially cool climate since less O_3 is produced; however this effect decays and is replaced by a warming due to less destruction of methane. For VOC and CO emissions the O_3 and methane changes both cool climate. The initial dip (more pronounced for the VOC emissions) is due to the impact on O_3, the longer tail due to the impact on methane. As for Naik et al. [2005] and Berntsen et al. [2005], the O_3 is most sensitive to NO_x emissions in a cleaner atmosphere (S. Asia) although it is least sensitive to the VOC emissions here. The methane destruction is most sensitive to tropical emissions (S. Asia). For CO, the only noticeable regional dependence is for European emissions, which being at the highest latitude have the least impact on methane. This study does not include the impact of O_3 on plants and the carbon cycle, and assumes a constant forcing per unit column burden and constant climate sensitivity regardless of location. Berntsen et al. [1997] and Shindell and Faluvegi [2009] suggest these quantities have a latitudinal dependence.

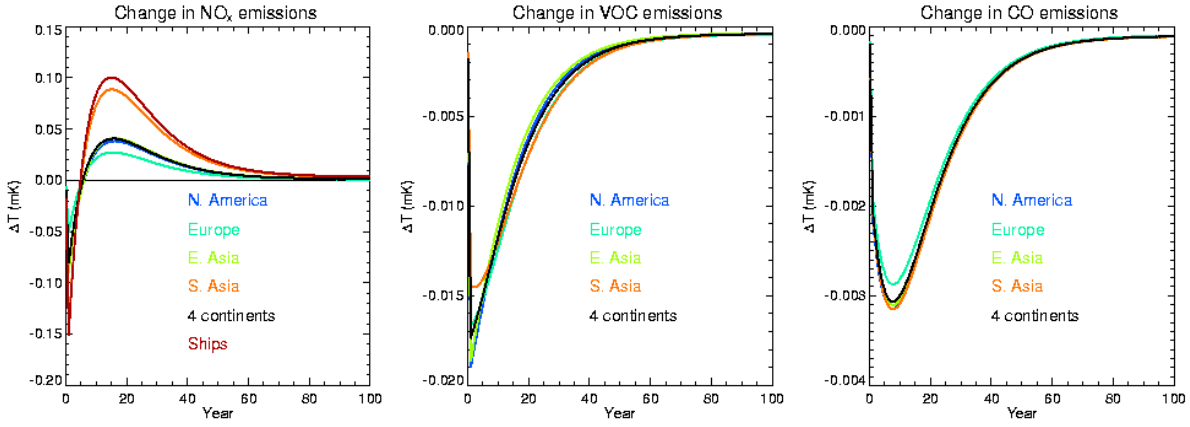


Figure 5.22. Change in global surface temperature for the years following a 1 Tg emission reduction in the first year. The normalisation is per Tg N, C and CO for NOx, VOC and CO. [Adapted from Figure 7 in Collins, W. J., et al. (2010), How vegetation impacts affect climate metrics for ozone precursors, *Journal of Geophysical Research, 115* (D23308).]

FINDING: Among ozone precursors, widespread reductions in emissions of CH_4, CO, and NMVOCs better decrease net climate forcing than reducing NO_x, which may increase forcing.

FINDING: Except for methane, the effects of ozone precursor emissions on global mean RF differ with the location of those emissions.

PM

Like O_3, the distribution and magnitude of the aerosol direct and indirect RFs are mainly determined by the spatial patterns of PM concentrations, but also depend on the absorption and scattering properties of those aerosols, their possible roles as cloud condensation nuclei, and environmental factors such as surface albedo and the presence of clouds [*Forster et al.*, 2007]. Industrialised regions of North America, Europe and Asia, and biomass-burning regions of Africa and South America are associated with large, negative aerosol direct forcing. Positive aerosol direct

forcing occurs where absorbing aerosols overlie bright surfaces, such as in the stratocumulus regions and over ice surfaces of the polar regions.

The spatial extents of aerosol direct RF from regional emissions and the role of aerosol LRT are revealed by a preliminary analysis comparing the HTAP PM SR6 perturbation runs (20% reductions in regional anthropogenic emissions), relative to SR1. We calculate direct RF from the archived aerosol optical depth fields for each component (i.e., sulphate, BC, or POM) from SR6 and SR1, in combination with RF efficiencies (forcing per unit optical depth) from the GOCART model [*Yu et al.*, 2006]. The seven models used in this preliminary analysis are: CAMCHEM (version 3311m13 and version 3514), ECHAM5-HAMMOZ, GISS-PUCCINI modelE, GMI, GOCART, and SPRINTARS. Figure 5.23 shows that emissions from North America, Europe, and East Asia exert significant RF on intercontinental and hemispheric scales. Emissions from South Asia have relatively small impacts mainly over the tropical Pacific. For all four regions, the extent of direct RF is much smaller in winter than in other seasons (not shown).

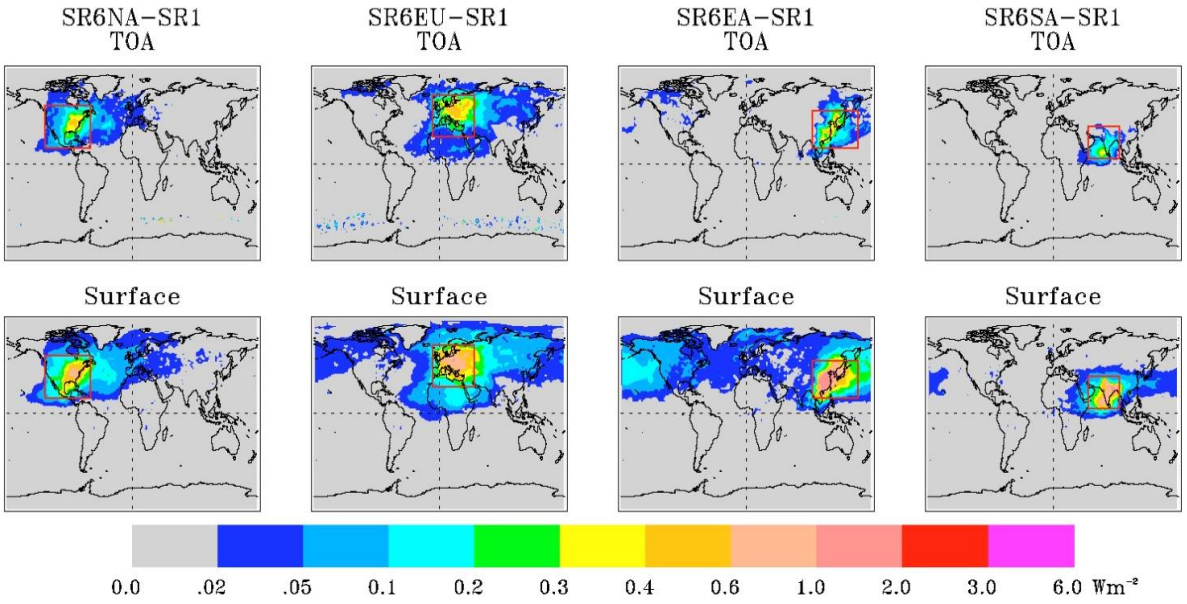


Figure 5.23. The annual average all-sky aerosol direct RF (Wm^{-2}) at the top of atmosphere (TOA) (top panel) and at the surface (bottom panel) resulting from 20% reductions of anthropogenic emissions over North America (NA), Europe (EU), East Asia (EA), and South Asia (SA), respectively. A positive RF represents that the aerosol direct RF is weakened by the reduction of emissions. Each of the four source regions is overlaid on corresponding SR6xx-SR1 maps.

As in Chapter A4, we calculate the relative annual intercontinental response (RAIR) as the direct RF response in a receptor region to the combined influence of 20% emission reduction in the three foreign emission regions, divided by the response to 20% emission reduction in all four regions. RAIR in a region represents the percentage contribution of LRT of foreign emissions relative to a sum of foreign and domestic emissions. RAIRs for aerosol optical depth (AOD) (Table 5.6) and direct RF (Table 5.7) depend on both region and species. For all regions and species, import from LRT is significant, with RAIR ranging from 9 to 30%. South Asia is most influenced by import of sulphate aerosol, and North America is most influenced by import of BC, followed by POM. For sulphate, POM, and BC combined, South Asia RF is most strongly influenced by foreign sources, and North America and Europe is the least. These rankings also reflect the strength of local emissions, relative to world emissions. Interestingly, RAIR values for optical depth and direct forcing of sulphate are much smaller than that for aerosol column loading [see Table 4.4 in Chapter A4]. This probably stems from transported sulphate aerosols experiencing lower relative humidity at high altitude (resulting in lower

optical depth) than local aerosols, which generally remain at lower altitudes. Finally, model variability is very large, highlighting the significant uncertainties in modelling aerosol processes.

Table 5.6. Multi-model derived relative annual intercontinental response (RAIR) (mean ± std. dev) for aerosol optical depth for four HTAP regions, in SR6 simulations relative to SR1, by chemical component. Standard deviations measure the model variability in simulating aerosol transport. BC stands for black carbon aerosol, and POM for particulate organic matter aerosol.

Receptor	NA	EU	EA	SA
BC	27%±16%	19%±12%	15%± 3%	17%± 6%
POM	21%±14%	19%±11%	19%± 4%	12%± 6%
Sulphate	14%±8%	12%±6%	16%±6%	30%±8%
BC+POM+Sulphate	16%±8%	14%±5%	16%±5%	25%±7%

Table 5.7. As Table 5.6, but for RAIR for TOA all-sky aerosol direct RF.

Receptor	NA	EU	EA	SA
BC	29%±17%	22%±13%	16%± 4%	21%± 6%
POM	21%±15%	19%±12%	21%± 6%	11%± 6%
Sulfate	13%±8%	12%±6%	16%±6%	29%±8%
BC+POM+Sulphate	9%±10%	10%±8%	17%±10%	26%±9%

These general findings on the extent of RF from regional emissions are supported by other studies. Reddy and Boucher [2007] and Shindell et al. [2008c] found that BC emitted in Asia contributes 20% of the BC burden in North America. Bond [2004] found that BC exerts a regional scale radiative forcing much larger than its local influence on air quality. On a regional scale again, Lau et al. [2006] suggest that locally large BC and mineral dust concentrations in India, combined with the specific orographic features of the Indian subcontinent, profoundly affect the Asian monsoon and Himalayan climate. Wang and Christopher [2006] found that the transport of Central American smoke aerosols to the southern U.S. in spring can decrease the monthly-averaged air temperature in Texas near the surface by 0.2°C, but enhance the upper boundary layer air temperature by ~0.1°C.

FINDING: In the four HTAP regions, the contribution of foreign regions to aerosol RF may be roughly 10-30% of the total.

The global mean anthropogenic RF has been attributed by the IPCC AR4 to emissions of particular species (in Wm^{-2}): 0.44 for BC, and -0.19 for OC. In general, RF is expected to respond roughly linearly to changes in emissions for primary PM species, but may be nonlinear for secondary aerosols. In Figure 5.21, estimates of RF attributable to anthropogenic sulphur dioxide and ammonia include the interaction between the resulting sulphate and nitrate aerosols, finding forcings of -0.25 for SO_2 and -0.09 for ammonia, and the -0.29 for NO_x includes both gas-phase and aerosol changes. Future research will need to evaluate anthropogenic contributions to global secondary organic aerosols, both through emissions of VOCs and through changes in atmospheric oxidants.

Just as aerosols exert a RF regionally, the effects of changes in emissions of PM and its precursors on global mean RF also varies with region. Table 5.8 lists changes of global annual average TOA direct

RF in response to the 20% reduction of anthropogenic emissions in four HTAP source regions. The global mean annual average TOA direct RF from baseline (SR1) simulations of 6 models is -0.74±0.27, -0.26±0.09, and +0.36±0.12 Wm^{-2} for sulphate, POM, and BC, respectively, where both anthropogenic and natural sources are included. Uncertainties are likely to be systematically underestimated due to the use of a single model to calculate RF from all models' simulated aerosol changes. The combined impact of 20% emission reductions in the four regions is to weaken the RF by 9%, 4%, and 9% for sulphate, POM, and BC, respectively. Relative contributions from individual regions vary. For sulphate, the change of global average RF due to the reduction of emissions in South Asia is substantially smaller than that due to the emission reductions from the other regions. For POM and BC, the reduction of emissions in East Asia makes the largest contribution to the change of global average RF. Table 5.9 shows the global annual average TOA all-sky aerosol direct RF efficiency per unit emissions from the source regions. The forcing efficiency for BC is an order of magnitude higher than that for sulphate and POM. While some models show large regional dependence of the forcing efficiency for some components, the multi-model average forcing efficiency is not strongly dependent on region. Note that these results do not include aerosol indirect effects, nor warming resulting from BC deposition on snow and ice.

Table 5.8. Global annual average TOA all-sky aerosol direct RF (unit: mWm^{-2}) in response to the 20% reduction of anthropogenic emissions in 4 source regions, for the mean ± std. dev for SR1 and SR6 simulations of six HTAP models.

Source Region	Sulfate	POM	BC	Sulphate+POM+BC
NA	16.1±5.6	1.6±1.0	-4.5±1.9	13.2±5.2
EU	26.7±9.5	1.9±1.2	-7.4±2.3	21.2±9.5
EA	19.6±7.2	3.2±1.8	-14.5±8.0	8.4±10.2
SA	6.1±1.9	2.5±1.3	-5.5±2.4	3.1±3.2
NA+EU+EA+SA	68.4±22.9	9.1±5.0	-31.9±13.7	45.9±24.6

For BC, the relative impact on global mean RF for emissions from different areas has been calculated [*Fuglestvedt et al.*, 2010; *Reddy and Boucher*, 2007; *Rypdal et al.*, 2009]. Reddy and Boucher [2007] found that BC transport to the Arctic is dominated by European emissions. The RF per unit emissions of BC can thus be significantly larger for emissions from Europe than for other regions, because of the higher forcing efficiency of BC over bright surfaces such as in the Arctic, while other results find greater RF for emissions from open burning in Northern Asia [*Bond et al.*, 2010]. The diversity in BC transport and deposition among numerical simulations is large, especially in Northern Eurasia and the Arctic [*Koch et al.*, 2009]. Source-receptor relationships are therefore likely to be model dependent. Nonetheless, analysis of these relationships in 17 HTAP models showed that even for transport to the Arctic, the models are quite consistent in the relative importance of transport from different regions, despite the diversity of absolute transport amounts [*Shindell et al.*, 2008c]. Rypdal et al [2009] and Fuglestvedt et al. [2010] generally showed higher forcing per unit emission for BC emissions from South Asia and to a lesser extent South America, somewhat higher values for emissions from Africa in many models, and typically lesser values for emissions from East Asia, Europe and North America. Variations are roughly a factor of 2 from high to low regions. For sulphate, the response to emission changes in North America is roughly twice the magnitude of the response to emissions from South and East Asia [*Shindell et al.*, 2008a], indicating that regional variations appear to again be substantial. Note that for BC in particular, due to its impact on snow and ice, localized impacts may be highly sensitive to both the location and also the season of emissions. Similarly, sulphur dioxide and short-lived O_3 precursors may have different impacts during different times of year when emitted near polar regions.

Table 5.9 Global annual average TOA all-sky aerosol direct RF efficiency per unit change in emissions from 4 source regions (unit: mWm^{-2} per Tg yr^{-1}) as derived from analysis of SR1 and SR6 runs of six HTAP models. The forcing efficiency for sulphate is calculated with respect to SO_2 emissions, although some models with fully coupled chemistry include changes of sulphate resulting from the reductions of other emissions.

Source Region	Sulphate	POM	BC
NA	-3.6±1.2	-4.4±1.2	42.0±14.7
EU	-3.7±1.3	-4.5±1.5	47.8±16.0
EA	-2.5±1.1	-3.4±1.1	43.4±23.0
SA	-3.2±0.7	-3.7±1.0	37.6±17.5
NA+EU+EA+SA	-3.2±1.1	-3.8±1.1	43.1±19.0

The RF of any forcing can be compared using the GWP (global warming potential), which is defined as the RF from a unit mass pulse emission integrated over a number of years (typically 20 or 100) compared to that of CO_2. Thus the climate impact of any emission control measure for these species can be calculated. For shorter-lived species such as methane (lifetime ~9 years), or aerosols and O_3 precursors (lifetimes ~ 1 week), GWPs can be calculated, but are less easy to interpret. For instance, the GWP_{100} for BC aerosols has been estimated to be 680 [*Bond and Sun*, 2005], or 374 to 677 depending on emission region [*Reddy and Boucher*, 2007] (Figure 5.24). However the timescales that BC and CO_2 act on are so different that the GWP_{100} is less meaningful. The RF from 1 kg BC in the week after its emission is 6 orders of magnitude larger than that of CO_2 but within weeks drops to near zero, while CO_2 continues to exert a forcing for well over 100 years. Shine et al. [2007] introduced the concept of a global temperature potential as a more policy relevant metric. Boucher and Reddy [2007] applied this to BC and calculated a GTP_{100} of ~100, which is up to a factor of 7 lower than the GWP_{100}, since after 100 years much of the climate impact of BC has disappeared. Other methods have similarly been proposed to quantify the relative climate influence of short-lived forcers [e.g., *Bond et al.*, 2010].

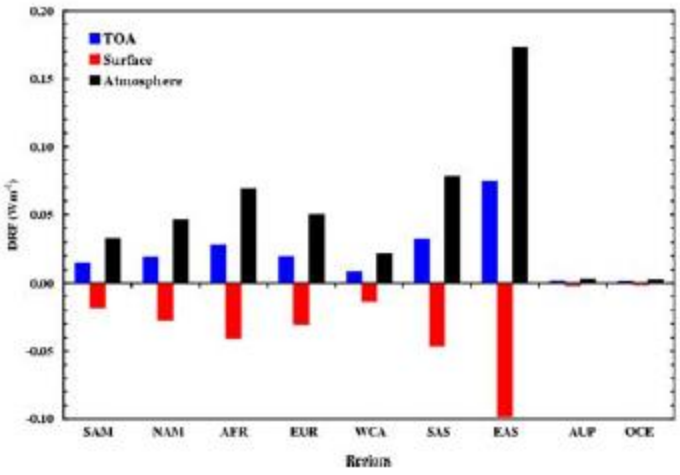


Figure 5.24. Global annual mean all-sky SW DRF at top-of-atmosphere (left bar), at surface (middle bar), and in the atmosphere (right bar) by BC emissions from different regions. [Reprinted from Figure 3 in Reddy, M. S., and O. Boucher (2007), Climate impact of black carbon emitted from energy consumption in the world's regions, *Geophysical Research Letters, 34* (L11802).]

FINDING: Reductions in PM would improve air quality and reduce premature mortality, but reductions in cooling aerosols, including sulphate, nitrate, and organic carbon would generally exacerbate global warming. Reductions in BC are likely to benefit both air quality and climate.

FINDING: The RF resulting from emissions of PM and its precursors is exerted regionally, and the effect on global annual mean RF varies with the location of the emissions.

5.3.3. Relevance of Regional Forcings for Regional and Global Climate

The regional RFs associated with O_3 and PM likely influence climate over a larger area than the forcing itself, as they influence atmospheric heating and dynamics. However, current understanding of regional climate sensitivities to regional forcings is incomplete. Both detection and attribution studies and climate models have examined the geographic patterns of surface temperature response to inhomogeneous RF. Most of these studies, however, have investigated the response to changes occurring over much of the globe (e.g. preindustrial to present-day changes in aerosols). Such studies indicate that at small scales (hundreds to thousands of km) climate responses are typically not closely correlated with the location of RF and instead the spatial pattern of response at these scales appears to broadly match the patterns of response to more homogeneous forcings such as the well-mixed long-lived greenhouse gases (LLGHGs) [*Boer and Yu*, 2003; *Hansen et al.*, 2005; *Levy et al.*, 2008; *Mitchell et al.*, 1995; *Shindell et al.*, 2008b; *Taylor and Penner*, 1994]. These studies also show that at larger scales, the location of RF does influence the response. For example the change in NH temperature exceeds the SH response when the NH RF is greater.

More insight comes from several recent studies. One showed clearly that climate in the Arctic and over the high-latitude Southern Ocean was far more sensitive to non-local influences than climate in most other areas [*Boer and Yu*, 2003]. Chou et al. [2005] indicated that climate response to localized forcing could extend well beyond the forcing location. Another study looked at the effect of regional forcings on global and regional temperatures in a systematic way [*Shindell and Faluvegi*, 2009]. It found that global mean temperatures follow the global mean RF fairly closely, with an enhancement of ~45% for extratropical forcings relative to tropical ones for idealized CO_2 changes [similar to results in *Hansen et al.*, 1997]. This reflects the influence of strong climate feedbacks in higher latitude regions, particularly snow and ice albedo feedbacks. For CO_2, global sensitivity to NH extratropical forcing was ~75% larger than to SH forcing, due to the greater area covered by land and strong positive snow and ice albedo feedbacks. For sulphate, values were similar to those for CO_2 at mid-latitudes, but for high-latitudes the response appears to depend upon aerosol indirect effects: when they were included, the global mean sensitivity was again ~50% greater than for tropical forcing, but when they were not, the sensitivity decreased to only one-quarter of the sensitivity to tropical forcing. This is because the reflective properties of sulphate (the direct effect) have less impact over the bright snow and ice surfaces found at high latitudes.

Shindell and Faluvegi [2009] further demonstrated that tropical mean temperatures follow the global mean forcing fairly closely regardless of where the forcing is imposed, though the response is enhanced (~40%) for local forcings. For other regions, however, the response to non-local forcings is not necessarily simply proportional to either the global mean forcing or the mean forcing within the remote area. The response in extratropical regions decreases as the forcing becomes more remote. These results suggest that the tropics, with its enormous thermal mass, acts as an effective buffer between the two extratropical regions. Hence the uneven distribution of forcing over the Earth's surface has little effect on the global or tropical climate response, but can profoundly affect the extratropical responses. This is especially true in the Arctic (Section 5.3.4), consistent with the earlier results [*Boer and Yu*, 2003]. A new study quantifying the spatial scales over which RF influences climate found that over land areas from 30S-60N, the response extended ~3500 km (30 degrees) in the meridional direction, and at least 12,000 km in the zonal direction, with consistent results across several climate models [*Shindell et al.*, 2010].

For other aspects of climate change, such as precipitation, fewer results are available. Those studies that do exist, however, suggest that at the largest spatial scales, the asymmetric distribution of aerosols between the two hemispheres may affect the location of the intertropical convergence zone

and its associated rainfall [*Chou et al.*, 2005; *Chung and Seinfeld*, 2005; *Ming and Ramaswamy*, 2009; *Rotstayn and Lohmann*, 2002]. Several detailed studies of the Asian monsoon suggest that regional forcing by absorbing aerosols may substantially alter precipitation patterns [e.g., *Chung and Seinfeld*, 2005; *Meehl et al.*, 2006; *Ramanathan and Carmichael*, 2008; *Wang et al.*, 2009].

FINDING: Ozone and PM exert radiative forcings that are not globally uniform, but extend from the location of (precursor) emissions over regional and intercontinental scales. These heterogeneous forcings affect global climate change, and can influence patterns of regional climate change including temperature and precipitation. In general, high latitudes are more sensitive to RF, as a result of snow/ice feedbacks.

5.3.4 Impacts of Long-Range Transport on Climate in the Arctic

The Arctic is experiencing rapid climate change, as global warming is amplified in this region due to the large albedo feedbacks associated with changes in snow and ice cover. O_3 and PM have particular RF effects in the Arctic due to the high surface albedo and low atmospheric moisture content, which increases aerosol lifetimes. In addition to the forcings from O_3 and PM directly in the Arctic region, changes in circulation driven by forcings outside of the Arctic are important to Arctic climate. Because emissions of air pollutants within the Arctic itself are low, it is clear that emissions from outside of the Arctic dominate Arctic climate changes, including LRT to the Arctic. Air pollution and climate change in the Arctic are significant concerns for possible increased human activities, including shipping, in the region [e.g., *Granier et al.*, 2006].

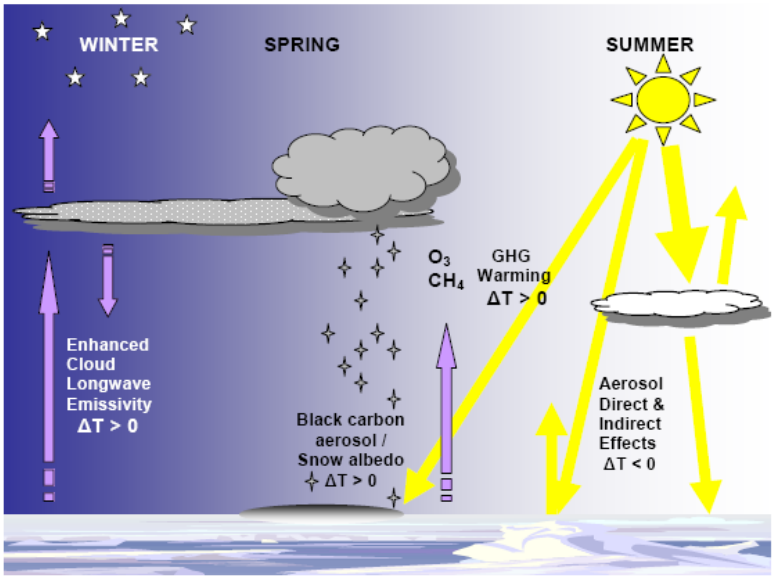


Figure 5.25. Forcing mechanisms in the Arctic environment resulting from the poleward transport of mid-latitude gas and particulate phase pollutants. The season of maximum forcing at the surface (F_S) is indicated for each forcing agent. ΔT indicates the surface temperature response. [Reprinted from Figure 1 in Quinn, P. K., et al. (2008), Short-lived pollutants in the Arctic: Their climate impact and possible mitigation strategies, *Atmospheric Chemistry and Physics*, *8*(6): 1723-1735.]

A summary of the radiative effects of various pollutants on the Arctic is given by Quinn et al. [2008], and shown schematically in Figure 5.25. An estimate of the anthropogenic temperature change in the Arctic (Figure 5.26) shows that reflective aerosols, BC, and ozone have influences that are comparable to the warming from CO_2. BC aerosol from fossil fuel or fire plumes has strong shortwave absorbance and can therefore warm the Arctic troposphere and stratosphere as well as reduce the net irradiance at the surface, especially above a high-albedo surface such as snow. The magnitude of this forcing, however, is highly variable in space and is not well-constrained [*Baumgardner et al.*, 2004]. The indirect RF associated with deposition of BC in Arctic snow (darkening) is receiving increased attention as ice-core and direct observational evidence suggest that soot levels, which had declined

through the late 20th century, may now be rising again [*Eleftheriadis et al.*, 2009; *McConnell et al.*, 2007; *Sharma et al.*, 2006]. Very small quantities of BC can significantly reduce the albedo of snow, thereby increasing snow aging and melt rates and further enhancing surface radiative warming through a positive feedback. It is thought that this mechanism may be contributing to accelerated glacier melting and sea ice cover reduction since the late 1990s [e.g., *Kim et al.*, 2005; *Qian et al.*, 2009]. As a result, the climate forcing efficacy due to BC in snow is estimated to be substantially larger than that of CO_2 [*Hansen and Nazarenko*, 2004]. Flanner *et al.* [2007] estimated that the anthropogenic contribution to the overall RF associated with BC deposition in the Arctic is in excess of 80%, and the resulting warming in the Arctic in the 20th century may have been as large as ~0.1-0.2 °C [*Flanner et al.*, 2007; *Hansen and Nazarenko*, 2004; *Koch et al.*, 2009].

High concentrations of pollution-derived sulphate aerosols may also impact Arctic climate by modifying the size distribution of ice nuclei in low-level clouds during the cold season, resulting in a reduced infrared radiation flux reaching the surface [*Girard et al.*, 2005]. Kristjansson et al. [2005] point out that the response of the Arctic climate system to direct and indirect effects of aerosols may also manifest itself in circulation anomalies induced by changes in sea-level pressure, but recent climatological observations have yet to validate these predictions. Model simulations and recent observations at Barrow, Alaska, indicate that the interaction of haze and thin water clouds in spring or summer can lead to a local surface temperature warming of +1° to 1.6°C, due an increase in longwave emissivity from these clouds [*Garrett and Zhao*, 2006; *Girard et al.*, 2005]. The combined effect of Arctic Haze (a combination of reflective and absorbing aerosols) and biomass burning aerosols is an estimated surface cooling of −0.93°C [*Isaksen*, 2009; *Quinn et al.*, 2008].

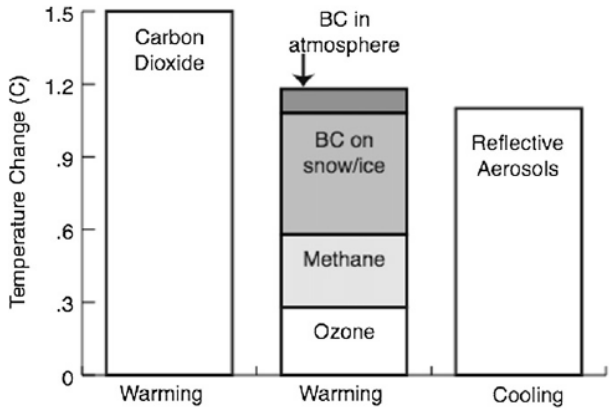


Figure 5.26. Estimates of the contribution of particular species to preindustrial to present-day Arctic (60° to 90° N) surface temperature trends. Values are based on the assessment of modelling and observations of Quinn et al., and do not include aerosol indirect effects. Reflective aerosols include sulphate and organic carbon. [Reprinted from Figure 41 of Isaksen, I. S. A., et al. (2009), Atmospheric composition change: Climate-chemistry interactions, *Atmospheric Environment, 43*(33): 5138-5192, with permission from Elsevier.]

The strong influence of meridional heat transport into the Arctic also leads to substantial climate sensitivity to forcing outside the Arctic [*Boer and Yu*, 2003; *Shindell and Faluvegi*, 2009]. Additionally, both O_3 and BC aerosols absorb solar radiation and thus heat the atmosphere, altering meridional circulation in ways that forcing from greenhouse gases or sulphate does not. Thus, for forcing in the Arctic, the climate response per unit tropopause or surface RF can be quite different for short-lived species than the global mean response. Surface temperatures in the Arctic are strongly influenced by both local forcing and forcing at Northern Hemisphere mid-latitudes (Chapter A4). As the latter is typically much greater, it can often play a more important role in driving Arctic climate changes for regionally inhomogeneous forcing [*Shindell and Faluvegi*, 2009].

Shindell and Faluvegi [2009] estimate that between 1976 and 2007, aerosols may have contributed ~1.09 °C to the observed Arctic surface temperature increase of 1.48 °C. In contrast, tropospheric O_3 increases since preindustrial times are estimated to be responsible for about 0.3°C

annual average and about 0.4-0.5°C during winter and spring to the 20th-century Arctic warming [*Shindell et al.*, 2006].

FINDING: The Arctic is experiencing rapid climate change. Arctic climate is affected by ozone and PM that are the result of long-range transport from other regions, as well as by ozone and PM outside of the Arctic. Deposition of black carbon in snow is an important contributor to warming in the Arctic.

5.3.5 Future Changes in Forcings

The future forcing from short-lived pollutants is very scenario dependent since (unlike for CO_2) control measures have a very rapid impact on atmospheric concentrations. Stevenson et al. [2006] analysed three possible future scenarios for O_3 precursors and found that the forcing from O_3 between 2000 and 2030 varied from -45 to 63 to 155 mWm^{-2}. The three scenarios were IIASA MFR (maximum feasible reduction), IIASA CLE (current legislation) and SRES A2 (unrestricted growth). The methane forcing over the same period varied from 0 to 116 to 141 mWm^{-2} for these three scenarios. Note the methane forcings were from an earlier transient run [*Dentener et al.*, 2005] and included the impacts of the non-methane O_3 precursors on the methane lifetime. The forcings compare with values for CO_2 - ranging from around 800 (SRES B2) to 1,050 (SRES A2) mWm^{-2} over the 2000-2030 period [*Stevenson et al.*, 2006]. Although the absolute numbers are higher for CO_2, the range in the combined forcing of methane and ozone was 341mWm^{-2} from cleanest to dirtiest. This compares to a range of 250mWm^{-2} for the two CO_2 scenarios. This suggests that controls on short-lived pollutants could be at least as effective for climate mitigation in the short term as controls on CO_2. Controls on particulate matter were not considered by Stevenson et al. [2006], but are also expected to have a large short term climate impact.

A new set of scenarios (RCPs) have been produced and reflect options to mitigate long term climate change [*Moss et al.*]. First results from these scenarios are presented in chapter A4.

The impact of climate change on the Stevenson et al. [2006] forcings was to mitigate the RF of methane by 26 mWm^{-2}, since methane is removed more efficiently in a warmer climate. The impact of climate change on the O_3 forcing was much more uncertain. Those models that simulated an increase in stratosphere-troposphere exchange [*Collins et al.*, 2003; *Sudo et al.*, 2003; *Zeng and Pyle*, 2003] with climate predicted a larger O_3 forcing in future. Those that did not predicted a lower O_3 forcing due to more efficient removal of O_3 [*Johnson et al.*, 2001]. These studies did not explicitly look at the impact of climate change on the response to emissions, but from the signs of the changes reported (more rapid removal of methane and O_3, greater stratosphere-troposphere exchange), we might expect the forcing attributable to emissions changes to decrease in the future.

Future climate is likely to increase overall precipitation and hence increase aerosol removal rates in many regions. However it will also increase the rate of oxidation of SO_2 to sulphate. Decreases in snow cover will make aerosol forcings less positive (more negative). Changes in clouds due to the combination of climate change and changes in PM may be very important.

5.3.6. Future Research Needs

Estimates of the RFs due to O_3 and PM are among the most important uncertainties in our current understanding of climate change. For both O_3 and PM, there is a need to understand the RF for present day concentrations relative to concentrations in the preindustrial period, for which there are limited observations. In addition, the possible effects of PM on cloud formation and lifetime pose particularly strong uncertainties. Substantial reduction in the aerosol RF uncertainty estimates requires continued, focused efforts on acquiring better observations, improving model representations of atmospheric processes, and developing a synergistic strategy [*U.S. CCSP*, 2009].

RECOMMENDATION: The radiative forcings due to ozone and PM are highly uncertain, and are among the most important uncertainties in our understanding of climate change. Observations of aerosol absorption, vertical distributions, and cloud properties (the indirect effect) are particularly needed to reduce these uncertainties.

RECOMMENDATION: The forcings resulting from changes in emissions of aerosols, their precursors, and ozone precursors (except methane) depend strongly on location, timing, and the background composition, and these dependencies merit further research.

There are also several research needs which are not particular to improving understanding of the climate effects of O_3 and PM, but would be shared with the health and ecosystems communities, and observational capabilities should be improved and designed with these different endpoints in mind.

RECOMMENDATION: Deficiencies in current observations of air pollutants limit the ability to understand effects on health, ecosystems and climate in large regions of the world. Improvements to observations through ground-based and satellite measurements, and in emissions and models, can be important for improving understanding of the health, ecosystem and climate effects of air pollutants. Improvements to these systems should be made with health, ecosystem and climate effects in mind.

RECOMMENDATION: Assessment of impacts of intercontinental transport of air pollutants on human health, ecosystem, and climate relies on global models with coarse resolution. There is a pressing need to establish modelling systems that link the local, regional, intercontinental, and global scales.

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Chapter 6
Summary

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6.1. Observational Evidence for Intercontinental Transport

Observations from the ground, aircraft, and satellites provide a wealth of evidence that ozone (O_3) and particulate matter (PM) concentrations throughout the Northern Hemisphere are influenced by intercontinental and hemispheric transport of pollutants. Transport in the Northern Hemisphere mid-latitudes is dominated by the westerly winds that transport emissions from East Asia across the North Pacific Ocean to North America, from North America across the North Atlantic Ocean to Europe, and from Europe into the Arctic and central Asia. The signature of long-range pollutant transport in measurements made downwind of sources depends on the lifetime of the particular pollutant. Discrete plumes of enhanced concentrations characterize pollutants with short lifetimes and no photochemical sources, while continuous distributions of concentrations represent transport of pollutants with longer lifetimes. In the latter case, the entire troposphere can be envisioned as completely filled with plumes of continuously varying concentrations in the process of intermixing and dispersing.

For PM, important evidence of transport is provided by satellite imagery of visible plumes of smoke and dust from wildfires and deserts that extend for thousands of kilometres. Over the last few years satellites have begun to provide quantitative information on intercontinental aerosol transport, including estimates of the amount of pollution transported, the altitude of transport and, in some cases, aerosol properties. Ground-based lidar networks and mountain top measurement sites in Europe, North America and Asia provide large continuous data sets that characterize the frequency of occurrence of aerosol transport events, the meteorological conditions responsible for them, and important information on aerosol particle properties. Evidence of intercontinental transport is also provided in the form of long-term trends in surface-site observations from remote islands, which in some cases compare well with the trends in emissions in upwind areas. Some in situ measurements have illuminated the importance of secondary aerosol formation from transported precursors. Long-range transport of aerosols can have significant air quality and environmental implications, especially in the outflow of the Asian and African continents and in the Arctic regions. Observations of the trace element composition and stable isotope ratios of aerosols have been useful in assessing the importance of natural and anthropogenic sources and studying processing of aerosols in transit.

For O_3, the evidence is provided in plumes of elevated O_3 observed in the free troposphere or at high elevation sites and, more importantly, in an increasing trend in baseline O_3 concentrations measured consistently at a number of remote sites across the Northern Hemisphere. Measurements suggest that during the latter half of the 20th century, concentrations of O_3 at northern mid-latitudes increased by a factor of two or more. It is likely that much of this increase is due to increases in anthropogenic emissions of O_3 precursors. Within the limits of the measurement records, the increase has been comparable throughout all longitudes, and has occurred in all seasons. More recently, more rapid increases have been observed downwind of eastern Asia in the free troposphere, whereas the increases within the boundary layer of central Europe and North America have slowed down. Measurements at some locations on the western coasts of Europe and North America clearly show that trans-oceanic air flows can carry O_3 concentrations that approach or exceed air quality standards and objectives, and that air can mix to the surface and contribute substantially to air quality standard violations. This is particularly noticeable in areas with low emission. The impact on surface air quality depends upon vertical mixing of air into the boundary layer, which is enhanced by complex topography.

Although the observational record is sufficient to demonstrate that intercontinental transport occurs, to understand the contribution of the intercontinental transport of these pollutants to air pollution impacts and the significance of these flows for air quality management now and in the

future, the observational evidence must be combined with quantitative models that describe the processes of emission, transport, transformation, and removal that drive the observed concentrations and deposition.

6.2. Modelling Analyses of Intercontinental Transport

Current global models of O_3 and PM reproduce much of the observed regional and seasonal variability in surface concentrations and deposition relatively well and have been used to predict the path of pollutants during individual intercontinental transport events. This ability gives us some confidence that we can quantitatively represent the key processes controlling the formation, transport and removal of O_3, PM, and their precursors. However, on finer spatial and temporal scales, significant discrepancies exist among estimates from different models and between model estimates and observations, indicating weaknesses in our representation of many processes at the resolution of current models. Furthermore, current global models are not able to reproduce entirely historical measurements and observed trends, raising concern about our ability to predict changes in the future.

Even with these limitations, current models provide useful insights regarding what emissions sources contribute to observed concentrations and how observed concentrations will change with emissions changes. Using various source attribution techniques, current models can estimate the contribution that different precursors, emissions source categories, or source regions make to observed concentrations or deposition. Source-receptor sensitivity analyses can be used to estimate how each of these fractions will change with changes in emissions or other factors.

The Task Force on Hemispheric Transport of Air Pollution (TF HTAP) coordinated several sets of multi-model experiments to better understand the ability of current models to describe intercontinental transport. As part of this effort, a set of emission perturbation experiments were conducted to compare model estimates of how emission changes in one region of the world impact air quality in other regions of the world. Multiple models were used to examine the global impacts of 20% emission reductions of relevant anthropogenic pollutants in four regions, approximately covering the major populated areas of North America, Europe, South Asia, and East Asia. These names are used hereinafter to refer to these rectangular regions, which encompass more than 75% of the anthropogenic emission sources in the Northern Hemisphere and, in some cases, include significant areas of ocean. Specific analyses were also made to quantify the impact in the Arctic of emission changes in these four source regions. These simulations provide information about source-receptor sensitivities as well as some information about source attribution.

6.2.1. Source Attribution

Attributing tropospheric O_3 back to the emission sources that contribute to its formation is complicated by the fact that O_3 is not emitted, but is formed in the atmosphere through a non-linear system of photochemical reactions of emitted precursors, including nitrogen oxides (NO_X), volatile organic compounds (VOC), carbon monoxide (CO), and methane (CH_4). Conceptually, observed surface O_3 concentrations may be thought of as composed of four fractions:

- O_3 that is formed in the stratosphere
- O_3 that is formed in the troposphere from
    - natural precursor emissions, including lightning, soil, fire, and vegetation emissions
    - anthropogenic precursor emissions that have been transported on intercontinental scales
    - anthropogenic precursor emissions from local or regional sources

The O_3 precursors from different sources interact in the atmosphere such that O_3 may be produced from natural or anthropogenic, local or transported, or a combination of natural and anthropogenic or local and transported emissions.

Each of the fractions that comprise observed O_3 differs in terms of (1) sensitivity to changes in precursor emissions, (2) the extent to which they can be controlled by a national or sub-national

jurisdiction, and (3) the magnitude of the contribution. The relative contribution of each fraction varies widely by location and season and has evolved over time. We have confidence that human activities contribute to the observed positive trend in surface O_3, and that the anthropogenic component to mean O_3 levels in the Northern Hemisphere has grown significantly since about 1850. However, there remain large uncertainties in our estimates of the source attribution for these changes.

Based on modelling performed for the HTAP multi-model experiments, the annual average ground-level O_3 mixing ratio averaged across the four study regions (North America, Europe, South Asia, and East Asia) and the ensemble of participating models is about 37 parts per billion by volume (ppbv)(± 4 ppbv standard deviation). This annual average, region-wide O_3 concentration masks large seasonal and geographic variability, and large differences across models. However, it provides a useful point of reference for considering the magnitude of intercontinental transport. Based on estimates in the literature, 20%-25% of this annual average ground-level concentration originates from the stratosphere, and a similar fraction is formed from natural precursor sources. The remainder is due to anthropogenic sources of precursors from within the region itself and transported from outside the region. The relative contribution of anthropogenic and natural, regional and extra-regional sources varies by location, season, and year.

Models indicate that in the northern mid-latitudes, intercontinental transport of air pollution typically peaks in boreal spring and fall, and is smallest during the summer months when O_3 levels are highest, due to the peak in production from local and regional emissions. Over the South Asia region, both the local and regional and the intercontinental influences are largest in late fall through winter and early spring. Intercontinental contributions to surface O_3 in the Arctic are largest in April through June, with a secondary maximum in October and November.

The maximum influence of intercontinental transport on ground-level O_3 generally occurs on mid-range pollution days (i.e., near the middle of the O_3 probability distribution, typically 50-70 ppbv), suggesting that import is less significant on days with the highest O_3 levels. These high O_3 conditions are typically due to trapping of local precursor emissions under stagnant meteorological conditions which also suppress the influence of distant sources. These results indicate that decreasing local or regional emissions is more effective at decreasing the highest O_3 levels, but that O_3 associated with stratospheric origins, natural emissions sources, and intercontinental transport comprise a significant fraction of tropospheric O_3 that is not within the control of local and regional political jurisdictions.

As with O_3, PM concentrations or deposition can also be apportioned into several fractions based on emission sources. These PM fractions are associated with emissions from:

- volcanic eruptions, vegetation, and wind-blown dust (the last of which can be exacerbated by anthropogenic factors)
- open biomass (vegetation) burning, some of which is natural and some of which may be anthropogenic
- anthropogenic emissions that have been transported on intercontinental scales
- local and regional anthropogenic sources

The relative magnitudes of these fractions and their sensitivity to emission changes differs by location, season, year, and chemical component. Primary particles such as dust and black carbon, which are directly emitted into the atmosphere, respond linearly to changes in their emission sources. Secondary particles such as sulphate and organic aerosols, have a slight non-linear response due to their dependence on oxidation and other reactions. The fractions also differ to the extent to which they can be controlled by a local or regional jurisdiction.

There is a wide range of surface aerosol concentrations predicted by current models, reflecting large uncertainties in emissions and atmospheric processes as represented in the models. The HTAP multi-model experiments suggest that, in the four regions studied, ground-level concentrations of wind-blown soil dust from deserts in Africa, Asia, and the Middle East can be a

factor of 1.5 to almost 20 higher than that from anthropogenic and open biomass burning sources on a region-wide, annual average basis. Of the ground-level PM concentration originating from anthropogenic and open biomass burning sources, intercontinental transport of anthropogenic emissions accounts for between 5% and 35% and transport from open biomass burning contributes between 4% and 14% on a region-wide, annual-average basis in each region.

In terms of the chemical components of PM, the HTAP multi-model experiments provided information for four components: sulphate, black carbon, particulate organic matter (POM), and mineral (or soil) dust. For North America, Europe, South Asia, and East Asia, anthropogenic sources of emissions within each region account for 60-90% of the sulphate and black carbon concentrations. For POM, the contribution of anthropogenic sources within the region is less than for sulphate and black carbon, with biomass burning and biogenic emissions also contributing to the estimated concentrations.

In the Arctic, European pollution is the largest contributor to surface sulphate, followed by volcanic emissions. Biomass burning in the boreal forests of Eurasia plays a major role in determining the surface concentrations of black carbon and POM in the Arctic. Nearly half of the mineral dust at the Arctic surface is from Asia, with smaller amounts from Africa and the Middle East.

The fraction of PM associated with intercontinental transport increases with altitude, indicating the importance of transport above the boundary layer. As a result, the contribution of intercontinental transport to total column loading tends to be 15 to 25% higher than the contribution to surface concentrations or deposition.

As is the case with O_3, annual average, region-wide statistics mask significant variability between seasons and within a given region. The seasonal cycles of aerosol concentration and intercontinental transport vary by chemical component and by region. However, there are substantial differences between the seasonal cycles in surface concentration predicted by models participating in the HTAP multi-model experiments. The model differences are larger for dust than they are for sulphate, black carbon, and POM, and are larger for the Arctic than for the mid-latitude regions.

In many regions, deposition of oxidised sulphur and nitrogen is large, and exceedance of critical loads occurs widely over many ecosystems. The intercontinental contribution to these exceedances is small. However, particular attention should be paid to the effects of long-range transport to polar regions where ecosystems are likely to be more vulnerable.

6.2.2. Source-Receptor Sensitivity

As part of the HTAP multi-model experiments, a set of emission perturbation experiments were conducted to compare model estimates of how emission changes in one region of the world impact air quality in other regions of the world. These experiments have provided the first set of comparable estimates of intercontinental source-receptor relationships from multiple models.

In the emission perturbation experiments, multiple models were used to examine the global impacts of 20% emission reductions of relevant anthropogenic pollutants in the four study regions.

For O_3, the impact of 20% changes in anthropogenic emissions of NO_X, VOC, CO, sulphur dioxide (SO_2), and direct PM in one region on surface O_3 in the other regions varies from 0.07 to 0.37 ppbv on an annual average, region-wide basis, as estimated by the mean of the model ensemble. These values are significant in comparison to the response of surface $O_3$to 20% decreases of emissions within the region itself, which vary from 0.8 to 1.3 ppbv. These annual average, region-wide values mask large temporal and geographic variability. For example, the values can vary by up to a factor of two from season to season and from grid cell to grid cell within a region.

The largest source-receptor relationship, in an absolute sense, is the impact of North American emissions on European surface O_3 levels. This is followed by the impact of European emissions on South Asian and East Asian surface O_3. The annual average impact of East Asian emissions on North American surface O_3 is similar to the impact of North American emissions on East Asian surface O_3, but with peaks in different seasons. European emissions have the largest influence on Arctic surface O_3 followed by North American emissions.

To quantify the relative importance of emissions changes outside each of these regions, as compared to emissions changes inside each of these regions, we defined the Relative Annual Intercontinental Response (RAIR) metric. RAIR is defined as the sum of the changes in the annual average, regionally-averaged concentration within a region due to a 20% decrease in emissions in the three other regions, divided by the sum of the changes in concentration within a region due to a 20% decrease in emissions in all four regions. The value of the metric ranges from 0%, indicating no intercontinental influence, up to 100%, indicating that air quality in a region is completely dominated by intercontinental sources. Thus, the RAIR is a measure of how much benefit a region may receive from emission reductions in other regions when emission reductions are coordinated on an intercontinental scale.

The RAIRs for O_3 estimated by the HTAP multi-model experiments range from 43% for Europe, to 40% for East Asia, to 32% for South Asia and North America. These values suggest that, in all four of the source-receptor regions, at least 30% of the total concentration changes within each of the regions is related to emission changes in the other three regions. Substantial O_3 transport takes place above the boundary layer in the free troposphere, where it can be brought to lower altitudes over distant receptors during subsidence, and mix with local emissions. The RAIRs are larger for column O_3 than for ground-level O_3, and the column RAIRs exceed 50% in Europe and East Asia.

Imported O_3 and precursors may have qualitatively different impacts on urban areas than rural areas due to the strong dependency of O_3 chemistry on the ratio of NO_X to VOCs. Results from the global models used in the HTAP multi-model experiments suggest that the response of annual average O_3 concentrations in large cities to changes in intercontinental transport may be as large or larger than the response to changes in regional emissions. Global models are not well suited to estimate responses at urban scales, as the resolution of current models is too coarse to resolve the strong chemical contrasts associated with urban regions. New studies using regional air quality models show that the higher resolution models are better able to resolve local topography, finer variations in land cover and use, and O_3-VOC-NO_X chemical non-linearities, which all have effects on the strength of surface O_3 responses to emission changes. For example in East Asia, results using a regional model predicted a smaller mean response in $O_3$from European sources, but with much larger spatial variability.

The HTAP multi-model experiments also examined the sensitivity of O_3 to changes in CH_4 by decreasing the globally fixed CH_4 concentration by 20%. From those simulations, we estimated the response to 20% decreases in regional anthropogenic CH_4 emissions. While local and regional emission controls are clearly most effective for lowering local and regional O_3, the O_3 response to anthropogenic emissions of CH_4 from distant source regions is nearly as large as that to emissions of the traditional O_3 precursors in these regions. The O_3 response to changes in CH_4 emissions requires several decades to be fully realized, given the relatively long atmospheric lifetime of CH_4. Thus, controlling CH_4 is an important component of a strategy to limit increases in baseline surface O_3, and as an important greenhouse gas, has additional benefits for climate change mitigation.

For PM, the HTAP multi-model experiments estimate RAIRs for PM surface concentrations of 20% for South Asia, 9% for East Asia, 7% for North America, and 5% for Europe. The RAIRs for surface deposition of sulphate, reactive nitrogen, black carbon and POM are similar to those for surface concentrations. The RAIRs for aerosol column loadings are generally larger (24%-37% for sulphate, 15%-24% for black carbon, and 12% to 23% for POM), reflecting the importance of transport above the boundary layer and implying more significant contributions to visibility and radiative forcing impacts.

Ground-level PM concentrations or deposition in the Arctic are most sensitive to emission changes in Europe. However, total column loadings of particulate matter over the Arctic are equally sensitive to changes in emissions from Europe or Asia, due to the fact that Asian emissions have a stronger tendency to be lifted and transported aloft than do European emissions.

Ground-level PM concentrations generally respond linearly to changing emissions from both local and upwind source regions. However, sulphate and some fraction of particulate organic matter are not directly emitted, but are formed in the atmosphere through oxidation, and can be affected by

non-linear systems of chemical reactions. For example, present-day North American emissions of NO_X and CO, together, are estimated to increase surface PM in Europe and East Asia by up to 0.5 $\mu g/m^3$. These impacts are not due to direct transport of secondary PM produced in the source region and then transported to other continents, which is negligibly small. These increases are due to increases in oxidants, leading to enhanced production rates of secondary aerosols during long-range transport and in the receptor region. These results underscore the multifaceted nature of the consequences of a rising background of pollution levels in the Northern Hemisphere on local pollution levels.

6.3. The Impacts of Intercontinental Transport

O_3 and PM pollution are serious public health and environmental problems in many parts of the world. Ample experimental and epidemiological evidence indicates that exposure to ambient PM and O_3 concentrations cause adverse health effects that range from minor sensory irritation to premature death. O_3 also causes damage to a variety of different ecosystems including crops, forests and grasslands, which are also damaged by PM through the processes of acidification. These ecosystem impacts have important implications for productivity, biodiversity, and food security. O_3 and PM pollution also significantly contribute to climate change on regional and global scales. The intercontinental transport of O_3 and PM contributes to each of these adverse impacts.

As explained above, the highest concentrations of O_3 and PM are typically associated with stagnant conditions, when the contribution from intercontinental transport is low and the contribution of local and regional sources are most important. However, intercontinental transport has increased baseline O_3 concentrations to the point where they exceed thresholds for protection of vegetation in many locations and exceed thresholds for the protection of human health occasionally in some locations. Intercontinental transport events associated with forest fires or dust storms produce exceedances of short-term PM public health standards (e.g., the impact of Saharan dust events on PM levels in Southern Europe). On a longer-term basis, current levels of intercontinental transport of PM interfere with the ability to meet natural visibility targets in western North America. If public health-based air quality standards continue to be tightened based on new health effects research, the contribution of intercontinental transport to exceedances of such standards will continue to increase.

Relatively few studies have tried to quantify the human health impacts of intercontinental transport of O_3 and PM specifically. Those studies that have been conducted have focused on the impact on the relationship between annual average concentrations and premature mortality. These studies suggest that intercontinental transport can contribute significantly to health impacts of air pollution within a given receptor region. For O_3, one study based on the HTAP multi-model experiments estimated that intercontinental transport of O_3 contributes from 20% to more than 50% of O_3-related premature adult mortalities in a given receptor region, subject to large uncertainty. For PM, contributions to PM from emissions within any given region are expected to be much more important for human health than emissions from intercontinental transport. Based on the HTAP multi-model experiments, intercontinental transport of PM may be responsible for 3% to 5% of the PM-related mortalities in a region.

The sum of the health impacts of transported pollution in downwind foreign regions can be larger than the health impacts of emissions in the source region itself. Although the impact on ambient concentrations in downwind foreign regions may be much less than in the source region itself, the total population exposed in those downwind regions is much greater. For O_3, three studies have suggested that emission reductions in North America and Europe will avoid more mortalities outside these source regions than within the regions themselves. For PM, one study based on the HTAP multi-model experiments has suggested that 15% and 12% of the total PM-related mortalities associated with emissions from North America and Europe, respectively, are estimated to be realized outside of these source regions.

Several previous studies have found that global and intercontinental transport of sulphur and nitrogen contribute to the acidification and eutrophication of natural ecosystems with consequences for productivity and biodiversity. For O_3, recent experimental studies on field crops, adult forest

stands and different grassland ecosystems have found significant impacts associated with ecologically realistic free-air O_3 fumigations that mimic the observed increases in background O_3 concentrations.

Global crop yield losses of four staple crops due to exposure to O_3 are estimated to be between 3% and 16%, depending on the crop, and are valued at $14 billion - $26 billion per year. Based on the HTAP multi-model experiments, intercontinental transport may be responsible for about 5% to 35% of the estimated crop yield losses depending on the location, crop, and response function used. However, there is significant uncertainty in these estimates, part of which is due to the limited representativeness of available exposure-response functions based on threshold indices (e.g. AOT40 and SUM06).

On a global basis, O_3 and PM are significant contributors to climate forcing. Intercontinental transport influences the distributions of O_3 and PM, and therefore the shape and magnitude of their forcings. O_3 is a greenhouse gas that causes warming directly, and damage to plants by O_3 inhibits the natural uptake of CO_2. PM is a mixture containing components that mainly cool, including sulphate and organic aerosols, and black carbon that warms, while aerosols also influence climate by changing clouds. Anthropogenic emissions of black carbon, CH_4, carbon monoxide, and non-methane volatile organic compounds (which are O_3 precursors) are estimated to have caused a climate forcing since 1750 roughly as large as that from anthropogenic CO_2. Because these compounds (and O_3) have short lifetimes compared to the long-lived greenhouse gases, reducing these emissions will both improve air quality and slow the rate of climate change in the near term.

The climate forcings resulting from changes in emissions of PM, its precursors, and O_3 precursors (except CH_4) depend strongly on location, timing, and the background composition. The radiative forcings and climate effects exerted by O_3 and PM are not globally uniform, but extend from the location of precursor emissions over regional and intercontinental scales. Based on the HTAP multi-model experiments, the Relative Annual Intercontinental Response (RAIR) of aerosol optical depth and direct radiative forcing was calculated for each of the four continental regions and each PM component. For all regions and PM components, emission changes outside the region had a significant effect relative to emission changes within the region, with RAIR of radiative forcing ranging from 9% to 30%. The change in radiative forcing over South Asia is most influenced by the import of sulphate aerosol, and North America is most influenced by the import of black carbon. For the change in radiative forcing due to sulphate, black carbon, and POM combined, South Asia is the region most strongly influenced by foreign sources, and North America and Europe are the least. The variability between models is very large, highlighting the significant uncertainties in modelling aerosol processes and transport.

This inhomogeneous forcing affects climate change at the global scale and at the regional scale, influencing atmospheric heating and dynamics and ultimately patterns of temperature and precipitation. The largest climatic impacts do not necessarily occur where the radiative forcing occurs and may occur downwind of the source region.

The Arctic is experiencing rapid climate change. Arctic climate is affected by O_3 and aerosols that are transported into the Arctic from other regions, as well as by the climate forcing of O_3 and aerosols outside of the Arctic. Deposition of black carbon on snow and ice is understood to be an important positive (warming) forcing in the Arctic.

Among O_3 precursors, widespread decreases in emissions of CH_4, CO, and VOCs will likely reduce climate warming. Decreasing NO_x may increase climate warming over decadal time scales because less NO_x leads to less hydroxyl radical, increasing the lifetime of CH_4. The increase in radiative forcing from the increased lifetime of CH_4 is greater than the decrease in radiative forcing from decreased O_3 formation. Reductions in PM would improve air quality, but for cooling aerosols, including sulphate, nitrate and organic carbon, this would generally exacerbate global warming. Reductions in black carbon would typically benefit both air quality and climate.

6.4. Future Scenarios

The significance of intercontinental transport for the achievement of environmental policy objectives may change in the future due to changes in the magnitude and spatial distribution of emissions. These changes may be caused by the continuing implementation of pollution control measures, regional differences in the pace of economic development, the growth in shipping and aviation emissions, and the implementation of climate change mitigation measures. In addition, changes in transport patterns, chemical evolution, depositional losses in transit, and emissions sources due to climate change may change the magnitude of intercontinental transport.

The current levels of intercontinental transport and hemispheric baseline concentrations of O_3 and PM are a result of emissions that, on a global basis, increased rapidly between 1950 and 1990. Since 1990, global emissions leading to O_3 and PM concentrations have experienced little change or have begun to decrease. In Europe and North America, which have been the dominant sources of anthropogenic emissions until recent decades, emissions of most precursors are constant or declining, due to the implementation of air pollution control policies. In East Asia and South Asia, emissions of precursors have risen dramatically in recent years, due to economic growth and development in these regions.

The implications of changes in anthropogenic emissions were explored in the HTAP multi-model experiments by considering a set of global emission scenarios developed to inform the Intergovernmental Panel on Climate Change's fifth assessment report, known as the Representative Concentration Pathways (RCPs). Three of the four scenarios assume some climate change mitigation policy will be adopted, and all four assume that the implementation of air pollution control policies will increase as development and income increase. As a result, all four of the scenarios suggest that between now and 2050, global emissions of most O_3 and PM precursors will decline, up to 76% for SO_2 emissions. However, the regional distribution of emissions in the Northern Hemisphere is expected to shift, with steeper and earlier declines in Europe and North America and shallower declines or actual increases in South and East Asia. Under the lowest emissions scenario, NO_x emissions between 2000 and 2050 decline by 78%, 63%, and 48% in North America, Europe, and East Asia, respectively, but increase in South Asia by 42%. Under the highest emissions scenario, NO_x emissions peak in 2030 with decreases of 43% and 16% in North America and Europe, respectively, and increases of 65% and 91% in East Asia and South Asia, respectively.

As part of the HTAP multi-model experiments, the impact of this redistribution of future emissions and expected changes in future global CH_4 concentrations, as specified by the RCP scenarios, was explored using linear approximations of the intercontinental source-receptor sensitivities determined under current conditions. To illustrate the range of future levels of air pollution, we assessed the RAIRs for 2050 under the lowest emissions scenario and for 2030, when global emissions peak, under the highest emissions scenario. For North America ground-level O_3 concentrations, the RAIR is estimated to increase to around 50% under both the high and low emissions scenarios, suggesting that, in the future, changes in emissions of O_3 precursors outside the region may be as important as changes within the region. For Europe, the RAIR for ground-level O_3 increases relatively little under the highest and lowest scenarios, due to simultaneously declining air pollution emissions in North America. For East Asia, the RAIR decreases under a high emission scenario, under which emissions within the region increase, and increases under a low emission scenario, under which emissions in the region decline. For South Asia, the RAIR for ground-level O_3 declines under both the low and high scenarios. Note that the sensitivity to changes in geographical distribution of emissions within a particular region has not been adequately assessed yet.

Using the linear approximation, the influence of changing CH_4 concentrations can be separated from the influence of changing intercontinental transport and local and regional emissions over the historical emission trends and future emission scenarios. Roughly 40% of the O_3 increase since the preindustrial period is believed to be due to anthropogenic CH_4. By 2050, the RCP scenarios suggest a wide range of possible changes in CH_4 concentrations, ranging from an increase greater than 50% to a decrease greater than 15%. Under the highest scenario, expected increases in CH_4

concentrations have a large influence on ground-level O_3 changes, in some cases offsetting significant decreases in O_3 formation associated with local and regional emissions.

The HTAP multi-model experiments also examined the potential impacts of changes in meteorology and transport patterns expected as a result of climate change. Future changes in climate are expected to increase the effect of O_3 precursor emissions over source regions and reduce the effect over downwind receptor regions. However, the magnitude of these effects is relatively small, and is driven by changes in atmospheric chemistry and not by changes in transport patterns. The effect of natural emission changes and wider climate-related feedbacks have not been evaluated fully yet.

6.5. Implications for International Policy

Taken together, O_3 associated with natural sources and anthropogenic intercontinental transport comprise a large fraction of observed O_3. By themselves, these sources create O_3 concentrations that are damaging to human health and ecosystems, contribute significantly to near-term climate change, exceed ambient standards for the protection of ecosystems in many areas, and, in some cases, exceed ambient standards for the protection of public health. Likewise, PM associated with natural sources and anthropogenic intercontinental transport can often exceed short-term ambient standards for the protection of public health and contribute to acidification, eutrophication, near-term climate change, and visibility degradation. The sources of these pollutants are not within the control of national or sub-national political jurisdictions. Thus, without further international cooperation to mitigate intercontinental flows of air pollution, many nations are not able currently to meet their own goals and objectives for protecting public health and environmental quality.

Mitigation of intercontinental transport is not a substitute for emission reductions at the local and regional scale. In most cases, concentrations within a source region are more sensitive to emission changes within that region. However, as emissions at the local and regional scale are reduced, the relative importance of intercontinental transport increases. Furthermore, as emissions increase in some parts of the world and decrease in others, the absolute importance of intercontinental transport increases. The import of pollution from outside a region increases the control costs needed to meet fixed ambient air quality objectives, offsetting progress made by emission reductions within the source region. Modelling performed under the HTAP multi-model experiments has demonstrated that comparable mitigation efforts carried out in key regions across the Northern Hemisphere can have significant benefits within source regions, as well as downwind across the entire Northern Hemisphere. Thus, international cooperation to reduce intercontinental transport of pollution is an effective and necessary complement to local and regional emission controls.

The anthropogenic emission sources that contribute most to intercontinental transport are often the same, regardless of whether considering O_3 or PM. Typically, these sources are associated with combustion of fossil fuels in the electricity, transportation, industrial, and residential sectors, or with anthropogenic biomass burning. These are also important sources of long-lived greenhouse gases. Different mitigation strategies will have different implications for the mix of emissions that result and the associated impacts on public health, agriculture, natural ecosystems, and climate change on the local, regional, intercontinental, and global scales. Examining these issues together, there is a potential to find solutions that maximize multiple benefits on multiple spatial scales.

6.6. Further research and analysis needs

The variability in current model estimates of transport magnitudes and the inability to explain some of the observed trends suggests that more research is needed to satisfactorily assess the significance of intercontinental transport. In particular, further efforts are needed to improve: the accuracy and spatial and temporal resolution of emissions estimates; the spatial, temporal, vertical, and chemical resolution of the current observing system; and the description of some chemical and physical processes in current models.

Improving our assessment of intercontinental and hemispheric transport will require an integrated approach where the best available knowledge from observations, emissions, and models is combined. A robust observational system, using multiple observational platforms and methods, is

needed to provide data for the evaluation and improvement of chemical transport models and emissions inventories. Further analytical efforts are needed to decrease the range of current model estimates for source-receptor relationships and improve our confidence in the assessment of intercontinental source-receptor relationships.

Key Challenges

Some of the key challenges that we face are outlined below. Addressing each of these challenges requires linking information across the areas of observation, emissions, and modelling to:

- Improve the modelling of transport processes using existing and new field campaign data. Focused evaluation of models using field campaign data is needed to improve descriptions of small-scale boundary layer venting, atmospheric subsidence of transported polluted air masses into the boundary layer, wet scavenging, and transport processes.
- Advance understanding of the role that deposition of pollutants to vegetated land and ocean surfaces plays in determining atmospheric pollution mass balance and hence intercontinental transport. These same deposition processes also represent the pathways by which terrestrial and marine ecosystems are impacted by air pollution. As such, an integrated approach to improve our understanding of these deposition processes may benefit our understanding of intercontinental transport as well as impacts.
- Improve global emissions inventories, using existing information at the national and sub-national scales, inverse modelling, and other methods to compare emissions estimates to ground-based, aircraft, and satellite observations.
- Identify and explain observed long-term trends by filling gaps in the observing system, developing reliable emission trends, and improving model descriptions. The current observational system has limited coverage and resolution in most regions of the world and provides limited information about the vertical distribution of pollutants. Better observational information is essential for improving the ability to detect and explain long-term changes.
- Develop a robust understanding of current source-receptor relationships using multiple modelling techniques and analyses of observations. The initial results of the HTAP multi-model experiments provide some useful information about the significance of intercontinental transport, but further detailed analyses are needed.
- Estimate future source-receptor relationships under changing emissions and climate. Such scenarios should consider future years from 2020 to 2050 and 2100 and be coordinated with efforts under the Intergovernmental Panel on Climate Change.
- Improve organizational relationships and information management infrastructures to facilitate necessary research and analysis. Efforts should further the implementation of the strategy for Integrated Global Atmospheric Chemistry Observations , building upon the World Meteorological Organization’s Global Atmospheric Watch program and contributing to the Global Earth Observation System of Systems.

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