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Megacities and climate change – A brief overview

Gerd A. Folberth ^{a, *}, Timothy M. Butler ^b, William J. Collins ^c, Steven T. Rumbold ^{a, d}

^a UK Met Office Hadley Centre, Exeter, UK

^b Institute for Advanced Sustainability Studies, Potsdam, Germany

^c Department of Meteorology, University of Reading, Reading, UK

^d NCAS-Climate, University of Reading, Reading, UK

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ABSTRACT

Cities have developed into the hotspots of human economic activity. From the appearance of the first cities in the Neolithic to 21st century metropolis their impact on the environment has always been apparent. With more people living in cities than in rural environments now it becomes crucial to understand these environmental impacts. With the immergence of megacities in the 20th century and their continued growth in both, population and economic power, the environmental impact has reached the global scale. In this paper we examine megacity impacts on atmospheric composition and climate. We present basic concepts, discuss various definitions of footprints, summarize research on megacity impacts and assess the impact of megacity emissions on air quality and on the climate at the regional to global scale. The intention and ambition of this paper is to give a comprehensive but brief overview of the science with regard to megacities and the environment.

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1. Introduction

It has long been recognized that urban areas affect air quality significantly on all scales. The impacts of urban agglomerations have been subject to numerous modelling studies and observation campaigns (c.f., e.g., White et al., 1976; Hov et al., 1978). The potential of large cities to contribute significantly to air pollution over large distances, even up to the hemispheric scale, has also been established (HTAP, 2010).

The largest cities in the world have been labelled "megacities" (UN, 2006). Megacities are urban agglomerations exceeding ten million inhabitants. The high population density and large number of businesses and production facilities not only turn these megacities into hotspots of economic activity but also into large sources of pollutants that impact on their environment. Emissions to the atmosphere of trace gases and aerosol species such as carbon dioxide (CO₂), nitrogen oxides (NO_x), and volatile organic compounds (VOCs), ammonia (NH₃), sulphur dioxide (SO₂) and black and organic carbon (BC, OC), arguably, prominent by-products of human activity in megacities. These emissions of greenhouse gases and pollutants impact both, the composition of the atmosphere and the climate.

In this paper we will discuss briefly some of the most prominent aspects of megacity impacts on the environment. In the first section a few of the basic concepts used in the discussions that follow are explained. Then, the impact of megacities on the environment is discussed applying several different definitions of so-called "footprints". The third section discusses specifically the impacts of megacities on atmospheric composition and climate. The paper concludes with a brief discussion and final remarks.

Due to the vastness of the subject and the limited space available the discussion presented here must remain brief and somewhat superficial. However, we have made an effort to present the main ideas in each case and to include useful references where a detailed discussion would require us to go beyond the scope of this paper. Notwithstanding, it is our hope and ambition to provide the reader a succinct overview of the impact that megacities can have on the environment, particularly on atmospheric composition and climate, and encourage further interest in the subject.

2. Explaining a few important common concepts

The discussion of commonly used concepts in this section is deliberately kept to the size of a very short introduction only. It is intended to facilitate understanding of the arguments presented here. The reader will find a more detailed discussion of most of the subjects by referring to the cited literature (and references therein) if they wish so.

* Corresponding author. E-mail address: gerd.folberth@metoffice.gov.uk (G.A. Folberth).

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2.1. What is a "megacity"?

In a paper published in 1950 Vere Gordon Chlide defined a human settlement to qualify as a "city" by applying ten general metrics that essentially reflect the trend to an increasing population density which were accompanied by a high degree of differentiation and specialization in occupations (Chlide, 1950). With this definition in mind we can say that the first cities appeared after the Neolithic Revolution in Mesopotamia and Egypt nearly 10,000 years ago (e.g., Davis, 1955). For most of human history since then the vast majority of the population continued to live in a rural context engaging in subsistence agriculture while in small centres of population trade and manufacture on a small scale represented the dominant economic occupation. At the beginning of the 19th century, however, a steep increase in urbanization occurred and this trend has continued ever since.

Since the industrialization the growth in the urban population was driven by both continued migration from rural areas to the cities and the demographic expansion following tremendous achievements in medicine, the sciences and engineering. This global trend has reached a landmark point in 2007 (UN, 2008) when for the first time in human history the urban population surpassed in number the human population in rural areas. The 2007 UN report "UNFPA state of world population 2007 – Unleashing the potential of urban growth" (UNFPA, 2007) has argued that this trend to global urbanization is inevitable and likely to continue in the future.

The largest cities have reached a population in excess of several million people. Cities of this size are called "mega-cities" (UN, 2006). A formal, more comprehensive definition does not yet exist but more or less to general consensus – and following the UN population reports – a megacity is defined as "a metropolitan area with a total population in excess of ten million people". Megacities can either be a single metropolitan area or consist of two or more such areas that have converged. In the latter case the megacity is sometimes referred to as "conurbation", "metropolis" or "metroplex".

There exist several lists of the megacities.¹ According to these lists there are at present between 20 and 30 megacities in the world. In comparison, the biggest include Tokyo with 39,400,000 inhabitants in 2014 (~13,200,000 in the Tokyo Metropolis), Shanghai with 29,600,000 inhabitants (apparently all in the Metropolis but approximately 50% classified as permanent residents and 50% as commuters), New Delhi (25,300,000), Mexico City (22,200,000) and New York (21,800,000). While Tokyo is the largest metropolitan area Shanghai is the biggest city proper at present. Today, approximately 10% of the entire human population of the world lives in megacities. However, due to the lack of a strict definition of a megacity this population percentage carries a very high uncertainty.

2.2. What is "radiative forcing"?

Radiative forcing of the climate system is discussed in great detail in the IPCC's Fifth's Assessment Report (IPCC AR5, 2013) in Chapter 8 of the Contribution from Working Group 1 (Myhre et al., 2013). Here, we present a very concise summary of the very basics only. The reader is encouraged to follow up details in the IPCC AR5.

In short, "radiative forcing" (RF) refers to the "net change in the energy balance of the Earth system due to some imposed perturbation" (Myhre et al., 2013). RF is given in units of watts per square

metre (W m⁻² s⁻¹). It quantifies the imbalance in the energy flux as a consequence of the imposed change averaged over a specified period of time. With most other climate metrics based on RF, it has become one of the most widely used metrics in climate science and policy making. There exist a number of definitions for RF, all of which have limitations but also advantages. Here, we only discuss the most basic concepts.

At its simplest radiative forcing is defined as the "instantaneous change in the net radiative flux due to an imposed change" (Myhre et al., 2013). The *instantaneous RF* can be calculated as the flux changes either at the climatological tropopause or the top of the atmosphere (TOA). As depicted in Fig. 1 a, any responses of the climate system (represented by the temperature profile in Fig. 1a) are explicitly excluded from this definition of RF. On the other hand, instantaneous RF is generally easy to compute with radiation transfer and climate models. However, in the IPCC's TAR (Third Assessment Report) and AR4 the *stratospherically adjusted RF* in which stratospheric temperatures are allowed to readjust as shown in Fig. 1b has been adopted.

Quite importantly, the definition of the instantaneous RF implicitly assumes that the "change in net irradiance in response to the imposed forcing alone can be separated from all subsequent responses to the forcing" (Myhre et al., 2013). However, the net change in irradiance to the forcing alone is not always clearly separable from the responses, especially when the responses occur on (very) short time scales. Examples for rapid adjustments in the climate system are cloud effects which affect energy fluxes but are not strictly part of RF. Consequently, there remains some ambiguity in what is a forcing and what effect must be considered part of the response in the climate system. In order to alleviate some of these ambiguities several methods have been devised to calculate an *effective radiative forcing* (ERF) as discussed in Myhre et al. (2013).

If the climate system readjusts completely after the forcing so that the net flux in irradiance is zero again and the climate system has reached a new equilibrium state as indicated in Fig. 1 c a simple relationship between the sustained RF and the equilibrium global mean surface temperature response (ΔT) can be derived:

$$\Delta T = \lambda \cdot RF \tag{1}$$

where λ is the equilibrium climate sensitivity (ECS) parameter. However, for the climate system to reach a new equilibrium state for all agents and components several millennia may be required. The "adjusted" and "effective" radiative forcings give increasingly better estimates of ΔT .

An equilibrium change in the global mean surface temperature in response to a doubling of CO₂ according to Equation (1) is often referred to as the "climate sensitivity". In this case the sustained RF is caused by a doubling of the atmospheric CO₂ concentration. At the end of the 19th century, while working to find an explanation for the occurrence of ice ages, the Swedish scientist Svante Arrhenius calculated the potential temperature change due to a doubling of CO₂ in the atmosphere. His result of 5-6 °C of warming on a global average (Arrhenius, 1896) is surprisingly close to the findings presented in the last IPCC assessment report: "there is high confidence that ECS [equilibrium climate sensitivity] is extremely unlikely less than 1 °C and medium confidence that the ECS is likely between 1.5 °C and 4.5 °C and very unlikely greater than 6 °C" (Bindoff et al., 2013).

2.3. What is the "Global Warming Potential"?

Many efforts in climate science revolve around quantifying the human impact on the climate and Earth system. Emissions of trace gases and aerosols from human activities such as energy

¹ E.g., http://www.nationsonline.org/oneworld/bigcities.htm, http://l-lists.com/ en/lists/2mzkgs.html, http://www.citypopulation.de/world/Agglomerations.html.

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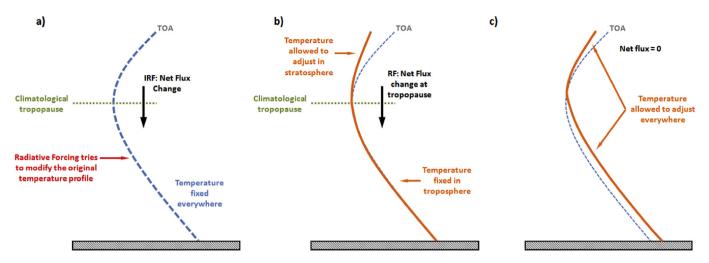


Fig. 1. Diagram comparing (a) instantaneous RF (b) stratospherically adjusted RF, commonly referred to as simply RF, and (c) the equilibrium response to climate forcing. Diagrams after Myhre et al. (2013).

production, industrial activities and transport are amongst the most prominent impacts. A suitable measure is required to quantify the impact of these emissions on the climate. For everyday use, when running complex climate or Earth system models is not an option, simpler measures or "metrics" are used. These metrics are based on complex models but do not require them to be applied. One such metric is the "Global Warming Potential" (GWP).

The GWP is defined as the "time-integrated RF due to a pulse emission of a given component (or trace gas or aerosol species), relative to a pulse emission of an equal mass of CO₂" (Myhre et al., 2013). Thus, the GWP can be interpreted as an index that quantifies the total energy that is added to the climate system through RF by a pulse of the specific component (or trace gas or aerosol species) relative to the energy added through RF by the equivalent mass of CO₂. It is best described as a "relative cumulative forcing index" (Myhre et al., 2013).

In general application GWP is usually determined over a 20, 100, or 500 year time horizon. It should be noted that these time horizons do not have any special significance and represent pure value judgements because they put relative weight of effects at different times. This is a consequence mainly of atmospheric lifetime and the effectiveness of individual trace gases to absorb long-wave radiation, respectively. Table 1 summarizes atmospheric lifetimes and GWPs over the 20 and 100 year time horizon (GWP₂₀ and GWP₁₀₀, respectively) for three of the most important anthropogenic greenhouse gases.

For example, methane (CH₄), even though it has a substantially lower atmospheric burden than CO₂, is significantly more effective as a greenhouse gas: $GWP_{100}(CH_4) > GWP_{100}(CO_2)$. Furthermore, it is apparent from Table 1 that, due to its short lifetime of only 12.4 years, CH₄ is even more effective over the shorter 20-year time horizon than over the 100 year time horizon:

Table 1

Atmospheric lifetimes and GWPs on the 20 and 100-year time horizon for carbon dioxide (CO_2), nitrous oxide (N_2O) and methane (CH_4). These data are taken from the IPCC AR5 WG1, Chp. 8, p. 714 (Myhre et al., 2013). The GWPs for CH₄ include impacts on tropospheric ozone and stratospheric water vapour.

	CO ₂	N ₂ O	CH ₄
T (years)	Variable	121	12.4
GWP ₂₀	1	264	84
GWP ₁₀₀	1	265	28

 $GWP_{20}(CH_4) > GWP_{100}(CH_4)$. This dependence of the RF effect on the time horizon has important implications for climate mitigation.

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3. Environmental impacts of megacities – megacity footprints

Megacities can affect the environment in several ways. The population consumes food and energy and produces waste in solid, liquid, and gaseous form in order to sustain themselves and pursue business activities such as manufacture or services. Due to the very high population density in many cases neither food nor raw materials or energy can be produced locally. As a consequence, additional impacts on the environment occur through transportation of goods and waste products in and out of the cities. The impact and its extension can be expressed in various forms of footprints reaching from the local to the regional and even to the global scale. Some of the footprints that will be discussed here are ecological, atmospheric and climate footprints. The emphasis will be on the latter.

3.1. The ecological footprint

The concept of the "ecological footprint" (Rees, 1992), in essence, is the size of area that is required to sustain the megacity population, i.e., the land that is required to provide the necessary resources (food, energy, raw materials, etc.) and to process the waste products. A detailed discussion can be found in Rees (1992) and on the internet.²

To give an example, the Tokyo metropolitan area extends over 2188 km² with a total population of 13,189,000 inhabitants amounting to a population density of 6028/km² (Tokyo Metropolitan Government, 2011). Assuming a biocapacity of the Earth of 1.8 persons per hectare (the global biological capacity available per person, 2008 status; taken from "The Global Footprint Network – Glossary"³) the Tokyo metropolitan area would require 237,402 km² or roughly two thirds of Japan to sustain itself. Of course, the same area is also a centre of innovation and productivity accounting for a major portion of Japan's economic output.

² The Global Footprint Network, http://www.footprintnetwork.org/en/index.php/ GFN/.

³ http://www.footprintnetwork.org/en/index.php/GFN/page/glossary/ #biocapacity.

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It has to be noted, however, that there is a large degree of uncertainty around numbers available for the biocapacity on the global scale. In addition, the biocapacity varies each year with ecosystem management, agricultural practices, ecosystem degradation, and weather, and population size. Thus, the biocapacity for a particular country or region may differ considerably from the global average available biocapacity. Nevertheless, the ecological footprint is a useful indicator for the potential impact of a megacity on its environment.

3.2. The dispersion footprint

Dispersion footprints of megacities are designed to quantify the outflow of trace gases and aerosols from their virtually point-like sources (point-like at least on the global and, to some extent, the regional scale). In other words, dispersion footprints essentially describe the maximum distance away from the source, i.e., the megacity, over which significant concentration levels of pollution can still be detected. Two major factors determine the efficiency of the pollution outflow relative to regional pollution accumulation: 1) the geographic location and 2) the atmospheric lifetime of the pollutant.

The atmospheric lifetimes of typical pollutants are between hours (for instance, in the case of some VOCs) and years (as in the case of methane, nitrous oxide and of course carbon dioxide). Important pollutants such as ozone and particulate matter typically exhibit atmospheric lifetimes in the range of a few hours up to a few weeks (Seinfeld and Pandis, 1998). The longer the lifetime the greater the chance for a particular trace gas or aerosol species to become subject to long-range transport. Meteorological conditions, on the other hand, determine the strength of the outflow and depend on geographic location of the megacity and the season during which pollution emission occurs.

In a study by Lawrence et al. (2007) two different types of metrics have been applied to quantify the export of pollutants from megacities. The one type of metric examines the distance over which transport occurs and measures the mass of the generic tracers which is exported a minimum distance away from the source, either in the horizontal or the vertical. This type of metric seems appropriate for studying the dispersion patterns of pollutants emitted by megacities and, thus, is appropriately described as a *dispersion footprint*. The other metric examines the contribution of megacity emissions to the composition of the atmosphere and is, thus, appropriately referred to as a composition or chemical footprint. This type of footprint shall be discussed in the following section.

3.3. The chemical footprint

Guttikunda et al. (2005) defined the ambient chemical footprint of a megacity to assess the impact of a particular species on atmospheric composition within a certain distance of the emission source. According to this paper the chemical footprint of a megacity is the "area where the megacity emissions contribute to 10% or higher of the monthly mean ambient concentrations" of a particular pollutant, say ozone, below 1 km altitude. In this case we would speak of the *ambient ozone chemical footprint of an emission species X* emitted from a megacity. The 10% threshold was chosen in Guttikunda et al. (2005) because in their study the megacity emissions accounted for 10% of the total emissions in the region under consideration. The chemical footprint then identifies the area where the emission source, in this case the megacity, has a disproportionate (larger or smaller) role on ambient concentrations of pollutants.

4. Megacity impacts on air quality and climate

As already discussed, large agglomerations of human population and production facilities can have a number of severe impacts on the environment. Arguably the most important and most farreaching impact of megacities is on the atmospheric environment. For this reason, and for the sake of manageability, this paper will limit its discussion to the impact of megacities on atmospheric composition and climate. The first section will present a brief overview over existing research. In the following three sections emissions from megacities and the impacts of megacities on atmospheric composition and on climate will be discussed separately.

4.1. A brief overview of existing work on megacity impacts

The assessment and quantification of the impact of megacities on atmospheric composition and climate is still a comparatively new subject within the atmospheric sciences. While studies of pollution outflow and long-range transport on the global, continental and regional scale are relatively numerous (c.f., e.g., Akimoto, 2003; Heald et al., 2003; HTAP, 2010; Pfister et al., 2005) similar efforts assessing the emissions from megacities and large population centres are much less available.

First studies on the impact of megacities on air pollution appeared shortly after the turn of the century. Molina and Molina (2004) published a critical review on megacities and atmospheric pollution which appears to be the first study to draw a connection between megacities and atmospheric pollution. Shortly afterwards several studies were published which expanded on the subject.

The first study on the effects of urban emissions on global air quality was published by Mayer et al. (2000). While not specifically focused on the impact of megacities on global atmospheric composition it analysed the connection between urbanization and key atmospheric trace components such as NO_x , ozone, and the hydroxyl radical. Around the same time, a small number of studies analysed specifically the impact of megacities on air quality on the regional scale (e.g., Gurjar et al., 2004; Guttikunda et al., 2005). The impact of megacities on the global scale was first examined in a series of modelling studies focussing on global atmospheric dispersion of pollutants (Lawrence et al., 2007) and global atmospheric chemistry (Butler and Lawrence, 2009; Butler et al., 2012). The potential impact of megacity emissions on the climate system was summarized in Folberth et al. (2012).

Several of the studies listed above have been conducted as part of large research efforts specifically initiated to study the connection between megacities and their environment from the local to the regional scale. Two of the largest, MEGAPOLI and CityZen, were funded by the European Union through Framework Programme 7. Both research programmes took place from 2008 to 2011 and were operated as sister projects.

MEGAPOLI (Megacities: Emissions, urban regional and Global Atmospheric POLlution and climate effects, and Integrated tools for assessment and mitigation) was a pan-European project that included a total of 23 research groups from 11 countries. The focus of this project has been on the impact assessment of megacities and large air-pollution hot-spots on local, regional and global air quality and to quantify feedbacks between air quality and climate. A comprehensive list of publications produced under MEGAPOLI can be found on the project web page (MEGAPOLI, 2011).

The CityZen project (megaCITY — Zoom for ENvironment) was designed to investigating air pollution distribution with a focus on a number of selected megacities and emission hotspots around the world. CityZen combined long-term satellite measurements and insitu observations with a series of modelling studies. Areas of focus

included the Eastern Mediterranean, the Po Valley region, the BeNeLux area and the Pearl River Delta agglomeration. 16 partner organizations in 11 countries were engaged in this project for a duration of three years. More project details and a comprehensive list of publications can be found on the CityZen web page (CityZen, 2011).

In 2006 an international collaborative research project took place which was focused on Mexico City, one of the world's largest megacities. The Megacity Initiative: Local and Global Research Observations (MILAGRO) campaign was aimed at conducting measurements of pollutants and other trace gases and particulate matter. It was to assess the atmospheric transport and transformation processes from the local to the global scale. The MILA-GRO Campaign was conducted in close collaboration with four other measurement campaigns taking place at the same time: Mexico City Metropolitan Area – 2006 Experiment (MCMA-2006), Megacity Aerosol Experiment (MAX-Mex), Megacity Impacts on Regional and Global Environments (MIRAGE-Mex), and Intercontinental Chemical Transport Experiment-B (INTEX-B). A comprehensive project description and publication list can be found on the project web site (MILAGRO, 2006).

A number of research projects have been conducted that were focused on particular regions or even cities, such as SAECAM/ ADAPTE with focus on South America, CAREBEIJING centred on the Beijing area, IMPACT with emphasis on the Tokyo Metropolitan Area and PRIDE-PRD focussing on the Pearl River Delta.

4.2. Anthropogenic emissions from megacities

Many of the human activities in megacities, such as industrial and energy production, transportation or residential heating, produce a variety of waste products. It is, however, the emission of pollutants in the form of trace gases and aerosols that have potentially the most wide-spread effect on the environment. The emission of nitrogen oxides (NO_x), carbon monoxide (CO) and volatile organic compounds (VOC) produces ozone in the troposphere (Haagen-Smit, 1952; Logan et al., 1981; Seinfeld and Pandis, 1998) and consequently leads to the formation of hydroxyl radical (OH) which determines the oxidation capacity of the atmosphere (Levy II, 1971; Warneck, 1974; Lelieveld et al., 2004). For most pollutants the reaction with the OH radical determines their atmospheric lifetime.

With approximately 10% of the entire human population living in megacities at the present these hotspots of economic activity and, consequently, waste production are significant sources of atmospheric pollutants. The dominant compounds emitted from megacities can be split into two major categories according to their atmospheric lifetimes, 1) greenhouse gases (or long-lived climate forcers, LLCFs) such as carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (N₂O) and 2) atmospheric pollutants (or short-lived climate forcers, SLCFs) such as nitrogen oxides (NO_x) , carbon monoxide (CO), volatile organic compounds (VOC) and particulate matter and their precursors (mainly sulphur dioxide, SO₂, organic carbon, OC, and black carbon, BC). With the timescale of effective inter-hemispheric mass exchange of the order of one year (e.g., Newell et al., 1969; Maiss et al., 1996) global mixing of SLCFs is effectively prevented while the much longer lifetimes of LLCFs allows an effective mass exchange across the ITCZ (inner-tropical convergence zone). Therefore, LLCFs are globally well mixed while SLCFs show considerable variation with latitude and even longitude within the respective hemisphere. Consequently, the impact of LLCFs appears globally homogeneous while SLCFs introduce significant regional variability in the Earth system with respect to radiative forcing and climate.

Table 2 summarizes the emissions for the dominant atmospheric pollutants emitted from megacities. Emissions are presented for the present-day (with base year 2005) and the future (base year 2050). The data is based on CMIP5 (Climate Model Intercomparison Project 5) emission scenarios for the historic (Lamarque et al., 2010) and future (Riahi et al., 2007) period and the table is a reproduction from Folberth et al. (2012). This latter paper also provides a more detailed discussion than can be presented here.

From Table 2 it is apparent that megacity emissions are dominated by greenhouse gases with CO_2 by far the major component. Ozone precursors (NO_x , CO, VOCs but also CH₄) and particulate matter (OC, BC, SO₂) represent a relatively minor portion of the emissions but are crucial for local and regional air quality. The contribution from megacities lies between 3% and 12% of the annual global total emissions (for these scenarios). With the exception of CO_2 all emissions are disproportionately smaller than would be expected from the megacity population fraction of 10% or, in other words, megacities underperform in term of emissions. This could be the case because some of the emissions are extra-mural in location. For instance, energy production could take place outside the cities themselves to some extend and this portion would not be accounted for in the megacity emission inventory (at least not in the simple modelling approach used in Folberth et al., 2012).

It is also apparent that emissions of long-lived greenhouse gases in the future scenario show a very different trend than the atmospheric pollutants. This is true for both the global base emissions and the emissions from megacities. While the emissions of greenhouse gases shows a substantial increase in 2050 relative to 2005 (consistent between global and megacity scenarios) the atmospheric pollutants show a disproportionately larger decrease in the megacity scenarios than in the global scenarios. In short, according to these scenarios megacities become significantly cleaner in the future. It must be noted, however, that in the scenarios presented in Table 2 no attempt has been made to extrapolate the development of megacities into the future, neither with respect to the number of megacities, their location or the increase in population size. For a detailed discussion of uncertainties the reader is referred to Folberth et al. (2012).

4.3. Megacity impacts on atmospheric composition

In a comprehensive modelling study Lawrence et al. (2007) have examined the tradeoffs between local and regional near-surface

Table 2

Summary of scenarios of anthropogenic emissions for present-day (reference year 2005) and future (reference year 2050) conditions. Relative contributions of megacities for each species are denoted in parenthesis for the present-day scenario. Relative differences (base and megacity contribution) for each species from present-day are given in parenthesis for the future scenario.

Species	2005		2050	
	Base	Megacity	Base	Megacity
Long-lived greenhouse gases (long-lived climate forcers, LLCFs) ^a				
$CO_2 (Tg yr^{-1})$	32,250.0	3870.0 (12%) ^b	68,280.0 (+112%) ^c	8194.0 (+112%)
CH_4 (Tg yr ⁻¹)	312.4	22.5 (7%)	676.8 (+110%)	47.4 (+111%)
$N_2O (Tg yr^{-1})$	8.0	0.3 (4%)	20.1 (+150%)	0.8 (+151%)
Short-lived climate-active pollutants (short-lived climate forcers, SLCFs) ^d				
NO_x (TgN yr ⁻¹)	43.4	2.0 (5%)	37.1 (-15%)	0.8 (-60%)
$CO (Tg yr^{-1})$	1080.4	35.8 (3%)	948.3 (-12%)	23.0 (-36%)
SO_2 (TgS yr ⁻¹)	28.5	1.5 (5%)	13.2 (-54%)	0.5 (-66%)
BC (Tg yr^{-1})	6.6	0.3 (5%)	4.5 (-32%)	0.1 (-66%)
$OC (Tg yr^{-1})$	34.2	1.0 (3%)	28.0 (-18%)	0.7 (-30%)

^a Based on EDGAR4.0 emission inventory (European Commission, 2009; Lamarque et al., 2010).

^b Percent contribution of megacities.

^c Change in emissions relative to present-day level (reference year 2005).

^d Based on CMIP5 RCP8.5 emission scenario (Riahi et al., 2007).

accumulation of pollution and extra-regional export of pollution through medium to long-range transport of pollutants emitted from megacities and major population centres. This pollution outflow occurs either through low-level advective processes or through deep convection to the free and upper troposphere. In both cases the outflow of air pollution from megacities and major population centres studied in Lawrence et al. (2007) is potentially significant for atmospheric composition and even climate (as will be discussed in the following section).

Lawrence et al. (2007) have demonstrated that long-range lowlevel advection is the dominant export mechanism at mid-to high latitudes while for megacity locations in the tropics deep convective transport seems to be more important. On the global scale, convection prevails over advection in terms of long-range transport mechanisms. Export of short-lived pollutants ($\tau \approx 1$ day in Lawrence et al., 2007) from megacities can distribute those species on the regional scale extending over several hundred to a few thousand kilometres. Pollutants with slightly longer atmospheric lifetimes ($\tau \approx 10$ days) can be dispersed on the continental scale extending over several thousand kilometres. Tracers with lifetimes longer than this ($\tau \approx 100$ days in Lawrence et al., 2007) can potentially reach dispersion on the hemispheric scale. This is typically the case for CO. Pollutants with even longer lifetimes, such as CH₄, N₂O and CO₂, are well-mixed on the global scale. Distribution altitude and distance varies considerable with the season. For further details we refer the reader to Lawrence et al., 2007.

Butler and Lawrence (2009) used a chemistry-transport model to study the impact of megacity emissions on the burden of key atmospheric pollutants (CO, NO_x, O₃, OH). In this modelling study the authors applied several scenarios, one for present-day conditions and three for future conditions at the 2030 time horizon. Scenario S1 refers to emissions for the year 2000. The future scenarios S2–S4 examine projected 2030 emissions under current emission control legislation (S2), maximum feasible reduction (S3) and worst-case evolution (S4) assumptions (Dentener et al., 2005, 2006). Table 3 lists the percentages of the Earth's surface for which at least a 10% increase in the annual mean concentration of specific key pollutants has been found in the model simulations under the various scenarios. The biggest impacts are found for NO_x at roughly 6% of the Earth's surface area affected under present-day conditions.

Overall, Butler and Lawrence (2009) found that global scale effects of megacities are generally quite small and also disproportionately smaller than the proportion of anthropogenic emissions from megacities. However, impacts on the regional scale can be quite substantial.

4.4. Megacity impacts on climate

Pollutant emissions from megacities do not only affect atmospheric composition. In changing the burden of key trace constituents significantly megacities can also have an impact on climate. As discussed above, megacity emissions divide into long-lived (e.g., CO_2 and CH_4) and short-lived climate forcers (e.g., O_3 , NO_x , CO, VOCs, OC, BC, $SO_{\overline{4}}$). Of the latter O_3 is an effective greenhouse gas

Table 3

Percentage of the Earth's surface area which experiences at least a 10% increase in the annual mean concentration of key pollutants due to anthropogenic emissions from megacities. Table reproduced from Butler and Lawrence (2009).

Emission	S1	S2	S3	S4
NO _x	6.25	4.96	1.94	3.27
O ₃	0.12	0.16	0.07	0.05
OH	0.52	0.51	0.53	0.44

contributing 350 mW m⁻² (with a range between 150 and 550 mW m⁻²; c.f., Myhre et al., 2013) in terms of additional, anthropogenic radiative forcing since the pre-industrial era. NO_x, CO and VOC are ozone precursors and also impact the amount of hydroxyl radicals in the atmosphere which, in turn, can change the atmospheric lifetime and, consequently, atmospheric burden of long-lived climate forcers such as, for instance, methane. These pollutants impact the climate indirectly and on the short time scale and are, therefore, called short-lived climate forcers, SLCFs.

Table 4 presents an overview and comparison of the radiative forcing of key pollutants emitted from megacities for present-day and future conditions. These data are reproduced from work accumulated during the MEGAPOLI project (MEGAPOLI, 2011) and summarized in Folberth et al. (2012). A detailed discussion of the methodology and scenarios applied in those studies can be found in this paper (and references therein). Here, we will only discuss the principal findings in term of the impact of megacities on the climate.

Carbon dioxide is the dominant species not only in the total anthropogenic emissions from megacities but also with respect to their impact on climate. While CO_2 is chemically inert and, thus, does not play a role in air quality, its long lifetime (c.f., Table 1) and effectiveness as a greenhouse gas turns it into a major factor with regard to climate impacts of megacities. CO_2 dominates the radiative forcing contribution of megacities both under present-day and future conditions (c.f., Table 4) contributing roughly 80% of the total radiative forcing from megacity emissions under both scenarios.

Interestingly, the total annual mean contribution from shortlived climate forcers – or atmospheric pollutants – is considerably smaller than that of each of the individual components: The individual effects seem to cancel each other out. Admittedly, even the radiative forcing of each of the individual components in the SLCFs class is dwarfed by the radiative forcing of carbon dioxide emitted from megacities. This is true both under present-day and future conditions.

Maintaining a constant 2005 level of emissions (i.e., no future increase or decrease in megacity emissions), megacities would be responsible for a global average annual mean warming of over 0.2 K over the next 100 years. This corresponds to 25% of the total global warming of 0.8 K since the pre-industrial era. Presented in another way, an increase of 0.2 K in the global surface temperature due to

Table 4

Annual mean top of the atmosphere (AMTOA) direct radiative forcing (DRF) due to emissions of long-lived greenhouse gases (long-lived climate forcers) and short-lived climate forcers from megacities. SW_{as} denotes "short-wave all-sky" TOA aerosol forcing and LW_{cs} refers to "long-wave clear-sky" TOA aerosol forcing. Table reproduced from Folberth et al. (2012.)

Species	2005 DRF (mW m^{-2})	2050 DRF (mW m^{-2})			
DRF from megacity emissions of long-lived climate forcers					
CO ₂ (tot AMTOA)	+120.0	+254.0			
CH ₄ (tot AMTOA)	+28.4	+59.8			
N ₂ O (tot AMTOA)	+3.3	+8.8			
Total forcing long-lived (AMTOA)	+151.7	+322.6			
DRF from megacity emissions of short-lived climate forcers					
CH ₄ (tot AMTOA)	-1.9 ± 0.04	-0.7 ± 0.02			
O ₃ (tot AMTOA)	$+5.7\pm0.02$	$+2.8\pm0.02$			
SW _{as} (tot AMTOA; SO ₂ , OC) ^a	-6.1 ± 0.21	-2.2 ± 0.10			
LW _{cs} (tot AMTOA; BC) ^b	$+1.5\pm0.01$	$+0.6 \pm 0.01$			
Total forcing short-lived (AMTOA)	-0.8 ± 0.24	$+0.5\pm0.09$			
Combined direct radiative forcing					
Total forcing	$+150.9\pm0.24$	$+323.1\pm0.09$			

^a Short-wave aerosol forcing is mostly due to sulphate (from in-situ oxidation of SO₂) and organic carbon (OC) aerosols and their impacts on clouds which results in efficient back-scattering of incoming sw-radiation.

^b Long-wave aerosol radiative forcing is mostly due to black carbon (BC) aerosols which efficiently absorb outgoing lw-radiation.

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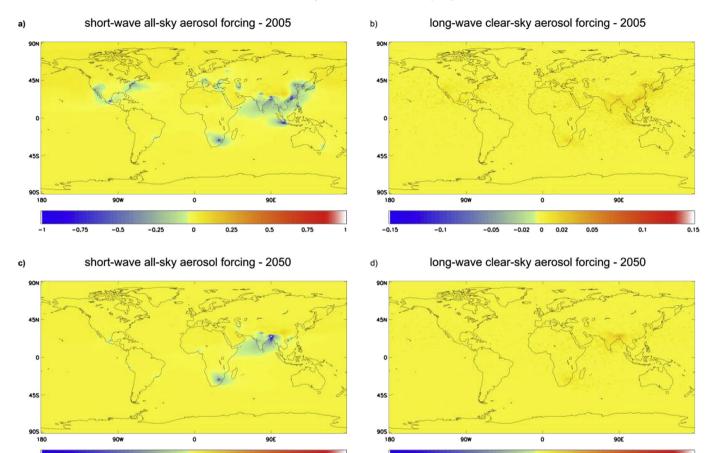


Fig. 2. Global distribution of AMTOA short-wave all-sky (left column) and long-wave clear-sky direct radiative forcing from aerosols emitted from megacities. Results are shown for present-day (upper row) and future conditions (lower row). Figure reproduced from Folberth et al. (2012).

-0.15

-0.1

megacity emissions compares to a 2.6-4.8 K increase of the total global surface temperature predicted for the end of the 21st century according to the worst case scenario in the IPCC fifth assessment report (Chapter 8 on radiative forcing; Myhre et al., 2013) – note: Folberth et al. (2012) have applied this worst-case scenario in their study.

0.25

0.5

0.75

As an illustration for the impact of SLCFs, Fig. 2 shows the geographic distribution of the AMTOA aerosol radiative forcing due to megacity emissions of OC, BC and SO₂. The radiative forcing of aerosols originating from megacities appears to be limited to the regional scale and is most pronounced in South and South-East Asia. The short-wave aerosol forcing is dominated by SO₂ emissions while the long-wave forcing is mostly due to black carbon (BC) aerosols. The latter, however, is only 25% of the short-wave forcing (on a global annual mean basis; c.f., Table 4) and of opposite sign. This holds true for both the present-day and the future.

5. Discussions and concluding remarks

-0.75

-0.5

-0.25

In this paper we have attempted to provide a brief introduction and overview of the effects and impacts megacities have on global atmospheric composition and climate. A number of important and commonly used concepts have been briefly introduced and extent and magnitude of the impact of megacities have been summarized. We have attempted to keep a balance between breadth and brevity. Consequently, the discussion has remained short and incomplete of necessity. For a deeper insight the reader is referred to the cited literature and references therein. It has to be repeated here that large uncertainties still persist around the analysis presented here and the referenced papers. Main factors of uncertainty persist around emissions and their geographic distribution, especially their future projections, and the extent and evolution over time of megacities. The principal tool for the assessment of megacity impacts on composition and climate are models, a fact that contributes not insignificantly to the overall uncertainty in the impact assessment presented. Further research is needed in order to fully understand the role that megacities play in the Earth system.

0 0.02

0.05

-0.02

-0.05

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References

Akimoto, H., 2003. Global air quality and pollution. Science 302, 1716–1719. Arrhenius, Svante, 1896. On the influence of carbonic acid in the air upon the temperature of the ground. Lond. Edinb. Dublin Philos. Mag. J. Sci. Ser. 5 41 (251), 237–276. April 1896.

0.15

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- Bindoff, N.L., Stott, P.A., AchutaRao, K.M., Allen, M.R., Gillett, N., Gutzler, D., Hansingo, K., Hegerl, G., Hu, Y., Jain, S., Mokhov, I.I., Overland, J., Perlwitz, J., Sebbari, R., Zhang, X., 2013. Detection and attribution of climate change: from global to regional. In: Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. (Eds.), Climate Change 2013: the Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Butler, T.M., Lawrence, M.G., 2009. The influence of megacities on global atmospheric chemistry: a modelling study. Environ. Chem. 6, 219–225. http:// dx.doi.org/10.1071/EN09110.
- Butler, T.M., Stock, Z.S., Russo, M.R., Denier van der Gon, H.A.C., Lawrence, M.G., 2012. Megacity ozone air quality under four alternative future scenarios. Atmos. Chem. Phys. 12, 4413–4428.
- Chlide, V.G., 1950. The urban revolution. Town Plan. Rev. 21 (1), 3-17.
- CityZen, 2011. megaCITY Zoom for the ENvironment, THEME FP7-ENV-2007.1.1.2.1: Megacities and Regional Hot-spots Air Quality and Climate. http:// cityzen-project.eu/.
- Davis, K., 1955. The origin and growth of urbanization in the world. Am. J. Sociol. 60 (5), 429-437.
- Dentener, F., Stevenson, D., Cofala, J., Mechler, R., Amann, M., Bergamaschi, P., Raes, F., Derwent, R., 2005. The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030. Atmos. Chem. Phys. 5, 1731.
- Dentener, F., Stevenson, D., Ellingsen, K., van Noije, T., Schultz, M., Amann, M., Atherton, C., Bell, N., et al., 2006. The global atmospheric environment for the next generation. Environ. Sci. Technol. 40 http://dx.doi.org/10.1021/ES0523845.
- European Commission, 2009. Emission Database for Global Atmospheric Research (EDGAR), Release Version 4.0. Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL). http://www.edgar.jrc.ec.europa.eu.
- Folberth, G.A., Rumbold, S.T., Collins, W.J., Butler, T.M., 2012. Global radiative forcing and megacities. Urban Clim. 1, 4–19. http://dx.doi.org/10.1016/ i.uclim.2012.08.001.
- Gurjar, B., van Ardenne, J., Lelieveld, J., Mohan, M., 2004. Emission estimates and trends (1990–2000) for megacity Delhi and implications. Atmos. Environ. 38, 5663–5681.
- Guttikunda, S.K., Tang, Y., Carmichael, G.R., Kurata, G., Pan, L., Streets, D.G., Woo, J.-H., Thongbooncoo, N., Fried, A., 2005. Impacts of Asian megacity emissions on regional air quality during spring 2001. J. Geophys. Res. 110 (D20301) http:// dx.doi.org/10.1029/2004JD004921.
- Haagen-Smit, A.J., 1952. Chemistry and physiology of Los Angeles Smog. Ind. Eng. Chem. 44 (6).
- Heald, C.L., Jacob, D.J., Fiore, A.M., et al., 2003. Asian outflow and trans-Pacific transport of carbon monoxide and ozone pollution: an integrated satellite, aircraft, and model perspective. J. Geophys. Res. 108 (D24), 4804. http:// dx.doi.org/10.1029/2003JD003507.
- Hov, Ø., Hesstveldt, E., Isaksen, I.S.A., 1978. Long-range transport, of tropospheric ozone. Nature 273, 341–344.
- HTAP, 2010. Hemispheric transport of air pollution 2010 part A: ozone and particulate matter. In: Dentener, F., Keating, T., Akimoto, H. (Eds.), Air Pollution Studies No. 17. ISSN: 1014-4625. United Nations Publication, Geneva.
- Lamarque, J.-F., Bond, T.C., Eyring, V., Granier, C., Heil, A., Klimont, Z., Lee, D., Liousse, C., Mieville, A., Owen, B., Schultz, M.G., Shindell, D., Smith, S.J., Stehfest, E., Van Aardenne, J., Cooper, O.R., Kainuma, M., Mahowald, N., McConnell, J.R., Naik, V., Riahi, K., van Vuuren, D.P., 2010. Historical (1850–2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. Atmos. Chem. Phys. 10, 7017–7039. http://dx.doi.org/10.5194/acp-10-7017-2010.
- Lawrence, M.G., Butler, T.M., Steinkamp, J., Gurjar, B.R., Lelieveld, J., 2007. Regional pollution potentials of megacities and other major population centres. Atmos. Chem. Phys. 7, 3969–3987. http://dx.doi.org/10.5194/acp-7-3969-2007.

- Lelieveld, J., Dentener, F.J., Peters, W., Krol, M.C., 2004. On the role of hydroxyl radicals in the self-cleansing capacity of the troposphere. Atmos. Chem. Phys. 4, 2337–2344. http://dx.doi.org/10.5194/acp-4-2337-2004.
- Levy II, H., 1971. Normal atmosphere: large radical and formaldehyde concentrations predicted. Science 173, 141–143.
- Logan, J.A., Prather, M.J., Wofsy, S.C., McElroy, M.B., 1981. Tropospheric chemistry: a global perspective. J. Geophys. Res. 86 (C8), 7210–7254. http://dx.doi.org/ 10.1029/JC086iC08p07210, 20.
- Maiss, M., Steele, L.P., Francey, R.J., Fraser, P.J., Langenfelds, R.L., Trivett, N.B.A., Levin, I., 1996. Sulfur hexafluoride – a powerful new atmospheric tracer. Atmos. Environ. 30 (10/11), 1621–1629.
- Mayer, M., Wang, C., Webster, M., Prinn, R., 2000. Linking local air pollution to global chemistry and climate. J. Geophys. Res. 105 (D18), 22,869–22896.
 MEGAPOLI, 2011. Megacities: Emissions, Urban, Regional and Global Atmospheric
- MEGAPOLI, 2011. Megacities: Emissions, Urban, Regional and Global Atmospheric POLlution and Climate Effects, and Integrated Tools for Assessment and Mitigation, THEME FP7-ENV-2007.1.1.2.1: Megacities and Regional Hot-spots Air Quality and Climate. http://megapoli.dmi.dk/.
- MILAGRO, 2006. Megacity Initiative: Local and Global Research Observations. https://www.eol.ucar.edu/node/4949.
- Molina, M.J., Molina, L.T., 2004. Megacities and atmospheric pollution. J. Air Waste Manag. Assoc. 54, 644–680.
- Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhang, H., 2013. Anthropogenic and natural radiative forcing. In: Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. (Eds.), Climate Change 2013: the Physical Science Basis, Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Newell, R.E., Vincent, D.C., Kidson, J.W., 1969. Interhemispheric mass exchange from meteorological and trace substance observations. Tellus XXI (5), 641–647.
- Pfister, G., Hess, P.G., Emmons, L.K., Lamarque, J.-F., Wiedinmyer, C., Edwards, D.P., Pétron, G., Gille, J.C., Sachse, G.W., 2005. Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data. Geophys. Res. Lett. 32 (L11809) http://dx.doi.org/10.1029/2005GL022995.
- Rees, W.E., 1992. Ecological footprints and appropriated carrying capacity: what urban economics leaves out. Environ. Urban. 4 (2), 121–130. http://dx.doi.org/ 10.1177/095624789200400212.
- Riahi, K., Gruebler, A., Nakicenovic, N., 2007. Scenarios of long-term socio-economic and environmental development under climate stabilization. Technol. Forcast. Soc. Change 74, 887–935.
- Seinfeld, J.H., Pandis, S.N., 1998. Atmospheric Chemistry and Physics from Air Pollution to Climate Change. John Wiley & Sons, Inc, New York, p. 1113.
- Tokyo Metropolitan Government, 2011. website: http://www.metro.tokyo.jp/ ENGLISH/PROFILE/overview03.htm.
- UN World Urbanization Prospects Revision 2005, Factsheet 7: Mega-cities, 2006. United Nations, Department of Economic and Social Affairs, Population Division. World Urbanization Prospects: The 2005 Revision. Working Paper No. ESA/ P/WP/200, 2006.
- UN World Urbanization Prospects the 2007 Revision, Executive Summary, 26 February 2008. United Nations, New York. http://www.un.org/esa/population/ publications/wup2007/2007WUP_ExecSum_web.pdf.
- UNFPA, 2007. UNFPA State of World Population Unleashing the Potential of Urban Growth. United Nations Population Fund. Thoraya Ahmed Obaid (Executive Director). http://www.unfpa.org/webdav/site/global/shared/documents/ publications/2007/695_filename_sowp2007_eng.pdf. © UNFPA.
- Warneck, P., 1974. On the role of OH and HO, radicals in the troposphere. Tellus 26 (1–2), 39–46.
- White, W.H., Anderson, J.A., Blumenthal, D.L., Husar, R.B., Gilani, N.V., Husar Jr., J.D.W.E.E., 1976. Formation and transport of secondary air pollutants: ozone and aerosols in the St. Luis urban plume. Science 194, 187–189.