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Max J. Suarez, Editor

Estimates of AOD Trends (2002 – 2012) over the World's Major Cities based on the MERRA Aerosol Reanalysis

Simon Provençal, Pavel Kishcha, Emily Elhacham, Arlindo M. da Silva and Pinhas Alpert

March 2014

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Simon Provençal
Département de géographie, Université Laval, Quebec City, Quebec, Canada

Pavel Kishcha
Department of Geophysical, Atmospheric and Planetary Sciences, Tel Aviv University, Tel Aviv, Israel

Emily Elhacham
Department of Geophysical, Atmospheric and Planetary Sciences, Tel Aviv University, Tel Aviv, Israel

Arlindo M. da Silva
Global Modeling and Assimilation Office, NASA/GSFC, Greenbelt, Maryland, USA

Pinhas Alpert
Department of Geophysical, Atmospheric and Planetary Sciences, Tel Aviv University, Tel Aviv, Israel

National Aeronautics and
Space Administration

Goddard Space Flight Center
Greenbelt, Maryland 20771

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Abstract

NASA's Global Modeling and Assimilation Office has extended the Modern-Era Retrospective Analysis for Research and Application (MERRA) tool with five atmospheric aerosol species (sulfates, organic carbon, black carbon, mineral dust and sea salt). This inclusion of aerosol reanalysis data is now known as MERRAero. This study analyses a ten-year period (July 2002 – June 2012) MERRAero aerosol reanalysis applied to the study of aerosol optical depth (AOD) and its trends for the aforementioned aerosol species over the world's major cities (with a population of over 2 million inhabitants). We found that a proportion of various aerosol species in total AOD exhibited a geographical dependence. Cities in industrialized regions (North America, Europe, central and eastern Asia) are characterized by a strong proportion of sulfate aerosols. Organic carbon aerosols are dominant over cities which are located in regions where biomass burning frequently occurs (South America and southern Africa). Mineral dust dominates other aerosol species in cities located in proximity to the major deserts (northern Africa and western Asia). Sea salt aerosols are prominent in coastal cities but are dominant aerosol species in very few of them. AOD trends are declining over cities in North America, Europe and Japan, as a result of effective air quality regulation. By contrast, the economic boom in China and India has led to increasing AOD trends over most cities in these two highly-populated countries. Increasing AOD trends over cities in the Middle East are caused by increasing desert dust.

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1 Introduction

Microscopic airborne aerosols have long been a prominent topic of study in the field of environmental science, predominantly in the atmospheric sciences. A considerable amount of literature has emerged in order to better understand the nature of these particles, but more specifically, to assess their impact on various spheres of life and the environment. Aerosols are found in highly variable space and time distribution, size and chemical composition, and they originate from many sources, both natural and anthropogenic [Poschl, 2005].

Aerosols affect considerably the environment and its living organisms. It is well documented that aerosols are a serious health hazard to humans, fauna and flora. They are linked to cardiovascular, respiratory and allergic diseases, as well as enhanced mortality [Poschl, 2005; Tager, 2013]. Aerosols affect weather and climate. Acting as cloud condensation nuclei, aerosols are an essential element of cloud formation. As such, they play an indirect role in increasing clouds' and the Earth's albedo as a whole [Haywood and Boucher, 2000; Lohmann and Feichter, 2005]. They also affect the Earth's radiation budget as absorbers of radiation, contributing to a warming of the atmosphere, and as reflectors of radiation, in which case they act as a cooling agent [Haywood and Boucher, 2000]. Finally, in high enough concentration, they can significantly reduce visibility [Charlson, 1969; Cheng and Tsai, 2000]. This is often associated with episodes of haze and smog.

The seriousness of the impacts listed in the previous paragraph is dependent on the aerosol concentration and size, but particularly on its chemical composition. It is therefore relevant to distinguish between different aerosol species:

- Sulfate (SU) aerosols are the product of fossil fuel and, to a much smaller extent, biomass burning, and therefore are vastly considered anthropogenic. However, small natural contributions originate from volcanoes and the oceans [Haywood and Boucher, 2000; Forster *et al.*, 2007, sect. 2.4.4.1];
- Organic carbon (OC) is the result of fossil fuel and biomass burning. The former is an anthropogenic source while the latter can be both a natural and an anthropogenic source. As a whole, sources of OC are widely considered to be anthropogenic [Haywood and Boucher, 2000; Forster *et al.*, 2007, sect. 2.4.4.3];
- Black carbon (BC) originates from the same sources as OC [Haywood and Boucher, 2000; Forster *et al.*, 2007, sect. 2.4.4.2];
- Sea salt (SS) aerosols originate from the oceans. The release of salt particles from the oceans depends on meteorological factors such as surface wind speed [Denman *et al.*, 2007, sect. 7.5.1.2];
- Mineral dust (DU) is the product of wind erosion predominantly in arid environments. Therefore, sources are in majority considered natural. However, agricultural and industrial practices, as well as deforestation, are responsible for a portion of anthropogenic dust aerosols in the atmosphere [Haywood and Boucher, 2000; Forster *et al.*, 2007, sect. 2.4.4.6; Denman *et al.*, 2007, sect. 7.5.1.1].

There is scientific interest in studying aerosol pollution in cities because associated industries and urban road traffic are major sources of particulate matter. Cities do offer a wide

variety of opportunities, such as better employment, education, health care, entertainment and other services, which stimulate an ongoing and accelerating urbanization movement around the world [Moore *et al.*, 2003]. In 1950, the United Nations estimated that 29% of the world's population lived in an urban area (http://esa.un.org/unup/CD-ROM/Urban-Rural-Population.htm). This proportion grew to 47% in 2000 and is expected to reach 67% in 2050. Particularly in developing countries, where the rate of urbanization is the greatest [Subbotina, 2004, chap. 10], cities are lacking the means to adjust fast enough to fulfill the demand of its rapidly growing population and economic development. In this respect, urbanization comes with its fair share of environmental consequences, among other problems, such as degradation of air quality [Shao *et al.*, 2006]. Indeed, atmospheric aerosol concentration is significantly higher in populated cities as opposed to rural or remote areas [Cheng and Tsai, 2000], and the cities' population growth in developing countries tend to correlate with an increase of aerosol concentration [Kishcha *et al.*, 2011]. Rapid urbanization and development in India and China resulted in a sharp increase of air pollutant emissions during the last decade [Lu *et al.*, 2011, Kishcha *et al.,2014*] and frequently recurring episodes of air pollution and haziness. On the other hand, urbanization in developed countries, albeit at a slower rate, hasn't had such a negative impact. Developed countries did indeed struggle with severe air pollution issues in the past, but their economical and democratic situation provides them with the means to enforce clean air regulations and develop green technologies. As a result, air quality has significantly improved over the last decades in the United States [Hand *et al.*, 2013], Europe [Vestreng *et al.*, 2007] and Japan [Kanada *et al.*, 2013], even though their population and GDP kept on growing.

Alpert *et al.* [2012] estimated aerosol optical depth (AOD) trends over the 189 largest cities in the world during the period from 2002 to 2010. They used three remote sensing instruments of the National Aeronautics and Space Administration (NASA): the two Moderate Resolution Imaging Spectroradiometers (MODIS) and the Multiangle Imaging SpectroRadiometer (MISR) on board the Terra and Aqua satellites. They found that AOD trends exhibit some geographical dependence: trends were increasing in Indian, Middle Eastern and Chinese cities, while they were decreasing in European, north-eastern American and south-eastern Asian cities. The clear advantage of using satellites to study aerosols, compared to ground-based measurements, is the global coverage. The major deficiency, however, is the inability to distinguish between aerosol species.

Several years ago, NASA's Global Modeling and Assimilation Office (GMAO) introduced the Modern-Era Retrospective Analysis for Research and Application (MERRA), a reanalysis tool incorporating satellite data to reproduce spatially consistent observations for many environmental variables. While the original MERRA included only meteorological parameters (winds, temperature, humidity, etc.), it has recently been extended to include assimilation of AOD from the MODIS sensor on both Aqua and Terra, which let to its rebranding now known as MERRAero. Although only the total AOD is constrained by MODIS observations, the data assimilation algorithm in MERRAero provides speciated monthly average, with the relative contribution from all major aerosol species (dust, sea salt, sulfates and carbonaceous).

The current study is aimed at estimating the contribution of these aerosol species to total AOD and their trends over the world's largest agglomerations (with population over 2 million people, hereafter simply referred to as "cities") using the obtained ten-year (July 2002 – June

2012) assimilated aerosol dataset. We focus our attention on dominant aerosol species and their trends over the specified cities located in different geographical regions.

2 Methodology and data

2.1 MERRAero aerosol reanalysis

The NASA GEOS-5 model was used to extend the MERRA reanalysis with five atmospheric aerosol components (sulfates, organic carbon, black carbon, mineral dust and sea salt). GEOS-5 is the latest version of the NASA GMAO Earth system model. GEOS-5 contains components for atmospheric circulation and composition (including atmospheric data assimilation), ocean circulation and biogeochemistry, and land surface processes. GEOS-5 includes a module representing atmospheric aerosols [Colarco *et al.*, 2010, and references therein]. This aerosol module is based on a version of the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model [Chin *et al.*, 2002]. GOCART treats the sources, sinks and chemistry of dust, sulfate, sea salt, and black and organic carbon aerosols. Aerosol species are assumed to be external mixtures. Except for biomass burning sources, aerosol emissions are based on the AeroCom version 2 hindcast inventories [Dr. Thomas Diehl, personal communication, and http://aerocom.met.no/emissions.html]; daily biomass burning aerosols are from the Quick Fire Emission Dataset (QFED, Darmenov and da Silva, 2013). Total mass of sulfate and carbonaceous aerosols are tracked, while for dust and sea salt, the particle size distribution is explicitly resolved across five non-interacting size bins for each. Both dust and sea salt have wind speed dependent emission functions, while sulfate and carbonaceous species have emissions principally from fossil fuel combustion, biomass burning and bio-fuel consumption, with additional biogenic sources of organic carbon. Sulfate has additional chemical production from oxidation of SO_2 and dimethylsulfide (DMS), as well as a database of volcanic SO_2 emissions and injection heights. GEOS-5 also includes assimilation of AOD observations from the MODIS sensor on both Terra and Aqua.

2.2 Method

In order to identify the cities of interest, Alpert *et al.* [2012] used data from City Population (http://www.citypopulation.de/world/Agglomerations.html), where all cities with a minimum population of 2 million inhabitants were selected. We use the same list of cities, even though the data from City Population is constantly being updated and its current list differs slightly from Alpert *et al.*'s [2012] list. A given city's population is only a mere criteria for selection, and therefore we have no reason to believe that neither Alpert *et al.*'s [2012] list, nor the current list from City Population, would produce significantly different results.

In order to retrieve data from MERRAero, a consistent resolution of $0.5^{o} \times 0.5^{o}$ was used over every city, which is approximately MERRAero's finest resolution. In order to define meridional and zonal boundaries, the coordinates (x, y) of a given city's center were subtracted

and added by 0.25º, i.e. for longitudinal boundaries: x ± 0.25º; and similarly for latitudinal boundaries: y ± 0.25º. First, MERRAero data were extracted for a period of 10 years, from July 2002 to June 2012. Then, the ten-year mean AOD was obtained for total and every one of the aerosol species. The AOD trend values in percentage form (for total and every one of the modeled aerosol species) correspond to the difference between the AOD averaged over the last 5-year period (July 2007 – June 2012) and the AOD averaged over the first 5-year period (July 2002 – June 2007), using as reference the AOD average over the first period.

3 Results

We first analyzed the raw data of total AOD averaged over the whole 10-year period. Table 1 illustrates 20 cities with the highest total AOD. Interestingly, Chinese cities occupy the entire list except for two Pakistani cities. If the list went down further, we would notice that all of the 50 cities with highest AOD are in Asia, except for six African cities. By contrast, cities with the lowest AOD don't seem to exhibit any particular geographical resemblance, spread over all other regions of the world.

Table 1. Cities with the highest average total AOD values

Rank	City	Country	AOD
1	Wuhan	China	0.723
2	Changsha	China	0.719
3	Zhengzhou	China	0.706
4	Nanjing	China	0.655
5	Nanchang	China	0.654
6	Shantou	China	0.642
7	Wuxi	China	0.613
8	Jinan	China	0.610
9	Tientsin	China	0.601
10	Suzhou	China	0.600
11	Zibo	China	0.584
12	Shijiazhuang	China	0.580
13	Shanghai	China	0.561
14	Nanning	China	0.557
15	Qingdao	China	0.552
16	Beijing	China	0.550
17	Lyallpur	Pakistan	0.537
18	Guangzhou	China	0.528
19	Lahore	Pakistan	0.521
20	Xi'an	China	0.511

Next, we analyzed the same data for every one of the aerosol species. These are illustrated in Tables 2 to 6 for the 10 cities with highest AOD respective to the species. Values of SU AOD (Table 2) are the highest among all species with Chinese cities occupying the first 19 cities with highest values and Asian cities occupying the first 52 cities, confirming high consumption of

fossil fuels in this region. Cities with the highest OC AOD values (Table 3) are mostly in China and India, with a few African cities included in the list, indicative of fuel and/or biomass burning. This is somewhat confirmed by BC AOD values (Table 4). The top 10 cities for this particular species are all in China while the top 22 cities are either in China or India. Relatively, BC AOD values are much lower compared to other species. SS AOD values (Table 5) are also fairly low. Nonetheless, cities with highest SS AOD values tend to be located close to the ocean and/or on a small island. DU AOD values (Table 6) are highest in cities of northern Africa and the Middle East, areas characterized by arid deserts. This brief analysis highlights, as expected, among the cities with the highest AOD, the fact that anthropogenic aerosols, mostly SU, are responsible for these high values. Tables 1 to 6 also exhibit an obvious geographical dependence not only for total AOD, but also for AOD of all aerosol species.

Table 2. Cities with the highest average SU AOD values

Rank	City	Country	AOD
1	Changsha	China	0.496
2	Wuhan	China	0.494
3	Zhengzhou	China	0.449
4	Nanchang	China	0.446
5	Nanjing	China	0.437
6	Shantou	China	0.416
7	Wuxi	China	0.403
8	Suzhou	China	0.394
9	Jinan	China	0.385
10	Tianjin	China	0.372

Table 3. Cities with the highest average OC AOD values

Rank	City	Country	AOD
1	Kinshasa	Congo	0.206
2	Luanda	Angola	0.197
3	Nanning	China	0.139
4	Changsha	China	0.122
5	Asunción	Paraguay	0.117
6	Nanchang	China	0.112
7	Wuhan	China	0.111
8	Guangzhou	China	0.110
9	Ibadan	Nigeria	0.110
10	Patna	India	0.108

The geographical dependence is also illustrated by Fig. 1 which represents the proportion of aerosol species in the ten-year mean total AOD over the world's megacities (i.e. the cities with population over 10 million). One can see that (a) megacities in the industrialized regions of North America, Europe and eastern Asia exhibit a strong presence of SU aerosols; (b) dust aerosol was dominant in megacities in northern Africa and the Middle East; and (c) South American megacities were characterized by a high proportion of organic carbon. In the following subsections, we concentrate on specific geographical regions.

Table 4. Cities with the highest average BC AOD values

Rank	City	Country	AOD
1	Zhengzhou	China	0.074
2	Shijiazhuang	China	0.066
3	Tianjin	China	0.064
4	Beijing	China	0.064
5	Wuhan	China	0.060
6	Jinan	China	0.059
7	Shantou	China	0.058
8	Changsha	China	0.056
9	Nanjing	China	0.055
10	Zibo	China	0.055

Table 5. Cities with the highest average SS AOD values

Rank	City	Country	AOD
1	Fortaleza	Brazil	0.058
2	San Juan	Puerto Rico	0.054
3	Taipei	Taiwan	0.052
4	Karachi	Pakistan	0.052
5	Santo Domingo	Dominican Republic	0.052
6	Salvador	Brazil	0.051
7	Dar es Salaam	Tanzania	0.051
8	Belem	Brazil	0.049
9	Manila	Philippines	0.048
10	Mumbai	India	0.048

Tables 6. Cities with the highest average DU AOD values

Rank	City	Country	AOD
1	Kano	Nigeria	0.378
2	Dakar	Senegal	0.317
3	Khartoum	Sudan	0.290
4	Baghdad	Iraq	0.275
5	Faisalabad	Pakistan	0.269
6	Lahore	Pakistan	0.248
7	Hyderabad	Pakistan	0.243
8	Gujranwala	Pakistan	0.232
9	Jidda	Saudi Arabia	0.218
10	Karachi	Pakistan	0.216

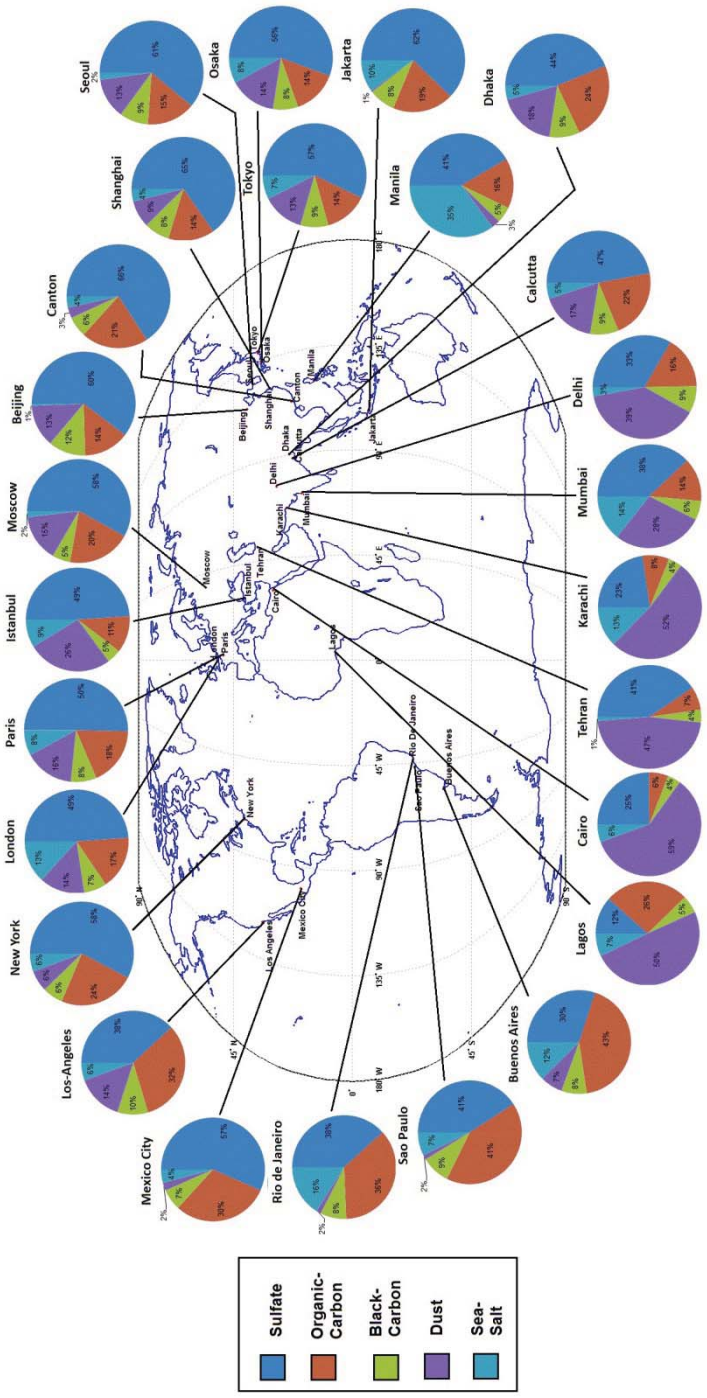

Fig. 1. Proportion of aerosol species in the MERRAero 10-year mean total AOD for the world's megacities.

3.1 Eastern and central United States and Canada

Total AOD values for cities in this part of the world are given in Table 7. The northeast is recognized as the most industrialized part of North America. According to Hand *et al.* [2012], the eastern states emit a substantially higher amount of SO_2 compared to the western states. This can explain why the majority of total AOD comes from SU aerosols in most cities displayed in Fig. 2. Note that cities in Florida, because they are surrounded by the Atlantic Ocean and the Gulf of Mexico, experience some significant contribution from SS aerosols. OC is second in importance and contributes about a quarter to total AOD in all the cities except Miami (19.0%).

Table 7. List of cities in eastern and central United States and Canada, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

State or province	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Québec	Montréal	0.163	-5.0	-3.2	-12.9	-5.5	4.9	9.2
Ontario	Toronto	0.170	-5.0	-5.9	-7.7	-4.1	7.6	6.9
Massachusetts	Boston	0.172	-6.4	-6.0	-12.4	-5.6	-3.7	8.8
New York	New York City	0.195	-4.0	-5.1	-6.1	-3.1	4.5	6.5
Pennsylvania	Philadelphia	0.199	-5.3	-7.0	-6.7	-3.7	4.5	6.0
	Pittsburgh	0.191	-7.1	-9.4	-6.5	-4.5	3.8	7.7
D.C.	Washington	0.198	-9.2	-11.2	-10.4	-6.6	-0.2	5.5
Michigan	Detroit	0.183	-4.0	-4.9	-6.2	-4.1	12.0	7.7
Ohio	Cleveland	0.188	-5.9	-7.4	-7.5	-4.4	11.0	8.0
	Cincinnati	0.198	-7.1	-9.1	-7.6	-3.8	3.0	8.0
Illinois	Chicago	0.183	-2.2	-3.3	-4.0	-2.2	10.8	10.0
Minnesota	Minneapolis	0.161	-1.3	-1.4	-4.7	-2.5	12.7	7.9
Missouri	St. Louis	0.187	-4.9	-5.7	-8.5	-4.2	10.6	8.9
	Kansas City	0.176	-1.1	-1.6	-5.1	-1.5	21.6	9.1
Colorado	Denver	0.096	-5.9	-7.5	-10.4	-2.8	3.6	4.9
Texas	Dallas	0.185	-4.2	-8.2	-6.0	-1.2	28.6	5.2
	Houston	0.197	-6.3	-9.5	-12.3	-4.0	13.6	4.5
	San Antonio	0.183	-5.5	-12.2	-4.8	1.6	13.0	6.4
Georgia	Atlanta	0.176	-9.1	-10.5	-11.0	-6.0	2.3	1.5
Florida	Orlando	0.164	-7.6	-7.7	-10.7	-5.8	-7.7	3.4
	Tampa	0.158	-9.6	-8.5	-17.6	-9.3	-6.8	2.7
	Miami	0.137	-7.5	-4.9	-15.5	-8.9	-6.2	-4.3

We found that, with respect to aerosol trends, every single city in this region noticed a decrease in total AOD as well as in AOD for every anthropogenic aerosol species (SU, OC and BC), except for the 1.6% BC AOD increase in San Antonio (Table 7). Our results on AOD trends are in line with previous studies [de Meij *et al.*, 2012; Hsu *et al.*, 2012] founded on ground-based and satellite data which indicated that AOD has decreased across the United States and in North America over the last decade. The declining AOD trends are most likely the result of effective air quality regulation in the United States known as the Clean Air Act, first adopted in 1970 and significantly amended in 1990. The latest air quality status report of the

Environmental Protection Agency (EPA), which is responsible for enforcing the Act and monitoring its outcomes, acknowledged the success of this regulation at a national scale by reporting a 50% decrease in SO_2 concentration, a precursor of SU aerosols, between 2001 and 2010 [EPA, 2012]. Hand *et al.* [2012, 2013] analyzed ground-based sulfate concentration across the U.S. from the IMPROVE (rural) and CSN (urban/suburban) air quality networks between 2000 and 2010. They reported a decrease at nearly all stations and in all regions of the U.S.; urban and rural concentrations decreased at a similar rate. Even more so, Hand *et al.* [2012] reported that sulfate concentration has decreased much more substantially over the eastern U.S. than over the western U.S., while Hand *et al.* [2013] and Xing *et al.* [2012] reported that the reduction in SO_2 emissions is the highest over the eastern U.S. as well. They also compared these data with SO_2 emissions obtained from the EPA's National Emissions Inventory and found a positive correlation between SO_2 emissions and SU concentration. The IPCC reported a 24% decrease in SO_2 emissions between 2000 and 2010 in the U.S. [de Meij *et al.*, 2012]. Granier *et al.* [2011] conducted a review of different inventories of anthropogenic and biomass burning emissions on both global and regional scales between 1980 and 2010. For SO_2 emissions in the U.S., the inventories agree well with decreasing trends since 2000. While analyzing air quality trends in New York City, Buckley and Mitchell [2011] found some significant decrease in both SO_2 concentration and $PM_{2.5}$ concentration between the years 2000 and 2007.

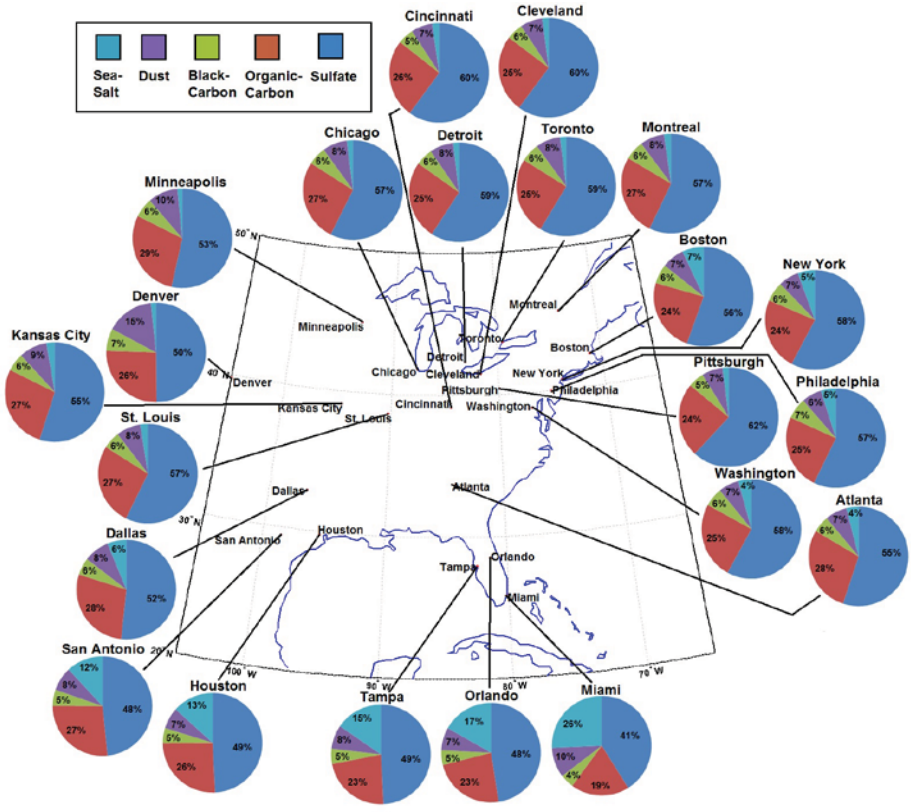

Fig. 2. Cities in eastern and central United States and Canada with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Although southern Ontario has its own active industrial sector, it is documented that air advection from the United States to Canada leads to poor air quality when meteorological conditions are appropriate [Chung, 1978]. In this respect, it is possible to link decreased AOD values in north-eastern U.S. cities to decreased AOD values in Canadian cities in proximity, such as Toronto and Montréal. As well, reports from both Ontario's Ministry of the Environment [2013] and Québec's Ministère du Développement Durable, de l'Environnement, de la Faune et des Parcs [MDDEFP, 2011] indicated a substantial drop in SO_2 emissions in their respective province since the year 2002.

3.2 Western United States and Canada

For cities in this part of the world, SU aerosols account for most of total AOD, as shown in Fig. 3. The second biggest contribution is from OC, which accounts from a quarter to a third of total AOD. DU aerosols account for a significant portion of total AOD, especially in Phoenix, located in a particularly arid area.

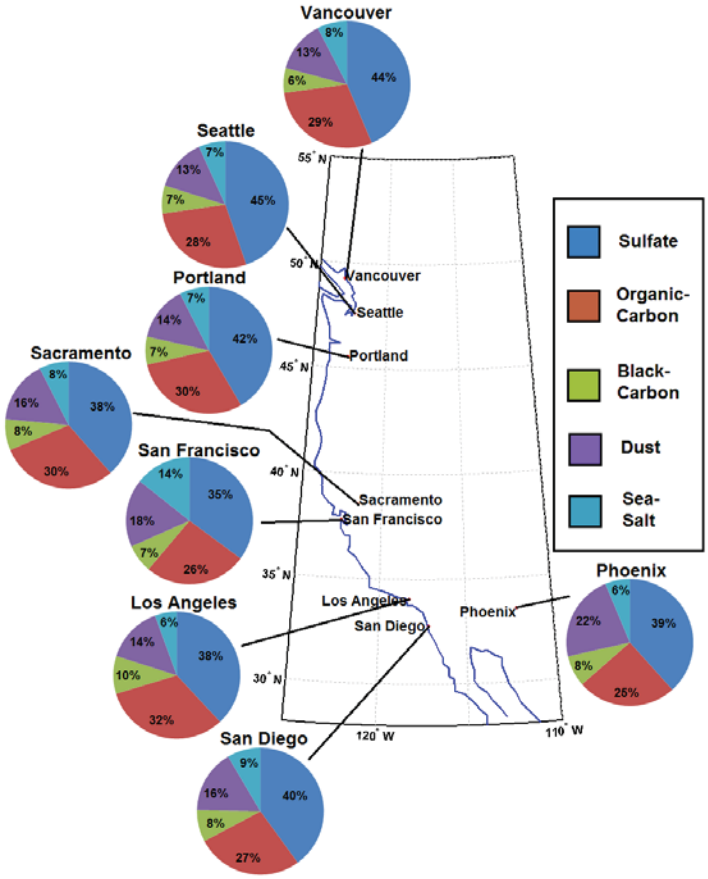

Fig. 3. Cities in western United States and Canada with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Five of the eight cities listed in Table 8 have increasing total AOD trends. This region was also targeted by air quality regulations, and therefore arguments presented in Section 3.1 should apply here as well. In some cases, the total AOD increase is entirely attributed to an increase in OC AOD. The most probable cause could be increasing wildfire activity in this region over the last decade. Wildfire activity is measured by two variables: the number of wildfires and the area burned. The former isn't a reliable metric to measure carbon emissions because the relative size of wildfires is an important factor to consider. As noted by Miller and Safford [2012], wildfires 400 ha or larger account for only 5% of all wildfires in the western U.S. since 1984, but are responsible for 95% of total area burned. The latter is the more reliable metric but still insufficient because the type and density of burning biomass affect the amount of carbon released. Spracklen *et al.* [2007], who modeled OC emissions from summer wildfires in the western U.S. between 1980 and 2004, found some correlation between the area burned and OC emissions. However, varying wildfires in different ecosystems is the reason why these two variables don't always correlate. They arrived at the conclusion that variability of OC concentration in the western U.S. was largely due to the variability in wildfire emissions. It is worth noting, however, that their study period barely overlaps with our own, so we cannot extrapolate their findings to our results. Nevertheless, they offer a likely explanation that justifies the positive OC AOD trends in Table 8. Since wildfires usually occur in forested (remote) areas, the extent to which they affect air quality in cities is dependent on regional meteorology. This could explain why some cities in Table 8, such as San Francisco and San Diego, were not affected by wildfire emissions.

Table 8. List of cities in western United States and Canada, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust.

State or province	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
British Columbia	Vancouver	0.122	10.2	6.3	21.1	8.5	8.5	2.6
Washington	Seattle	0.125	5.9	4.3	8.3	3.4	14.8	3.3
Oregon	Portland	0.121	5.5	3.7	6.0	2.8	17.6	4.9
California	Sacramento	0.116	1.5	-4.0	9.2	3.8	-9.3	5.6
	San Francisco	0.108	-2.1	-2.2	-4.7	-0.9	-6.1	5.2
	Los Angeles	0.126	8.0	-5.6	31.9	9.4	-3.5	2.1
	San Diego	0.114	-6.9	-5.8	-13.4	-8.9	-4.9	1.9
Arizona	Phoenix	0.130	-0.8	-3.7	-6.3	-0.6	11.0	7.7

3.3 Mexico and Central America

Total AOD values over cities in this region are listed in Table 9. The three Mexican cities, as well as the sole continental Central American city, Guatemala City, have the most important contribution from SU aerosols. OC aerosols account for 27.4% to 33.8% of total AOD (Fig. 4). Cities in the Caribbean have a more heterogeneous distribution of aerosol species, with SS, DU and SU aerosols being the most important.

Table 9. List of cities in Mexico and Central America, with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust.

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Mexico	Monterrey	0.139	-3.6	-9.2	4.8	4.2	-1.5	-1.8
	Guadalajara	0.137	-9.4	-11.6	-7.5	-5.0	-4.8	-8.2
	Mexico City	0.143	-6.4	-9.5	-2.5	2.4	-4.3	-9.2
Guatemala	Guatemala City	0.161	1.0	4.9	0.4	-5.1	-11.0	-23.0
Haiti	Port-au-Prince	0.110	-6.7	-4.4	-12.4	-4.8	-2.7	-11.2
Dominican Rep.	Santo Domingo	0.141	-6.2	-4.8	-16.6	-6.6	-2.3	-9.4
Puerto Rico	San Juan	0.129	-2.4	-4.7	-11.7	-6.9	3.5	-6.5

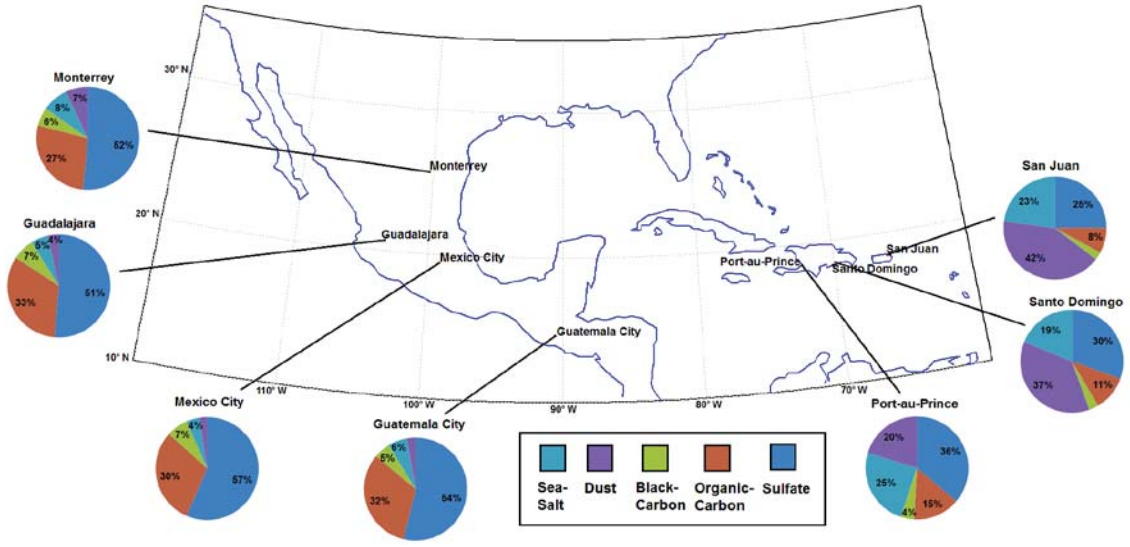

Fig. 4. Cities in Mexico and Central America with proportion of aerosol species in the MERRAero 10-year average total AOD.

The trends in total AOD over all the cities are decreasing, except for the trend over Guatemala City which showed a slight increase of 1.0% (Table 9). In Mexican cities, the decrease in total AOD is attributed to the decrease in SU AOD. Much like the United States government, the Mexican government implemented management programs and incentives to improve air quality in the 1980's. This effort has proven to be fairly effective, as the concentration of SO_2 and that of particulate matter have decreased in Mexico City since then [Molina and Molina, 2004a, b]. Parrish *et al.* [2011] showed that the trend in SO_2 concentration keeps decreasing until 2010.

The decline in total AOD over the three Caribbean cities is due to a decrease of DU, OC and SU aerosols AOD. The dust load is the result of transport from the Sahara desert. The only

locations where dust aerosols are monitored in the Caribbean are Barbados and Puerto Rico. Prospero and Mayol-Bracero [2013] published a portion of this record, for Barbados, between 2004 and 2009. The seasonal variability of dust load is quite large, but overall, there seems to be a slight decrease over the period. Unfortunately, according to Prospero and Mayol-Bracero [2013], the meteorological dynamics that drive dust transport are not well understood.

3.4 South America

For the distribution of aerosol species, Caracas is the city with the most heterogeneous distribution (Fig. 5). Since it is located close to the Caribbean Sea and quite far from all the other major cities in South America, SS aerosols account for 30.0% of its total AOD. SU and OC aerosols AOD account together for 50.8% of the total. Santiago is the only city west of the Andes and the only one where SU aerosols account for the majority of total AOD. The three north-most cities in Brazil (Salvador, Fortaleza and Belém), located on the eastern coast of the country, have some significant contribution from SS aerosols. All other cities between Buenos Aires and Brasilia have the significant contribution to total AOD from OC aerosols. In most of them, OC is the major contributor to total AOD while SU is second in importance (Fig. 5). Deforestation and biomass burning for agricultural purposes in the Amazon rainforest could be responsible for such a substantial presence of OC aerosols in the air.

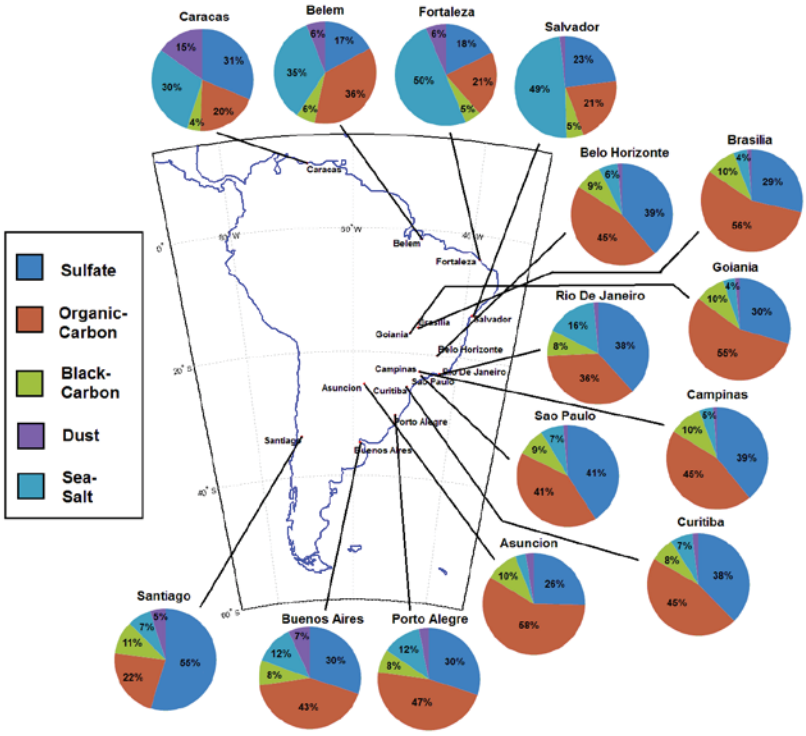

Fig. 5. Cities in South America with proportion of aerosol species in the MERRAero 10-year mean total AOD.

All the cities in the eastern part of South America experienced a declining trend in total AOD during the 10-year study period (Table 10). Moreover, with rare exceptions, all aerosol species exhibit declining (negative) AOD trends. Fortaleza is the only city with an increase (0.5%) in OC AOD, and Asunción is the only city with an increase (1.0%) in SU AOD (Table 10).

Table 10. List of cities in South America, with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Venezuela	Caracas	0.134	-9.5	-2.3	-17.5	-12.5	-12.4	-6.0
Brazil	Belém	0.141	-10.6	-28.9	-15.4	-11.2	5.4	-9.5
	Fortaleza	0.116	-8.9	-25.8	0.5	1.8	-7.2	-8.7
	Salvador	0.106	-7.1	-16.1	-5.4	2.1	-4.6	1.3
	Brasilia	0.107	-12.1	-13.4	-12.6	-4.8	-12.1	-12.1
	Goiânia	0.117	-9.9	-8.6	-11.0	-6.2	-11.8	-13.4
	Belo Horizonte	0.101	-13.8	-9.7	-18.4	-8.0	-11.7	-10.6
	Rio de Janeiro	0.139	-9.0	-3.2	-17.0	-3.2	-7.1	-3.8
	Campinas	0.140	-9.1	-3.9	-14.0	-6.0	-9.5	-8.9
	São Paulo	0.143	-9.4	-4.3	-14.9	-5.2	-10.5	-6.5
	Curitiba	0.127	-13.3	-4.3	-21.3	-12.4	-9.6	-1.0
	Porto Alegre	0.164	-8.5	-5.9	-13.8	-7.5	2.1	7.1
Paraguay	Asunción	0.200	-7.3	1.0	-11.7	-7.0	4.4	0.6
Argentina	Buenos Aires	0.162	-0.8	-4.7	-6.1	0.7	13.8	27.4
Chile	Santiago	0.095	4.3	4.4	-2.4	5.5	16.7	15.1

Previous publications support our findings on the declining AOD trends in South America. According to de Meij *et al.* [2012], between 2000 and 2009, ground-based observations from stations spread across South America showed a decline in total AOD over this region. Between 1997 and 2009, van der Werf *et al.* [2010] modeled carbon mass emissions from wildfires in South America by using active fire data, area burned data and vegetation productivity indices from satellite. They detected some decrease in mass emissions from wildfires between 2002 and 2009 (their Table 7 and Fig. 9). However, Cifuentes *et al.* [2005] discussed the fact that air quality data in Latin America is scarce, lacking, incomplete, outdated, unreliable or difficult to access. This complicates the analysis and explains why most papers on this topic use remote sensing instruments. Further research is needed to understand the probable reasons for the obtained declining AOD trends.

3.5 Northern Africa

The standout feature of northern Africa is obviously the Sahara desert. Desert DU aerosols account for the most important contributor to total AOD in all the cities in this region (Fig. 6). In

the cities located in immediate proximity to the desert (Cairo, Khartoum, Kano, Adis Abeba and Dakar), the contribution of DU to total AOD exceeds 70% in some of them. SU aerosols are the second most important contributor to total AOD in the four cities on the northern coast, while the four coastal cities near the equator (Abidjan, Accra, Lagos and Ibadan) experience a significant contribution from OC aerosols to total AOD.

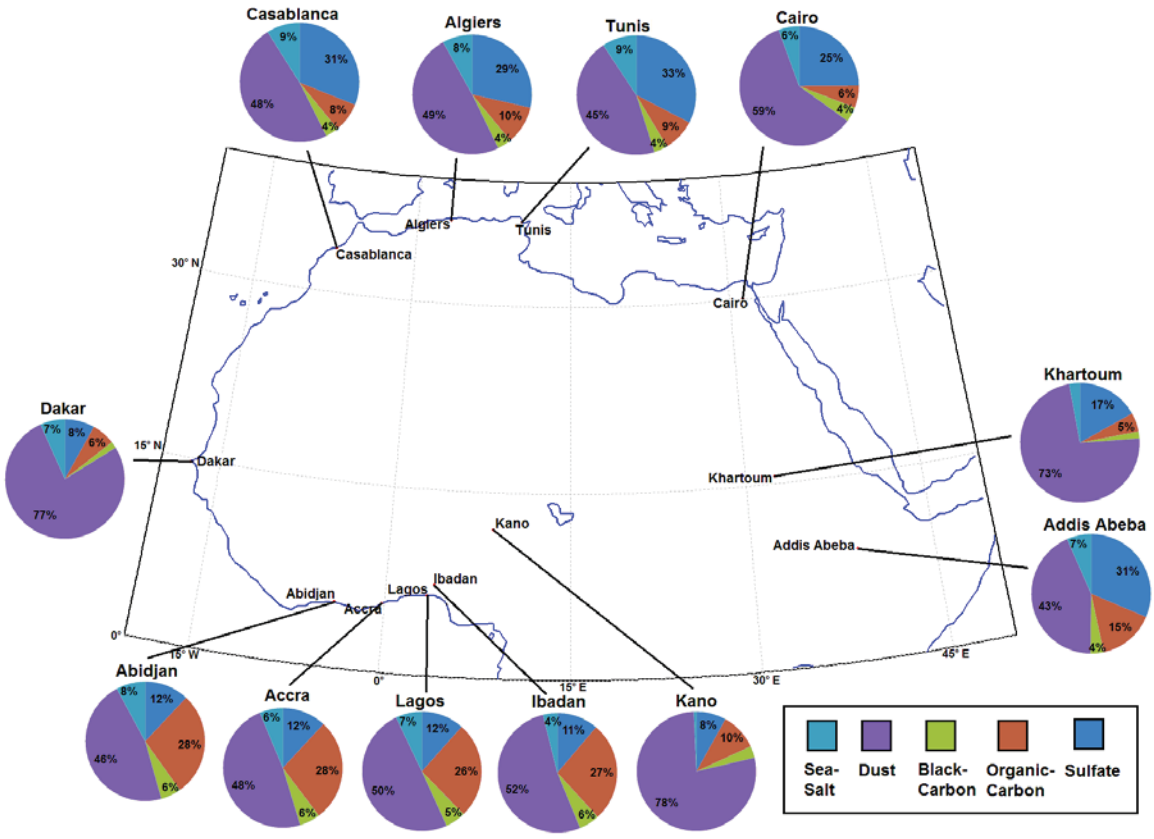

Fig. 6. Cities in northern Africa with proportion of aerosol species in the MERRAero 10-year mean total AOD.

As shown in Table 11, in the cities located in immediate proximity to the desert (Cairo, Khartoum, Kano, Adis Abeba and Dakar), the increasing trend in total AOD is mostly driven by the increasing trend of DU AOD. The cities near the equator saw mainly a decrease in total, DU and OC AOD; the cities in the vicinity of the desert (Cairo, Khartoum, Kano, Adis Abeba) saw an increase in total and DU AOD; the northwestern coastal cities saw mainly a decrease in total, DU and SU AOD.

It is important to mention that MERRAero doesn't assimilate AOD data over bright surfaces such as deserts. Therefore, over the source region, the data is constrained primarily by parameterized emissions determined by wind speed. However, the darker surface of cities might have been sufficient to provide assimilated data in and around them.

Table 11. List of cities in northern Africa, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Morocco	Casablanca	0.178	-1.9	0.3	-15.6	-4.2	1.0	-1.1
Algeria	Algiers	0.190	-9.4	-0.8	-0.8	1.0	0.4	-17.9
Tunisia	Tunis	0.200	-10.7	-5.6	-13.7	-4.1	-7.5	-14.7
Egypt	Cairo	0.257	2.4	2.8	-2.3	3.0	-0.7	3.0
Senegal	Dakar	0.411	0.5	-3.0	-16.3	-11.0	5.7	2.3
Côte d'Ivoire	Abidjan	0.300	-9.3	-21.4	-5.2	-8.0	12.6	-11.8
Ghana	Accra	0.353	-8.6	-21.5	-5.0	-7.1	15.4	-10.1
Nigeria	Lagos	0.394	-6.1	-19.7	-3.1	-5.3	12.4	-6.9
	Ibadan	0.406	-3.2	-13.2	1.4	-4.0	24.1	-5.0
	Kano	0.485	2.2	6.4	0.5	-3.7	13.9	2.2
Sudan	Khartoum	0.396	7.0	11.4	8.5	4.6	8.5	6.0
Ethiopia	Addis Abeba	0.164	15.5	5.7	15.5	9.1	17.4	23.6

3.6 Southern Africa

AOD values for cities in this part of the world are listed in Table 12. South Africa, being the most industrialized country in this region, is where SU aerosols account for the largest proportion of total AOD (Fig. 7). The two coastal cities of Cape Town and Durban are exceptions with a significant contribution from SS aerosols. Total AOD for the other cities further north is dominated by OC aerosols, except for Dar es Salaam located on the Indian Ocean coast.

Table 12. List of cities in southern Africa, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Congo	Kinshasa	0.344	-9.0	-52.4	9.4	12.3	16.5	-24.2
Angola	Luanda	0.350	-1.9	-38.2	6.6	7.8	26.8	-4.6
Kenya	Nairobi	0.090	3.4	-21.2	14.3	6.8	17.7	29.4
Tanzania	Dar es Salaam	0.129	-1.2	-27.4	10.9	9.8	1.3	30.0
Zimbabwe	Harare	0.116	-2.9	-13.6	2.8	8.3	1.4	4.2
South Africa	Pretoria	0.163	2.4	-2.8	8.0	10.9	0.1	12.1
	Johannesburg	0.163	1.8	-2.4	6.5	9.6	0.4	11.5
	Durban	0.180	-1.4	-4.5	1.2	3.3	-1.2	6.0
	Cape Town	0.085	-1.9	-6.5	-6.2	-1.1	7.1	2.1

Total AOD trends in this region are rather weak, except for the 9.0% decrease in Kinshasa (Table 12). Much like South America, the predominance of OC aerosols in the air could be

explained by biomass burning in the African savannah and rainforests during the burning season (May to October). Although extensive fire data are available in six protected and confined areas in southern Africa [Archibald *et al.*, 2010], forest cover and loss statistics are generally lacking [Mercier, 2012], as well as fire data [Ito *et al.*, 2007]. This leaves only remote sensing and model output as means to study this issue broadly. Archibald *et al.* [2009] claimed that human activities are the main sources of ignition in this region, as opposed to natural causes, but also affirmed that increased human activities doesn't necessarily imply an increase in burned area, most likely because climatic factors determine to which extent ignited fires will burn [Archibald *et al.*, 2010]. Although forest preservation policies have been implemented in Africa, Mercier [2012] calls them a failure and assures that deforestation is ongoing; from 2005 to 2010, the deforestation rate in Africa was the second highest among all continents, behind Central America. van der Werf *et al.*'s [2010, their Table 7 and Fig. 9] modeled carbon emission results suggested slight increasing trends in OC between 2002 and 2009. All of these findings support the OC AOD trends shown in Table 12.

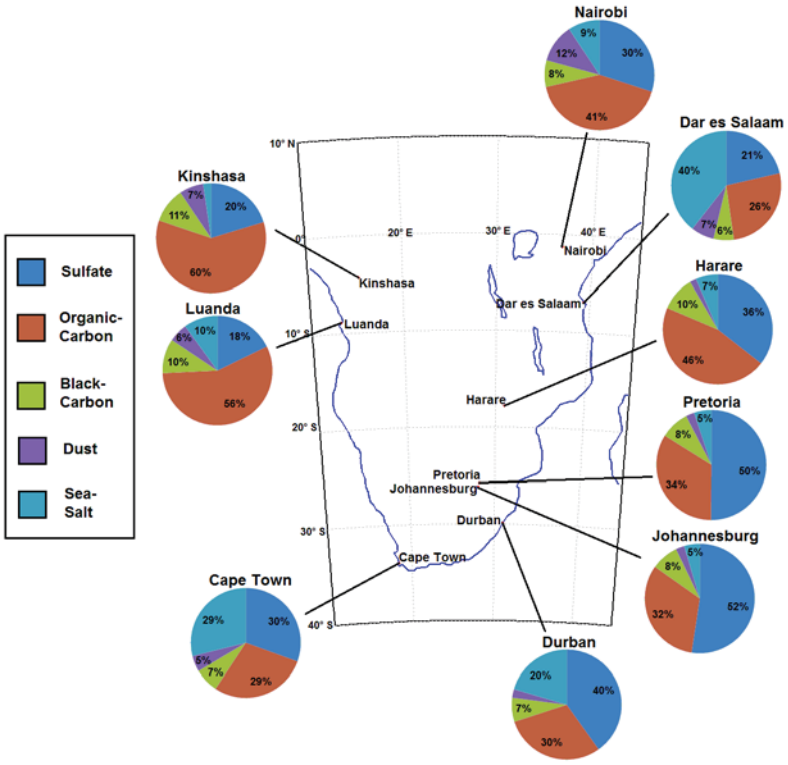

Fig. 7. Cities in southern Africa with proportion of aerosol species in the MERRAero 10-year mean total AOD.

3.7 Western Europe

The situation in western Europe is similar to that in North America. As seen in Fig. 8, SU aerosols are the most important aerosol species there. Over cities located in southern Europe,

DU aerosols are the second most important contributor to total AOD (Fig. 8). This is in line with Querol *et al.* [2009], who analyzed observations of Saharan dust transport over the Mediterranean.

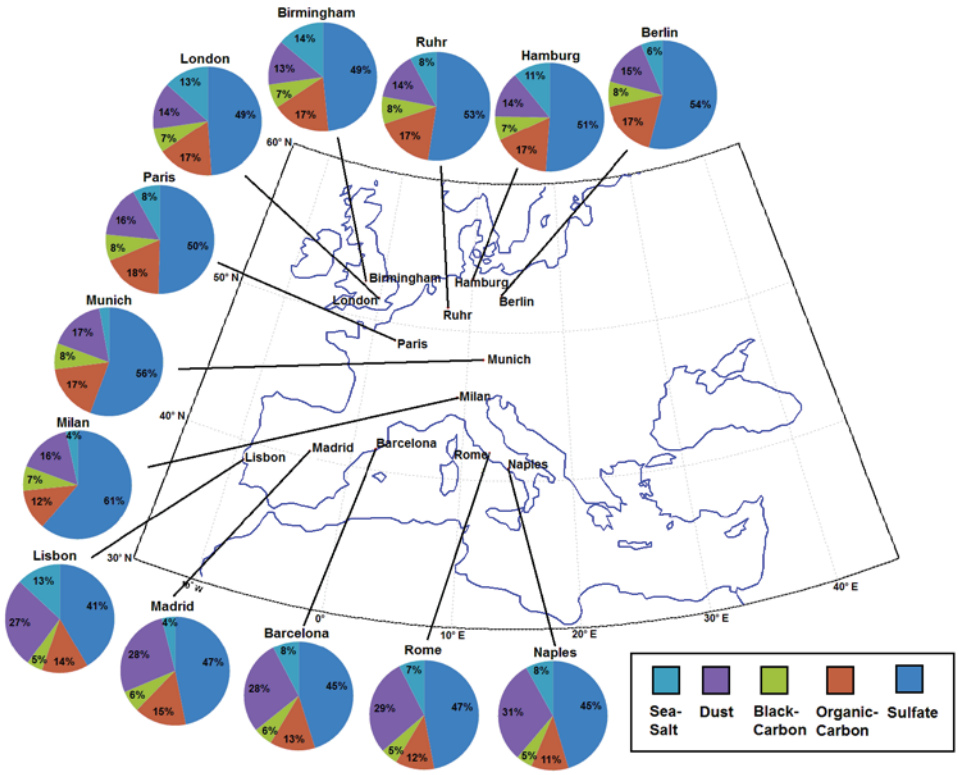

Fig. 8. Cities in western Europe with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Cities in this region saw mainly decreasing trends for total AOD and for every aerosol species (except for Lisbon and Munich's slight SU AOD increases of 0.6% and 1.7% respectively) (Table 13). This is in line with de Meij *et al.* [2012] and Hsu *et al.* [2012], who found that ground-based and satellite-based remote sensing observations showed decreasing AOD trends over Europe as a whole. The European Environmental Agency [EEA, 2013] reported that implementation of air quality policies and greener fuels and technology contributed to a 55% reduction of SO_2 emissions between 2000 and 2011 among all members of the European Union. SO_2 emissions data are shown in Fig. 9 for the six countries listed in Table 13. A near constant decrease is noticeable for all of them. Emissions of total suspended particulates also decreased by 14% during the same period. Numbers from the IPCC for the whole of Europe are more conservative (20% decrease between 2000 and 2010 for SO_2 emissions [de Meij *et al.*, 2012]), but substantially decreasing nonetheless. Granier *et al.*'s [2011] emissions inventory revue claimed that most inventories agree with a decreasing trend since 2000 as well. Cusack *et al.* [2012] analyzed data from many monitoring stations across Europe also found decreasing $PM_{2.5}$ concentration trends over the last decade.

Table 13. List of cities in western Europe, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Portugal	Lisbon	0.131	-7.8	0.6	-24.0	-10.4	-1.2	-13.7
Spain	Madrid	0.108	-9.5	-3.5	-15.7	-7.5	-8.4	-16.3
	Barcelona	0.148	-9.4	-2.7	-13.0	-3.6	-3.2	-20.2
United Kingdom	Birmingham	0.126	-10.2	-5.7	-24.2	-10.3	-6.9	-10.2
	London	0.132	-9.1	-5.1	-20.5	-9.1	-6.0	-11.4
France	Paris	0.143	-7.1	-3.3	-16.8	-5.5	-6.8	-8.4
Germany	Ruhr	0.149	-6.8	-1.4	-20.0	-6.9	-10.7	-6.3
	Hamburg	0.145	-8.4	-4.5	-21.7	-9.9	-4.1	-7.7
	Berlin	0.152	-7.2	-1.6	-19.6	-8.8	-10.1	-9.7
	Munich	0.143	-2.5	1.7	-9.4	-3.1	-11.9	-6.8
Italy	Milan	0.223	-7.6	-6.1	-16.1	-6.1	-8.1	-6.7
	Rome	0.165	-6.8	-3.5	-17.6	-6.4	-6.4	-7.8
	Naples	0.177	-4.7	-2.1	-14.6	-5.4	-2.2	-5.2

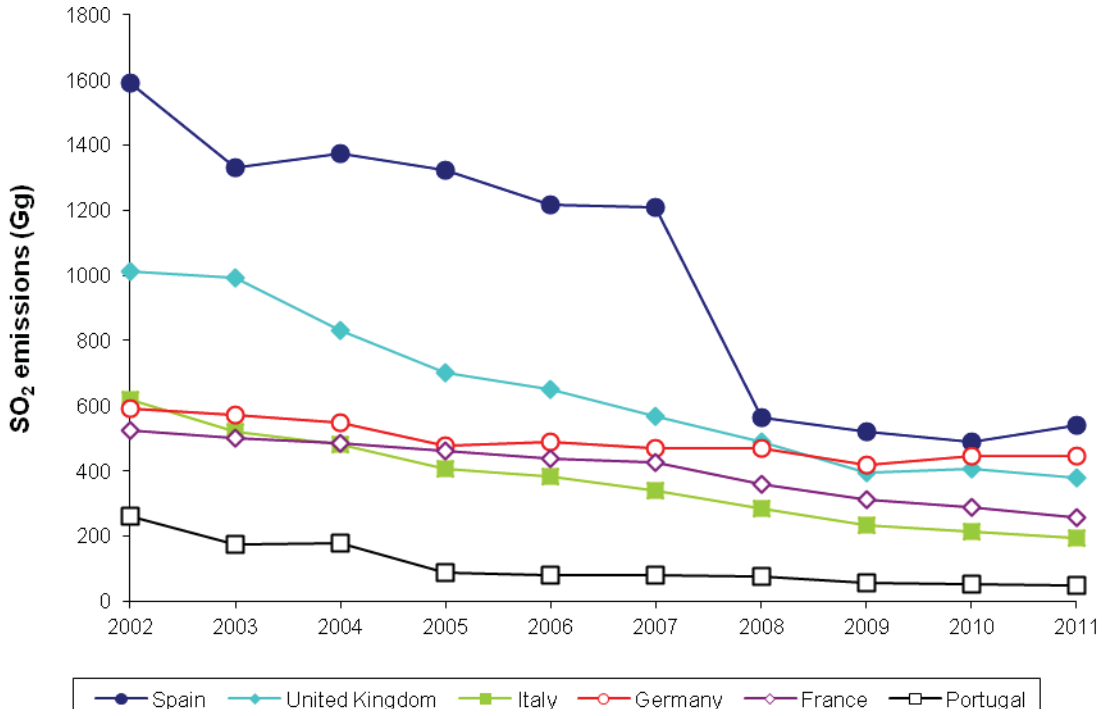

Fig. 9. SO_2 emissions in western European countries. Data is from the EEA: 2002-2004 data is from the 2012 report; 2005-2011 data is from the 2013 report.

Air quality regulations most likely helped decrease carbon aerosol loads as well, but the decrease of OC AOD observed in Table 13 clearly stands out in most cities. Reduced wildfire

activity would explain such a decrease. Note that year-to-year variability is high and extraordinary wildfire activity in a given season can drag the trend considerably. This appears to have been the case in the exceptionally warm and dry summer of 2003, when raging wildfires in Portugal, southern France, Italy and the eastern coast of the Adriatic sea, combined with stagnant weather conditions, led to high aerosol load and high AOD values (> 0.3) in the Mediterranean basin during August [Hodzic *et al.*, 2007]. AOD values were also relatively high in urban centers of northern Europe during August due to accumulated urban pollutants and smoke dust from the wildfires [Hodzic *et al.*, 2007]. Advection of smoke from Portugal affected areas in Great Britain, France and Germany where AOD values above 0.8 were occasionally recorded from satellites [Vautard *et al.*, 2007]. The evolution of our OC AOD results for selected cities during the period from 2003 to 2011 is shown in Fig. 10. The peak in OC AOD in the year 2003 is clearly seen.

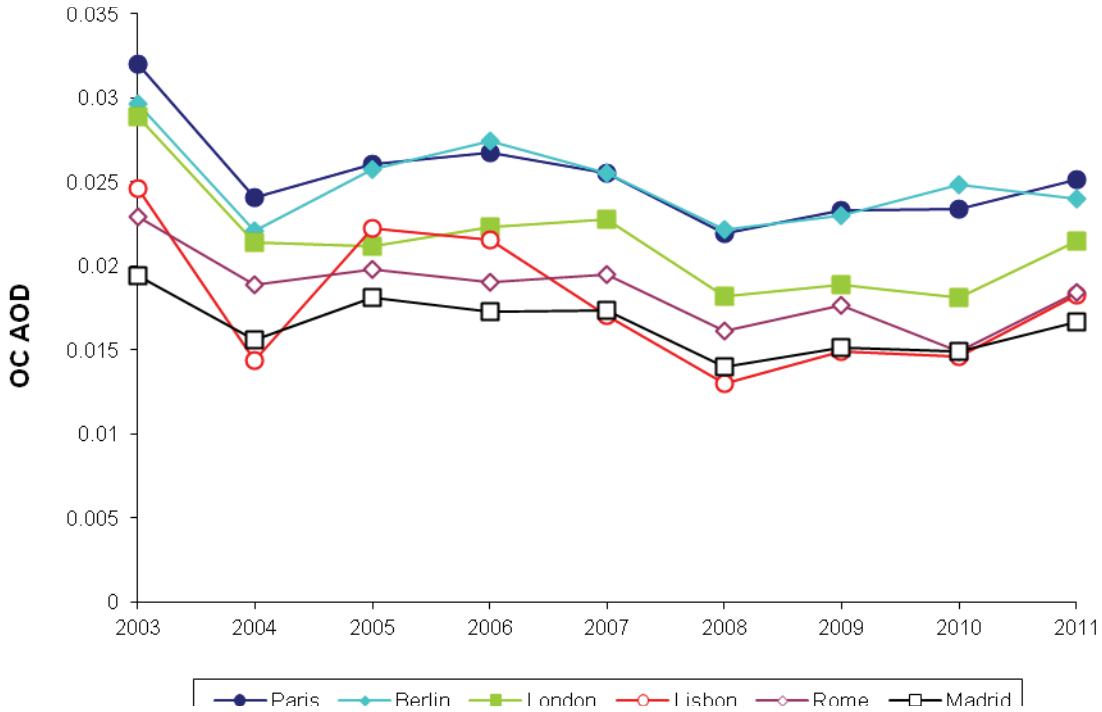

Fig. 10. Evolution of MERRAero yearly mean OC AOD for a selection of cities in western Europe.

By analyzing African dust outbreak trends at different locations in Spain, France, Italy and Greece (Greece is covered in Section 2.8) between 2001 and 2011, Pey *et al.* [2013] concluded that northeastern Spain, southeastern France and northern Italy experienced a decreasing trend from 2006 onward, explained by a negative shift of the North Atlantic Oscillation index during the summer since then. Their results support our findings on decreasing DU AOD trends based on MERRAero data (Table 13).

3.8 Eastern Europe, including Russia and Turkey

For the majority of cities in this region, SU aerosols are the main contributor to total AOD (Fig. 11). There are exceptions, however, such as Athens and the three Turkish cities which have a significant contribution from DU aerosols. This indicates that dust transport from the Sahara to eastern Europe could be more significant than that from the Sahara to western Europe, which is in line with Querol *et al.* [2009].

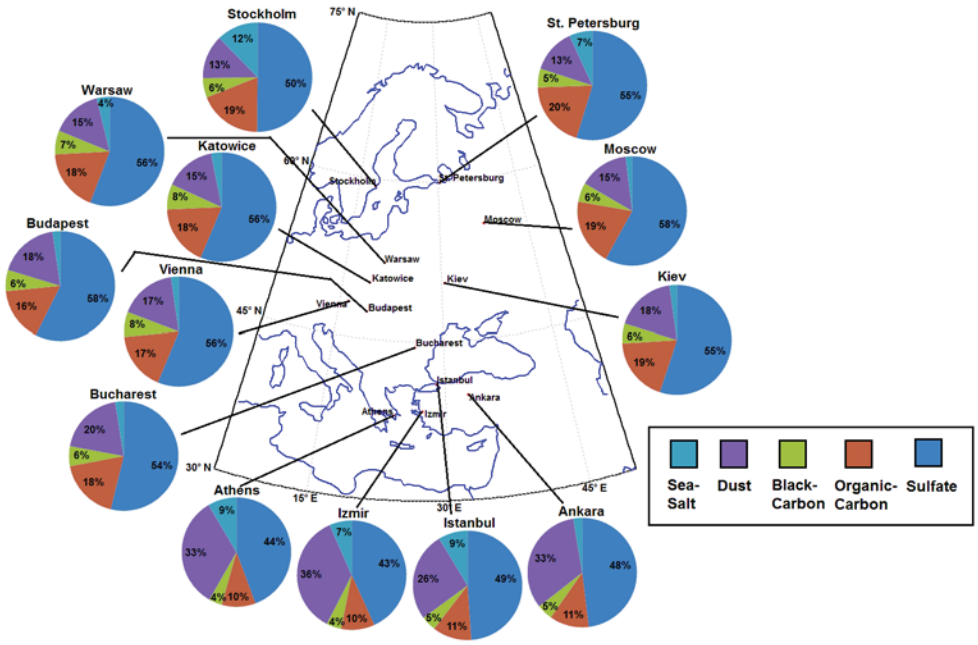

Fig. 11. Cities in eastern Europe with proportion of aerosol species in the MERRAero 10-year mean total AOD.

As shown in Table 14, in the majority of cities, total AOD trends were insignificant (between -3% and 3%). Exceptions are the increasing trend in Moscow (8.2%) and Katowice (5.6%), and the decreasing trend in Budapest (6.4%) and Vienna (4.9%). There was a strong decrease in OC AOD for all cities except Moscow. Athens and Budapest are the cities with decreasing SU AOD trends (Table 14), while in Katowice, Bucharest and Vienna, SU AOD trends were insignificant.

Although the wildfire season of 2003 wasn't as dramatic in Eastern Europe compared to its western counterpart (see Section 3.7), advection of smoke to this region and the severe weather conditions likely had an impact on the carbon aerosol load. Model simulations by Mues *et al.* [2012] reproduced an abnormally high concentration of PM_{10} over most of Europe in the summer of 2003, compared to the average summer concentration for the years between 2003 and 2007. Mues *et al.* [2012] concluded that a combination of warm temperatures, low wind speed and little precipitation was sufficient to produce prolonged episodes of air pollution. Adding carbon emissions from the wildfires would only worsen this matter.

Table 14. List of cities in eastern Europe, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Sweden	Stockholm	0.123	-1.2	4.3	-16.6	-6.8	-0.8	4.3
Poland	Warsaw	0.173	-3.8	2.8	-18.8	-7.1	-13.9	-3.5
	Katowice	0.175	-5.6	0.2	-18.2	-6.8	-14.3	-8.2
Austria	Vienna	0.169	-4.9	0.5	-15.4	-6.7	-11.1	-9.5
Hungary	Budapest	0.187	-6.4	-2.1	-17.5	-7.7	-8.9	-8.5
Russia	St. Petersburg	0.139	2.7	7.3	-10.3	-2.2	-5.4	11.3
	Moscow	0.174	8.2	6.8	7.7	3.8	-7.4	18.6
Ukraine	Kiev	0.181	0.9	3.5	-9.2	-5.1	-5.3	7.3
Romania	Bucharest	0.225	-1.2	0.1	-5.8	-2.0	-6.6	0.3
Greece	Athens	0.189	-2.7	-2.5	-9.1	-3.0	-1.3	-1.2
Turkey	Istanbul	0.214	-1.0	-0.1	-10.6	-4.9	2.7	1.4
	Ankara	0.145	0.9	4.4	-5.0	-0.4	0.7	-1.7
	Izmir	0.181	-0.9	0.2	-10.6	-2.0	2.1	0.2

The eastern-most city in this region, Moscow, has an increasing OC AOD trend (Table 14). Western Russia was affected by its own severe wildfire season during the summer of 2010 [Konovalov *et al.*, 2011]. Indeed, average AERONET AOD observations in Moscow during July and August 2010 were above 1 for many days and reached a daily average peak of 3.6 on August 7, while observed concentrations of combined black and organic carbon were over 500 µg per kg of air during many days as well [Huijnen *et al.* 2012]. As shown in Fig. 12, the evolution of MERRAero data for Moscow during the period from 2003 to 2011 was able to reproduce the strong OC AOD peak in 2010. This peak, however, is probably underestimated. Occasionally, the smoke from the wildfires was so thick that MODIS incorrectly identified it as clouds [van Donkelaar *et al.*, 2011].

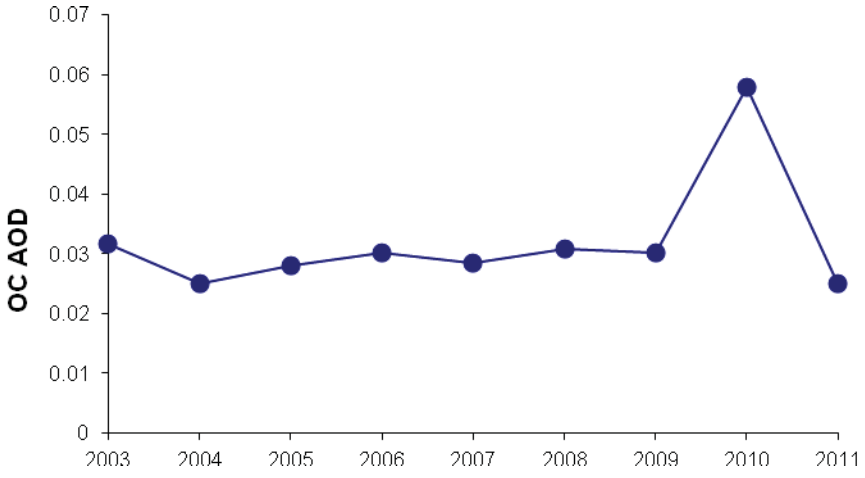

Fig. 12. Evolution of MERRAero yearly mean OC AOD in Moscow.

3.9 Western Asia

This region includes all Asian cities west of India and China. Their AOD values are listed in Table 15. As shown in Fig. 13, DU aerosols account for most of total AOD due to dust transport from deserts in Saudi Arabia and the Middle East. SU aerosols are the second most important species. All the cities in Table 15 showed increasing trends in total AOD during the 10-year study period, accompanied by increasing trends in DU AOD.

Table 15. List of cities in western Asia, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Yemen	Sana'a	0.254	6.4	-4.8	2.3	2.1	-2.3	14.7
Saudi Arabia	Jeddah	0.372	13.8	0.7	2.7	3.9	12.3	17.7
Jordan	Amman	0.221	4.6	4.1	-2.0	2.0	-0.6	6.6
Israel	Tel Aviv-Jaffa	0.223	3.3	3.6	-2.3	1.4	0.5	4.6
Syria	Damascus	0.167	5.0	3.1	-4.6	2.0	-0.6	8.3
	Aleppo	0.204	2.5	-1.5	-4.3	1.3	-2.4	7.9
Iraq	Baghdad	0.396	10.4	1.0	-1.2	4.8	-2.0	14.8
Iran	Isfahan	0.202	9.0	3.4	1.3	1.4	10.7	13.3
	Tehran	0.238	7.8	3.5	-1.4	2.7	10.6	13.6
	Mashhad	0.218	12.6	8.5	-0.9	1.7	15.6	17.3
Uzbekistan	Tashkent	0.244	9.2	5.2	-11.5	-6.5	7.7	17.3
Afghanistan	Kabul	0.154	4.8	1.5	-13.5	-7.9	7.9	12.3
Pakistan	Rawalpindi	0.363	2.5	-4.4	-5.3	-0.3	0.1	9.8
	Gujranwala	0.482	2.0	-2.2	-0.8	2.2	-3.5	5.6
	Lahore	0.521	1.4	-1.6	0.5	2.8	-5.9	3.8
	Faisalabad	0.537	0.7	-3.0	-1.7	2.2	-3.9	3.3
	Hyderabad	0.449	2.9	2.3	4.9	4.6	-1.3	3.2
	Karachi	0.414	3.4	1.0	6.0	6.7	0.5	4.5

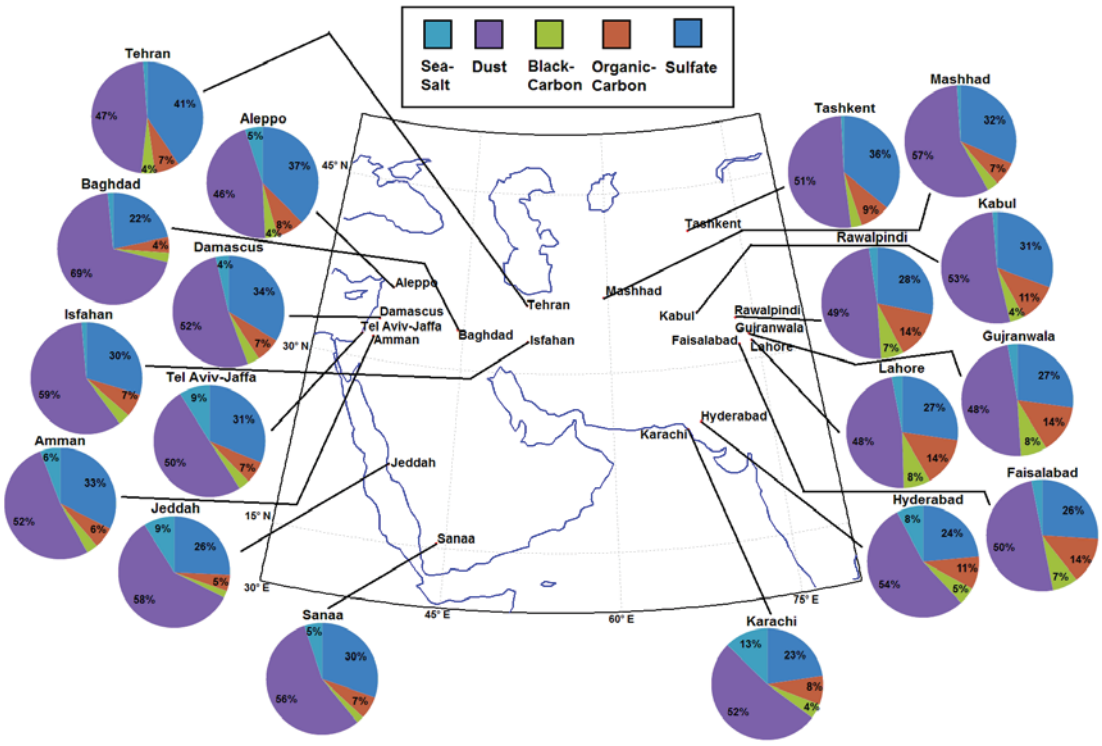

Fig. 13. Cities in western Asia with proportion of aerosol species in the MERRAero 10-year mean total AOD.

3.10 India and Bangladesh

The cities in the northwestern part have the largest contribution from DU aerosols to total AOD, while SU aerosols are the second most important contributor (Fig. 14). By contrast, SU aerosols are the dominant species in southern and northeastern cities, while DU aerosols are the second in importance.

We obtained increasing trends in total AOD in every single city listed in Table 16. Similarly, increasing trends were obtained for SU, OC and BC AOD. This is the probable result of industrial development and population growth in India and Bangladesh. Indeed, Lu *et al.* [2011] estimated that SO_2 emissions increased by 51% between 2000 and 2010 in India, black carbon by 35%, and organic carbon by 29%. In Bangladesh, the government deployed actions in order to reduce air pollution. However, these incentives seem to be failing since measurements of fine particulate matter mass concentration in Dhaka indicated an increase of 69% between the 2001-2002 period and the 2007-2009 period [Begum *et al.*, 2013].

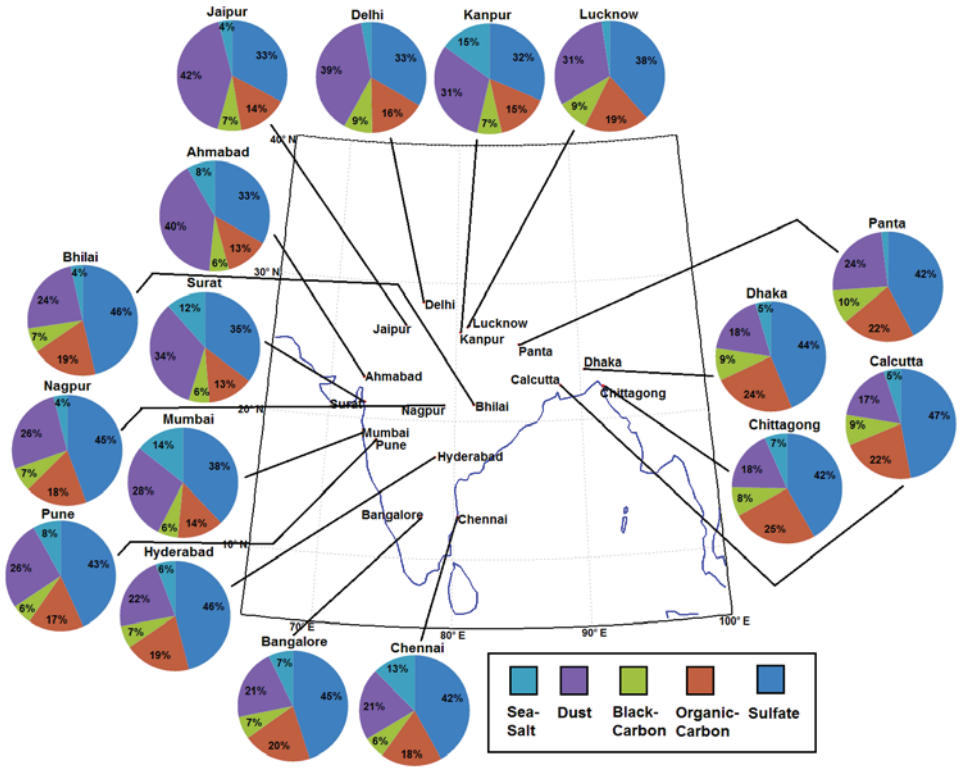

Fig. 14. Cities in India and Bangladesh with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Table 16 List of cities in India and Bangladesh, together with the 10-year mean total AOD values and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
India	Delhi	0.505	1.6	2.4	6.4	6.7	-8.3	-1.3
	Jaipur	0.355	4.4	5.4	10.0	8.6	-4.4	1.9
	Ahmadabad	0.360	6.9	7.3	12.4	10.4	-3.5	6.7
	Surat	0.343	8.9	11.0	15.7	14.1	-4.0	7.9
	Mumbai	0.336	11.4	12.2	18.4	16.5	-0.3	12.2
	Pune	0.276	12.5	11.1	19.4	16.6	0.2	13.9
	Lucknow	0.502	5.2	10.3	10.5	5.9	-11.5	-2.7
	Kanpur	0.502	6.1	10.3	11.6	7.5	-9.2	-1.1
	Bhilai	0.335	12.1	8.7	16.4	15.5	5.3	15.2
	Nagpur	0.323	14.2	12.2	19.0	18.5	2.0	15.2
	Hyderabad	0.305	11.8	7.6	16.4	17.2	3.8	17.3
	Bangalore	0.199	8.9	5.0	11.4	10.9	-3.9	19.8
	Chennai	0.272	8.2	5.6	10.1	12.8	-3.9	18.7
	Patna	0.503	9.3	14.0	13.6	7.4	-4.3	0.1
	Calcutta	0.431	11.8	11.4	14.4	13.1	7.4	10.4
Bangladesh	Dhaka	0.390	10.5	10.6	13.2	11.1	14.7	5.5
	Chittagong	0.292	9.0	8.0	11.7	11.8	13.2	5.1

3.11 China

China is the most populated country in the world and therefore has the highest number of cities in our list: a total of 33. As mentioned earlier, total AOD values for Chinese cities are among the highest in the world (Table 1). Unsurprisingly, SU aerosols account for the majority of total AOD: over 60% in most cities (Fig. 15).

Table 17. List of cities in China, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Province	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Heilongjiang	Haerbin	0.322	-0.6	3.8	-13.6	2.6	9.1	2.5
Jilin	Changchun	0.360	0.3	3.5	-11.1	4.2	5.5	2.5
Liaoning	Shenyang	0.360	1.6	5.7	-11.7	4.6	6.9	2.0
	Anshan	0.464	0.3	2.6	-11.1	3.1	-1.2	3.5
	Dalian	0.508	1.4	3.0	-9.7	5.0	2.6	4.6
Beijing	Beijing	0.550	4.8	8.5	-9.1	8.5	7.4	1.4
Tianjin	Tianjin	0.601	4.9	7.8	-8.3	8.0	6.5	3.5
Hebei	Shijiazhuang	0.580	4.9	6.3	-3.8	10.0	13.0	2.9
Shanxi	Taiyuan	0.393	4.7	6.2	-4.0	11.5	11.7	2.3
Shandong	Jinan	0.610	3.9	4.6	-3.5	10.2	-0.2	4.4
	Zibo	0.584	5.3	6.3	-2.0	10.8	-0.8	4.8
	Qingdao	0.552	3.4	3.7	-2.0	9.3	-4.3	5.3
Gansu	Lanzhou	0.304	0.8	0.6	-5.9	4,6	4.8	2.3
Shaanxi	Xi'an	0.511	2.3	0.7	3.2	9.2	25.7	3.1
Henan	Zhengzhou	0.706	1.9	1.1	-2.2	10.3	8.9	4.2
Jiangsu	Nanjing	0.655	6.8	6.5	6.5	12.9	-8.3	6.6
	Wuxi	0.613	7.5	7.4	7.7	12.8	-6.6	6.9
	Suzhou	0.600	7.4	7.3	8.1	12.6	-7.3	7.0
Shanghai	Shanghai	0.561	7.5	7.6	8.2	13.2	-6.6	7.7
Hubei	Wuhan	0.723	5.4	3.8	9.0	13.2	-4.4	5.0
Hunan	Changsha	0.719	3.3	1.4	8.1	10.1	-0.7	4.2
Jiangxi	Nanchang	0.654	3.6	2.2	7.1	9.4	-9.5	4.2
Zhejiang	Ningbo	0.490	5.1	5.2	6.5	10.8	-8.3	5.8
	Wenzhou	0.425	1.3	0.7	2.7	6.0	-6.7	5.5
Fujian	Fuzhou	0.436	-0.8	-1.3	-1.5	2.2	-0.1	5.0
	Quanzhou	0.410	-4.6	-4.7	-8.1	-1.1	-3.5	4.9
Guangdong	Shantou	0.642	5.5	4.9	4.3	13.4	-7.8	6.3
	Guangzhou	0.528	-8.8	-8.8	-12.6	-2.4	-9.7	5.1
	Shenzhen	0.452	-9.3	-8.3	-16.5	-4.8	-6.1	5.0
Hong Kong	Hong Kong	0.443	-9.0	-7.8	-16.7	-4.9	-5.0	4.9
Guangxi	Nanning	0.557	-2.9	-3.7	-4.4	3.6	-0.4	6.8
Yunnan	Kunming	0.203	-1.1	-2.4	-2.7	3.0	9.9	8.2
Xinjiang	Urumqi	0.227	6.3	-3.5	-16.1	1.8	18.3	21.3

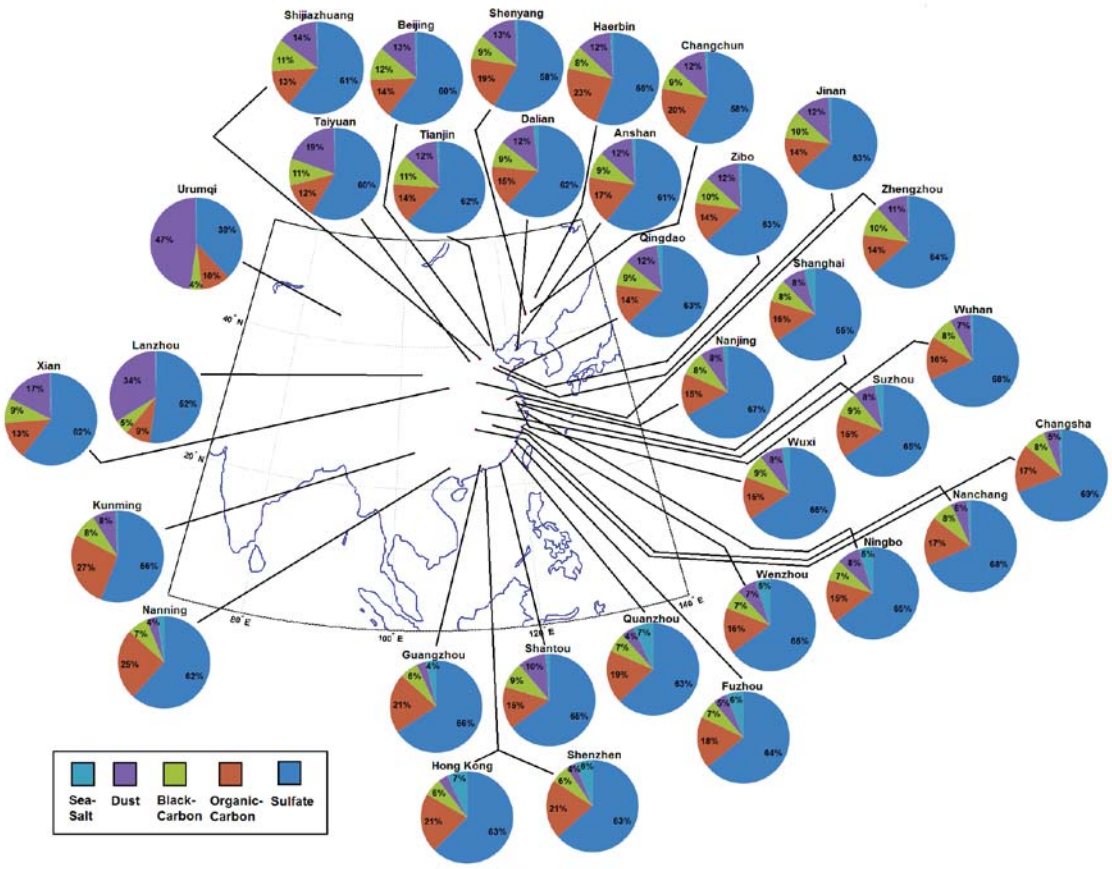

Fig. 15. Cities in China with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Total AOD trends are increasing in most cities which are usually accompanied by an increase in SU AOD (Table 17). The opposite is true for the cities with decreasing trends, all of which are located in the southeastern border and coast between Myanmar and the Taiwan Strait, except for Urumqi located farter to the northwest. OC AOD is decreasing in many cities.

Like India, China is emerging economically and is consuming consistently more coal since 2000 [Lu *et al., 2011*]. However, trends in total and SU AOD are generally weaker over cities in China than over cities in India. The reason for this is most likely due to emission patterns observed in China. Lu *et al.*'s [2010, 2011] estimates of Chinese SO_2 emissions have clearly risen between 2002 and 2010, overall by 28%. However, this is the result of a strong 38% increase from 2002 to 2006 (peak year) and a slow decrease since then to 2004 levels by 2010 (7% decrease between 2006 and 2010). Other estimates vary by numbers but all agree with peak emissions in 2006 and a slow decrease since then [Wang and Hao, 2012; Zhang *et al.*, 2012; Klimont *et al.*, 2013; Zhao *et al.*, 2013a; Zhao *et al.*, 2013b]. Implementation of national comprehensive policies by the Chinese government in 2005 has been successful in this respect. The Government also took advantage of international events, such as the 2008 Beijing Olympics, to temporarily and regionally push stricter air quality guidelines [Lin *et al.*, 2013].

The trend of SO_2 emissions is quite disproportionate throughout China. In order to evaluate our results for SU AOD trends, we compared them with Zhao *et al.*'s [2012a] and Zhao *et al.*'s [2013b] SO_2 emission estimate trends in Chinese provinces between 2005 and 2010. There is good agreement between these two publications and our results in the cities of Haerbin, Wuhan, Guangzhou, Shenzhen and Hong Kong. Given China's disproportionate SO_2 emissions in time as well, some cities with an overall increasing trend actually observed a decreasing trend between 2005 and 2010, which is consistent with Zhao *et al.* [2012a] and Zhao *et al.* [2013b]. This is indeed the case for the cities of Xi'an, Zhengzhou, Changsha and Wenzhou. The evolution of MERRAero SU AOD for these four cities is shown in Fig. 16.

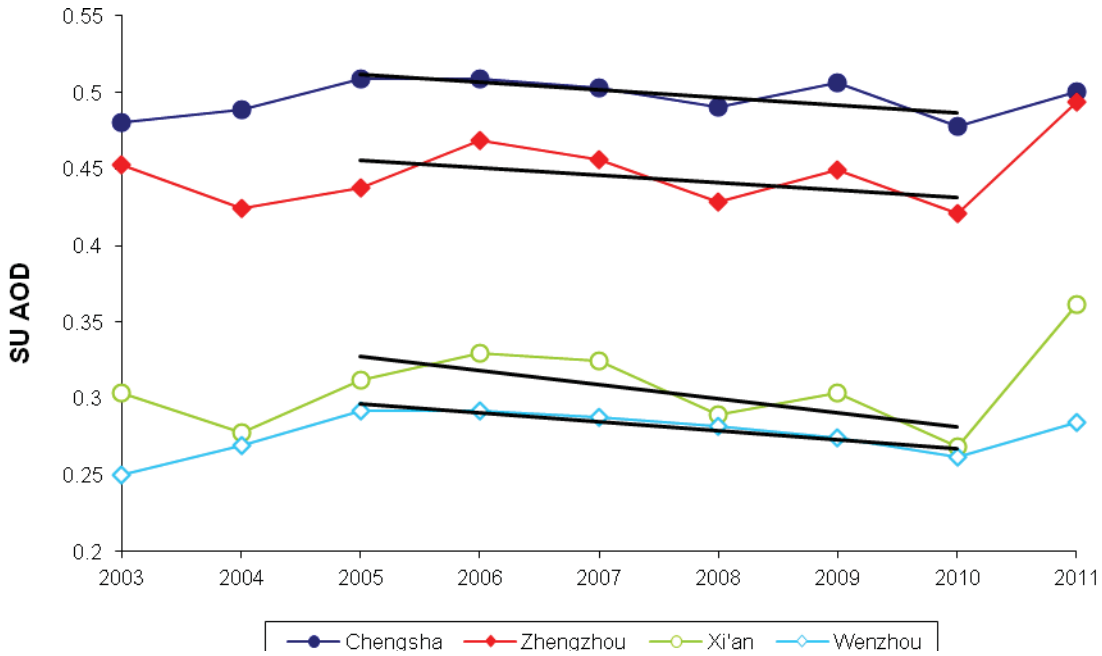

Fig. 16. MERRAero yearly mean SU AOD for Chengsha, Zhengzhou, Xi'an and Wenzhou. Trend lines are included for the years 2005-2010.

3.12 Eastern Asia

This region includes all Asian cities east of China. SU aerosols are the major contributor to total AOD in all these cities (Fig. 17). The four cities in Japan all have decreasing total AOD trends caused by a decrease in SU AOD (Table 18). In Japan, air quality policies have been implemented since the 1960's. As a result, SO_2 emissions and concentrations decreased substantially since the 1970's [Kanada *et al.*, 2013]. Moreover, national air quality statistics data from the Ministry of the Environment (http://www.env.go.jp/en/statistics/contents/index_e.html#iousankabutu) show that anthropogenic SO_2 emissions have decreased by 15% during the period from 2002 to 2008, while SO_2 concentration decreased from 0.005 ppm in 2002 to 0.003 ppm in 2010. It is important to mention that volcanoes release some significant

amount of SO_2 in Japan's atmosphere. Between 2000 and 2007, both natural and anthropogenic SO_2 emissions decreased, and this resulted in decreasing SO_2 concentrations observed at most monitoring sites in Japan [Lu *et al.*, 2010]. Lu *et al.* [2010] also noticed that most sites with increasing SO_2 concentration between 2000 and 2007 (period corresponding with China's industrialization boom) were located in the southwestern tip, closest to China. Environmental policies in Japan had a positive impact on Tokyo's air quality as well. Tokyo has a well-developed air quality network of monitoring stations. Data from this record has been published by Hara *et al.* [2013] for suspended particulate matter. The average concentration based on measurements from 45 ambient monitoring stations showed a decrease of 36% between 2002 and 2010, while the average concentration based on measurements for 33 roadside monitoring stations indicate a decrease of 42% between the same two years. These observed decreasing tendencies support the obtained decreasing MERRAero AOD trend in Tokyo shown in Table 18.

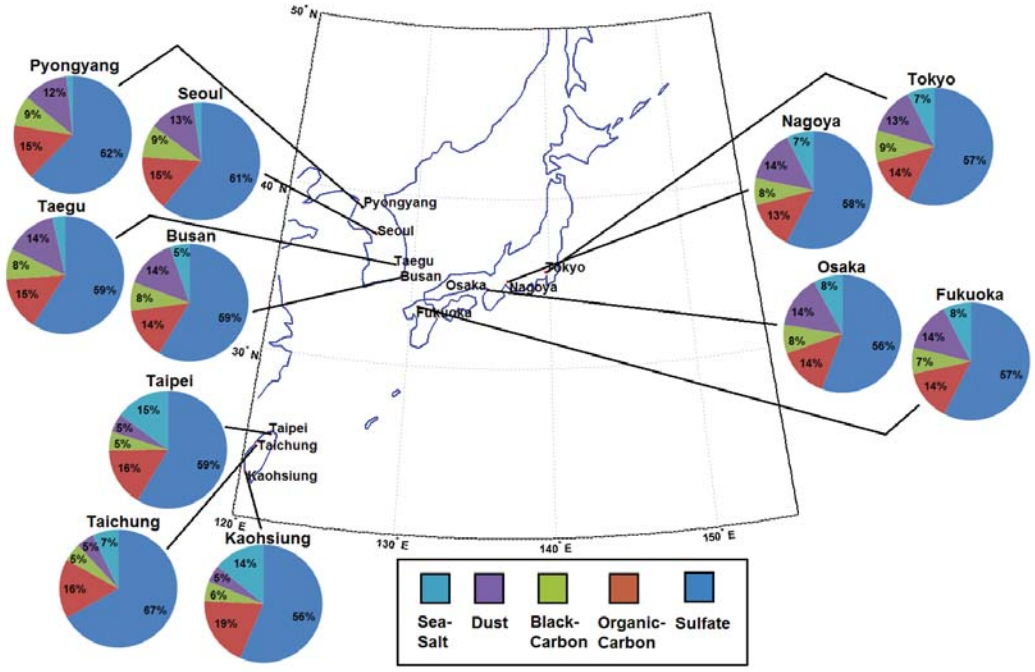

Fig. 17. Cities in eastern Asia with proportion of aerosol species in the MERRAero 10-year mean total AOD.

The three cities in South Korea have rather insignificant trends in both total and SU AOD (Table 18). SO_2 emissions have slightly decreased between 2000 and 2007 in that country [Kim *et al.*, 2011]. Although, Lu *et al.* [2010] estimated that a 1% increase in Chinese SO_2 emissions leads to 0.71% and 1.15% increase in "background" concentration in Japan and South Korea respectively. SO_2 emissions in Seoul between 2001 and 2008 were quite sporadic and did not show any significant trend [Jeong *et al.*, 2013]. The aforementioned facts could explain the obtained insignificant MERRAero AOD trends in South Korea's cities as shown in Table 18.

Table 18. List of cities in eastern Asia, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Japan	Tokyo	0.272	-7.6	-11.7	-6.9	-0.2	-1.7	2.2
	Nagoya	0.263	-7.2	-12.1	-2.2	-1.0	0.0	2.3
	Osaka	0.260	-5.5	-9.5	-2.7	-0.5	-2.5	3.4
	Fukuoka	0.305	-1.7	-3.0	0.9	2.7	-9.6	3.9
North Korea	Pyongyang	0.453	1.1	1.5	-5.3	3.6	5.4	4.6
South Korea	Seoul	0.427	0.8	0.2	-2.7	4.2	5.1	4.8
	Daegu	0.337	0.5	-0.8	1.1	3.9	0.5	3.4
	Busan	0.340	0.0	-1.3	1.5	3.7	-3.9	3.3
Taiwan	Taipei	0.352	-3.8	-4.9	-11.7	-4.0	6.6	6.0
	Taichung	0.305	-7.1	-6.1	-18.4	-6.9	3.7	5.3
	Kaohsiung	0.247	-10.3	-9.5	-22.5	-9.9	1.1	2.4

Taiwan faces the southeastern region of China, where all the Chinese cities with a decreasing AOD trend are located (Section 3.11). This could explain the obtained decreasing MERRAero AOD trend in Taiwanese cities shown in Table 18.

3.13 Southeastern Asia and Oceania

This subsection includes cities east of India and south of China, as well as cities in Oceania. Cities outside of Australia have the highest contribution from SU aerosols to their total AOD (Fig. 18), although the contribution of OC aerosols to total AOD is also significant. SS aerosols are important in Filipino cities given their insular location. Cities in Australia have a heterogeneous distribution of aerosol species; SU, OC and SS aerosols being the most important. Total AOD showed decreasing trends in every city listed in Table 19. This is mainly due to decreasing trends in SU and OC AOD. Wildfire activity attenuation could be a likely cause but we don't have data in this part of the world to support this assertion. Although, according to van der Werf *et al.* [2010, their Table 7 and Fig. 9], modeled carbon emissions from wildfires in Equatorial Asia (including Indonesia) and Oceania suggest some decreasing tendencies between 2002 and 2009.

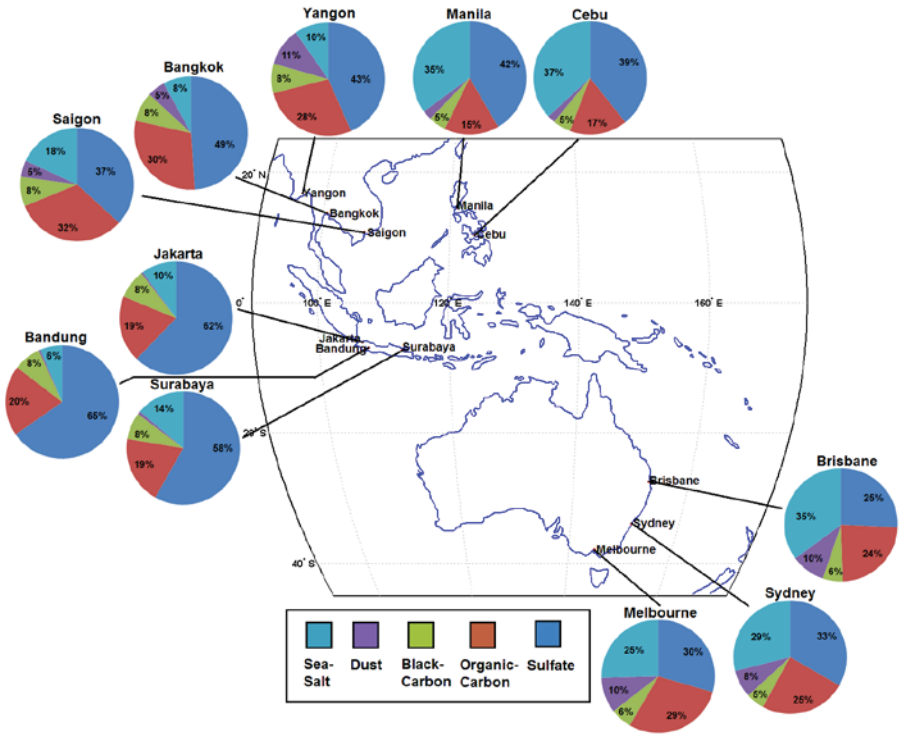

Fig. 18. Cities in southeastern Asia and Oceania with proportion of aerosol species in the MERRAero 10-year mean total AOD.

Table 19. List of cities in southeastern Asia and Oceania, together with the 10-year mean total AOD and its trend for various aerosol species.
Legend – SU: sulfate; OC: organic carbon; BC: black carbon; SS: sea salt; DU: dust

Country	City	Total AOD	AOD Trend (%)					
			Total	SU	OC	BC	SS	DU
Myanmar	Yangon	0.237	-1.1	-4.8	3.0	-0.4	-2.4	4.8
Thailand	Bangkok	0.284	-1.9	-0.3	-2.9	1.8	-18.4	12.1
Vietnam	Saigon	0.163	-10.6	-9.1	-13.8	-9.7	-11.6	4.2
Philippines	Manila	0.138	-9.9	-8.9	-26.6	-19.1	-0.8	-14.1
	Cebu	0.101	-17.5	-19.9	-33.6	-25.6	-5.0	-19.5
Indonesia	Jakarta	0.260	-11.5	-2.8	-32.4	-16.2	-15.0	9.4
	Bandung	0.193	-12.9	-5.4	-31.0	-17.2	-18.9	2.9
	Surabaya	0.191	-10.7	-3.6	-27.1	-14.9	-12.7	1.9
Australia	Brisbane	0.096	-11.8	-8.1	-27.2	-18.0	-5.1	0.9
	Sydney	0.114	-16.0	-0.9	-43.0	-22.5	-6.8	-2.6
	Melbourne	0.093	-6.7	-1.0	-20.9	-8.0	1.6	1.5

4 Discussion

The recently developed ten-year (July 2002 – June 2012) MERRAero aerosol reanalysis was applied to the study of AOD of various aerosol species and their trends over the world's major cities. The model reproduced high SU AOD values over cities in industrialized regions such as North America, Europe, India, China and eastern Asia, caused by fossil fuel consumption. The model also reproduced high DU AOD values over cities in proximity to the major deserts (northern Africa and western Asia), and high OC AOD values over cities in regions where biomass burning is common practice (South America and Africa south of the Sahara).

MERRAero also reproduced declining trends in SU AOD over North American, European and Japanese cities, where effective air quality regulation was implemented. The situation in China is more delicate since air quality regulation in that country has only been implemented recently. MERRAero was able to show increasing trends of SU AOD over Indian cities which is consistent with increasing anthropogenic emissions.

Since OC and BC are also by-products of fossil fuel combustion, in many cities, MERRAero showed correspondence between AOD trends in carbonaceous aerosols and those in sulfate aerosols. In some cities, however, AOD trend in carbonaceous aerosols was disproportionate or opposite to SU AOD trend. This is probably due to varying wildfire and biomass burning activities. We observed such trends in cities on the western coast of the U.S. and Canada, Europe, western Russia, Indonesia and Australia.

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6 Appendix: Acknowledgements

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