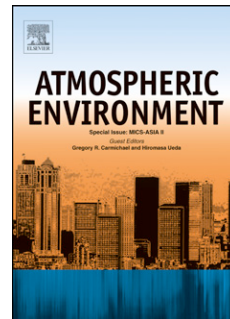


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## 1 Megacities as hot spots of air pollution in the East

## 2 Mediterranean

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29 **Abstract**

30 This paper provides a comprehensive overview of the actual knowledge on the  
31 atmospheric pollution sources, transport, transformation and levels in the East  
32 Mediterranean. It focuses both on the background atmosphere and on the similarities  
33 and differences between the urban areas that exhibited important urbanization the past

34 years: the two megacities Istanbul, Cairo and the Athens extended area. Ground based  
35 observations are combined with satellite data and atmospheric modeling. The overall  
36 evaluation pointed out that long and regional range transport of natural and  
37 anthropogenic pollution sources have about similar importance with local sources for  
38 the background air pollution levels in the area.

39

40 *Keywords:* megacities, East Mediterranean, air pollution, transport, anthropogenic impact

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## 41 **1. Introduction**

42 The increasing need of humans for facilities, security, health care and  
43 employment have been the driving forces for increasing urbanization that gave birth  
44 to the Megacities, urban agglomerations with more than 10 million of inhabitants  
45 (<http://www.worldclimate.com>). This increasing urbanization not only affected the  
46 neighboring landscape, air quality, regional climate and ecosystems in the megacities  
47 but also downwind of these regions. During the last decades, the Mediterranean,  
48 following the general trend, has experienced a rapid growth in urbanization, vehicle  
49 use and industrialization as being reflected in pollutant emissions to the atmosphere.

50 The Eastern basin of the Mediterranean and the surrounding regions, include  
51 two megacities: the Greater Cairo area (GCA) (>15 million, Egypt) at the south edge  
52 of the basin and the Greater Istanbul Area (GIA) (>12 million inhabitants, Turkey) at  
53 the North East edge, as well as several large urban centers like to its northern part the  
54 Greater Athens area (GAA) (>4 million) in Greece (Table 1, Figures 1 and 2a) that  
55 exhibited important urbanization the past years. The region covers rural (inland Greek  
56 and Anatolian peninsulas), maritime (Crete and Cyprus islands) and desert (Anatolian  
57 plateau, north Africa, Middle East) sites.

58 The Mediterranean located at the boundary between the tropical and mid-  
59 latitudes, is subject to large (about 50%) changes in the total O<sub>3</sub> column (Ladstaetter-

60 Weissenmayer *et al.*, 2007), which have been attributed to changes in the location of  
61 the sub-tropical front (Hudson *et al.*, 2003). It is also a crossroad of air masses coming  
62 from Europe, Asia and Africa, where anthropogenic emissions, mainly from Europe,  
63 Balkans and the Black Sea, meet with natural emissions from Saharan dust (e.g.  
64 Kallos *et al.*, 1993, Kanakidou *et al.*, 2007), vegetation (e.g. Liakakou *et al.*, 2009)  
65 and the sea (e.g. Kouvarakis *et al.*, 2002), as well as from biomass burning (e.g. Balis  
66 *et al.*, 2003), which present a strong seasonal pattern. The transport of anthropogenic  
67 pollutants from America also exerts a significant influence in the free troposphere  
68 (Lelieveld *et al.*, 2002).

69 The typical Mediterranean climate is characterized by hot, dry summers and mild,  
70 rainy winters. Evaporation is especially high in its eastern half basin, greatly  
71 exceeding precipitation and river runoff in this region. This causes the sea water level  
72 to decrease and salinity to increase eastward (Demirov and Pinardi, 2002). As a  
73 consequence of its unique location and emissions, the Mediterranean is a climatically  
74 sensitive region, often exposed to multiple stresses, such as a simultaneous water  
75 shortage and air pollution exposure (IPCC, 2007) that is favored by the Mediterranean  
76 climate and is likely to grow in the future due to the rapid urbanization.

77 Air pollution is one of the challenging environmental problems in the whole  
78 East Mediterranean basin since both ozone and aerosol air quality limits are often  
79 exceeded, in particular during summer. In contrast to Central and Northern Europe,  
80 photochemical episodes can also occur during winter since at these latitudes solar  
81 radiation is intensive year-around, driving photochemical reactions that favour air  
82 pollution. The contribution of natural emissions to these exceedences seems  
83 significant and remains to be determined. High ozone and aerosol concentrations are

84 harmful for human health and ecosystems, and they also cause agricultural crop loss  
85 and climate change.

86 This paper summarizes the actual knowledge on the atmospheric pollution  
87 sources, transport, transformation and levels in the Eastern Mediterranean. It first  
88 outlines characteristics of the two megacities Istanbul and Cairo and the Athens  
89 extended area, air transport patterns and meteorology. Then it discusses the  
90 similarities and differences between these major pollution sources in the region and  
91 compares them to the background atmosphere. Areas where further research is needed  
92 to support mitigation strategy development are pointed out.

## 93 **2. The Megacities characteristics**

94 The studied urban areas are distributed over three continents: Europe, Asia and  
95 Africa and present some common features as well as significant differences (Table 1).  
96 Istanbul extends on two continents with the European part of the city being the oldest  
97 one. It is separated from the Asian part by Bosphorus strait of 30-km length that  
98 connects the Marmara Sea at the south with the Black Sea at the north.

99 The air circulation patterns at all three urban locations are affected by the  
100 existence of hills: seven hills in GIA, the Mogattam hill to the east and the southeast  
101 of GCA and the Parnes, Penteli and Hymettus mountains, all three over 1000 m,  
102 surrounding mainly the North- and East boundaries of GAA. In Istanbul northeasterly  
103 winds prevail during summer (Kindap, 2008) whereas southwesterly occur mainly  
104 during winter (Koçak *et al.*, 2010). Istanbul is vulnerable to trans-boundary transport  
105 of air pollutants from Europe, because of its location on the eastern end of the  
106 continent in the zone of westerly synoptic air flow (Kindap *et al.*, 2006). Cairo  
107 experiences two dominant wind sectors: the North sector and the South–West sector.  
108 Although prevailing all year long, the north sector presents maximum occurrence

109 frequency in summer. The winter and spring seasons are significantly impacted by  
110 south-western winds (Favez *et al.*, 2008a,b). Finally, in Athens, the prevailing wind  
111 axis is north-east/ south-west and the ventilation takes place at northeasterly  
112 directions (Melas *et al.*, 1995).

113 GIA and GAA are both subject to sea and land breeze local circulation  
114 phenomena, favored during the weakening of the synoptic wind. During summer, the  
115 southern part of GIA close to the Marmara Sea experiences such circulation patterns  
116 that influence pollutants transport and accumulation in the boundary layer (Im *et al.*,  
117 2006). The northern part of GIA is affected by the colder northern air masses and the  
118 cooler Black Sea. In Athens sea/land breezes appear along the axis of the basin (NE to  
119 SW) and anabatic/catabatic flows from the surrounding mountains. Under these  
120 circumstances the ventilation of the basin is poor; the boundary layer is shallow and  
121 the air pollution potential increases (Melas *et al.*, 1995 and references therein). The  
122 sea breeze system from the Saronic Gulf, located to the south of GAA, sweeps  
123 primary pollution from the city center, combined with O<sub>3</sub> titration, and favors  
124 pollutant accumulation to the northern suburbs where significant episodes are  
125 encountered. Air pollution episodes may occur in Athens during all seasons of the  
126 year but most of these episodes are associated with the development of sea-breeze  
127 (Kallos *et al.*, 1993).

### 128 2.1. Istanbul

129 The city of Istanbul (Table 1) is hosting almost 17% of Turkey's population.  
130 Since the southern part of the GIA is the most urbanized, further growth will intensify  
131 pressure on industrial and residential uses in the northern part of the metropolitan  
132 region, where the natural protection areas and the watersheds are located (OECD,  
133 2008). Average wind speed is highest in winter and lowest in summer with annual  
134 average of about 2.7m/s. The humidity is high during all seasons (Ezber *et al.*, 2007).

135 The heating effect due to urbanization was found to produce two-cell structure during  
136 summer, one on the European and one on the Asian side of the city. The cells extend  
137 to about 600–800 m height in the atmosphere over the city and combine aloft (Ezber  
138 *et al.*, 2007).

139

## 140 2.2. Cairo

141 Cairo (Al-Qāhirah), Egypt's capital (Table 1) situated south of the delta in the  
142 Nile basin, is the largest rapidly expanding city in Egypt facing many environmental  
143 problems. GCA's main populated area of about 200 km<sup>2</sup> is 4 km wide stretching 50  
144 km along the banks of the Nile River. Outside GCA desert areas extend in the west  
145 and east directions. Dust and sand storms frequently occur in spring and autumn  
146 (Zakey and Omran, 1997). Hot desert cyclones known as the "Khamasin" depressions  
147 pass over the desert during spring, always associated with strong hot and dry winds  
148 often carrying dust and sand that increase particulate matter (PM) levels. During  
149 winter the climate is generally cold, humid and rainy; while during the summer season  
150 the predominant weather is hot and dry (Zakey *et al.*, 2008). The mean wintertime  
151 wind is weaker than during summer, implying a lower ventilation of the area during  
152 winter that could favor pollutant accumulation in the vicinity of the sources (Abu-  
153 Allaban *et al.*, 2009). Robaa (2003) showed that rural and suburban parts of the city  
154 have higher ventilation due to higher wind speeds than urban parts, which may lead to  
155 higher pollutant levels in the urban regions of GCA. Cairo has a very poor dispersion  
156 factor because of the advection patterns, its layout of tall buildings and narrow streets  
157 and the lack of rain (Table 1). This results in a permanent haze over the city with PM  
158 in the air reaching over three times the background levels.

159

### 160 2.3. Athens agglomeration

161 The GAA gathers about 40% of Greece's total population in a basin on the  
162 west coast of the Attica peninsula. During the warmer part of the year, the mean wind  
163 pattern in the atmospheric boundary layer is a persistent northeasterly flow of  
164 relatively high constancy. GAA is also exposed to the summer monsoon circulation of  
165 the Eastern Mediterranean. Etesians, a system of semi persistent summer northerly  
166 winds, favor good ventilation of the basin prohibiting pollution episodes.

167

### 168 2.4. Outflow of pollution

169 Trajectories at approximately 700m height have been used to define air  
170 pollution transport patterns from Istanbul, Cairo and Athens, in a regional scale. They  
171 are based on 30-year (1961-1990) reanalysis data (NCEP/NCAR), available for every  
172 six hours at a 2.5° resolution (Kindap *et al.*, 2009). The computed probability depends  
173 on the grid size and increases with the trajectories length, with very small changes for  
174 trajectories longer than 8 days (Kindap *et al.*, 2009). Figure 1 depicts the probability  
175 of air masses originating from GIA, GCA and GAA to reach various locations in the  
176 East Mediterranean, demonstrating the regional importance of air pollution from these  
177 megacities. Istanbul pollution is exported mainly in the North East- South West  
178 direction (Koçak *et al.*, 2010) whereas Cairo outflow is mainly affecting the south-  
179 southwest locations and the Arabian Peninsula. Similarly, Athens plume is transported  
180 mainly towards South East over the East Mediterranean Sea. These results are in good  
181 agreement with the global modeling study by Lawrence *et al.* (2007).

## 182 3. Emission sources of air pollutants

183 All three cities experience heavy pollution from the transportation sector with  
184 more than 2 million of cars in Athens and Istanbul and more than 1 million in Cairo,

185 of variable age and technical characteristics with the older ones in Cairo. A large  
186 fraction of their country's industrial activities is also located in their vicinity.

187 The emissions inventories available for the entire East Mediterranean have  
188 relatively coarse resolution (e.g. EMEP in 50 km resolution, Vestreng *et al.*, 2006,  
189 and global inventories down to 1°x1° Granier *et al.*, 2005). The new EDGAR v4  
190 inventory now becoming available, is making significant improvement increasing the  
191 resolution to 0.1°x0.1° (<http://edgar.jrc.ec.europa.eu/>). However for large urban  
192 agglomerations such as GIA, GAA and GCA higher resolution detailed emission  
193 inventories would greatly improve our understanding of air pollution levels in the  
194 area. Such inventories of anthropogenic sources have been developed by Markakis *et*  
195 *al.* (2009; 2001a,b), in high spatial (2x2 km<sup>2</sup>) and temporal resolutions for the GIA  
196 (reference year 2007) and for the GAA (reference year 2003), but appropriate  
197 information is still missing for Cairo (Table 2). Weekend emissions are lower than  
198 week days and diurnal profile fits with the rush hours due to the highest contribution  
199 of traffic emissions (Markakis *et al.*, 2009). Application of the Markakis *et al.* (2009)  
200 inventory has significantly improved the simulations of PM<sub>10</sub> levels (Im *et al.*, 2010)  
201 in GIA.

202 Table 2 shows the annual sectoral distribution of pollutants. Industrial  
203 activities are important sources of PM and responsible for almost 30 % of the SO<sub>2</sub>  
204 emissions. On-road traffic is the major contributor to CO, NO<sub>x</sub> and non methane  
205 volatile organic compounds (NMVOCs) in Istanbul and Athens. Residential  
206 combustion and cargo shipping are significant pollution contributors in GIA and  
207 GAA. Similar conclusions are reached for Istanbul by Koçak *et al.* (2010), based on  
208 Positive Matrix Factorization (PMF) analysis of aerosol chemical characterization  
209 observations (Theodosi *et al.*, 2010) from an urban background site in Istanbul.  
210 Almost 20% of PM emissions in GAA originate from non-exhaust sources, including

211 tire, break wear and road abrasion. The central heating operations do not account for  
212 more than a few percent in the annual totals (with the exception of  $\text{SO}_2$  ~ 15%  
213 contribution), but in the winter months they make a significant contribution.

214 Cairo shows different emissions fingerprint: Residential Combustion and  
215 Industries being the major emitters of CO and  $\text{NO}_x$  whereas NMVOC emissions are  
216 mostly from solvents use seconded by road transport. A significant portion of  $\text{NO}_x$   
217 (~50%) and  $\text{SO}_2$  (~71%) originates from industrial activities. On-road traffic is also  
218 an important source for CO (35%), NMVOC (37%) and  $\text{PM}_{2.5}$  (36%). Anthropogenic  
219  $\text{PM}_{2.5}$  in GCA originates mainly (54%) from residential combustion and open  
220 burnings. Open fire burnings is a common practice and a major contributor to air  
221 pollution in Egypt, as also seen on aerosol optical depth (AOD) seasonality derived  
222 from satellite data with peaks in fall (Hatzianastassiou *et al.*, 2009).

223 To limit air pollution, measures were taken in all three urban centres around  
224 1990-1995 with different level of implementation success.

225

### 226 3.1. Istanbul

227 Between 1980 and 1990 the consumption ratio of coal to fuel-oil increased  
228 from 0.68 (in 1980) to 3.09 (in 1990; Tayanc, 2000). There has been the use of higher  
229 quality coal and a shift from coal to natural gas for domestic heating purposes starting  
230 from early 90s, leading to a decrease in the concentrations of primary pollutants such  
231 as sulfur oxides ( $\text{SO}_x$ ) and an increase in secondary pollutants such as secondary  
232 aerosols and ozone (Tayanç, 2000). From the beginning of 1998 liquefied petroleum  
233 gas (LPG) has been widely used in traffic. Low quality solid and liquid fuels with  
234 high sulfur content, natural gas and LPG are the most commonly used fuel types in  
235 the industrial activities that comprise 37% textile, 30% metal, 21% chemical, 5% food  
236 and 7% other industries (Istanbul Chamber of Industry reports cited by Im *et al.*,

237 2006). Under these dense and various industrial activities, the region experiences very  
238 complex air quality conditions.

239

### 240 3.2. Cairo

241 About 52% of the industries and 40% of the electricity production in Egypt are  
242 located in the GCA (Nasralla, 2001). Cairo has many unregistered lead and copper  
243 smelters which heavily pollute the city. GCA accommodates 50% of Egypt's road  
244 transport fleet, 60% of which is over 10 years old, lacking modern emission cutting  
245 features like catalytic converters (Mowafi and Atalla, 2005). The information  
246 regarding the amounts of pollutants released in the atmosphere of Cairo is very  
247 limited (El Mowafi and Atalla, 2005; Gurjar *et al.*, 2008; Table 2). Source  
248 apportionment analysis based on simultaneous observations of several non methane  
249 hydrocarbons (NMHC), including aromatics, and of aerosol components, including  
250 metals, (Abu-Allaban *et al.*, 2002, 2007, 2009), pointed to mobile and industrial  
251 emissions (lead smelting and LPG, considering that industrial processes may be  
252 fueled by LPG) as the major source of NMHC during both summer and winter.

253 In 1995, the first environmental acts were introduced and the situation has  
254 seen some improvement, with 36 air monitoring stations and emissions control on  
255 cars. 20,000 buses have also been commissioned to the city to improve congestion  
256 levels. In 2003, Egypt initiated an enforced vehicle emission-testing program in  
257 Greater Cairo. The limits of CO, hydrocarbons and opacity for the vehicles have been  
258 significantly reduced in 1995. However, the publicized information indicated an  
259 overall failure rate of about 10% (El Mowafi and Atalla, 2005).

260

262 The massive number of registered vehicles in circulation, growing at a rate of  
263 7% yearly, is allegedly the major cause of air pollution related problems in the area,  
264 taking into account the large proportion of non-catalytic (0.8 million) or powered by  
265 old technology diesel engines vehicles (0.2 million). Athens experiences very severe  
266 congestion phenomena with the average speed not exceeding 12 km/h during rush  
267 hours. Although the use of natural gas for domestic heating purposes has increased  
268 lately, combustion of fuel oil is still primarily used for central heating. The large  
269 industrial complexes are located in the Thriassion plain, several kilometres to the west  
270 of the GAA. They are separated from the Athens basin by mount Aigaleo (up to 450  
271 m) that acts as a physical barrier preventing most of the exchange of air pollutants  
272 between the industrialized area and the city (Melas *et al.*, 1998).

#### 273 4. Air pollution in the East Mediterranean

274 Enhanced levels of pollution (Figure 2) and increasing trends over the last  
275 decade are seen by satellites over East Mediterranean and over the Middle East and  
276 Cairo (Lelieveld *et al.*, 2008; Vrekoussis *et al.*, 2009). Background tropospheric O<sub>3</sub>  
277 levels in the area are high, particularly in spring and summer, depending on the  
278 meteorological conditions since they are controlled by large-scale, long-range  
279 transport and photochemical formation (Gerasopoulos *et al.*, 2005). Background PM  
280 levels are also high due to a significant contribution of Sahara dust aerosol (Querol *et*  
281 *al.*, 2009) but also transported pollution (Mihalopoulos *et al.*, 2007). In the urban  
282 atmosphere due to the high levels of primary pollutants, like PM and NO<sub>x</sub>, maintained  
283 by the anthropogenic emissions, O<sub>3</sub> titration by reaction with NO is leading to very  
284 low O<sub>3</sub> levels over city centers, whereas NO<sub>x</sub> and PM remain high. Primary pollutants  
285 decrease downwind where O<sub>3</sub> and secondary aerosols build up photochemically. In

286 the urban regions, the temporal variability of primary gaseous pollutants reflects the  
287 high emissions during winter time and the faster photochemical destruction during  
288 summer time. Figure 2b depicts the tropospheric NO<sub>2</sub> columns as observed by  
289 SCIAMACHY (SCanning Imaging Absorption SpectroMeter for Atmospheric  
290 CHartographY) for the period 2005-2006 and highlights the local pollution sources all  
291 around the Mediterranean. SCIAMACHY observations of NO<sub>2</sub> tropospheric column  
292 over the region (Figure 2b) indicate high tropospheric columns of NO<sub>2</sub> over urban  
293 sites around the Mediterranean with those over both the GIA and the GCA increasing  
294 over the last years (Vrekoussis *et al.*, 2009). This distribution nicely contrasts to the  
295 O<sub>3</sub> distribution shown in Figure 2a, that presents the largest enhancement actually  
296 over the water, covering the whole East Mediterranean basin, which acts as receptor  
297 of the surrounding pollution. Total columns of CO over the region range between 1.5  
298 and  $3 \times 10^{18}$  molecules.cm<sup>-2</sup> maximizing in late winter/early spring (high emissions)  
299 and minimizing in late summer/early fall (high photochemical destruction) (MOPITT:  
300 Measurements of Pollution in The Troposphere; [ftp://14ftl01.larc.nasa.gov/MOPITT/  
301 MOP03M.003/](ftp://14ftl01.larc.nasa.gov/MOPITT/MOP03M.003/)).

302 Mean satellite observations of short lived trace gases (NO<sub>2</sub>, CHOCHO, HCHO  
303 and O<sub>3</sub>) and AOD over the region during the recent years are summarized in Figure 3.  
304 High tropospheric columns of NO<sub>2</sub>, HCHO, CHOCHO are observed over urban  
305 locations (GIA, GCA, GAA) and low levels over the background receptor site of  
306 Finokalia. The progressive reduction of tropospheric columns of NO<sub>2</sub> from Istanbul to  
307 Athens and then to Cairo can be noticed together with a similar trend in CHOCHO  
308 and HCHO, used as proxy for NMVOC levels. Remarkably, CHOCHO peaks over  
309 GCA pointing to a higher NMVOC/NO<sub>x</sub> ratio than over GIA and GAA. This  
310 indicates higher O<sub>3</sub> formation potential of NO<sub>x</sub> in GCA due to high NMVOC  
311 loadings, in agreement with ground-based observations (Abu-Allaban *et al.*, 2009).

312 The HCHO/CHOCHO ratio appears different over GCA than over GIA and GAA,  
313 indicating a different NMVOC speciation in this region, most probably strongly  
314 marked by biomass burning emissions. Tropospheric O<sub>3</sub> columns indicate the elevated  
315 O<sub>3</sub> background towards the south that maximizes over the Finokalia receptor site.  
316 However, they minimize over GCA that is closer to tropics and thus affected by a  
317 much lower total O<sub>3</sub> column (~18 DU lower than over Finokalia, based on TOMS /  
318 OMI 2005-2008 data in 0.25°x0.25° grid; <http://gdata2.sci.gsfc.nasa.gov>).

319

#### 320 *4.1. Ozone and its precursors*

321 Table 3 recapitulates the available measurements of ozone in the Eastern  
322 Mediterranean at urban and regional background locations. A clear North to South  
323 increasing gradient is evident. In particular, surface O<sub>3</sub> increases when moving from  
324 rural background sites of Istanbul to Athens and then to Cairo, indicating significant  
325 contribution from long-range transport sources in air masses that age in the region.  
326 Ozone measurements along the Aegean Sea (NE Mediterranean, Kourtidis *et al.*,  
327 2002; Kouvarakis *et al.*, 2000) confirmed that transport from the European continent  
328 is the main mechanism controlling ozone levels in the region, especially in summer  
329 (or spring depending on the prevailing air transport patterns), when ozone presents a  
330 maximum of about 60±10 ppbv (Gerasopoulos *et al.*, 2005).

331 Kalabokas *et al.* (2007) analyzing aircraft data found that during summer in  
332 the middle troposphere of the eastern basin, O<sub>3</sub> was only 5–10% higher than over  
333 Central Europe and high tropospheric ozone values were mainly confined in the low  
334 troposphere. Gerasopoulos *et al.* (2006b) analyzing 7 years of surface O<sub>3</sub> observations  
335 at Finokalia, found that the entrainment of O<sub>3</sub> rich air masses from the free  
336 troposphere (4–6% of the observed ozone levels) maximizes during summer, when  
337 the chemical production of O<sub>3</sub> is also enhanced by photochemistry and long-range

338 transport. This summertime high regional source term of O<sub>3</sub> is almost balanced by the  
339 enhanced O<sub>3</sub> destruction via deposition and chemistry. Below a brief presentation of  
340 the ozone measurements at the various cities is presented.

341

#### 342 4.1.1. Istanbul

343 Im *et al.* (2008) reported O<sub>3</sub> observations at two different urban locations  
344 within GIA located at both its European (8±7 ppbv) and the Asian (11±8 ppbv) parts  
345 from 2001 to 2005. The highest ozone levels were observed during sunny and warm  
346 summer days (maximum temperatures >25 °C) with southwesterly surface winds.  
347 Recent observations of ozone levels in semi-urban and rural stations in the GIA  
348 during the period 2007-2009 (Im *et al.*, 2009), provide insight to the background  
349 levels of ozone in the extended area. They show higher ozone levels than the urban  
350 stations, reaching 30-35 ppbv on average, for high ozone seasons.

351

#### 352 4.1.2. Cairo

353 Ozone in the southwestern Cairo area has been observed to exhibit a seasonal  
354 and diurnal cycle with levels reaching 70 ppbv in summer (Egyptian Environmental  
355 Affairs Agency, <http://www.eeaa.gov.eg/eimp/news8.html>). Year-long, mean levels  
356 often exceed the Egyptian and European Union air quality standards of 60 ppbv for  
357 daytime (8-h) O<sub>3</sub> mixing ratios. Khoder (2009) reported a year (Dec 2004-Nov 2005)  
358 of observations of ground level O<sub>3</sub>, nitrogen dioxide (NO<sub>2</sub>) and nitric oxide (NO)  
359 concentrations at Giza in the GCA with daytime mean O<sub>3</sub> values of 91 ppbv during  
360 summer (Table 3). Air masses reaching Cairo during summer originate from the  
361 Aegean and the Cretan Seas. Thus, considering the Finokalia regional background  
362 values (60 ppbv), the observed mean value of 91 ppbv in Cairo indicates that despite  
363 O<sub>3</sub> titration from the local NO<sub>x</sub> emissions, significant photochemical O<sub>3</sub> production

364 occurs. This is additionally supported by high VOC levels (Abu-Allaban *et al.*, 2009)  
365 in the GCA, in agreement with the satellite observations shown in Figure 3. Maxima  
366 in O<sub>3</sub> levels occur in summer due to local photochemical production and long range  
367 transport whereas the highest levels of NO<sub>x</sub> are found in winter. The diurnal cycles of  
368 O<sub>3</sub> revealed a uni-modal mid-day peak year-around. The diurnal variations in NO<sub>x</sub>  
369 concentrations during the winter and summer showed two daily peaks linked to traffic  
370 density.

371

#### 372 4.1.3. Athens

373 Kalabokas *et al.* (1999a,b) analyses of 11-year observations from the Greek  
374 Ministry of Environment air pollution network in Athens since 1987, show a  
375 significant downward trend for almost all primary pollutants in all stations.  
376 Comparison between the 3-year periods 1988-1990 and 1995-1997 gave the highest  
377 reduction in the center of GAA of 52%, 34%, 26% and 20% for SO<sub>2</sub>, CO, NO<sub>x</sub> and  
378 black smoke, respectively. The concentrations of the secondary gaseous pollutants  
379 remained essentially at the same levels since 1990, even though different  
380 characteristics (e.g. in ozone trends) may be observed for different site types  
381 (Hatzianastassiou *et al.*, 2007). Observations of O<sub>3</sub> prior to 2000 (Kalabokas and  
382 Repapis, 2004) at three stations in the GAA and the surroundings were found to  
383 exhibit characteristic seasonal variation of rural ozone concentrations, with lowest  
384 winter afternoon values at about 25 ppbv in December–January and average summer  
385 afternoon values at about 60 ppbv in July–August. These values are comparable to  
386 observations at Finokalia (Gerasopoulos *et al.*, 2005; 2006b) and indicate significant  
387 contribution from long range transport sources rather than local photochemistry.

388 The increased regional background in Athens is also supported by the CO-  
389 NO<sub>x</sub> molar ratios in GAA (Figure 4, derived from Table 3) that are between 20 and

390 30, whereas in GIA are lower ranging from 9.8 (Sarachane) to 12.6 (Kadikoy) close to  
391 those in Mexico City (11) and higher than for Tokyo (8.5) and US cities (6.7 in 2003)  
392 (Parrish *et al.*, 2009). Both in GIA and GAA, CO-to-NO<sub>x</sub> molar ratios are lower than  
393 the mean ratio of 41 observed in Beijing that has been attributed to significant  
394 regional contribution to CO levels in that megacity (Parrish *et al.*, 2009). Ratios  
395 higher than 50 are derived from the observations by Elminir *et al.* (2005) for a GCA  
396 residential site and point to the different CO sources characteristics (like older cars,  
397 domestic combustion and open fires) in GCA than in the other megacities. These  
398 ratios are however much lower than those of about 100 to more than 300 observed  
399 during summertime at Finokalia where long range transport is the dominant source for  
400 CO.

401

#### 402 4.2. Airborne particulate matter

403 The Mediterranean is one of the areas with the highest AOD in the world, also  
404 seen from space (Hatzianastassiou *et al.*, 2009), which presents high temporal  
405 variability due to the short lifetime of PM in the troposphere (of the order of a week).  
406 Two-year (2005-2006) mean observations of AOD at 443nm over the area from  
407 MISR (Multiangle Imaging Spectro Radiometer) and of the aerosol small mode  
408 fraction derived from MODIS (Moderate Resolution Imaging Spectroradiometer,  
409 using the Giovanni daily data of NASA GES DISC), are depicted in Figures 2c and  
410 2d. Although the annual mean AOD distribution is marked by the Sahara dust  
411 contribution, relatively high levels of AODs are also seen over the Aegean and the  
412 Black Sea. In addition, Figure 2d indicates the existence of significant fraction (about  
413 0.5 to 0.6) of fine particles in the region that are commonly associated with pollution  
414 sources. Synergistic analysis of MODIS AOD and aerosol index TOMS data, used as  
415 proxy for absorbing dust aerosol, enabled a first evaluation of the local anthropogenic

416 contribution to the AOD over the GAA and GCA at 15-30% and 25-50%,  
417 respectively, during summer (Hatzianastassiou *et al.*, 2009).

418 Ground- based observations over the area show high concentrations of aerosols, in  
419 both PM<sub>10</sub> and PM<sub>2.5</sub> fractions (Querol *et al.*, 2009), with PM<sub>2.5</sub>/PM<sub>10</sub> ratios around  
420 0.5 (Table 3), in agreement with the satellite observations in Figure 2d. In the Eastern  
421 Mediterranean, PM<sub>10</sub> has a similar seasonal behavior as PM<sub>2.5</sub>, with maxima in spring  
422 and fall in the eastern basin due to African dust transport. This is also seen by lidar  
423 (Papayannis *et al.*, 2008), sun photometer (Fotiadi *et al.*, 2006) networks and satellite  
424 based-sensors (Papayannis *et al.*, 2005; Kalivitis *et al.*, 2007). PM<sub>1</sub> behaves  
425 differently showing small maxima during summer and is mainly dominated by  
426 pollution components (Gerasopoulos *et al.*, 2007; Koçak *et al.*, 2008).

427 In the background coarse mode aerosol (PM<sub>1.3-10</sub>) dust and ionic components  
428 contribute about 40% and 50%, respectively and organics about 10% (Koulouri *et al.*,  
429 2008a). Mineral dust transport events are found to contribute about 8-12  $\mu\text{g m}^{-3}$  to the  
430 background PM<sub>10</sub> annual mean levels in the East Mediterranean, whereas an  
431 additional 5-10  $\mu\text{g m}^{-3}$  is attributed to transported anthropogenic regional sources and  
432 sea-spray loads (Querol *et al.*, 2009). Re-suspension of dust is likewise a significant  
433 and highly uncertain component of aerosols in the cities. Recent aerosol mass  
434 spectrometer measurements of ultra fine aerosols on Crete Island during late spring  
435 (Hildebrandt *et al.*, 2010), revealed highly oxidized background organic aerosol  
436 throughout the campaign, regardless of the source region. These observations of aged  
437 particles in air masses that circulated and were photochemically processed over the  
438 extended region, support the role of the East Mediterranean basin as the ‘pressure-  
439 cooker’ of transported air pollution. Compared to the colder Central and North  
440 Europe, the high temperatures in the Mediterranean impose a low thermal stability of

441 ammonium nitrate in summer and favor the formation of nitric acid rather than  
442 ammonium nitrate in the area (Querol *et al.*, 2009; Mihalopoulos *et al.*, 1997).

443 High sulfate background loadings in the East Mediterranean are mostly  
444 attributed to the long-range transport of SO<sub>2</sub> (Zerefos *et al.*, 2000). In addition,  
445 significant interactions exist in the Mediterranean between natural and anthropogenic  
446 components in the atmosphere, both in the gas and aerosol phases. Observations and  
447 modeling have shown that on a mean yearly basis, marine biogenic emissions  
448 contribute up to 20% to the total sulphate production (Kouvarakis and Mihalopoulos,  
449 2002). They also demonstrate that the reaction of dimethyl sulfide of marine origin  
450 with nitrate radicals, which are mainly of anthropogenic origin, is responsible for  
451 about 17% of the total HNO<sub>3</sub> production plus particulate nitrate formation  
452 (Vrekoussis *et al.*, 2006). The deposition of these species is of great environmental  
453 significance since it provides nutrients to the ocean. During summer in the eastern  
454 Mediterranean, sulphate on fine particles is produced via gas phase reactions whereas  
455 almost 90% of the supermicron nss-sulphate is formed via heterogeneous pathways,  
456 coating natural aerosols (Mihalopoulos *et al.*, 2007).

457

#### 458 4.2.1. Istanbul

459 Hourly PM<sub>10</sub> levels are monitored by the metropolitan Municipality of  
460 Istanbul at the urban network stations of GIA since late 90's. GIA experiences high  
461 and variable levels of PM<sub>10</sub> and PM<sub>2.5</sub> particles (Table 3). Ozdemir *et al.* (2009)  
462 reported average PM<sub>10</sub> levels of about 66 µg·m<sup>-3</sup> observed at 10 Istanbul municipality  
463 stations during the last 10 years with values ranging from 47 µg·m<sup>-3</sup> to 115 µg·m<sup>-3</sup>.

464 A significant fraction of studied PM<sub>10</sub> episodes has been attributed to regional  
465 transport of African dust and anthropogenic emissions. Kindap *et al.* (2006)  
466 calculated that almost 50% of the wintertime PM<sub>10</sub> episodes in 2002 are associated

467 with air masses coming from Eastern Europe. Karaca and Camci (2010) attributed  
468 about half of the studied high PM<sub>10</sub> levels in Istanbul in 2008 to distant source  
469 contributions. On the other hand, Im et al. (2010) studied the effect of local emissions  
470 on a 5-day PM episode in January 2008 using the high resolution emission inventory  
471 of Markakis et al. (2009) and attributed 90% of the elevated PM<sub>10</sub> levels to local  
472 anthropogenic emissions, combined with very low persisting vertical mixing. This is  
473 in agreement with Koçak *et al.* (2010), who evaluated the contribution of the  
474 anthropogenic sources to PM<sub>10</sub> levels at about 90%, in an independent analysis of the  
475 same episode.

476 Recently, more than one year of aerosol observations at the background  
477 Bögaziçi University sampling station in Bosphorus strait coast, provided the first  
478 complete chemical characterization measurements in GIA (Theodosi *et al.*, 2010).  
479 They measured 9 different water-soluble ions, water soluble organic carbon (WSOC),  
480 organic and elemental carbon (OC, EC) and several trace metals, between November  
481 2007 and June 2009. Trace elements related to human activities obtained peak values  
482 during winter due to domestic heating, whereas natural origin elements peaked during  
483 the spring period due to dust transport from Northern Africa. During winter, OC was  
484 found to be mostly primary and strongly linked to fuel oil combustion and traffic, as  
485 EC. Both OC and EC concentrations increased during winter due to domestic heating.  
486 The mean OC/EC ratio was about 2, lower than those in Athens and Finokalia, but  
487 close to those observed in GCA (Table 3), indicating an overall dominance of primary  
488 pollution. The higher WSOC to OC ratio observed during summer was mostly  
489 attributed to the presence of secondary, oxidised and more soluble organics. Source  
490 apportionment PMF analysis of these long term observations indicates that  
491 approximately 80 % of the PM<sub>10</sub> in Istanbul is anthropogenic in origin (Koçak *et al.*,  
492 2010). Secondary aerosols maximize during summer and are mainly due to long-range

493 transport sources that account for 20% of the PM<sub>10</sub> mass over the studied 1.5-years  
494 period. Adding the contributions of crustal and sea salt (10.2 and 7.5 % of the  
495 observed mass, respectively), regional sources can explain at least 38% of PM mass,  
496 in line with the earlier mentioned studies.

497

#### 498 4.2.2. Cairo

499 There have been a number of studies that evaluated the long-term surface  
500 aerosol observations in Cairo (Abu-Allaban *et al.*, 2002; 2007) along with chemical  
501 composition (Favez *et al.*, 2008a,b). These studies showed that the area is  
502 characterized by elevated levels of surface PM, with annual averages around 100 µg  
503 m<sup>-3</sup> and above (Table 3). Favez *et al.* (2008a,b) reported more than 2 years (Jan.  
504 2003- May 2006) of weekly observations of bulk aerosols at two GCA urban sites  
505 (Table 3), along with their chemical characterization with respect to selected ionic  
506 species and carbonaceous aerosols (sum of EC and OC). Dust aerosols displayed high  
507 background levels (50 µg m<sup>-3</sup>) all year long, maximizing during the dust storm periods  
508 (Favez *et al.*, 2008a). About 40% of Ca<sup>2+</sup> on these dust aerosols was found to be  
509 associated with ions of anthropogenic origin like SO<sub>4</sub><sup>=</sup>, NO<sub>3</sub><sup>-</sup> and/or Cl<sup>-</sup>, pointing out  
510 human driven processes that alter the chemical characteristics of dust and thus its  
511 climatic impact on a regional scale. High concentration levels of non-sea-salt Cl<sup>-</sup> (up  
512 to 15 µg m<sup>-3</sup> on a monthly basis), likely of industrial origin, were observed in autumn  
513 and winter. During autumn, biomass burning aerosols originating from rice straw  
514 burning in the Nile Delta, known as the “Black Cloud” event, have been estimated to  
515 account for 12%, 35% and 50% of Cairo EC, water insoluble organic carbon (WIOC)  
516 and WSOC mass concentrations, respectively.

517 Overall, non-dust aerosols were equally distributed between carbonaceous  
518 aerosols and ions, and their concentrations were about 100 µg m<sup>-3</sup> in autumn and

519 winter, and  $60 \mu\text{g m}^{-3}$  in spring and summer. Remarkably, relatively low WSOC/OC  
520 ratios (about 1/3) were obtained all the year-long. Favez *et al.* (2008b) further  
521 investigated the carbonaceous content in the sub micron fraction of aerosols at an  
522 urban site in GCA in spring 2005. They found well-marked diurnal patterns for the  
523 WSOC/EC and WIOC/EC ratios, with minima during the traffic-influenced morning  
524 period and maxima during the intense photochemical periods, suggesting significant  
525 formation of both WSOC and WIOC during the afternoon. Applying the EC-tracer  
526 method, they evaluated that freshly-formed secondary OC accounts for more than  
527 50% of OC concentrations measured during the early afternoon period. This fresh  
528 SOC was calculated to be mainly (~60%) composed of WIOC species. The latter  
529 (unexpected) result has been tentatively attributed to low ambient relative humidity  
530 and high anthropogenic volatile organic compounds in Cairo (Favez *et al.*, 2008b).

531

#### 532 4.2.3. Athens

533 Grivas *et al.* (2008) analysed  $\text{PM}_{10}$  concentration data collected by the Greek air  
534 quality monitoring network at 8 sites over the GAA, for the period of 2001-2004.  
535 Daily concentrations, averaged over the whole study period, ranged between 32.3 and  
536  $60.9 \mu\text{g m}^{-3}$  and the four-year average concentration of  $\text{PM}_{10}$  at five sites exceeded the  
537 annual limit value of  $40 \mu\text{g m}^{-3}$ , while most of the sites surpassed the allowed  
538 percentage of exceedances of the daily limit value ( $50 \mu\text{g m}^{-3}$ ). The urban sites were  
539 mainly affected by primary, combustion-related processes and especially vehicular  
540 traffic, as deduced from the examination of the diurnal distribution of particulate  
541 levels and by factor analysis. On the contrary, suburban background sites were subject  
542 to particle transport from more polluted neighbouring areas and secondary particle  
543 formation through gaseous precursors, both processes supported by favourable  
544 meteorological conditions. The association of the  $\text{PM}_{10}$  levels with backward

545 trajectories indicated that a notable part of area-wide episodic events could be  
546 attributed to trans-boundary transport of particles (Querol *et al.*, 2009b).

## 547 **5. Air pollution and impacts.**

548

### 549 *5.1. Climate*

550 In the Mediterranean, aerosols reduce the solar radiation absorption by the sea  
551 by about 10%, alter the heating profile of the lower troposphere and exert a cooling  
552 effect five times higher than the warming induced by the greenhouses gases  
553 (Lelieveld *et al.*, 2002; Vrekoussis *et al.*, 2005). As a consequence, evaporation and  
554 moisture transport, in particular towards North Africa and the Middle East, are  
555 reduced. Satellite observation analysis (Rosenfeld, 2000) supported that aerosols  
556 caused important perturbations to cloud microstructure and convection, probably  
557 decreasing precipitation. Querol *et al.* (2009) analysis of available aerosol data in the  
558 Mediterranean pointed out three very important climate relevant features of the  
559 aerosols in the area: the increasing gradient of dust from the west towards the east; the  
560 change of hygroscopic behavior of mineral aerosols (dust) via nitration and  
561 sulphation; and the abundance of highly hygroscopic aerosols during high insolation  
562 (low cloud formation) periods. Radiative forcing by aerosols also influences the  
563 energy budget of the Mediterranean and the Black Sea, however the consequences of  
564 this are still poorly understood. A changing energy budget and anomalous winds are  
565 expected to influence the ocean circulation (Tragou and Lascaratos, 2003). Therefore,  
566 aerosols may affect several components of the eastern Mediterranean atmosphere-  
567 ocean system including the regional water cycle. These aerosol-generated effects are  
568 already substantial today, even though sulphate from Europe has actually decreased in  
569 the past two decades (Smith *et al.*, 2010) through the abatement of acidification.

570

572 During summer the persistent northerly winds carry large pollution loads from  
573 Europe that can deposit onto the Mediterranean sea, for instance, nitrate and  
574 phosphorus containing aerosols, which affect the water quality and could contribute to  
575 eutrophication (Kouvarakis *et al.*, 2001; Markaki *et al.*, 2003). In addition, O<sub>3</sub> levels  
576 in the regions downwind pollution sources are also often exceeding phytotoxicity  
577 levels (Kourtidis *et al.*, 2002).

578 Furthermore, ageing of aerosols, such as coating of dust by pollution  
579 compounds (Falkovich *et al.*, 2004) or chemical trapping of nitrogen on pollen  
580 particles (Franze *et al.*, 2005), can be harmful for human health. Katsouyanni (1995)  
581 points out that air pollution effects on health, partly determined by specific mixtures  
582 of air pollutants, may be altered by other environmental, behavioural and social  
583 patterns. She also points out that the health effects of the interactions between  
584 pollutants and photochemical oxidants can be enhanced in the Mediterranean under  
585 high temperatures and humidity patterns. She stresses that even if the health effects of  
586 air pollution only slightly increase the risk to an individual, they are likely to be  
587 important for public health because of the ubiquitous exposure of the population.

588 El Mowafi and Atalla (2005) cited that approximately 3% of the GCA  
589 population is chronically exposed to PM<sub>10</sub> levels above 100  $\mu\text{g}\cdot\text{m}^{-3}$ , compared to  
590 48% exposed to 100-50  $\mu\text{g}\cdot\text{m}^{-3}$  and 49 % exposed to 50-5  $\mu\text{g}\cdot\text{m}^{-3}$  PM<sub>10</sub>. Based on  
591 ambient air pollutant concentrations Gurjar *et al.* (2008) have classified Cairo as a  
592 megacity with extremely poor air quality, where measures for air pollution reduction  
593 need to be taken urgently. It is estimated that 10,000 to 25,000 people a year in Cairo  
594 die due to air pollution-related diseases. These findings indicate the significant  
595 benefits that could be achieved by implementing the proper abatement measures to  
596 improve air quality in Cairo.

598 Significant effort is recently paid on understanding atmospheric composition  
599 change in the East Mediterranean due to human activities, supporting the role of the  
600 basin as the ‘pressure-cooker’ of transported air pollution from distant anthropogenic  
601 sources but also from surrounding urban centres. Air masses are mixed and aged in  
602 the area under favourable meteorological conditions with high solar radiation.  
603 Background O<sub>3</sub> observations show an increasing gradient towards the south that  
604 partially compensates O<sub>3</sub> titration by NO<sub>x</sub> in the urban sites. The increased regional  
605 background contribution in Athens, Cairo and Finokalia compared to GIA are in line  
606 with the observed CO/NO<sub>x</sub> molar ratios. In GIA, CO/NO<sub>x</sub> molar ratio is close to that  
607 observed in Mexico City and Tokyo whereas in GCA is double or triple, indicating  
608 significant regional contribution to CO levels. This ratio maximizes at the background  
609 atmosphere ranging from about 100 to more than 300 observed during summertime at  
610 Finokalia, where long range transport is the dominant source for CO. GCA  
611 experiences also high levels of NMVOC that point to a high O<sub>3</sub> formation potential of  
612 NO<sub>x</sub> in this region. Satellite observations of HCHO and CHOCHO seem to indicate  
613 different NMVOC speciation and sources over GCA than over GIA and GAA. Due to  
614 the non linear dependence of O<sub>3</sub> on NO<sub>x</sub> and NMVOC levels, control of NO<sub>x</sub>  
615 emissions is expected to lead to higher O<sub>3</sub> levels and thus O<sub>3</sub> exceedences in the cities.  
616 Available information on NMVOC total amounts, reactivity and chemical speciation  
617 is scarce, although the NMVOC/NO<sub>x</sub> ratio and VOC reactivity is critical for the  
618 build-up of air pollution. CO observations in rural areas are also limited, despite the  
619 key role of CO in O<sub>3</sub> production. There is a clear need of such reliable and systematic  
620 measurements of NMVOC, NO<sub>x</sub> and CO in the region to support modelling of air  
621 pollution and climate impacts.

622 PM, even in the urban regions, is also shown to have a significant contribution  
623 by long range transport of African dust or distance anthropogenic pollution sources  
624 over the region. Data analysis has shown that a significant number of PM  
625 exceedences, registered in Istanbul and Athens as long range transport episodes, are  
626 associated with regional pollution or natural dust transport. PMF analysis of ground  
627 based aerosol chemistry observations indicates that local anthropogenic sources  
628 account for about 60% of PM levels in GIA and an additional 20% of PM levels is  
629 associated with transported anthropogenic pollution. Based on satellite derived AOD,  
630 the local anthropogenic emissions in GAA and GCA have been estimated to  
631 contribute by 15-30% and 25-50% to the total AOD, respectively. These estimates  
632 need to be reconciled with ground based observations. On an annual mean basis, in  
633 the East Mediterranean the background  $PM_{10}$  contains about  $8-12 \mu g m^{-3}$  of  
634 transported mineral dust and an additional  $5-10 \mu g m^{-3}$  is attributed to transported  
635 anthropogenic regional sources and to sea-spray loads. Dust transport increases  
636 towards the east of the basin and dust aerosols are coated by pollution components  
637 that modify their climate relevant properties. The climatic impact of this mixture  
638 remains to be determined. The first limited number of available  $PM_1$  data show that  
639 their composition and variability is tightly linked to the anthropogenic sources in the  
640 area. OC/EC observations help elucidating the ageing of pollution air masses and the  
641 contribution of photochemistry versus primary sources. Further studies of  $PM_1$  mass  
642 and chemical characterisation will elucidate the sources and impact of PM pollution in  
643 the area.

644

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654

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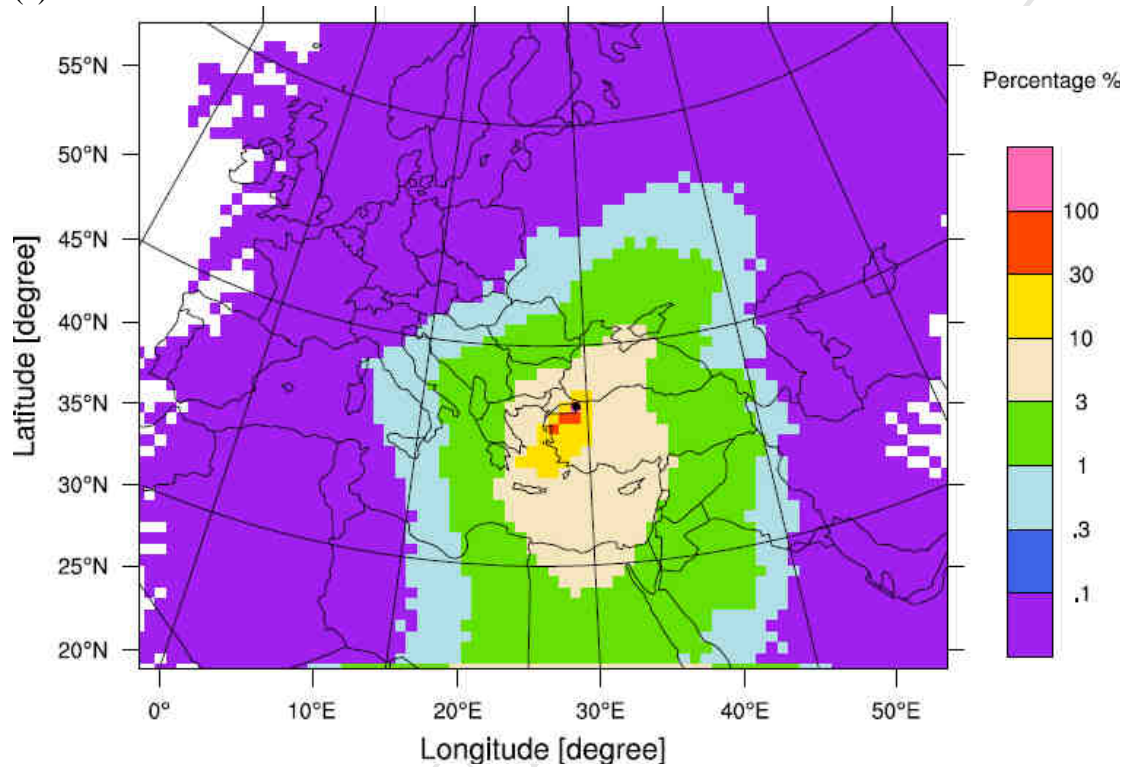
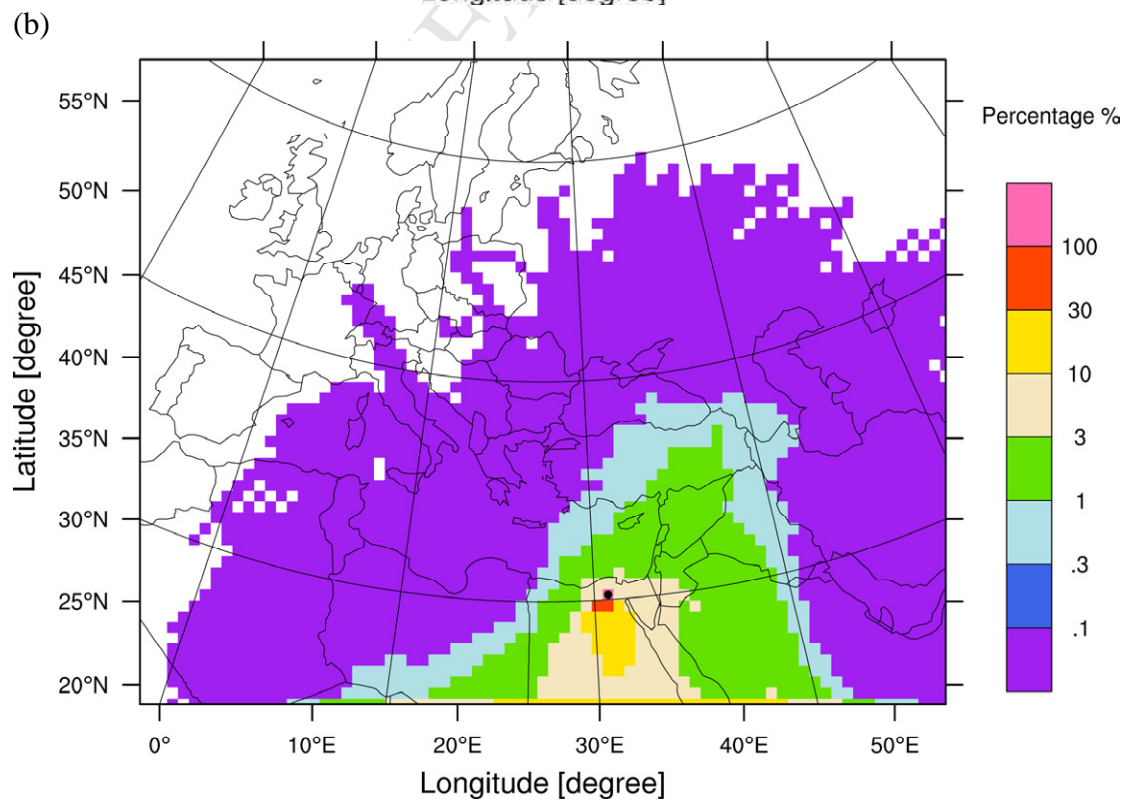
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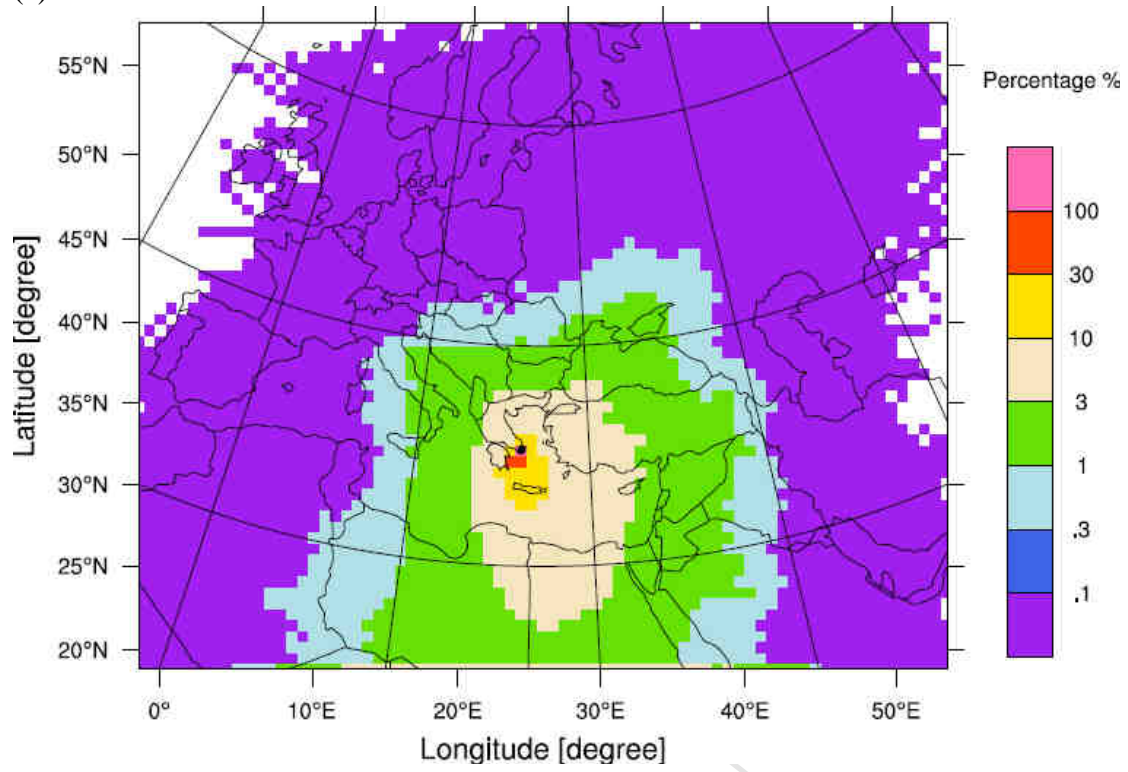
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942 **Figures**

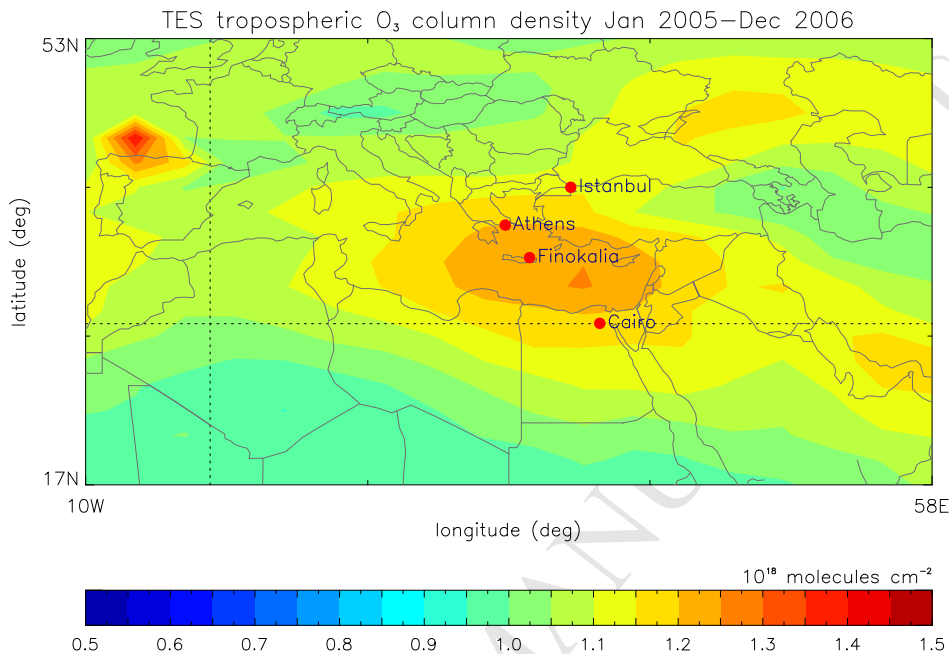
943 Figure 1: Map for the probability of arrival of trajectories starting from (a) Istanbul,  
 944 (b) Cairo, (c) Athens, over the 30 years period based on NCEP 6-hourly  
 945 meteorological data at  $2.5^\circ$  resolution, see text. Dot points indicate the city of  
 946 Istanbul, Cairo and Athens respectively.

947 (a)

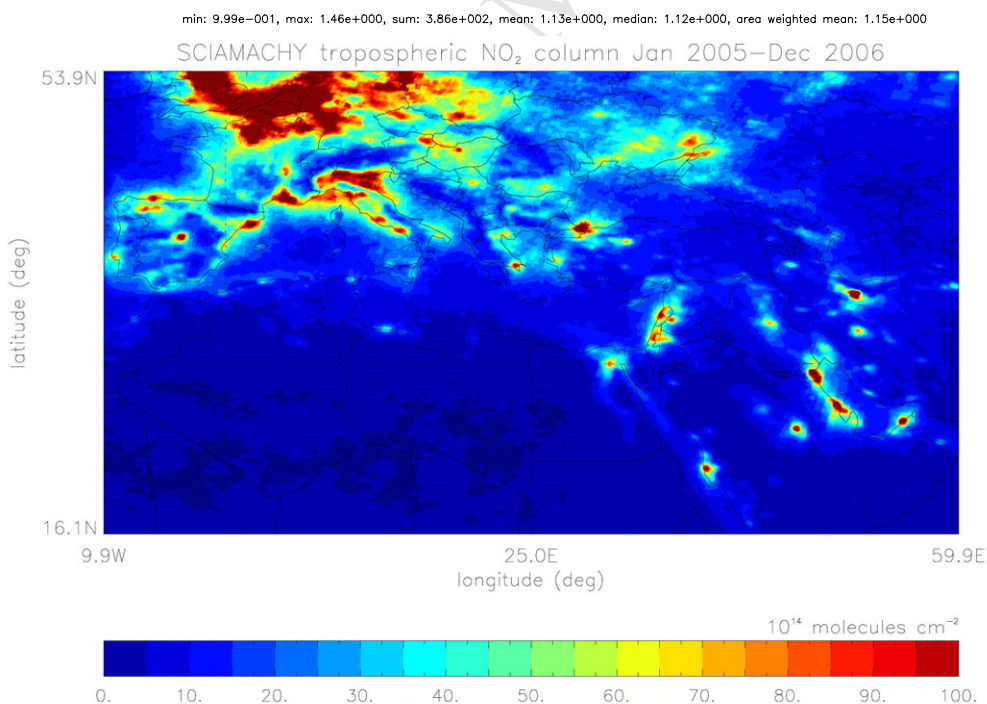
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955 Figure 2: (a) Tropospheric O<sub>3</sub> column as deduced from TES (Tropospheric Emission  
 956 Spectrometer) satellite sensor gridded in 2°x4° lat x lon – The locations of Istanbul,  
 957 Athens, Cairo and Finokalia are indicated; (b): Tropospheric NO<sub>2</sub> column from  
 958 SCIAMACHY; (c) MISR aerosol optical thickness (AOT) at 443 nm in 0.5°x0.5° and  
 959 (c) MODIS aerosol small mode fraction in 1°x1° resolution. Mean columns for the  
 960 years 2005-2006. (a, c, d) have been derived from daily data using the Giovanni  
 961 visualization tool of NASA (Acker and Leptouck, 2007).



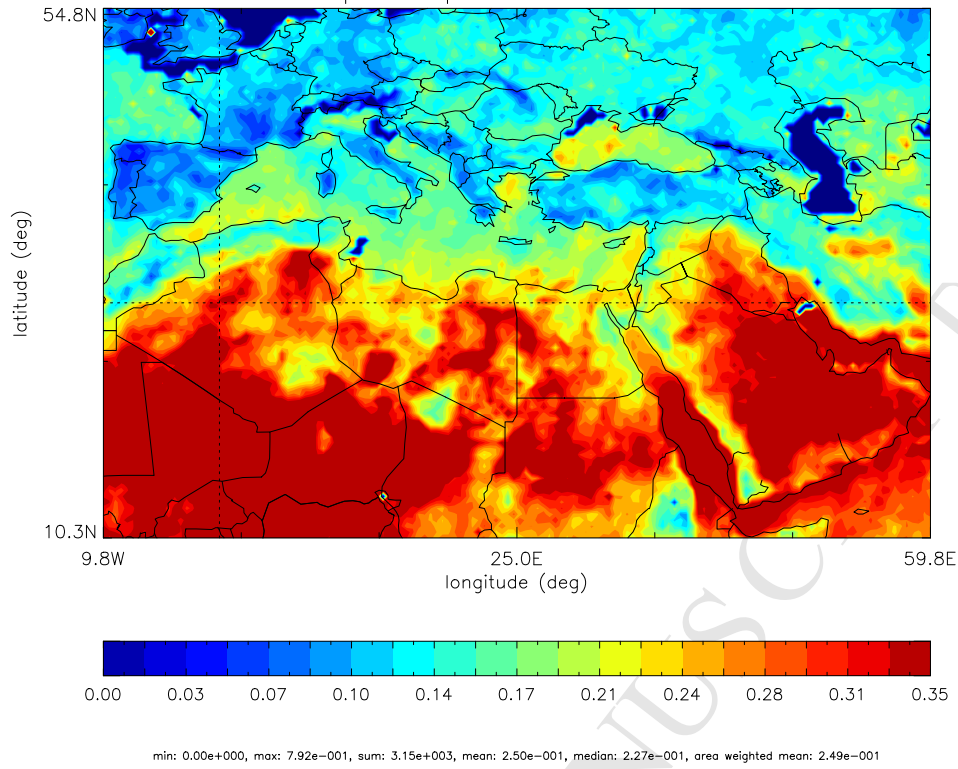
962 (a)



963 (b)

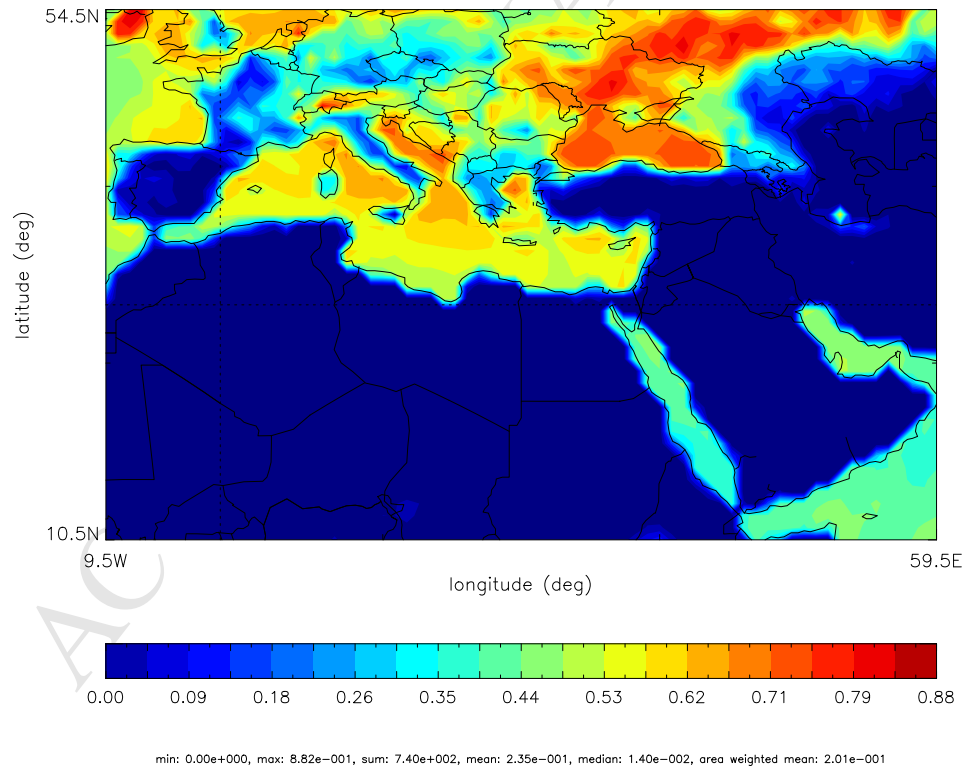
min: \*\*\*\*\*, max: 3.33e+002, sum: 2.78e+006, mean: 1.63e+001, median: 8.33e+000, area weighted mean: 1.44e+001

MISR aerosol optical depth at 443nm Jan 2005–Dec 2006



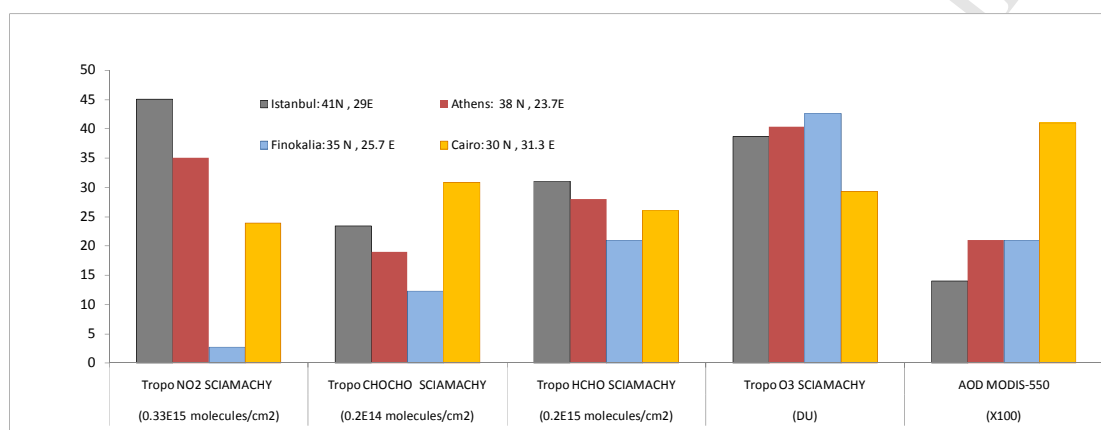
964 (c)

Fraction of fine aerosols MODIS Jan 2005–Dec 2006



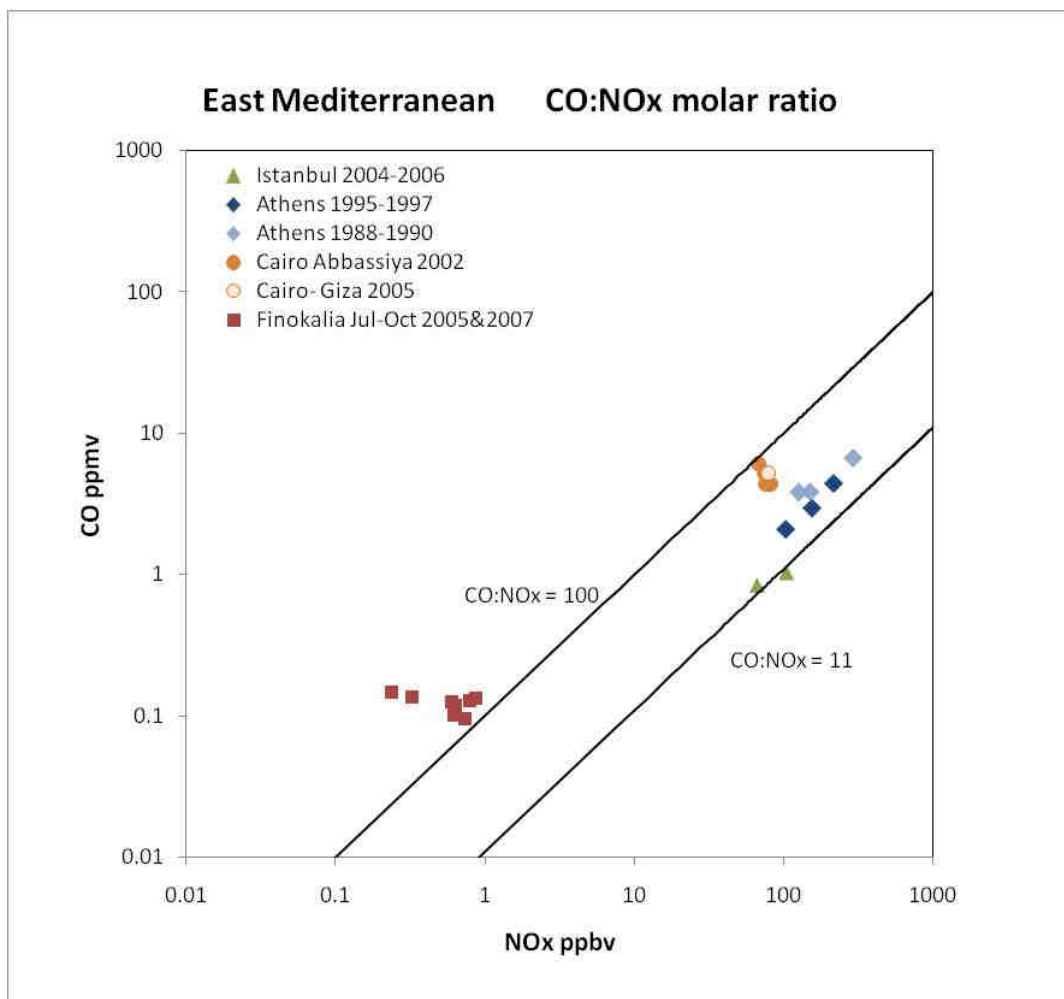
965 (d)

966 **Figure 3.** Satellite observations of air pollutants over GIA, GCA, GAA and Finokalia  
 967 in the East Mediterranean. Mean over the period 2003-2009 from SCIAMACHY:  
 968 Tropospheric columns of NO<sub>2</sub> in 10<sup>15</sup> molecules cm<sup>-2</sup> and CHOCHO and HCHO in  
 969 10<sup>14</sup> molecules cm<sup>-2</sup> (multiplied by 3, 5 and 5 respectively) in a grid of 0.25°x0.25°  
 970 covering the city. Mean tropospheric column of O<sub>3</sub> as deduced from SCIAMACHY  
 971 (2003-2009) based on limb-nadir-matching and mean AOD at 550 nm from MODIS  
 972 (2000-2008) in 1°x1° grid (multiplied by 100).  
 973



974

975 **Figure 4.** Relationship between mean observations of CO (ppmv) and NO<sub>x</sub> (ppbv)  
976 levels in Istanbul, Athens, Cairo and Finokalia based on data reported in Table 3 and  
977 references therein. Lines correspond to the CO:NO<sub>x</sub> molar ratios of 11 and 100.



978

979 Table 1 – Megacities and receptor location (Finokalia, Crete, Greece) characteristics (reference year 2009). General sources: Thomas  
 980 Brinkhoff, 2009; <http://www.worldclimate.com/> (from data prior to 1990) \* Extended region, in brackets highly populated area.

Characteristic	Istanbul	Cairo	Athens	Finokalia
Latitude, longitude	41.01°N, 28.97°E	30.03°N, 31.3°E	37.96°N, 23.71°E	35.33°N, 25.66°E
Continent	Europe-Asia	Africa	Europe	Europe
Surface (km <sup>2</sup> )	6220	8815* (200)	3808* (450)	-
Population (Millions)	12.5	15.2	4.4	-
Ranked as Megacity	21 <sup>st</sup>	16 <sup>th</sup>	-	Background
population growth % over the last decade	45 29.6 (urban parts) 81 (rural parts)	16.4 (all Egypt) 18 (urban – Egypt)	6	-
Typical air temp. (°C)				2001-2009
winter average	8	15	10	11.6
summer average	28	28	26	24.2
Wind speed m/s	last 30 years	1995-2000	1984-2004 – Thissio	2001-2009
Annual mean	2.7	Urban; suburban; rural	3.3	5.8
Winter	3.0	2.2 3.7 3.1	3.4	5.8
summer	2.4	2.1 4.3 3.5	3.5	6.6
Mean precipitation (mm/yr)	800	25	400	350
Type of climate	Mediterranean (southern part) Cooler + wetter (northern part)	Sub tropical	Mediterranean Hot dry summer Wet mild winter	Mediterranean
Heat island (°C max surface temp. change)	1°	≤2.1°	exceeding 4° in 20% of studied cases	-
References	Ezber et al., 2007; Kindap, 2008	Zakey and Omran, 1997; Khoder, 2009; Zakey et al., 2008; Robaa, 2003	Kallos et al., 1993; Melas et al., 1995; Kassomenos & Katsoulis, 2006	Gerasopoulos et al., 2005 ; 2006; Vrekoussis et al., 2006

981 Table 2 – Anthropogenic emissions from Istanbul (reference 2007: Markakis *et al.*, 2009), Athens (reference year 2003; Markakis *et al.*,  
 982 2010a,b) and Cairo (reference year 2005; van Aardenne *et al.*, 2009, Duering *et al.*, 2009; # Cairo inventory concerns PM2.5 emissions)  
 983 greater areas.

	Residential Combustion %	Industry %	Fuel Extr./ Distribution %	Solvent Use %	Road Transport %	Off-road %	Maritime %	Waste %	Energy %	Total Ktons/yr
CO										
Istanbul	10.8	3.7	-	-	83.1	-	0.3	0.7	0.7	437
Athens	8.0	3.2	-	-	75.6	13.0	0.2	-	-	473
Cairo	28.8	31.2			35.5			2.2	2.4	285
NO <sub>x</sub>										
Istanbul	2.1	2.4	-	-	79.4	2.8	9.5	-	3.2	305
Athens	3.1	22.4	-	-	51.0	17.8	3.1	-	2.6	78
Cairo	4.0	50.2			11.4	3.37		0.12	30.9	222
SO <sub>2</sub>										
Istanbul	14.7	23.2	2.3	-	2.3	4.1	17.6	-	35.6	91
Athens	14.9	29.1	8.4	-	3.2	7.2	11.3	-	25.9	31
Cairo	7.6	71.5			4.4					135
NMVOC										
Istanbul	2.6	0.5	-	29.8	44.8	0.4	0.6	20.4	0.2	77
Athens	3.2	2.1	2.0	13.8	70.6	5.7	0.5	-	2.1	93.2
Cairo	11	2.6		43.8	36.9			0.8		62.3
PM10 #										
Istanbul	7.1	64.9	0.1	-	17.4	3.9	3.1	1.7	1.8	61
Athens	18.0	62.7	-	-	13.0	0.8	1.9	3.6	-	21
Cairo #	53.4	4.3			35.9			4.4		6.4

984 Table 3 – Comparison of surface air pollution levels in Istanbul, Cairo, Athens and Finokalia -Crete (background site) in the East  
 985 Mediterranean. PM<sub>10</sub> and PM<sub>2.5</sub> are particles of diameter smaller than 10 and 2.5 microns, respectively.

Pollutant	Season/Date	Average	Location	Reference
O <sub>3</sub> ppbv	1998-2008 2001-2005 2008-2009	<30 8 ± 7 11 ± 8 25.3 ± 16.8 19.9 ± 14.2	Istanbul * Saraçhane- Europe Kadikoy-Asia Buyukada Kandilli	Ozdemir et al. 2009 Im et al., 2009 Im et al., 2009 “
O <sub>3</sub> ppbv	Winter 2005 Spring 2005 Summer 2005 Fall 2005 2002	Day / Dial 44 / 30 65 / 48 91 / 64 58 / 43 23.4	Cairo (Giza)    Abbassiya	Khoder 2009    Elminir et al., 2005
O <sub>3</sub> ppbv	1987–1996 winter (Dec.–Jan.) summer (Jul-Aug)	(12:00-18:00 LT) ~ 25 ~ 60	Athens	Kalabokas and Repapis, 2004
O <sub>3</sub> ppbv	1997-2004 July-Aug. Dec	49 ± 11 58 ± 10 36 ± 7	Finokalia-Crete	Gerasopoulos et al., 2006b
NO <sub>2</sub> ppbv	2001-2005	25± 18.9 (NO: 24± 46.3) 8.8±7.8 (NO: 2 ± 5.8)	Kadıköy Sarachane	Im et al., 2008

NO <sub>2</sub> ppbv	Dec. 2004 - Nov. 2005 (hourly) Winter (hourly) Summer (hourly) 2002	60-150 80-200 (NO: 95-200) 60-130 (NO: 45-125) ~40	Cairo- Giza Cairo- Giza Cairo- Giza Abbassiva	Khoder, 2009 Khoder, 2009 Elminir et al., 2005
NO <sub>2</sub> ppbv	1987-1997	57±5.3 (NO: 140.5±9.6) 18 ±4 (NO:31.9±18.0) 42.6±4.3 (NO:73.5±18.0)	Athens-Patission Maroussi Athinas	Kalabokas et al., 1999b
NO <sub>2</sub> ppbv	June 2001 – Sept. 2003	0.35±0.31 (NO:0.033±0.020)	Finokalia-Crete	Vrekoussis et al., 2006
CO mg m <sup>-3</sup>	2004-2006	1.181± 0.957 0.956±1.233	Sarachane Kadikoy	Im et al., 2008
CO mg m <sup>-3</sup>	2002	~6 (4-10)	Cairo- Abbassiya	Elminir et al., 2005
CO mg m <sup>-3</sup>	1987-1997	6.2± 1.2 1.9±0.6 3.8±0.5	Athens-Patission Maroussi Athinas	Kalabokas et al., 1999b
CO mg m <sup>-3</sup>	July-Oct 2005 and Jul-Oct 2007	~ 0.143	Finokalia-Crete	Unpublished data
SO <sub>2</sub> µg m <sup>-3</sup>	1998-2008	~22 (7.5 - 45)	Istanbul *	Ozdemir et al., 2009
SO <sub>2</sub> µg m <sup>-3</sup>	Winter 1999-2000 Summer 2000	125±21.6 83±17.6	Cairo (Giza)	Khoder, 2002
SO <sub>2</sub> µg m <sup>-3</sup>	1995-1997	25±3 40±4	Athinas- Athens Patission-Athens	Kalabokas et al., 1999b
SO <sub>2</sub> µg m <sup>-3</sup>	1997-1999	2.7±0.9	Finokalia -Crete	Kouvarakis et al., 2002
PM <sub>10</sub> µg m <sup>-3</sup>	Jul 2002-Jul 2003 1998-2008 Nov 2007- Jun 2009	47.1 66 (47 – 115) 39.1	Istanbul * Background- Boğaziçi Univ.	Karaca et al., 2005 Ozdemir et al., 2009 Theodosi et al., 2010
PM <sub>10</sub> (bulk aerosol)	2005: Win., Spr., Sum., Fall	215, 190, 115, 165	Cairo (Giza & El-Gomhoreya)	Favez et al., 2008

$\mu\text{g m}^{-3}$	2001-2002	170 $\pm$ 25 140 $\pm$ 40	Cairo (17 sites) Background -Cairo	Zakey et al., 2008 «
PM <sub>10</sub> $\mu\text{g m}^{-3}$	Jun1999-May 2000	75.5 $\pm$ 27.5	Athens	Chaloulakou et al., 2003
PM <sub>10</sub> $\mu\text{g m}^{-3}$	2001-02 & 2004-05 2004-2006	28 $\pm$ 30 32.5 $\pm$ 27.7	Finokalia-Crete	Gerasopoulos et al, 2006a ; 2007; Koulouri et al., 2008
PM <sub>2.5</sub> $\mu\text{g m}^{-3}$	Jul 2002-Jul 2003	20.8	Istanbul	Karaca et al., 2005
PM <sub>2.5</sub> $\mu\text{g m}^{-3}$	2001-2002	85 $\pm$ 12	Cairo (17 sites)	Zakey et al., 2008
PM <sub>2.5</sub> $\mu\text{g m}^{-3}$	Jun1999-May 2000 2004-2006	40.2 $\pm$ 16.7 23.7 $\pm$ 10.7 29.3 $\pm$ 10.4	Athens-Aristotelous Athens-Lykovrissi Athens-Goudi	Chaloulakou et al., 2003 Koulouri et al., 2008b “
PM <sub>2.5</sub> $\mu\text{g m}^{-3}$	2004-2006	18.2 17.9 $\pm$ 12.4	Finokalia-Crete	Gerasopoulos et al., 2007 Koulouri et al., 2008
OC/EC	Nov 2007- Jun 2009	1.98 (PM <sub>10</sub> )	Istanbul – urban Background	Theodosi et al., 2010
OC/EC	March- April 2005 2005	1.4 $\pm$ 0.3 (morning) 2.9 $\pm$ 0.5 (early afternoon) 2.5 - 5.0 Bulk aerosol	Cairo : El-Gomhoreya and Giza	Favez et al., 2008a « Favez et al., 2008b
OC/EC	June–July 2003	3.9 $\pm$ 0.9 (PM <sub>2.5</sub> ) 24 $\pm$ 17 (PM <sub>2.5-10</sub> )	Athens	Sillanpaa et al., 2006
OC/EC	July 2004-July 2006	4.0 (PM <sub>1.3</sub> ) 4.0 (PM <sub>1.3-10</sub> non-dust cases)	Finokalia- Crete	Koulouri et al., 2008a

986 \*10 municipality stations