CityZen

megaCITY - Zoom for the Environment

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PP	Restricted to other programme participants (including the Commission Services)	
RE	Restricted to a group specified by the consortium (including the Commission Services)	
CO	Confidential, only for members of the consortium (including the Commission Services)	

Model evaluated trends of ozone, PM and deposition patterns over the recent 10 to 20 years, discrimination between anthropogenic and natural contributions.

1. *Air Pollution in the East Mediterranean*

Enhanced levels of pollution (Figure 1, Kanakidou *et al.,* 2011) and increasing trends over the last decade are seen by satellites over East Mediterranean and over the Middle East and Cairo. Background tropospheric ozone (O_3) levels in the area are high, particularly in spring and summer, depending on the meteorological conditions since they are controlled by large-scale, long-range transport and photochemical formation. Background particulate matter (PM) levels are also high due to a significant contribution of Sahara dust aerosol but also transported pollution. In the urban atmosphere due to the high levels of primary pollutants, like PM and nitrogen oxides (NO_x), maintained by the anthropogenic emissions, O_3 titration by reaction with nitrogen oxide (NO) is leading to very low O_3 levels over city centers, whereas NO_x and PM remain high. Primary pollutants decrease downwind where O_3 and secondary aerosols build up photochemically. In the urban regions, the temporal variability of primary gaseous pollutants reflects the high emissions during winter time and the faster photochemical destruction during summer time.

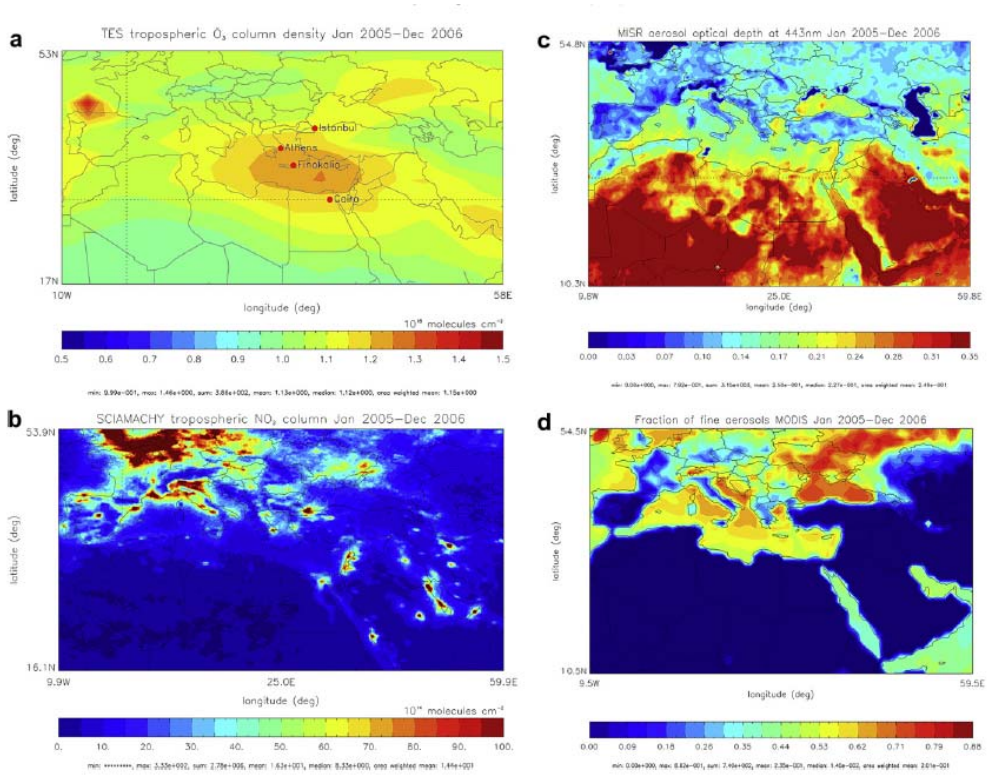

Figure 1: (a) Tropospheric O_3 column as deduced from TES (Tropospheric Emission Spectrometer) satellite sensor gridded in 2^ox4^o lat x lon – The locations of Istanbul, Athens, Cairo and Finokalia are indicated; (b): Tropospheric NO_2 column from SCIAMACHY; (c) MISR aerosol optical thickness (AOT) at 443 nm in 0.5^ox0.5^o and (c) MODIS aerosol small mode fraction in 1^ox1^o resolution. Mean columns for the years 2005-2006. (a, c, d) have been derived from daily data using the Giovanni visualization tool of NASA Figure from Kanakidou et al. (2011).

Mean satellite observations of short lived trace gases (nitrogen dioxide: NO_2, glyoxal: CHOCHO, formaldehyde: HCHO and ozone: O_3) and aerosol optical depth (AOD) over the East Mediterranean

during the recent years are summarized in Figure 2 (Kanakidou *et al.*, 2011). High tropospheric columns of NO_2, HCHO, CHOCHO are observed over urban locations (GIA: Greater Istanbul Area, GCA: Greater Cairo Area, GAA: Greater Athens Area) and low levels over the background receptor site of Finokalia. The progressive reduction of tropospheric columns of NO_2 from Istanbul to Athens and then to Cairo can be noticed together with a similar trend in CHOCHO and HCHO, used as proxy for non methane volatile organic compounds (NMVOC) levels.

Available measurements of ozone in the Eastern Mediterranean at urban and regional background locations show a clear North to South increasing gradient (Kanakidou *et al.*, 2011). In particular, surface O_3 increases when moving from rural background sites of Istanbul to Athens and then to Cairo, indicating significant contribution from long-range transport sources in air masses that age in the region.

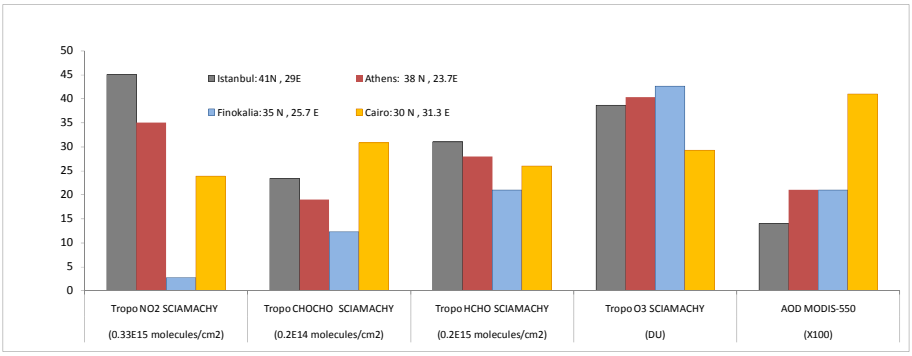

Figure 2. Satellite observations of air pollutants over GIA, GCA, GAA and Finokalia in the East Mediterranean. Mean over the period 2003-2009 from SCIAMACHY: Tropospheric columns of NO_2 in 10^{15} molecules cm^{-2} and CHOCHO and HCHO in 10^{14} molecules cm^{-2} (multiplied by 3, 5 and 5 respectively) in a grid of 0.25^ox0.25^o covering the city. Mean tropospheric column of O_3 as deduced from SCIAMACHY (2003-2009) based on limb-nadir-matching and mean AOD at 550 nm from MODIS (2000-2008) in 1^ox1^o grid (multiplied by 100). Figure from Kanakidou *et al.* (2011).

2. Summertime air quality levels and their dependence on temperature

The origin of the gaseous and particulate air pollutant levels in the Eastern Mediterranean has been investigated using the WRF/CMAQ mesoscale modeling system coupled with the MEGAN model for the processing of biogenic volatile organic compound emissions (Figure 3a).

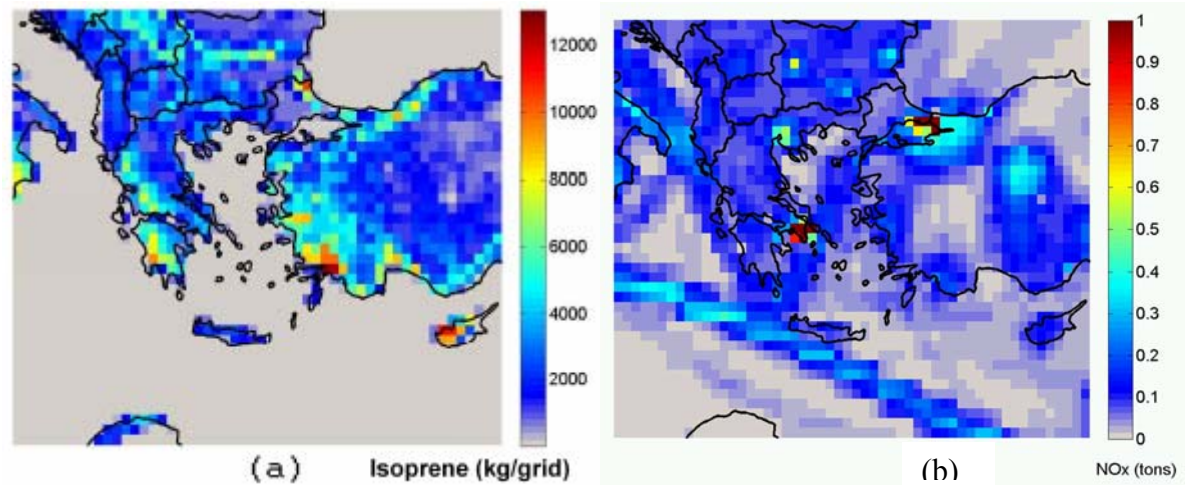

Figure 3. (a) Isoprene emissions as computed by the MEGAN model and (b) NOx emissions used in the WRF/CMAQ model for July 2004. NOx emission distribution clearly depicts the shipping lines and the large urban agglomerations of the region.

Im et al. (2011a) focused on July 2004 when isoprene observations are also available for the region. In agreement with observations (Kanakidou et al., 2011), the model results indicate that the Eastern Mediterranean basin acts as a reservoir of pollutants and their precursor emissions from large urban agglomerations (Figure 3b). During summer, chemistry is a major sink at these urban areas near the surface, and a minor contributor at downwind areas.

The CO/NOx ratio is an indicator of emission composition and air mass ageing (Figure 4). Due to the short lifetime of NOx compared to CO, low CO/NOx ratios indicate high contribution by local emissions whereas high ratios point to important contribution of transported air masses. The model-calculated CO/NOx ratios (figure 4) increase from below 50 in the large agglomerations to above 150 downwind, due to influence from the surrounding region, which is consistent with the observed pattern (Kanakidou et al., 2011). The low ratio in Istanbul indicates significant local influence whereas in Athens, regional influence is much stronger.

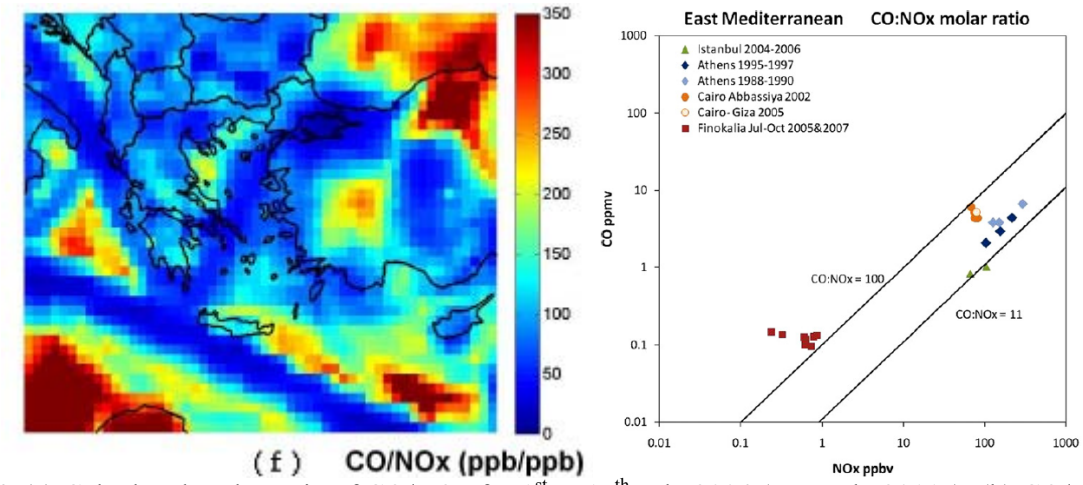

Figure 4. (a) Calculated molar ratio of CO/NOx for 1st to 15th July 2004 (Im et al., 2011a); (b) CO/NOx ratio derived from observations, lines correspond to molar ratios of 11 and 100 (Kanakidou *et al.*, 2011).

Furthermore, Im et al. (2011b) have simulated $PM_{2.5}$ to PM_{10} ratios ($PM_{2.5}$ and PM_{10} are PM with diameters smaller than 2.5 μm and 10 μm, respectively) that decrease from >80% over the urban regions to 45% downwind (Figure 5) and that the organic aerosols dominate urban sites and inorganic aerosols downwind sites.

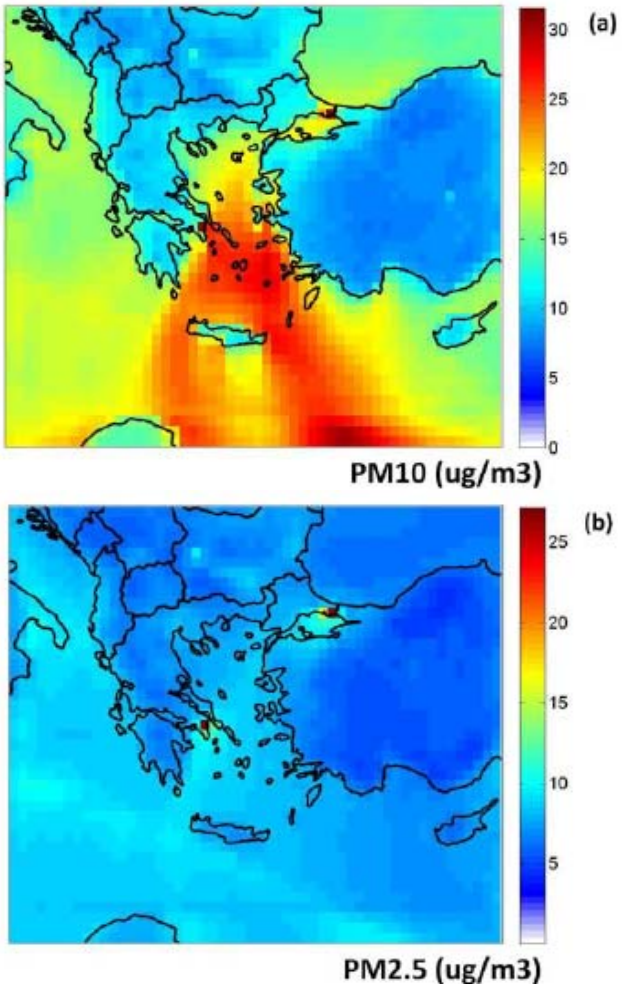

Figure 5. Simulated PM10 and PM2.5 for 1st -15th July 2004 that depicts the maximum of fine particles over the urban locations and high contribution of the coarse mode over the south Aegean during southward transport (Im et al., 2011b).

The Mediterranean is a climate sensitive region and includes megacities like Istanbul and large urban agglomerations such as Athens. **Changes in temperature (of relevance in a warmer future climate)** due to variability in meteorology and climate change are expected to significantly impact atmospheric composition. The effect of temperature changes on gaseous and particulate air pollutant levels in the Eastern Mediterranean has been investigated using the WRF/CMAQ mesoscale modeling system coupled with the MEGAN model for the processing of biogenic volatile organic compound emissions. A set of temperature perturbations (spanning from 1 to 5 K) has been investigated for July 2004 conditions.

Higher temperatures increase biogenic volatile organic and sea-salt emissions. Thus biogenic emissions increase by 9% K^{-1}. For the studied temperature perturbations, the **regional O_3 mixing ratios** increase almost linearly with the increases in ambient temperatures **by 1±0.1 ppb O_3 K^{-1}**. Smaller increases (about half) are computed for the highly polluted Istanbul area (Im et al., 2011a).

Temperature increase also lead to higher biogenic secondary organic aerosol (by 10±2 ng m^{-3} K^{-1} that is about 0.8% K^{-1}) and nitrate aerosol concentrations (by 23±16 ng m^{-3} K^{-1} that is about 3.2% K^{-1}). On the opposite, the impact of temperature increases on sulfate aerosols is calculated to be -70±110 ng m^{-3} K^{-1} (that is about -1.1% K^{-1}), induced by changes in the aqueous-phase production resulting from high reduction in the cloud cover (90% K^{-1}). Overall, **a small increase of domain-mean PM_{10} concentrations by 46±43 ng m^{-3} K^{-1} (about 0.3% K^{-1}) is calculated**. Results show that changes in temperatures modify not only the aerosol levels but also the chemical composition.

3. Natural versus anthropogenic contributions in the area.

The Mediterranean is one of the areas with the highest AOD in the world, also seen from space (Hatzianastassiou et al., 2009), which presents high temporal variability due to the short lifetime of PM in the troposphere (of the order of a week). Two-year (2005-2006) mean observations of AOD at 443 nm over the area from MISR (Multiangle Imaging Spectro Radiometer) and of the aerosol small mode fraction derived from MODIS (Moderate Resolution Imaging Spectroradiometer, using the Giovanni daily data of NASA GES DISC), are depicted in Figs. 1c,d.

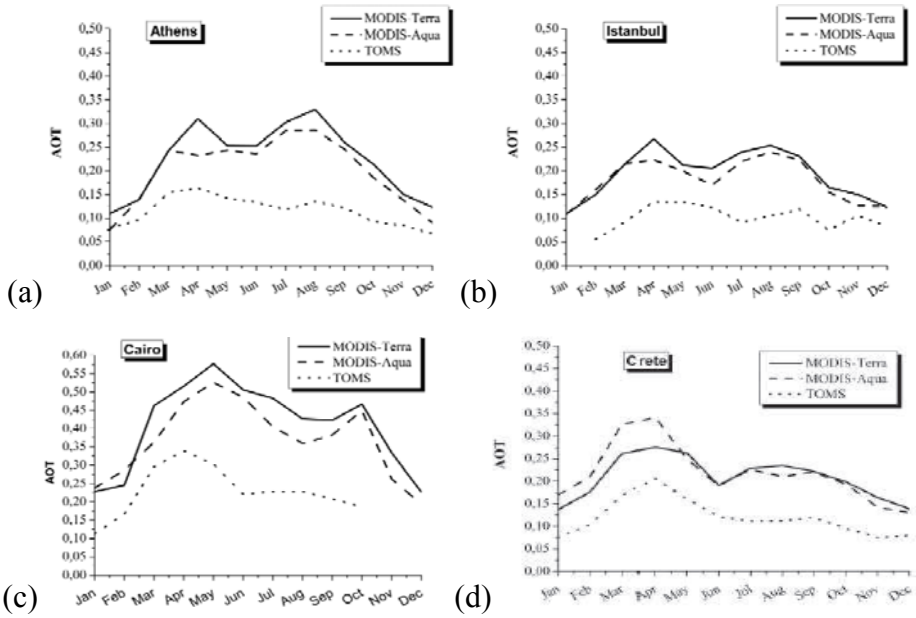

Figure 6 Intra-annual variation of visible aerosol optical thickness (AOT) over: (a) greater Athens area (37°N-39°N, 23°E-24°E), (b) greater Istanbul area (40°N-42°N, 28°E-30°E), (c) greater Cairo area (29°N-31°N, 30°E-31°E), and (d) Crete (34°N-36°N, 24°E-26°E), based on data taken from MODIS-Terra (2000-2005, solid lines), MODIS-Aqua (2002-2005, dashed lines) and TOMS (1980-2001, dotted lines). The AOT values are provided at λ = 500 nm and λ = 550 nm for TOMS and MODIS, respectively.

Synergistic analysis of MODIS-AOT (AOT: aerosol optical thickness) and aerosol index TOMS data, used as proxy for absorbing dust aerosol, enabled a first evaluation of the local anthropogenic contribution to the AOT (since the difference of MODIS-TOMS is indicating the anthropogenic aerosol component) **over the Greater Athens Area and Greater Cairo Area at 15-30% and 25-50%, respectively. This contribution maximizes during summer when in Cairo urban sites local anthropogenic emissions contribution to PM levels can reach almost 100%** (Figure 6; Hatzianastassiou et al., 2009).

Water-soluble ions, water soluble organic carbon, organic and elemental carbon, and trace metals were measured in aerosol PM_{10} samples above the **megacity of Istanbul between November 2007 and June 2009** (Theodosi et al., 2011). Source apportionment analysis using Positive Matrix Factorization indicates that approximately 80% of the PM_{10} is anthropogenic in origin (secondary, refuse incineration, fuel oil and solid fuel combustion and traffic). Crustal and sea salt account for 10.2 and 7.5% of the observed mass, respectively. In general, anthropogenic (except secondary) aerosol shows higher concentrations and contributions in winter. Mean concentration and contribution of crustal source is found to be more important during the transitional period due to mineral dust transport from North Africa. **During the sampling period, 42 events exceeding the limit val-**

ue of 50 μg m^{-3} are identified. A significant percentage (91%) of these exceedances is attributed to anthropogenic sources, 1% to natural sources and the remaining 8% has mixed origin. Potential Source Contribution Function analysis highlights that Istanbul is affected from distant sources from Balkans and Western Europe during winter and from Eastern Europe during summer. On the other hand, Istanbul sources influence western Black Sea and Eastern Europe during winter and Aegean and Levantine Sea during summer (Kocak et al., 2010).

Furthermore, simulations of the impact of anthropogenic emissions from Istanbul with WRF/CMAQ for July 2004 have shown that **Istanbul anthropogenic emission have significant regional impact** (Figure 7).

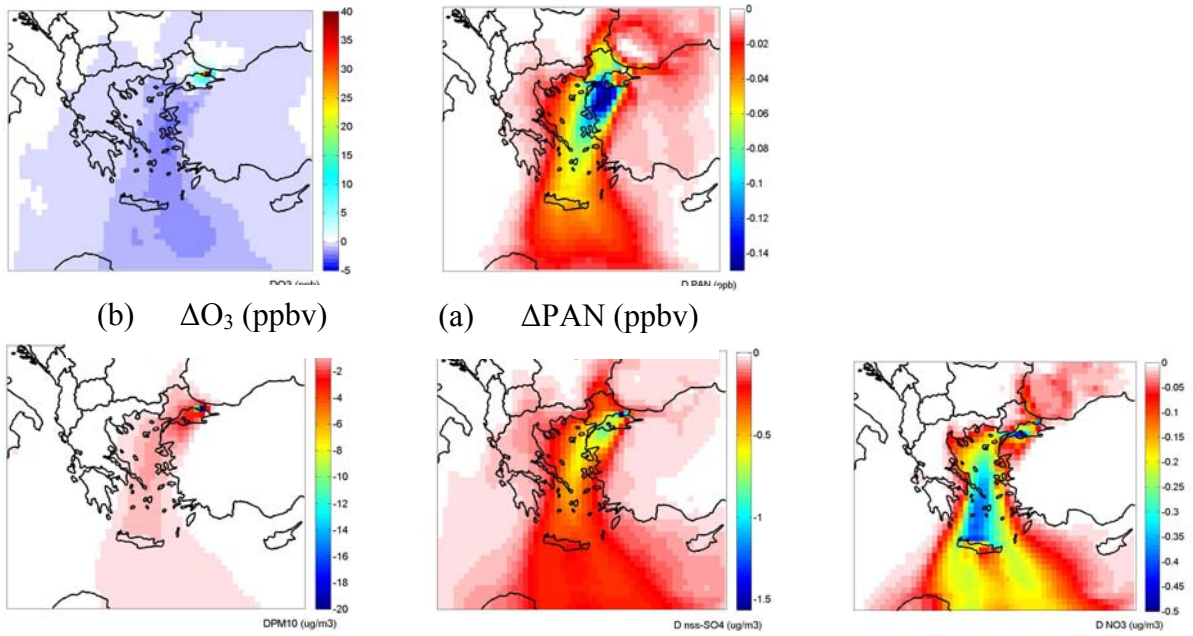

Figure 7. Impact of anthropogenic emissions of Istanbul on mean surface (a) O_3 (ppbv), (b) PAN (ppbv) , (c) PM10 (μg m^{-3}), (d) Δnss-SO_4^{2-} (μg m^{-3}), (e) ΔNO_3^- (μg m^{-3}) levels averaged over the period 1st-15th July 2004. (Im et al., in preparation 2011c).

Istanbul anthropogenic emissions inside the megacity itself are responsible for about 20% reduction in O_3 due to titration by the local NO_x anthropogenic emissions (Figure 7a), whereas in the outflow downwind the megacity over the Aegean Sea they appear to contribute to the 15-day averaged simulated air pollution levels by about 2 ppbv of O_3, 0.1 ppbv of PAN, 1-2 μg m^{-3} of PM_{10}, more than 0.5 μg m^{-3} of nss-SO_4^{2-} and more than 0.5 μg m^{-3} of NO_3^-, during summer. These increases deteriorate the air quality of the region and can lead to increased number of exceedences of the allowed by European Union limits for O_3 and PM_{10} since these pollutants present their seasonal maxima during summer. Note also that the main outflow from Istanbul occurs in the North-East to South-West direction and coincides with the corridor of the Etesian winds that are typical wind patterns for the Aegean Sea in the East Mediterranean during summer.

4. Interannual changes in air pollutants

Trends of **aerosols** (PM_{10}, AOD) in Athens, Istanbul, Cairo and Finokalia (as estimate of regional background) have been calculated from in situ measurements and satellite (MODIS) observations

(Figure 8). In-situ measurements in each megacity were first grouped to urban traffic (Urb-TRF), suburban industrial (Sub-IND) and suburban background (Sub-BG), whereas MODIS Aerosol Optical Depth (AOD) is used for the quantification of the columnar atmospheric content in aerosols, additionally discriminated to fine and coarse particles.

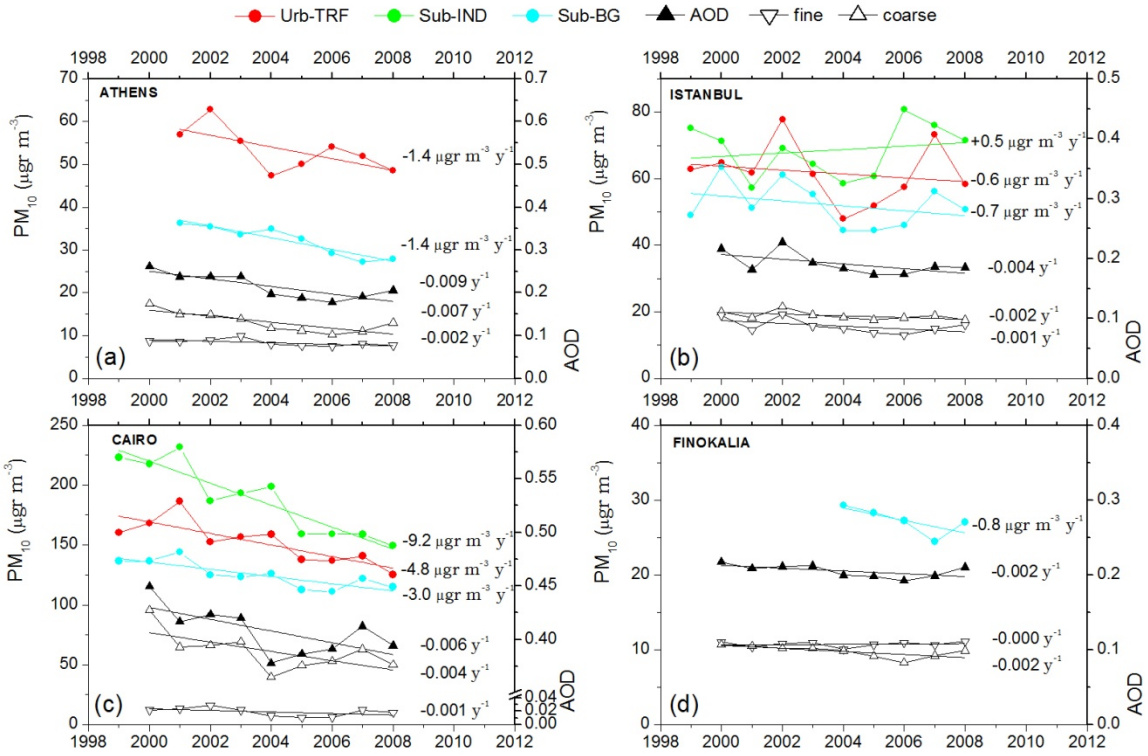

Figure 8. Observed changes in annual mean PM_{10} levels at various sites in (a) Athens (Greece), (b) Istanbul (Turkey), (c) Cairo (Egypt) and at (d) Finokalia on the island of Crete, Greece since 2000. Urb-TRF (Urban-traffic sites), Urb-BG (Urban background), Sub-IND (Suburban-Industrial), Sub-BG (Suburban- Background) (Vrekoussis et al., in preparation 2011).

In Athens a similar decreasing trend (1.4 μg m^{-3}) is found in both available site-type clusters in conjunction with the decrease in AOD, which is attributed to a decline in coarse particles rather than fine.

In Istanbul a decreasing trend is derived only for urban traffic and suburban background sites, with a slope half than that observed in Athens. The same result is also confirmed from AOD observations revealing a slight decrease in both fine and coarse particles. On the contrary, industrial sites show an overall tendency to increase (0.5 μg m^{-3}), however, the enhanced interannual variability of the values should not be disregarded during the evaluation of the trends.

Significant trends are revealed in Cairo which presents the highest levels in PM_{10} compared to the other cities. The decrease recorded at industrial sites is 9.2 μg $m^{-3}yr^{-1}$ and this seems to affect also particulate matter levels in the neighboring sites. Even though the trends of surface PM_{10} are remarkably greater than e.g. in Athens, this is not seen in AOD which shows moderate decline, once more attributed to coarse particles.

The case of Finokalia, studied as a regional background reference but also as receptor point of the regional megacities pollution, reveals small decline in both surface and columnar aerosol loads.

The trends of **trace gases** (NO, NO_2, O_3, CO, SO_2) have been investigated for the Greater Istanbul Area (GIA) and the Greater Athens Area (GAA) (Figure 9). For GAA, the observations since 1985 from fourteen monitoring stations of the Greek Ministry of Environment air pollution monitoring network have been collected and categorized into four different groups namely urban traffic (Urb-Trf), urban-Background (Urb-Bg), suburban industrial (sub-ind) and suburban background (Sub-BG). For the GIA the observations of ten stationary monitoring stations operating since 1998 have been categorized to up to three subgroups (urban traffic data, suburban industrial and suburban background) depending on the availability of the observations.

NO_2 : Nitrogen dioxide over Athens shows a decreasing trend for the urban traffic regions. The highest decline in NO_2 levels is found for the period 1985-1994. For the present period (2003-2008) this decline is almost negligible. The other three categories encountered an overall increase in NO_2 levels from 1985-1994 and a decrease of about 1 μg m^{-3} NO_2 per year for the years 1995-2008. Contrary, for GIA an important increase of 8 μg m^{-3} NO_2 per year is recorded for the suburban background areas during 2003 to 2008.

NO: The mean annual values of nitrogen oxide over Athens indicate a significant decrease on their levels from 1985 to 2008. Especially for the urban traffic regions a reduction by 42% has been observed during this period; the highest reduction in μg m^{-3} occurred from 1994-2002. In Istanbul the NO measured at the suburban traffic areas decreased significantly (≈60%).

O_3: Ozone trend is less profound than the nitrogen oxides' one. The highest values are observed for the suburban background regions and the lowest for the urban traffic ones due to the titration with the NOx species. On average ozone remained constant throughout the last 25 years except for the urban background areas where an important increase of about 35 μg m^{-3} has been recorded. A more straightforward behaviour of the ozone monitored at the urban traffic areas of Istanbul showing an overall increase of 2 μg $m^{-3}y^{-1}$.

CO: Carbon monoxide levels are constantly dropping for the urban traffic areas of Athens presenting a decrease of 65% during the last 25 years. A smaller scale decrease is observed for the other three categories. For GIA, CO presented an increasing trend until 2003 and a decreasing one until present time. For both areas the CO levels of the urban traffic regions are greater than the ones of the other categories.

SO_2: For the GAA the increase of the sulfur dioxide levels during 1998-1992 in almost all groups of interest has been followed by a drastic decrease mainly during the decade 1993-2002. Then the SO_2 levels have been stabilized at about 15 μg m^{-3}. A continuous decrease in SO_2 levels has been recorded also for the GIA from 1998 to 2008. Especially for the industrialized areas this decrease is drastic and equals to about 90%.

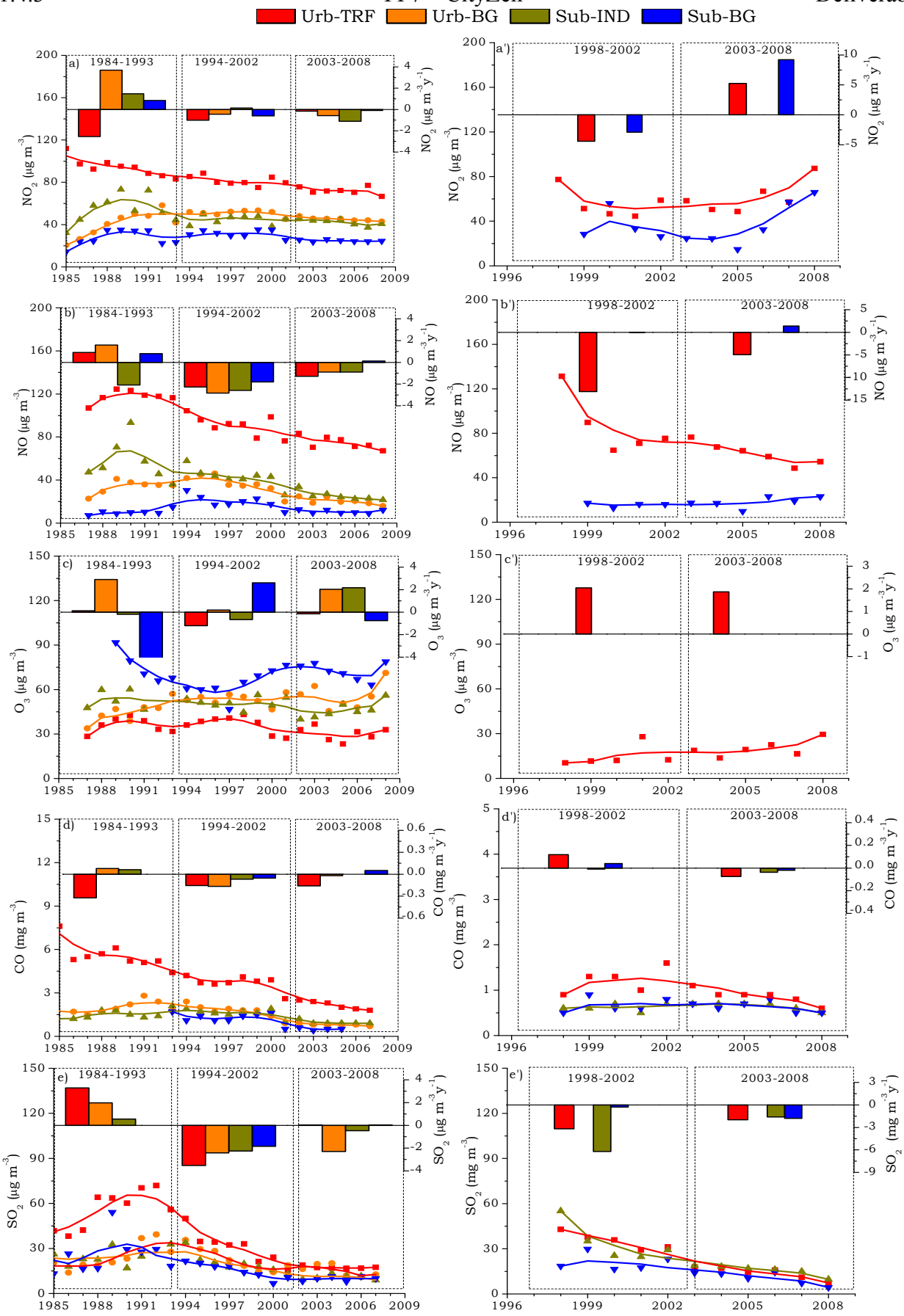

Figure 9: Panels (a) to (e) depict the interannual variability of the concentrations of NO_2, NO, O_3, CO and SO_2 over Athens and panels (a') to (e') the same over Istanbul. The upper part of each pannel shows the observed changes of the trace gases concentration per year attributed to the dotted time period. Red colour scattered lines and bars correspond to urban-traffic sites, orange to urban background sites, green to suburban-industrial sites and blue c to suburban-background sites (Vrekoussis *et al.,* in preparation, 2011).

Interannual Simulations for the East Mediterranean- Simulations have been performed with the global model TM4-ECPL (Myriokefalitakis et al., 2010; 2011) using ERA-interim meteorology for the years 2000-2008 (Daskalakis *et al.,* in preparation 2011). Updated anthropogenic emissions (in particular for the Mediterranean), biomass bunring emissions from gfed-v2.0, the biogenic emissions from the POET database, monthly and annually varying dust emissions compiled for AEROCOM have been adopted and seasalt and marine organic aerosol emissions have been interactively calculated.

TM4-ECPL model results are compared in Figure 10 with O_3 observations at EMEP stations with focus on coastal sites in the Mediterranean and its Eastern basin. The model is satisfactorily capturing the O_3 variability and levels in the studied background stations. It also satisfactorily simulates the PM_{10} levels at Finokalia, Greece (remote receptor site, Figure 11). Similar conclusions can be drawn for background stations in the suburbs of Athens (Figure 12) where, however, PM10 levels are somehow underestimated partially due to dust re-suspension that is not satisfactorily represented in the global model. Aerosol chemical composition investigation will highlight theses issues. Further targeted simulations with different emission scenarios/meteorology and comparison to observations are being performed to analyze these observed and simulated patterns and link them to past anthropogenic emission changes due to measures taken for air quality improvement.

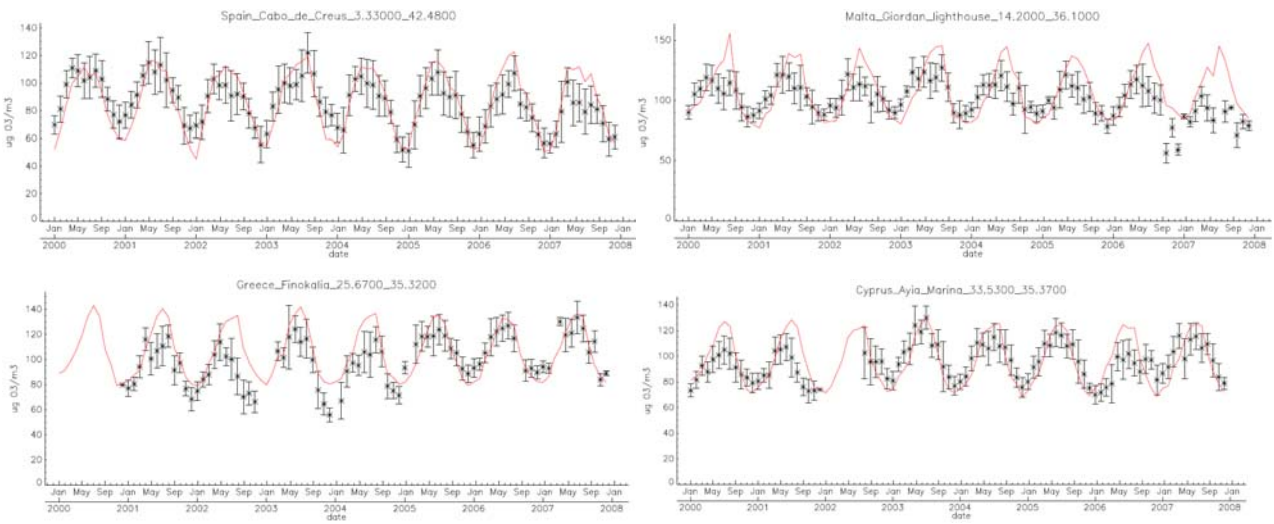

Figure 10. Simulated and observed interannual variability in background surface O_3 in the Mediterranean –coastal locations. top left: Cabo de Creus, Spain; top right: Malta; bottom left: Finokalia, Greece; bottom right: Agia Marina, Cyrpus (in $\mu g/m^3$) (Daskalakis *et al*., in preparation 2011)

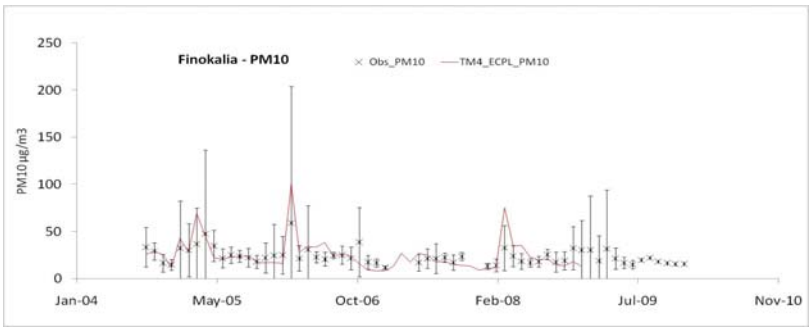

Figure 11. Simulated and observed interannual variability in background surface PM_{10} at Finokalia, Greece.

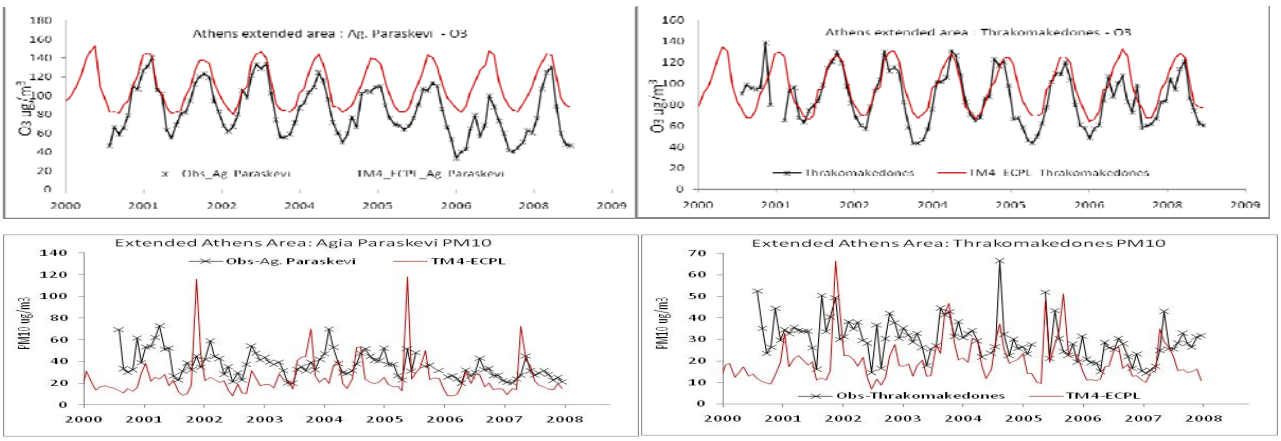

Figure 12. Simulated interannual variability of surface O_3 (top) and PM_{10} (bottom) in background stations (suburbs) in the Athens extended area. Left panels: Agia Paraskevi; right panels: Thrakomakedones.

5. Atmospheric Deposition in the East Mediterranean

In the Eastern Mediterranean basin, where nutrient riverine inputs are negligible, atmospheric inputs of nitrogen (mainly as nitrate, NO_3^-, and ammonium, NH_4^+) and phosphorus (as phosphate, PO_4^{-3}) are believed to be the main source of nutrients in the euphotic zone of the open sea, other than the vertical mixing of water during winter. Deposition of airborne Dissolved inorganic phosphorus (DIP) present in atmospheric aerosols as phosphate has been measured from June 2001 to May 2005 (Markaki et al., Marine Chemistry, 120 (1-4): 187-194, 2010). Wet deposition fluxes of Dissolved Inorganic Nitrogen (DIN) have been derived from rainwater samples (collected on an event basis). Particulate nitrate (NO_3^-) and ammonium (NH_4^+) have also been measured. Thus, dry deposition fluxes have been inferred either from observations of size-segregated atmospheric aerosol concentrations and deposition velocities obtained from literature, or after collection of bulk atmospheric aerosols on glass beads. Details for the sampling and analytical procedures are given in Markaki et al. (Limnology Oceanography, 48(4):1557-1568, 2003) and Kouvarakis et al. (Global Biogeochem. Cycles, 15: 805–818, 2001).

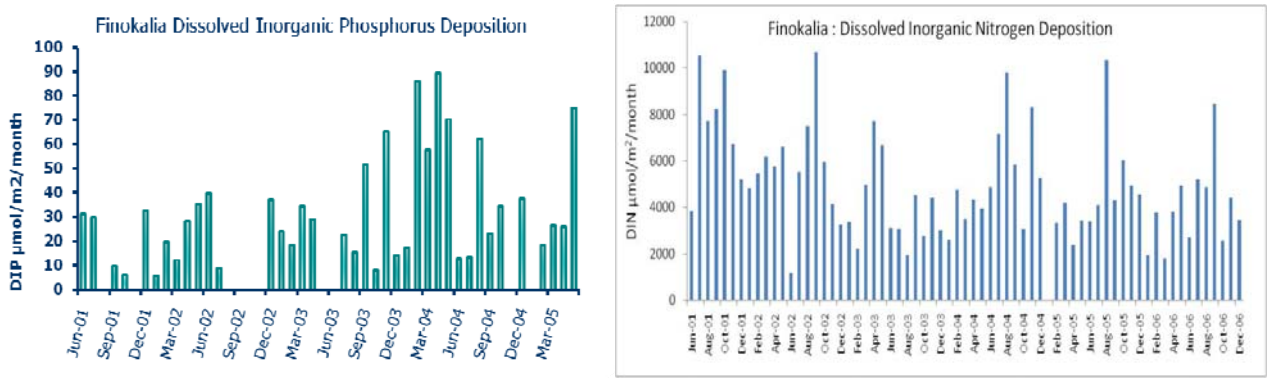

Figure 13. Observed inorganic nitrogen deposition monthly and interannnual variability at Finokalia in the East Mediterranean

These observed deposition rates are used in marine ecosystem models for the region to evaluate their impact on ecosystems. They are also used to evaluated TM4-ECPL (Figure 14, left panel) and WRF/CMAQ (Figure 14, right panel) modeled deposition rates over the region.

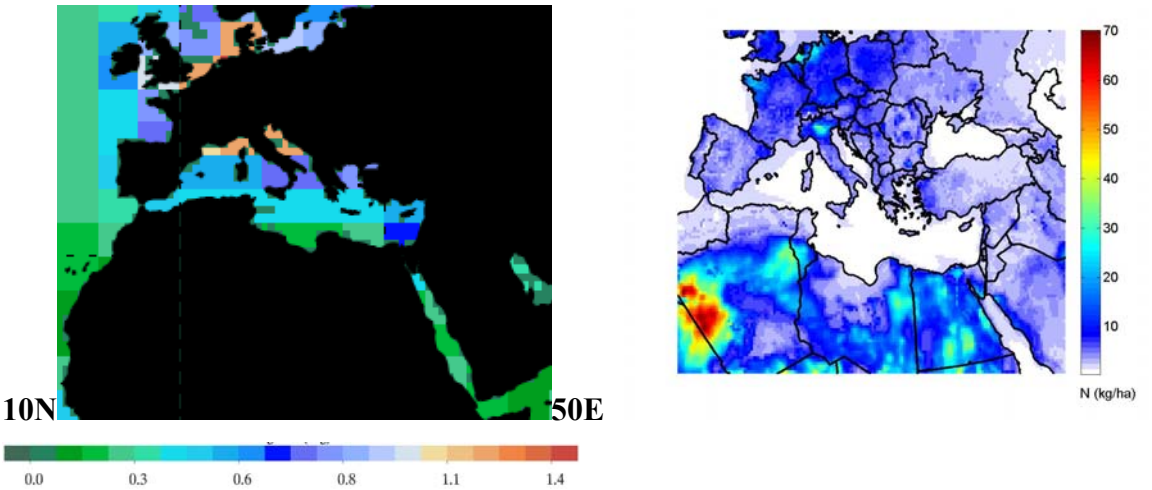

Figure 14. left panel : Total annual N deposition (g-N/m2/y) over the sea in Europe and in the East Mediterranean calculated by TM4-ECPL model; right panel: Total annual N deposition (kg/ha) over Europe and in the East Mediterranean calculated by WRF/CMAQ model.

Publications (issued from the project)

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