CityZen

megaCITY - Zoom for the Environment

Collaborative Project

7th Framework Programme for Research and Technological Development

Cooperation, Theme 6:

Environment (including Climate Change)

Grant Agreement No.: 212095

Deliverable D1.4.4, type R

Report on import/export budgets from the East-Mediterranean area at the regional and global scales

Due date of deliverable: project month 36

Actual submission date: project month 36

Start date of project: 1 September 2008 Duration: 36 months

Scientist(s) responsible for this deliverable: ECPL

Authors: Maria Kanakidou (ECPL), Stelios Myriokefalitakis (ECPL), Nikos Daskalakis (ECPL), Ulas Im (ECPL), Nikolaos Mihalopoulos (ECPL)

Project co-funded by the European Commission within the Seventh Framework Programme (2007-2013)		
Dissemination Level		
PU	Public	X
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)	

Report on import/export budgets from the East-Mediterranean area at the regional and global scales

Executive summary

The East Mediterranean is a cross road of civilizations and air masses from the surrounding regions. Due to its location and the particularly warm and sunny climate, the East Mediterranean is receptor and chemical cooker of transported air pollution.

Main local anthropogenic sources in the region are from traffic, combustion and energy generation. Megacities local pollution emissions are added to high background air pollutant levels. This high regional background is due 1) to long range transport from upwind pollution or dust sources, 2) to interactions of transported air masses with regional emissions.

Among the most important interactions are those of nitrogen oxides with emissions from vegetation, further increasing secondary pollutant levels such as O_3, nitrate, sulphate and secondary organic aerosols, and those of dust with acidic pollutants, like nitrate and sulphate that change aerosol properties and impacts.

Overall, air quality in the East Mediterranean is strongly affected by sources other than the local anthropogenic sources. Thus inter-country influences and natural contributions need to be documented. Improvement in air quality in the East Mediterranean requires coordinated effort between the countries surrounding the basin as well as the upwind located countries since their emissions affect the basin via long range transport and chemical aging of air masses.

The results clearly demonstrate important imports of pollutants to the boundary layer via long range transport within the FT followed by subsidence to the BL. In particular for boundary layer O_3, local pollution contributes by about 8% to the annual mean and by about 35% to its net chemical production; whereas it has smaller impact on the O_3 budget in the free troposphere (FT). Furthermore, 30-40 times higher amounts of O_3 are transported in and out the FT over the region than are chemically produced in the BL. Transport from the FT to the BL is a major source for the BL O_3 two to three times the net chemistry.

1. Evaluation of import/export fluxes to/from the East Mediterranean – Global TM4-ECPL model results

1.1 Method

The budgets of air pollutants in the East Mediterranean region have been investigated using the global chemistry-transport model TM4-ECPL (Myriokefalitakis et al., 2011 and references therein). The model is able to simulate ozone chemistry including non-methane volatile organics together with all major aerosol components, including secondary aerosols like sulphate, nitrate and secondary organic aerosols. It also accounts for multiphase chemistry that affects secondary organic aerosol formation in clouds and in the aerosol water. The model has been evaluated for its ability to simulate atmospheric composition in the global atmosphere and in particular, glyoxal levels (Myriokefalitakis et al., 2008), organic aerosols, including marine organics and methanesulfonate (Myriokefalitakis et al., 2010; 2011a) and water soluble organic fraction of organic aerosol and specifically oxalate (Myriokefalitakis et al., 2011a). For the present study, the model has been also evaluated for its ability to simulate gas phase air pollutants in rural and remote atmosphere (see Deliverable 1-4-3 and Daskalakis et al., 2011 in preparation).

TM4-ECPL driven by ECMWF ERA-interim meteorology for the year 2008 has been used to calculate tropospheric budget terms for all air pollutants taken into account in the model. Budgets have been calculated separately for the boundary layer (up to 850 hPa) and for the free troposphere (up to 250 hPa). Specifically, for the present study, pollutant fluxes through the boundaries of the studied region have been also calculated as well as between the boundary layer and the free troposphere and between the free troposphere and the upper layers of the model (Fig. 1). All terms are calculated every model time step and averaged or integrated appropriately.

Two emission scenarios have been studied with this model configuration: The base case simulation (S0) using anthropogenic and biomass burning emissions specific for the year 2008 (Daskalakis et al., 2011 and Myriokefalitakis et al., 2011b in preparation) and natural emissions specific of the year 2000, as described in Myriokefalitakis et al. (2011a) and references therein. For the second scenario (S1), all anthropogenic emissions in the East Mediterranean region have been masked. The region defined for the present study is the area from 17°E to 40°E in longitude and from 25°N to 47°N in latitude.

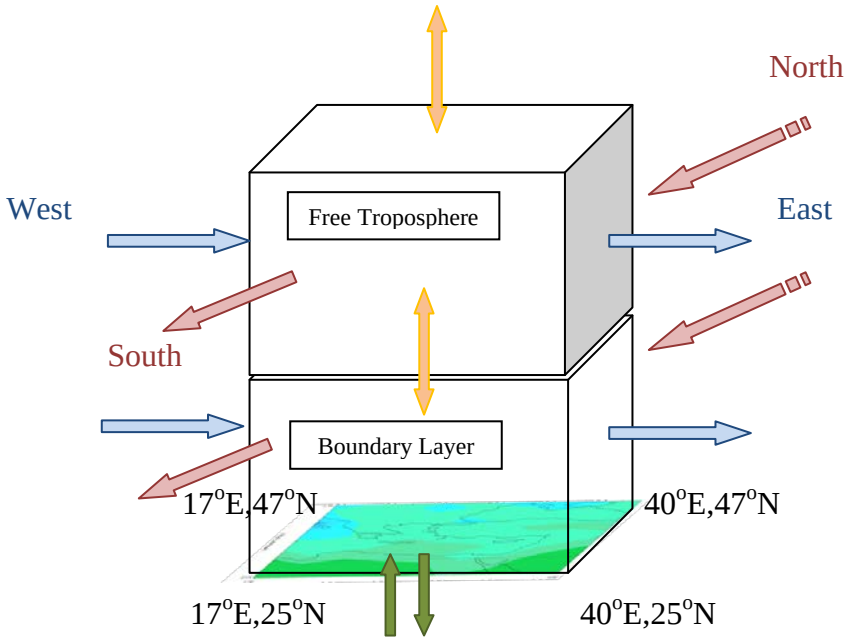


Figure 1. Schematic representation of the fluxes calculated in TM4-ECPL model for each pollutant. Blue arrows indicate West /East fluxes; red (North/South fluxes) orange marks the vertical fluxes/exchanges between the two considered compartments. Green arrows indicate surface exchanges: emissions and deposition. The studied area extends from 17°E to 40°E in longitude and from 25°N to 47°N in latitude; from surface to about 100 hPa.

1.2. Results – ozone budget terms

TM4-ECPL is able to simulate air pollutant levels in the rural and remote atmosphere in the East Mediterranean as demonstrated in Deliverable 1.4.3 for O_3 and PM_{10} levels (Daskalakis et al., 2011). Figure 2 further compares annual mean tropospheric O_3 column derived from TES satellite (Fig. 2a) with those calculated by TM4-ECPL model for the year 2008 for the base case simulation S0 (Fig. 2b) and for simulation S1 (Fig. 2c). This comparison shows that the model is able to capture the observed pattern of tropospheric O_3 column with maximum over the East Mediterranean and high levels to the North East of the black sea and to the South East over the Persian Gulf.

When removing the regional /East Mediterranean anthropogenic emission (simulation S1), the tropospheric O_3 column is locally reduced more than 2% and over the whole region by about 1% on

annual mean basis. Fig. 2c clearly shows that the high tropospheric O_3 columns are not due to the anthropogenic emissions in the region. Several factors contribute to them (investigation of these factors is ongoing) and the regional anthropogenic emissions support the observed O_3 pattern by the modest amounts earlier mentioned.

Focusing on the annual mean surface O_3 mixing ratios (Fig. 3) (for evaluation see Deliverable 1-4-3) it can be seen that about 45-50 ppb O_3 in the East Mediterranean are maintaining by sources other than the anthropogenic regional emissions. Anthropogenic East Mediterranean emissions contribute by up to 15% to the high O_3 levels in the region.

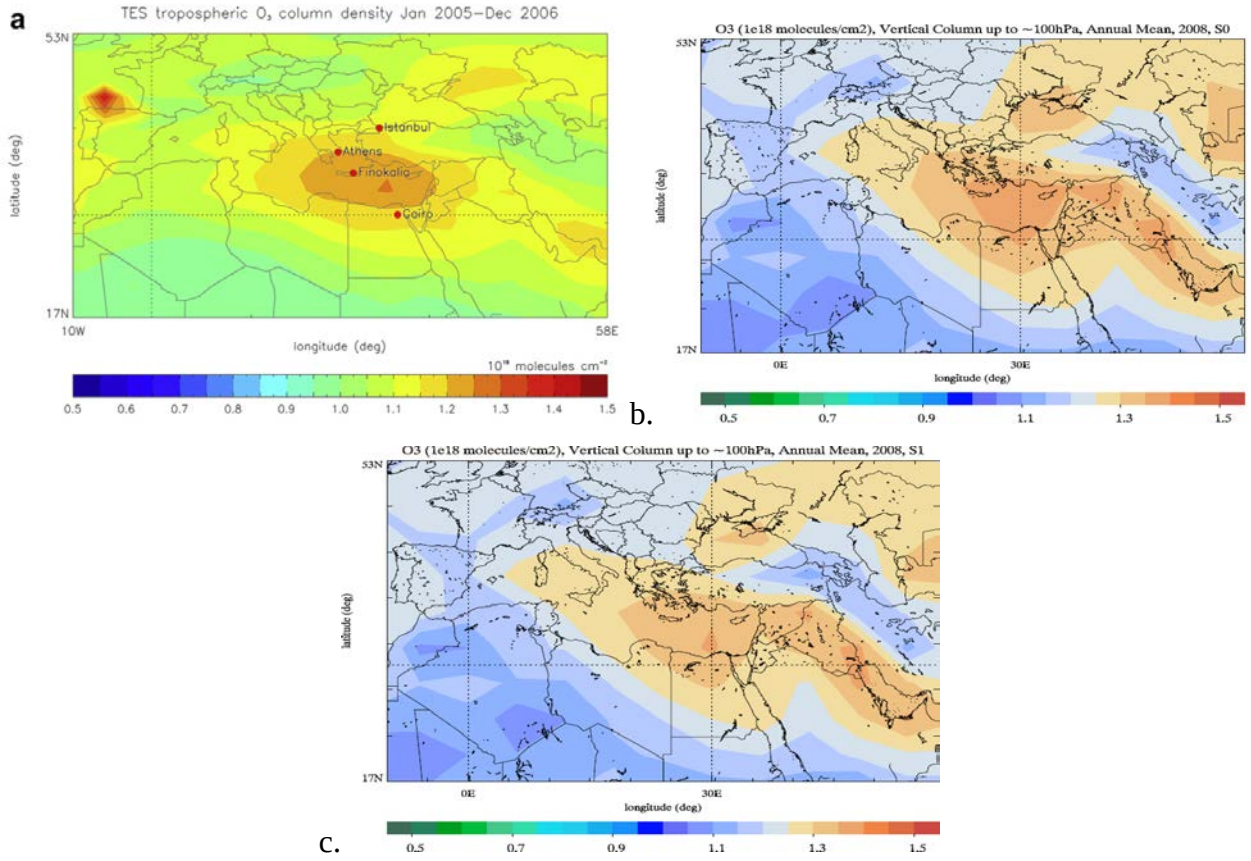


Figure 2. Annual mean tropospheric O_3 column over the Mediterranean showing the high columns over the East Mediterranean basin (a) TES- satellite derived observations (from Kanakidou et al., 2011), (b) calculated by the TM4-ECPL model, (c) calculated by the TM4-ECPL masking the anthropogenic emissions in the East Mediterranean (this study).

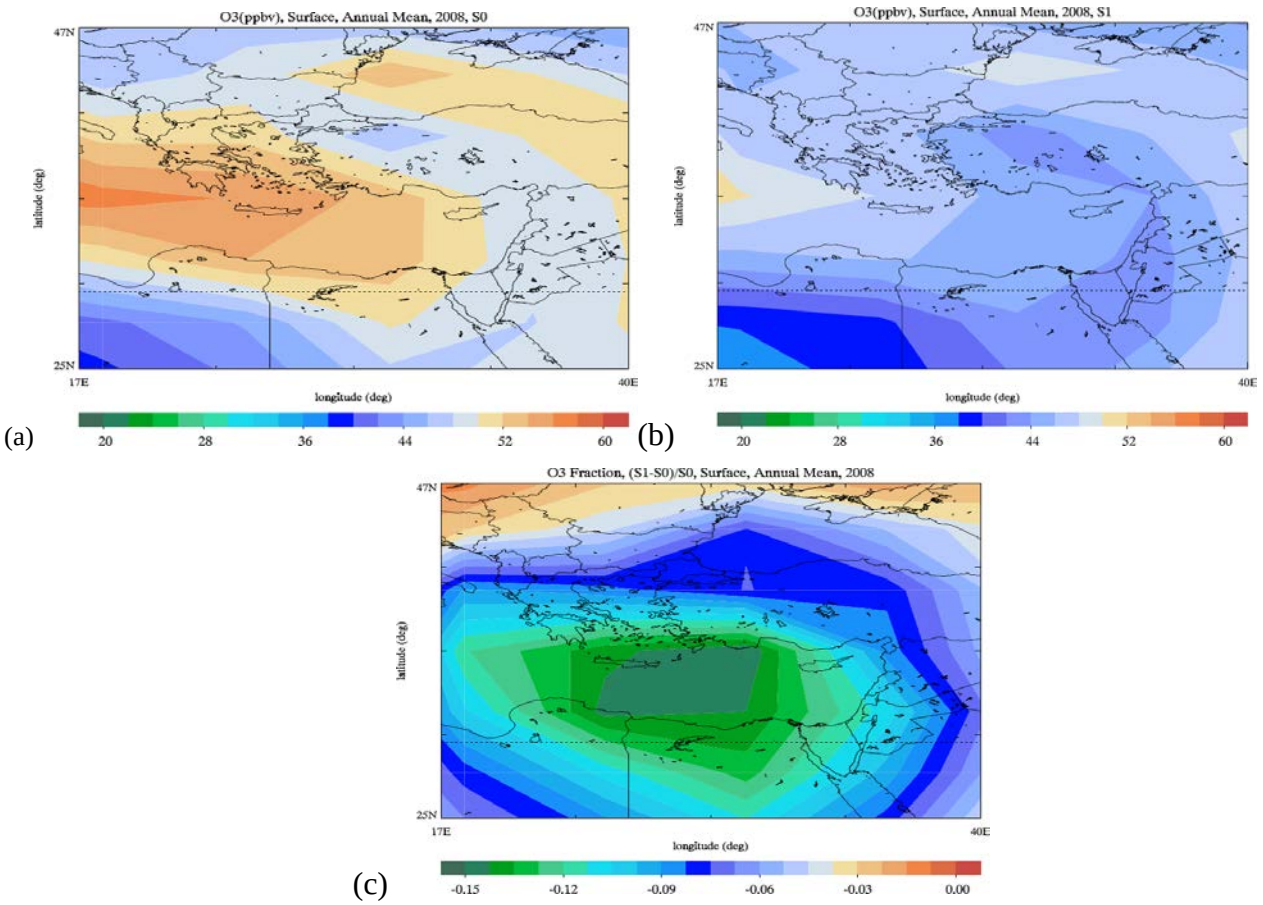
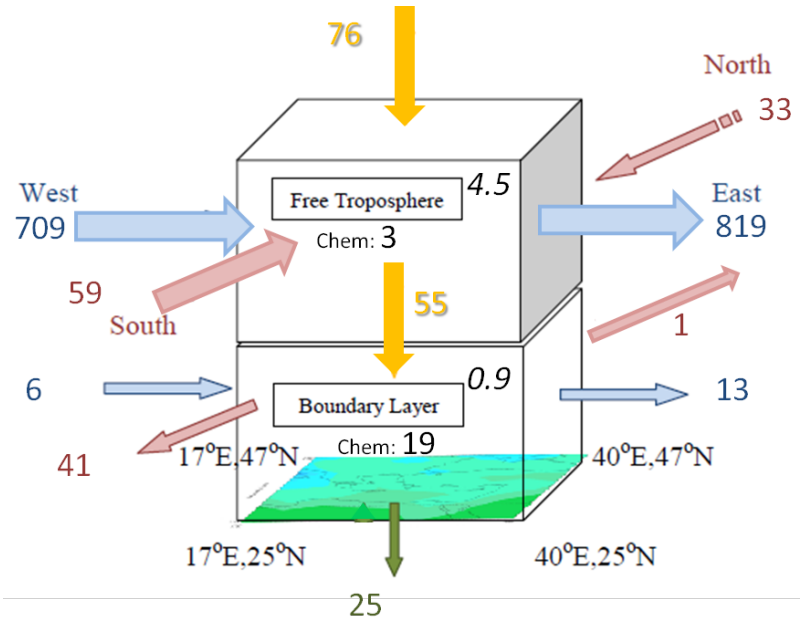


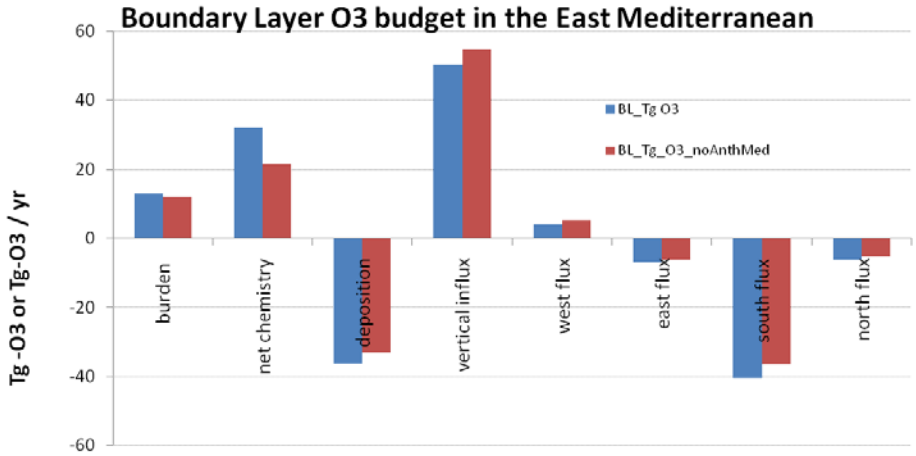
Figure 3. Annual mean surface O_3 mixing ratios in the East Mediterranean (a) for the base case simulation S0, (b) masking the East Mediterranean anthropogenic emissions (S1), (c) fractional decrease in surface O_3 mixing ratios due to Results from the global chemistry transport model TM4-ECPL (this work).

The O_3 budget has been investigated for the boundary layer (Fig 4b) and for the free troposphere (Fig. 4c) in the East Mediterranean for both simulations S0 and S1 as explained in section 1.1. The simulations show that in the boundary layer (BL), local pollution contributes by about 8% to the annual mean O_3 and by about 35% to its net chemical production; whereas it has smaller impact on the O_3 budget in the free troposphere (FT). As seen in Fig. 4a, 30-40 times higher amounts of O_3 are transported in and out the FT over the region than are chemically produced in the BL. On the other hand transport from the FT to the BL is a major source for the BL O_3 two to three times the net chemistry.

a.



b.



c.

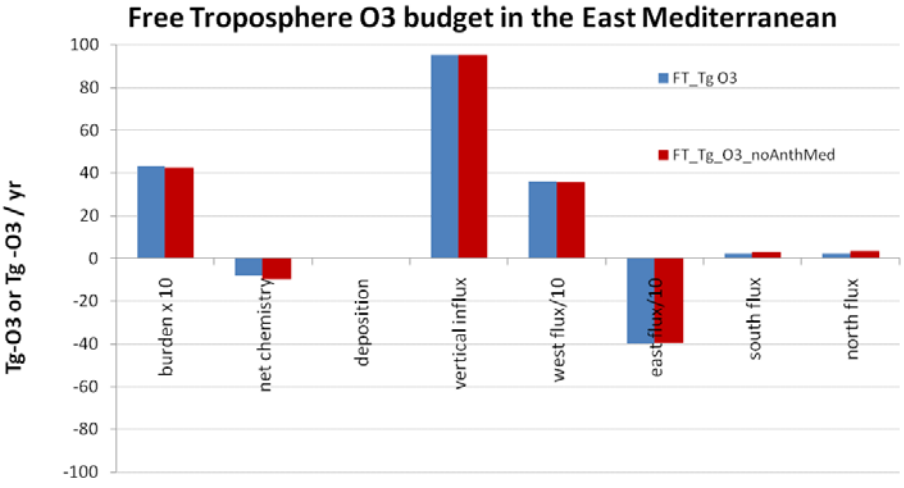


Figure 4. Annual O_3 budget terms in the East Mediterranean for the base case simulation (blue) and for simulation S1, masking the East Mediterranean anthropogenic emissions (red). Results from the global chemistry transport model TM4-ECPL (this work) (a) outline for both boundary layer and free troposphere budgets in Tg-O_3/yr as calculated by the high resolution simulations (2°lat x 3°lon x 25 layers) for 2008; (b) for the boundary layer (up to 850hPa) in Tg-O_3/yr; (c) for the free troposphere in Tg-O_3/yr. (b) and (c) as calculated by the low horizontal resolution simulations (4°lat x 6°lon x 34 layers) for 2008. Blue: for the base case scenario. Red: for scenario S1 without Anthropogenic Emissions from the East Mediterranean region.

1.3. Results- PM budget terms

PM_{10} surface levels in the region maximize over Africa due to significant contribution of dust (Fig. 5a) whereas PM levels, excluding sea-salt and dust (PM-SS-DU; Fig. 5b) maximize over continents showing a clear outflow from pollution centers to South-East. Masking anthropogenic emissions over the Mediterranean (simulation S1) the PM-SS-DU values are reduced to below the 2 $\mu g/m^3$ levels (Fig. 5c) that locally correspond to more than 80% reduction (Fig. 5d).

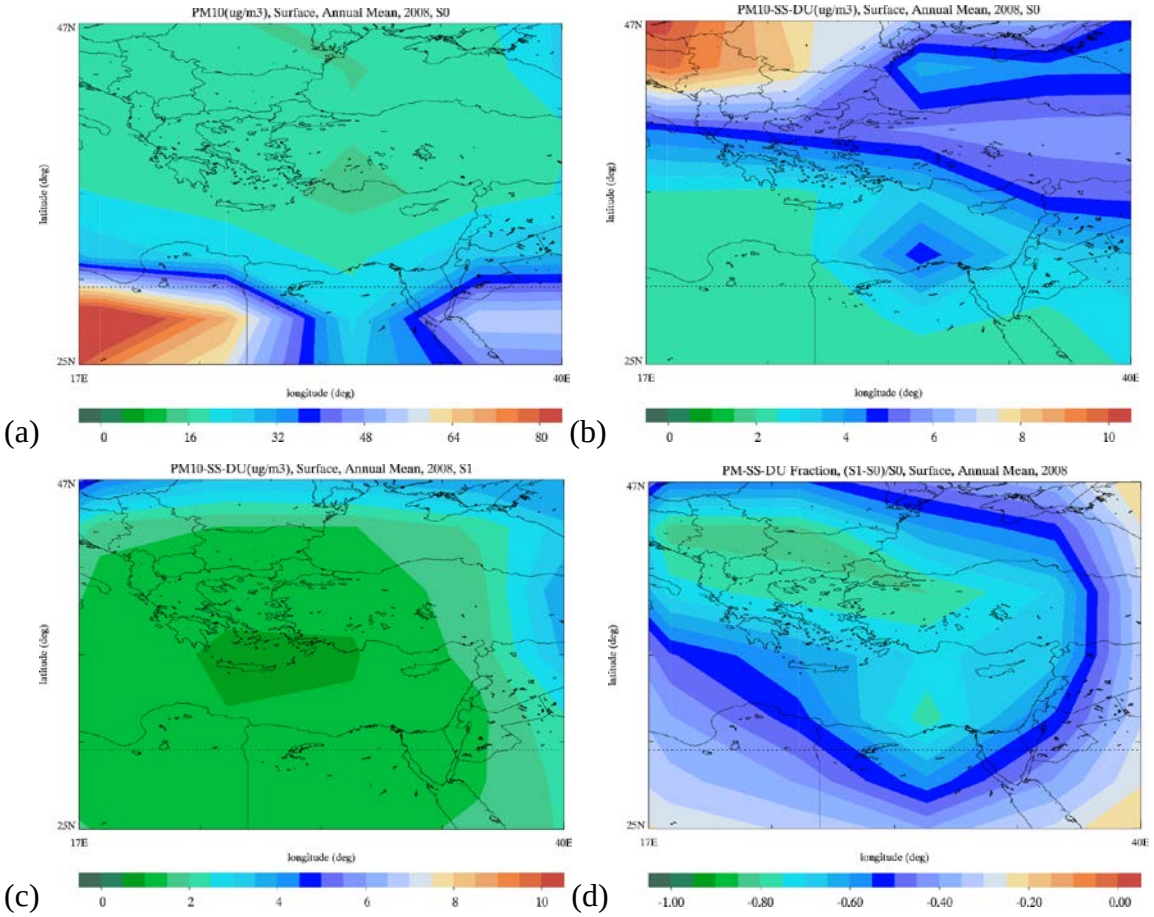


Figure 5. (a) Annual mean PM levels for the base simulations, (b, c) annual mean PM levels (sea-salt and dust aerosol components are not included in this figure for clarity – since on annual mean basis these natural aerosols dominate on the PM levels in the region due to the proximity of the African continent) (b) for the base simulation S0, (c) for S1 when masking the anthropogenic emissions in the East Mediterranean, (d) the fractional change between (c) and (b) relative to (b).

In Fig. 6, the annual budget terms of dust (Fig. 6a), of sea-salt as NaCl (Fig. 6b) and of PM-SS-DU (Fig. 6c) are shown for the East Mediterranean as calculated by the TM4-ECPL for the base case. Figure 6d compares the annual budget terms for PM-SS-DU for the base case (S0) with those computed after masking all anthropogenic emissions in the East Mediterranean (S1).

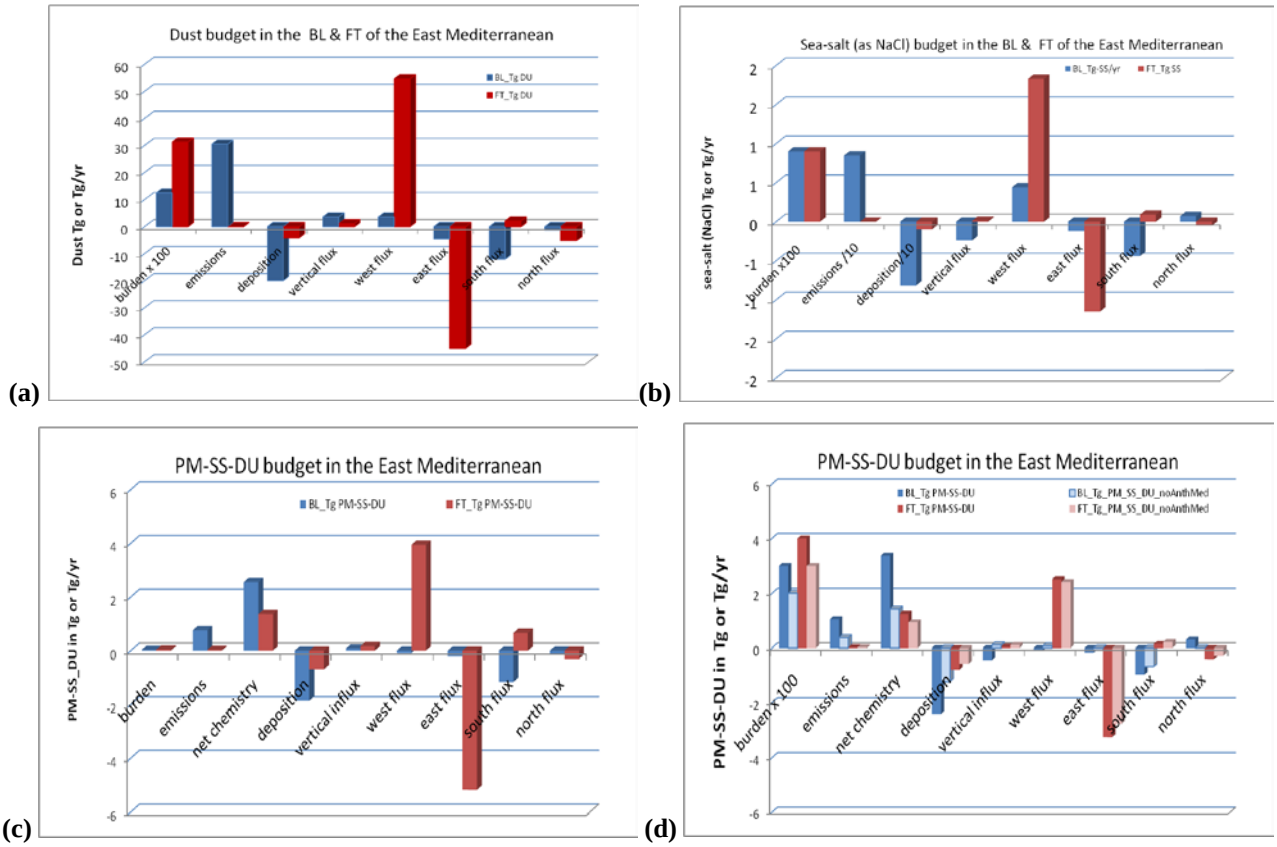
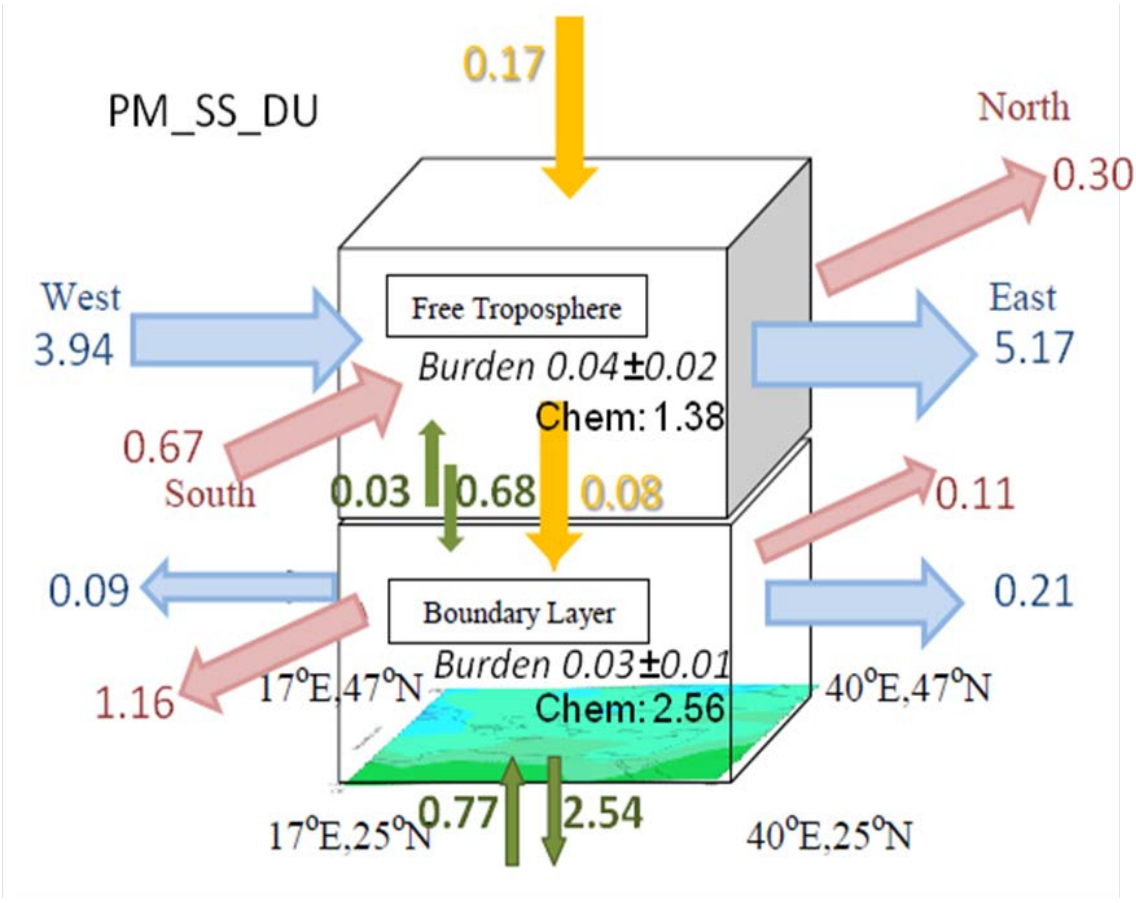


Figure 6. PM annual budget terms for the East Mediterranean for the year 2008: (a) for dust PM (b) for sea-salt (as NaCl), (c) for all PM except dust and sea-salt for the base case S0; (a),(b),(c) as calculated by the high horizontal resolution simulations (2^olat x 3^olon x 25 layers); (d) as (c) but in dark colors S0 and in light colors S1 and as calculated by the low horizontal resolution simulations (4^olat x 6^olon x 34 layers).

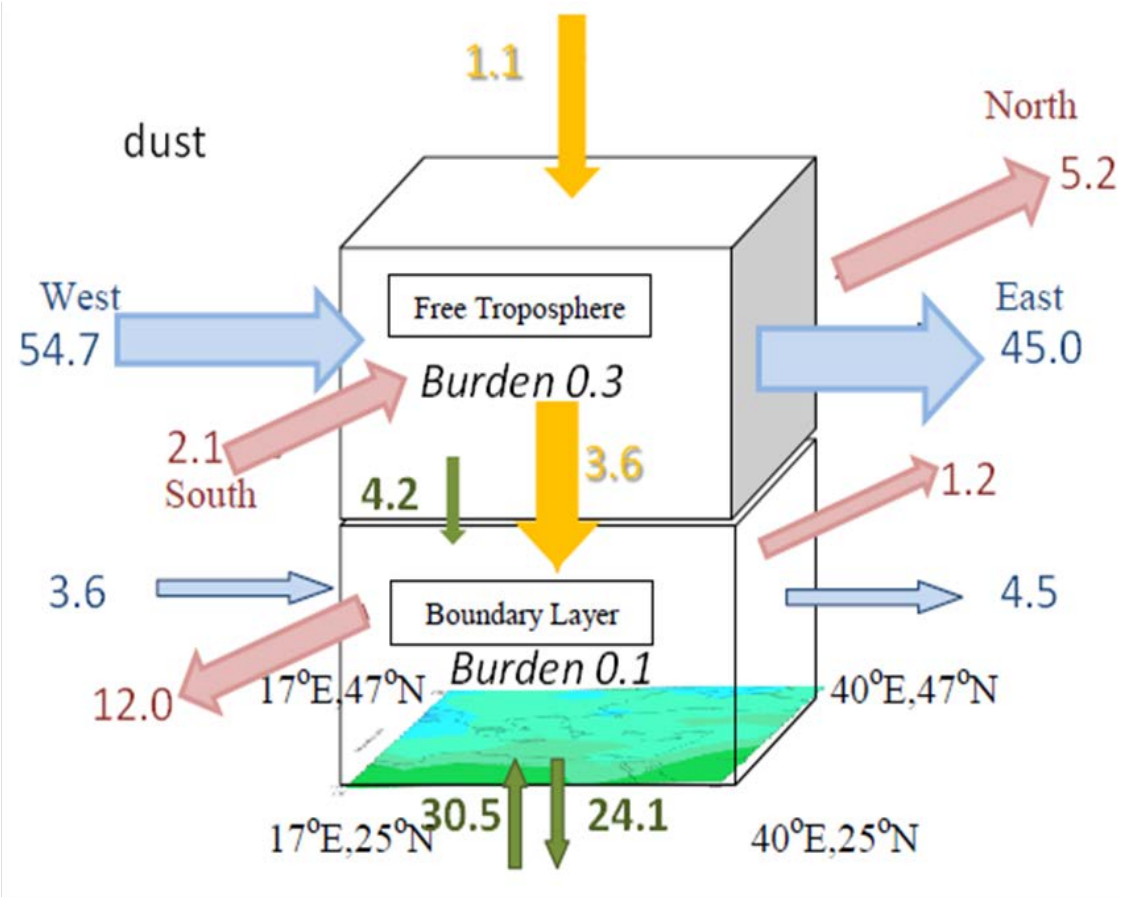
Figure 7 summarizes the aerosol budget in the area. In Figure 7a, dust and sea-salt aerosol components have been excluded from the aerosol budget (PM-SS-DU) for clarity. Dust and Sea-salt are the major natural aerosol components in the area and are marking (in particular dust due to the vicinity of the African continent) the budget of the total aerosol mass. Their budget terms are provided separately in Figure 7b for dust and 7c for sea-salt aerosols.

Figure 7a clearly shows the role of the boundary layer of the East cooker of pollution that arrives mainly via transport through the free troposphere and subsequent subsidence to the region as well as that is emission inside the region. Then, it exports the aged polluted air masses to all surrounding regions with dominant export path to the East and the South of the region. The free tropospheric column of the region is subject to strong west winds that carry significant amounts of pollutants, as clearly shown for O_3. These aerosols are partially scavenged and transported to the boundary layer in the region but also unreached by secondary aerosols formed or primary aerosol emissions in the region, before export following mainly to the east and to a lesser extent export through the north boundary of the region.

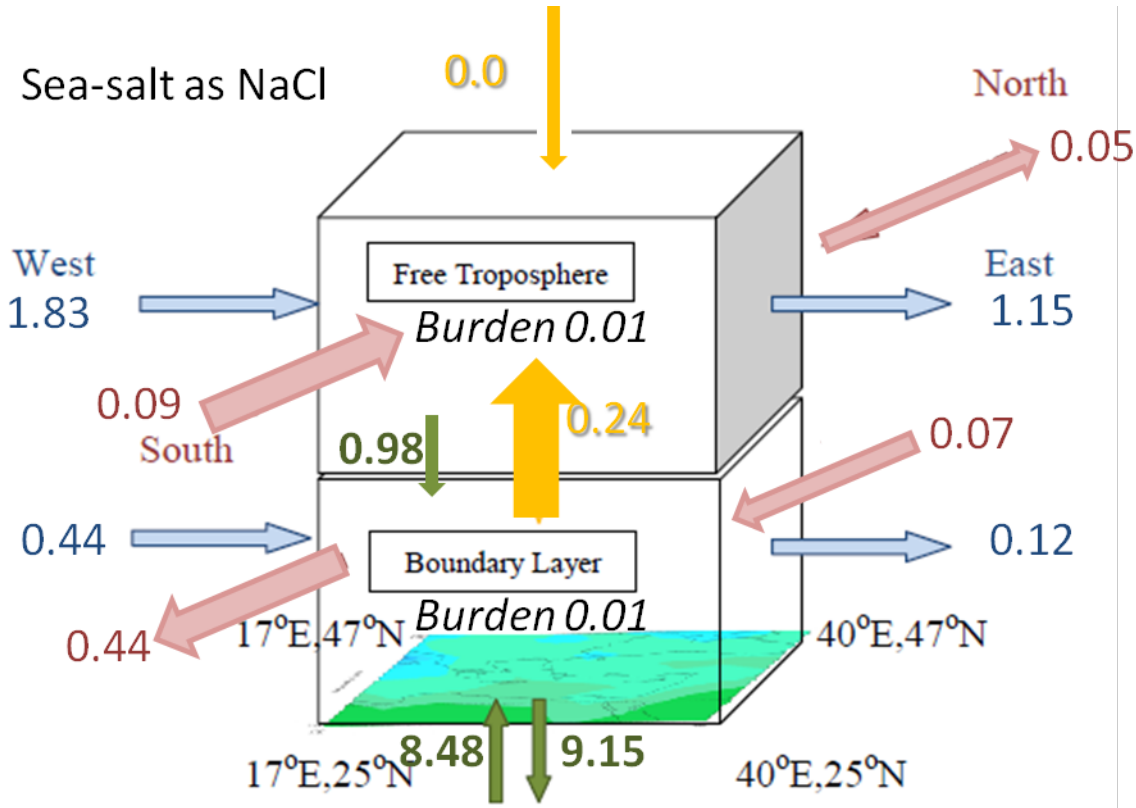
Figure 7b shows the regional budget terms for the dust aerosol component. Similarities but also significant differences exist with the earlier described PM-SS-DU aerosol component budget, since there is no chemical production of dust. In addition, DU has strong emissions in the boundary layer since the studied region covers part of the North East African Continent. However as calculated for O3 (Figure 4a) and for PM-SS-DU (Figure 7a) significant amounts are entering the region from the west and from the free troposphere. In addition, the free troposphere of the region is a receptor of additional dust entering from its south and west boundaries. The dust mass exported from the East and North is less than that entering the region since dust is partially subsiding over the region and removed by precipitation, sedimentation and deposition.


PM_SS_DU
0.17
North
0.30
West
3.94
Free Troposphere
Burden 0.04±0.02
East
5.17
Chem: 1.38
0.67
South
0.03
0.68
0.08
0.11
0.09
Boundary Layer
0.21
Burden 0.03±0.01
1.16
17°E,47°N
Chem: 2.56
40°E,47°N
17°E,25°N
0.77
2.54
40°E,25°N

(a)


dust
1.1
North
5.2
West
54.7
Free Troposphere
Burden 0.3
East
45.0
2.1
South
4.2
3.6
1.2
3.6
Boundary Layer
4.5
Burden 0.1
12.0
17°E,47°N
40°E,47°N
17°E,25°N
30.5
24.1
40°E,25°N

(b)



(c)

Figure 7. Aerosol budget for the East Mediterranean: (a) for PM excluding SS and DU; (b) for DU; (c) for SS (sea-salt as NaCl). Fluxes are in Tg/yr as calculated by the high resolution simulations (2°lat x 4°lon x 25 layers) for the base case scenario S0 for 2008.

Figure 7c depicting the sea-salt budget terms shows two major differences from that of DU (Figure 7b) above discussed: 1) Sea-salt is transferred to the free troposphere by advection and convection from the top of the boundary layer, although precipitation and sedimentation are bringing back to boundary layer 3-4 times more sea-salt that directly reaches the FT of the region via LRT from up-wind region to the west. 2) sea-salt is entering the BL over the region also from its north boundary, due to the vicinity with the Black Sea.

2. *Evaluation of import/export fluxes inside the East Mediterranean region – WRF/CHEM/CMAQ mesoscale modeling*

Im et al. (2011a,b) mesoscale modeling studies, using the WRF/MEGAN/CMAQ system, have investigated the response of gaseous and particulate air pollutants in the East Mediterranean, to increases in air temperature during summertime, susceptible to occur during climate warming. These air quality studies had both regional focus on the East Mediterranean and local focus on Istanbul and Athens extended areas as well as on the remote costal location of Finokalia on Crete Island in the East Mediterranean, acting as receptor site of air pollution.

Im and Kanakidou (2011) applied the same mesoscale modeling system to further investigate the summertime impacts of anthropogenic emissions from Istanbul and Athens on local and regional air quality in the Eastern Mediterranean. In order to accomplish this, mesoscale simulations have been performed accounting for and masking the anthropogenic emissions from Istanbul megacity and/or Athens urban agglomerations. The results (Fig. 8) show that Athens emissions have larger regional (0.8%) and downwind (2.7% at Finokalia) impacts on O_3 than Istanbul emissions that contribute to

surface O_3 by 0.6% to the domain-mean and 2.1% to the levels at Finokalia. On the opposite, regarding fine particles ($PM_{2.5}$) levels, Istanbul emissions have larger contribution both inside the megacity itself (75%) and regionally (2.4%) compared to Athens emissions, which have a local contribution of 65% and domain-wide contribution of 0.4%. Biogenic emissions are found to limit the production of secondary inorganic aerosol species due to their impact on oxidant levels.

Table 1 summarizes the budget terms for the gaseous pollutants (O_3, SO_2, NO_x, HNO_3 and PAN) and the particulate pollutants (nss-SO_4^{2-}, NO_3^- (fine and coarse), NH_4^+ (fine and coarse), OC, EC and PM_{10}) for Istanbul (IST), Athens (ATH) and Finokalia (FKL) in the entire boundary layer and for the period July, 1-15th, 2004. The results show that horizontal advection (HADV) is mostly a sink term for most pollutants in Istanbul and Athens whereas at Finokalia, it is a source term. This suggests that pollutants from the surrounding areas are transported to Finokalia. In Istanbul and Finokalia, O_3 is advected to the city whereas in Athens, it is horizontally transported from the city. A remarkable distinction between the two large urban areas is that Istanbul megacity atmosphere acts as chemical sink for O_3, suggesting significant titration by fresh NO_x emissions. On the other hand, in Athens, chemistry is a source term for O_3. This difference is particularly due to the different NMVOC/NO_x emission ratios in these cities (Im and Kanakidou, 2011). In Athens, O_3 formation is favored by relatively high NMVOC emissions compared to NO_x emissions. At Finokalia, chemistry (CHEM) is a source term for all ported gaseous species (Table 1). Vertical advection (ZADV) contributes relatively less than HADV at all sites and is generally a source term, except over Athens where O_3 and other secondary gaseous pollutants are exported to the free troposphere. Emission contribution to aerosol species is larger in IST than ATH due to higher particulate emissions (Im and Kanakidou, 2011). Negative CHEM budgets show destruction of NO_x and SO_2 to produce HNO_3 and H_2SO_4. Indeed, the CHEM term for HNO_3 is positive showing chemical production. Moreover, the results also suggest fine to coarse interaction of particles in the region. For instance, as seen in the Table 1, the aerosol processes (AERO) budget, which includes physical processes like condensation, coagulation etc…) are calculated negative for the fine nitrate particles whereas they are positive for the coarse mode nitrate, suggesting condensation of fine particles over the large particles like sea-salt and dust. An important result is that cloud production (CLDS) of nss-SO_4^{2-} is as important as the primary emissions and is even a larger source, particularly at FKL.

Our calculations suggest that the large urban areas are important sources of PM_{10} at their downwind locations. Unspecified anthropogenic particulate emissions and re-suspended dust emissions are important fraction of particulate emissions in big cities that are transported to downwind areas (Markakis et al., 2010a,b; Im and Kanakidou, 2011). Additionally, natural contributors, such as dust and sea-salt, account for a large portion of the aerosol levels in the Eastern Mediterranean. This also explains why although HADV is a sink term for most aerosol species presented in Table 1, it is a source term for the total PM_{10} mass at Finokalia that is representative for the remote East Mediterranean.

These global and mesoscale model results are under further interpretation and are the topic of a publication by Myriokefalitakis et al., 2011b in preparation.

Table 1. Budget terms for the gaseous pollutants (O_3, SO_2, NO_x, HNO_3 and PAN) and the particulate pollutants (nss-SO_4^{2-}, NO_3^- (fine and coarse), NH_4^+ (fine and coarse), OC, EC and PM_{10}) in Istanbul (IST), Athens (ATH) and Finokalia (FKL) integrated in the entire boundary layer for the 15-day period (July,1-15th, 2004): Horizontal advection (HADV), vertical advection (ZADV), emissions (EMIS), dry deposition (DDEP) chemistry (CHEM), aerosol processes (AERO) and cloud processes (CLDS). All terms reported in tons per 15 days. For region definition see in Im et al. (2011a).*

Species	IST							ATH							FKL						
	HADV	ZADV	EMIS	DDEP	CHEM	AERO	CLDS	HADV	ZADV	EMIS	DDEP	CHEM	AERO	CLDS	HADV	ZADV	EMIS	DDEP	CHEM	AERO	CLDS
O_3	4372	14	*N/A*	-297195	-4884	*N/A*	1	-4124	-40	*N/A*	-330997	3451	*N/A*	0	411	106	*N/A*	-409447	433	*N/A*	-1
SO_2	-853	0	1045	-82971	-33	*N/A*	-87	-224	-1	404	-43109	-18	*N/A*	-116	-239	2	341	-12855	-29	*N/A*	-73
NO_x	-12645	-2	14260	-261252	-1209	*N/A*	-2	-1895	-1	4730	-57961	-2616	*N/A*	0	-240	1	318	-2528	-107	*N/A*	0
HNO_3	-856	0	*N/A*	-192279	917	*N/A*	-55	-30	-1	*N/A*	-112441	140	*N/A*	-15	52	3	*N/A*	-74101	57	*N/A*	5
PAN	14	0	*N/A*	-9622	-19	*N/A*	0	42	-1	*N/A*	-6610	-40	*N/A*	0	-2	2	*N/A*	-9477	19	*N/A*	0
nss_SO_4^{2-}	-131	1	34	-11037	*N/A*	49	47	-162	2	24	-27085	*N/A*	27	122	-137	2	1	-36903	*N/A*	43	111
NO_3^- (2.5)	-18	0	165	-621	*N/A*	-146	-1	-6	0	28	-423	*N/A*	-22	0	-1	0	9	-27	*N/A*	-8	0
NO_3^- (2.5-10)	-15	0	*N/A*	-30411	*N/A*	49	-2	10	0	*N/A*	-35462	*N/A*	27	1	9	0	*N/A*	-74679	*N/A*	41	0
NH_4^+ (2.5)	-43	0	*N/A*	-2549	*N/A*	50	-9	-12	0	*N/A*	-5210	*N/A*	19	-5	1	0	*N/A*	-6055	*N/A*	2	0
NH_4^+ (2.5-10)	0	0	*N/A*	-228	*N/A*	0	0	0	0	*N/A*	-156	*N/A*	0	0	0	0	*N/A*	-309	*N/A*	0	0
OC	-269	0	274	-5890	*N/A*	10	-14	-33	0	30	-4591	*N/A*	7	-5	-3	0	1	-5481	*N/A*	3	0
EC	-114	0	120	-2083	*N/A*	0	-5	-49	0	52	-2714	*N/A*	0	-1	-5	0	4	-1438	*N/A*	0	0
PM_{10}	-793	2	1185	-374730	*N/A*	-58	-18	-410	5	802	-571720	*N/A*	21	45	266	6	34	-564663	*N/A*	-102	269

* Zero (0 values point to very small budget terms whereas N/A denotes the budget terms that are not calculated by CMAQ for the given gaseous and particulate species.

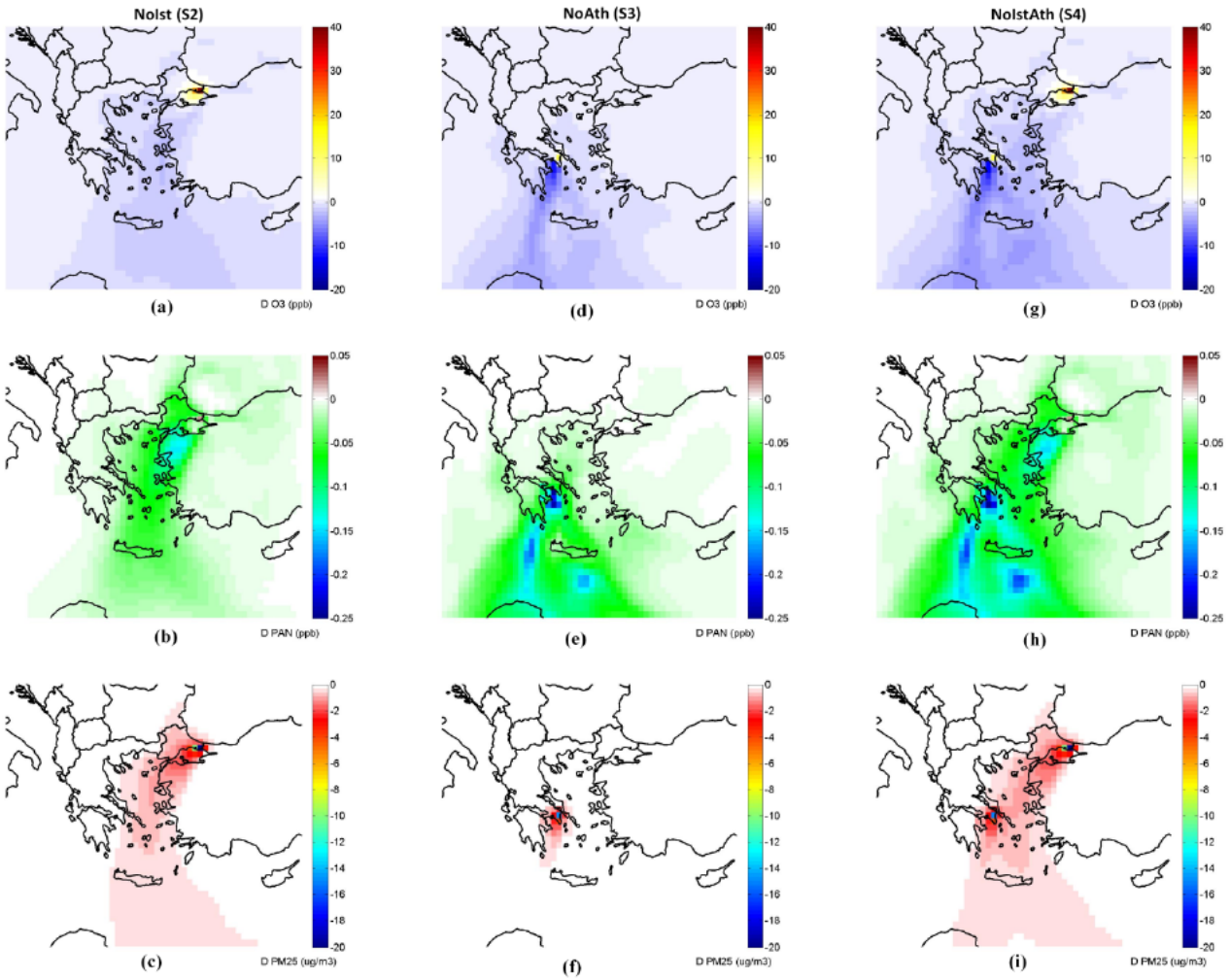


Figure 8. Surface O_3 (top panels), PAN (mid- panels) and $PM_{2.5}$ (bottom panels) concentrations calculated from the base case scenario and the differences (%) of scenarios neglecting emissions from Istanbul (NoIst), from Athens (NoAth) and from both Istanbul and Athens extended areas (NoIstAth) relative to the base case scenario, averaged over the 15-day simulation period (see details in Im and Kanakidou, 2011).

References issued from the CityZen project

Daskalakis N., Tsigaridis K., Myriokefalitakis S., Kanakidou M. et al., Observed and simulated ozone and organic aerosol changes in the global troposphere during the last decade, to be presented at the ACCENT plus Conference, Urbino, 13-16 Sept 2011; *and paper in preparation.*

Im, U., Markakis, K., Poupkou, A., Melas, D., Unal, A., Gerasopoulos, E., Daskalakis, N., Kindap, T., and Kanakidou, M.: The impact of temperature changes on summer time ozone and its precursors in the Eastern Mediterranean, Atmos. Chem. Phys., 11, 3847-3864, doi:10.5194/acp-11-3847-2011, 2011a.

Im, U., K. Markakis, M. Koçak, E. Gerasopoulos, N. Daskalakis, N. Mihalopoulos, A. Poupkou, T. Kindap, A. Unal, M. Kanakidou. Summertime aerosol chemical composition in the Eastern Mediterranean and its sensitivity to temperature: A modeling case study, *under review* Atmospheric Environment, 2011b.

Im, U., and Kanakidou, M: Summertime impacts of Eastern Mediterranean Megacity Emissions on air quality, *submitted to* Atmos. Chem. Physics Discuss., acp-2011-639, 2011.

Kanakidou M., Mihalopoulos N., Kindap T., Im U., Vrekoussis M., Gerasopoulos E., Dermitzaki E., Unal A., Kocak M., Markakis K., Melas D., Kouvarakis G., Youssef A.F., Richter A., Hatzianastassiou N., Hilboll A., Ebojie F., Wittrock F, von Savigny C., Burrows J.P., Megacities as hot spots of air pollution in the East Mediterranean, Atmospheric Environment, 45, 1223-1235, http://dx.doi.org/10.1016/j.atmosenv.2010.11.048, 2011.

Myriokefalitakis, S., Daskalakis, N., Im, U., Kanakidou M. et al: Air pollutant budgets in the East Mediterranean – the importance of long-range transport, *in preparation,* 2011b.

Other references

Markakis, K., Poupkou, A., and Melas, D., Tzoumaka., P and Petrakakis, M.: A computational approach based on GIS technology for the development of an anthropogenic emission inventory of gaseous pollutants in Greece, Water Air Soil Pollut., 207, 157-180, 2010a.

Markakis, K., Poupkou, A., and Melas, D. and Zerefos, C.: A GIS based anthropogenic PM10 emission inventory for Greece, Atmos. Poll. Res., 1, 2, 71-81, 2010b.