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Supplement of

Aerosol chemistry above an extended Archipelago of the Eastern Mediterranean basin during strong northern winds

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S1 Supplementary model details

The meteorological inputs for the PMCAMx applications are provided by the WRF/ARW model (Skamarock et al. 2008) (http://www.wrf-model.org/index.php). The model is driven by the National Centers for Environmental Prediction (NCEP) operational Global Final (FNL) Analyses (1.0º×1.0º spatial resolution), in combination with Sea Surface Temperature (SST), from the Real-Time Global SST (0.5º×0.5º spatial resolution). The 25-category USGS land-use classification scheme was adopted in order to provide land-cover data. The planetary boundary layer (PBL) parameterization used is the 'first-order' closure scheme developed by the Yonsei University (YSU) (Hong et al., 2006).

The WRF numerical simulations were performed using triple two-way nesting, with the first domain covering the extended area of Europe (23.0 to 77.0ºN, 14.5 to 44.5ºE) with 0.5º horizontal resolution, the second covering the extended area of Greece and Italy (29.3 to 50.2ºN, 4.8 to 32.2ºE) with 0.167º horizontal resolution and the third following the horizontal grid system as PMCAMx. In the vertical axis, 35 full sigma levels resolve the atmosphere (model top at 50hPa or 20 km), with a finer resolution near the surface.

The global model GEOS-CHEM (Bey et al., 2001; http://acmg.seas.harvard.edu/) is applied in order to cope with the transported air masses from outside the PMCAMx simulation domain. The model is driven by assimilated meteorological data from the Goddard Earth Observing System, Version 5 (GEOS-5) of the NASA Global Modeling and Assimilation Office (http://gmao.gsfc.nasa.gov). The version applied for the current study (v8-03-01) uses the SOA chemical mechanism, which includes the NOx-Ox-hydrocarbon-aerosol species module and a SOA module based on the framework proposed by Chung and Seinfeld (2002) and Henze et al. (2008). Moreover, the ISORROPIA II package for aerosol thermodynamical equilibrium is used (Fountoukis and Nenes, 2007).

A 9-month (January to September 2011) simulation of the global model is initially performed (4º latitude × 5º longitude). Three-hour boundary conditions (BCs) are calculated for May-September 2011 (allowing 5-month model spin up) around a domain centered in Europe (22 to 74º N and -20 to 45º E). Then, a nested run over this window is applied, with 0.5º (latitude) × 0.67º (longitude) horizontal resolution (Protonotariou et al., 2013). The vertical grid is composed by 47 levels up to 0.01 hPa.

S2 Emission model inputs

The PMCAMx applications are provided with hourly emissions for a series of pollutants from different source categories. Data for the area of Greece include NO_x, SO_2, NMVOC, CO, NH_3 and bulk PM_{10} emissions from several types of industries, road transport, central heating, maritime activities, railways, air traffic, agricultural activities and isoprene, terpenes, NO from forests (provided by the Ministry of Environment for the year 2002). This emission database has been refined to account for the changes related to the newer motor highway inside the Athens basin. These emissions vary against season and weekday/weekend of the week.

In the frame of this study, emission rates for the area of Turkey are retrieved from the EMEP emission dataset (http://www.ceip.at/webdab-emission-database/officially-reported-emission-data/). NO_x, SO_x, NMVOC, CO, NH_3 $PM_{2.5}$ and PM_{coarse} emissions from the 10 recommended SNAP sectors at 0.5º x 0.5º are horizontally downscaled to the model grid following the land use (eg. coarse ship emissions are equally split only to the fine cells covered by the sea). The vertical downscaling of the EMEP emissions for PMCAMx is done using the disaggregation factors proposed by Bieser et al. (2011), as described in Mailler et al. (2013). The temporal disaggregation of the EMEP emissions from the yearly totals to hourly values for a winter and a summer weekday and weekend is performed using the temporal factors extracted from Greek National emissions.

The daily emission rates from anthropogenic and natural sources of gaseous and particulate atmospheric constituents used in the PMCAMx model are given in Table S1. The emissions of most species in the two countries are in similar levels (e.g., nitrogen species are ca. 50 megaton/day). Differences (e.g. in particulate sulfate) are attributed to difference in the surface coverage between the two countries, and the fact that a significant part of Istanbul (and emission sources therein) is outside the PMCAMx simulation domain (treated as BCs by GEOS-CHEM). At this point, it should be noted that particulate sulfate is the most abundant anthropogenic emitted specie. This is in line with the significance (above 60%) of the industrial contribution in PM_{10} emissions in Athens and Istanbul reported in Kanakidou et al. (2011) and explains the predominance of sulfate in the total aerosol content in the EM (Kopanakis et al., 2012; Im et al., 2012; Tagaris et al., 2013).

Sea-salt particles, road (tire, brake, road wear and re-suspension) and soil dust emission fluxes are calculated online with meteorology, following the methodology developed and applied by Athanasopoulou et al. (2008, 2010). The size and chemical distribution of total aerosol emissions to the species and size bins used by the current PMCAMx model applications are also described therein. An update is related with the conversion of organic carbon (OC) to total organic aerosol (OA). In particular, industrial and motorway OC emissions are multiplied by a factor of 1.25 and by 1.54, respectively (Bergstrom et al., 2012; Brown et al., 2013), so that the modeled organic concentrations are directly comparable to the measured organic mass. The chemical speciation of the non-methane hydrocarbons from transportation and industry for SAPRC99 is described in Bossioli et al. (2002).

The emission databases used for the GEOS-CHEM applications are from fossil fuel burning (including ships) from the EMEP inventory for the European domain (Vestreng and Klein, 2002), biofuel emissions (Yevich and Logan, 2003), biogenic VOC emissions from vegetation based on the MEGAN model (Guenther et al., 2006) and natural sources from the oceans (Spracklen et al., 2008), volcanic activity (Chin et al., 2000) and lightning (Price and Rind, 1992). Global biomass burning emissions are not included, because the updated emissions were not available for the year 2011 in the currently applied GEOS-CHEM version.

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Table S1. Emission rates for the gaseous and aerosol species in PM_{40}. Numbers correspond to daily sums (summer weekday) for each of the two countries within the PMCAMx simulation domain.

	Gas and aerosol (PM_{40}) species	Total emission rates			
		Greece		Turkey	
		Area sources	Point sources	Area sources	Point sources
Gases (megamols day^{-1})	Carbon monoxide	140	9.2	51	54
	Sulfur dioxide	3.4	27	0.2	45
	Nitric oxide	44	5.7	12	35
	Nitrogen dioxide	3.2	0.8	0.8	2.5
	Ammonia	35	-	34	0.6
	Non-methane hydrocarbons	52	0.9	37	0.6
	Large chain hydrocarbons	0.5	-	0.2	-
Aerosol components (tons day^{-1})	Organics	31	9	16	17
	Sulfate	362	522	14	38
	Elemental carbon	61	47	28	13
	Sodium chloride[a]	23,867	-	23,867	-
	Crustal species[b]	750	-	750	-
	Rest aerosol components (species not treated by the PMCAMx)	703	210	2.6	219
	Nitrate[c]	0.029	-	0.011	-

[a] Sodium chloride corresponds mainly to sea-salt emissions of the whole simulation domain, developed as described in Athanasopoulou et al. (2008). It is noted that sea-salt emissions are calculated online with meteorology, thus they differ from day-to-day. This value corresponds to the date 29 August 2011.

[b] Aeolian soil dust emissions (mainly calcium, potassium, magnesium) from the whole simulation domain are calculated online with meteorology, thus they differ from day-to-day (Athanasopoulou et al., 2010). This value corresponds to the date 29 August 2011.

[c] Nitrate is only produced due to road dust re-suspension (Athanasopoulou et al., 2010).

Table S2. Mathematical formulation and statistical benchmarks, goals and criteria for evaluating the model system used in this study.

Metrics	Formulas [a]	Benchmarks, Goals, Criteria	Notes
Mean Bias	$MB = \frac{1}{N}\sum_{i=1}^{N}(E_i - O_i)$	≤ ±0.1g/kg, ≤ ±0.5K, ≤ ±10deg, ≤ ±0.5m/s	
Mean Absolute Gross Error	$MAGE = \frac{1}{N}\sum_{i=1}^{N}\lvert E_i - O_i\rvert$	< 2g/kg, ≤ 2K, ≤ 30deg, -	Statistical benchmarks for water vapor mixing ratio, air temperature, wind direction and wind speed, respectively (Tesche et al., 2001; Emery et al., 2001).
Root Mean Square Error	$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\lvert E_i - O_i\rvert^2}$	-, -, -, ≤ 2m/s	
Index of Agreement	$IA = 1 - \left[\frac{N \cdot RMSE^2}{\sum_{i=1}^{N}\left(\lvert E_i - \overline{O}\rvert + \lvert O_i - \overline{O}\rvert\right)^2}\right]$	≥0.6, ≥ 0.8, -, ≥ 0.6	
Mean Fractional Bias	$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{(E-O)}{\left(\frac{O+E}{2}\right)}100\%$	±30, ±64, ±47, ±187, ±32, ±31, ±122, ±30 (goals[b]) ±60, ±88, ±74, ±189, ±62, ±61, ±135, ±60 (criteria[b])	Aerosol performance metrics for sulfate, nitrate, ammonium, chloride, organics, OC, EC and PM_{10}, respectively. When goals (criteria) are met, performance is good (acceptable).
Mean Fractional Error	$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{\lvert E-O\rvert}{\left(\frac{O+E}{2}\right)}100\%$	50, 101, 83, 192, 58, 56, 149, 50 (goals[b]) 75, 118, 102, 193, 81, 80, 158, 75 (criteria[b])	
Normalized Mean Bias	$NMB = \frac{1}{N}\sum_{i=1}^{N}\frac{(E_i - O_i)}{O_i}100\%$		
Normalized Mean Error	$NME = \frac{1}{N}\sum_{i=1}^{N}\frac{\lvert E_i - O_i\rvert}{O_i}100\%$		
Correlation coefficient	$r = \left[\frac{\sum_{i=1}^{N}(E_i - \overline{E})(O_i - \overline{O})}{\sum_{i=1}^{N}(E_i - \overline{E})\sum_{i=1}^{N}(O_i - \overline{O})}\right]$		
Standard deviation	$Stdev = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\lvert x_i - \overline{x}\rvert^2}$		xi and $\overline{x}$ are the raw and mean estimation or observation values

[a] E is the estimated (modeled) and O is the observed value of each parameter, paired in space and time for each i of N data pairs. $\overline{E}$ and $\overline{O}$ are the mean values of estimations and observations, respectively.

[b] The goals and criteria for aerosol components are given by the equations below (Boylan and Russell, 2006):

$MFB = \pm 170e^{-0.5\left(\frac{\overline{O}+\overline{E}}{0.5}\right)+30}$ and $MFE = 150e^{-0.5\left(\frac{\overline{O}+\overline{E}}{0.75}\right)+50}$ (goals),

$MFB = \pm 140e^{-0.5\left(\frac{\overline{O}+\overline{E}}{0.5}\right)+60}$ and $MFE = 125e^{-0.5\left(\frac{\overline{O}+\overline{E}}{0.75}\right)+75}$ (criteria), where $\overline{E}$ and $\overline{O}$ are the mean values of estimations and observations, respectively.

Table S3. Prediction skill metrics of the basic meteorological parameters and submicron aerosol concentrations (**prd**, in **bold**) against airborne measurements (obs) from the nine flights during the period of interest (29 August – 09 September 2011). Performance metrics with green (red) fonts indicate good (poor) model performance, according to the selected criteria of evaluation, given in Table S2. The rest model outputs (performance metrics with black fonts) are acceptable (average model performance).

Metrics	Water vapor mixing ratio g/kg	Temperature K	Wind speed m/s	Wind direction deg	Sulfate	Nitrate	Ammonium	Chloride	Organics
					μg m^{-3} (below/above 2.2 km asl)				
Mean prd	**4.2**	**278.9**	**8.0**		**4.8 (5.5/3.8)**	**0.9 (1.9/0.4)**	**1.1 (1.7/0.5)**	**0.02 (0.05/0)**	**1.4 (2.3/0.9)**
Mean obs	4.2	279.2	8.4		5.0 (5.8/3.7)	0.1 (0.2/0.04)	1.0 (1.5/0.5)	0.01 (0.02/0)	2.4 (4.4/1.1)
Max prd	**14.2**	**300.3**	**19.5**		**12.1 (12.1/9.7)**	**7.9 (7.9/2.7)**	**4.2 (4.2/1.5)**	**0.4 (0.4/0.03)**	**6.8 (6.8/4.8)**
Max obs	14.4	304.2	24.1		23.4 (23.4/15.0)	1.9 (1.9/1.5)	5.2 (5.2/2.5)	0.8 (0.3/0.8)	10.7 (10.7/9.4)
Stdev prd	**3.7**	**11.7**	**2.9**		**2.4 (2.4/2.2)**	**1.5 (2.1/0.4)**	**0.8 (0.8/0.4)**	**0.07 (0.1/0)**	**1.3 (1.4/0.9)**
Stdev obs	3.9	11.1	4.1		3.8 (3.8/3.4)	0.1 (0.1/0.1)	0.9 (0.8/0.5)	0.03 (0.03/0.03)	2.5 (2.2/1.9)
IA	0.96	0.99	0.70		-	-	-	-	-
MB	0.05	-0.26	-0.40	8.07	-0.2 (-0.3/0.1)	0.8 (1.7/0.3)	0.1 (0.2/-0.02)	0.01 (0.03/0)	-0.9 (-2.1/-0.2)
MAGE	1.10	1.63		23.44	2.2 (2.4/1.8)	0.9 (1.9/0.4)	0.6 (0.8/0.3)	0.03 (0.1/0)	1.4 (2.2/0.8)
RMSE			3.64		3.1 (3.5/2.4)	1.7 (2.7/0.6)	0.8 (1.0/0.4)	0.07 (0.1/0.03)	2.0 (2.7/1.3)
MFB (%)					20.4 (6.0/41.2)	67.8 (17.1/93.8)	21.9 (17.0/27.1)	-150.1 (-74.7/-194.3)	2.0 (-64.2/42.9)
MFE (%)					55.2 (43.8/71.6)	184.0 (173.7/189.3)	63.4 (57.4/69.9)	18.3 (162.2/-65.7)	83.3 (74.5/88.7)
NMB (%)					-0.03 (-0.06/0.02)	9.3 (9.3/9.1)	0.1 (0.1/-0.03)	1.8 (1.3/-2.7)	-0.4 (-0.5/-0.2)
NME (%)					0.4 (0.4/0.5)	10.3 (10.2/10.5)	0.6 (0.6/0.6)	3.9 (2.7/-7.7)	0.6 (0.5/0.7)
r					0.6 (0.4/0.7)	0.3 (-0.1/-0.1)	0.6 (0.2/0.6)	0.2 (0.1/0.03)	0.8 (0.6/0.8)

r^2					0.3 (0.2/0.5)	0.1 (0.01/0.01)	0.3 (0.03/0.4)	0.03(0/0)	0.6 (0.4/0.6)
% within a factor of 2					70.2 (79.8/56.4)	4.5 (3.7/4.9)	59.2 (65.1/52.9)	6.6 (3.7/8.3)	41.9 (48.2/37.9)
No of samples	16052	16052	16052	15133	1335 (787/548)	2244 (765/1479)	1518 (788/730)	1624 (599/1025)	2066 (788/1278)

Table S4. Prediction skill metrics of the aerosol concentrations (**prd**, in **bold**) against ground measurements (obs) during the period of interest (29 August – 09 September 2011). MFB and MFE with green fonts indicate good model performance, according to the selected criteria of evaluation, given in Table S2.

Metrics	PM_1 Sulfate	PM_{10} OC[a]	PM_{10} EC	PM_{10}
	μgm^{-3} Vigla (N. Aegean) / Finokalia (S. Aegean)			
Mean prd	**-/ 6.4**	**2.0/2.1**	**0.32/0.26**	**-/29.1**
Mean obs	-/5.9	2.3/2.9	0.25/0.38	-/30.6
Max prd	**-/12.5**	**4.8/4.3**	**0.6/0.4**	**-/47.8**
Max obs	-/14.3	5.0/8.1	0.8/1.2	-/59.8
Stdev prd	**-/3.1**	**1.0/0.8**	**0.13/0.0.8**	**-/8.6**
Stdev obs	-/3.5	1.1/1.3	0.20/0.23	-/12.8
MB	-/0.5	-0.2/-0.8	0.07/-0.12	-/1.5
MAGE	-/2.4	0.7/1.2	0.16/0.19	-/13.9
RMSE	-/2.9	0.9/1.5	0.21/0.27	-/17.9
MFB (%)	-/12.9	-11.3/-28.2	40.8/-18.6	-/10.6
MFE (%)	-/45.1	36.6/46.1	63.1/55.6	-/47.4
NMB (%)	-/0.1	-0.1/-0.3	0.3/-0.3	-/0.0
NME (%)	-/0.4	0.3/0.4	0.7/0.5	-/0.5
r	-/0.6	0.6/0.2	0.3/0.1	-/-0.4
r^2	-/0.4	0.4/0.05	0.1/0.0	-/0.2
% within a factor of 2	-/78.8	83.9/80.9	54.8/71.4	-/76.9
No of samples	-/184	31/42	31/42	-/26

[a] Organic aerosol predictions are divided by 2.1 (Turpin and Lim, 2001), to extract the carbon mass (OC).

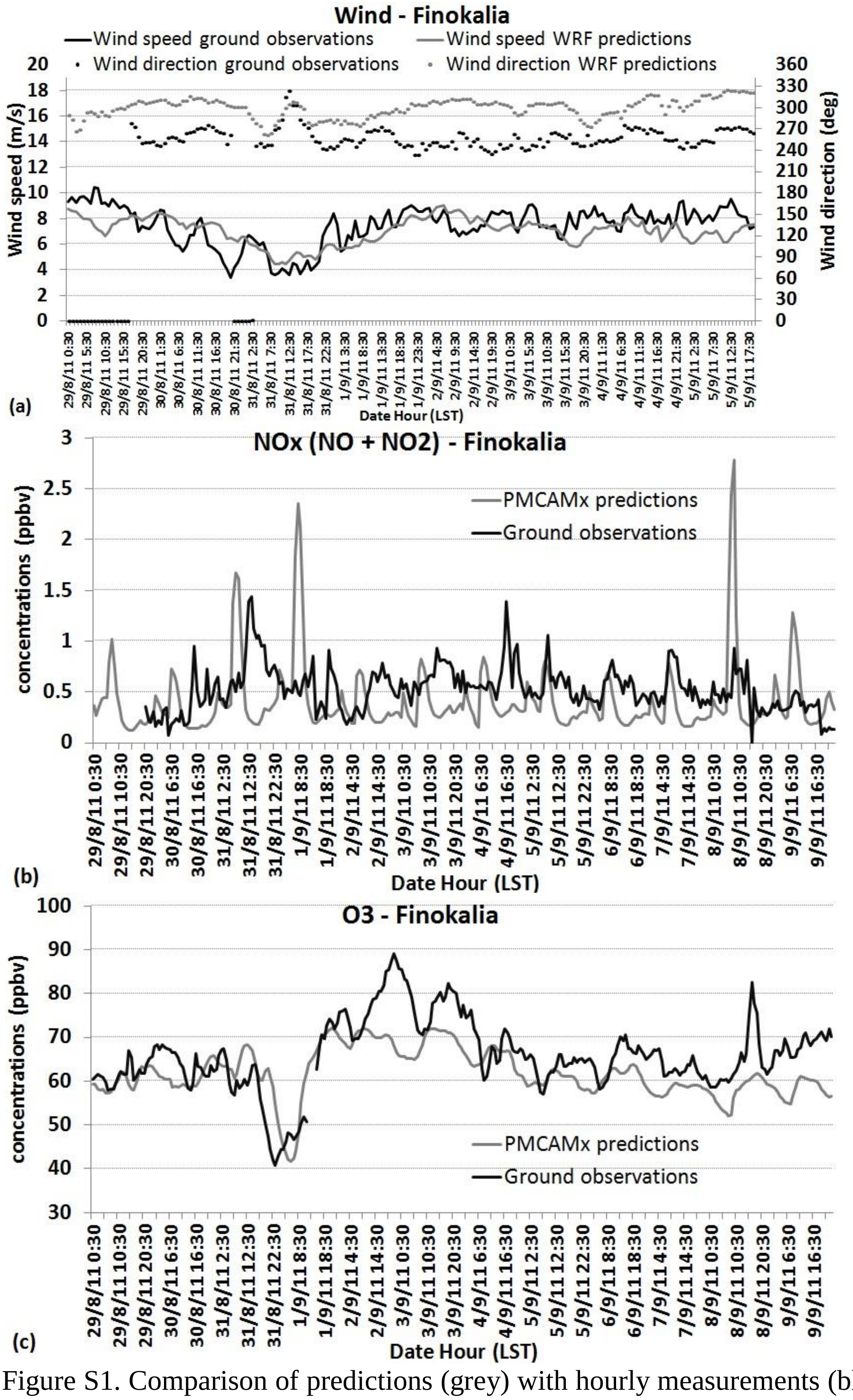

Figure S1. Comparison of predictions (grey) with hourly measurements (black) of: (a) wind speed (m s^{-1}) and wind direction (deg), (b) NO_x concentrations (ppbv) and (c) O_3 concentrations (ppbv) over Finokalia (Crete).

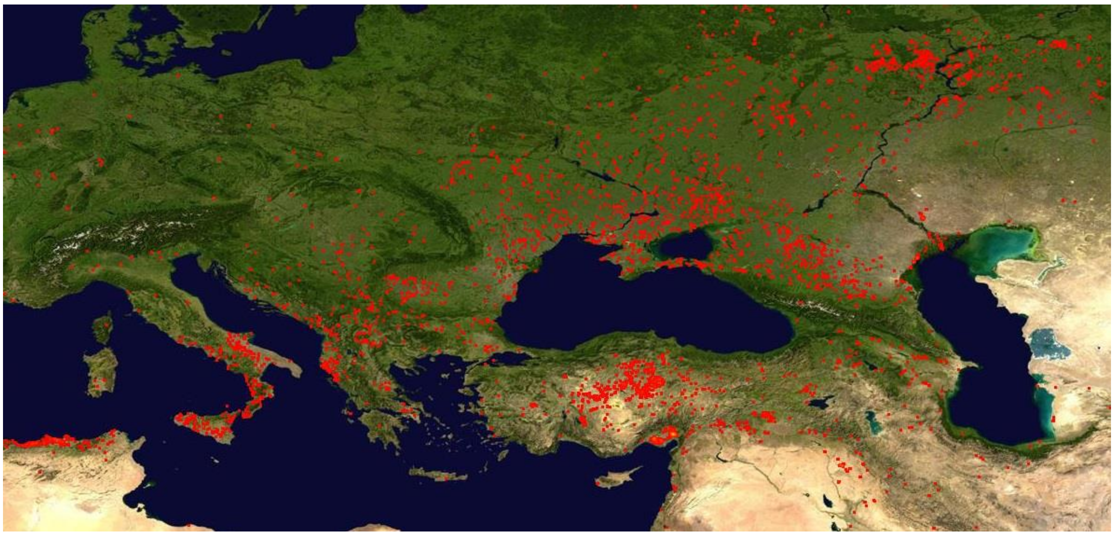

Figure S2. MODIS image acquisition, showing the fire events in Turkey, south and east Europe from 29 August until 07 September 2011.