1.27 VALIDATION AND INTER-COMPARISON OF CALPUFF REGULATORY MODEL TO EULERIAN MODELS AND MEASUREMENTS. AN APPLICATION OVER THE GREATER ATHENS AREA, GREECE.

Anna Protonotariou [1], Elisavet Bossioli [1], Eleni Athanasopoulou [1], Aggeliki Dandou [1], Maria Tombrou [1], Vassiliki D. Assimakopoulos [2], Helena A. Flocas [1] and, Costas G. Chelmis [1]
[1]Division of Applied Physics, Department of Applied Physics, University of Athens, Greece
[1]Institute of Environmental Research and Sustainable Development, National Observatory of Athens, Greece

INTRODUCTION

Models description

In this study, the validation of a regulatory dispersion model towards two Eulerian and experimental measurements is presented over the Greater Athens Area (GAA) in Greece. The models being inter-compared are: (1) CALPUFF (*Scire et al.*, 2000), an advanced 3D Lagrangian-Gaussian non-steady-state regulatory modeling system that consists of the diagnostic meteorological CALMET model and the air quality transport and dispersion CALPUFF model, (2) the Urban Airshed Model, UAM, (*SAI*, 1992), a 3D Eulerian photochemical model, designed to calculate the concentrations of both inert and chemically reactive pollutants by simulating the physical (advection, dispersion, deposition) and chemical processes (Carbon Bond chemical mechanism, *Gery et al.*, 1989) in the atmosphere, and (3) the Eulerian Regional Modeling System For Aerosol and Deposition, REMSAD, (*ICF Consulting*, 2002), incorporating the micro-CB-IV mechanism for the photochemistry (*Gery et al.*, 1989), the MARS-A algorithm for the formation of the secondary inorganic particulate matter (Binkowski F.S. and U. Shankar, 1995) and the Pankow approach for the secondary organic aerosol formation (*Odum et al.*, 1997). The meteorological fields required by CALPUFF were provided by CALMET and in the case of the Eulerians, by the Penn State/NCAR Mesoscale Model MM5 (*Anthes et al.*, 1978). The wind fields and mixing heights generated by CALMET are also compared towards those calculated by MM5.

EXPERIMENTAL SET UP

The measurement data were collected during the experimental campaign, carried out in the GAA within the framework of the ICAROS-NET European project covering the period from 13 September to 7 October 2002. Three experimental sites were employed (see Fig. 1): PEN (background suburban, being characterized by complex terrain), KAP (rural) and ZOG (background suburban area, near the city centre). Data of NO_2 were collected at all three stations, while data of PM_{10} were collected only at PEN. Moreover, mixing height estimations were available with the aid of a SODAR-RASS system at ZOG and of a SODAR system at PEN. The above mentioned experimental data were combined with NO_2 and PM_{10} data provided by existing air quality monitoring stations (18 stations of the official air pollution monitoring network of the Greek Ministry of Environment and 6 stations of Athens International Airport) being evenly distributed within the GAA (see Fig. 1). The radiosonde data, were obtained twice (0200 and 1400 LST) daily by the Greek Meteorological Service, at Hellinikon (HEL) station. These data are used supplementary to SODAR-RASS data to provide the wind and temperature profile above the atmospheric boundary layer. The meteorological surface data were provided by the National Observatory of Athens at two stations: a) Thission (NOA), close to the city centre and b) at Penteli Mountain (PEN). This study was performed for the 27th of September 2002.

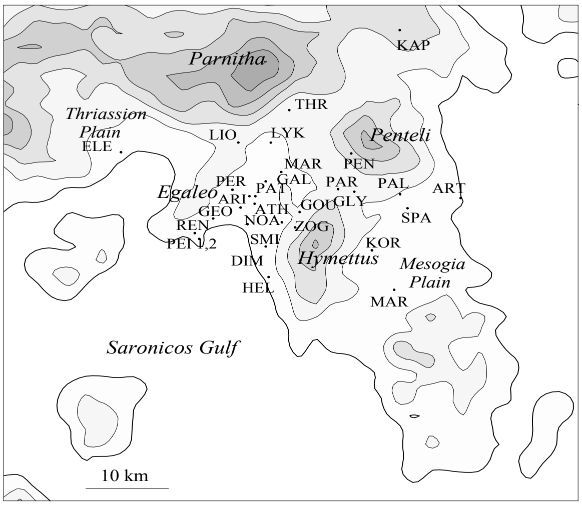


Figure 1. Map of the Greater Athens area

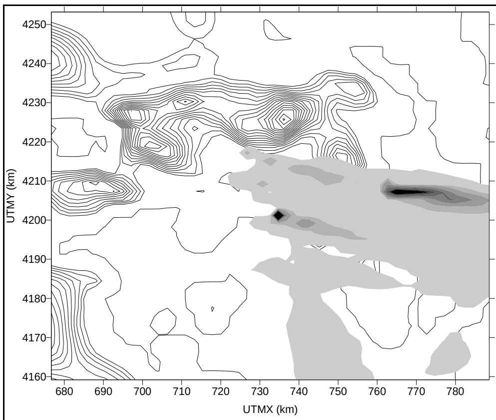
Emissions inventory
For the numerical simulations, the latest emission inventory of the GAA is adopted, having as reference year 1998, covering the transportation sector (on and off road activities, railway, airport, sea transport) (*Symeonidis et al*., 2003) and the industrial activities (Greek Ministry of the Environment). The spatial resolution of the area sources is 2kmx2km, while the temporal basis is seasonal and hourly. Point sources are also included. The input emission species in CALPUFF are NO_2, NO, SO_2 and PM_{10}, while hydrocarbons and CO were included only in UAM and REMSAD.

METEOROLOGICAL MODELS APPLICATION

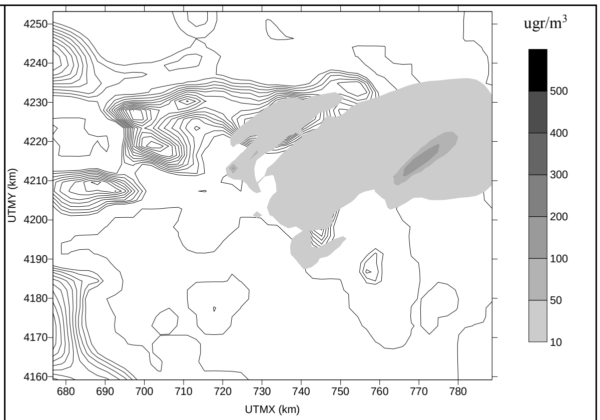
The MM5 model runs by using the ECMWF input data, (from the European Centre for Medium-range Weather Forecasts), while, in this study, CALMET simulated twice the meteorological fields by using two input data sets: (a) surface meteorological data measured at NOA and PEN stations and upper air data from the HEL station and the SODAR-RASS system, at ZOG station and (b) as in the previous case but with additional surface meteorological data measured at six more stations of the Greek Ministry of the Environment, in the eastern part of the GAA and close to the airport as well as by upper air data calculated by the MM5 model in ELE, LIO, PEN and SPA stations. It was found that, CALMET produces a rather homogenous wind field compared to the one calculated by the MM5, especially during stable conditions. This is mainly due to the interpolation procedure that CALMET applies to the measured meteorological data. Thus, it is not able to resolve the local circulation patterns generated by the topographical features, as in the case of MM5 model. This was the main reason to deliver another simulation by CALMET, supported by the second data set. The diurnal variation of Mixing Height (calculated and measured) was studied at PEN and ZOG stations. It was found that during the daily hours, the calculated values (from both models) were higher than the experimental ones. However, during the afternoon transition period the values from CALMET reached almost zero values, when the experimental ones varied around 300m for ZOG and 200m for PEN. This discrepancy could be explained by the fact that more than 5 hours passed from the previous sounding. MM5 results were much improved when the Mixing Height was allocated at the height of the inversion base, during the day.

DISPERSION MODELS APPLICATION

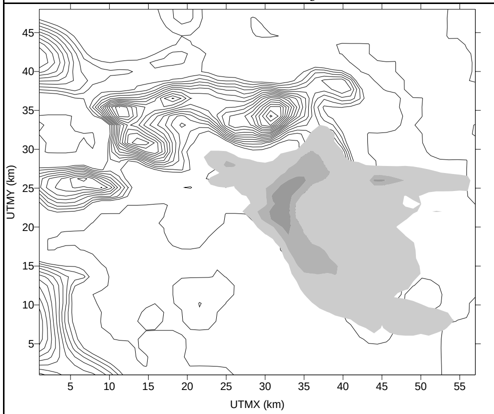
The output species common for CALPUFF, UAM and REMSAD are NO_2, NO and SO_2, while for CALPUFF and REMSAD are SO_4, NO_3, HNO_3 and PM_{10}. From these species, only the NO_2 and PM_{10}, produced by the three models, are presented in Table 1, for two hours (03:00 LST and 15:00 LST), at the sites where measurements were available. In particular, NO_2 concentration data were collected at 24 stations, while PM_{10} were measured only at seven stations (shown in Table 1). Also, their spatial distribution is shown in figures 2,3,4 and 5. The air quality stations are characterized as urban traffic (UT), urban background (UB), suburban industrial (SI), suburban background (SB) and suburban (S).



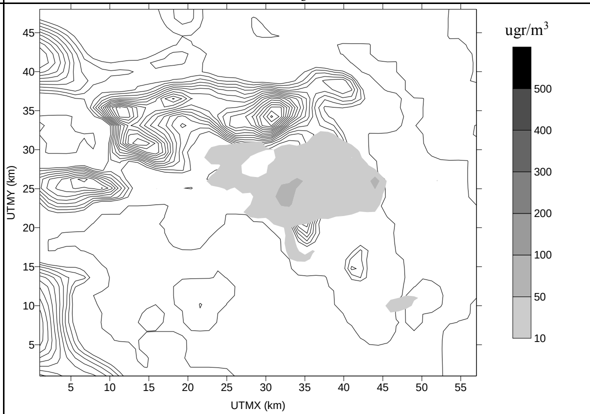
2a. NO_2 at 03:00 LST from CALPUFF



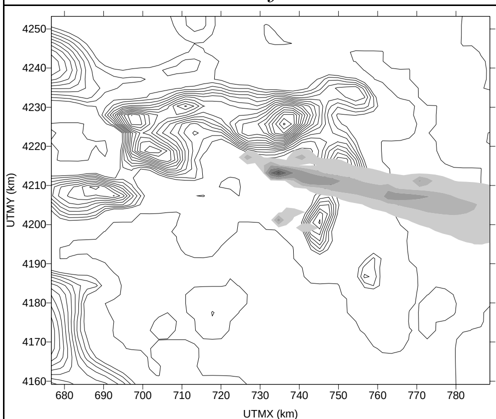
2b. NO_2 at 15:00 LST from CALPUFF



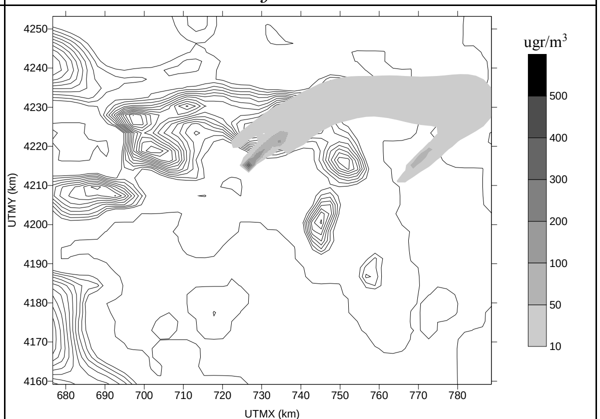
3a. NO_2 03:00 LST from UAM



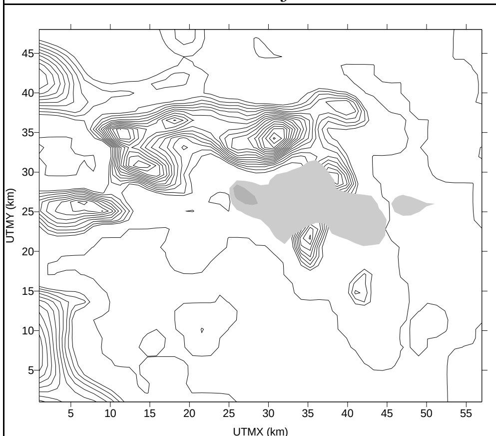
3b.NO_2 15:00 LST from UAM



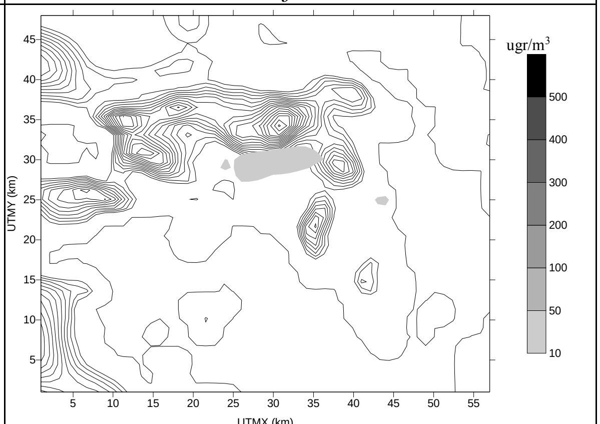
4a. PM_{10} 03:00 LST from CALPUFF



4b. PM_{10} 15:00 LST from CALPUFF



5a. PM_{10} 03:00 LST from REMSAD



5b. PM_{10} 15:00 LST from REMSAD

Figures 2-5. Spatial distribution of NO_2 and PM_{10} over GAA.

In figures 2-5 it is shown that there is an agreement between the regulatory model (CALPUFF) and eulerian models calculations, especially during unstable atmospheric conditions, when strong vertical movements prevail. In contrast, there are significant discrepancies during stable conditions that can be attributed to the strong local circulations. In the case of CALMET, the formation of plumes is apparent, which can be partly attributed to the homogeneous wind fields and also to the fact that the emissions from the point sources retain the plume characteristics. In contrast, the eulerian models immediately disperse the emissions in the whole grid cell, where the point sources are located. Thus, the model either overestimates or underestimates the meteorological parameters in areas where the local topographical features generate significant circulation patterns. As a result, the validity of the model is expected to be less accurate in areas with apparent topographical features where the existing meteorological network does not adequately cover.

Table 1. NO_2 and PM_{10} concentrations (ugr/m^3) at 3:00 LST and 15:00 LST at all stations EXP: experimental measurements, CAL: CALPUFF and REM: REMSAD.

STAT	C	EXP NO_2 3:00	**EXP NO_2 15:00**	CAL NO_2 3:00	**CAL NO_2 15:00**	UAM NO_2 3:00	***UAM* NO_2 15:00**	REM NO_2 3:00	**REM NO_2 15:00**	EXP PM_{10} 3:00	**EXP PM_{10} 15:00**	CAL PM_{10} 3:00	**CAL PM_{10} 15:00**	REM PM_{10} 3:00	**REM PM_{10} 15:00**
PAT	UT	84	**121**	43	**28**	116	**59**	45	**45**						
ATH	UT	32	**39**	38	**24**	124	**62**	43	**35**						
MAR	UT	16	**21**	58	**31**	89	**43**	39	**32**						
PEI1	UT	52	**80**	25	**12**	36	**26**	2	**3**	29	**33**	1	**0**	10	**0**
GAL	UB	55	**32**	41	**34**	102	**48**	41	**45**						
GOU	UB	15	**25**	42	**26**	120	**62**	30	**32**	31	**20**	2	**2**	20	**2**
SMI	UB	18	**17**	71	**9**	112	**22**	22	**2**						
PEI2	UB	34	**43**	9	**6**	50	**36**	6	**4**						
PER	UB	9	**14**	27	**14**	41	**26**	22	**11**						
VOT	SI	15	**24**	38	**26**	113	**47**	48	**19**						
ELE	SI	5	**11**	2	**6**	24	**7**	7	**3**						
ZOG	SB	9	**12**	50	**25**	47	**35**	15	**18**	24	**34**	2	**2**	11	**1**
THR	SB	7	**4**	0	**10**	1	**4**	1	**5**	13	**13**	0	**16**	0	**4**
LIO	SB	10	**15**	5	**11**	4	**8**	1	**11**						
PAR	SB	13	**13**	51	**18**	36	**29**	16	**17**	27	**19**	2	**1**	15	**1**
PEN	SB	7	**9**	7	**28**	43	**21**	21	**19**	21	**19**	5	**3**	12	**2**
KAP	SB	4	**7**	1	**10**	7	**7**	6	**9**						
SPA	SB	18	**37**	28	**7**	23	**20**	15	**21**						
PAL	SB	21	**29**	52	**13**	31	**27**	23	**20**						
GLY	SB	20	**17**	49	**20**	34	**31**	16	**21**						
KOR	SB	8	**21**	34	**8**	29	**7**	13	**4**						
MAR	SB	7	**12**	37	**6**	30	**5**	16	**6**						
ART	SB	11	**13**	44	**16**	25	**14**	15	**15**						
LYK	S	14	**17**	4	**32**	39	**31**	21	**20**	34	**43**	55	**3**	32	**11**

The comparison of models results to the measurements, confirms the previous findings (Table 1). During the night, the CALPUFF predicted reasonable NO_2 values, at half of the stations, which are characterized mainly as suburban industrial (SI) and suburban background (SB) concentrations. At the same time, REMSAD gave the closer calculations to the measurements, while UAM calculated the higher concentrations for all the urban stations (traffic and background). The discrepancy between the eulerian models is mainly due to the differences in ozone concentration predictions caused by the two versions of the Carbon Bond chemical mechanism. Nevertheless, UAM simulations were the most appropriate at the locations with the higher emissions, city center (PAT station) and harbor (PEI). During daytime, the results were substantially improved, for all the three models, while the two eulerian simulated comparable values. Regarding the PM_{10}, either CALPUFF or REMSAD gave lower values, especially during the day. The values predicted by REMSAD are very close to the measurements during the nighttime. The concentrations at a suburban station (LYK) were very close between the two models and the measurements, during the night. This was anticipated, as there is a substantial transport of pollution to this area from the industrial area of Thriassion plane. Summarizing, the inter-comparison between the models results and the measurements was better than 35% for UT stations, up to 80% for UB stations, up to 75% for

SI stations and up to 65% for SB stations, during the daytime hours than during the night time. As for CALPUFF, it is interesting to mention that the results were substantially improved, especially during the night, when the additional surface stations, as well as the vertical meteorological profiles generated by the MM5 model, were also introduced as input data. Some miscalculations from all the models may occur due to the non-recently updated emissions inventory. In particular, the emissions are corresponded to the inventory recorded in 1998 when the new Athens airport was not on service, while experimental campaign took place in 2002 with the additional burden of the increased number of vehicles. This is more obvious in the PM_{10} calculations, as the increase of PM_{10} emissions due to the Athens 2004 Olympic Games (construction of significant infrastructures in GAA) and the absence of the seaborne aerosol simulation are not recorded in this inventory, but there are included in the experimental measurements. It is believed that an up-to-date emissions inventory, would improve all three simulations.

Acknowledgments – This study was supported by the European Union, within the ICAROS-NET project

REFERENCES

Anthes R. A., 1972: The development of asymmetries in a three-dimensional numerical model of the tropical cyclone. *Mon. Wea. Rev.,* **100**, 461-476.

Athanasopoulou, E., E. Bossioli and M. Tombrou, 2004: Modeling of particulate matter in the Greater Athens Area by REMSAD model, 9th International Conference on Harmonization within atmospheric dispersion modeling for regulatory purposes. Garmisch-Partenkirchen, Germany.

Binkowski, F.S. and U. Shankar, 1995: The regional particulate matter model, 1: model description and preliminary results. *J. of Geophysical Research*, **100**, 26191-26209.

Gery, M.W, G.Z. Whitten and J.P. Killus, 1989: A photochemical kinetics mechanism for urban and regional computer modelling, *J. Geophysical Research,* **94**, 12, 925-12,956.

ICF Consulting, 2002: User's guide to the Regional Modeling System for Aerosols and Deposition (REMSAD).

Odum, J.R., T.P.W. Jungkamp, R.J. Griffin, R.C. Flagan and J.H. Seinfeld, 1997: The Atmospheric Aerosol-Forming Potential of Whole Gasoline Vapor, Science, Vol. 276, pp 96-99. See also Environmental Science & Technology, **31**, 1890-1897.

Prosmitis, A. B., Diapouli E., Chaloulakou A., Spyrellis N., 2003: Organic and elemental carbon particulate continuous field measurement in Athens urban area. 8th Int. Conference on Environ Sci and Tech

SAI, 1992: User's Guide for the Urban Airshed Model, Volume IV: Emissions Preprocessor System (Version 2.0). U.S. Environmental Protection Agency (EPA-450/4-90-007D(R)).

Scire J. S., Strimaitis D. G., Yamartino R. J., 2000: A user's guide for the CALPUFF dispersion model.

Symeonidis, P, I. Ziomas and A. Proyou, 2003: Development of an emission inventory system from transport in Greece, Environmental Modelling & Software XX (2003) XXX-