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EVALUATION OF TWO MESOSCALE MODELING SYSTEMS USING DIFFERENT CHEMICAL MECHANISMS

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1. INTRODUCTION

High levels of photochemical pollutants, like ozone, frequently affect south European countries and the concern with this subject substantially increased during the last decade. In the West coast of Portugal, where the human activities are concentrated, several episodes of photochemical pollution have been verified (Borrego *et al*., 2000). This coastal zone is strongly influenced by the nearby Atlantic Ocean, with frequent sea/land breeze circulation.

Aiming to contribute to a better understanding of these mesoscale phenomena a field campaign was carried out in a coastal region, Aveiro, from 25th June to 2nd July of 2001, which covered the last ozone episode noticed in the region. Measurements of all the main meteorological parameters and of ozone and its precursors concentrations were taken at surface in five different locations and in altitude.

In addition, two mesoscale modeling systems (MEMO-MARS and MAR IV), with three different chemical mechanisms (EMEP, KOREM and CB-IV) were applied to the study area, for the two most representative days of the field campaign. The main purpose was to investigate the origin and formation of the measured high air pollution levels, to evaluate these systems, and to analyze the influence of the chemical mechanism type in the simulations.

2. CHEMICAL MECHANISMS

A critical component of air quality models is the chemical reaction mechanism that describes how volatile organic compounds (VOCs) and nitrogen oxides (NO_X) interact to produce ozone and other oxidants. Chemical mechanisms were first used in models more than 20 years ago. Since that time, there has been an enormous growth in our understanding of the chemical processes that lead to oxidant production, especially in the role that organic species play in the process.

The MAR IV system integrates two models: the Systems Applications International Mesoscale Model (SAIMM), and the photochemical Urban Airshed Model (UAM-IV). UAM-IV is a three-dimensional Eulerian model that contains the photochemical Carbon Bond Mechanism (CB-IV). This version uses 85 reactions and 23 species to solve chemical kinetics, and explicitly treats the isoprene. CB-IV is a lumped structure mechanism used for both urban and regional scale modeling. In the lumped structure approach, organics are divided into smaller reaction elements based on the types of carbon bonds in each species. Organic reactions and products in the CB-IV are represented using 12 species (USEPA, 1990).

In what concerns the other mesoscale system, the meteorological MEMO model was applied jointly to the photochemical dispersion MARS model. Two chemical mechanisms were analyzed: EMEP, which describes the tropospheric gas-phase chemistry with 66 species, 139 photochemical reactions including 34 photolysis reactions; and the KOREM mechanism, which is a simplest one, including 39 chemical reactions and 20 reactive pollutants. KOREM is the combination of anorganic reactions of the CERT mechanism and of organic reactions of the compact mechanism of Bottenheim and Strausz. The chemistry of methane is also taken into account by including the reaction of methane with the hydroxyl radical. In KOREM mechanism the VOCs are lumped in five classes (Moussiopoulus, 1992). On the other hand, the EMEP mechanism considers 13 categories as VOC speciation (including ketones, alcohols and isoprene).

3. APPLICATION AND VALIDATION

Analysis of temporal evolution of the meteorological and air quality parameters data series revealed that high levels of ozone (exceeding 180 $\mu g.m^{-3}$) were registered on 30th of June and 1st of July, under the influence of a thermal surface low-pressure system, common over the Peninsula during summer and responsible for some ozone episodes. For this reason these days were selected for the simulation using a 200x140 km^2 domain with 5 km resolution applied to both modeling systems, MEMO-MARS and MARIV.

In order to obtain the resolution required by air quality models, the anthropogenic emissions data (NOx, CO and VOC) have been disaggregated (geographically and temporally) from the national emissions values of the most recent CORINAIR inventory using adequate statistical indicators (top-down methodology). On the other hand, the biogenic emissions (isoprene and monoterpene) from forest were calculated using local and detailed dataset and emission factors (bottom-up methodology) (Monteiro *et al*., 2001). In what concerns to the VOC emissions, they were split in different species, according to each

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chemical mechanism, using adequate literature profiles.

In order to better evaluate model performance, and regarding the quantity of data sets that have to be compared, a statistical analysis was applied. The statistical parameters S, S_{obs}, E and E_{UB} were calculated. It is possible to say that the simulation presents a good skill when $S \approx S_{obs}$, $E < S_{obs}$ and $E_{UB} < S_{obs}$ (*E* = root mean square error, rmse, E_{UB}=rmse after removal of a certain deviation, *S* and S_{obs}=standard deviation of modelled and observed data) (Keyser and Anthes, 1977).

Table 1 presents the statistical results performed. Globally, UAM-IV simulations present better results than MARS simulations. Concerning the different locations one can say that the worst statistical correlations were verified for Covelo, specially regarding parameters E/S_{obs} and E_{UB}/S_{obs} for MARS simulation. Comparing KOREM and EMEP simulations, they are quite similar, but EMEP results tend to be better than KOREM results.

TABLE 1 – Statistical analysis for the 3 different model applications, at 3 surface stations.

		Aveiro	Sangalhos	Covelo
UAM-IV **CBIV**	S/S_{obs}	0,99	1,07	0,84
	E/S_{obs}	0,82	0,51	0,65
	E_{UB}/S_{obs}	0,77	0,52	0,51
MARS **KOREM**	S/S_{obs}	0,80	0,59	0,41
	E/S_{obs}	1,20	1,00	2,08
	E_{UB}/S_{obs}	0,76	0,72	1,27
MARS **EMEP**	S/S_{obs}	0,88	0,63	0,73
	E/S_{obs}	1,21	0,99	1,38
	E_{UB}/S_{obs}	0,98	0,90	1,33

In Figure 1 the spatial distribution of concentration differentials for CBIV and EMEP, and EMEP and KOREM, for 13 UTM are presented.

As it would be expected, differences in Figure b) are smaller, since KOREM and EMEP mechanisms are part of the same modeling system. Regarding the 3 studied stations, the absolute differences between the mechanisms are not so significant as in other areas of the domain with higher ozone levels (near Porto urban and industrial area).

4. CONCLUSIONS

Both modeling systems presented a reasonable performance from the photochemical air pollution point of view. Concerning the chemical mechanisms, preliminary conclusions point to a non-significant improvement of results associated to a more complete description of photochemical reactions. Regarding the CB-IV mechanism results, it can be concluded that chemical speciation (VOC in particular) could be a more relevant factor than the number of chemical reactions included in the mechanism.

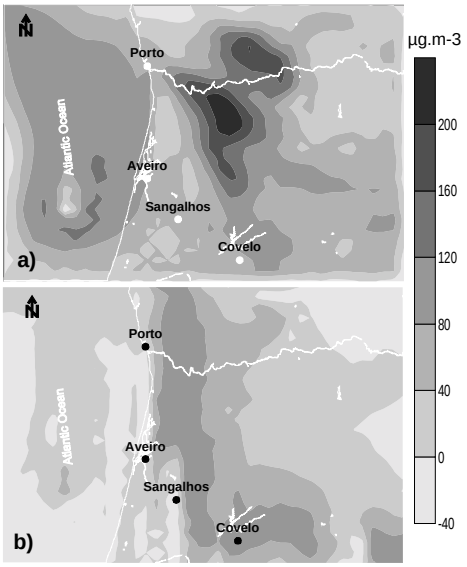

FIGURE 1 – Spatial distribution of concentration differentials for a) CBIV-EMEP, b) EMEP-KOREM

Comparison of results obtained for the 3 different chemical mechanisms should take into consideration that the meteorological model used is not the same for both modeling systems. Therefore, differences verified could, in some extent, be due to the meteorological simulation, in spite of the quite good performance of both models. This study should be regarded as a first attempt to compare model performance using different chemical mechanisms.

5. REFERENCES

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