1.23   A COMPARISON OF MODAL AND SECTIONAL APPROACH IN AEROSOL MODELING IN THE MILAN AREA.

Monforti-Ferrario F.[1], Pederzoli A.[1], Silibello C.[2]
[1]ENEA, Bologna, Italy, [2]ARIA-NET srl, Milan, Italy

INTRODUCTION
Two box models implementing respectively the sectional and the modal approaches have been used to simulate aerosol concentrations in Milan area.The first box-model, called AERBOX, follows the sectional approach.In this framework, particle size distribution is fully described by means of 64 dimensional classes (size bins) and the time evolution of each one of the 14 chemical components of each size class is governed by a specific progonostic equation.
The second box-model include the so-called aero3 module, implemented in US-EPA CMAQ modeling system, that follows the modal approach. In this module, particle size distribution is described by the superposition of three lognormal distributions, named "modes": Aitken mode (i-mode), accumulation mode (j-mode) and coarse mode (c-mode). The core of the model consists in a set of prognostic equations for some integral properties of the particle size distribution, namely the total particle number concentration, the total surface area concentration and the total mass concentration of the individual chemical components in each mode.
Both AERBOX and Aero-3 take into account the main chemical and physical processes involving the particulate matter and the gaseous pollutants in the atmosphere such as nucleation, condensation/evaporation and deposition, whereas only Aero-3 deals with coaugulation.
Particles and gases hourly concentrations produced by the two models have been compared in medium term simulation on the Milan area. The simulation involved a 50 km x 50 km square centered on the town of Milan.
Comparison results are expected to give useful suggestions for the development of the MINNI system (Zanini et al., The MINNI integrated modeling system. Harmo9 abstract book).

THE MODELS
AERBOX
AERBOX is a box model dealing with PM evolution and taking into account the following physical and chemical processes:
1. condensation/evaporation of inorganic compounds through the AIM model (Wexler and Seinfeld, 1991)
2. nucleation of system H_2O-H_2SO_4 applying the method of critical concentration (Wexler et al, 1994)
3. production of SOA (secondary organic aerosols) through Pandis's theory (1992)
4. evolution of the boundary layer height according to Batcharova and Gryning (1990) for instable atmospheres and according to Zilitinkevich (1990) for stable and neutral atmospheres.

The chemistry of pollutants in gaseous phase is treated by means of the Carter's SAPRC90 method (1990).
AERBOX produces the hourly concentrations of 14 chemical aerosol components for 64 dimensional classes and the hourly concentrations of 52 gaseous compounds, expressed in $\mu g/m^3$.

aero-3

To compare AERBOX performance with a model based on the modal approach, aero-3, the module of Models-3/CMAQ (community multy-scale Air Quality system) dealing with dynamic and chemistry of particles has been included in a box model framework very similar to AERBOX as far as meteorological features and gas phase chemistry are concerned. These routines represent the particle size distribution as the superposition of three lognormal sub-distributions called "modes". The smaller, called Aitken mode, represents fresh particles either from nucleation or from direct emissions, while the accumulation mode represents aged particles. Primary emissions may also be distributed between these two modes. The two modes interact with each other through coagulation. The coarse mode species include sea salt, wind-blown dust and other unspecified material of anthropogenic origin. Each mode may grow through condensation of gaseous precursors and is subject to wet and dry deposition. The most important processes treated by Aero-3 are:

1. condensation/evaporation as described by Whitby (1991)
2. nucleation of system H_2O-H_2SO_4 applying Kulmala's method (1998)
3. production of SOA (secondary organic aerosols) applying Schell's method (1992)
4. coaugulation using numerical quadratures (Whitby *at al.*, 1991)

MODEL COMPARISON

Simulation domain

The performances of Aero-3 and AERBOX were compared on a 50 km X 50 km domain including the town of Milan and its suburbs (figure 1).

Figure 1. Map of Lombardy (Northern Italy) showing the simulation domain (in grey), including the town of Milan and its province (in black)

Meteorology

Meteorological data were obtained from soundings made by Italian Air Force in Milano-Linate; these six-hourly measures provide the most important meteorological parameters (temperature, pression, wind, relative umidity...) as a function of altitude. A linear interpolation in time has been executed to obtain missing data and vertical averages have been used as input for box models.

Emissions

In order to obtain the detailed input needed, the emission pre-processor THOSCANE (Tool for Hourly Speciation of CORINAIR Annual Emissions, Monforti *et al.*, 2003) was applied on emission inventories used in the City Delta exercise.
THOSCANE allows the user to split annual emissions into hourly emissions taking into account the features of each source category (SNAP). It calculates also the detailed chemical composition of hourly emitted VOC and it applies the lumping method suggested in the frame of SAPRC-90 (Carter, 1990) chemical mechanism to reduce to a limited VOCS categories.

Finally THOSCANE differentiates PM chemical composition as a function of particles size giving different percentage values for $PM_{2.5}$, $PM_{(2.5-10)}$, PM_{10}.

Boundary conditions

The boundary concentrations of gaseous pollutants have been provided by the EMEP model. For PM, instead, background concentrations provided by the urban station of Limito have been used and a typical chemical speciation of the Po valley background aerosol coming from the PIPAPO campaign (Pianura Padana Produzione Ozono) has been supposed.

RESULTS

PM_{10} concentrations

AERBOX and Aero-3 performances were tested by simulating PM_{10} concentrations during two months: January 1999 and April 1999.
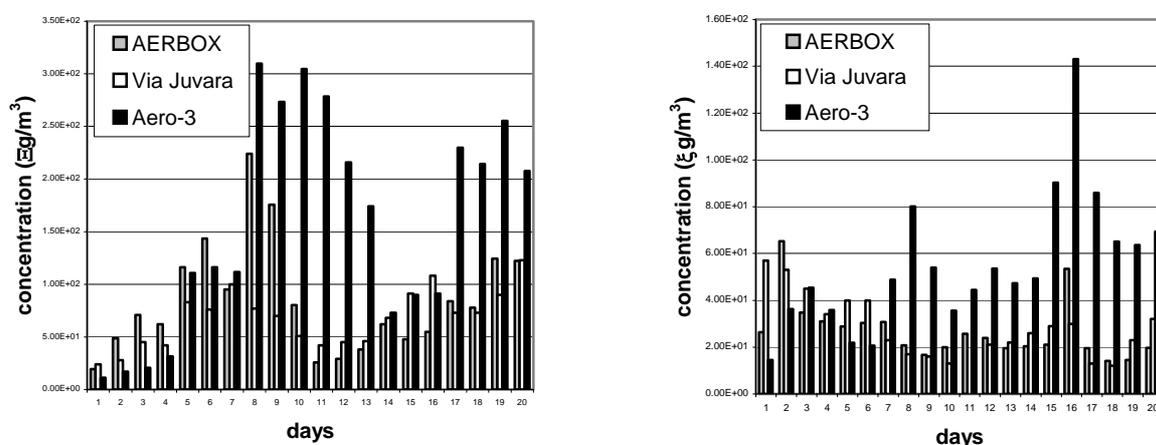

Figure 2. Daily means of PM_{10} concentrations in January (left) and April (right). AERBOX (grey), Aero-3 (black) and urban background station in via Juvara (white).

Figure 2 shows daily means of PM_{10} concentrations as modeled by AERBOX and Aero-3, compared with PM_{10} concentrations measured in the urban background station in Milan (Via Juvara).

Performance analysis

The usual performance indicators have been computed for two hours modeled PM_{10} concentrations in comparison with Via Juvara station. Results are shown in table 1 and 2.

Table 1. Performance statistics obtained from simulation in January 1999

	average	Sigma	bias	nmse	Cor	fa2	fb	fs
Milano(Via Juvara)	68.95	37.32	0.00	0.00	1.00	1.00	0.00	0.00
Aero-3	149.58	114.73	-80.62	1.86	0.224	0.455	-0.738	-1.018
AERBOX	89.28	92.17	-20.33	1.37	0.268	0.581	-0.257	-0.847

Table 2. Performance statistics obtained from simulation in April 1999

	Average	sigma	bias	nmse	Cor	fa2	fb	fs
Milano (Via Juvara)	31.06	19.78	0.00	0.00	1.00	1.00	0.00	0.00
Aero-3	55.18	42.18	-24.11	1.87	-0.276	0.443	-0.539	-0.723
AERBOX	28.75	23.50	2.32	0.81	0.238	0.741	-0.077	-0.172

Table 1 and 2 evidence that all indexes indicate that AERBOX performs better than Aero-3. In order to identify possible influences of meteorological parameters on the model performance, the residual analysis has been applied to Aero-3 results.
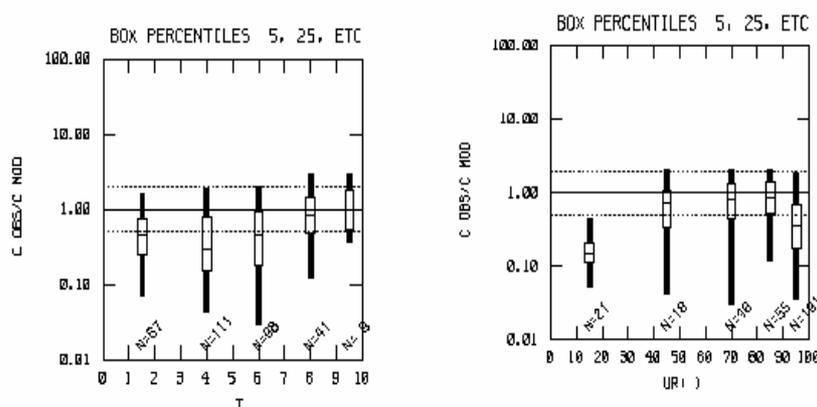Figure 6 shows the residual plots when temperatures and relative humidity are considered.

Figure 6. Residual analysis for PM$_{10}$ concentration by Aero-3 varying relative humidity and temperature

CONCLUSIONS
Both AERBOX and aero-3 box models use the same chemical mechanism for gaseous compounds, named SAPRC90 (Carter, 1990) and share almost completely the treatment of meteorological variables.
They mainly differ in the description of the most important chemical and physical processes involving particles, such as nucleation, condensation/evaporation, deposition, and in the rapresentation of the granulometric spectrum of particles: sectional in AERBOX, modal in Aero-3. Two simulations performed on the Milan area were compared together and with sperimental data in january 1999 and april 1999. In both cases Aero-3 overestimates PM$_{10}$ concentrations, while AERBOX shows a better agreement to experimental data. Residual analysis suggests that a reason could be found in the difficulty of Aero-3 in evaluating H$_2$O concentration in aerosol phase, especially during periods of high relative humidities (~ 90%). On the other side, the sectional approach adopted in AERBOX requires a large numbers of variables (equal to the product between the number of bins and the number of chemical components) and equations. Together with the more careful numerical representation of the water dynamics, this approach makes AERBOX sensibly heavier from the point of view of the computation time.
In the future more detailed tests will be carried on to confirm or confute these findings.

REFERENCES

City Delta emission inventory for the Milan Area. Available at http://rea.ei.jrc.it/netshare /thunis/citydelta

Carter W P L, A detailed mechanism for the gas phase atmospheric reactions of organic compounds, *Atm Env*. 1990, **24A**, 481-518

Wexler A S, Lurmann F W, Seinfeld J H, Modeling urban and regional aerosols – I. Model development. *Atm. Env*. 1994, **28**, 531-546

Batcharova E, Gryning S-E, Applied model for the growth of the daytime mixed layer. *Boundary layer meteorology*, **56**, 261-274.

Pandis S N, Wexler A S, Seinfeld J H, Secondary organic aerosol formation and transport – II. Predicting the ambient secondary organic aerosol size distribution. *Atm Env*. 1993, **27A**, 2403-2416

Zilitinkevic S S, Velocity profile, the resistance law and the dissipation rate of mean flow kinetic energy in a neutrally and stably stratified planetary boundary layer. *Boundary layer Meteorology* , **46**, 367-387