INVITED

The spatial and temporal pattern of ozone in the provinces of Milano, Varese, Bergamo and Como (Italy)

E. ANGELINO (1), M. BEDOGNI (2), E. BRAVETTI (3), R. GUALDI (1), G. LANZANI (4), C. LAVECCHIA (5), A. MUSITELLI (6), M. VALENTINI (1)

(1) Environmental Protection Office of Milan (P.M.I.P of Milan), Via Juvara 22, 20129 Milano, Italy
(2) Environmental Consultant, Via Lanfranco della Pila 57/e, 20162 Milano, Italy
(3) Environmental Protection Office of Varese (P.M.I.P. of Varese), Via Campigli 5, 21100 Varese, Italy
(4) Province of Como, Via Borgovico 148, 22100 Como, Italy
(5) Environmental Consultant, Via Bosatra, 20064 Gorgonzola (Mi), Italy
(6) Environmental Protection Office of Bergamo (P.M.I.P. of Bergamo), Via C. Maffei 4, 24100 Bergamo, Italy

In the last years photochemical pollution has been recognised as a critical environmental problem. Summer smog episodes occur over large parts of Europe and also in the North of Italy high ozone concentrations are measured. A working group is investigating the ozone distribution and estimating the contribution of breeze circulations transport to the actual ozone levels measured in the territories of the provinces of Milan, Bergamo, Varese and Como in the Northern Italy. In this paper the principals results of this outstanding study are presented. Times series of ozone concentrations measured in the last years by the four provincial air pollution networks, including 18 monitoring sites, have been analysed by comparing trends and frequency of exceedances of air quality standard limits. Moreover a statistical analysis has been made to evaluate similarity among measured ozone concentrations in terms of levels and temporal variability (frequency and peaks distribution, daily shape etc.). This study covers the summer period from June to September 1992 and uses the measured 1-h ozone concentrations in two different test cases. The first case considers day-time ozone data (9.00-20.00); the second one includes also night time ozone data (0.00-24.00). The method developed is based on hierarchical Cluster Analyses techniques. Every group has been well characterized by an averaged hourly concentration and relative variance. The statistical analyses has shown a summer time gradient of surface ozone concentrations going from South to North of the area under study and interesting similarities among monitoring sites in the day time maximum peak occurrence.
Wind field and the typical breeze circulations have been described on the base of measured data and on gridded wind fields simulated by using a mesoscale meteorological model, CSUMM. Principal ground-level wind circulation has been pointed out; the next phase of the study, which concerns the backward trajectories analysis, will investigate this phenomenon thoroughly.

1. INTRODUCTION

High levels of photochemical smog have been pointed out by a more and more developed air quality monitoring network. Particularly critical episodes occur in regions exposed to high insolation, downwind to highly urbanized towns, intense sources of precursors: oxides of nitrogen, primarily from automobile and industrial emissions, and hydrocarbons. Ozone and precursors can be transported up to hundreds of miles downstream from urban sources and persist on the order of hours to days. In some instances this transport is nearly the sole cause of the exceedances of the state ambient ozone air quality standard in downwind areas, far from precursors sources. Then photochemical phenomena require to be studied on mesoscale basin.

To investigate the ozone distribution on the western territories of Lombardia region, located in the North of Italy, a working group has been established during summer 1993. The final aim of the work is to characterize the impact on ozone levels and the contribute to exceedances in downwind areas, in the north of the domain under study, of precursors emitted in the densely urbanized upwind areas located in Milan and surroundings.

2 THE AREA OF STUDY

The area of study is located in a basin of approximately 150 x 120 Km and includes the territories of the provinces of Bergamo, Como, Milano and Varese. At the northern side Alps Mountains are the physical barrier between Italy and Switzerland. The topography of the study area is complex, the altitude increases from 30 m to 3000 m above sea level going from South to North. Moreover numerous valleys cross the basin: the Valtellina to the North, along East - West direction; the Po river valley to the South, and the Ticino river valley to the West. The area comprehends also many lakes. The chief ones are the Lake of Como, the Lake Maggiore, and the Lake of Lugano. The most inhabited areas are located in the South, around the city of Milan, where numerous industrial plants are concentrated. Going from South to the North, inhabitants and industries density decreases.

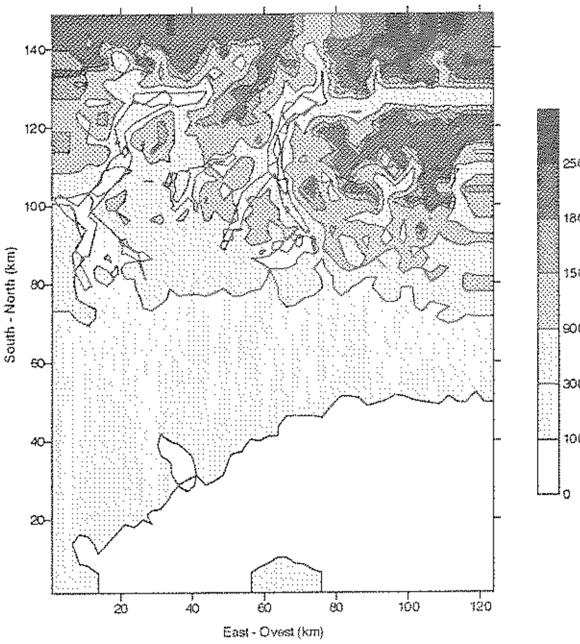

Figure 1: Topography of the study area. The domain is 150x125 km and altitude isopleths are contoured at 100, 300, 900, 1800 and 2500 m above sea level.

3. TYPICAL WIND FIELDS

Alps mountains surrounding the area to the North are the physical barriers that protect the regional under study by the intense circulations becoming from Europe and from Mediterranean Sea. Only with the presence of great pressure gradients between the two opposite sides air flows can overpass the orographic obstacle, generating the typical phenomena of the "Foehn", a strong dry wind coming down from the Alps to the plain. During summer period the weather conditions are characterized by mountain-valley circulations. These circulations are due to the temperature discontinuities between mountains and the lowland plains, in the southern regions. The typical breeze circulations tend to come from South during daytime (valley breeze) and from North during the night (mountain breeze). Moreover, on local scale, inside the numerous valleys, circulations generated by altimetric gradient prevail over mesoscale breezes. Calms or weak circulations characterize wind field in the southern region of the area, in the province of Milan, occurring approximately 80% of the days in the cold season and approximately 40% during summer.

Figure 2 presents the wind field pattern in the late afternoon derived from measures at meteorological monitoring sites of the provincial networks. It is evident the typical flow of the valley breeze: winds are directed from South to North. The anemological field near Milan is affected by "urban heat island" effect. In fact the wind blowing to the city is rotated in relation to the city centre and tends to settle in its original direction once they have passed the city limits. Wind field in PreAlps and Alps blow along the principal valley's axes. For example it is evident the flow along the Lake of Como, turning to the right, in the North, towards Valtellina valley.

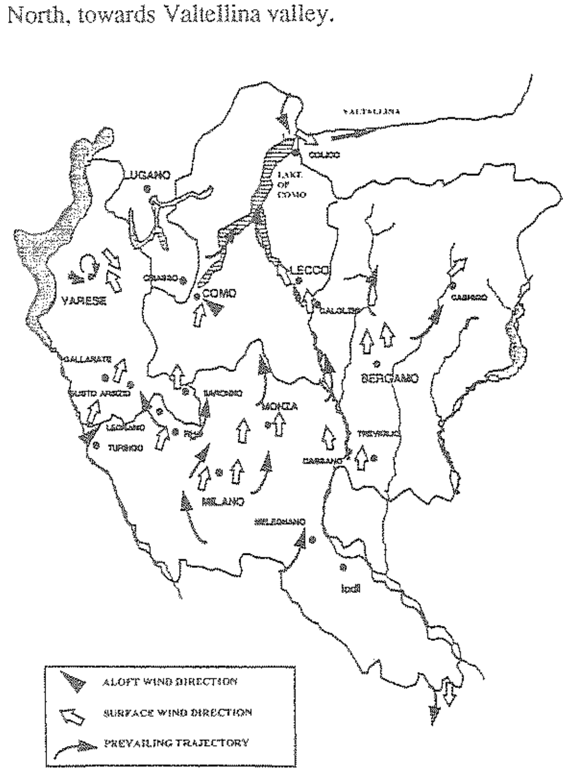

Figure 2: Typical breeze circulations in summer time in the late afternoon

Figure 3 shows the wind vector field calculated by CALMET (diagnostic meteorological model; Scire, 1990) on the base of measures of monitoring sites and the vector fields generated by CSUMM, a tridimensional prognostic meteorological model, developed by the Colorado State University (Kessler, 1989). After a detailed sensitivity analysis, the model CSUMM has been used in the study area. For the wind field calculation a numerical grid of 50x60 grid points was used. A horizontal grid spacing of 2.5 km was applied. The figure is referred to the anemological field at the same time of figure 2, in the late afternoon. Comparing the flow patterns qualitatively, it could be evidenced that the calculated wind field follows the measured one both in intensity and in direction.

4. DATA ANALYSIS

4.1 Description of the measurements sites

The study area is monitored by four provincial air quality networks. In particular, ozone is measured in 18 monitoring sites. UV absorption photometric analysers are used for the measurement of ambient levels at all considered sites, except for the station of Bergamo Opsis, where a D.O.A.S. (Differential Optical Absorption Spectroscopy) technique is used.

Monitoring sites are linked to a data telemetry system which allows the transmission of results over telephone lines, to a computerized network control centre, one for each of the four provinces. Data are also validated and processed before the subsequent public dissemination via press and TV.

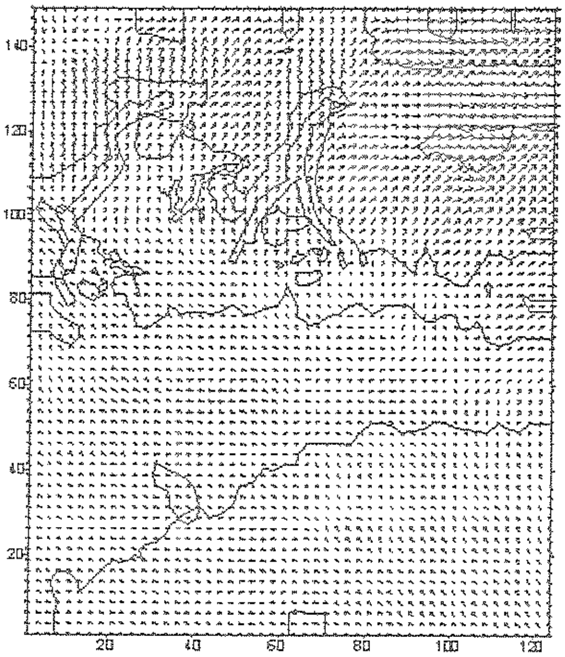

Figure 3: Surface wind field computed by CALMET and CSUMM at the late afternoon during summer time.

The stations are located in very different areas. Some stations are placed in densely urbanized centres. Other sites are in suburban locations. Two stations may be considered rural (Varenna and Varese Vidoletti). One of the sites is located in urban park, in the city of Saronno (in the Province of Varese). Another one is situated in the countryside, but near a chemical plant (Limito, East of Milan). Some new locations of monitoring network, not yet currently operating, will be placed in sites remote from areas of intense primary pollutant emissions.

4.2 Ozone measurements

The air quality standard ozone value defined from Italian laws is 200 µg/m^3 as hourly averaged concentration, not to be overpassed more than once in a month.

In the study area frequent exceedings of this limit occur during summer time. A summary of guideline exceedences covering all monitoring sites and 1992 and 1993 years is provided in table 1. Table 2 shows monthly averaged concentrations. It should be noted that the minimum values are measured in the urban stations, located in Milan and the other towns of the study area (Bergamo, Legnano, Como, Lecco, Corsico, Busto). Obviously, the higher values are registered in rural stations. Moreover, it is evident that, generally, there is an increasing trend from South to the North, probably related to transport of ozone and its precursors by the breeze circulation discussed before. This gradient will be more evident from the applications of the cluster analysis, to the detailed study of spatial variations.

Figure 4. Ozone monitoring network

During summer time, the typical diurnal pattern presents maximum values one - two hours after the time of higher temperatures and insolation.

STATION	June		July		August		September	
	1992	1993	1992	1993	1992	1993	1992	1993
BG S. Giorgio	0	0	2	0	0	3	0	0
BG Opsis	N.A.	20	N.A.	13	N.A.	16	N.A.	0
Como Centro	0	0	0	0	0	1	0	0
Lecco	0	0	0	0	0	0	0	0
Merate	0	0	6	4	0	2	0	0
Varenna	3	26	77	21	53	8	2	0
MI Juvara	0	0	0	0	0	0	0	0
MI Verziere	0	N.A.	0	0	0	0	0	0
Legnano	0	0	0	3	0	0	0	0
Monza	0	0	1	0	3	7	0	0
Corsico	2	0	0	0	0	0	0	0
Cinisello	0	N.A.	0	0	8	8	0	0
Limito	0	3	7	3	5	8	0	0
VA Artistico	1	N.A.	18	N.A.	4	N.A.	0	N.A.
VA Vidoletti	19	N.A.	102	N.A.	20	N.A.	4	N.A.
Saronno	0	N.A.	20	N.A.	9	N.A.	1	N.A.
Busto A. Pal.	1	N.A.	0	N.A.	0	N.A.	0	N.A.
Busto A. Mag.	0	N.A.	8	N.A.	10	N.A.	0	N.A.
Gallarate	0	N.A.	13	N.A.	4	N.A.	0	N.A.

Table 1: Ozone guideline exceedences (200 µg/m3) in 1992 and 1993 (NA: not available).

STATION	June		July		August		September	
	1992	1993	1992	1993	1992	1993	1992	1993
BG S. Giorgio	36	47	48	52	54	82	18	23
BG Opsis	N.A.	92	N.A.	92	N.A.	103	N.A.	48
Como Centro	23	32	39	38	38	55	12	N.A.
Lecco	25	47	41	46	58	63	30	30
Merate	31	40	41	46	53	57	20	19
Varenna	66	90	106	88	112	94	66	48
MI Juvara	30	39	34	42	47	53	23	14
MI Verziere	36	N.A.	52	43	47	45	25	19
Legnano	N.A.	32	39	37	39	46	20	12
Monza	28	45	41	44	48	63	28	18
Corsico	38	29	27	29	33	43	18	12
Cinisello	38	N.A.	34	N.A.	50	60	20	16
Limito	N.A.	66	65	68	72	75	34	37
VA Artistico	86	71	75	70	N.A.	77	40	38
VA Vidoletti	95	N.A.	136	99	119	103	75	62
Saronno	49	83	64	N.A.	67	85	35	31
Busto A. Pal.	30	47	54	N.A.	54	58	28	17
Busto A. Mag.	48	55	67	55	74	73	38	20
Gallarate	60	84	87	59	91	66	57	28

Table 2: Monthly averaged 1-h concentrations 1993 (NA: not available).

5. CLUSTER ANALYSIS

5.1 The method

The determination of similarities among monitoring sites in terms of temporal trends and spatial variation has been deepened by cluster analysis application. The objective of the method is to group the data units into cluster such that members within a cluster have a high degree of association among themselves while the cluster are relatively distinct from one another. The similarity among members is established by a distance function, characterized by peculiar analytical properties. The association measures may be used to construct a similarity matrix describing the strength of all pairwise relationships among the entities in the data set. The hierarchical clustering methods operate on this similarity matrix in an agglomerative way. At the beginning each group consists of exactly one entity; at any subsequent stage the two closest clusters are merged and the similarity between the new cluster and any other group is calculated. The cascade of merges continues until all the objects belong to a unique group. The sequence of unions is described by a tree-diagram, that shows the associations computed at any step, and the distance at which they occur. Once a tree is constructed for N objects, analyst may choose from as many as N sets of clusters, looking for the optimal location for each of the objects and the final cluster assignment in which the discordance between the objects in each cluster is minimized. Each cluster is described by a representative of all the elements, chosen according to the type of distance function considered (cluster centroid).

In this work the cluster analysis objects are the 18 monitoring sites. The variables are all homogeneous and are constituted by the hourly averaged ozone concentrations measured at the different locations. The objective of the study is to quantify the similarities among monitoring sites in terms of concentrations levels and temporal trends. So the Euclidian distance and the correlation factor have been respectively chosen as similarity function. In fact, the Euclidian distance permits to point out the similarities from a quantitative point of view, while the correlation factor allows to focalize similarities among temporal trend phases.

The analysis has been applied to the months from June to September 1992 and to the week 28.07.1993 - 04.08.1993. For every period, two different temporal series have been studied; the first one includes the hourly concentrations measured during all the day (1.00 a.m.-12.00 p.m.), while the second one just only those measured during day-time period (9.00 a.m.-08.00 p.m.). The choice of the day-time temporal interval permits to avoid the influence of the night-time concentrations, focalizing on the insolated hours, when peaks of ozone occur. The selected week has been chosen for the elevated concentrations measured in those days.

The clustering algorithm used is the Complete Linkage, that on the base of preliminary tests is resulted the best method compared with the other ones (Average Linkage, Single Linkage, Centroid Method, Ward Method) (Lavecchia, 1992). The number of clusters has been determined by tree-diagram analysis and the study of the within variance. Every group has been characterized by some representative parameters: numbers of elements; monthly averaged concentrations; internal group variance; centroids ozone concentrations.

5.2 Results

The application of the cluster analysis has shown similar results considering the daily period (00.00- 24.00) and day-time one (08.00-20.00). In both cases the identification of four or five clusters has allow to characterize groups as in terms of homogeneity of members as in terms of differences among clusters. The comparison of results relative to different months has shown different values, but similar partitions of the stations. In particular the spatial distribution of members belonging to each group evidences in the study area the following regions:

- southern region includes the province of Milan, of Bergamo and the South of the province of Como;
- intermediate region includes the southern and the prealpine territories of the province of Varese;
- northern region comprehends the high province of Varese and the high province of Como.

In reality this classification is more complex, as it will be described in the following part of the paragraph. The presence of intense NOx emission sources, as highly traffic street, or the existence of parks located near the monitoring sites can strongly influence measured data. So, in these cases, the location of the monitoring sites may be less important.

The comparison of clusters included into the three regions points out an increasing gradient of ozone concentrations going from the South to the North.

Similar results are obtained for the 1993 week. The analysis has been extended to 22 monitoring stations, including 4 located in Switzerland, on the border of Italy. The results of clustering for this week are reported in table 3 and in figures 5 and 6. Codes included in table 3 refer to figure 4, while the codes CH.. are referred to Switzerland stations. Five clusters have been identified. The number of objects varies for the different clusters, from one to seven. The internal variance is obviously 0 for one-element cluster but it is very similar for the other groups, about 20 - 25 $\mu g/m^3$. The cluster with the lower ozone concentration (54 $\mu g/m^3$) groups the stations located in Milan, Como and Lecco towns, near dense traffic streets. Not much higher values characterize members belonging to cluster 4, referred to stations located in the South of study area. The final 3 clusters show the most elevate values, respectively 92, 97, 139 $\mu g/m^3$. The three cluster group the stations located in the north of the study area, excepted for the stations of Saronno and of Limito. The first one is located in a park, in the intermediate region of study. The second, on the contrary, is located in the countryside surrounding Milan, but near a chemical plant. Moreover, it is evident that Switzerland sites belong to the same group that includes also Italian stations out of the border at the same latitude. In figure 5 are reported the ozone concentrations computed for the centroids of the 5 clusters. The differences are well underlined, above all during daytime. Moreover a night peak is well characterized in every cluster, even if in different hours. The nature of such a peak is nowadays not well understood, even if possible causes are related to horizontal advection, due to the change in the breeze direction in nightime from North to South or vertical transport in troposphere, deriving from aloft atmospheric layers. Another possible reason can be related to the reduced atmospheric mixing height in the first hours of the morning.

In table 4, and in figures 7 and 8, the cluster including monitoring site in terms of similar ozone concentration temporal trends are shown, for the partition with a correlation factor of 0.8 in each group. Seven groups have been identified, four with only one element. It is confirmed the tendency of grouping according to the spatial location: south-east stations belong to one group, while north-west stations belong to another. Also the stations located in the Milan area are included in only one cluster. It must be noted that in this case the station of Limito (088) groups with the other stations of Milan. Moreover the Saronno station (905) is jointed with the group comprehending the other stations belonging to the same area.

6. CONCLUSIONS

The first step of the research was a descriptive characterization of photochemical smog in Lombardia region, North of Italy, which suffers of high ozone concentrations during summertime.

cluster	objects number	internal variance	concentr. ($\mu g/m^3$)	stations codes
1	1	0.0	139	903
2	2	20.2	97	905 088
3	6	25.2	92	902 008 CH1 CH2 CH3 CH4
4	7	23.3	74	001 005 015 086 907 906 915
5	6	23.7	54	003 002 014 074 085 006

Table 3: The results of clustering for the week 28.07 - 04.08.1993 in terms of ozone concentration levels (euclidian distance).

cluster	objects number	stations codes
1	1	001
2	1	002
3	1	003
4	9	005 905 014 902 915 907 903 CH2 CH4
5	3	008 CH1 CH3
6	1	906
7	6	006 085 015 086 088 074

Table 4: The results of clustering for the week 28.07 - 04.08.1993 in terms of ozone concentration temporal trends (correlation factor).

Particular attention regarded the study of summer anemological fields, responsible of ozone and precursors transport phenomena. The mesoscale mount-valley breezes, which dominate in high pressure conditions due to the persistence of Azores Anticyclone on Europe, cause transports of ozone from the northern mountains during the night and of precursors from the flat urbanized area to the North. A mathematical model was successfully applied to reproduce intensity and direction of wind breezes. More efforts are to be spent to include the effects of local circulations related to the presence of mountains, lakes, river valleys and towns.

The values and spatial distribution of ozone in the aerological basin were statistically analysed in relation to anemological fields, sources locations and altitude. A gradient of ozone concentration exists along the south-north direction, corresponding to the decreasing of the inhabitants, traffic and factories densities and to the geography. Three areas can be considered homogeneous relative to the hourly ozone concentrations: the southern and northern zones differ in the frequency and intensity of the guideline overcomes and in the daily shape of the trends (different nocturnal and diurnal peak hours, high or low night-time concentrations); the intermediate area presents both the tendencies according to the rural or urban nature of the monitoring sites.

The next efforts of the research will concern the objective characterization and numerical computation of the transport trajectories and the study of the cause-effect relationships among ozone, precursors and meteorology. To collect more significant data, some new monitoring stations were installed and the mobile laboratories were located in mountain and rural sites during the 1994 and 1995 summers. The analysis of these data will improve the overall knowledge about the secondary pollution in the Lombardia aerological basin.

7. REFERENCES

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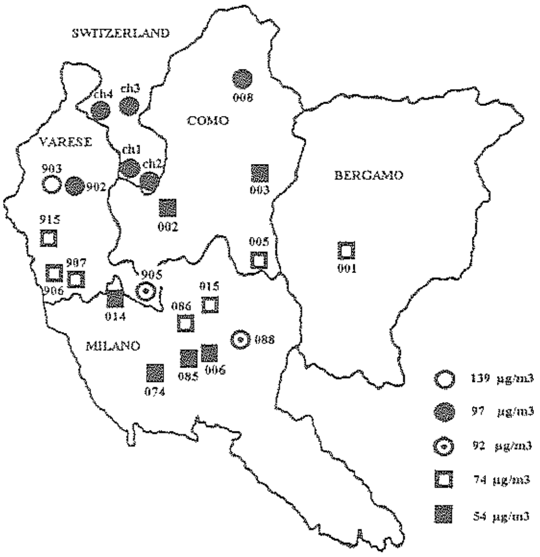

Figure 5. Cluster Analysis (Euclidean Distance): groups composition relative to hourly ozone concentrations in the weekly period 28/07/93 - 4/08/93

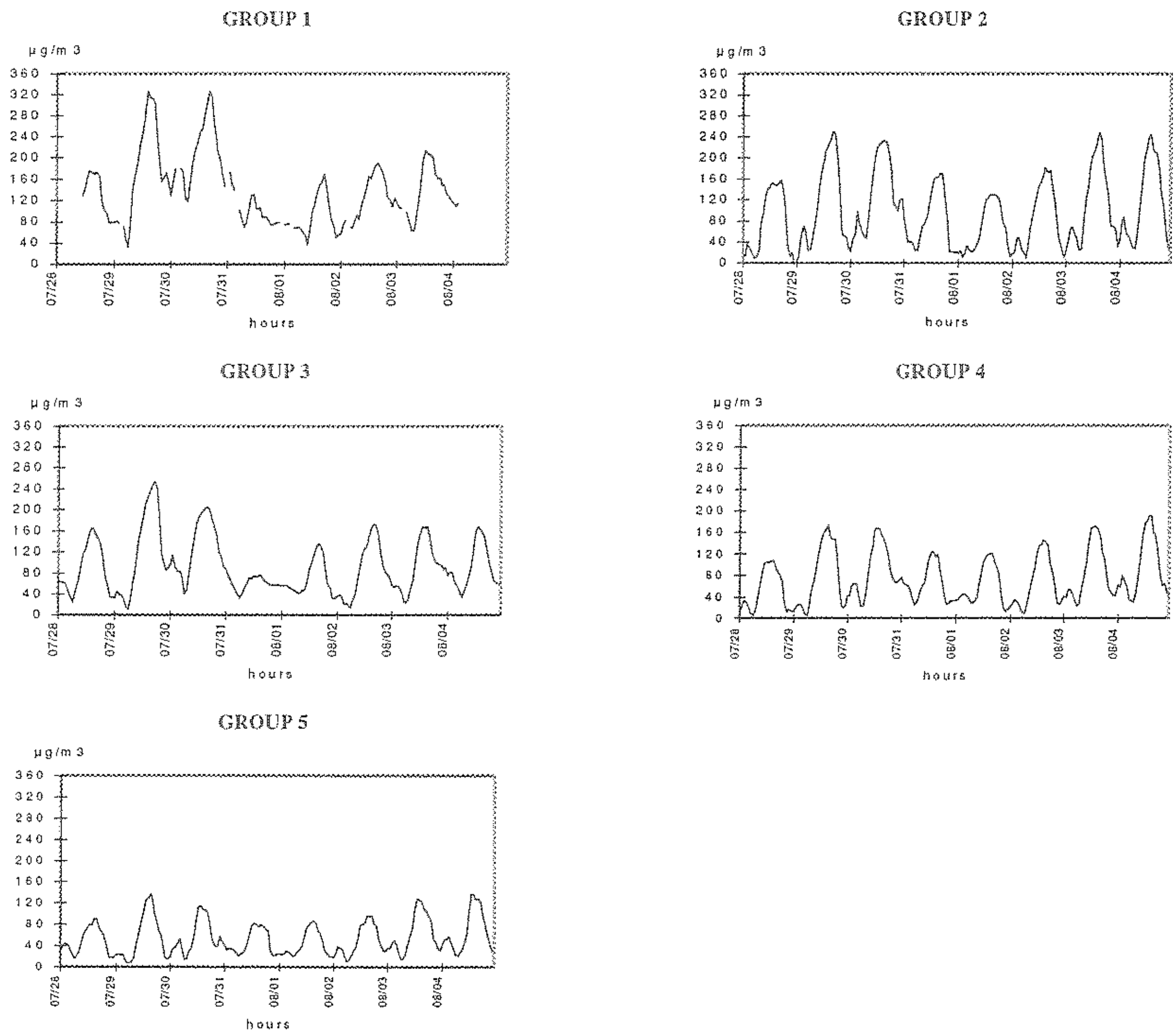

Figure 6. Cluster Analysis (Euclidean Distance): groups centroids relative to hourly ozone concentrations in the weekly period 28/07/93 - 4/08/93

Figure 7. Cluster Analysis (Correlation Factor): groups composition relative to temporal ozone concentrazions trends in the weekly period 28/07/93 - 4/08/93

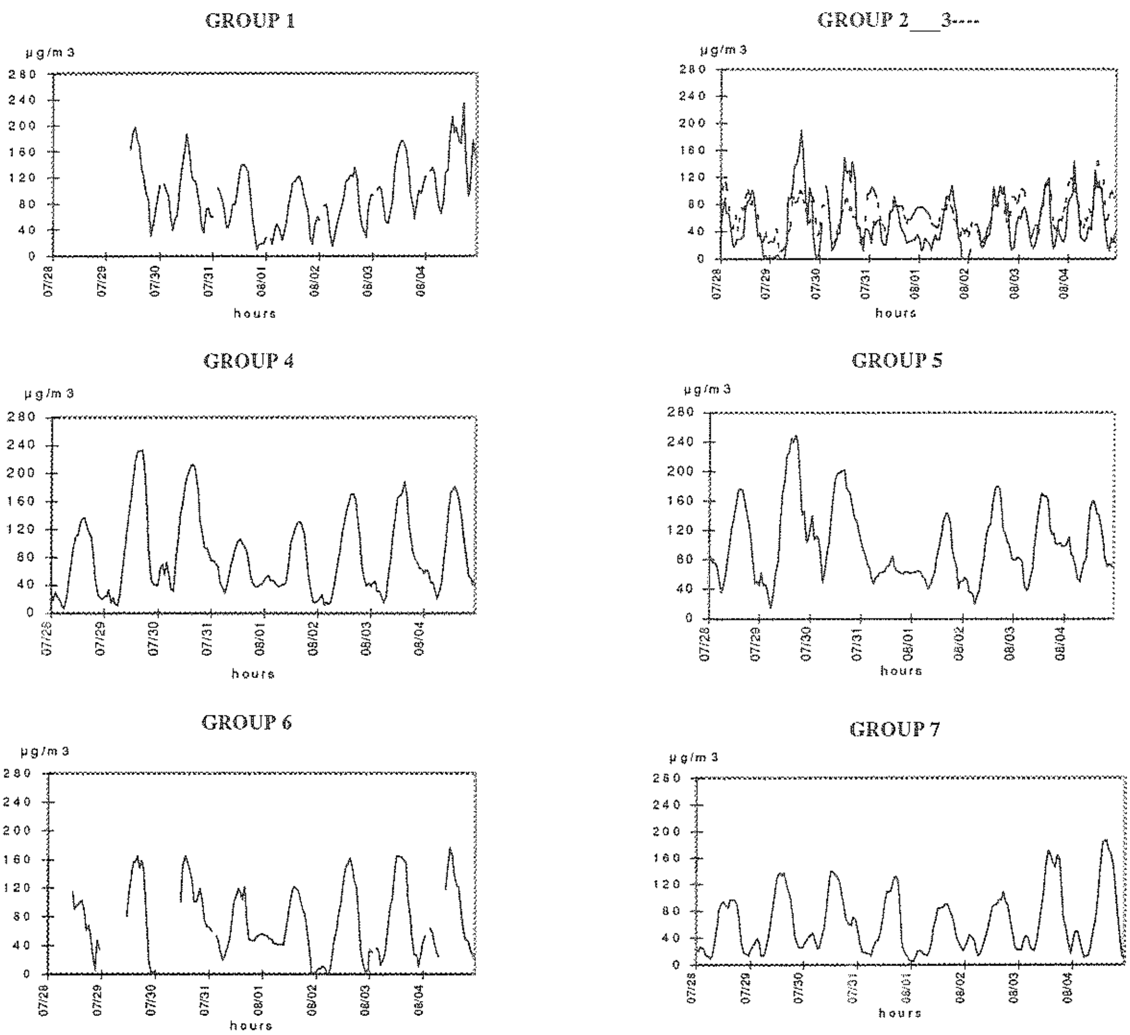

Figure 8. Cluster Analysis (Correlation Factor): groups centroids relative to temporal ozone concentrazions trends in the weekly period 28/07/93 - 4/08/93