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# Evolution of Process Coefficient of the Sulphur Dioxide in Urban Zone

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We have selectioned the sulphur dioxide, between other urban pollutions for its great importance in the high imision level measured, as for its strong and harmful effect. We have calculated the process coefficient by using of a revelant algorithm and analysed its behavior for strong and light issue of pollution. The spectral analysis is used for detecting some periodicity. The dates used are horary record of sulphur dioxide and rainfall during three years.

V práci je studován $SO_2$ jak pro jeho význam, tak i naměřenou vysokou hladinu imise i pro škodlivý vliv. Byl vypočten procesní koeficient použitím vhodného algoritmu a analýzou jeho chování pro případ silného a slabého znečištění. Spektrální analýzou byla odkryta určitá periodicita pozorovaného jevu. Použitá data představují hodinové záznamy množství $SO_2$ a vodních srážek v průběhu tří let.

Нами подобраный диоксид серы между другими городскими загрязениями имеет большее значение изза высокого уровня имиссии и также изза сильного загразнеющегося эфекта. Расчитан коэфициент процесса при помощи релевантных алгоритмов и проведен анализ его поведения для сильного и слабого потока загрязнения. Спектральный анализ применяется для детекции некоторой периодичности. Даты применяемые в работе из часовой записы диоксида серы и из спада воды в течении трех лет.

## 1. Introduction

Among the many and complicated processes that can take place in the atmosphere and can have an effect on the air pollution, those which have a direct influence on the removal or on the decrease of the concentration are the most important. These processes are generally known as processes of contaminant removal. In these processes we can distinguish between those carried out without the participation of rainfall as a removal factor called DRY removal processes, and those that have the rainfall as the main meteorological factor, named WET removal processes. We must take in mind that what seems a clear and emphatic division in the removal processes it is not so, because the predominant meteorological factor coexists with others although the process name is bound to this factor. The injection of contaminants in the atmosphere is due to two kinds of phenomens, the natural type (volcanic eruptions and others) and those of the antropogenic origin as result of the different human activities which range from the production of energy to the obtaining of a great number of product, for the food industry, parachemistry.

Basically since 1950 to nowadays a great number of scientifics have been concerned with the processes of contaminants removal. Meethan in 1950 carried out the calculation of the residence time of sulphur dioxide in the atmosphere and the measurement of particles as an effect of the rainfall by analysing the rain. Also are important the Engelman's studies (1965) about the rainout and washout coeficient. Makhon'ko (1967) proposed to take into account the following variables for calculating the fraction of contaminant removal by precipitation or rainfall, minimum height of the cloud, intensity of precipitation, contaminant concentration at the beginning of precipitation and duration of it. In 1970 Rhode obtained the residence time of Anthropogenic Sulphur Dioxide. Also in 1970 Moller and Shuman carried out an important study about the mechanisms of the transport of contaminant from the atmosphere to the earth's surface. In 1975, Dana and his colaborators analysed in their study the aerosols removal by precipitation. Davies T. D. (1976) forms his studies evaluating the importance of precipitation as a factor of contaminant removal in the atmosphere. Slinn (1978), Maul (1978), H. Michael Davenport (1978), B. C. Scott (1978), Henning Rodhe (1980), and others have carried out important studies for the knowledge of the removal processes in the last decade.

## 2. Description of the studied zone

This paper has been carried out in the city of Madrid and more specifically in the Plaza de Castilla zone. Madrid is situated between 40°80′ North latititude, on a irregular plateau of 19 × 18 square kilometres and a medium height above the Mediterranean sea level of 650 m. The southern part of the city is limitated by an industrial zone and the northern part by residential suburbs that spread towards Sierra de Guadarrama which constitutes the orographic limit. The prevailing winds are the South West which blow away contaminants from the industrial zone to the lowest parts of the city and towards zones of the negative divergence (M. A. Pascual and E. Hernández, 1981). On the other hand the height of the inversion layer varies between 500 m and 1000 m. These factors added to a slow, dense traffic cause pollution levels above the permissible levels in specific months of December and of January fundamentally.

In a city like Madrid, where atmospheric pollution is fundamentally due to heat generators and traffic, the Sulphur Dioxide contaminant is of great importance for the high imision level measured as the harmful effect of this contaminant on the environment (people, animals and buildings) (Drufuca G. and Giuliano M., 1977). If we have in mind the acid rain (Nitric acid and sulphur Dioxide dissolved in rain water) in 1980 the sulphur emision was 500.000 t in Spain what gives us the magnitude and importance of the sulphur dioxide contaminant chosen as subject of this paper on removal. The data used in Sulphur Dioxide concentration imision has been given to us by the Ayuntamiento (Council) of Madrid, taken from the automatic pollution control. This sampling corresponds to the years 1979, 1980, 1981 and around Plaza de Castilla as it is said before and with an interval of $\frac{1}{2}$ hour, which amounts to 52.508 data. Because we didn't have statistic programs to handle such a number of data, the sampling time increased to 1 hour, 26.304 data, what enabled to carry out calculations per months (less than 800 samples) in programs that admited a maximum of 1000 samples. Besides these data, the precipitation data have been handled, corresponding to the same zone during the same period of time with variable intervals of sampling according to the duration of the precipitation rainfall.

In order to check the meteorological information given by the automatic network control, data have been used from the O. M. Retiro that correspond to the periods on which there was some precipitation during these 3 years.

## 3. Process Coefficient

The atmosphere, in the processes of changing and dischanging of contaminants, responds with accuracy to the behavior that in specific conditions present physics systems already known, as that of the capacitor in the charging and discharging process through a serie resistor or the energy lost by an oscillator, which is left free after submitting it to the effect of a force.

In the two physical systems mentioned above, the resistence in the first system and the constant of the oscillator in the second are to condition the speed of increase and loss of charge and energy respectively, and their physical effect could be compared to that of "The resistence to the change transference in the atmosphere (V. Platt, 1978)"; the mathematic study of these systems evolution allow us to define a coefficient known as loosening time, that results a function of the system physical characteristics and the inverse of which represents the speed of change or loss of the energy. This coefficient can be used as typical of the process.

In the change and dischange processes of contaminants in the atmosphere, similitude is defined by a coefficient to which units $t^{-1}$ are given, and which is going to be the inversion of relaxation time of the atmospheric system in the removal processes called PROCESS COEFFICIENT.

The mathematic pattern will be:

$$\frac{dc}{dt} = -\lambda c ,$$

where $c$ = contaminant concentration
$t$ = sampling time
$\lambda$ = process coefficient.

The integration of the differential equation between two moments $t_i$ and $t_{i+1}$ in which concentration is respectively $c_i$ and $c_{i+1}$, will give the calculation algoritm for the Process Coefficient.

$$\int_{c_i}^{c_{i+1}} \frac{dc}{c} = - \int_{t_i}^{t_{i+1}} \lambda \, dt$$

Supposing $\lambda$ constant in the interval $t_{i+1} - t_i$, after integration we have

$$\ln \frac{c_{i+1}}{c_i} = -\lambda(t_{i+1} - t_i)$$

and

$$\lambda = -(t_{i+1} - t_i)^{-1} \ln \frac{c_{i+1}}{c_i} .$$

If $c_{i+1} > c_i$ the contaminant concentration increases and $\lambda < 0$. In this case $\lambda$ will be called CONTRIBUTION COEFFICIENT during the time $t_{i+1} - t_i$. If $c_{i+1} < c_i$ the contaminant concentration decreases and $\lambda > 0$. In this case $\lambda$ will be called REMOVAL COEFFICIENT, during the time $t_{i+1} - t_i$. If $c_{i+1} = c_i$ the contaminant concentration is constant in the time and value $\lambda = 0$ will be called BALANCE COEFFICIENT, during the time $t_{i+1} - t_i$. The $\lambda$ generic denomination will be Process Coefficient, which typifies in a global way the evolutions with regard to contaminant removal (M. A. Pascual, 1983).

## 4. Analysis of Process Coefficients Calculated

### 4.1. Annual and global analysis

The values obtained for the process coefficient are in the range $-30.475 \times \times 10^{-5}\ s^{-1}$ and $+29.480 \times 10^{-5}\ s^{-1}$, similar figure to those given by other authors like R. E. Man and others (1971), R. J. Engelman (1965).

The first series of coefficient process values studied were the corresponding to these three years aforementioned with samplings intervals of 1 hour. The histogram of relative frequencies (fig. 1) shows the likeness in a Moivre's distribution. Submitted the sample to nonparametric testing K–S, the result was the normal distribution with S = 90%. We must point out that in 43.8%, cases the process coefficient was positive, which corresponds to global situations of contaminant removal. In 38.6% cases the process coefficient was well corresponding to stationary total situations. In 17.6% cases the process coefficient was negative, corresponding to global situations of contaminant contribution. The process coefficient distributions corresponding to each of the years 1979, 1980, 1981 present Moivre's distributions which the T–S gives as normal with S = 99%'.

The coefficient analyses indicate that negative $\lambda$ are always displayed in inferior proportion to positive coefficient $\lambda$ and null, they are in proportion less than 33%

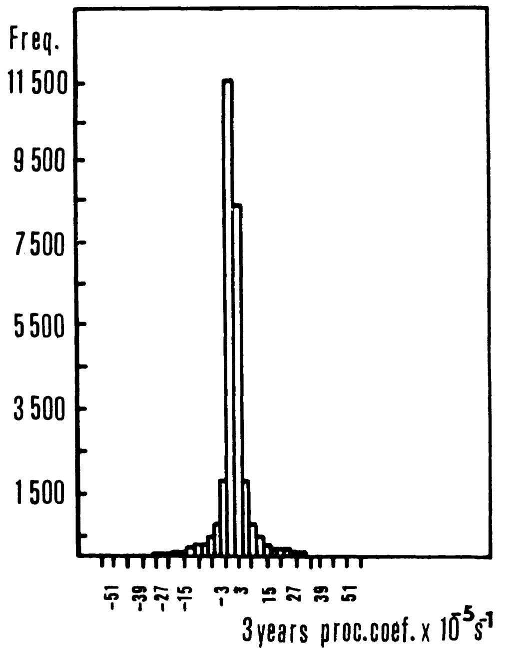


*Fig. 1.*

of null coefficient or positive coefficient in the year 1981 and above 50% in the years 1979 and 1980. With regard to the removal coefficients, these are alive among the 3 years.

#### 4.2. Analysis of non-heating periods

The analysed periods were:

1. From 01.04.1979 to 31.10.1979 – 214 days 5136 samples
2. From 01.04.1980 to 31.10.1980 – 214 days 5136 samples
3. From 01.04.1981 to 31.10.1981 – 214 days 5136 samples.

All the distributions were Moivre's and normal.

During the non-heating periods, which are spring and summer, the proportion of negative contaminants, situations of contaminant contribution are essentially smaller than the correspondent to positive coefficient, removal situations becoming even a 33% smaller for the year 1981.

### 4.3. Analysis of heating periods

The analysed periods were:

1. From 01.01.1979 to 31.03.1979 – 90 days 1160 samples
2. From 01.11.1979 to 31.03.1980 – 152 days 3648 samples
3. From 01.11.1980 to 31.03.1981 – 151 days 3624 samples
4. From 01.11.1981 to 31.12.1981 – 61 days 1464 samples.

The 1st and 4th periods are shorter due to having handled data of 3rd whole years and the 3rd period has a day less than the 2nd because 1980 was a leap year. All distribution are Moivre's and normal. The proportion of positive coefficients (removal) is slightly superior to that of the negative coefficients.

## 5. Precipitation

The different factors that take part in the contaminant removal by precipitation make presented phenomena very difficult to evaluate. The two most characteristic process in removal by precipitation are the RAINOUT process and the WASHOUT. In these specific cases the process coefficient will be called WASHOUT COEFFICIENT and represents the fraction of contaminant removed from the atmosphere by the effect of rainfall. As the washout processes constitute a specific case within the general processes, the algorithm used for the general cases can be applied here to obtain the washout coefficient. The washout coefficient obtained has been used for all the precipitation periods in these three years of research. The table I shows the global data of rainfall together with some more information to evaluate it. Data has been checked with other taken in the Retiro O.M.

*Table 1.* Rainfall Castilla circus and Retiro

| Month-day | Rainfall Retiro | Total (mm) Castilla C. | Type of clouds | Incidence |
|---|---|---|---|---|
| 01-18 | 35 | 41 | Ns | |
| 19 | 18.9 | 29 | Ns | |
| 21 | 8.8 | 23 | Sc-C | |
| 22 | 2.4 | 4 | Sc-C | |
| 23 | 0.1 | NO | Sc-C | |
| 26 | 7.2 | 8 | Sc-C | |
| 27 | 24.8 | 27 | Sc-C | |
| 28 | 1.8 | 7 | Sc-C | |
| 29 | 0.1 | NO | Sc-C | |
| 02-01 | 1.5 | 1 | Sc-C | |
| 02 | 10.1 | 14 | St-C | |

Table 1. (Continued)

| Month-day | Rainfall Retiro | Total (mm) Castilla C | Type of clouds | Incidence |
|---|---|---|---|---|
| 04 | 1.8 | 3 | Sc-C | |
| 05 | 0.3 | NO | Sc-C | |
| 06 | 1.1 | 1 | Sc | |
| 07 | 1.4 | 3 | Sc | |
| 08 | 0.9 | 2 | Sc | |
| 09 | 9 | 9 | Sc | |
| 10 | 22.6 | 29 | Sc | |
| 11 | 13.9 | 15 | Sc | |
| 12 | 6 | 9 | Sc | |
| 13 | 5.5 | 2 | Sc | |
| 14 | 5.9 | 6 | Sc | |
| 16 | inap. | NO | | |
| 21 | 0.1 | NO | Ns | |
| 28 | 1.8 | 1 | Ns | |
| 06-08 | 1.7 | NO | Cu-CuCb | |
| 09 | 0.8 | 1 | Ac-Ab | |
| 27 | 1.1 | 10 | Cu-CuCb | |
| 30 | 0.4 | NO | Cu | |
| 07-01 | 10.7 | 14 | Cu-Cb | Strong storm |
| 11 | 2.3 | 3 | Ac-As | |
| 16 | inap. | NO | | |
| 19 | 29.8 | 10 | Cu-GCu | Torrential rainfall |
| 08-31 | 1.0 | NO | Cu | |
| 10-04 | 1.9 | 1 | Ac-Ns | Could |
| 05 | inap. | NO | Cu-Sc | Could |
| 06 | 20.5 | 21 | Cu-Sc | |
| 07 | 0.4 | NO | Sc-Cu | |
| 10-08 | 0.4 | 1 | Sc-Cu | |
| 09 | 13.4 | 19 | Sc-Cu | |
| 10 | 0.2 | 2 | Sc-Cu | |
| 11 | 0.8 | NO | Sc-Cu | |
| 12 | 2.3 | 10 | Sc-Cu | |
| 13 | 11.8 | 9 | Sc-Cu | |
| 14 | 8.5 | 13 | Sc-Cu | |
| 15 | 5.2 | 12 | Cu | |
| 16 | 0.2 | 1 | Sc | |
| 19 | inap. | NO | | |
| 20 | inap. | NO | | |
| 21 | 13 | 9 | Cu | |
| 23 | 0.3 | NO | Sc | Smog |
| 25 | 4.5 | 4 | Sc | |
| 26 | 7.9 | 7 | Sc-Cu | Hail at 0 : 30 |

## 5.1. Analysis of WASHOUT COEFFICIENT

The results of analysing the washout coefficient are shown in table 2. The proportion of positive washout coefficient is larger than the negative, being the former a 15% superior to those obtained for the total population in 3 years. It should be motived too, the strong relation between the global quantity of rainfall and the proportion of positive coefficients. Although the determination of the process coefficient has only been established for the precipitations cases, there is a proportion of "washout" coefficient with negative value, that is to say, situations of contaminant contribution. When these cases have been specifically observed, we have noticed that most of them happen in 1 or 2 hours around the daily maximum relative of sulphur dioxide.

*Table 2.* Distribution of process coefficient for rainfall during three years.

| Year | Washout C. $< 0$ | Washout C. $= 0$ | Washout C. $> 0$ | Total Rainfall in mm |
|---|---|---|---|---|
| 1979 | 19.6% | 20.1% | 59.3% | 502 |
| 1980 | 35.8% | 14,6% | 49.6% | 357 |
| 1981 | 28.4% | 16.6% | 55% | 525 |

Why this phenomenon happens is explained by the calculation algorithm, who on seeing measures of relative contaminants, doesn't notice the small increase of concentration by effect of rainfall. In these cases, the washout can be obtained like the area between the curves that represent the media concentrations between the times $t_i$ and $t_{i+1}$.

The other negative coefficients which are out of the surroundings of the contaminant maxima can be ascribed to contaminant depression phenomena in the rainfall zone; such depression causes a migration of contaminant towards the rainfall zone. This phenomenon can also be explained by the contaminant desertion process, on which a falling raindrop, goes through a clear zone.

## 5.2. Relationship between the WASHOUT COEFFICIENT and rainfall

Relations of polynomic types have been studied, but none can be established as a whole in a satisfactory way. Different authors propose relations which range from the direct line to the negative powers. (Henning Rodhe – 1972) (R. J. Engelmann – 1965). The relation found by us is:

$$\lambda = bR^{-0.4}s^{-1}$$

$b = 0.87 \times 10^{-5}$ is a constant and $R$ = rainfall in mm .

This expression has been obtained for those facts of precipitation with positive washout. Engelman uses for the exponent a value slightly higher – 0.7, although his zone studied presents characteristics rather different. The relation obtained shows that strong rains produce a small washout and that light rains cause a larger effect of contaminant removal. The cause seems to be that the sulphur dioxide has to spread itself in the dragging zone and in the precipitation water very swiftly. The residence time of sulphur dioxide in the atmosphere, calculated as $1/\lambda$ proved to vary from 1 h. to 25 hours. As it was hoped, there is a clear decrease of the medium value with regard to that obtained in general conditions, which ranges from 1/2 h. to 15 days.

### 6. Spectral treatment

The values obtained for the coefficient process constitute a temporal series with sampling interval of 1 h., so that a isophorphism can be established between the numeric position that a certain coefficient occupies and the time passed since the beginning of the sampling. As a starting hypothesis we consider that the process is stationary. The power spectrum can be related to that of autocovariance function, through that transformed by Fourier. The knowledge of autocovariance function of a process is equivalent to know the power spectrum.

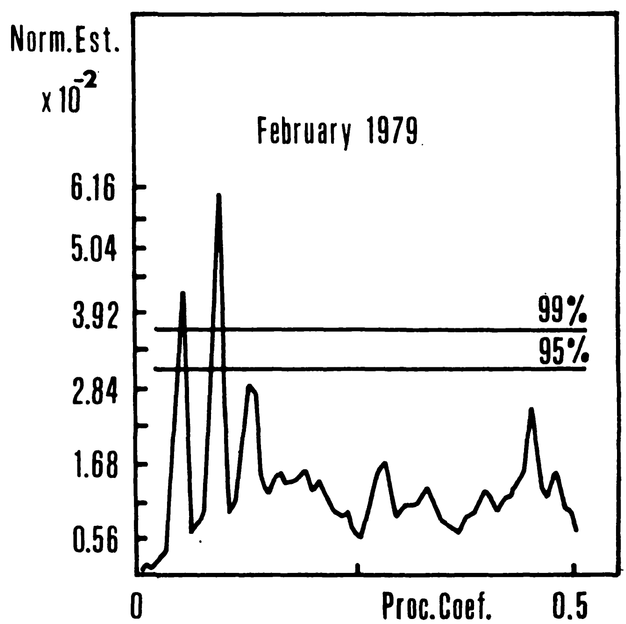


*Fig. 2.*

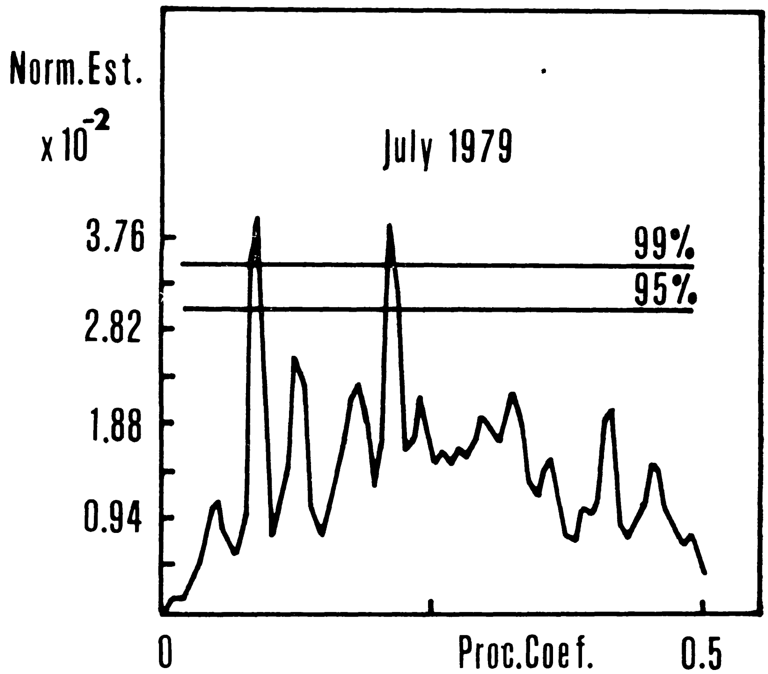
Norm.Est.
x10^-2
July 1979
3.76
2.82
1.88
0.94
99%
95%
0
Proc.Coef.
0.5

*Fig. 3.*

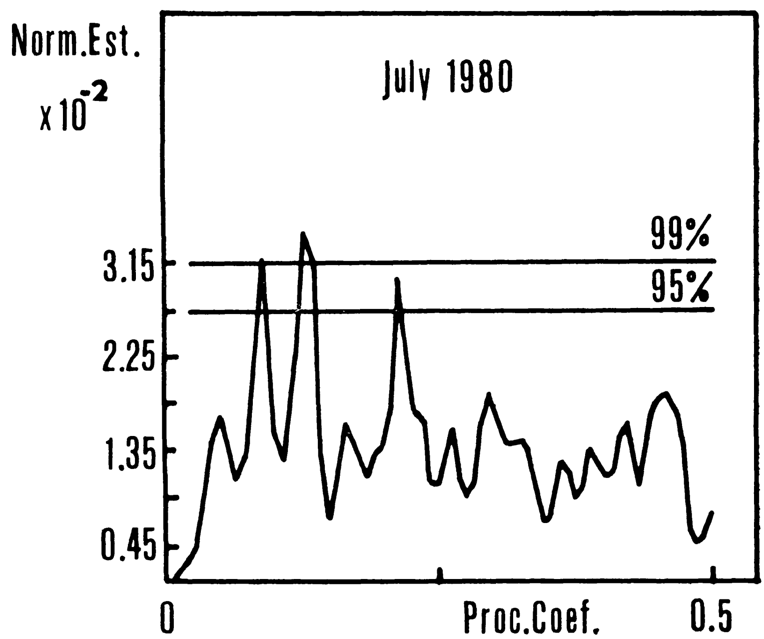
Norm.Est.
x10^-2
July 1980
3.15
2.25
1.35
0.45
99%
95%
0
Proc.Coef.
0.5

*Fig. 4.*

### 6.1. Determination of the spectrum

The spectrum of the process coefficient has been calculated by using R. B. Blackman and J. N. Tukey's method (1958). 36 power spectra have been obtained, corresponding to every month of the sampling. In fig. 2, 3, 4 and 5 some of the most significant appear. Being $\Delta t = 1$ h, Nyquist frequency will be $1/2 = 0.5$ therefore phenomenon of an inferior duration of 2 hours cannot be detected. The delay has been $u = 70$, and the spectral window used, that of Hanning's, which gives an adequate resolution and stability for the spectra. The simple visual inspection of the spectra of the coefficient process tell us that these do not correspond to a Markov's red sound. The red sound test carried out by the calculation of the autocorelation coefficient enable us to verify that above mentioned. The kind of graphic that the serie presents and the spectrum shape make us to think on a white sound, which was tested making a white sound test.

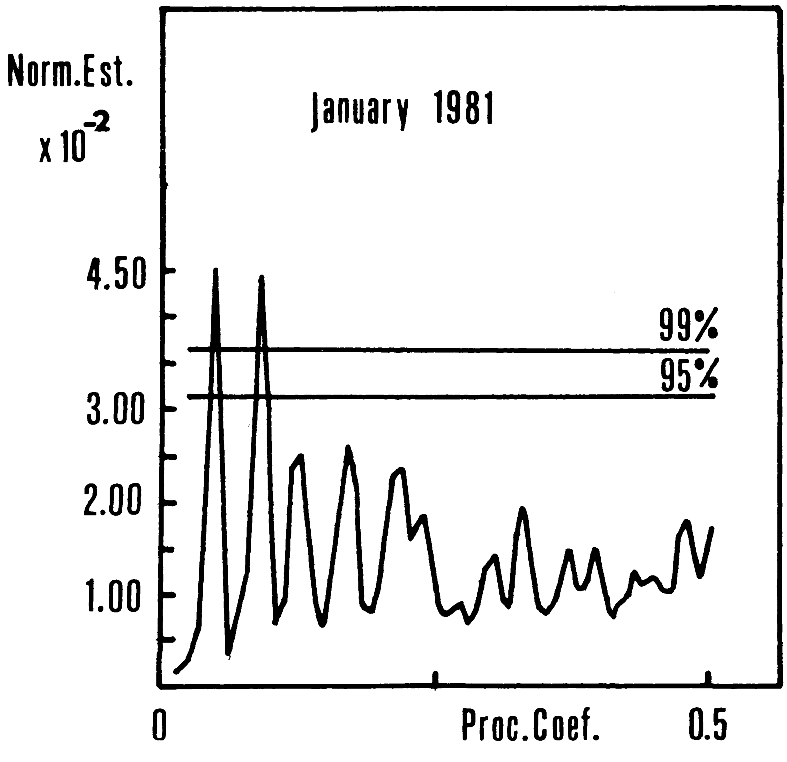


*Fig. 5.*

### 6.2. Periodicity in the spectrum of the process coefficient

Once the different spectra of the process coefficient are settled down the levels of null continuous, of 95% and 99% for each of the one spectra were drawn, in order to identify the possible periodicities that were significant. In the analyses of spectra appeared at the 95% level the following periodicities: One, which is maintained along the 3 years. Such periodicity corresponds to the frequency centred in the band between 0.086 cycles $h^{-1}$ and 0.072 cycles $h^{-1}$ (11.6 and 14) h., being its main value 12.8 h. The second periodicity observed corresponds to the band 0.25 cycles $h^{-1}$, 0.16 cycles $h^{-1}$ (4 and 6.25) h. and its estimate cental value 5 h. Periodicities of 24 hours are detected and so is a spurious periodicity due to the sampling in low frequencies during the month of January, 1981, which corresponds to the 0.029 cycles $h^{-1}$ frequency, that is 34 hours.

The periodicity of 12 h. can be explained as associated with the relative maximum of sulphur dioxide therefore they have a correspondence with the atmosphere stratification stability, which was studied by E. Yu. Bezuglaya and others (1970).

The periodicity of 5 h. is mainly linked to the summer months and could be associated with the average duration of summer storms.

And finally, the periodicity of 24 h. is linked to the winter course spectra and corresponds totally to the daytime's activity, which is strongly relationed with those months emissions.

## 7. Conclusions

By similarity with known physical systems, a calculation algorithm has been obtained to establish a coefficient, called process coefficient. Such coefficient enables us to characterize the atmosphere with regard to the evolution of the pollution (removal, contribution or stationary). The medium residence times of sulphur dioxide have been obtained, and it follows that for the genetal system of meteorological conditions it ranges from 1/2 h. to 15 days, and for rainfall situations it varies from 1 h. to 25 h. The distribution presented by the process coefficient was normal and normal were also the corresponding to the process coefficient for periods of heating and non-heating. The process coefficient was totally of removal or null, which represents the atmospher tendency towards selfcleaning or balanced situations. During the heating periods the stationary or contribution processes are more frequent, therefore the pollution level is maintained or increased in a general way. The rainfall periods break the stationary situation of the process, causing a larger number of removal situations. The cases of rainfall with negative washout are linked to these hours on which the sulphur dioxide presents a maximum, with a delay or advance of 2 h. It is proved that light and persistent rainfall produces a larger contaminant washout than the hard rainfall of short duration. Periodicities of 5, 12 and 24 hours have been detected in the temporary serie of the process coefficient.

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