MULTIFRACTALS AND NETWORK EFFECTS IN SEVESO DIOXIN POLLUTION

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ABSTRACT

In this work we analyze the Dioxin (TCDD) pollution of the Seveso (Milan, Italy) territory, using contemporaneously the measurements collected from 1976 up to 1981. First we present the mathematical framework of *Universal Multifractals,* discussing their practical importance as a new statistical parametrization of pollution intensities at different scales; we also point out the relevance of a multifractal approach in connection with the problem of toxicity and the measure of pollutant on a sparse (fractal) network. Then we apply the *Double Trace Moment (DTM)* multifractal technique in order to estimate both α (the degree of multifractality) and C_1 (the codimension of the mean field) and (using spectral analysis) we also calculate H (the degree of non-conservation of the process). Finally we discuss the problems of undersampling and network sparseness and provide a way to statistically correct for these effects. We conclude that the ground distribution of Dioxin shows clear multifractal features and can be classified as an *unconditionally hard universal multifractal process.*

1. Introduction

Many geophysical phenomena show extreme variability over a wide range of scales. This behaviour is the result of non-linear interactions between different processes at various scales and involves the appearance of complex (multi)fractal structures (see e.g. the papers in Ref. [1]). Developments in multifractals[2] provide the link between experimental observations of natural phenomena and mathematical ideas about scaling. Whereas fractals are sufficient for dealing with scale invariant *sets*, multifractals are now understood as the natural theoretical framework for scale invariant *fields*. There is now considerable evidence[1-2] that various atmospheric fields (such as rain, wind, clouds, temperature and radiation fields) are multifractals.

In this paper we analyse the Dioxin pollution of the Seveso territory which occurred on July 10th, 1976, when a chemical reactor of the Icmesa factory in Seveso (Milan, Northen Italy) exploded, spreading a large amount of Tetra-Chloro-Dibenzo-p-Dioxin (TCDD) over an area of about 8 km^2.

As is well known, Dioxin is a very stable toxic heavy molecule and the accident has led to several epidemiological studies and health countermeasures[3]. Toxicity is normally discussed in terms of *mixing ratios*; however, due to the extreme variability of the *in situ* measurements, mixing ratios are usually averaged over the scale of the detecting network. Nevertheless, the physical effects of toxicity often depend on the strictly local concentration of pollutant: areas of high concentration ("hot spots") may be invisible to the detecting network[4] (due to either insufficient *spatial* or *dimensional* resolution) but may nonetheless be highly significant from a health perspective. The advantage of multifractals is that they can characterize the detailed structure of the pollutant distribution over the entire range of scales, from the strictly local concentration up to the largest spatial average.

Our aim is to provide a description of the ground pollution in terms of multifractals: in fact, over the relevant scales, all of the non-linear mechanisms involved in dispersing the Dioxin - turbulent diffusion, fallout (scavenging, dry deposition), infiltration in the soil - are expected to be cascade processes (especially the turbulent diffusion) operating over wide ranges of scales and it is now known that cascade mechanisms generally lead to multifractal fields. The measuring network is also likely to be scaling (this can be understood since it is sparse with "holes" at all scales) and we shall see that it is fruitful to consider the density of stations itself as a *multifractal measure.*

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2. The Pollution

The collection of measurements took several years starting in 1976 and was performed during a series of sampling campaigns. In this paper we use the data collected during different campaigns, starting from 1976 soon after the accident up to 1981, for a total of 2424 measurements. A basic difference from a previous work[5] is that we now use contemporaneously all the available data, without splitting them into separate data bases corresponding to different campaigns. The justification for doing so stays in that the range of variability of measured pollution is almost the same for each campaign; hence the multifractal analyses are not affected by considering all the data at the same time.

The Dioxin distribution is extremely variable (intermittent), which is a typical multifractal "signature": in fact, the amount of pollution ranges from $\approx 10^{-1}$ μg/m^2 to $\approx 2.7 \cdot 10^4$ μg/m^2.

The locations of the sampling points form a sparse network having fractal (correlation) dimension[6] $D_C \approx 1.4$. This roughly indicates that (over the corresponding scales) the network is not *space-filling*, i.e. "holes" occur at all scales. It is important to stress that the *scaling* of the network[1] holds over a fairly large range: from $\approx$20m up to at least $\approx$2km, i.e. about two orders of magnitude; this, in turn, justifies the application of multifractal analysis techniques over the corresponding range of scales.

Several parametric descriptions[7] of the TCDD distribution have already been attempted. All of such models smoothed the original field by means of regular surfaces, washing out any extreme values ("hot spots"). Of course any description aiming to be more realistic should not disregard such values as being anomalous, especially since they contribute significantly to the mean of the process. Some of us have already proposed a (mono)fractal description[8] of the Dioxin distribution; we now want to take advantage of the more powerful opportunities offered by a *universal multifractal* approach. In fact (mono)fractals can be used, at best, to approximate a multifractal process near the mean, but will generally miss the extremes. Moreover, in the following we will take advantage of the existence of stable and attractive *universality classes* for multifractal processes: this means that many of the details of all the complex non-linear interactions will be "washed out" when they occur over a large enough number of interacting structures, ultimately only a few parameters will matter.

In the following sections we will present the mathematical framework of multifractals, improving the brief description we gave of them in a previous work[5] and extending the discussion to the network effects.

3. The Multifractal Analysis

For a multifractal field[2g-i] the following (scaling) relation holds:

$$Pr(P_\lambda \geq \lambda^\gamma) \propto \lambda^{-c(\gamma)} \qquad (1)$$

where P_λ is the field intensity (here the Dioxin concentration) at a given resolution λ, γ is the order of singularity and $c(\gamma)$ is the codimension function describing the "sparseness" of the field intensities. Eq. (1) relates the intensity of the field P_λ to its probability of occurence through the function $c(\gamma)$, which is independent of any particular resolution λ.

For *universal multifractals* $c(\gamma)$ may be written in terms of three parameters[2g-i]:

$$c(\gamma - H) = C_1 \left(\frac{\gamma}{\alpha' C_1} + \frac{1}{\alpha} \right)^{\alpha'} \quad \text{for } \alpha \neq 1 \qquad (2a)$$

$$c(\gamma - H) = C_1 \, e^{\frac{\gamma}{C_1} - 1} \quad \text{for } \alpha = 1 \qquad (2b)$$

and $\frac{1}{\alpha} + \frac{1}{\alpha'} = 1$. The parameter α is the *"degree of multifractality"* of the field, varying from α=0 (monofractal) to α=2 (Gaussian generator) and specifying the class of the probability distribution; C_1 is the *"codimension of the average field"*; H is the *"degree of non-conservation of the process"*. An

1 *Scaling* means that the average number of stations in a circle of radius R has the power law form $\langle n(R) \rangle \propto R^{D_C}$.

equivalent probabilistic description of the distributions can also be given[9] in terms of the statistical moments of order q:

$$<P_{\lambda}^{q}> \propto \lambda^{K(q)} \tag{3}$$

where K(q) is the q^{th}-moment scaling function and "<·>" stands for "ensemble average". It has also been shown[2e] that c(γ) and K(q) are related through the following "Legendre Transformations":

$$c(\gamma) = \max_{q} (q\gamma - K(q)) \tag{4a}$$

$$K(q) = \max_{\gamma} (q\gamma - c(\gamma)) \tag{4b}$$

which also establish a one-to-one correspondence between moments and order of singularities, since $c'(\gamma_q)=q$ and $K'(q_\gamma)=\gamma$. Applying the Legendre transform to eq. (2) we obtain a three-parameter expression:

$$K(q) - qH = \frac{C_1}{\alpha-1}(q^{\alpha} - q) \qquad \text{for } \alpha \neq 1 \tag{5a}$$

$$K(q) - qH = C_1\, q\, Log(q) \qquad \text{for } \alpha = 1 \tag{5b}$$

where q≥0 for α<2 and $\frac{1}{\alpha} + \frac{1}{\alpha'} = 1$. A direct estimate of c(γ) for several values of γ can be obtained using the *Probability Distribution Multiple Scaling (PDMS)* technique[2h]. Then, given c(γ), the parameters α, C_1 and H could be estimated by regressions on eqs. (2) directly. However, since α and C_1 are highly correlated, non-linear regressions would lead to poor estimates of them.

In order to obtain robust estimates of both α and C_1 (H will be discussed later) we apply the *Double Trace Moment (DTM)* technique[10]. The introduction of DTMs is straightforward. Let us define the "η-flux" $\Pi_{\tilde{\lambda}}^{(\eta)}$ of the multifractal field $P_{\tilde{\lambda}}$ (at the maximum available resolution $\tilde{\lambda}$) through boxes B_λ resolution $\lambda < \tilde{\lambda}$:

$$\Pi_{\tilde{\lambda}}^{(\eta)}(B_\lambda) = \int_{B_\lambda} P_{\tilde{\lambda}}^{\eta}\, d^D x \tag{6}$$

where D is the dimension of the observing space. Then the DTMs can be defined as:

$$Tr_{A_\lambda}(P_{\tilde{\lambda}}^{\eta})^q = <\sum_i \left[\Pi_{\tilde{\lambda}}^{(\eta)}(B_{\lambda,i})\right]^q> \approx \lambda^{K(q,\eta)-D(q-1)} \tag{7}$$

where A_λ is the region of interest covered by boxes $B_{\lambda,i}$ resolution λ and "<·>" stands for "ensemble average". The introduction of a second moment order η is non-trivial: in fact[10] the (single) moments scaling function K(q) becomes a function K(q,η) of both q and η: $K(q,\eta)=K(q\eta)-qK(\eta)$. Applying this to universal multifractals we obtain (exploiting eqs. (5)):

$$K(q,\eta) = K(q,1)\, \eta^{\alpha} = K(q)\, \eta^{\alpha} \tag{8}$$

Now, since the DTMs scale as $\lambda^{K(q,\eta)-D(q-1)}$, keeping q fixed and calculating the DTMs for different values of η and λ we may eventually estimate K(q,η) for a whole range of η. Taking logs of both sides of eq. (8) leads to a linear relation with slope α, which gives a direct estimate of it; then, using eqs. (5), we may also calculate C_1.

It is worth noting that, when moments of sufficiently high order are taken, two effects will lead to the breakdown of eq. (8). On one hand, the finite sample size poses a limit to the maximum order of singularity γ_s that can be found in the sample itself: clearly it is impossible to estimate codimensions of singularities that are never encountered! Introducing the *sampling dimension* D_s

(given by $\lambda^{D_s}=N_s$, where N_s is the number of independent realizations[2] of the process) then γ_s can be found from $c(\gamma_s)=D+Ds$. The *sampling moment* $q_s=c'(\gamma_s)$ (that is, the highest order of moment that can be reliably estimated in a finite sample[2i,10]) is $q_s=\left(\frac{D+D_s}{C_1}\right)^{\frac{1}{\alpha}}$, where D is the dimension of the observing space. On the other hand, the very singular behaviour of a process fully developed down to the smallest scales and then spatially averaged over larger-scale sets of dimension D (the "dressed properties") may prevent moments of sufficiently high order from converging. It turns out that, whenever $\alpha\geq1$, there exists a finite order of divergence q_D, given by the solution of $K(q_D)=D(q_D-1)$, above which all of the moments diverge[3]. Such violent statistics is associated with "hard" behaviour. Taking into account both undersampling and divergence of moments a precise criterion is that eq. (8) breaks down whenever $max(q\eta,q) > min(q_s,q_D)$.

4. Estimating H

In cascade processes (such as those leading to multifractals) it is convenient to isolate a conserved quantity having a basic physical significance. In terms of scaling conservation means $<P_\lambda>=constant$ (i.e. independent of λ) and hence $K(1)=0$. The energy spectrum of P_λ is of the scaling form $k^{-\beta}$, k being the frequency and β the spectral slope; moreover, the exponent for conserved multifractal processes[11] is $\beta=1-K(2)$. In our case we can easily estimate β: in fact we can calculate the Fourier Transform of the TCDD distribution, take its square modulus and calculate the (isotropic) spectrum. Finally, we need a power law filter *(fractional integration)* k^H to obtain the conserved quantity from the observed concentration, where H is calculated exploiting eqs. (5) and the above relation between β and the moments scaling function K.

5. The Effects of the Network

Up to now the structure of the network has played no role. Actually, the (typically sparse) nature of a network may significantly alter the infered statistical properties of a phenomenon measured on it. Treating the *density of stations* as a *multifractal measure* (rather than the stations themselves as a *fractal set*) it is possible to statistically correct[1,2] for such effects. Consider N_λ sampling points on a grid resolution λ; the local density of measurements can be estimated as $\rho_\lambda \approx N_\lambda \lambda^2$. The fundamental idea is to consider the measured quantities as a *product measure* $M_\lambda=\rho_\lambda P_\lambda$, where M stands for "measured" (observed) intensity and P for "true" intensity. In the i[th] grid element $B_{\lambda,i}$ the value of M_λ can be estimated as follows:

$$M_{\lambda,i} \approx \lambda^2 \sum_{(B_{\lambda,i})} P_j \approx \lambda^2 N_{\lambda,i} P_{\lambda,i} = \rho_{\lambda,i} P_{\lambda,i} \qquad (9)$$

where the sum is over all the pollution intensities measurements P_j in the i[th] box. Now, supposing statistical independence of ρ and P (i.e. the network density and the phenomenon are not "correlated"), taking the q-power and ensemble averaging, we obtain:

$$Tr_{A_\lambda} M_\lambda^q \approx \lambda^{K_M(q,\eta)-D(q-1)} \qquad (10)$$

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2 The *number N_s of independent realizations of a process* should not be confused with the *number of measurements* (e.g. of pollution) collected: here N_s=1 and hence[10] D_s=0.

3 Actually, a classification[1,2i] of universal multifractals has only recently been provided based on the value of α:
- $1\leq\alpha\leq2$: we have *unconditionally hard* multifractals, i.e. the corresponding process will show divergence of moments (also called *hard behaviour*) above a (critical) order, since q_D remains finite for all finite D;
- $\alpha<1$: we have *conditionally soft/hard multifractals* (for large enough but finite values of the dimension D of the observing space all of the moments converge, i.e. $q_D=\infty$).

with $K_M(q)=K_P(q)+K_\rho(q)$, i.e. the K's for P and ρ add (this is because they are the second characteristic functions of Log P and Log ρ). This formula is easy to generalize[12] to Double Trace Moments by summing over P_j^η in eq. (9). This yields the following DTM scaling function relations:

$$K_P(q,\eta) = K_M(q,\eta) - K_\rho(q,1) = K_M(q,\eta) - K_M(q,0) \qquad (11)$$

Such a formula expresses the corrected "true" $K_P(q,\eta)$ in terms of the measured $K_M(q,\eta)$. This correction leads, in turn, to estimates of α and C_1 not affected by the geometry of the network over which the phenomenon is measured. It is worth noting that such a technique can be applied using different values of the q-moment to improve the statistical accuracy.

The parameter H also needs corrections. In fact, using the "*" notation to indicate *conserved* quantities, we may write:

$$\lambda^{H_M} M_\lambda^* = \lambda^{H_\rho} \rho_\lambda^* \lambda^{H_P} P_\lambda^* = \lambda^{H_\rho + H_P} \rho_\lambda^* P_\lambda^* \qquad (12)$$

Hence the degree of non-conservation H for the "true" process (i.e. H_P) is simply given by the difference H_M-H_ρ, which can be easily calculated using the techniques explained in the previous section: from β_M and $K_M(2)$ we obtain H_M, while H_ρ is estimated using β_ρ and $K_\rho(2)$.

6. Analysis of the Results

A preliminary corrected-DTM analysis of the TCDD distribution shows clear multifractal features (see table 1 - refinements are expected in the near future): in fact α turns out to be ≈1.8, quite different from monofractality (α=0). Given the estimated value of α≥1, we may classify (see also footnote 3) the actual distribution of Dioxin as an *unconditionally hard universal multifractal process*. This means that the process shows "hard singularities" that cannot be tamed in any way.

The parameter C_1 has value ≈0.6, indicating a rather sparse mean-field intensity.

Fourier analysis allows us to estimate the parameter β for both the network and the data; then the corresponding values of H_ρ and H_M are readily calculated. From the present analysis (see table 1) we obtain, respectively, β_ρ≈1.2 and H_ρ≈0.4 for the network and β_M≈0.1 and H_M≈0.2 for the data; then the amount of fractional integration needed for the "true" process to become stationary is H_P≈-0.2.

# data	α	C_1	q_s
2424	≈ 1.8	≈ 0.6	≈ 1.9
D_C	β_ρ	H_ρ	H_P
≈ 1.4	≈ 1.2	≈ 0.4	≈ -0.2
	β_M	H_M	
	≈ 0.1	≈ 0.2	

Table 1: Values of all the estimated Universal Multifractals parameters (see text).

7. Conclusions and Future Perspectives

Using universal multifractal analysis we achieved two main results. On one hand, the statistical characterization of the TCDD pollution is readily accomplished by means of simple procedures (namely, DTMs and FFTs). On the other hand, we gain a deeper insight into the intermittent behaviour of the pollutant distribution at the smallest scales: the wild fluctuations are not regarded as anomalous and discarded, on the contrary they are kept as an essential feature of the phenomenon. While the former conclusion has implications for generating fast computer codes, both have important theoretical and practical consequences in environmental sciences. We have provided a simple methodology to extract the full statistics from sparse measurements, preserving their intrinsic features such as intermittency (i.e. no "a priori" regularity or smoothness hypotheses are required as in conventional objective analyses), and we have investigated the phenomenon at the highest available resolution. Furthermore, we stress that the estimates of the multifractal parameters α, C_1 and H allow us to generate (stochastic) simulations of the process. Finally, we are exploiting the mathematics of universal multifractals to create statistical procedures able to estimate the intensity of a field in the "gaps" of a (multi)fractal network. Such *"multifractal objective analysis"* could have many applications, since geophysical measurements are often collected on sparse (multi)fractal networks.

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