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HEAVY METALS IN SEWAGE SLUDGE UTILIZED IN AGRICULTURE

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Summary. - *To evaluate the fertilizing and polluting potential of sewage sludge and to establish whether the latter limits the former or not, sludge must be characterized, application rates chosen, and sludge effects monitored. This work emphasizes that chemical analysis aimed at total metal quantification cannot by itself ensure reliable answers about the agricultural value sludge. The EEC directives about heavy metals in sewage sludge provide the total content limitation of each element without taking into consideration the respective fractions of metals present in their free state constituting the chemical state most directly related to sanitary risk. To evaluate the role of the Environmental Impact Assessment (EIA) in a such disposal system, a scheme is proposed.*

Riassunto (Metalli pesanti nei fanghi utilizzati in agricoltura). - *Per valutare da un lato la potenzialità fertilizzante di un fango e dall'altro quella inquinante, e stabilire se e in che termini quest'ultima limiti la prima, è necessario caratterizzare il fango, stabilirne le dosi di impiego e controllare i suoi effetti ambientali. E' da notare però, che la sola analisi chimica mirata al contenuto totale in metalli non è sufficiente a fornire risposte provanti al fine dell'impiego agricolo di un fango, in quanto le frazioni di metalli presenti nel loro stato libero e disponibile sono quelle più direttamente legate al rischio sanitario. A tal fine, per una valutazione più completa del rischio sanitario viene proposto uno schema di Valutazione di Impatto Ambientale (VIA).*

Introduction

The enactment of Law 319 (10 May 1976) [1] has brought about an extensive and progressive diffusion of wastewater treatment plants and, consequently, an ever increasing need to dispose of sewage sludge that represents the byproduct of the treatment process itself. The pollutants removed from the liquid phase and later concentrated in sludge (whose volume is relatively reduced) represent a cumbersome mass of undesirable and potentially dangerous products requiring hygienically safe and ecologically suitable means of disposal.

Initially, a simple and convenient solution to disposal, also constituting one of the solutions proposed (in generic and vague terms) by Law 319 and its successive normatives [2, 3] by Presidential decree No. 915 of 10 September 1982 [4] and subsequent directives of 27 July 1984 [5] can be as agricultural utilization. In contrast, the problem of heavy metals has been regulated by means of a EEC directive [6]. This directive provides a total content limit for each element without taking into account the respective fractions of metals present in their free state which constitute, in fact, the chemical state most directly related to sanitary risk.

Actually, for ecological and environmental reasons, agricultural utilization of sludge is not regarded as a perfect solution to the sludge disposal problem. It is a practise that is not suitable in all cases [7, 8] and, thus, should be considered only an auxilliary part of disposal, but not a substitute to other disposal techniques. Nevertheless, the agricultural disposal of sludge may result in considerable environmental damage to soil, water (superficial and subterranean), cultivated farmland, not to mention the harm it may cause in man either indirectly through the food chain or directly through contact [9-11].

Hygienic aspects and effects on human health

Table 1 illustrates a number of pollutants according to type of contamination and possible effects on human health. Fig. 1 represents a schematic diagram of the general cycle by means of which agents responsible for contamination produce their harmful effects.

Although it is possible to reduce the level of microbiological contamination through sophisticated technological treatments, proper methods of sludge deposition and appropriate use of agricultural terrain, the risk of contamination still exists, also in relation to the survival time of sludge-derived pathogens in soil (Table 2) [12]. Therefore, disposal of sludge in soil is conditioned by limitations imposed by survival time of pathogenic microrganisms on temporary use of soil and the type of cultivation considered.

Table 1. - *Pollutants found in sludge and their potential effects on human health [12]*

Type of contamination	Agents of contamination	Effects on human health
Microbiological	Bacteria	Infections
Virus		
Protozoa		
Helminths		
Micropollutants	Elements	Acute toxicity
	Organic substances	Chronic toxicity
	Nitrates	Mutagenesis
		Cancerogenesis
		Teratogenesis

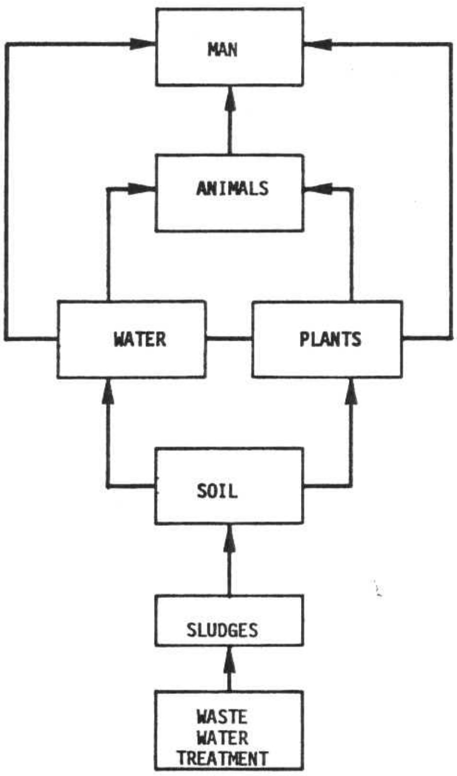

Fig. 1. - General cycle by means of which agents responsible for contamination produce their harmful effects.

A list of the principal organic substances and their frequency of occurrence is reported in Table 3 [13]. Presently, the concentration of substances are presumed not to be at danger level (except for some unforseen occurrence or event) with a consequent reduced risk to human health (one must also take into account the microflora present in soil that could further degrade these substances). With respect to heavy metals particular attention has been given to Cd [14] since, in addition to being accumulated by plants and transferred to humans by means of the food chain, it has a half-life in humans of about 25 years. Regarding studies carried out on other metals one can briefly note the following:

- Ni does not present a bioaccumulation phenomenon partly because it tends to be eliminated rapidly from the organism [15];
- Pb tends to form insoluble compounds and the radicular system acts as an ulterior barrier [13];
- Cu and Zn can cause health problems only in very high doses since both are essential elements and phytotoxic as well [16];
- Hg is present in such modest quantities in sludge that it has not been proven as a hazard [13];
- Cr is tollerable even at quite high levels and is generally absorbed by the radicular system [13].

Of course this refers to the behaviour of metals taken as singular entities, whereas a realistic evaluation of their insertion into the food chain should take into consideration the presence of more than one metal and the multiple interactions between them and the soil and plants. Eviden-

Table 2. - *Survival time of sludge-derived pathogens in soil [12]*

Pathogen	Maximum time	Average time
Bacteria	1 year	2 months
Virus	6 months	3 months
Protozoa	10 days	2 days
Helminths	7 years	2 years

Table 3. - *Major organic substances present in sludge [13]*

Compound	Frequency (%)
Toluene	94
Dichloromethane	73
Ethylbenzene	63
Benzene	61
1,2 Dichloroethylene	60
Trichloroethylene	54
Pyrene	53
Phenanthrene	53
Phenol	50
Anthracene	48
Butylphthalate	45
Butylbenzyl	43
Dichloroethane	34
Trichloroethane	24
Others	10-20

tly monitoring of sludge both before and during agricultural utilization should be done to ascertain variations in concentration of the different metals.

Composition variability

The principal sources of variability in chemical testing and sampling of sludge can be summarized as follows:

1) variation among different portions of the same mass due to differences at sampling site and between different masses due to differences in sampling time (sampling error);

2) variation among subsamples derived from the same sample (error in distribution of samples into subsamples);

3) variation in testing procedures on same subsample (testing error).

Testing and subsampling errors are not difficult to avoid or, at least, control by means of sophisticated statistical methods. To control sampling errors is more complicated. It involves choosing the most appropriate sampling procedure that takes into account the physical properties of sludge. Such a procedure can be adapted or borrowed from systems that have already been successfully tested.

A very important aspect of sampling procedure concerns the alteration of sludge with time. Numerous factors influence the metallic content of urban sludge; among these are type of "wastewater", "flowing" [14] and type of "treatment" [17, 18]. Presuming that the methodologies employed in sludge treatment are constant (though this is not always the case), the variability of sludge composition in time is prevalently ascribed to the other two factors mentioned [19].

On the other hand, the importance of "flowing" on the metallic content of urban sludge is well established as is the influence of climate [20] on the qualitative state of the metallic components and indirectly on sludge. Furthermore, a certain variability in sludge composition can be attributed to plant operating conditions [13, 18].

Table 4 indicates some measured data for metal removal in a typical treatment plant. The data reported in Table 5 refer to the variability in the concentrations of metals in urban sludge over a three year period; the variability is expressed in terms of percentage variability coefficients and refer to samples tested every year at the same period. Table 6 reports the results of a study made by the authors on sludge at a small to medium size plant; variations in the concentration of metals in sludge are indicated after a one-year interval. The percentages of elution with acetic acid (for the same sludge sample) are also listed (Table 7). The analytical conditions have been discussed elsewhere [23, 24]. An analysis of data found in the tables leads to the following observations: although heavy metals concentrations do not exceed the values established by the EEC directive, the variability of the chemical composition of sludge originating from the same treatment plant (evidenced by other researchers [25-27]) testifies to the need for a constant monitoring of sludge whenever agricultural deposition is carried out systematically and in great quantities. Furthermore, even if testing in an acid environment (acetic acid) represents an extreme form of elution, from the environmental point of view, one cannot deny that sludge, when in constant contact with environmental factors, undergoes alteration. The latter must be taken into account whenever deposition of sludge in acid terrain is done.

Table 8 lists the metal concentration in sludge established by the EEC directive. Table 9 presents the limits on heavy metal concentration provided by the EEC directive compared to those set by the Italian government. Table 10 compares maximum metal concentration in sludge, fertilizer and manure.

Table 4. - *Typical efficiency of metal removal in activated sludge [21]*

Metal	Concentration in raw sewage	Average (mg/l)	Removal (%)
Cd	0.008-0.142	0.02	20-45
Cr	0.020-0.700	0.05	40-80
Cu	0.020-3.360	0.10	0-70
Hg	0.001-0.044	0.0013	20-75
Ni	0.002-8.800	0.10	15-40
Pb	0.050-1.270	0.20	50-90
Zn	0.030-8.310	0.18	35-80

Table 5. - *Variability over a number of years of heavy metal concentrations in urban sludge, expressed in terms of coefficients of % variability (% CV)*

Metal	Cd	Cr	Cu	Ni	Pb	Zn
% CV	23	35	28	22	15	19

Table 6. - *Monthly variation of heavy metal concentrations in urban sludge expressed in terms of coefficients of % variability (% CV) [22]*

Metal	Cd	Cr	Cu	Ni	Pb	Zn
% CV	13	21	18	18	12	18

Table 7. - *Heavy metal concentrations obtained by elution test with acetic acid 0.5 M expressed in terms of coefficients of % variability (% CV) [22]*

Metal	Cd	Cr	Cu	Ni	Pb	Zn
% CV	8	1	4	18	4	27

Table 8. - *Maximum concentration of heavy metals in a few European countries compared with the EEC directive [22]*

	Cd	Cu	Ni	Pb	Zn	Hg	Cr
Belgium	10	500	100	300	2000	10	500
France	20	1000	200	800	3000	10	1000
Germany	20	1200	200	1200	3000	25	1200
Switzerland	30	1000	200	1000	1000	10	1000
Italy	10	600	200	500	2500	10	500
The Netherlands	5	600	100	500	2000	5	500
EEC	40	1750	400	1200	4000	25	-

Table 9. - *Heavy metal concentration in compost, sludges and soil after disposal, compared with average concentration of heavy metals in sludges originated from Italian municipal plants [22]*

	Maximum concentration in soil after disposal		Maximum concentration		Average concentration in sludge in a few Italian municipal plants
	compost	sludge	compost	sludge	
Cd	3	3	10	40	4.7
Cu	100	140	600	1750	351
Ni	50	75	200	400	59
Pb	100	300	500	1200	293
Zn	300	300	2500	4000	2199
Hg	2	1.5	10	25	2.3
Cr	50	-	500	-	161

Table 10. - *Heavy metal concentration provided in sludge compared with average concentration of heavy metals found in fertilizers and manures [22]*

	Maximum concentration in sludge	Range concentration in fertilizer	Avarage concentration in manure
Cd	40	0.1-9.3	1
Cu	1750	1-138	62
Ni	400	2.1-43	29
Pb	1200	0-48.7	16
Zn	4000	1-566	71
Hg	25	-	-
Cr	-	0.8-178	56

Principles and techniques in the agricultural utilization of sludge

Careful evaluation of the different environmental effects, desirable or not, of the agricultural usage of sludge, can be made by considering a series of essential parameters:

- the treatment plant features from which sludge originates;
- characteristics of sludge;
- characteristics of terrain;
- type of cultivated land;
- mode of sludge deposition;
- period of deposition of sludge.

This evaluation must be made parallel to an experimental phase articulated according to the scheme presented in Fig. 2. For a correct approach it is necessary first of all to evaluate the various effects of the operative system and second to plot the possible measures in order to minimize them. From this standpoint the possible measures can be grouped into three categories:

1) prohibition of use;
2) criteria and mode of use;
3) instruments of management and control.

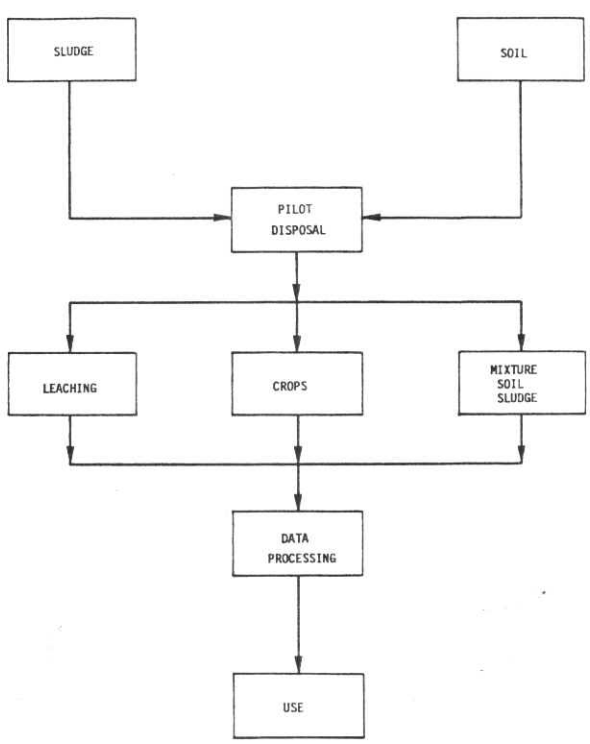

Fig. 2. - Analytical and experimental verifications for evaluating the suitability of use.

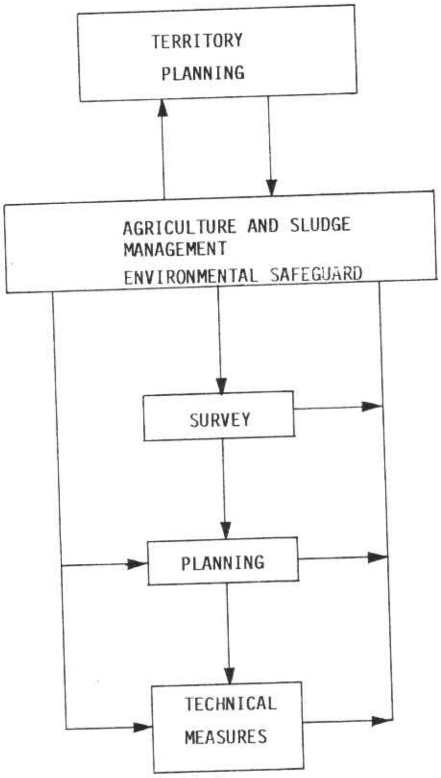

Fig. 3. - Strategy for agricultural utilization of sludge.

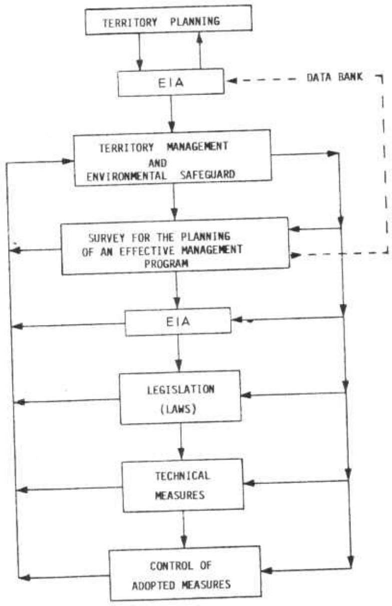

Fig. 4. - Role of the EIA in the political management of agricultural utilization of sludges.

Therefore, a generalized strategy regarding agricultural utilization of sludge involves three fundamental stages:

1) a survey of the problem;

2) definition of political safeguards;

3) the program of technical procedures to be adopted.

Fig. 3 illustrates a general methodological scheme including all necessary criteria for the implementation of such a strategy. In the development of environmental policy, EIA [28-30] plays an essential role of pivot point between the planning (land use) and technical operative stages.

Furthermore, within the framework of strategies regarding balance point territory management and environmental protection, EIA is the necessary tool for calibration of technical action to be adopted, as shown in Fig. 4.

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