

WASTEWATER AND SLUDGE REUSE MANAGEMENT IN AGRICULTURE

Ioannis K. Kalavrouziotis, Prodromos Koukoulakis

School of Science and Technology, Hellenic Open University, Patras, Greece

Corresponding author E.mail: ikalabro@eap.gr

Abstract

Huge quantities of treated wastewater (TMWW) and biosolids (sludge) are produced every day all over the world, which exert a strong pressure on the environment. An important question that is raised is “what to do with them?”. An effort is put by the scientific community to eliminate the concept of “waste” and to replace it with the concept of “recycling of resources”, by means of effective management, which does not concern only the users, but all the other groups involved in the problem, such as facility administrators, operations, politicians, scientific community and the general population. Sludge concentration data showed that there exist 516 chemicals in biosolids which create a serious health risk. It is pointed out that this risk will be greatly exacerbated by chemical toxins present in the sludge which can predispose skin to infection by pathogens. Consequently, the need for science-based policies are necessary to effectively protect public health. The risk assessment due to sludge, is difficult to evaluate due to the large number of unknown interactions involved. People living near the sludge application sites may suffer from such abnormalities as: eye, nose, and throat irritation, gastrointestinal abnormalities, as nausea, vomiting, diarrhea, including cough, difficulty in breathing, sinus congestion, skin infection and sores. Many problems seem to be related to biosolid and wastewater application in agriculture, which should be solved. A universal one, acknowledged as an “international health crisis” is the resistance of pathogens to antibiotics and to the evolution of multidrug resistance of bacteria”. Certain anthropogenically created environments have been identified as major sources of multidrug resistance bacteria such as in water treatment plants, concentrated animal feeding operations etc. All these, and many other health problems, render the safety of sludge and biosolid and wastewater agricultural reuse, for the time being questionable even though the application is done according to official guidelines. It is therefore necessary that more research work be conducted on the short and long term application effects of sludge on human health, and on the environment so as to successfully address these problems. Also, the existing guidelines must be reconsidered on the basis of the research findings to be attained. It is only then that the application of these inputs to land could be as safe as possible.

Introduction

Huge quantities of treated municipal wastewater (TMWW) and biosolids (sludge) are produced every day all over the world, which exert a strong pressure on the environment. An important question that is raised is “what to do with them?”.

As the natural resources are being depleted, the demand for clean potable water increases, the irrigation water needs also increase and the climate is warming, as a result the management of wastewater in this context, is becoming a challenging situation. Both developed, and developing countries are struggling to effectively reuse wastewater and especially the treated biosolids (sludge).

An effort is put by the scientific community to eliminate the concept of “waste” and to replace it with the concept of “recycling of resources”, by means of effective and modern management, which includes issues of converting for example the biosolids into useful products, such as in organic fertilizers, building materials, production of biogas (UN-HABITAT, 2008) etc.

On the other hand, the wastewater, and biosolids reuse in agriculture, is an attractive option, which contributes to the relative release of the exerted pressure on the ecosystem, thus offering a partial solution to the problem, which is faced by many countries, but at the same time may create other problems which must be effectively addressed. Yet, as the production of wastewater and biosolids increases, especially in the developing countries due to the increase of production and to the changing habits of contemporary human beings, who spent more water for various uses, and consumes more food of plant and animal origin, the problem of disposal of the above byproducts is becoming more complex and its solution more urgent. Consequently, the management of these wastewater treatment byproducts must concern not only the users, but all the other groups involved in the problem, such as facility administrators, operators, politicians, scientific community and the general population.

Effects of TMWW and biosolids on soils and plants

In recent years the scientific community has shown a great concern about the effects of TMWW and biosolids on the soil quality, and plant growth. (Kalavrouziotis and Koukoulakis, 2009a; NRC, 1996). Numerous experiments have been conducted aiming at the study of these effects on various types of soil and plant species. The results of this work has indicated that both short term and long term reuse of TMWW, and especially of biosolids may lead to the accumulation of heavy metals in the soil and plants. However, an important issue that is connected with the reuse, is that of safety, as a number of problems related to health risk effect are directly associated with the reuse of both TMWW and of biosolids. Harisson et al. (2006) reported that sludge concentration data showed that there exist 516 chemicals in biosolids. These workers identify various areas where more research is needed on the level of fate, and toxicity of chemicals to “human and non human receptors”. Such information is of vital importance for the biosolid application to be considered safe. It is obvious that the issue of TMWW

and biosolid safety is very important for human health. The biosolid reuse is affected by various factors, which may influence the degree of safety. An important factor related to safety is the metal plant uptake. McBride (2003) reported that the uptake varies with the metal type and plant species. He underlines in relation to the role of biosolids in plant uptake that “several critical generalizing assumptions about the behavior of sludge-borne metals in soil crop-system included into the EPA (Environmental Protection Agency) risk assessment for metals, tend to underestimate the risks and do not seem to be justified by the published research”. These authors advocate the need for a more science-based precautionary approach to toxic metal additions to soils. Lewis and Gattie (2002) reviewed and assessed the decrease of risk from known pathogens in land applied sewage sludge. They point out that this risk will be greatly exacerbated by chemical toxins present in the sludge. They state that the chemical irritants can predispose skin to infection, and they emphasize on the need of science based policies which can effectively protect public from such pathogen interactions, which must be taken into account in relation to protecting the human health.

Efromymson et al. (1998) outlined the complexity of the ecological interactions involved in relation to the ecological risk due to toxic contaminants and to excess of N in sewage sludge. They concluded that a definite risk assessment is difficult to accomplish as there are too many unknown interactions and many uncertainties. Therefore, the application of sludge to these environments cannot be considered safe. According to the above workers the accurate risk assessment will require the understanding of short and long risk term sludge application effects on soil microorganisms and, plant and wild life. Another important problem has been reported by Ghosh (2005), related to sludge application. According to it, while the bacterial levels 13 days after application decrease to control levels, *Staphylococcus aureus* increases to its highest level after 13days from application.

The appearance of symptoms reported by Lowman et al. 2013 in the people living near the sludge application sites, such as: of eye, nose, and throat irritation, gastrointestinal abnormalities, as nausea, vomiting, diarrhea, also cough, difficulty in breathing, sinus congestion, skin infection and sores, (Khuder et al., 2007), have been attributed to sludge application and more specifically to the influx of *staphylococcus* during the “post application period” of sludge. Similar results have been reported by Lewis et al. (2002a) and Harrison, and Summer (2005) stating that there have been numerous reports in the literature documenting health incidents in people living around the sites of sludge application. They point out that this situation exists even in sites where the sludge application was done in “compliance with the regulations”. They conclude that “compliance with the regulations does not ensure protection of public health”, and they suggest reconsideration of the regulations controlling sludge application.

An important problem, related to biosolid application in agriculture is the resistance of pathogens to antibiotics and to the evolution of multidrug resistance of bacteria, which is universally recognized and acknowledged as an “international health crisis”. It has been reported in this respect that certain anthropogenically

created environments have been identified as major sources of multidrug resistance bacteria (water treatment plants, concentrated animal feeding operations etc.).

Snyder, (2005) reported that “serious illness, including deaths and adverse environmental impacts have been linked to land application of sewage sludge”.

The US clean Act defines sewage sludge as pollutant. As the wastewater contains many contaminants, during the treatment process all these contaminants concentrate in the sewage sludge. On the other hand the US National Academy of Sciences (NAS) has warned that sewage sludge being a complex and unpredictable mix of biological and chemical wastes, its risk when used for agricultural purposes, cannot be assessed. Therefore, the NAS panel concluded that “standard strategies to manage the risk of land application do not protect public health (NRC, 2002).

Based on the above and on many other relevant publications, which appeared in the literature, the so called “safety” of sludge and biosolid and wastewater reuse seems to be for the time being partly accomplished. It is therefore necessary that the research work conducted towards addressing effectively all the barriers involved and also existing guidelines must be reconsidered in the light of the research result to be attained.

The properties of treated wastewater and biosolids

In order to achieve safe reuse, it is necessary to acquire a deep insight of the TMWW and biosolids (sludge) properties. Both of these inputs are characterized by negative and positive characteristics. The wastewater and the sludge contain macro, micronutrients and organic matter. However, sludge and biosolids have a much higher concentration of plant nutrients and organic matter. Especially they are very rich in heavy metals. Also, both of them contain toxic organic compounds, pharmaceuticals, and xenobiotics, at the same time they are carriers of microbes, and pathogens, helminth spores and viruses, fungi, bacteria etc. In fact, it is this composition that creates a high health risk and makes dangerous their reuse and strongly problematic. On the other hand the TMWW and the biosolids or sludge may be very useful in supplying plants with macro and micronutrients and additionally the soil with organic matter. The existence of pharmaceuticals, also creates serious universal problems related to the increase of some microorganisms such as *Acinetobacter spp* (Zhang et al., 2009) and the resistance to antibiotics, a problem related to human health risk. It has been reported that the wastewater treatment plants and biosolids concentrate antibiotics and pathogens from hospitals and industry etc. They state that “the activated sludge process probably provides a favorable environment with high microbial biomass and selective process for horizontal gene transfer of antibiotic resistant genes” (Czekalski Nadine et al., 2012

Elemental interactions and their contribution to soils and plants

Significant interactions between heavy metals, plant macro and micronutrients as

well as soil physical and chemical properties, are taking place in soil and plants (Kalavrouziotis and Koukoulakis, 2012).

Interactions between elements occurring in soils and plants have been reported long ago by Olsen (1972), Robson and Pitman (1983), and later on by Marschner (2002) and Bollan et al. (2005). Originally, the work on the interactions was focused on the “balanced supply” of nutrients in crops (Shear et al., 1946), which was considered a “sinus non quam” condition for optimum plant growth, and maximum yield. This concept showed that the interactions are related to maximum plant yield, as they play an important role in plant growth. In fact, the interactions according to Frey et al., (2010) affect the components of the rhizosphere microbial population, such as the ammonia oxidizing bacteria (AOD).

Hundreds of interactions are occurring in plants and soils. For example about 362 interactions took place in cabbage plant and in its different organs (roots, leaves, heads and stems). More specifically, the percent distribution in terms of these interactions, were as follows: antagonistic 16,85%, and synergistic 70,71% (Kalavrouziotis and Koukoulakis, 2012).

Similarly, 182 interactions occurred in *Brassica oleracea* var *gemmifera* (Brussel's sprouts) of which 71,62% were synergistic, 6,70% antagonistic and 4,07% biphasic (Kalavrouziotis et al., 2009d). The effect of the interactions irrespective of the plant organ it can be either positive or negative, depending on whether they are synergistic or antagonistic. It also depends on the kind of the interacting elements, that is whether they are plant nutrients or heavy metals. The interactions may contribute plant nutrients. If they are synergistic and occur between heavy metals and essential nutrients, if they occur between heavy metals and are synergistic they may contribute more heavy metals, hence the result will be negative. On the other hand, if the interactions are antagonistic, the result will be positive as less heavy metals will be supplied to soils or to plants.

Therefore, it becomes obvious that some interactions may cause a risk effect on human health and this must be borne in mind. For example, the interaction ClxCd being synergistic, may contribute significant quantities of Cd to treated wastewater, thus increasing the health risk effect on human beings where the wastewater is used for crop irrigation (Kalavrouziotis et al., 2011a).

It must be underlined that the reuse of TMWW is a multifactor problem related to social, economical, and environmental parameters. Consequently, it is directly connected and associated with the environmental quality due to the fact that it is in an immediate association and contact with the soil and plant system (Cheng et al., 1998). It has been reported by Kalavrouziotis et al. (2009b) in relation to the occurrence of the interactions in the soil system that out of 40 interactions which occurred under the effect of the TMWW, almost all of them were synergistic, and most of them were between heavy metals, a fact that suggested that the soil was supplied with heavy metals, which could have significant environmental impact. The presence of TMWW intensifies the elemental interactions, increasing significantly their number of occurrence (Kalavrouziotis et al., 2011a).

The elemental contribution of the interactions occurring in the plant, may not be always the same in a given plant organ. In other words, in the roots an interaction may be synergistic and in the leaves antagonistic. In many however cases, it may be in two or more plant organs constantly the same i.e. antagonistic or synergistic. For example, the interaction Cd x Pb has been found antagonistic in Broccoli leaves and synergistic in Broccoli roots. Similarly, the interaction Pb x Cd is synergistic in Broccoli roots (Kalavrouziotis et al., 2009a). As to the contribution of the interactions, in terms of heavy metals or plant nutrients, and their accumulation in the plants, a number of factors affect it, with the plant genotype being the most important (Kalavrouziotis et al., 2009c).

Elemental contribution of interactions

One important question that is raised with respect to the interaction's contribution is: "What is the quantitative contribution of the interactions in terms of metals or plant nutrients?". The answer to this question is based on the following rational: "The difference between two sites in the field in elemental concentration may be due to various reasons such as: variability of mineral content, microbial activity; water content, fixation, aeration, pH, organic matter, oxidation reduction potential, calcium carbonate, etc. All these factors affect the soil availability of the metals and plant nutrients. To these, the interactions must also be added, which undoubtedly occur continuously in the soil and affect the availability of heavy metals and plant nutrients. Consequently, to quantify the effect of interactions of the concentrations of the elements in general in the soil, it is necessary to run a regression analysis between the heavy metals, plant nutrients, and possibly the soil chemical and physical properties. The statistical approach to solution of this problem is analyzed below as follows (Kalavrouziotis et al., 2010; Koukoulakis et al., 2013).

1. Based on the analytical experimental data of soil heavy metals (Zn, Cu, Mn, Cd, Co, Cr, Ni, Pb), essential elements i.e. macro and micronutrients (N, P, K, Ca, Mg, Cl) and soil physical and chemical soil properties (pH, Organic Matter (OM), Calcium Carbonate (CC), and Electrical Conductivity (EC)), regression analysis is run according to the interactions given in Table 1. Among the regression equations produced, the statistically significant ones are chosen.

2. These significant regressions equations are taken into account for the quantification of the elemental contribution of each chosen interaction in terms of Fe, Zn, etc. Let us assume that the following interactions and their corresponding regression equations are statistically significant, i.e.

- | | | |
|----|---------|--|
| 1. | Fe x Zn | $Fe = \pm a(Zn)^2 \pm b(Zn) \pm c$ |
| 2. | Fe x Cu | $Fe_1 = \pm a_1(Cu)^2 \pm b_1(Cu) \pm c_1$ |
| 3. | Fe x Cd | $Fe_2 = \pm a_2(Cd)^2 \pm b_2(Cd) \pm c_2$ |
| 4. | Fe x Cr | $Fe_3 = \pm a_3(Cr)^2 \pm b_3(Cr) \pm c_3$ |
| 5. | Fe x Ni | $Fe_4 = \pm a_4(Ni)^2 \pm b_4(Ni) \pm c_4$ |

6. Fe x P $Fe_5 = \pm a_5(P)^2 \pm b_5(P) \pm c_5$
7. Fe x K $Fe_6 = \pm a_6(K)^2 \pm b_6(K) \pm c_6$
8. Fe x OM $Fe_7 = \pm a_7(OM)^2 \pm b_7(OM) \pm c_7$
9. Fe x pH $Fe_8 = \pm a_8(pH)^2 \pm b_8(pH) \pm c_8$
10. Fe x CC $Fe_9 = \pm a_9(CC)^2 \pm b_9(CC) \pm c_9$
11. Fe x EC $Fe_{10} = \pm a_{10}(EC)^2 \pm b_{10}(EC) \pm c_{10}$

Table 1. Interactions between heavy metals, macro and micronutrients and soil physical and chemical properties.

	Fe	Zn	Mn	Cu	Cd	Co	Cr	Ni	Pb	N	P	K	Ca	Mg	Cl	pH	OM	CC	EC
Fe	FeZn	FeMn	FeCu	FeCd	FeCo	FeCr	FeNi	FePb	FeN	FeP	FeK	FeCa	FeMg	FeCl	FeH	FeOM	FeCC	FeEC	
Zn	ZnFe	ZnMn	ZnCu	ZnCd	ZnCo	ZnCr	ZnNi	ZnPb	ZnN	ZnP	ZnK	ZnCa	ZnMg	ZnCl	ZnH	ZnOM	ZnCC	ZnEC	
Mn	MnFe	MnZn	MnCu	MnCd	MnCo	MnCr	MnNi	MnPb	MnN	MnP	MnK	MnCa	MnMg	MnCl	MnH	MnOM	MnCC	MnEC	
Cu	CuFe	CuZn	CuMn	CuCd	CuCo	CuCr	CuNi	CuPb	CuN	CuP	CuK	CuCa	CuMg	CuCl	CuH	CuOM	CuCC	CuEC	
Cd	CdFe	CdZn	CdMn	CdCu	CdCo	CdCr	CdNi	CdPb	CdN	CdP	CdK	CdCa	CdMg	CdCl	CdH	CdOM	CdCC	CdEC	
Co	CoFe	CoZn	CoMn	CoCu	CoCd	CoCr	CoNi	CoPb	CoN	CoP	CoK	CoCa	CoMg	CoCl	CoH	CoOM	CoCC	CoEC	
Cr	CrFe	CrZn	CrMn	CrCu	CrCd	CrCo	CrNi	CrPb	CrN	CrP	CrK	CrCa	CrMg	CrCl	CrH	CrOM	CrCC	CrEC	
Ni	NiFe	NiZn	NiMn	NiCu	NiCd	NiCo	NiCr	NiPb	NiN	NiP	NiK	NiCa	NiMg	NiCl	NiH	NiOM	NiCC	NiEC	
Pb	PbFe	PbZn	PbMn	PbCu	PbCd	PbCo	PbCr	PbNi	PbN	PbP	PbK	PbCa	PbMg	PbCl	PbH	PbOM	PbCC	PbEC	
N	NFe	NZn	NMn	NiCu	NiCd	NiCo	NiCr	NiNi	NiPb	NiN	NiP	NiK	NiCa	NiMg	NiCl	NiH	NiOM	NiCC	NiEC
P	PFe	PZn	PMn	PiCu	PiCd	PiCo	PiCr	PiNi	PiPb	PiN	PiP	PiK	PiCa	PiMg	PiCl	PiH	PiOM	PiCC	PiEC
K	KiFe	KiZn	KiMn	KiCu	KiCd	KiCo	KiCr	KiNi	KiPb	KiN	KiP	KiCa	KiMg	KiCl	KiH	KiOM	KiCC	KiEC	
Ca	CaFe	CaZn	CaMn	CaCu	CaCd	CaCo	CaCr	CaNi	CaPb	CaN	CaP	CaK	CaCa	CaMg	CaCl	CaH	CaOM	CaCC	CaEC
Mg	MgFe	MgZn	MgMn	MgCu	MgCd	MgCo	MgCr	MgNi	MgPb	MgN	MgP	MgK	MgCa	MgMg	MgCl	MgH	MgOM	MgCC	MgEC
Cl	ClFe	ClZn	ClMn	ClCu	ClCd	ClCo	ClCr	ClNi	ClPb	ClN	ClP	ClK	ClCa	ClMg	ClCl	ClH	ClOM	ClCC	ClEC
pH	pHFe	pHZn	pHMn	pHCu	pHCd	pHCo	pHCr	pHNi	pHPb	pHN	pHP	pHK	pHCa	pHMg	pHCl	pHH	pHOM	pHCC	pHEC
OM	OMFe	OMZn	OMMn	OMCu	OMCd	OMCo	OMCr	OMNi	OMPb	OMN	OMP	OMK	OMCa	OMMg	OMCl	OMH	OMOM	OMCC	OMEC
CC	CCFe	CCZn	CCMn	CCCu	CCCd	CCCo	CCCr	CCNi	CCPb	CCN	CCP	CCK	CCCa	CCMg	CCCl	CCH	CCOM	CCCC	CCEC
EC	ECFe	ECZn	ECMn	ECCu	ECd	ECCo	ECr	ECNi	ECPb	ECN	ECp	ECk	ECa	ECmg	ECcl	ECpH	ECOM	ECcc	ECec

3. Each one of the above equations is solved for the maximum and minimum value of the independent variable X (Xmax, Xmin), which in these examples is represented by the elements: Zn, Cu, Cd, EC, respectively. These values of the above elements can be easily found from the available analytical data of the experimental soil. Thus, by solving the above equations, the difference between the corresponding maximum, and minimum values of the dependent variable (Y) i.e. Y_{max} and Y_{min} , is found, for each regression equation, respectively, which in the present example is Fe_{max} and Fe_{min} respectively. These values are the results of purely interactions effect.

This difference for each of the above ten interactions given above as a general example, are as follows: $(Fe_{1max} - Fe_{1min})$, between $(Fe_{2max} - Fe_{2min}) \dots (Fe_{10max} - Fe_{10min})$.

4. From the same experimental data, the max and min values of the actual value of Fe can be found, and the difference $(Fe_{acmax} - Fe_{acmin})$ can be calculated which is due to the factors mentioned previously including the interaction effect. This difference is equal to 100% and represents the concentration of the element by which two sampling points in the same field differ in their metal content. Therefore, the contribution of interaction in terms of “Percent Elemental Contribution (PEC)” of a single element based on one regression equation, is given by the following relation (Koukoulakis et al., 2013):

$$PEC = \frac{(Y_{tmax} - Y_{tmin}) \times 100}{(Y_{amin} - Y_{amax})} \quad [1]$$

Where

- PEC = Percent Elemental Contribution (%)
- $(Y_{tmax} - Y_{tmin})$ = difference between the theoretical independent values of the given metal concentration in mg/Kg as determined by the solution of the respective regression equation.
- $(Y_{amin} - Y_{amax})$ = difference between the actual values of the dependent variable as it is given by the soil analytical data, representing a given metal in mg/Kg.

The importance of the interactions

Based on the results obtained so far in relation to the usefulness and importance of the elemental interactions (Koukoulakis et al., 2013; Kalavrouziotis and Koukoulakis 2012; Kalavrouziotis and Koukoulakis 2009a; Kalavrouziotis et al., 2009b; Kalavrouziotis et al., 2009c; Kalavrouziotis et al., 2008), the following may be summarized. The elemental interactions, which unfortunately have not been paid due attention by the scientific community, play an important economical, environmental, and social role.

The interactions between metals and macro and micronutrients under the effect of TMWW and sludge or biosolids are continuously taking place in the soil and plants, affecting soil fertility, productivity, plant growth, and yields. Also, the interactions affect forest plant growth, pasture's animal crops or evergreen plants of the ecosystem, thus influencing the environmental quality.

Furthermore, the positive heavy metal interactions (synergistic) may contribute toxic metals, which, in the long run may affect adversely human and animal health, causing the so called "silent toxicity" (Papaioannou et al., 2015). Also, the interactions can unfavorably affect the microflora and the macroflauna diversity in soil influencing positively or negatively soil quality.

Another important aspect of interactions is that they may be used to explain observed positive effects of heavy metals on plants. Such positive effect on plant growth may be due to contribution of the interactions in terms of plant nutrients (Kalavrouziotis and Koukoulakis 2011b; Bollard, 1983).

Soil metal pollution by the biosolid and wastewater reuse

Both, the biosolids and wastewater reuse, provide the closing of the plant nutrient cycle (Shomar et al., 2013). In fact, by the reuse a sustainable and ecological sound wastewater and sludge management can be accomplished, provided that the known health risk barriers are removed from these wastewater treatment byproducts. In other words, the agricultural application of biosolids or sludge must be

accompanied by concrete and well defined safety measures in order to secure a healthy environment and to protect the consumer's life. Such an application of both wastewater and biosolids could be very advantageous in the sense that it will be economic and a good supplier of plant nutrients, and friendly to the environment.

Biosolid application to soil may contribute heavy metals which could accumulate in the soil either in short term being applied at high levels, (Shomar et al., 2013), or in long term, applied in lower levels but repeated for many years.

Since our basic aim is to accomplish safe reuse of wastewater and of sludge or biosolids, we have developed pollution indices for the assessment of soil pollution. These indices, can be used to evaluate not only the wastewater and biosolid based, but of any other source which may contribute heavy metal to soil. For example industrial activities, metal smelting, mine operations, including crop fertilizer application. All these activities may cause soil metal pollution of the ecosystem. By definition the soil pollution is highly complex, elementally collective and very interactive process. Therefore, the pollution must be examined and be evaluated in relation not only to one metal, but collectively to all metals, which are involved in the process of pollution. Furthermore, not only the main effect of this element is related to soil pollution but also to the interrelationships, which should be paid special attention..

The pollution indices that have been developed by our research team, take into account all the metals involved, and in this respect, they are dynamic, but the result of the assessment is static, in the sense that the index assesses the metal pollution level at the moment of soil sampling, and its chemical analysis. Irrespective of this characteristic, the indices reflect a soil pollution caused collectively by all the existing soil metals, and this is a very useful information with respect to decision making as to the facing of the problem of soil amelioration.

The pollution indices that have been developed by our research team are as follows (Kalavrouziotis et al., 2012):

1. Elemental Pollution Index (EPI)

$$EPI = \sqrt[n]{M_1 \times M_2 \times M_3 \times \dots \times M_n} \quad [2]$$

Where

$M_1, M_2, M_3, \dots, M_n$ = the metals involved in the pollution in mg/Kg

2. Heavy Metal Load (HML)

$$HML = M_1 + M_2 + M_3 + \dots + M_n \quad [3]$$

Where

$M_1, M_2, M_3, \dots, M_n$ = the metals involved in the pollution in mg/Kg

3. Total Concentration Factor (TFC)

$$TCF = \frac{M_1 + M_2 + M_3 + \dots + M_n}{M_{1r} + M_{2r} + M_{3r} + \dots + M_{nr}} \quad [4]$$

Where

$M_1, M_2, M_3, \dots, M_n$ = the metals involved in the pollution in mg/Kg
 $M_{1r}, M_{2r}, M_{3r}, \dots, M_{nr}$ = the corresponding reference values of these metals.

The above pollution indices have been calibrated on the basis of the percent dry matter loss of *Beta vulgaris L.*, and the relevant calibration data has been tabulated to assess the level of soil pollution (Papaioannou et al., 2016).

The basic characteristic of the above pollution indices, is that they are easy and simple to use and that EPI and HML do not need to have the reference values of metals, but only TCF

The PLI index of Cabrera et al., (1999) has been used as a reference index for the above three, indices for reasons of comparison.

Conclusion

Based on the above the following conclusions can be drawn:

- in spite of the fact that the agricultural reuse of wastewater and biosolids (sludge) is a routine practice in several countries, especially in Mediterranean area, the “safety” of land disposal of these inputs is in many cases questionable.
- the replacement of the concept of “waste” by the: “recycling of resources” will be possible only upon successfully addressing the risk health problems involved in the reuse of wastewater and especially of sludge. This will be possible with more systematic research and continuous effort towards accomplishing a science - based reuse;
- also it is a “sinus quam non” necessity, that the existing guidelines, controlling the reuse of sludge and wastewater, must be reconsidered, in the light of the new research results, due to the fact that health risk problems appear even in cases where the reuse is applied in compliance with the existing official guidelines and regulations;
- health problems related to pathogens have appeared in the many cases in people living around and near the sites of application of sludge. Furthermore, the increase in antibiotic resistance of bacteria is an “international health crisis”, toxic metals accumulate in the edible plant parts. Also the existence of many unknown interactions and uncertainties render the application of sludge not safe. Unless these problems are successfully addressed the “safety” of reuse will be always questionable;
- the scientific community must be supplied with all necessary research infrastructure and needed by the states and by the Organizations involved in the reuse, to work towards accomplishing the safety of sludge and wastewater reuse, necessary for the effective management of these inputs, so as to release the

pressure exerted by the huge quantities of wastewater and sludge produced universally every year;

- the research team of the School of Science and Technology of the Open University of Patras has been working for the last 20 years with the accumulation of heavy metals in soils and plants with emphasis in the edible parts, heavy metal interactions, their elemental contribution to soils and plants, and also with the establishment of pollution indices for the assessment and forecasting soil pollution from the long term reuse of wastewater and sludge.

References

- BOLAN N., ANDRANO D.C., NAIDU R., LA RUZ MOVA M, SANDIAGO M. (2005) Phosphorous trace element interactions in plants system. In: Phosphorous in Agriculture and Environment, Madison, WI, ASA, SSSA, pp: 317-350.
- BOLLARD E.G. (1983) Involvement of unusual elements in plant growth, and nutrition. In: Lauchli, A., Bieleski, R.L. (Eds.) Encyclopedia of plant Physiology, New Series 15B, pp: 695-755.
- CABRERA F., CLEMENTE L., DÍAZ BARRIENTOS E., LÓPEZ R., MURILLO J.M.(1999) Heavy metal pollution of soils affected by the Guadiamar toxic flood. The Science of Total Environment, 242:117–129.
- CHANG A.C., PAGE A.L., ASANO T., HESPAHOL J. (1998) Evaluating methods of establishing health-related guidelines for crop land application of municipal water. In: Asano, T., Wastewater reclamation and reuse. Technomic Publishing Lancaster, Basel, USA.
- CZEKALSKI N, BERTHOLD T, CAUCCI S, EGLI A, BÜRGMANN H. (2012) Increased levels of multiresistant bacteria and resistance genes after wastewater treatment and their dissemination into Lake Geneva, Switzerland. *Frontiers in Microbiology*, 3:106.
- EFROMYMSON R.A., SAMPLE B.E. et al., 1998. Evaluation of ecological risks associated with land application of municipal sewage sludge. Oak Ridge national Laboratory ORNL/TM-13703 Sep. 30,1998.
- FREY B., RUDT A., WIDMER F. (2010) Changes in rhizosphere bacterial population during phytoextraction of heavy metal contaminated soil with poplar. *Plant Biosystems*, 144:392-395.
- GOSH J. (2005) Bioaerosols generated from biosolids applied farm fields in wood county, Ohio 2005. Dissertation, Bowling Green State University.
- HARRISON E.Z., OAKES S.R. (2002) Investigation of alleged health incidents associated with land application of sewage sludges. *New Solutions: A Journal of Environmental and Occupational Health Policy*, 12:387-408.
- HARRISON E.Z., OAKES S.R., HYSSELL M., HAY A. (2006) Organic chemicals in sewage sludges. *The Science of Total Environment* 367, 481–497.
- KALAVROUZIOS I.K., KOUKOULAKIS P.H., ROBOLAS P., PAPADOPOULOS A.H., PANTAZIS V. (2008) Macro and micronutrients interactions in soil under the effect of Brassica oleracea var. Italica, irrigated with treated wastewater. *Fresenius Environmental Bulletin*, 17:1270-1282.
- KALAVROUZIOS I.K., KOUKOULAKIS P.H. (2009a). Distribution of elemental interactions in Brussels sprouts plants, under the Treated Municipal Wastewater. *Journal of Plant Interactions*, 4:219–231.

- KALAVROUZOTIS I.K., KOUKOULAKIS P.H., PAPADOPOULOS A.H. (2009b) Heavy metal interrelationships in soil in the presence of treated wastewater. *Global Nest J.*, 11:497-509.
- KALAVROUZOTIS I.K., KOUKOULAKIS P.H., MANOURIS G., PAPADOPOULOS A.H. (2009c) Interactions between Cd, Pb, Co and Ni in broccoli, irrigated with treated municipal wastewater. *European Water Journal*, 26:13-23.
- KALAVROUZOTIS I.K., KOUKOULAKIS P.H., MEHRA A. (2010) Quantification of elemental interaction effects on Brussels sprouts under treated municipal wastewater. *Desalination*, 254:6-11.
- KALAVROUZOTIS I.K., KOSTAKIOTI E., KOUKOULAKIS P.H., PAPADOPOULOS A.H., LEOTSINIDIS M., SAKAZLI E. (2011a) The impact of ClxCd interrelationship on planning wastewater reuse in cabbage. *Water Air and Soil Pollution*, 214:565-573.
- KALAVROUZOTIS I.K., KOUKOULAKIS P.H. (2011b) Plant nutrition aspects under treated wastewater reuse management. *Water, Air, Soil pollution*, 218:445-456.
- KALAVROUZOTIS I.K., KOUKOULAKIS P.H. (2012) Contribution of elemental interactions in total essential nutrient and heavy metal content in cabbage under treated wastewater irrigation. *Plant Biosystems - An International Journal Dealing with all Aspects of Plant Biology*, 146:491-499.
- KALAVROUZOTIS I.K., KOUKOULAKIS P.H., NTZALA G., PAPADOPOULOS A.H. (2012) Proposed indices for assessing soil pollution under the application of sludge. *Water, Air, Soil pollution*, 223:5189-5196.
- KHUDER S., MILZ S.A., BISESI M., VINCENT R., McNULTY W., CZAJKOWSKI K. (2007) Health survey of residents living near farm fields permitted to receive biosolids. *Archives of Environmental and Occupational Health*, 62(1):5-11.
- KOUKOULAKIS P.H., CHATZISSAVVIDIS C., PAPADOPOULOS A.H., PONTIKIS D. (2013) Interactions between leaf macronutrients and micronutrients and soil properties in pistachio (*Pistacia vera* L.) orchards. *Acta Botanica Croatica* 72, 295-310.
- LEWIS D. L., GATTIE, D.K., 2002. Pathogen risks from applying sewage sludge to land. *Environmental Science of Technology* 36, 286-293.
- LEWIS D.L., GATTIE D.K., NOVAK M.E., SANCHEZ S., PUMPHREY C. (2002a) Interactions of pathogens and irritant chemicals in land-applied sewage sludges (biosolids). *BMC public health* 2, 11.
- LOWMAN A., MCDONALD M.A., WING S., MUHAMMAD N. (2013) Land Application of Treated Sewage Sludge: Community Health and Environmental Justice. *Environmental Health Perspectives* 121, 537-542.
- MARSCHNER, H., 2002. Mineral Nutrition of Higher Plants. An Elsevier Science Imprint, Amsterdam, Academic Press, pp: 38.
- McBRIDE M.B. (2003) Toxic metals in sewage sludge-amended soils: has promotion of beneficial use discounted the risks? *Adv. Environ. Res.*, 6:5-19.
- NRC (1996) Use of reclaimed water and sludge in food production. National Academy Press, Washington, D.C. 1996.
- NRC (2002) National Academy of Sciences(NAS) Biosolids applied to land; Advancing standards and practices, Washington D.C. National Academy press (prepublication copy)
- OLSEN S.R. (1972) Micronutrient Interactions. In: *Micronutrients in Agriculture*, Mortved, J.M.J.J., Goirdano P.M., Lindsay, W.L. (Eds.). SSSA, Madison, WI, pp: 243-264.
- PAPAIOANNOU D., KALAVROUZOTIS I.K., KOUKOULAKIS P.H., PAPADOPOULOS, F. (2015) A proposed method for assessment of the interactive heavy metal accumulation in soils. *Global NEST Journal*, 17:835-846.

- PAPAIOANNOU D., KALAVROUZOTIS I.K., KOUKOULAKIS P.H., PAPADOPOULOS F. (2016) Critical ranges of pollution indices: a tool for predicting soil metal pollution under long term reuse of wastewater. *Toxicological and Environmental Chemistry* (In press)
- ROBSON A.D., PITMAN J.B. (1983) Interactions between nutrients in higher plants. In: Läubli, A., Bielecki, R. L. (Ed.). *Inorganic plant nutrition*. New York: Springer, pp:147-18.
- SHEAR C.B., CRANE H.L., MAYERS A.T. (1946) Nutrient element balance: A fundamental concept in plant nutrition. *Proc. Amer. Hort. Sci.*, 47:237-245.
- SHOMAR B., KALAVROUZOTIS I.K., KOUKOULAKIS P.H., VAHYA A. (2013) Soil pollution indices under the effect of sludge. *Water, Air, Soil pollution*, 224:1436
- SNYDER C. (2005) The Dirty Work of Promoting “Recycling” of America’s Sewage Sludge. *International Journal of Occupational and Environmental Health*, 11:415-427.
- UN-HABITAT (2008) *Global Atlas Of Excreta, Wastewater Sludge, And Biosolids Management: Moving Forward The Sustainable And Welcome Uses Of A Global Resource*. Ronald J. LeBlanc, Peter Matthews, Roland P. Richard (eds). UN HABITAT POBOX 30030, Nairobi, Kenya.
- ZHANG Y., MARRS C.F., SIMON C., XI C. (2009) Wastewater treatment contributes to selective increase of antibiotic resistance among *Acinetobacter* spp. *The Science of Total Environment*, 407:3702–3706.